EUROPEAN SPALLATION SOURCE

Science and Engineering Research Council
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Instrumentation & Techniques for the European Spallation Source

Report of an Expert Meeting
held at
The Cosener's House, Abingdon
24-27 February 1992

A D Taylor (Ed.)

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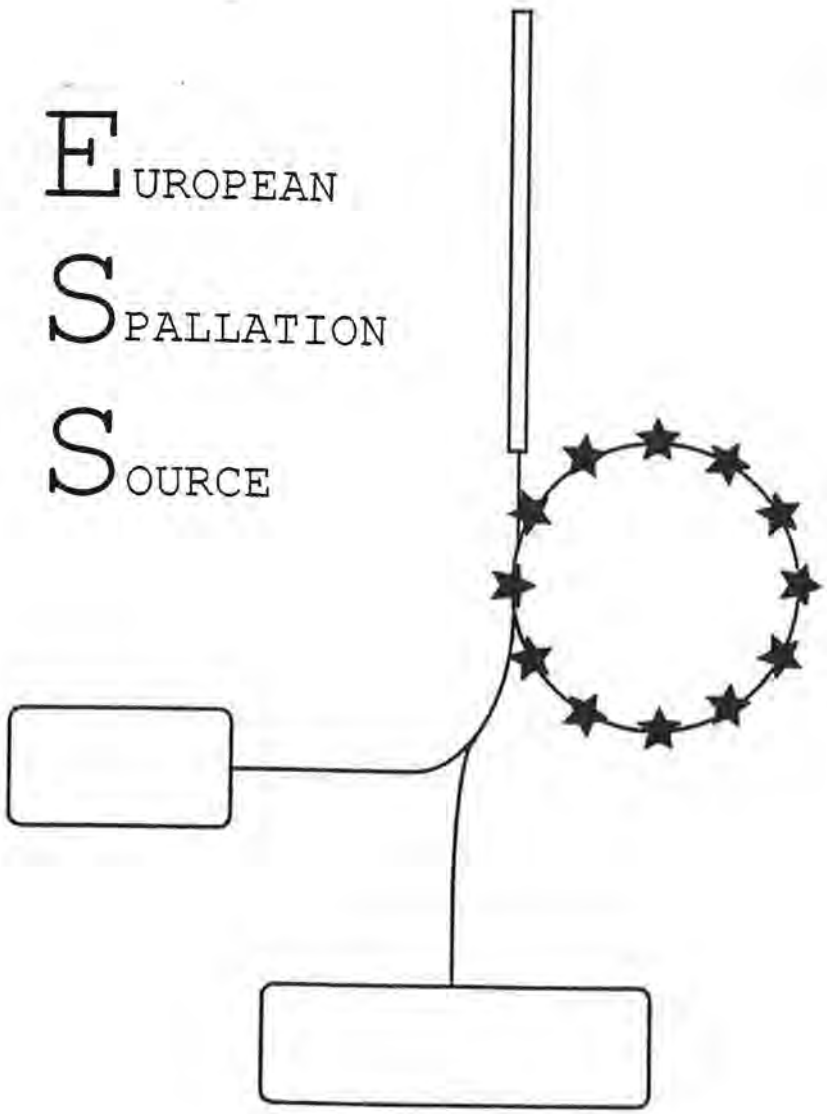

Prologue

Neutron scattering provides basic microscopic information on the structure and dynamics of materials which underpins our understanding of condensed matter in fields as diverse as biology, materials science, chemistry, the earth sciences and physics. In the Large Facilities Report to the CEC in 1990, the Neutron Study Panel underlined the continuing need for neutron scattering as a microscopic probe of the condensed state, and recognised that a major initiative was necessary to secure an effective ongoing neutron science programme in Europe for the year 2000 and beyond. The Panel recommended that a design study be initiated for a next generation neutron source. Subsequently, in a joint initiative by KFA Jülich and Rutherford Appleton Laboratory, a series of meetings have been held to explore options for such a next generation European Neutron Source. The CEC has provided support for two of the meetings, an Expert Meeting on Accelerators (held at Simonskall near Jülich in September 1991) and an Expert Meeting on Instrumentation and Techniques (held at Abingdon near RAL in February 1992). A further meeting on Target Technologies jointly organised with PSI Villigen took place in early February 1992.

These meetings focused on the provision and utilisation of an advanced high-powered accelerator-driven pulsed spallation source and the scientific opportunities that it would herald. Discussions of the Expert Meeting on Accelerators (and subsequent workshops on various aspects of the accelerator design) are summarised in a Jülich Report, those of the Target Technologies and Moderators Workshop in the PSI Report PSI-Proceedings-92-03. This is the Report of the Expert Meeting on Instrumentation and Techniques.

The source specification for the European Spallation Source (ESS) is based on a proton accelerator producing a pulsed beam with an average beam power of 5 MW in ~1 μs pulses at a repetition rate of 50 Hz. Two target stations are envisaged, one for operation at 10 Hz for high resolution and long wavelength instruments and one operating at 50 Hz for high intensity applications. The neutronic performance anticipated from this specification will make the European Spallation Source some 30 times brighter than ISIS, presently the world's most powerful source of this type. The ESS will have a time averaged thermal flux equivalent to that of the High Flux Reactor at the Institut Laue Langevin (ILL), but because of its sharp pulse structure, the peak flux of the ESS will be some 40 times higher than this average.

An accelerator concept has been identified which will meet this specification using technologies which are considered by the Expert Meeting to be within attainable goals. A target design concept to dissipate 5MW and an appropriate moderator system did not appear to pose any insurmountable problems. The scientific benefit that the ESS would bring to a wide range of scientific disciplines was considered in detail at the Expert Meeting on Instrumentation & Techniques. It was confidently predicted that the enhanced temporal brightness of the ESS would allow the source to make both a major impact on established fields and substantial contributions to new areas of research.

A proposal for a two year design study which would investigate various aspects of accelerator technology and confirm the feasibility of the target design and neutronic performance of the moderators is being prepared, Neutron scattering technique development identified at this Meeting will be the subject of a parallel European-wide development request.

Construction of the European Spallation Source would maintain Europe in the pre-eminent position that it has held for the past 20 years in the field of neutron scattering. In addition, it is anticipated that the ESS would have a major impact on other areas such as neutrino physics and muon science, and would provide a strategic opportunity to develop high-powered accelerator technology with potential applications in nuclear waste management and isotope production.

Instrumentation & Techniques for the European Spallation Source

1 Introduction

An Expert Meeting on Neutron Instrumentation and Techniques for the European Spallation Source was held at the Cosener's House, Abingdon in the week 24-27 February 1992. About 70 experts in the applications of neutron scattering to problems in condensed matter science from some 14 countries participated. The meeting was organised jointly by Rutherford Appleton Laboratory (RAL) and Kernforschungsanlange (KFA) Jülich.

This was the third of a series of recent meetings which have explored the options for a next generation European Neutron Source. The CEC has provided support for two of the meetings, an Expert Meeting on Accelerators (at Simonskall near Jülich in September 1991) and the present Expert Meeting on Instrumentation and Techniques. A further meeting on Target Technologies was held jointly with PSI Villigen in early February 1992.

These meetings focused on the provision and utilisation of an advanced high-powered accelerator-driven pulsed spallation neutron source and the scientific opportunities that it would herald. Discussions of the Expert Meeting on Accelerators (and subsequent workshops on various aspects of the accelerator design) are summarised in a Jülich Report and those of the Target Technologies and Moderators Workshop in the PSI Report PSI-92-03.

This Report outlines the scientific potential offered by the European Spallation Source. Over a period of three days, individual Working Groups covered the following areas:

- Crystallography;
- Liquids Amorphous and Disordered Systems;
- Large Scale Structures;
- Excitations;
- High Resolution Spectroscopy.

The Working Groups discussed the impact the ESS would have on their fields, specified a reference suite of instruments and identified the R&D required to exploit the source fully. Their detailed discussions are summarised in the Working Group Reports which are attached as Appendices to this document.

Although the Meeting confined its discussions almost entirely to the utilisation of the ESS for neutron scattering, clearly a major facility like the ESS would have a much wider scientific

impact. Relevant areas include fundamental physics studies of the neutron itself (electric dipole moment, neutron lifetime, neutron-antineutron oscillations etc.), interferometry, nuclear physics studies and neutrino production. It would also have a major impact as a muon source, not only for condensed matter studies, but also for fundamental physics and fusion studies. Other applications include irradiation, materials damage studies, isotope production, medical and advanced accelerator studies.

2 Neutron Scattering In Condensed Matter Research

Together with other established techniques such as X-ray, electron and light scattering, and emerging techniques such as the scanning tunnelling microscope, neutrons provide a powerful probe of condensed matter on an atomic level. It should be stressed that all of these are complementary probes, and often more than one must be brought to bear to answer real scientific and technical questions. As we move towards the 21st century the complexity of technology, and of the scientific questions posed, will increasingly require an understanding of materials at the atomic level. Examples range from the production of a vaccine for AIDS, and understanding how to make higher density data storage devices, to questions of the chemistry of pollution, and the production of stronger, lighter and more reliable engineering components. While we would never suggest that neutrons alone can provide the answers to these problems, they are one of the key microscopic techniques with which the base materials of so much of our 'real' world will be examined, understood and improved.

In proposing a new neutron source we must be aware not only of the vast amount of work presently being done at neutron sources around the world, but also of the potential for progress if higher intensities can be achieved. Neutron scattering is a technique which is severely constrained by the weak intensities available. In comparison with present or future X-ray synchrotrons, the source brightness is many orders of magnitude lower. The strong motivation for continuing with neutron scattering is the unique information that can be extracted from this technique. This uniqueness arises from a number of reasons:

- Neutrons interact with atoms principally through the short range nuclear force, which varies irregularly from nucleus to nucleus. Light atoms may often be studied in the presence of heavy ones, and significant differences in the interaction generally exist between different isotopes of the same element. Undoubtedly the most widely exploited isotopic difference is that between hydrogen and deuterium. By the process of selective deuteration, neutron experiments are able to examine parts of complex molecules in many fundamental areas of chemistry, polymer science and biology. Soft condensed matter studies using neutrons have grown explosively over the last decade, and the problems being addressed now impinge significantly on the commercial world.

- The energy and momentum of neutrons may be matched simultaneously to energy and length scales appropriate for excitations in condensed matter. Neutron wavelengths of 1 - 10 Å are ideal for the study of interatomic correlations and the available neutron energies of 1 to 1000 meV allow the measurement of excitations over nine decades from neV to eV.

- The neutron has a magnetic moment which couples to spatial variations of the magnetisation of materials on the atomic scale. Neutrons are thus ideally suited for the study of magnetic structures (and hence microscopic magnetism) and short wavelength magnetic fluctuations. A recent example of this kind of measurement is the mapping of the dynamical susceptibility of high T_c superconductors, thus directly addressing the question of electronic correlations in this important class of new materials.

- The neutron is a weak probe, and does not significantly perturb the system. The resultant data may be directly compared with theory. In many cases absolute cross-sections may be obtained. This must be contrasted with the difficulties in interpreting information from strongly interacting probes such as photons or electrons.

- The low absorption of neutrons and the lack of charge allows neutrons to probe the bulk of materials. This feature, which has made possible the construction of sophisticated sample environment equipment, is now being exploited in the study of residual stress in engineering components and composite materials

3 The Advantages of a Pulsed Neutron Source

There are presently two main types of source used for neutron research, (see figure 1), steady state thermal sources based on fission reactors using highly enriched ^{235}U fuel and accelerator driven pulsed sources which produce neutrons by spallation of a non-fissile target. The pre-eminent steady state source is the 58 MW High Flux Reactor at the Institut Laue Langevin in Grenoble. Although the reactor was constructed in the early 1970s, with the appropriate refurbishment it will continue to operate at some 10^{15} n/cm^{2}s for many years. More recently, pulsed sources have been developed and the most powerful, ISIS at the Rutherford Appleton Laboratory, is now approaching its design current of 200 μA at 800 meV (160 kW beam power). These two sources are generally considered to be complementary, each having areas where it excels, but with a broad middle ground where both are able to make equivalent contributions.

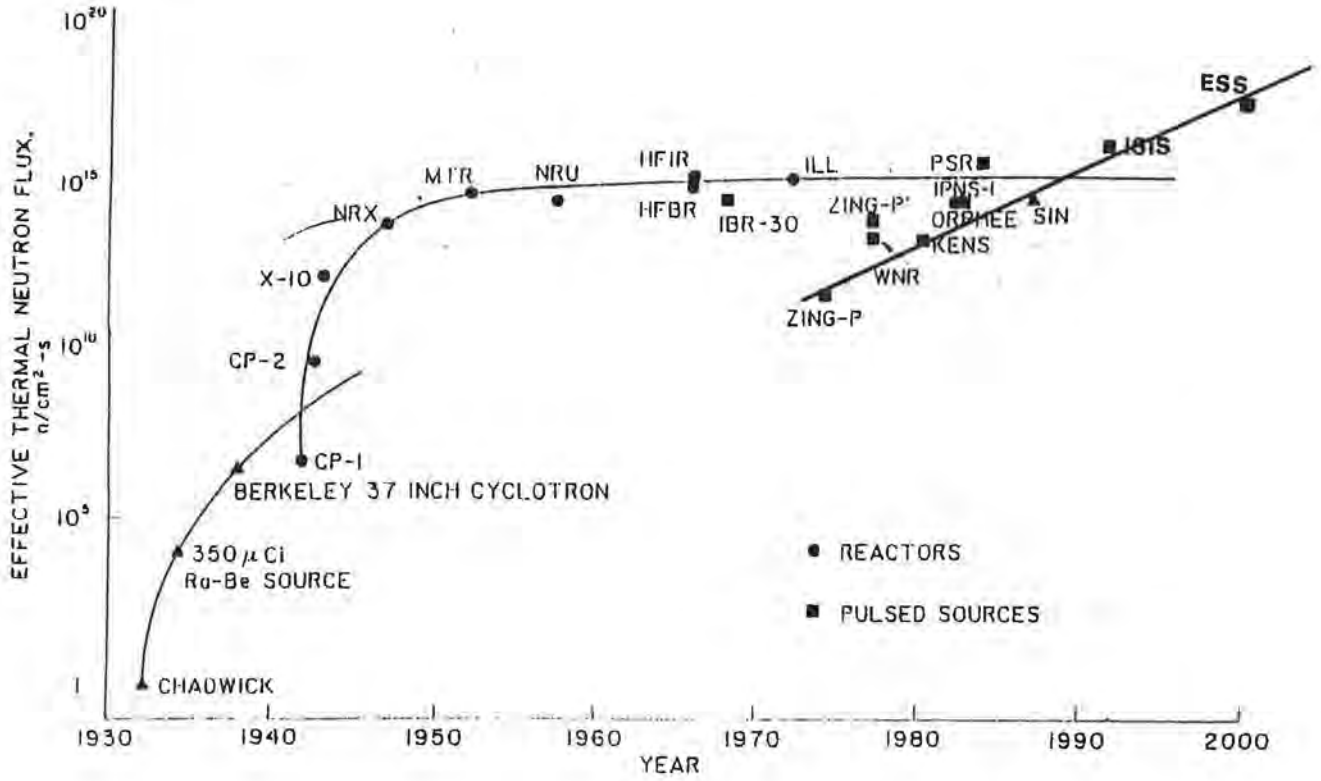

Figure 1 The growth in steady state and pulsed neutron sources in the past 50 years.

The two types of source do, however, differ significantly in their potential. Reactor technology is limited by thermo-mechanical constraints to only modest advances. The Advanced Neutron Source project at the Oak Ridge National Laboratory, USA offers a gain over the ILL some 30 years later of only a factor 5, whereas the potential gains for an accelerator-based source are much more substantial. Compared with the earlier spallation sources, ISIS shows gains already of some orders of magnitude, and, given the continued development of accelerators and recent technological interest in them for isotope production and transmutation of nuclear waste, this progress is expected to continue. In addition to increased raw brightness -- the proposed new source will be a factor of 30 brighter than ISIS -- there are many ways to exploit the pulse structure that are as yet relatively unexplored .

Pulsed sources produce and use neutrons in a qualitatively different way. The scientific opportunities resulting from these differences have contributed to the rapid progress that has

been made in the past few years, particularly at ISIS. Exploitation of these new opportunities has led to the emergence of new techniques and areas of research. Examples include reflection studies at the air-liquid interface, high quality structural data from complex polycrystalline materials, high energy excitations in low-dimensional quantum spin systems, diffuse scattering studies of disordered materials, H...H correlations in aqueous liquids, demonstration of the Bose condensate in liquid helium, and wide band tunnelling spectroscopy.

The known advantages of pulsed sources include:

- the ability to use of time-of-flight in a natural way;
- high intrinsic resolution in both momentum transfer **Q** and energy transfer ε, resulting in very high precision structural and dynamical studies;
- excellent signal to background : the source is effectively 'off' when data are being collected, with fast neutron backgrounds being easily eliminated by time-of-flight techniques. This allows subtle effects to be observed;
- wide spectral range in both **Q** and ε: with the trend towards ever more complex systems, access to data over as wide a dynamical range as possible is more and more essential;
- use of fixed scattering geometries which greatly facilitate the use of complex and extreme sample environment;
- the pulsed nature itself which leads naturally to the study of time-dependent perturbations, kinetic experiments such as *in situ* chemical reactions etc., and the exploitation of extreme conditions which cannot be achieved in the steady state;
- determination of absolute neutron scattering cross-sections: this is very powerful when combined with theory or computer simulation, since the cross-section for neutron interactions may often be calculated exactly;
- ability to perform simultaneous diffraction while studying excitations. This has been shown to be very important in many fields, particularly in soft condensed matter (liquid crystals, molecules adsorbed or intercalated) where a complex phase has been prepared in situ and must be monitored simultaneously with the measurement of the excitations.

New advantages will emerge as detector technologies improve (large area detectors can in general be more effectively exploited with time-of-flight) and beam polarisation devices based on ^{3}He become a reality. The potentially vast amount of data available from a pulsed source is

both a problem and a challenge. Developments in computing technology are growing even faster than pulsed neutron source brightness, and they will play a major role in developing further scientific exploitation through advanced data handling and visualisation techniques. In certain cases these advantages will lead to a further order of magnitude increase in the scientific effectiveness of pulsed sources.

4 Source Specification

In its fifty year history, there have been substantial developments of the technique of neutron scattering both in terms of the sophistication of its instrumentation and the breadth of its impact. At one time neutron scattering was solely a tool of solid state physics -- now its applications range from biology, through chemistry and earth sciences to materials research and engineering. Neutron scattering however is an intensity limited technique, and consequently there is ever increasing scientific demand for brighter sources.

A new type of neutron source has recently emerged based on particle accelerators rather than the traditional nuclear fission reactors. Advances in accelerator technology have given this type of source a very strong growth curve over the past two decades, Figure 1. These pulsed spallation sources produce intense bursts of fast neutrons by spallation of high atomic number materials with energetic protons. The fast neutrons are subsequently moderated to thermal energies in small hydrogenous moderators with little degradation in pulse structure. By using instrumentation which exploits time-of-flight (TOF) techniques, these sources now rival the best fission reactors for condensed matter research.

There has been extensive research and development in recent years in the field of high power accelerators for both the Strategic Defence Initiative and for studies of accelerator transmutation of nuclear waste. Much of this work will be of direct benefit to the proposed study for a next generation spallation neutron source.

The source specification for the ESS is based on a proton accelerator producing a pulsed beam with an average beam power of 5 MW in ~1 μs pulses at a repetition rate of 50 Hz. Two target stations are envisaged, one for operation at 10 Hz for high resolution and long wavelength instruments and one operating at 50 Hz for high intensity applications. Although much detailed work remains to be done, the conclusion of the Simonskall and PSI meetings is that an accelerator concept can be identified which would meet this specification, and engineering solutions can be envisaged for all aspects of the target-moderator system.

The neutronic performance anticipated from this specification is summarised in the neutron intensity and pulse width characteristics of Figures 2 and 3. In its high resolution mode, the source would be some 30 times brighter than ISIS, presently the world's most powerful source

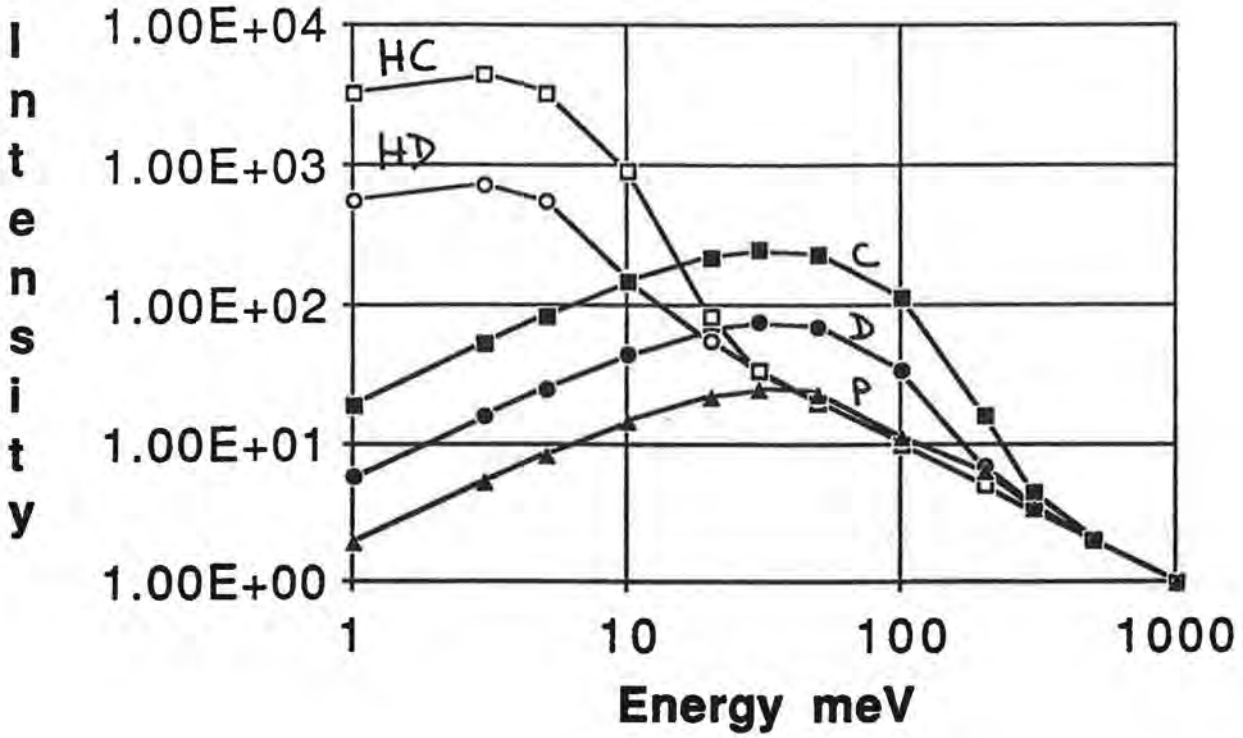

Figure 1 The intensity of the ambient moderators (closed symbols): Poisoned Water (triangle), Decoupled Water (circle), Coupled Water (square) and the cryogenic moderators(open symbols): Decoupled Hydrogen (circle) and Coupled Hydrogen (square) moderators. For a 5 MW spallation source the units are 2.2 10^{14} n.eV.sr.100cm^2.s

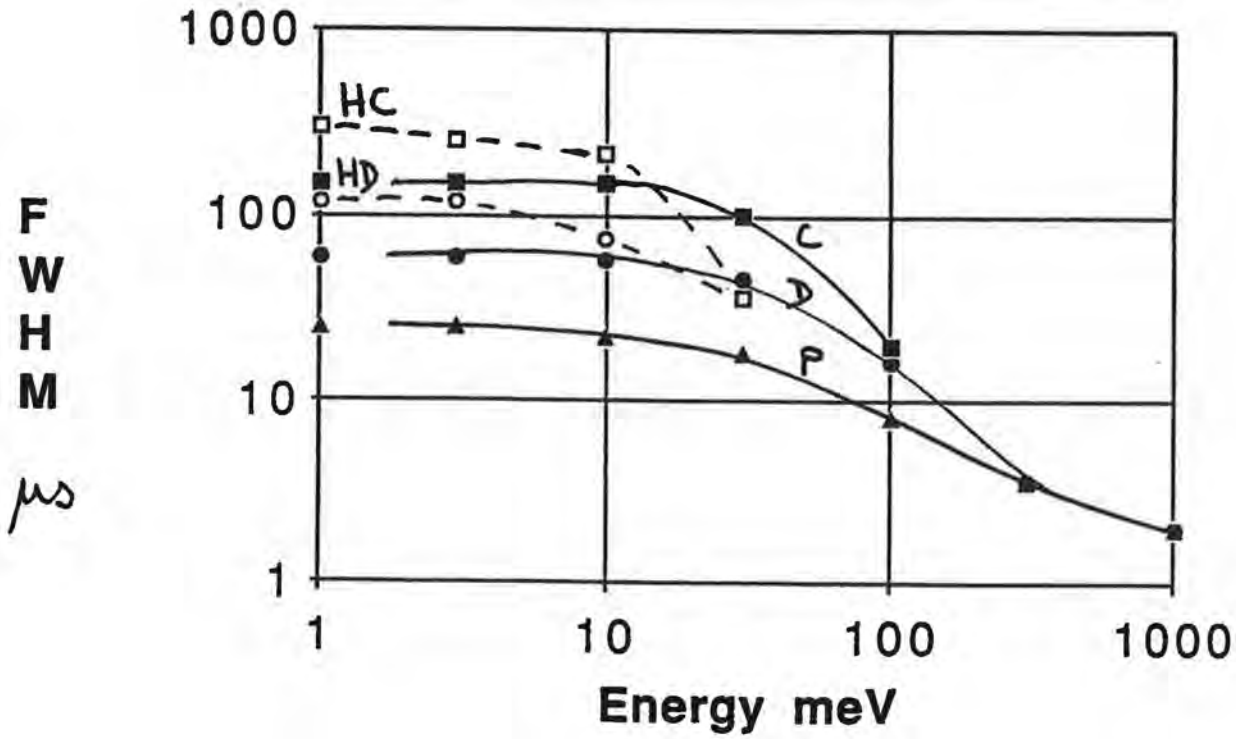

Figure 2 The pulse width (FWHM μs) for the above moderators. The pulse shapes are asymmetric with a sharp rising edge and long time tail.

of this type. It would have a time averaged flux equivalent to that of the High Flux Reactor at the Institut Laue Langevin (ILL), but, because of its sharp pulse structure, with a peak to average ratio greater than a factor 40.

The benefits of a neutron source with such an increase in source brightness and the instrumental opportunities presented by the sharp pulse structure are discussed in the following sections.

5 The Scientific Case for the European Spallation Source

The outstanding temporal brightness of the ESS is the key factor which will open up new opportunities in physics, chemistry, earth and materials science and biology. The intensity, resolution and signal-to-noise of instruments on the ESS will all show significant improvements over present generation sources and will not only expand opportunities in established fields but also open up new areas of research.

Neutron scattering is an intensity limited technique. The ESS will be a substantially brighter neutron source and a qualitatively different type of source from the traditional fission reactor source. One of the major issues discussed at the Abingdon meeting was instrumentation and techniques required to exploit this source fully. Another major question addressed was the qualitatively new science that might emerge from its exploitation.

The increase in source brightness will significantly change both the quantity and quality of information that can be obtained from neutron scattering. With more intensity, experiments can be performed on smaller samples or in a shorter time. It is not simply that samples 'smaller' - in the physical sense - can be studied (although this is an important aspect with new exotic materials, with systems subjected to extremes of pressure, temperature or external field, and with systems that are inherently small such as a thin film or the stress at a crack tip). Higher neutron intensities will also allow more dilute systems - systems at the natural state concentrations of interest for example to biologists - to be investigated. Parametric studies (in composition, temperature, pressure, humidity, external fields and combinations of the above) will be possible in hours for measurements which today would take days or weeks and are therefore not presently feasible.

Higher sensitivity and better signal-to-noise will generalise the isotopic substitution technique and allow new weak cross-section phenomena to be investigated. Pulsed techniques will be employed to provide extremes of sample environment (pressure, magnetic and electric fields). Time-dependent relaxation phenomena and kinetic processes will be investigated effectively. Higher resolution and an increased range in both space and time will be accessible, inevitably leading to a better and more detailed understanding of complex phenomena.

An overwhelming theme in the discussions of the Working Groups was the trend - for technological as well as scientific reasons - in all areas towards greater complexity. The systems under investigation at the forefront of modern biology, material science, chemistry and physics are becoming so complex that all possible microscopic probes must be used in an attempt to understand the phenomena: only in the simplest of systems does one probe give all the answers. Synchrotron X-ray and neutron sources will both have a complementary role to play in studying new materials. Examples abound in:

- the ceramic high T_c superconductors. These are complex structures with defect oxygens playing a crucial role in determining their specific properties;
- the new industrially-important hard magnetic materials (being investigated under the CEC's CEAM initiative) such as $Nd_2Fe_{14}B$ have a large, 64 atom unit cell and multiple exchange constants;
- biology where one trend is towards investigating biological function from dynamical as well as structural measurements;
- chemistry where increasing complexity in the organisation of polymers and surfactants at interfaces and in aggregates such as micelles is being studied;
- in materials science where multi-component ceramics will be the structural materials of choice for many high technology applications in the 21st Century;
- liquids where dilute aqueous and non aqueous solutions are being studied for both biotechnological as well as chemical reasons;
- molecular solids where interest has progressed from molecules, through simple polymers to proteins and complex biological systems such as crystalline insect flight muscle.

Although we can be very confident that the new scientific opportunities offered by the ESS will lead to major scientific and technological developments, some of the most dramatic are, by their very nature, not predictable. High temperature superconductors provide a very good example of why we cannot predict the future: they would not have been considered in such discussions six years ago, but nevertheless their study has been a major part of the programmes at all neutron sources in the past few years. Our current understanding of these materials is due in no small measure to the neutron technique, and the fact that the required sources and appropriate instrumentation existed and could be immediately brought to bear on these problems. The same will be true in the future: nature will continue to present us with new exotic materials with interesting and sometimes technologically important properties -- the

exotic form of carbon, buckminster fullerene is one recent example -- which require a diverse portfolio of probes to elucidate their underlying microscopic properties. It is important that an intense neutron source is part of that armoury.

The Working Groups identified a large number of scientific areas which will require a source of the power and characteristics of the ESS to be exploited effectively. The most exciting challenges are summarised as follows:

The **Crystallography Working Group** were excited at the breadth of research that would become possible with the ESS. In particular, major advances were anticipated in

- *biology* where high resolution single crystal structural studies of proteins would be possible, enabling the routine location of functionally important hydrogen atoms thus complementing X-ray synchrotron studies;

- *materials science* and engineering, where the increased ESS flux would be used to study the emerging complex "21st century" materials with the finer beams and higher resolution that are required;

- *physics* where for example our understanding of magnetism and superconductivity would be considerably enhanced by the ability

  - to determine precise magnetic moments and detect weak incommensurate signals ;
  - to measure simultaneously the average and local nuclear and magnetic structure of a material;
  - to utilise the increased flux to probe surface and thin layer magnetic structure;
  - to investigate time-dependent phenomena and magnetic and structural studies in high applied fields and/or pressures;

- *chemistry* where the intense ESS flux coupled with the ability to obtain a full diffraction pattern at fixed angle offer unrivalled possibilities for the detailed study of in situ catalysis, materials formation, reaction kinetics, decomposition and thermal diffractometry.

New 'hot' topics such as the high T_c superconductors and fullerenes will no doubt arise. Ten years ago the examples would have been different (heavy fermions, zeolites, organometallics) but the need to characterise these new materials, and the importance of neutron scattering would be unchanged. We cannot predict what the new materials of tomorrow will be, but we can safely assert that neutron powder diffraction will contribute significantly to our understanding of their microscopic properties.

The **Liquids and Amorphous Materials Working Group** predicted that the ESS will provide qualitatively new data on the microscopic properties of the disordered state which will challenge current theoretical thinking:

- Diffraction measurements will expand to four orders of magnitude in Q leading to unprecedented real space resolution in studies of non-crystalline materials. Intensities will allow derivatives of the pair correlation function g(r) with respect to external variables to be determined, which give information on the three-body potential in the system.

- Simultaneous studies of structures over a wide range of spatial scales, which will be of increasing importance in understanding the behaviour -- and hence tailoring the design of -- complex industrial materials.

- The long anticipated field of Neutron Brillouin Scattering (NBS) will be realised, probing the dynamics of liquids and disordered materials in the non-hydrodynamic regime.

- The determination of the vibrational densities of states will become a routine tool for chemists, biologists and material scientists and will contribute to their understanding of the behaviour of materials.

- Increased intensity will widen the range of possible isotopic substitution experiments by including some further dozen elements. This will make an impact on complex biological systems such as the structure and dynamics of water around selective bases in DNA, as well as in solution chemistry.

- The use of polarisation analysis to separate coherent and incoherent scattering.

The **Large Scale Structures Working Group** believed that the field of soft condensed matter will become an increasingly important area of research activity, with significant technological applications. Neutron reflection and Small Angle Neutron Scattering (SANS) are techniques which will play a central role in the wider understanding of many of the outstanding problems in this field. The brightness of ESS will offer unparalleled opportunities for these techniques.

The ability to probe a wide range of spatial scales with high resolution will be important in both SANS and reflection studies on the ESS. In addition to providing a real opportunity for many of the newly emerging fields (such as surfactant adsorbtion) to mature, the ESS will provide a large growth potential for new areas such as the *in situ* study of the electrochemical interface, and adsorption at the liquid-solid and the liquid-liquid interfaces. The enhanced flux will

enable kinetic experiments, allow better spatial resolution, and present real opportunities to study off-specular scattering, grazing incidence diffraction and surface inelastic scattering.

If the ESS were available today it would make a major impact on our understanding of the following areas:

- the structure of adsorbed organic molecules at air-liquid, liquid-liquid and liquid-solid interfaces;
- the nature of the electrochemical interface;
- the structure of pure liquid surfaces;
- interactions and moment distributions in thin magnetic films;
- complex colloidal systems, including liquid crystalline polymers and anisotropic systems;
- kinetic processes such as relaxation during extrusion.
- technologically important materials such as detergents, paints, inks, coatings; composite materials such as polymers and alloys; areas of food technology such as emulsion stability, foams, gels as well as lubricants and fuel additives.

The **Excitations Working Group** warmly embraced the development of new types of spectrometer to study excitations in single crystals (a quintessentilally reactor-based experiment using the ubiquitous triple axis spectrometer) through using an extensive, highly pixellated detector, with the resulting data arrays subsequently analysed using (yet to be developed) powerful computing techniques. They anticipated gain factors in favourable cases of 1000 (30 from instrument x 30 from source) over existing facilities which will lead to *qualitatively* new science. There was also great enthusiasm for the potential scientific impact of extended dynamical range data -- which will provide a complete picture of the excitations in a system rather than the specific view afforded by traditional (three-axes) techniques.

They noted:

- The trend in material science is towards the study of ever more complex materials. Pulsed sources are naturally suited to such studies since the complexity demands a characterisation technique which can cover simultaneously a wide dynamic length and time range in one measurement.
- In addition to incremental progress, the ESS will provide qualitative improvements in our understanding of some of the fundamental problems in physics, such as magnetism and the evolution of metallic behaviour from a Mott-Hubbard insulator.

- There are many examples of *measurements* which can be done with present generation neutron sources but only on time scales which forbid any possibility of systematic parametric studies. On ESS, experiments which with existing facilities would take a week will be feasible in a fraction of a day: this again leads to a qualitative change in the *experiments* - as a function of temperature, concentration, pressure, humidity, magnetic field elapsed time or some other parameter - that may be envisaged with neutrons.

- Developments in computing over the last 20 years have been a prerequisite for the full exploitation of pulsed neutron sources: the instrumentation on the ESS should anticipate this trend continuing.

- In some fields -- e.g. 2d magnetic excitations -- the brightness of the ESS represents a step forward which is comparable to that between using a rotating anode and a synchrotron source for x-ray structural studies.

In addition they highlighted:

- The ability to measure weak (mbarn or μbarn) cross-sections -- which will allow a new range of electronic phenomena to be observed.

- The ability to perform *full* polarisation analysis which will impact on soft condensed matter studies through separation of the coherent and incoherent scattering as well as in magnetism.

- The unexplored fields of the dynamics and kinetics of surfaces and thin films which the ESS would make possible.

- The development of a new field of neutron scattering in the mid-infrared energy range, where a weakly perturbing probe giving Q-dependent information will be invaluable to our understanding of a wide range of phenomena.

The **High Resolution and Molecular Spectroscopy Working Group** regarded the ESS as an opportunity to realise the novel ideas in instrumentation on a high flux next generation neutron source. Europe undoubtedly has an innovative record in instrument design and the ILL and ISIS (the leading reactor and the leading pulsed source) provide a unique springboard from which to consolidate European domination of this branch of science.

Scientific advances in these areas which the group anticipates the ESS will facilitate include :

- The study of the dynamics of transmission of chemical species by biological membranes, such as the diffusion of sodium in nerve tissue and the routes whereby carcinogens penetrate skin.

- The timely application of inelastic neutron scattering to a rich variety of biological problems including the dynamics of proteins and solitons along DNA chains.

- Improvements in resolution which will put neutron spectroscopy on a par with IR and Raman scattering but with all the usual associated advantages of being able to relate intensities to atomic displacements and the lack of selection rules. One example will be the study of the dynamics of industrial catalysis processes *in real industrial environments*, rather than in idealised laboratory conditions.

- Contrast effects equivalent to those achieved by isotopic substitution of H with D will be possible by specific *in situ* spin contrast variation using dynamical nuclear polarisation. Such labelling of the biologically active sites in an enzyme would revolutionise our ability to probe biological activity at the molecular level.

The Working Group proposed a broad scientific programme for the ESS based upon a novel range of state-of-the-art spectrometers, which include:

- A spin echo spectrometer with a resolution of 100 peV.

- A purpose built spectro-diffractometer which *simultaneously* has 1 μeV energy resolution and 2 10^{-4} $\Delta d/d$ resolution.

- A gravity spectrometer with neV resolution.

- A zero-field spin echo machine.

- An inelastic spectrometer with resolutions equivalent to those of light scattering over a range up to 500 meV, but with all the advantages of neutron spectroscopy.

- A direct geometry cold neutron spectrometer with fluxes at the sample which exceed those presently available by a factor of 40.

- Crystal analyser spectrometers with resolutions below 100 neV and with momentum transfer less than 20 $m\text{Å}^{-1}$.

- A polarisation analysis spectrometer with 15 μeV resolution.

- A large-scale long-wavelength neutron interferometer and other fundamental neutron physics instruments.

6 Instrumentation for the European Spallation Source

The Working Groups identified a reference set of some 70 instrument types -- see Table I -- for the ESS. They concluded that the vast majority of areas currently studied by neutron scattering using ILL or ISIS would experience substantial gains of between one and three orders of magnitude. Instruments on the ESS would benefit from the technology developed over the past 40 years on all steady state sources and should build on the experience gained during the last decade on pulsed sources such as IPNS, KENS, LANSCE and ISIS. In areas of instrumentation where pulsed sources have not yet realised their full potential, specific efforts should be made to develop advanced instrument concepts during the design phase of the ESS. The areas specifically identified in this category were:

- Long wavelength instrumentation, particularly SANS, on a 10 Hz Target;
- Structural studies using single crystals;
- Developing a time-of-flight analogue of the 3-axis spectrometer -- the MAPS concept;
- Polarisation techniques appropriate to a pulsed source.

Critique of source specification

The reference source specification of a 5 MW proton beam in 1 μs pulses delivered at 50 Hz and 10 Hz to two separate target stations was not an arbitrary choice, but was based on expert advice (later confirmed by the Accelerator and Target meetings) as to what was technologically feasible. Although the Working Groups recognised that this specification represented a significant advance, nevertheless they wished it to be noted that a higher beam power was desirable as was a greater choice of target types. This should be borne in mind during the design phase, and the option to upgrade at a later stage incorporated, as technological advances are made, if at all possible.

Whilst the two-target 10 and 50 Hz specification was felt to offer a reasonable compromise, it was noted that a significant number of instruments could utilise effectively higher frequencies, if this increased their time-average neutron flux: higher frequencies *per se* at constant beam power is not advantageous.

EUROPEAN SPALLATION SOURCE: INSTRUMENT SUITE

	Moderator	Hz	FP (m)
CRYSTALLOGRAPHY			
High Resolution Powder Diffractometer	M+	10	150
Structural Refinement Diffractometer	P	25-50	50
Special Environment Diffractometer	P	10.-50	60
Small Sample Powder Diffractometer	P	20-50	20
Time Resolved Powder Diffractometer	P	50	12
Long Wavelenghth Powder Diffractometer	DH	10.-50	40
Materials & Engineering Science Diffractometer	P	100	25
Ultra High Resolution Powder Diffractometer	M+	50-100	250
High Resolution Physics Single Crystal Diffractometer	P	100	20
Chemical Chrystallography Diffractometer	M+	50	15
Biological Molecules Single Crystal Diffractometer	CH	50	7
Diffuse Scattering Diffractometer	M-	50	15
LIQUID, AMORPHOUS & DISORDERED SYSTEMS			
Neutron Brillouin Scattering (2 - 30 meV)	DH	20-	10 , 7-12
Neutron Brillouin Scattering (20 - 100 meV)	M+	50	10 , 7-12
Neutron Brillouin Scattering (80 - 1000 meV)	P	100	10 , 7-12
HIgh Resolution Spectrometer (2 - 30 meV)	DH	20-	15 , 4
High Resolution Spectrometer (20 250 meV)	M+	50	15 , 5
High Productivity Spectrometer	M+	100	9 , 3
Special Environment Spectrometer	M+	50	8
High Energy Spectrometer (1 - 20 eV)	P	200	10 , 2-7
HIgh Resolution Diffractometer	M+	20-	25
High Intensity Diffractometer	M+	50	10
Special Environment Diffractometer	M+	50	10
Anomalous Dispersion Diffractometer	P	50	12
Single Pulse Diffractometer	P	?	7
LARGE SCALE STRUCTURES			
Wide Q Range Flasgship SANS	DH	10	20-40
Very Low Q Anisotropic Sans	CH	10	40
Very Low Q Isotropic SANS (Double Crystal)	CH	50	15
High Flux SANS	CH	50	15
Small Sample SANS	DH	10	20-40
High Resolution Reflectometer (Horizontal)	DH	10	15-20
High Resolution Reflectometer (Vertical)	DH	10	15-20
High Intensity Reflectometer (Horizontal)	CH	10	15
High Intensity Reflectometer (Vertical)	CH	10	15
Evanescent Wave Reflectometer	DH	10	15-20
Off-Specular Reflectometer	DH	10	15-20

	Moderator	Hz	FP (m)
EXCITATIONS			
Siingle Crystal Low Energy TOF Spectrometer (2 - 20 meV)	DH	25-50+	10-15 , 4
Siingle Crystal Low Energy TOF Spectrometer (2 - 20 meV) + PA	DH	25-50+	10-15 , 4
Single Crystal Med Energy TOF Spectrometer (20 - 400 meV)	M+	50	10-15 , 4
Single Crystal Med Energy TOF Spectrometer (20 - 400 meV) + PA	M+	50	10-15 , 4
Siingle Crystal High Energy TOF Spectrometer (0.2 - 5 eV)	P	100	10-15 , 4-10
Siingle Crystal High Energy TOF Spectrometer (0.2 - 5eV) + PA	P	100	10-15 , 4-10
Poly Crystal Low Energy TOF Spectrometer (2 - 20 meV)	DH	25-50+	10-15 , 4
Poly Crystal Low Energy TOF Spectrometer (2 - 20 meV) + PA	DH	25-50+	10-15 , 4
Poly Crystal Med Energy TOF Spectrometer (20 - 400 meV)	M+	50	10-15 , 4
Poly Crystal Med Energy TOF Spectrometer (20 - 400 meV) + PA	M+	50	10-15 , 4
Poly Crystal High Energy TOF Spectrometer (0.2 - 5eV)	P	100	10-15 , 4-10
Poly Crystal High Energy TOF Spectrometer (0.2 - 5eV) + PA	P	100	10-15 , 4-10
Low Energy Crystal Analyser Spectrometer	DH	10	7
Medium Energy Crystal Analyser Spectrometer	M+	100	7
Low Energy Triple Axis Spectrometer	CH	50+	7
Medium Energy Triple Axis Spectrometer	C	50+	7
High Magnetic Field Inelastic Spectrometer	M+	50	10-15 , 4
High Pressure Inelastic Spectrometer	M+	50	10-15 , 4
Rotating Analyser Inelastic Spectrometer	P	50	8
Critical Scattering Spectrometer	DH	10	100
Surface Inelastic Spectrometer	DH	10	10
High Resolution Spectroscopy			
MUSICAL - Multi-Monochromator Microvolt Machine	CH	50	10,2
VIRUS - Very High Resolution Quasi-elastic Spectrometer	DH	25-	150,2
NSE*1 - Long Wavelength Spin-Echo Spectrometer	CH	15	12,3
NSE*2 - Short Wavelength Spin-Echo Spectrometer	DH	50	10,2
CASSANDRA - Highest Res'n Crystal Analyser Molecular Spectrometer	DH	15-	50,1
CHINTZ1 - Direct Geometry Spectrometer for Medium Energies	DH	100	10,4
CHINZ2 - Microvolt Resolution Direct Geometry Spectrometer	CH	50	10,6
OPERA - Magnetic Bunching Spectrometer	DH	100	10,10
BUNCHING - Phase Space Transformation Instrument	DH	100	12,3
PYRRHUS - Polarisation Analysis QuasiElastic Spectrometer	DH	50	25,1
IRIS - Good Resolution QuasiElastic Spectrometer	DH	50	40,1
NRSE - Resonance Spin Echo Spectrometer	CH	25	12,8
FOTOF - Time of Flight Focussing Machine	DH	400	10,10
Diffracto-Spectrometer - Simultaneous Diffraction and Spectroscopy	DH	50	20,1
UCN - Ultra-Cold Neutron Gravity Spectrometer	CH	10	10,5
Fundamental Physics Station	CH	10	10
RECOIL - eV Molecular Spectroscopy	M+	100	12,1
High Intensity Crystal Analyser	CH	50	10,1
Interferometer	DH	25	10
CHRYSALIS - Wide Range Molecular Spectrometer	M+	50	25,1

The Working Groups recognise that the instruments identified in Table I and discussed in the Appendices will not necessarily be those eventually deployed on the ESS. They do provide a reference set which gives some indication of the likely requirements for both target frequency and moderator type. Several points are worth noting:

- Firstly the large number (70) of instrument types identified. Some reduction may result from melding the requirements of the individual groups, but some duplication may also be necessary. At least two target stations are required to support this number of instruments.

- Secondly, there was a substantial demand for a liquid methane (or equivalent) moderator and a desire for a low temperature moderator with improved pulse characteristics. Technical difficulties (radiolysis and deposition of solid hydrocarbons) associated with the operation of iiquid methane at 5 MW beam power should be addressed with some urgency in the Target R&D programme.

- The specification of 10 Hz and 50 Hz targets appears to be a good compromise. A significant number of instruments require a less than 50 Hz target and the majority find 50 Hz optimal. Many might benefit from a 100 Hz source if it provided a higher time-average neutron flux. If however, as is likely, heat dissipation in the target dominates, then a 100 Hz target would produce the same time-average neutron flux as a 50 Hz target, but could achieve it with a lower peak proton current in the accelerator, which would have some benefit in the accelerator design. Such a compromise would however disadvantage the majority of instruments which require a 50 Hz or 10 Hz target.

Clearly these findings are relevant to the Accelerator and Target Studies.

Moderator	10 Hz	10 - 50 Hz <<---.	10 - 50 Hz .--->>	50 Hz	>50Hz	TOTAL
Coupled Water	-	-	-	-	1	1
Coupled Hydrogen	5	2	0	6	1	14
Decoupled Hydrogen	9	6	4	4	4	27
Methane or DH	0	0	0	1	0	1
Methane or P Water	1	1	-	13	4	19
Poisoned Water	0	0	3	3	8	14
TOTAL	15	9	7	27	18	75

TABLE II Analysis of instruments by source frequency and moderator type

7 R&D Needs for the European Spallation Source

At the ESS, considerable expense, effort and skill will have gone into the production of protons, their conversion to neutrons and subsequent spectral tailoring. It is equally important that similar effort is applied to their detection and analysis. Before addressing the R&D needs for instrumentation at the ESS in detail, it is worth considering the lessons learnt from the construction and operation of both ILL and ISIS.

- A significant fraction of the construction budget (about 30 %) must be devoted to instrumentation and the operating budget must include provisions for enhancing instrumentation and developing new concepts;
- The instrumentation must be designed and developed in an innovative atmosphere;
- The development of neutron instrumentation and associated equipment requires long lead times. Instruments are sufficiently complex that the true performance is difficult to predict from theoretical design studies. It is therefore vital to develop and test ideas on existing sources.

The Instrumentation & Techniques meeting identified several R&D issues that are likely to impact directly on the success of the ESS. Three outstanding areas are

- detectors
- polarising filters
- data visualisation.

If these areas are not addressed at a very early stage and if significant manpower is not committed to them, it is likely that a number of the spectrometers proposed for the ESS will have performances far below their peak value when the source is commissioned. The participants at the meeting agreed that the expertise necessary to address these R&D topics exists within the European neutron scattering community. They recommended that a programme should be pursued under the Human Capital and Mobility initiative to utilise the expertise which, (partly because of the present situation at the ILL), is currently available in Europe. It was agreed that a separate bid will be made to the CEC for resources to enable this R&D programme to be pursued.

Detectors

The optimal design of the ESS requires that each instrument has a large detector complement. The requirement to provide position-sensitive detectors of dimension 1 - 10 m^2 is in itself a challenge, although possible using current technology. When this is combined with the need

for low cost, very short dead-times (10 - 100 ns), very small spatial and temporal cross-talk, together with the usual requirements of low intrinsic backgrounds and gamma sensitivity, the task takes on another dimension and clearly requires a significant R&D programme. There are several promising approaches that could be used to build better neutron detectors including the use of improved scintillators, the use of boron-containing semiconductors, and ^{3}He micro strip detectors.

^{3}He Polarisers

Another area in which R&D is clearly and urgently needed is in the production and analysis of polarised beams of neutrons. Fully 50% of the spectrometers proposed for ESS intend to utilise polarised neutrons. Since most instruments plan to detect neutrons with a range of wavelengths that are scattered into large detectors, the only appropriate technique for neutron polarisation appears to involve the use of polarised ^{3}He. Although very promising developments have occurred recently in this field (e.g. the group at Mainz has succeeded in polarising ^{3}He at one torr and then increasing the pressure by a factor of ten), much more work will be needed before the technique can be applied widely to neutron scattering.

Visualisation

From high-flux reactor institutes with a total of about 10 detector channels (one per spectrometer) in the early 1970s, neutron scattering has progressed to a few hundred thousand data channels at the present ILL. If the spectrometers imagined at this workshop are built, ESS may have more than 1,000,000 detector channels, each involving between 200 and 2000 time slices. When time-dependent experiments are taken into account, the number of information channels could approach 10^{10}. This proliferation of information poses a challenge to the data acquisition electronics which we believe can be met by the year 2000. It also poses a challenge to the software required to present this vast quantity of information in a form that can be readily assimilated by the experimenter.

The only way the impedance mis-match between detector chain and human can be reduced is by implementing imaginative data visualisation algorithms that allow experimenters to view their data from many points of view and to "zero in" on important regions. Such tools are essential not only for extracting science from data in the long term but also for experimental management. Once again, a development of this sort will take a concerted effort by the community to define and refine its needs, as well as considerable manpower to write and implement appropriate computer algorithms.

Supermirrors

Supermirrors have now been produced to reflect neutrons incident at angles up to about 3 times the critical angle of nickel ($3\gamma_c$) with reasonable efficiency. While this achievement opens exciting possibilities for neutron guides and other optical elements with performances better than those found at ILL, even further gains can probably be achieved with modest R&D funding. It is believed that $5\gamma_c$ supermirrors could probably be made. However, this is likely to take some time because improvements will probably be made in a serial fashion rather than by many parallel studies.

Capillary Guides

It has long been apparent that neutron scattering suffers enormously in comparison with other spectroscopies from the use of plane-wave optical elements rather than focusing lenses or mirrors. The reason for this is the very basis for the success of the neutron as a probe—it interacts weakly with matter (or fields). Nevertheless there are three methods by which neutrons can be focused—large-field hexapoles, Fresnel lenses and curved bundles of capillary guides. Although guides of this sort have been made in Russia, they are not yet efficient enough to be used in an optical element. A modest R&D effort can probably solve this problem or at least indicate the probable limits of the technology.

Generalised Polarisation Analysis

Recent work at the ILL has shown the great importance of the application of generalised neutron polarisation analysis to work on magnetic structures. It is likely that the same conclusion will be reached for inelastic scattering. Development work is required to be able to extend this technique to spectrometers with wide angular coverage.

Phase Space Manipulators

One disadvantage of TOF spectrometers as they are currently designed is that resolution in Q-ε space cannot be modified easily. For example, it is not presently possible to achieve the type of variable reciprocal-space focusing currently used on triple-axis spectrometers at reactors. Focusing in any dimension (reciprocal space, time etc.) is a crucial technique for neutron scattering because it allows researchers to enhance signals in ways that are essential if the inherently low phase-space density of neutrons is to be mitigated. In almost all cases, focusing involves manipulating volumes of phase space that contain neutrons and altering the shapes of these volumes in an appropriate way. Often the extended axis of the phase space volume is made to coincide with a direction in phase space along which there is little variation of a phenomenon that is to be measured. At the same time, a smaller dimension of the phase-space volume is made to coincide with the scan direction, thereby enhancing the measured signal and

improving resolution. More research is needed into devices (e.g. Drabkin resonators, rotating crystals etc.) that can be used to make phase-space transformations. A great deal of this work could be carried out at smaller neutron centres or even in suitable university departments.

Tuneable Filters

For many experiments that involve elastic scattering, it is important to be sure that results are not contaminated by inelastic scattering. Obvious examples are to be found in the fields of small angle scattering and reflectometry, but the concept may be more generally applicable. For ESS, tuneable filters, perhaps making use of a time-ramped Drabkin resonator, will be needed for this purpose.

Pulsed Fields

Many of the exciting scientific horizons opened by the ESS involve the use of severe sample environments such as high magnetic fields, pressures, lasers etc. Time of flight spectrometers offer advantages in these areas of research because they provide more convenient access to samples contained within cryostats, pressure cells, and other sample environment devices. Nevertheless, extreme environments can often only be applied for short periods of time and in a pulsed manner. Such environments are often well-suited for use at a pulsed neutron source such as the ESS and should be developed further.

Other R&D Needs

There are a number of other areas of R&D that should be addressed before construction of the ESS is initiated and can be carried out at existing sources. For example, ISIS should promote the construction of those new spectrometers that increase the diversity of scientific research carried out at pulsed spallation sources, thereby increasing the base of experience which will be used to identify spectrometers for the ESS. They should encourage the evaluation of direct time-of-flight methods for obtaining data of the type that is conventionally measured with triple axis spectrometers at reactors. The community that uses such spectrometers must be assured, before ESS is built, that the science they do can be fully supported by the ESS.

Other important areas of R&D that will involve single investigators at various centres include: studies of spectrometer optimisation based on proper evaluations of the information content of the data that can be obtained; work on tuning the mosaic of crystals by ultrasonic excitation; the construction of an ultra-high temperature furnace; the investigation of devices that can efficiently flip the spins of neutrons over a wide range of wavelengths; and development of the zero-field spin echo technique.

8 Conclusion

The Expert Meeting recognised the major advantages that follow from the time structure of a pulsed neutron source. Although these are being explored at existing sources, further significant advances are anticipated as the technique matures. Building on this experience, the clear scientific potential of a next generation source was evident. Although a source of the specified power - 30 times ISIS, at present the most intense such source - would give a neutron source of the same time averaged flux of the best available reactor source (ILL at Grenoble), the expert meeting recognised that the exploitation of the time structure would result in gains in effective fluxes of up to two or more orders of magnitude.

The Working Groups confidently predicted that the enhanced temporal brightness of the ESS would allow the source to make both a major impact on established fields and substantial contributions to new areas of research. Not only would the flux increase be easily exploited in the most obvious ways (smaller samples, short time kinetic experiments), but would also allow even higher spatial and temporal resolution, coupled with the wider dynamic range in energy transfer (and hence time), and momentum transfer (and hence space) which are needed to probe the more complex systems that are increasingly central to, for example, new materials development and macromolecular systems.

The expert meeting strongly supported the concept of a next generation spallation source for the future European neutron facility, and it drew up the specifications for instrument suites on the two targets that were required to exploit fully the source potential. A number of R&D issues were raised, e.g. detectors, polarisation and data visualisation, on which further collaborative work was recommended on the international level. Those present proposed that mechanisms be found to assist this collaborative R&D in the near future.

Neutron scattering is an important probe of condensed matter. Its impact is constrained by the intensity of present day sources. An accelerator-based pulsed neutron source, the European Spallation Source promises a qualitative advance in this field. It appears to be technically feasible and environmentally acceptable. Predicting the future is always fraught with difficulties, but whilst all the scientific dreams aired may not be realised, this Expert Meeting has identified a vast amount of research which could be achieved with the ESS.

Expert Meeting on Instruments & Techniques for the European Spallation Source

Monday 24 February 1992

Agenda

12:30	Lunch	
14:00	Plenary Session	Chair -- T Springer
	Welcome	P R Williams
	Mission Statement	G Lander
	Discussion	
15:00	Plenary Session	
	Summary of Accelerator Expert Meeting	S Martin
	Summary of Target & Moderator Technology Workshop	G Bauer
	Source Characteristics & Performance	J M Carpenter
16:00	Tea	
16:30	Plenary Session	
	Formation of Science & Instrumentation Working Groups	
	Parallel Session	
	First Meeting of Working Groups	
19:00	Dinner	
20:30	Parallel Session	
	Second Meeting of Working Groups	
21:30	End of Parallel Session	

Expert Meeting on Instruments & Techniques for the European Spallation Source

Tuesday 25 February

Agenda

09:00 Plenary Session — Chair -- J Axe

Scientific Horizons & Technical Briefings I : a series of short talks and discussion:

J W White, G Aeppli, P Timmins, R Pynn, M W Johnson

10:30 Coffee

11:00 Parallel Session

Third Meeting of Working Groups

12:30 Lunch

14:00 Parallel Session

Fourth Meeting of Working Groups

15:30 Tea

16:00 Plenary Session — Chair -- R Pynn

Scientific Horizons & Technical Briefings II : a series of short talks and discussion:

W I F David, F Mezei, J Enderby, J B Forsyth, R Golub, S F J Cox

17:30 End of Plenary Session

19:00 Dinner

20:30 Plenary Session — Chair -- P A Egelstaff

Interim Reports from Co-ordinators

Discussion

22:00 End of Plenary Session

Expert Meeting on Instruments & Techniques for the European Spallation Source

Wednesday 26 February

Agenda

09:00 Plenary Session — Chair -- J W White

Scientific Horizons & Technical Briefings III : a series of short talks and discussion:

J Axe, R K Thomas, B D Rainford, J Higgins, M Steiner, J B Hayter

10:30 Coffee

11:00 Parallel Session

Fifth Meeting of Working Groups

12:30 Lunch

14:00 Parallel Session

Sixth Meeting of Working Groups

15:30 Tea

16:00 Parallel Session

Preparation of Reports

17:30 End of Parallel Session

19:30 Sherry

20:00 Dinner

Expert Meeting on Instruments & Techniques for the European Spallation Source

Thursday 27 February

Agenda

09:00	Plenary Session	Chair -- G H Lander
	Final Reports from Co-ordinators	
	Discussion	
11:00	Coffee	
11:30	Plenary Session	Chair -- J L Finney
	Summary of R & D Requests -	R Pynn
	Discussion	
	Meeting Summary	P A Egelstaff
12:30	Lunch	
14:00	End of Expert Meeting	

Participants at the Expert Meeting on Instrumentation & Techniques for the European Spallation Source

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J D	Axe	BNL, USA
G	Bauer	PSI, Switzerland
F J	Bermejo	Madrid, Spain
C J	Carlile	RAL, UK
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H	Conrad	KFA Julich, Germany
R A	Cowley	Oxford, UK
S F J	Cox	RAL, UK
U	Dahlborg	Stockholm, Sweden
W I F	David	RAL,UK
P	Day	RI, UK
A	Deriu	Parma, Italy
P A	Egelstaff	Guelph, Canada
J E	Enderby	Bristol, UK
B	Farnoux	LLB, France
J L	Finney	RAL, UK
J B	Forsyth	RAL, UK
M	Furusaka	KEK, Japan
I S K	Gardner	RAL, UK
R	Golub	Munich, Germany
L de	Graaf	Delft, Netherlands
J B	Hayter	ORNL, USA
G	Heger	Saclay, France
A	Heidemann	ILL, France
A W	Hewat	ILL,France
J S	Higgins	Imperial, UK
J A K	Howard	Durham, UK
W S	Howells	RAL, UK
M W	Johnson	RAL, UK
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R M	Richardson	Bristol, UK
D K	Ross	Salford, UK
F	Sacchetti	Rome, Italy
R	Scherm	Braunschweg, Germany
A J	Schultz	ANL, USA
D S	Sivia	RAL, UK
A K	Soper	RAL, UK
T	Springer	KFA, Germany
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T G	Walker	RAL, UK
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Scientific Horizons for the European Spallation Source

Scientific Horizons for the European Spallation Source

Introduction

A number of prominent neutron scattering scientists were asked, as a stimulus to subsequent discussion within the Working Groups, to present short accounts of their vision of the major scientific challenges which would likely face the world's premier neutron source at the beginning of the next millennium. Summaries of these talks and of the important points raised in discussion are presented here. Part of these sessions also addressed the technical and instrumental challenges which this source will face.

Session : Scientific Horizons I *Chairman J D Axe*

Scientific Horizons in Physical Chemistry

J W White, ANU, Canberra, Australia

The purpose of this session is to look to the middle and distant horizons of the science that might be done with the world's best neutron scattering facilities in the years from say 2010 onwards. The source brightness and moderator characteristics from the previous two Expert Meetings set the datum. I have been asked to be, and I will attempt to be, imaginative and provocative in three areas of interest.

Of the powerful characteristics of neutron scattering, those most unique and complementary to the likely developments at third generation X-ray synchrotrons are: (a) coherent and incoherent high resolution spectroscopy at low energies, and (b) isotopic and spin polarisation contrast variation, and (c) the use of the strong, pulsed, high energy neutron flux for measurement of molecular and atomic electronic excitations and related effects. Each is discussed briefly below.

High Resolution Spectroscopy

High resolution spectroscopy of adsorbed species at low energy transfers and an energy resolution of microelectron volts has revealed isolated and collective rotational tunnelling, surface diffusion at atomic resolution, adatom vibrations and characteristics of the incommensurate-commensurate transitions prevalent at all interfaces. The scope of this field is wide, and exploration by neutron scattering only limited (for example, in the extent to which

coherent scattering can be used) by intensity and technical aspects (e.g. the eventual need for polarisation analysis). An extremely useful feature of pulsed source instruments is the ability to measure structure using very high resolution diffraction (at say $\Delta d/d=10^{-3}$) simultaneously with dynamical studies using spectroscopy. Recent work on the domain structure of intercalation compounds illustrates this (1).

Figure 1 shows the rotational tunnelling spectrum of adsorbed hydrogen molecules in second stage caesium graphite alongside the c-axis diffraction near the (003) reflection. The smooth change in diffraction with hydrogen content indicates that the strong changes in tunnelling result from a two dimensional phase change in the adsorbed-layer.

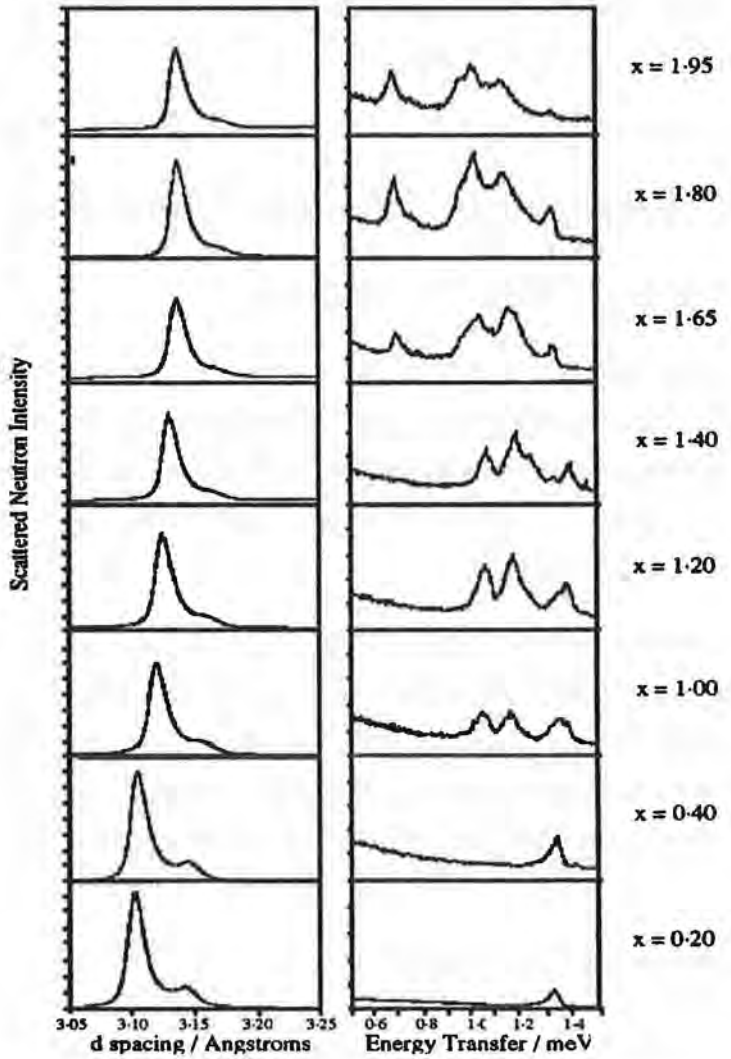

Figure 1. IRIS Neutron diffraction (a) and high resolution neutron inelastic scattering (b) from $C_{28}Cs(H_2)_x$.

Isotopic and Spin Contrast Variation

In the exploitation of small angle scattering, reflectometry and molecular spectroscopy for surface, polymer, sol-gel, biological and metallurgical problems, contrast variation by isotope substitution is vital to enable us to obtain detailed information. Contrast may also be varied by static or dynamic nuclear polarisation (2,3) and by using polarised neutron beams. Developments in magnetic resonance techniques, (with fields of 12T now routinely used), and lasers (e.g. for pumping the ESR or optical transitions) make this method potentially very attractive. It has been shown that flip ratio data can be used for single crystal structure refinement (4), and so the possibility of selective depolarisation (2) of e.g. proton pairs (identified from the very sharp line solid state NMR spectrum that arises at high nuclear polarisation) should be explored for both crystals and less ordered scatterers such as biological materials. It is possible that, by using dynamic polarisation at high magnetic fields, experiments might be done at temperatures approaching ambient.

Electronic Spectroscopy

Elegant examples of the use of high energy neutrons to observe crystal field transitions have come from current work at ISIS and IPNS. By means of the electron-phonon interaction and high energy neutron scattering at low angles (to control the momentum transfer), it should also be possible to access, for example, molecular and atomic electronic excitations (5). The cross-sections are very small (6) (10^{-4} barns) and would require the intensities expected from the ESS.

A possible scheme for observing these weak effects both as densities of states and to measure the exciton dispersion surfaces would be to detect $\Delta k=0$ optical fluorescence from exciton states stimulated away from the zone centre in coincidence with the scattered neutron (5). An analysis of the internal conversion to exciton k=0 and subsequent k=0 to k=0 transitions versus the in-zone $\Delta k=0$ fluorescence yield would also be interesting. Though such experiments can also be contemplated at third generation synchrotrons, neutrons have a unique aspect in that longitudinal as well as transverse excitations are accessible in principle.

References

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2. An asymmetric split-pair superconducting magnet for nuclear polarisation experiments, E Gilbert, P Hanley, J B Hayter and J W White, J Phys E 6 714-718 (1973)

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4. Precise location of hydrogen atoms in complicated structures by diffraction of polarised neutrons from dynamically polarised nuclei, S Cox, J B Hayter, G T Jenkin, M Leslie, G Warner and J W White, Phil Trans R Soc Lond B 290 497-503 (1980)

5. Advances in neutron scattering spectroscopy (plenary lecture to the Deutsches Physikalisches Gesellschaft, October 1975), J W White, Phys Rev B 12 633-650 (1976)

6. P Schofield (private communication)

Scientific Horizons in Solid State Physics

G Aeppli, A T & T Bell Laboratories, USA

When we contemplate expending 0.5×10^9 ecus of taxpayers money, it is important to consider what the project will return to its investors, presumably European taxpayers. More specifically, the project should be related to technological goals understood by politicians and business people. Examples of such goals include a room temperature superconductor, electrical batteries with as much stored energy per unit volume and mass as gasoline, and treatments for various diseases currently deemed incurable. The physicists, chemists and biologists here can and should be able to make honest arguments that the ESS will further progress towards goals of this nature, not only to enable funding of the ESS, but also because it is the achievement of such goals, rather than incremental progress in the small subfields we are currently engaged in, which motivated most of us to choose scientific careers. This does not mean that pure science, in the sense of experiments whose sole purpose is to satisfy our curiosity on a particular topic, has no place at the ESS. However, such science should have intellectual vigour apparent to others.

Apart from its contribution to the general scientific culture and technology, an expensive new project must represent a qualitative improvement over existing facilities. I give here an example of how the ESS will yield such an improvement in my own field, namely the physics of strongly interacting electrons, an understanding of which may eventually lead to more efficient searches for new magnets and superconductors. Arguably the most important problem in this field is to understand how a metal evolves from a Mott-Hubbard insulator. The latter is an antiferromagnet, and an important characteristic of the metal is the nature of the magnetic fluctuations which replace the order in the insulator. Neutron scattering probes the wavevector (Q)- and frequency (ω)-dependent fluctuations directly, so that an intensity map in Q-ω space completely represents the geometry and dynamics of the magnetism in a particular compound. Figure 2 shows such a map obtained for $La_{2-x}Sr_xCuO_4$ with x=0.075, a sample just beyond the insulator-metal transition in the alloy series which for x=0.15 yielded one of the first high-T_c (=38K) superconductors. The frequency is fixed such that $h\omega$ = 1 meV, while the x and y coordinates represent points in the two-dimensional reciprocal space for the CuO_2 planes,

which are the fundamental building blocks shared by this compound with all high temperature superconductors. The important feature of the data is that the magnetic scattering, which for the insulator is sharply peaked at (π,π), is redistributed to form a square with a centre at (π,π) and intensity peaks at the corners. The square grows with x, and represents a unique measure of the Fermi surface induced by doping into the metallic phase.

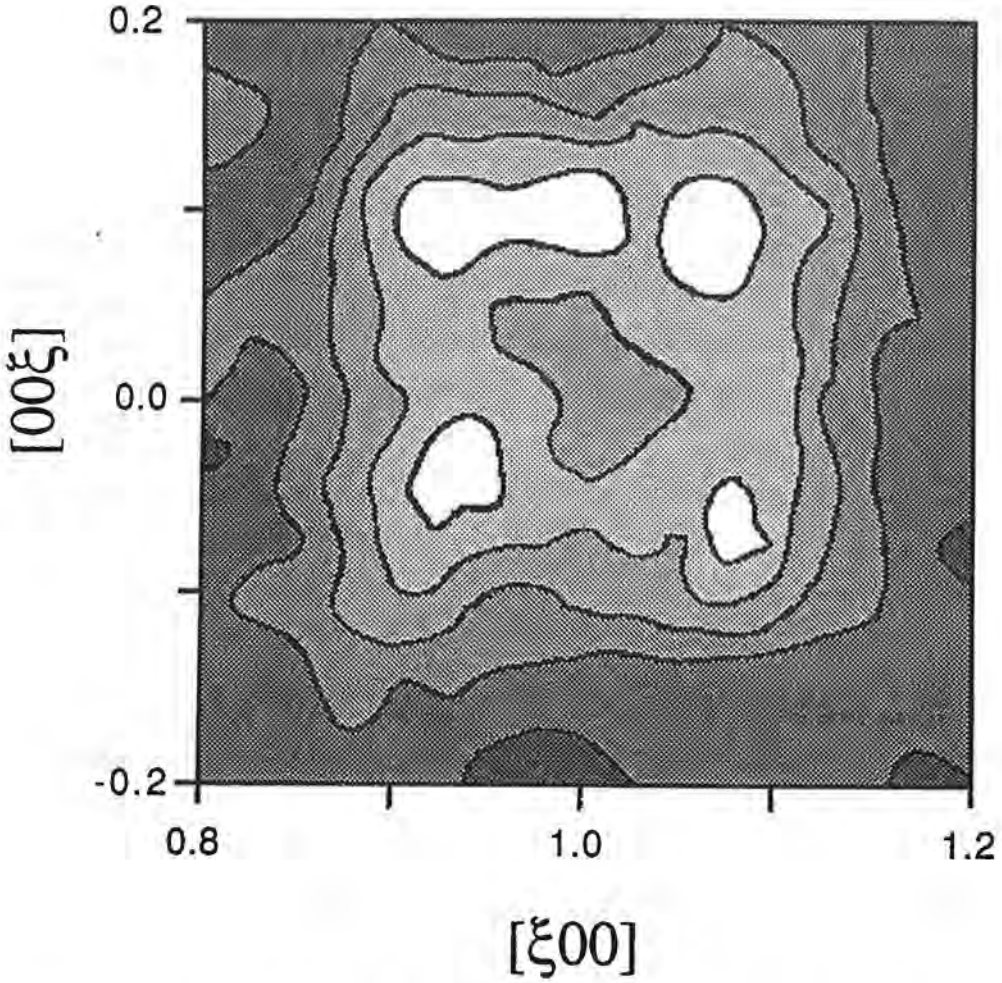

Figure 2 The Q,ω map for $La_{2-x}Sr_xCuO_4$ (x=0.075) at ω = 1 meV

While Figure 2 is informative, the experiment to obtain it was difficult. A modern three-axis spectrometer at the cold source of the Risø medium flux reactor visited each individual point on the two dimensional grid for 20 minutes to accumulate intensities. Furthermore, considerable effort was expended on discovering and eliminating spurious processes due to the rather poor rejection rates (noise/signal ~ 10^{-3}) of monochromators and analysers. Thus, the net measuring time was roughly a week. Because of the two-dimensionality of the sample, and the fact that the time-averaged incident flux will be larger than at Risø, using a position sensitive detector on a direct geometry chopper machine such as the MAPS type of instrument at the ESS would yield the *entire* pattern in Figure 2 in less than 20 minutes. Furthermore, in the same

period, analogous patterns at all other frequencies (consistent with kinematic restrictions) will be obtained. In other words, we would be accumulating useful data at a rate approximately three orders of magnitude larger than with today's reactor-based three-axis technology, which is considered ideally matched to problems such as identifying the magnetic fluctuation in single crystals of $La_{2-x}Sr_xCuO_4$. This represents a *qualitative* improvement in our ability to do science which is akin to that on going from a rotating anode X-ray source to the a synchrotron source.

Many real systems are not low-dimensional like the case discussed above, so it is not as easy to visualise how useful information can be extracted from a time-of-flight multidetector experiment as it is for $La_{2-x}Sr_xCuO_4$. However, in principle, it is possible to obtain arbitrary cuts through Q-ω space from a properly planned experiment on a MAPS-type instrument. The key problem is to develop software which allows scientists to explore Q-ω space during the experiment. Since this is a four-dimensional space, schemes for data display coming from fields ranging from high energy physics to aerospace ("virtual reality") should be imported and combined into a user-friendly package.

In summary, putting modern accelerator, spectrometer and computer technology together should increase the efficiency of many interesting experiments in solid state physics by two to three orders of magnitude. Since the last high flux sources were constructed, computing has become cheaper than shielding and mechanical devices used in spectrometers. Because of its pulsed nature, the ESS takes full advantage of this development, which will lead to opportunities in software engineering as large as those in accelerator and spectrometer design.

Scientific Horizons in Structural Biology

P Timmins, Institut Laue Langevin, Grenoble, France

The object of this talk is to try and identify some of the fields within modern biology which could benefit from a new high flux spallation source. This is of course an impossible task and we can only begin to guess what role neutrons might play by looking at the past and the present. It is first of all important to recognise the multidisciplinary nature of molecular biology and to realise that no single technique will provide the much searched for "cure for cancer" or "cure for AIDS". It is also important to realise the specificity of molecular biology and the relationship between structure and function. The ultimate goal of structural molecular biology is to understand function, not the determination of structure *per se*. An important adjunct whose importance has only more recently been recognised is the role of dynamics in the understanding of function.

High Resolution Structure

Here we refer to structures at a resolution of 3Å or better where, with stereo-chemical restraints, it is possible to obtain models at atomic resolution and therefore to elucidate details of intermolecular interactions, enzyme mechanisms, etc. It is clear that the overwhelmingly successful technique in this area has been X-ray crystallography, with over 400 structures now solved. NMR methods are progressing to the state where, for proteins of less than ~ 20,000 molecular weight, a solution structure may now be obtained by this method. Neutron diffraction, due mainly to the difference in scattering length between hydrogen and deuterium, can provide important complementary information. This may be about the protonation state of a particular group, the water structure or the orientation of for example an amide group. The use of hydrogen-deuterium exchange is also a very important method of probing the dynamics of a structure through proton accessibility. Such studies have been carried out to date on a very small number of proteins. Some others have been tried and failed, and many others have not even been tried. We should therefore ask the question why. The principal problem is a lack of flux and poor signal to noise on existing sources. In order to solve a significant number of protein structures by neutron crystallography it must be possible to work with :

- Crystals of volume < 1 mm^3;
- Unit cells up to 50x50x50$Å^3$.

The particular case of fibre diffraction, where larger samples are sometimes available, is also one where better structural resolution could probably be obtained with higher flux.

In view of the flux limitations of even a next generation spallation source, this implies the use of *Laue or quasi-Laue techniques.*

Low Resolution Structure

Here we are referring to real space resolutions of 3-50Å and the study of crystals and partially ordered systems.

There are a significant number of large macromolecular assemblies which, although essentially crystalline, are in part disordered. An X-ray crystallographic experiment on such systems will solve the structure of the ordered part of the assembly, but usually give no information about the disordered part. This is due to the fact that:

- Disordered protein, nucleic acid or lipid has very poor contrast *vis à vis* water;
- Any contribution to the scattering by the disordered material is concentrated in the very low resolution data, which are difficult to measure precisely with X-rays.

With neutron diffraction, the contrast can be varied (increased) easily, making the disordered component visible against the aqueous background. Moreover, with long wavelength neutrons, the low resolution data can be easily measured. This technique was developed at ILL and a dedicated diffractometer built for this purpose in collaboration with the EMBL.

Crystals of viruses, nucleosomes, membrane proteins, lipoproteins and several other complexes have already been investigated, and several others are awaiting adequate neutron scattering facilities. Due to the very small sample volumes available, such experiments will require the *highest possible flux of cold(ish) neutrons.*

Solutions

Traditionally, neutron small angle scattering has been used to study systems similar to those considered above but in solution. It is clear that at the same resolution crystallography provides more information than small-angle scattering, although many interesting systems cannot be crystallised, and a demand for such information will always be present. The unique power of small angle scattering, however, is the ability to investigate properties which are specific to the solution phase.

In this latter category examples are :

- macromolecular interactions;
- macromolecular assembly/disassembly;
- effects of temperature/pressure;
- effects of pH, ionic strength.

There are currently very few instruments in the world where such experiments can be routinely carried out, principally because they require a *very high flux of cold neutrons.*

Dynamics

The use of neutron inelastic and quasielastic scattering to investigate dynamics is a well developed technique both in solid state and in soft matter physics. Until the last few years, there has been virtually no successful applications of the technique to biological systems due in part to the lack of theoretical understanding of the dynamic behaviour of biological macromolecules. There have now been investigations of a few soluble proteins and of one membrane protein from which general rules are beginning to be elucidated.

For the future, the most exciting prospect would be to look at the dynamics of a specific area of a macromolecule. This would be even more exciting if it could be measured for different functional states of the molecule. We can imagine for example monitoring the dynamics of an RNA polymerase molecule as it makes its way along the DNA strand. Such a reaction can be

controlled very precisely in vitro, and intermediate states isolated. Such experiments are still in the realm of dreams and would require progress in neutron techniques to provide very high fluxes. They would however require very major advances in biochemistry in order to produce specific local deuterium labelling.

Conclusion

As we all know, long term prediction in science is impossible, which is why we all keep trying. If we can design instruments which increase the effective flux on the sample, it is highly probable that modern molecular biology, which has transformed the world in the last 15 years, will provide new techniques to allow us to manipulate biological macromolecules and macromolecular complexes in such a way as to enable the particular advantages of the neutron probe to be exploited in improving our understanding of biologically relevant processes

Parallel Spectroscopy, Data Visualisation and Analysis

R Pynn, LANSCE, Los Alamos, USA

ESS represents a large extrapolation beyond the capabilities of our current neutron scattering facilities. In particular, the deployment of tens of square metres of multidetectors with centimetre resolution, coupled with the use of time-of-flight techniques, will provide a facility with about 10^9 parallel information channels. This formidable quantity of data presents both a challenge and an opportunity. The challenge is to manage effectively an experiment which produces so much data. To meet the challenge requires that effective use be made of modern techniques for visualisation of data and image processing. And therein lies the opportunity. Computer and electronic techniques have advanced by many orders of magnitude since the first time-of-flight neutron spectrometers were abandoned in the sixties in favour of spectrometers that are easier to manage. Unfortunately, the latter, typified by the triple-axis spectrometer, sacrifice parallel data collection in favour of a slow serial process in order to make experiments more manageable. Such a methodology can be dangerous: to understand the reason, one only has to reflect on the fact that incommensurate structures were not discovered as a ubiquitous phenomenon until the seventies - by abandoning film for discrete counters. X-ray scatterers were led to ignore information that did not contribute to Bragg diffraction from one or other known space groups. It never occurred to them that their classification of space groups could be incomplete!

The complex materials that are of interest today are often typified by interactions that occur on widely different length and time scales. Such complexity mandates a characterisation technique that can cover a wide dynamic range of length and time simultaneously. Spallation neutron

sources that make use of time-of-flight techniques provide a tool with the sort of dynamic range that is required.

It is clear that many of the outstanding successes of neutron scattering during the past two or three decades have come from triple axis spectrometers. And indeed, when the region of Q-ε space of interest is known to be small (can one ever really *know* this?), the technique works very well. For this reason, it is essential that instrumentation be developed at pulsed sources to mimic the type of experiment done so well by triple axis machines, albeit as a subset of their total performance. This will require a considerable software effort, but is well within today's technology.

Resolution and Information

We have all heard, at one time or another, that the triangular resolution function of IN5 at the ILL offers advantages in the separation of elastic and quasielastic scattering. It is easy to understand why when one looks at a picture of typical data - a triangle sitting atop a broad Lorentzian. The two components can be easily separated because there is a clear change of slope between them. This observation can be quantified. One may ask the question "what resolution function should be used to get the most information out of scattering data?" It turns out that the answer depends on what information one wants to obtain. For example, to know the position of a peak in a spectrum, the resolution function should be made as narrow as possible. Conflicting requirements of this sort occur only when one knows exactly what question one wants to ask of the data. In general, one cannot define such a precise question for a given spectrometer - it will not always be used to measure peak positions rather than intensities, for example. If the "information question" is asked in a more general way - "how can one obtain the best estimate of $S(Q,\varepsilon)$ given that we don't know its form before the experiment" - Bayesian analysis can provide an answer. In particular, Sivia [1] has developed a figure of merit that measures "information content" and has shown that it depends on the Fourier transform of the resolution function as well as the level of detail sought from the data. The important ramification of Sivia's work is that we should no longer attempt to suppress asymmetric resolution functions - indeed, we should probably favour them. Such resolution functions occur naturally at a pulsed source because the time distribution of neutrons leaving a moderator is asymmetric. This should be exploited in future generations of instrumentation.

Another consequence of Sivia's work on figures of merit is that it allows us to estimate the performance of spectrometers that are transferred to ESS from CW sources. In particular, a triple axis spectrometer could be transferred and have its detector time-gated so that only the signal from the anticipated energy transfer is registered. Such an arrangement would eliminate many of the spurious higher order processes that plague the lives of triple axis spectroscopists. In addition, it would also help with experiments that are currently limited by backgrounds that

are not time-correlated (e.g. cosmic rays, electronic noise etc). For example, an experiment with a signal-to-noise ratio of 0.1 at the ILL would have a figure of merit that is seven times better at ESS than at ILL, even though ESS has the same average flux as ILL. This sort of argument shows that it is not completely stupid to transfer CW technology to ESS. My own preference would be to take advantage of the opportunities offered by the time structure of the ESS to extend dynamic range and obtain parallel data inputs. But the security blanket is there for those who want to use it.

Reference

1. D S Sivia, R N Silver, R Pynn, "The Baysian Approach to Optimal Instrument Design" in 'Neutron Scattering Data Analysis 1990', ed. M W Johnson, IOP Conf Seriers 107, 45-55.

Detectors, Electronics and Computing

M W Johnson, ISIS Facility, RAL, Chilton, UK

The Detector Chain

This talk is about the technical problems that might be encountered in the detector chain for instruments on the European Spallation Source. To place this problem in context, we should first look at the growth of intensity of pulsed sources. **Figure 3,** first drawn by Bob Bruger in 1968 to illustrate the growth and saturation of laboratory and reactor sources, has been augmented by Jack Carpenter and Bill Yellon to show the performance of pulsed sources. The ESS has been added with a completion date of 2001. This diagram is not intended to reflect accurately the relative performances of the two source types (they have been simply compared by plotting either the mean flux (reactors) or the peak flux (pulsed sources)), but to illustrate the recent growth of pulsed sources. In fact, the line joining ZING-P and the ESS in Figure 1 represents a growth factor of 1.7 per year - a quite remarkable performance.

By contrast, the technology of neutron detectors has remained relatively static for the last 20 years. This talk will therefore address the following three points :

- What are the components of the detector chain that play an important role in instrument design?
- Are there problems?
- Are there solutions?

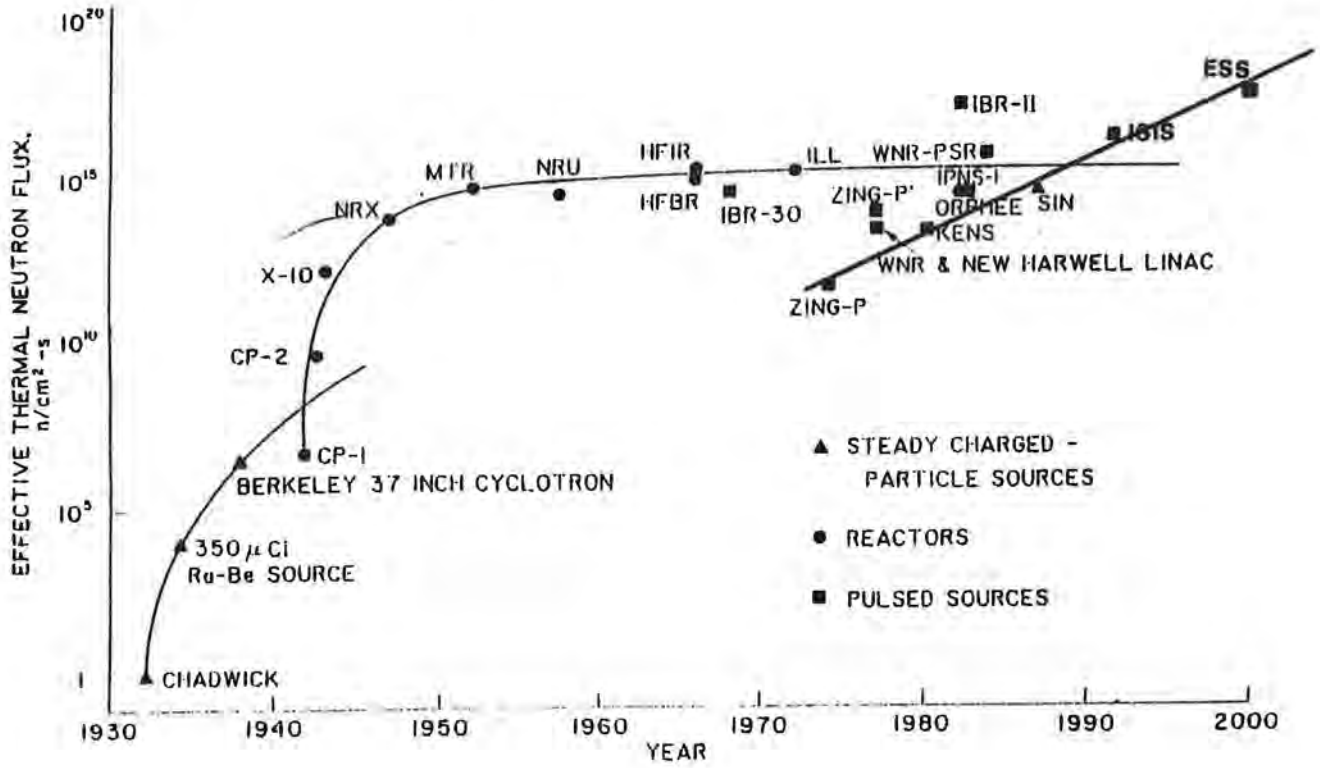

Figure 3 The growth in steady state and pulsed neutron sources in the past 50 years.

Figure 4 illustrates the principal components of the detector chain. The first component is the detector itself with its associated analogue electronics. The important characteristic of this part of the chain in the dead-time (Δt), the time within which two events cannot be resolved. The required dead time is, of course, dictated by the highest instantaneous rate of neutron arrival (designated r_1) and the acceptable dead time correction (usually a few %) given by :

$$f = \frac{r.\Delta t}{(1 + r.\Delta t)}$$

For pulsed sources, the neutron's detection must be 'time stamped' and the resulting descriptor containing position, time and possibly phase (for time-varying samples) information stored digitally. The number of detectors (n_d), number of channels (n_e) and number of phases (n_p), together with the mean count rate for the instrument (m), clearly define the bandwidth required of the recording electronics, together with the size of the histogram memory, and must be considered in instrument design.

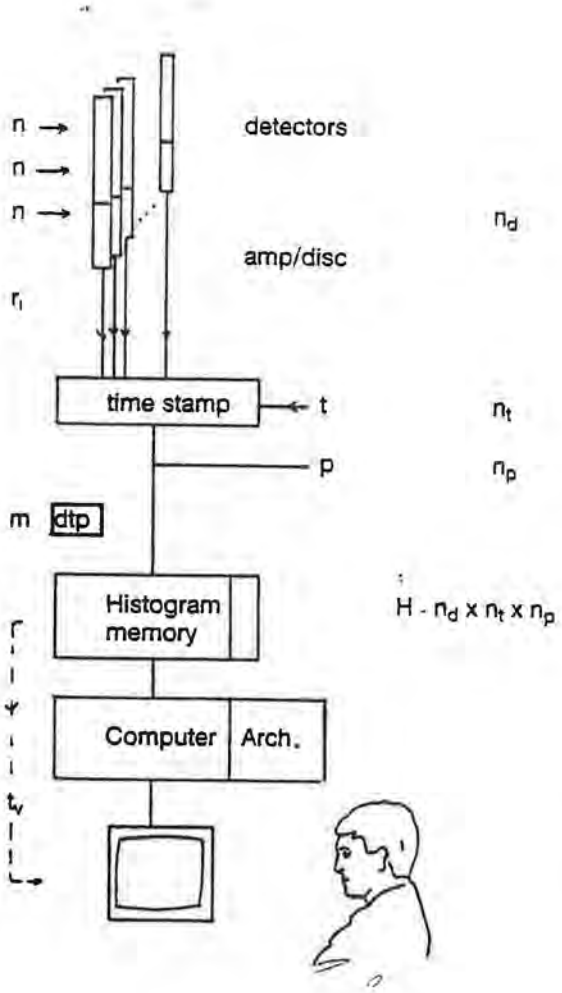

Figure 4 A schematic representation of the data chain.

Some computing element is required in this chain to turn the unintelligible raw data into information suitable for experiment assessment and publication. An important element in the chain is thus the experimenter, and a useful method of characterising the requirements of the computers needed is to measure the time taken to visualise the data, and ensure this is kept to a reasonably low value (t_v < 20s ideally, < 60s to be workable). Although the computing element is shown as a separate entity in Figure 4, it is very likely that to achieve the required t_v time a number of parallel computing components must be incorporated within the histogram memory. Examples of histogram memory incorporating transputer CPUs are already being tested within the current ISIS data acquisition electronics.

In the discussion which follows, I will look at the 'sharp pulse' instrumentation that might be employed at the ESS. This is because such instruments will probably have the highest instantaneous count rate, and the largest histogram memories, and will therefore pose the

hardest problems to solve. This approach also allows us to do some simple scaling from existing ISIS instruments and thus identify exactly where the problems might lie.

Table 1

	r_1 n.ms^{-1}	m n.ms^{-1}	H mbyte	t_v s
TFXA	0	3	0.5	20
IRIS	0.4	1	4	20
MARI	0.4	10	16	60
POLARIS	10	200	2	60
SANDALS	300	1000	24	2000
SXD	60	4	16	200
LIMITS (5% DTC)	**50**	**1000**	**16 x N**	**10-100**
'ESS TFXA'	3	60	0.5	20
'ESS IRIS'	12	30	4	20
'ESS MARI'	12	300	16	60
'ESS POLARIS'	300	6000	2	60
'ESS SANDALS'	9000	30000	24	2000
'ESS SXD'	1800	120	16	200
'ESS POLARIS' + Ω	300	30000	60	2000
ESS POL'+Ω+Strobe	300	30000	6000	200,000

The top half of Table 1 lists six of the existing ISIS instruments, together with four key parameters in defining their data acquisition chain. As expected, the three inelastic instruments (TFXA, IRIS and MARI) have relatively low instantaneous count rates, while the elastic instruments (POLARIS, SANDALS and SXD) have count rates (per detector) ranging from 0.01 to 0.3 MHz. (The rates for SANDALS are higher because the figure refers to the rate for each SANDALS *module*, equivalent to approximately 20 ^{3}He detectors). The mean rate (m) and histogram size is also shown, together with the time-to-visualisation (t_v) for the present ISIS instruments.

The current limits for these data chain parameters are shown in the central row. The limit of 0.05 MHz for r_1 stems from the current dead-time (Δt at 1 μs) for scintillator detectors, and is clearly barely adequate for some existing ISIS instruments. The mean count rates are less of a problem, but histogram memory sizes for the most demanding ISIS instruments already require

the use of multiple MULTIBUS crates. The time to visualisation is also beyond an acceptable range in some cases.

The lower half of Table 1 shows the simple consequence of increasing the ISIS flux on these instruments by a factor of 30, a reasonable approximation to 'transplanting' them to the new ESS facility.

The consequences for the inelastic instruments would seem to be quite tolerable. The instantaneous count rates, r_1, stay within the present limit, as do the other parameters shown. The elastic instruments are more problematic, and the shaded area at the bottom of Table 1 highlights those parameters which would exceed present limits.

Present ISIS instruments are, however, not fully equipped with detectors, and to complete the picture we should at least consider the effect on one instrument of providing a detector complement commensurate with the cost of the neutron beam line. This is shown on the penultimate row in Table 1 as 'ESS POLARIS + Ω'. An instrument such as this, quite capable of taking entire diffraction patterns in one second, will undoubtedly be used for time-dependent studies. The consequences of this are shown in the final row of Table 1. The histogramming memory requirements have now increased to 6 Gbyte and the time-to-visualisation has lengthened between 2000 and 20000 times the acceptable value.

The consequences that follow from the examples given in Table 1 are straightforward. We will require:

- detectors - both individual and area PSDs - with dead times in the range 10-50 ns. In fact, the requirements for single crystal instrument detectors will be the most demanding, and will require dead times towards the bottom end of this range. (Combined, of course, with the existing requirements for neutron efficiency, γ-ray inefficiency (of order 10^{-6}), quiet counts (of the order of one count/hr), uniformity of spatial response and long term stability).

- a new generation of data acquisition electronics capable of handling mean count rates of 30 M events/s, and providing histogramming memories of up to 6 Gbytes in size.

- additional computing power, to reduce the potential bottle neck of visualisation time, equivalent to 2000 to 20000 times the speed of current processors used in instrumental analysis.

Of these three requirements, the first is clearly the most difficult. Existing neutron detectors, based either on ^{3}He proportional counters or Li loaded ZnS scintillators, have dead times in the range 1-5 μs. Microstrip ^{3}He designs would appear to offer the possibility of lowering the

dead time to ~ 100 ns , while organic scintillators (with $\Delta t \sim 1$ ns) could, in principle, be developed for neutron detection if suitable methods can be found to incorporate neutron absorbing nuclei. Si detectors would also seem to offer the possibility of 'fast' detection, although - as with organic scintillators - the problem of coupling the neutron absorber remains.

By comparison, the problems of providing faster electronics, more computing power and larger memories appear straightforward. The speed of electronic components, the power of microprocessors and the size of memories (per unit cost) are all increasing at a factor of ~ 2 per year. Thus over the next 10 years the 'limits' shown in Table 1 for mean data rate and histogram size will easily increase to reach the values required by the ESS. The use of multiple processors, in combination with their increase in speed, will also provide the necessary CPU power to visualise the most demanding ESS data, and to provide the 'ease' of data handling now required with complex instrumentation.

Session: Scientific Horizons II *Chairman R Pynn*

Crystallography and Materials Science

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Introduction

Neutron powder diffraction is a powerful materials science technique that has made a significant impact in many areas of solid state research. Recent highlights have included high temperature superconductors and fullerenes. The unexpected nature and sheer surprise of these new areas of research make detailed predictions about the future of neutron powder diffraction almost impossible. However, general trends can be predicted and upgrading of instrumentation planned that will broaden the applicability of the method. Quantitative improvements in intensity, resolution and decreasing sample size will all lead to qualitatively improved new science. For example, optimisation of cold moderator technology at spallation neutron sources will lead to new science in the study of magnetic materials, structure determination and large structure refinement. In looking to the future, the following five areas give a broad flavour of the potential developments in both pure and applied neutron powder diffraction:

• new materials	chemistry
• superconductivity	chemistry/physics
• phase transitions and transformations	physics/chemistry
• drug molecules	pharmacy/biochemistry
• advanced materials	metallurgy/materials science

New Materials

New materials represent one of the main challenges of neutron powder diffraction. Often new compounds are initially only available as powders - this has happened both for high temperature superconductors and fullerenes and is usually the case with materials such as zeolites and battery electrodes that have a strategic technological interest. However, while high T_c superconductors were immediately available in gram size quantities, the fullerenes were only synthesised in quantities of less than ~100mm^3. These small quantities severely limit the neutron powder diffraction technique. A thirty-fold increase in flux coupled with optimised detector configurations would permit ≤10mm^3 samples to be investigated at good resolution ($\Delta d/d \sim 10^{-3}$) for full structural analysis. Improved flux will also permit the detailed investigation of novel heterogeneous as well as homogeneous materials.

Superconductivity

The importance of neutron powder diffraction as a tool for materials research has been highlighted most clearly over the past five years by its use in the investigation of high temperature superconductors. The correct structure of the prototypic superconductor, $YBa_2Cu_3O_{7-\delta}$, was first determined by neutron powder diffraction in a number of different laboratories less than a month after the identification of the pure phase, indicating the rapid nature and reliability of the technique. Perhaps more significantly, however, neutron powder diffraction has become established in the area of high temperature superconductivity because of the wide diversity of information that has been obtained. Selective highlights include (a) the site-by-site investigation of oxygen stoichiometry in $YBa_2Cu_3O_{7-\delta}$ as a function of temperature and partial pressure, (b) the location of excess oxygen in the superconductor, $La_2CuO_{4-\delta}$, (c) the correlation of Cu-O bond-length with superconducting transition temperature through investigations as a function of chemical composition, oxygen stoichiometry and hydrostatic pressure, (d) the investigation of disproportionation in $Nd_{2-x}Ce_xCuO_4$, correlation with Meissner fraction behaviour and the development, through line broadening analysis, of a model of phase segregation and (e) the in-situ study of formation of 'bismuth' superconductors to unravel the conditions for formation of various phases and to determine the optimal synthesis routes. This list is by no means exhaustive but indicates the wide variety of neutron powder diffraction applications in materials research.

Phase transitions and transformations

A structure determined by single crystal techniques will generally give more information and improved precision and accuracy over a similar powder diffraction determination. However, powder diffraction usually offers significant advantages in the study of trends whether it be structural variations as a function of chemical composition or the scanning of a thermodynamic

variable. Improvements in sample environment will lead to detailed in-situ studies of reaction kinetics, better high pressure measurements and more precise derivations of order parameters and associated critical exponents at structural phase transitions.

Drug molecules

Although neutron powder diffraction impacts upon a wide range of scientific disciplines, there are areas of research where the technique has not yet been used. These include pharmacy and biology. Neutron powder diffraction is never likely to make contributions to macromolecular biology. However, with improved neutron sources, the study of fully protonated small drug structures, such as cimetidine and ibuprofen become tenable. Many drug polymorphs cannot be synthesised as $>1mm^3$ crystals and powders offer the only route to structure solution. X-ray synchrotron powder diffractometers will, over the next 5 years, be routinely used to determine the non-hydrogen backbone of drug molecules. However, there is intense interest in proton positions which largely determine biochemical activity. High resolution cold-moderator neutron powder diffractometers on a future spallation neutron source will provide such answers.

Advanced Materials

Neutron diffraction is a bulk probe that is commonly used to study homogeneous single phase systems. However, many new materials, particularly of technological interest, have been specifically designed to be heterogeneous. These include γ-γ' Ni superalloys used in aeroengine turbine blades and SiC-metal matrix composites used as strong light materials. Higher intensity neutron sources will have a significant influence on the study of these advanced materials. Higher fluxes permit finer beams to be used to probe the spatial variation within a sample or close to a surface. Higher resolution not only allows the measurement of lattice parameter mismatch and residual macrostrain in composites and superalloys but also the strain mismatch and microstrain broadening. Large area detectors will be used to study texture, particularly in non-isotropic materials such as SiC-metal matrix composites. Fixed angle geometry can be optimised to investigate catalytic reactions and, for example, to study in-situ cathode, anode and electrolyte behaviour during battery charge and discharge cycles.

Scientific Horizons on a Pulsed Neutron Source

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In trying to guess the future shape of neutron scattering research of condensed matter in a time frame of 2 to 3 decades, I come to two major conclusions: (a) things will be very different from what we are used to today, but (b) neutron research will remain a key ingredient of condensed matter physics.

The changes will have to be due to the development of synchrotron radiation, on the one hand, and to the evolution on neutron sources, on the other. The old rule "Don't do it with neutrons if you can do it with X-rays" takes on a completely new meaning with the tremendous and still fully expanding capabilities of synchrotron methods, and it suffices to refer, as a recent example, to the progress of use of polarised X-ray methods in magnetism. Anomalous scattering effects near absorption edges are other examples of great opportunities. With respect to neutron sources, the novelty is to be expected from the maturation of pulsed spallation source techniques. The reactor approach can eventually provide a gain of a factor of 3-10 above ILL, but this appears to be the ultimate limit. Gain factors in data collection rates with the next generation of spallation sources in contrast are two orders of magnitude in many cases, and an order of magnitude overall. This is to say, that spallation sources in the future should not be envisaged as complementary to steady state ones (as they are today) but as the general neutron sources of the future.

In face of the increasing competition from synchrotron radiation, the significance of neutron research relies on its complementary, and this will remain so for the foreseeable future. We have to accept that synchrotrons will be the major partners in this complementary approach, while neutrons will be indispensable in special cases or for special purposes. Thus, the weak interaction between neutron and matter and the general validity of the first Born approximation in inelastic and diffuse scattering is a fundamental asset which will continue to play a key role in quantitative comparison with theory - the ultimate validation of progress in physics. Tracking light elements is another area where the inherent difficulty of very small cross sections for X-rays provides a long term need for neutrons. Inelastic scattering in the most relevant energy range (few meV and below) is, as of today, the monopoly - and a very important one - of neutron methods. Extending high resolution techniques to the study of elementary excitations is a major additional opportunity for neutrons. Although it is never prudent to exclude fully unexpected and apparently "impossible" future technical breakthroughs, neutron scattering methods do possess a tremendous advantage - both in principle and by the accumulated wealth of experience - in the field of dynamic studies, which by any educated guess will remain for a long time to come.

Prospects for Liquid State Physics

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Introduction

As is commonplace with many other branches of condensed matter science, the role of one particular technique, in this case neutron scattering, must be set firmly in context with other experimental methods. This is particularly important now that a new generation of synchrotron light sources is coming on stream. Care should be taken to ensure that the science carried out at the ESS is fully complementary to what can be achieved by X-ray methods, and not to 'overstate' the case. Nevertheless, the ability of neutrons to penetrate samples, the relative ease of inelastic studies, and the isotope dependence of the scattering length, give neutrons a decided competitive edge. Liquid systems or topics which will benefit from the ESS include the study of the structures of:

- liquid rare gases (p, V, T and mixtures)
- liquid metals ($S_{ii}(Q)$, $S_{ie}(Q)$ and $S_{ee}(Q)$
- liquid alloys at the metal to insulator transition (structural dependence of the mobility edge) including the formation of Zintl ions
- molten salts (pairwise + higher terms, exploiting $\delta S_{AA}(Q)/\delta\rho$)
- aqueous solutions (H/D substitutions) and behaviour at electrodes
- ordered liquids
- hydrophobic systems (proteins, solutions of rare gases, methane hydrates)
- liquids perturbed by transient, shear, single pulses
- liquids in pores, low dimensionality etc.
- biology on the few Å scale in solution, made possible by ^{14}N to ^{15}N exchange and the developments in biotechnology

The European Spallation Source

The CEC's Study Panel on Neutron Beam Sources estimated that the growth in 'real' demand for neutrons is likely to be 10% pa for the next few years, and proposed that the 'Figure of merit(s)' of pulsed source instruments versus possible steady state source instruments should

be at least comparable for the 'worst' cases, and in general > 300 in order to expose new science. This may involve the transfer of technology from steady state to pulsed sources (e.g. whole instruments - triple axes; instrumental methodology - spin echo; specific techniques - polarisers) as well as the transfer of technology/experience from ISIS, IPNS, LANSCE, etc.

A particularly interesting possibility for liquids is the notion of 'tuneable' resolution, particularly for quasi-electric work and liquid and glass structure at high Q.

Basic Requirements

(a) Structure

- high flux (good statistics, small or dilute samples)
- high stability (difference methods)
- wide range of Q (real space resolution)
- small angles of scatter (inelastic corrections)

These developments will lead to new standards of structural work, including the exploitation of small isotope differences (e.g. ^{12}C to ^{13}C), and/or a reliable synergy with X-ray methods.

(b) Dynamics

- maximum dynamic range
- adequate Q and ω resolution
- high signal to background
- optimised resolution functions

Likewise, these developments should lead to new standards for quasielastic work with meV resolution, high intensity, small Q steps and a wide energy window. So far as Brillouin scattering is concerned, studies close to the hydrodynamic limit but not actually at Q=0 at a precision capable of testing perturbation theories will form an important branch of liquid state physics over the next few years.

Scientific Horizons in Magnetic Structures

J B Forsyth, ISIS Facility, RAL, UK

The Study of Magnetism by Elastic Neutron Scattering

Although the advent of second generation sources of synchrotron radiation have produced a number of interesting studies which bear on the magnetic states of condensed matter, the low

cross sections involved in the absence of resonance enhancement ensures that magnetic photon scattering is many orders of magnitude weaker than the accompanying charge scattering. The contribution of synchrotron radiation to the study of magnetic structure is therefore likely to remain complementary and subsidiary to the results of neutron scattering, in which nuclear and magnetic cross sections are of the same order.

Neutron diffraction is a unique tool in the determination of magnetic structure. The vector nature of the interaction between the neutron and the magnetisation means that the scattered intensity contains information about the directions of the atom-centred moments, as well as their magnitudes. However, for all but the simplest collinear arrangements, the observed magnitudes of the magnetic interaction vectors may not lead to a unique model for the structure. This is particularly true for powdered samples of high symmetry materials, where it is well known that the combination of domain structure and the overlap of equivalent reflections limit the determination of moment directions.

Some of the fields where neutrons have made a major impact include:

- magnetic structure determination and its role in the discussion of exchange interactions;
- single-ion anisotropy;
- frustration and moment instability , eg Mn;
- Rare earth incommensurate structures, long-periodicity quasi ferromagnets e.g. MnSi, FeGe;
- fan structures;
- amplitude modulation.

Magnetic structure can also be studied as a function of temperature, pressure and magnetic field. The ESS should enable us to combine two or more of these variables more easily, since smaller samples can be used and more limited egress needs to be provided for the scattered radiation.

Since the complete description of the magnetic cross section includes the spin state of the neutron, both before and after scattering, it is clear that a knowledge of one or preferably both of these vectors must lead to more precise descriptions of the scattering system. A polarised incident neutron beam enables much more accurate measurements to be made of magnetic scattering amplitudes through their interference with their nuclear counterparts. This technique has been exploited for over three decades and is continuing to provide detailed information of prime interest to condensed matter physicists and chemists. Typical studies include the determination of magnetic form factors: these are influenced by the band structure in metallic systems, and may also serve to identify the ground state in actinide materials or the degree of delocalisation in weakly ferromagnetic intermetallic compounds.

The extension of the technique to exploit the white pulsed beam of the ESS lends weight to current developments in polarising filters, particularly ^{3}He. The efficiency and wavelength range covered by such a device are potentially superior to those of either the polarised proton or polarised Sm filters. ^{3}He offers the dream of a filter which, once polarised, can be removed from the polarising equipment and installed in a neutron instrument where it will remain effective for at least 24 hours. It should also be noted that such a filter will be more efficient, even at a single wavelength, than the monochromating polarising crystals used at reactor sources.

Since the so-called polarised neutron flipping ratio technique is sensitive as well as accurate, it is ideal for the study of weakly magnetic systems in which magnetic and nuclear scattering occur in the same Bragg reflection and are not in phase quadrature. All paramagnetic and diamagnetic materials comply with this requirement, and the accuracy with which they can be studied is governed by the complexity of the chemical structure and the magnitude of the moment that can be induced by an external applied field. The electrons responsible for magnetism in transition metal ions are also involved in their bonding to anions and their spatial distribution reflects the molecular orbitals responsible for covalency as well as the influence of the local crystalline field. Almost complete paramagnetic alignment can be achieved with a field of 5 T at liquid helium temperatures. An example of this type of investigation of prime significance to chemists is the comparative study of the hexaaquo ions of the divalent ions of the first group transition elements. A similar study of the more covalent trivalent anions is now underway and has already been extended to the second long period ions Mo(III) and Ru(III). Much work remains to be done to extend this type of work to complex anions. A more intense source and lower backgrounds should enable further progress to be made in the study of diamagnetism which has so far been limited to graphite and bismuth . Significant new chemical information has resulted from the polarised neutron studies of cluster compounds such as Nd_6I_{11} and compounds in which the paramagnetism arises from nitroso groups.

Whilst the polarisation dependent cross sections occur in many classes of magnetic material, the majority of antiferromagnets do not lend themselves to the flipping ratio technique. If the magnetic propagation vector describing the structure contains fractional or irrational components, then the magnetic scattering is separated from the nuclear and interference cannot occur. Similarly, the existence of antiferromagnetic domains within the sample may contribute in such a way as to diminish or remove the polarisation dependence. The magnetically scattered intensity must then be obtained by classical integrated intensity measurements with an unpolarised incident beam.

Recent experiments in which both the incident and elastically scattered neutron polarisation have been controlled and measured by a zero-field polarimeter have reinforced the demand for this type of measurement to be routinely available for the study of antiferromagnetic structures.

Polarisation analysis, with or without a magnetising field on the sample, will find increasing application in many types of magnetic investigation, whether it be the determination of magnetic structure, the inelastic measurement of coherent magnetic excitations, the separation of magnetic from nuclear diffuse scattering or the reflection from magnetic surfaces and multilayers.

Amongst other potential uses of the ESS in this area are:

- Measurement of magnetic diffuse scattering.
- Reflectometry from magnetic surfaces and layer structures.
- Topography.
- Domain structure.
- Nuclear magnetism.

The use of magnetic fields at the sample position in neutron elastic scattering studies

Many neutron studies of magnetic materials require a magnetic field to be present at the sample position. The strength and orientation of the field with respect to the scattering geometry vary depending on its purpose. A low magnetic field of some 10^{-2} T may simply be required to maintain the direction of polarisation of an incident polarised beam. A modest field of 1-2 T is sufficient to align ferro- and ferri-magnetic domains in materials with low coercivity. The highest achievable field which can conveniently be provided within the constraints of the scattering geometry is of great interest for aligning paramagnetic and diamagnetic samples, for aligning the moments in magnetically hard materials and for studying its influence on the magnetic structures of metamagnets and antiferromagnets, in which spin flop transitions may be induced in fields of order 10 T. Ultimately, pulsed field studies at >20 T using single pulse data acquisition will fully exploit the unique possibilities of the ESS.

The study of magnetic structure and disorder in ferro- and ferri-magnets using polycrystalline or powdered samples can achieve isothermal separation of magnetic and nuclear scattering by using the Q = 0 switch in which the moments are aligned along or close to the scattering vector κ. An angle of $\pm 5°$ to κ reduces the magnetic scattering to < 1%. The limitation in angle which the detector may subtend at the sample then clearly favours the t-o-f technique since a large range of d-spacings can be recorded simultaneously. The most convenient geometry would be backscattering into a detector subtending 20° on one side of the incident beam with H offset, or a detector from 170° 2θ on either side of the incident beam with **H** provided by a solenoid coaxial with the beam.

Q=1 measurements require the sample magnetisation to be perpendicular to the plane of scattering. In most reactor instruments the direction of magnetisation is vertical, the plane of

scattering horizontal and normal beam geometry with a single lifting detector is used to measure one reflection at a time. The static detection system associated with most pulsed white beam instruments suggests an alternative and more effective geometry in which the direction of magnetisation is parallel to the incident beam and measurements are made with 2θ at or near 90° (**Q**=1).

From the above considerations it would appear that **Q**=0 and **Q**=1 measurements may be made using a single design of superconducting magnet in the form of a Helmholz pair of coils. Such a magnet could also find application in the measurement of polarisation- dependent cross sections. A vertical field, produced by asymmetric windings to preserve the polarisation of the input beam, is normally adopted for monochromatic, continuous source instruments, so the incident beam lies in the plane perpendicular to the field direction. The polarisation-dependent cross section depends on **Q.P** and is therefore a maximum for reflections whose scattering vectors are perpendicular to the magnetising field. A lifting single detector and normal beam geometry accesses a reasonably large volume of reciprocal space for a single sample mounting. However, the angle between **Q** and **P** rapidly increases and the cross section reduces for reflections measured at low g and high (20°) γ.

Pulsed white beam flipping ratio measurements might also make use of this magnet. In this option, the incident beam would be polarized parallel and antiparallel to the beam direction. There would then be no difficulty in preserving the polarisation of the incident beam and the magnet coils could be identical, in contrast to the asymmetric design described above. A penalty of a factor of two reduction in the polarisation-dependent cross section would then occur for reflections measured at 2θ= 90°.

The present analysing system for zero-field polarimetry using a monochromatic incident beam is essentially limited to a single direction for the scattered beam. At a pulsed source orders of a given reflection could, however, be measured with reduced efficiency at a single position of the analyser. A considerable development programme is required to improve the efficiency of this type of measurement. In-field polarisation analysis based on measurements of three mutually perpendicular components of the scattered polarisation, though less complete, can be more easily adapted to exploit the potential of the ESS. All reflected beams which pass through a given position of the analysing system can be measured, so the efficiency is directly proportional to the solid angle subtended at the sample by the analyser.

Isothermal separation of magnetic and nuclear scattering can be performed by having $\mathbf{P}_i$ parallel to κ. In this situation, all magnetic scattering is spin flip and can be separated by in-field polarisation analysis. This is of principal interest in powder diffraction. Again, the limitation in angle which the detector may subtend at the sample favours the t-o-f technique and limits the area over which analysis must be performed.

UCN Research at an Advanced Pulsed Neutron Source

R Golub, Hahn Meitner Institut, Berlin, Germany

Some of the most successful work at the ILL has been in the field of fundamental physics with Ultra Cold Neutrons (UCN). It would be a pity if the planning for the ESS was to go ahead without making arrangements for similar work to be performed at this powerful spallation source.

The construction of ESS would offer an outstanding chance to produce a uniquely intense source of UCN - based on the super-thermal source principle of collecting the UCN produced by downscattering in a vessel with a long storage time, and hence producing higher densities than are available by more conventional methods which are limited by Liouville's theorem. Such a source would transform the field by an amount even greater than the transformation achieved by the current turbine UCN source at the ILL.

This source, where the UCN would be produced in a thin film of solid deuterium, would allow the presently most successful applications of UCN (the search for a neutron electric dipole moment and measurement of the β decay lifetime) to be performed at much higher accuracies as well as opening up a new range of measurements and instruments:

- angular correlations in neutron β decay (polarised UCN offer an extremely clean way of carrying out such experiments);
- the search for a gravitational dipole moment;
- UCN interferometers with a broad range of applications ranging from the neutron electric charge determination to investigating general relativistic effects;
- operation of a UCN microscope at much higher resolution than has been obtained to date.

Further developments of UCN are anticipated for inelastic and quasi-elastic scattering from condensed matter . Quasi-elastic energy changes as small as 10^{-11} ev/bounce have already been observed and an improvement of the NESSIE inelastic scattering instrument at the ILL allowing 2π collection of the scattered neutrons is under construction at Kyoto. A Neutron Resonance Spin Echo spectrometer for UCN is also being studied. All these points constitute a very strong case for the inclusion of a strong UCN programme at ESS.

Positive Muons as a Magnetic Resonance Probe of Structure and Dynamics

S F J Cox, ISIS Facility, RAL, Chilton, UK

This talk notes the ESS potential as a parallel source of low energy muon beams for condensed matter and chemical studies. The suggestions discussed extrapolate from the success of the ISIS Pulsed Muon Facility (established with the aid of a CEC grant) and exploits the fact that accelerators designed for spallation neutron production are close to optimal for muon production also. Huge muon intensities could be expected at the ESS even in parasitic parallel operation, with little effect on neutron yield.

At ISIS, muons are produced in a thin graphite target, working in transmission in the extracted proton beam, just upstream of the spallation neutron source. Less than 2% of the protons are intercepted, but this suffices to provide the world's most intense pulsed muon source. Scaling to the ESS specification would outperform even the purpose-built meson factories which presently are the major centres for implanted muon studies. The following is a first look at feasibility and potential of the ESS for muon studies.

μSR spectroscopy

It is rather surprising that a hydrogen atom, introduced into silicon, is more stable at the centre of a silicon-silicon bond than within the ample interstitial spaces which exist in the silicon lattice. It is surprising also that the screening charge of conduction electrons, which surrounds interstitial defects in metal lattices, cannot always follow the motion of the interstitial particle adiabatically, but may cause a significant drag on its diffusion at low temperatures. It is surprising that organic radicals, invisible to ESR spectroscopy in the gas phase, may be detected readily when one proton is replaced by an unstable particle with a lifetime of only two microseconds; it is also surprising that nuclear zero--point energy in certain of these radicals may be a deciding factor in the total energy balance and determine the molecular configuration.

These are some of the more remarkable findings of μSR spectroscopy, which exploits the similarity of the positive muon and the proton in their chemical interactions with matter. It may be likened to magnetic resonance spectroscopies, but with a sensitivity and selectivity which is much enhanced over conventional NMR or ESR: the muon probe may be implanted with an initial polarisation close to 100% (irrespective of the temperature of the sample or the magnetic field applied to it) and the subsequent evolution of its polarisation monitored *via* the asymmetry in the muon's radioactive decay (this is the classic example of parity violation), making use of single-particle detection methods. More routine applications involve the muon as

a sensitive and accurate magnetic probe: implanted as a microscopic magnetometer in magnetic or superconducting materials, its characteristic precession frequencies determine the interstitial local fields, its linewidths or relaxation functions characterise the distribution or fluctuation of these fields. Particularly important recently have been the reliable measurements of London penetration depth in a variety of superconductors (most notably the high-T_C cuprates, but also heavy-Fermion systems, Chevrel phases and even some organics) and the identification of antiferromagnetic parent compounds of the high-T_C superconductors.

Such studies ideally complement diffraction methods. The microsecond muon lifetime defines a new time-window for the study of dynamical phenomena, however, which neatly bridges the gap between neutron scattering and conventional magnetic resonance.

Pulsed *vs.* continuous sources

Some muon spin resonance measurements place no demand on the time structure of the muon beam. A simple integral counting of the forward-backward asymmetry in the muon decay allows full use of the intensity available at muon sources such as PSI and TRIUMF (and potentially also at LAMPF). Other muon spin relaxation measurements require modest timing resolution. At continuous sources this means only one muon in the sample at a time and represents a severe limit on data rate and a waste of available intensity; there is also an inevitable background count from false correlations. At a pulsed source, there is no such fundamental limit on data rate and the spectra are virtually background-free, allowing slow relaxation to be followed to long elapsed time. Both types of study could be envisaged at the ESS.

Pulsed sources are inherently unsuitable for muon spin rotation studies in which high frequencies are to be observed directly. Access to high frequency spin transitions is regained by the use of RF resonance, however, and this technique has the advantage that it is not confined to the study of species formed promptly on muon implantation, but may also be used to analyse for the final state following thermal conversion or chemical reaction.

Feasibility: muon production targets

The schematic layout of Figure 5 shows how all beams might be used. Thick targets as presently used at PSI and TRIUMF would undoubtedly be suitable for the 125 μA neutral beam available from partially striped H^-, and result in an order of magnitude gain over ISIS muon intensity. Here the long pulse length would imply integral counting spectroscopy. The muon beam might be split to increase usage.

A thin rotating transmission target could be envisaged for the 1250 μA proton beam, based on LAMPF experience. The microsecond pulse length would allow time resolved measurements

and might be further shortened by slicing (with successive slices directed to separate experimental stations). An order of magnitude increase over ISIS performance could also be expected here, by virtue of the increased current and improved muon collection.

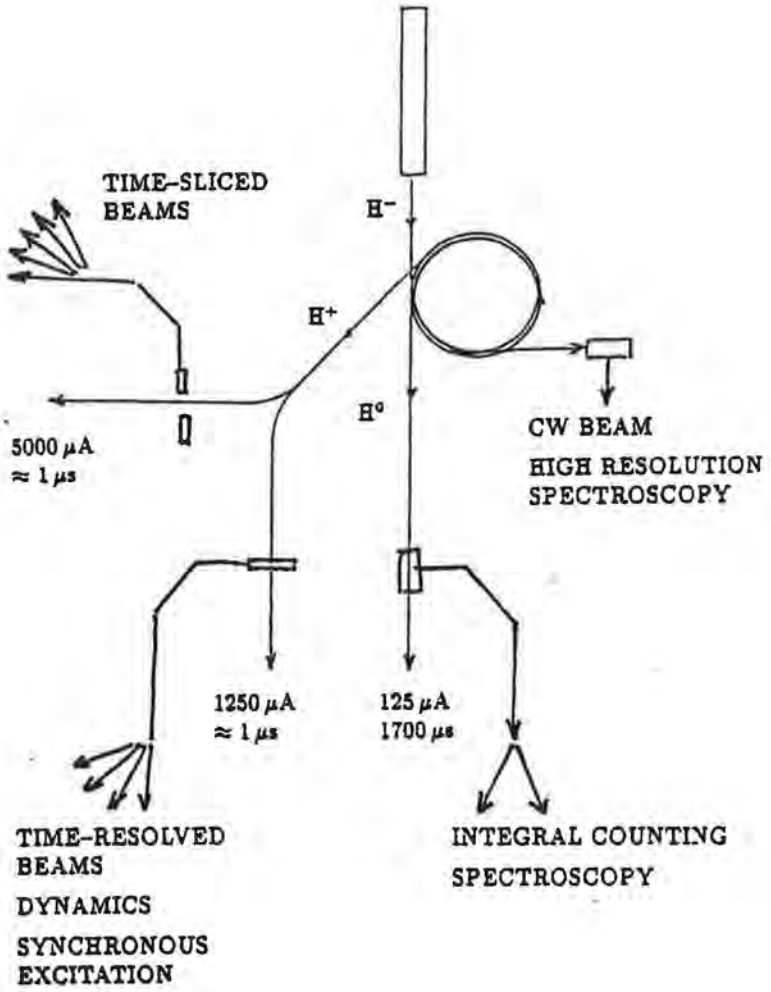

Figure 5 Conceptual scheme for muon beamlines and their usage at the ESS.

Substantial further gains would result from utilisation of the 5000 μA beam. Present intermediate target designs are inadequate here, and there is a concern that failure of the intermediate target would result in sudden shrinkage of the proton spot on the neutron spallation source. A safe solution might be the use of 'scraper' targets in the fringe of this beam, but this is an area which requires expert appraisal.

Finally, the possibility of specifying a fourth storage ring should not be overlooked, with this ring used to stretch rather than compress the linac pulses. Slow extraction could then provide a CW muon source so that high frequency muon spin rotation experiments could also be offered.

Feasibility: data acquisition

Pulsed μSR implies high instantaneous count-rates at the peak of the muon decay curve. With conventional digital detection, this can lead to distortion of the data from dead-time losses in the detector chains. Segmentation of the detectors is used at ISIS (as at KEK) to minimise this effect, but further segmentation could be prohibitively expensive. A promising solution is the examination of the anode-current signals using transient recorders. This analogue detection method proves to be far less sensitive to saturation. It has already been successfully used in the detection of RF resonance. Tests show that it is also suitable for the display of precession signals out to at least 8 muon lifetimes. Exploitation of the natural advantage of pulsed sources may require some hybrid scheme reverting to digital detection at long elapsed time. ISIS holds the record in providing useful data out to about $10\ \tau_\mu = 22\ \mu s$: this could be stretched to 30 μs at ESS intensity.

Scientific outlook

The present status of μSR spectroscopy can best be compared with that of magnetic resonance some decades ago: experiments on model systems have validated the technique - but also given some surprises which have served to eliminate complacency and upset established views in several areas (notably in defect electronic structure in semiconductors and quantum diffusion in metals). The spectroscopy can now be applied with confidence to a wide variety of materials, and in particular to the modelling of hydrogen in materials. In semiconductors as in metals, extension of studies from the crystalline to the amorphous materials now seems timely and promising, issues of hydrogen localisation and diffusion being accessible to μSR studies.

The large field of muonium chemistry now promises an important extension from liquid-phase to surface and gas-phase studies. The detection of transient radicals also offers the prospect of laboratory simulation of some aspects of atmospheric chemistry which are a cause for environmental concern.

RF resonance, already practised at ISIS, is one example of sample excitation which is synchronous with the muon pulse. More extreme environments which could not be sustained continuously include laser irradiation and high pulsed magnetic fields. The ESS would be ideal for such experiments and could even provide sufficient intensity for radiolytic excitation by the muon pulse itself.

Demand

The fast growing demand for muon beamtime for μSR programmes has put huge pressure on all the existing sources, and nowhere more than at ISIS. Extension of capacity on the scale offered by the ESS proposal is required for the full potential of this still novel spectroscopy to

be realised. The European μSR community would welcome the opportunity to participate fully in further design studies for the ESS and in establishing the scientific case for its realisation.

Session: Scientific Horizons III *Chairman J* W White

The Future of Collective Excitations

J D Axe, Brookhaven National Laboratory, USA

In this talk, the complementarity of X-rays and neutrons was stressed along with the present and likely future contiguity of synchrotron and high flux neutron sources. This is a desirable complementarity and users should be at least as clever as the carpenter who does not use the chisel as a hammer. The durability of this complementarity was also evident and the first example of the magnetic scattering at low temperature from $Tb_{1.2}Mo_6S_8$ where superconductivity and magnetism coexist at temperatures of ca 0.07K illustrates this point. Here the magnetic diffraction, though in principle observable by X-rays, is only obtainable by neutron diffraction because of form factor and background limitations.

A second enduring complementarity is that associated with triple-axis spectrometry for complex materials. While it is evident that the multiangle inelastic instruments at pulsed sources such as HET, MARI and PRISMA (at ISIS) have already a performance above 50 meV which surpasses triple-axis machines, there is an obvious problem of data analysis at lower energies and for the excitations of more complex structures such as the cuprate superconductors. Unless quite novel ways of data presentation and interfacing to the physics can be found there will still be the need for the highly selective scans in (Q,ω) space provided by the triple-axis method. Though the method is "mature" the pulsed source offers advantages in triple-axis operation so long as a mean flux near present steady state fluxes is available. Time of flight order sorting and background suppression by gating data collection into the pulse are avenues to be explored.

A final point of interest is to study phonon line widths, for example in superconductors, to follow the electron-phonon interactions. High resolution and a precisely defined resolution function are needed for this.

Scientific Horizons in Large Scale Structures

R K Thomas, Physical Chemistry Laboratory, Oxford, UK

In the study of 'large scale structures' using neutron reflectometry and small angle scattering there is a convergence of fundamental science and technological interest. A "wholeness" in both sorts of problem is attained by combining reflectometry e.g.. from single surfactant monolayers with small angle neutron scattering from the same system in dispersion or after aggregation. For this work isotopic contrast is essential, there being a need for systematic variation of the contrast of all system components to ensure a unique interpretation of the data. The complementarity with X-rays is minor since for complex systems their use does not provide adequate contrast variation to resolve the uniqueness problem. " X-rays are for the special occasion - neutrons are the more general and easy technique to use".

Three examples which point the way to future needs were discussed.

(a) The nafion membrane structure determined to high resolution, combining small angle scattering and high angle measurements out to $15Å^{-1}$ to determine partial structure factors for the pore-pore and atomic correlation functions in much the same way as Enderby has done for ionic solutions.

(b) Reflection studies of surfactant embedding at the air-water interface, where again the approach is to use the full scattering function and determine partial structure factors by appropriate isotopic combinations. The great importance of chemical synthesis and wide Q range in this work was stressed.

(c) The third way forward was through the extensive use of simulation combined with the systematic variation of the contrast of all system components and partial structure factor measurement. Advances in computing and progress in understanding the physics of reflection and small angle scattering make this promising.

Scientific Horizons in Magnetic Excitations

B D Rainford, University of Southhampton, UK

The first point stressed the future importance of 'parametric studies', the systematic variation of temperature, pressure, alloy composition etc. in $S(Q,\omega)$ studies of metallic magnetism. The first example discussed was the study of heavy fermion systems like $CeRu_2Si_{2-x}Ge_x$ alloys on IN6 at ILL. For such work the triple-axis spectrometer is an 'outdated concept' though still indispensable for some crystal studies. By such parametric studies the Q modulation of the

magnetic response, the single ion part of the linewidth and its scaling behaviour, can be extracted.

In the separation of magnetic and phonon scattering it was asserted that "polarisation analysis has not yet got anywhere" despite its enormous potential and the fact that, in principle, white beam polarisation (through dynamic proton polarisation) is a solved problem. The experimental problem is on the side of the analysis of the scattered beam. A valuable approach developed by Osborn has been to use the high Q density of states from a sample as the basis for Monte Carlo simulation (of the phonon part) of the scattering. An example of the present difficulty and challenge to experimental development was the work done on YMn_2 which needed about three weeks on D5 to do the polarisation analysis.

A final point was that neutron scattering has much to learn from the astronomers and crystallographers in data sharing and availability. The Cambridge data base is now a world resource. The same should be true of neutron scattering data and the opportunity now exists just as large data sets are beginning to be acquired to put a systematic archiving into place. Perhaps the rule should be that, like astronomy, neutron data should be the property of those who take it only for the first two years.

Horizons in Polymer Science

J S Higgins, Imperial College, London, UK

There is a word of caution to be noted in preparing any 'wish list' to new instruments for polymer studies. Fifteen years ago studies of the time dependence of polymer structure with the best source in the world at ILL were foreseen as a major future goal and reflectometry was not even imagined. The present and future impact of this latter technique for studies of interfacial mixing and adhesion are only in their infancy.

As concerns instruments, polymers are "large and slow"; appropriate ranges of momentum and energy transfer (10^{-3}Å^{-1} and 1 - 1000 neV) are required but as with "large scale structures" (above) extension of these ranges to higher energy transfers looks to be important. It was also emphasised that the neutron experiment (whether SANS or reflection) is unique as the use of isotopic contrast is essential to study single (tagged) polymer molecules in "crowded surroundings".

A second main theme for the future is that more systems must be studied. The paradigms of behaviour for polymer systems are more blurred than is the case for much of liquid and solid

state physics. Examples of technologically and fundamentally important new systems are stretched polymers, liquid crystal polymers and polymers trapped in networks.

Polymer engineering is likely to become a significant area of interest. This will not only involve studies of phenomena such as spinodal decomposition of mixtures (important in polymer recycling) but also examination of polymer strain, creep and adhesion in engineering applications in much the same way as residual stress is now being done on welds etc. using the penetrating power of neutron diffraction. Examples of the time dependent small angle scattering (using D11 at ILL) resulting from demixing of M_W 10^5 d8 polymethyl methacrylate / M_W 10^5 polymethyl styrene-co-acrylonitrile and the time dependent reflectivity from interfacial mixing were discussed.

Scientific Horizons in Magnetism

M. Steiner, Johannes Gutenberg Universität, Mainz, Germany

The need in future work on magnetism to address problems at the level of

(a) a microscopic understanding of the physics with complete theories capable of explaining all details of paradigm experiments which demonstrate the effects of dimensionality, symmetry etc. and

(b) very complex systems, such as the new "hard " magnets $Nd_2Fe_{14}B$ (63 atoms per unit cell) (is it localised or itinerant magnetism?) etc.

presents neutron scattering with a strong experimental challenge. Industry requires the latter knowledge which might best be provided by careful understanding of some paradigms in (a) with carefully chosen experiments in (b).

Examples of both types of work were presented. In the first, topological solitons, the nature of the differences (following predictions by Haldane) between integer and half integer quantum ground states and the value of e.g.. 2D Heisenberg S=1/2 models for high T_c systems were covered. For the second the need for full polarisation analysis and the determination of mode eigenvectors to "fingerprint" the state of a system were stressed.

Future directions identified were the investigation of (i) transitions between non-linear and chaotic behaviour, (ii) the effects of microscopic to macroscopic scale change. Instrument development was critical - the study of "hard" magnets being presently impossible for lack of intensity and data gathering power on present sources.

Optical Devices for an Advanced Neutron Source

J B Hayter, Oak Ridge National Laboratory, USA

The present, short term and long term availability of neutron beam insertion devices was considered in the context of developments in hand for the US Advanced Neutron Source at Oak Ridge. The four categories of availability were

- On Sale Today
- Available Real Soon
- New and Improved
- Vapourware

In the first category , see Table 2, are the guides, supermirrors, beam splitters and long focal length lenses. Polarisation is an available option at some level of beam divergence for all of these. There seems to be little justification to go to the expense of ^{58}Ni for the 18% gain in solid angle when supermirror guides will do much better. Stacked coated mirrors to form a channel guide, though of high theoretical performance for a truly parallel input beam, have greatly degraded polarisation performance , in practice, because of beam divergence.

"Available Real Soon" items include transmission polarisers (e.g.. ^{3}He) , toroidal lenses and crystal mirrors - some prototypes of which exist but are at the limits (e.g. for smoothness) of the best available technology - and supermirrors out to $5\theta_c$Ni . Such an item could not be produced by the relatively cheap evaporators currently in use for guides and supermirrors but require the $M5 - $M10 devices used by semiconductor companies. Two US companies have indicated interest. These items require strong development investments but are essential elements in future high performance instruments.

In the "New and Improved" domain are capillary guides (some are being tested at NIST , Washington), capable of bending neutron beams by 180° and of creating a short focal length convergence and the final category opens a vista of revolutionary instrumentation based upon current developments e.g. in neutron interferometry.

Table 2

On Sale Today

- Conventional Single Channel Guides
  - Ni or ^{58}Ni
  - Supermirror
- Variations on the Theme
  - Beam focussing (anti-trompette, spiral)
  - Divergence reduction (trompette)
  - Beam splitters
  - Beam benders
  - Wafer stacks
- Lenses
  - Hexapole (cold)
  - Fresnel (very cold)
- Polarised flavours are optional

Available Real Soon

- Transmission Polarisers
  - Various wafer stack geometries
  - ^{3}He
- Toroidal Lenses
- Spherical focussing crystal mirrors
- μ-Benders
- Supermirrors to $5\theta_c^{Ni}$

New and Improved

- Capillary Guides
  - Barrel lenses (convex and concave)
  - High curvature beam deviation
  - μ–focussing
- High-T_c hexapoles
- Aspherical focussing crystal mirrors
- Fast neutron filters

Vapourware

- Solid state fast white-beam switches
- Holographic insertion devices

  Lens hologram
  Crystal hologram
  Magnetic phase hologram

  - Spin-turn devices
  - Spin-echo focussing

- Magneto-optical devices

  Rapid phase modulation of precession
  Structural refinement by matching scattering from a phase modulated magneto-optical crystal hologram

European Spallation Source

Report of the Crystallography Working Group

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Report of the Crystallography Working Group

1. Introduction

Det er svært at spå, især om fremtiden *P.Storm Pedersen.*

Crystallography underpins all solid state research. A knowledge of atomic structure is an essential prerequisite to the full understanding of the properties of materials. Perhaps the most obvious recent example of this is the area of high temperature superconductivity: all the bulk properties of these materials are highly two-dimensional in nature and this is abundantly clear from a basic analysis of crystal structure, which reveals an atomic architecture that is dominated by square planes of copper and oxygen ions. Crystallographic techniques are used to study all aspects of crystalline material ranging from a description of the arrangement of the structural building blocks to an exact account of the atomic distribution. Any available radiation commensurate with the dimension of the object to be studied can be used, including X-rays, electrons and neutrons. For each of these there is of course constant development of new applications driven by new scientific fields and techniques. The goal of crystallography is to obtain structural information, and in many cases the use of several types of radiation is necessary to obtain a complete picture of the structure under investigation.

The emphasis of neutron diffraction has varied to encompass advances in the techniques. The first properties of the neutron exploited in diffraction were probably the nature of the atomic scattering factor and the magnetic interactions of matter with neutrons, and in the early days of neutron scattering with low flux sources the low absorption of most materials to neutrons were also of paramount importance. Subsequent to this, the combination of well understood reflection profiles coupled with new analysis techniques, especially the development in 1967 of the Rietveld method of profile refinement, invigorated the field of neutron powder diffraction and turned it into an indispensable tool for solid state chemists and physicists. Crystallographers also imported the techniques of H/D exchange first developed for small angle neutron scattering and in addition variable sample environments allowed structural studies to be performed in differing states of a material, particularly important in the use of neutrons for kinetic studies.

When planning new sources with new characteristics one must obviously base this on present day achievements, and a mixed blend of science and techniques naturally follows. It would be naive to believe that it is possible to predict what science shall be extant in ten years time, but the following is an attempt to list some of the types of science, the instruments that will enable it to be performed at ESS and most importantly the science that cannot be done now which will be feasible at the new intense source.

2. New Science on the European Spallation Source

2.1 Materials Research with Powder Diffraction

Powder diffraction

Neutron powder diffraction, like small angle scattering, is a relatively straightforward but indispensable technique. Powder techniques are indeed simple and well understood, but the applications are many and varied, ranging from high temperature superconductors to zeolite catalysts, from stress in engineering components to toughening of ceramic materials, from the chemistry of battery electrodes to the physics of fullerenes. Neutron powder diffraction, like small angle scattering, has made a major impact in the wider scientific community because it deals with 'real materials'.

The high penetrating power of the neutron, together with its strong scattering by light elements, are the two most important reasons for this success. In Rietveld refinement the former property means that a correct 'powder average' over many crystallites is normally obtained, where X-rays can suffer from systematic 'texture' errors. The latter quality means that light atoms such as oxygen can be precisely located in the presence of heavier elements such as those found in high temperature superconductors.

The numerous achievements of neutron powder diffractometers at both reactors and spallation sources are well documented. The development and rapid progress over the past 10 years of time-of-flight diffractometers at spallation neutron sources is remarkable. These achievements and developments have led to the establishment of the technique at the forefront of condensed matter research. The most notable examples are the early and highly significant contributions from neutron powder diffraction in the recent 'hot' topics of high T_c superconductors and fullerenes. However, many other examples can be sited that include template location in zeolites, in-situ discharging of electrode materials, the analysis of both the chemical and magnetic structures of important new ferromagnets such as $Nd_2Fe_{14}B$ and the study of structural phase transitions. Although the sheer range of science covered is impressive in itself, it is the proven precision and accuracy and consequent reliability of the technique over a broad range of physics, chemistry, materials science and geology that confirms and will

continue to ensure that neutron powder diffraction plays a central role in materials research. Consider, for example, high temperature superconductors. Neutron powder diffraction not only was used to determine the current crystal structures of the prototype superconductors $La_{2-x}Ba_xCuO_4$ and $YBa_2Cu_3O_{7-\delta}$, but has also been used to cover areas as diverse as

- the site-by-site investigation of oxygen stoichiometry in $YBa_2Cu_3O_{7-\delta}$ as a function of temperature and partial pressure;
- the location of excess oxygen in the superconductor, $La_2CuO_{4-\delta}$;
- the correlation of Cu-O bond-length with superconducting transition temperature through investigations as a function of chemical composition, oxygen stoichiometry and hydrostatic pressure;
- the investigation of disproportionation in $Nd_{2-x}Ce_xCuO_4$, correlation with Meissner fraction behaviour and the development, through line broadening analysis, of a model of phase segregation;
- the in-situ study of formation of 'bismuth' superconductors to unravel the conditions for formation of various phases and to determine the optimal synthesis routes.

This highlights the importance of neutron powder diffraction in a single area of research. Without doubt, the establishment of a vigorous programme of powder diffraction on the European Spallation Source will maintain and increase this high profile.

Ultra-high resolution neutron powder diffraction

Experience has shown that significant new science can always be achieved with increased instrument resolution, with hitherto hidden or ill-understood effects yielding to the improved instrument performance. The proposed High Resolution Power Diffractometer (HRPD; instrument POW-I) would improve the instrument resolution from the current best of $5x10^{-4}$ to $3x10^{-4}$ for d-spacings of 1 to 10Å from particles of dimension 1 to 10μm. By this time the resolution limit is imposed by the size of the powder particles themselves. As has already been shown on ISIS on the lower resolution HRPD, this would permit further advances in the field of direct solution of structures of moderately complex inorganic and organic materials. The improved signal/background ratio from the new HRPD should also allow work on hydrogenous materials without the need for deuteration. In the field of physics, it will permit the examination of critical scattering very close to the elastic Bragg peaks, as well as detailed examination of the peak shapes themselves to extract information about texture, strain, stacking faults, particle distribution and shape within the sample.

Materials in special environments

The subject of sample environments is dealt with in some detail below, but the general principles can be summarised briefly here with respect to neutron powder diffraction. As a consequence of their high penetrating power, neutrons are especially suitable for the study of materials in special sample environments providing high pressure and temperature. For example, the Argonne special environment powder diffractometer has been used to show how the high temperature superconductor $YBa_2Cu_3O_7$ is reduced to non-conducting $YBa_2Cu_3O_6$ on heating. With a higher intensity source, such techniques can be extended to higher pressures and temperatures, which can usually be obtained only for small samples. The proposed new Special Environment Powder Diffractometer (POW-III) will permit pressures of higher than 100 kbar to be reached at both high and low temperature. Pressure often has large effects on the properties of materials, for example changing the T_c of some superconductors, or altering the intermolecular interactions in fullerenes. Such studies provide vital clues to the basic understanding of these compounds.

Chemical kinetics

Chemical kinetics is well established as one of the major applications of neutron powder diffraction. Since neutrons are both sensitive to hydrogen atom scattering, and penetrative, the hydration-dehydration of materials such as cement can be followed *in situ* on the beamline. The availability of higher flux will allow the study of many more types of chemical reaction, including those that only occur at interfaces or in small volumes, for example at the surfaces of battery electrodes. The Time Resolved Powder Diffractometer TRPD (POW- IV) will address these problems, reducing count times for an experiment to the order of a few seconds, which is more appropriate to the time scale of many chemical reactions, while still maintaining a respectable resolution of some 4×10^{-3}. In addition to establishing reaction mechanisms, TRPD will permit the *in situ* mapping of phase diagrams, an essential step in the investigation of the stability regions of new materials as a function of composition, pressure and temperature.

Further technologically relevant research will be enabled by the increased flux which permits small quantities of reaction products to be detected, and allows the formation and decomposition of materials to be monitored at high temperatures in a uniquely clear manner. For example, a current problem in making practical thallium superconductors is in understanding all of the reaction steps, especially since thallium oxide is relatively volatile, and the stoichiometry of the TlO-layer greatly influences the superconducting properties. The proposed Small Sample Powder Diffractometer SSPD (POW-V) will be optimised to obtain full diffraction patterns from samples as small as $10mm^3$ in times as short as 1 hour.

Solid state chemistry

The main 'work-horse' instrument on the new source, the Structure Refinement Powder Diffractometer SRPD (POW-II), would be used for Rietveld refinement with a moderate resolution of 10^{-3}. Although such techniques are now well developed on existing sources, much higher intensity would permit much faster data collection cycles. This would allow the structure to be examined at small temperature intervals to investigate phase transitions, and many different sample compositions could be examined in one experiment. For example, subtle changes in the Cu-O bond lengths in high temperature superconductors as a function of the oxidation state of the sample have been mapped out on high intensity powder diffractometers at the ILL, and were used to demonstrate 'charge transfer' from charge reservoir layers to the superconducting Cu-O layers in $YBa_2Cu_3O_7$ on oxidation.

Long wavelength diffraction

It is becoming increasingly apparent that very long d-spacings are fairly common in nature, and that the orderings represented by these frequently influence physical properties. For example, the incommensurable structures of the Bi- and Tl-superconductors, or the quasi-crystalline order of new alloys are both of great significance. Neutrons are only one of a number of important and complementary physical techniques, which include electron microscopy and X-ray (synchrotron) diffraction, for studying quasi-crystals and incommensurable structures. However, neutrons are unique in the case of long (quasi-) period magnetic structures. To exploit this field a dedicated Long Wavelength Powder Diffractometer (LWPD; POW-VI) will be required on the ESS, operating with cold neutrons of wavelength 2-20 Å at a resolution of 1.5×10^{-3}.

Materials science and engineering

There is currently a significant amount of effort being expended towards the use of neutron diffraction techniques in the study of texture and stress in engineering materials. Neutrons are unique in this case because their high penetrating power permits investigation of the interior of bulk materials, for example machine components, rail pieces. Elastic and plastic deformation as well as phase transformations are of great importance for mechanical properties. Ideally these should be studied as a function of load, stretching, creep and heat treatment. With the increase in neutron flux these process dependent applications become realistic, with measuring times for one pole figure in texture analysis of the order of a few minutes, while retaining an adequate resolution $\Delta d/d$ of around 10^{-3}. The advantage of the new source will be that scattering can be obtained on a much finer grid of points, with scattering volumes as small as $1mm^3$. Automatic scanning of the stress fields will be possible with sufficiently high flux, which must be obtained while maintaining the relatively high resolution of 10^{-3}. The Materials Powder Diffractometer

(MPD; POW-VII) meets these specifications. With a pulsed neutron source it is possible to combine simultaneously texture with residual stress measurements in using several radial 90° detectors with special collimators, especially in a vertical 90° plane. The resolution properties of such a dedicated material science diffractometer are also sufficient to study textures in large crystallite minerals of geological interest.

2.2 Diffraction Physics

Much fascinating new science would be opened up by a source with sufficient flux to enable the construction of a high resolution physics diffractometer (SX-II) to allow the collection of single crystal data out to $\sin\theta/\lambda \sim 4$ Å^{-1}.

Order/disorder transitions

There is growing evidence that many (perhaps all?) structural phase transitions regarded as purely displacive in character are in fact order-disorder. That is to say, the position at which an atom is found in the low temperature phase is one of the two or more positions over which the atom is disordered above T_c. This is known to be the case for H in KH_2PO_4 where the H sites are 0.4 Å apart. Discontinuities in the thermal parameters at T_c strongly suggest the behaviour of the K and P atoms is like that of the H atoms but with a much smaller site separation. If it is in fact generally disorder, data to > 3 Å^{-1} would be needed to know this decisively. In this way a transition could be followed as the multisite distribution progressively orders in the temperature range just below T_c.

Long range order/disorder transitions

These transitions are known to take place with time in the oxygen chains in the high T_c superconductors, at or near T_c ,but have hitherto not been studied because of sample size, low intensity of the specific diffraction and the time scale of the experiment.. Such measurements become viable with the ESS and *vital* in the overall scheme in being able to understand the structure/property relationships in these fascinating materials. The existence of order/disorder which gives rise to diffuse scattering has begun to be studied at ISIS but the increased intensity of ESS and more detectors will enable this effect to be better observed and measured.

Anharmonicity

Even when an order/disorder transition is indiscernible, anharmonic distributions of atomic thermal motion are expected close to T_c, and data to a similar resolution would be required to follow these distortions as a function of temperature through T_c. Other examples of these types of studies are in the area of high T_c superconductors, searching for anharmonicity of the oxygen atoms in the vicinity of T_c. Studies of these kinds would greatly extend our

knowledge of transition processes, and provide the basis for a whole new level of interpretation of inelastic scattering. There is a further important class of problems concerning anharmonicity that has so far proved intractable: determinations of skewness directed along a polar axis. An example is ferroelectric $PbTiO_3$, where the indeterminacy of skewness along the 4-fold axis has prevented successful studies of ordering in this prototype system. Very high Q data should overcome this long standing problem, in part by reducing correlations among anharmonic terms.

Very small atomic shifts

Very high Q data are also necessary to determine small parameter shifts under electric fields or those due to magnetostriction in materials such as MnF_2, where shifts of less than 0.01Å between the paramagnetic and antiferromagnetic ordered states are of interest. Here the use of very short neutron wavelengths, with sufficient flux to also allow the use of a small sample crystal, would have the important additional benefit of minimising extinction effects. Many materials are disordered in that the same site is randomly occupied by more than one type of atom, such as hydride/H_2 disorder in some transition metal complexes. Very high Q data could reveal the disorder and provide models for the bonding in these molecules.

Diffuse scattering studies of disorder in crystals

This field includes thermal diffuse scattering and static structural disorder, for example fast ion conductors, disorder in alloys, melting and phase transitions.

Requirements are a wide Q range ($0.3 < Q < 15$ Å^{-1}) if possible, with a Q resolution equivalent to that of the current SXD being adequate. It is necessary to measure *total* scattering, i.e. energy integrated, not just elastic scattering. Using long wavelengths leads to 'distortion' of diffuse scattering by phonon dispersion and so neutrons of wavelength < 1 Å should be used. It may also be necessary to measure Bragg scattering *separately* with high resolution. There is likely to be some interest in using an instrument with these characteristics (Diffuse Scattering Diffractometer, SX-V) also for liquids or glasses with some characteristic orientation (applied field, stress, liquid crystals). The requirements for measurement of magnetic diffuse scattering depend on the type of scattering of interest. If the features are sharp and their Q location is required precisely then longer wavelengths and wider angles can be used. On the other hand, if the features are broad and the interest is in absolute intensity measurements, then the requirements are as for the above non-magnetic measurements. There will also be possibilities for very interesting work with applied fields using two detectors on opposite sides of the instrument (+/- 90°).

Ultra-high resolution single crystal studies

Critical scattering in magnetic or structural systems requires resolutions in the order of 10^{-4} ($\Delta d/d$). This is because the theories used in this area are valid only asymptotically close to T_c when the correlation lengths are very long and data are required over several decades for which the correlation length is longer than several atomic spacings. Good resolution, planned in the Ultra-High Resolution Diffractometer (SX-I) would then enable the line shape to be determined instead of assumed, which would provide a quantitative test of theories. High resolution X-ray measurements have shown discrepancies between theory and the data for some structural systems. Similar measurements are required on magnetic systems and also on structural systems to examine the effect of surfaces on the properties near to phase transitions.

A second area of interest is in incommensurate systems where the structure of the novel soliton phases can be determined from the weak higher harmonic satellites. It is therefore necessary to determine the intensity of these satellites, which can be as low as 10^{-5} of the fundamental peak intensity while being close in wavevector. Good resolution is therefore required to separate these peaks and determine the satellite intensities. These experiments require scans along lines in reciprocal space, which are not necessarily radial lines.

2.3 Molecular Chemistry

It is possible to draw the analogy between the present situation in chemical neutron crystallography and its prospects with a brighter source, to the situation a few years ago when NMR was limited to much lower fields, which limited resolution, but many obvious applications of such higher performance were known.

There is a dearth of instrumentation *anywhere* for medium to high resolution structural studies and it is proposed to construct a Chemistry Single Crystal Diffractometer at ESS (SX-III). The unit cell edge range in question is 15-40Å, the predicted minimum sample size 0.1-0.5mm^3 and a major gain will be a much reduced timescale for any experiment. Whether for a "routine" data collection, one of a series of T/P measurements in the study of phase transitions or real-time 'one-off' problems, this much shorter experimental period will be of paramount importance. Most of the problems in this field have a high H atom content and/or other low atomic number atoms for which rather low precision atomic coordinates and thermal parameters are retrieved from X-ray data. These problems are removed in neutron diffraction, thus providing detailed structure in the *critical* molecular regions.

Sample Size

Using crystal volumes *comparable* to laboratory X-ray experiments allows transferability which is simply not possible now and this will remove current problems in charge, spin and momentum density studies which follow directly from using two (or more) different (sized) samples. The application of X-N experiments to chemical problems has been somewhat superseded by X-X' in recent years, thereby ignoring and not solving the age old problem of mis-match of thermal parameters between the data sets. It is difficult (or impossible) to deconvolute thermal smearing from electron distributions and a faster turnaround on equivalent sized samples for the ESS neutron experiments would encourage a reconsideration of real (but shelved) problems. This complementing of size can be taken further to include gamma-ray diffraction where accurate studies are being made in areas of e.g. magnetostricture in MnF_2. The importance of obtaining very high resolution neutron diffraction structures is obvious in the fields of charge density studies, and the complexity of compounds studied in this area would significantly increase with smaller sample size and shorter experimental time consequent on the higher intensity of ESS.

Solid state reactions and polymorphism

These fields are not currently studied with neutron diffraction but would be easily accommodated on ESS since the experimental time comes within the time scale of the reactions in question, and the rather weak intensities resultant from the changes become more measurable. It should be remarked that there is no other single crystal technique which samples the entire crystal bulk and monitors the atomic motions in the lattice *non-destructively*. The options also exist for varying the sample environment *viz.* temperature and pressure in order to affect the reaction rate under study, in the cases where such external effects are known not to be catastrophic. Certain observed polymorphism has become important in the pharmaceutical industry where one polymorph may be biologically active and its transformed solid state structure is not. *A priori* assays of the tendency to exhibit polymorphism by applying the real time variables which are met under manufacturing and storage conditions are of obvious use in protecting against drug 'failure'.

Non-destructive solid state chemical rearrangements

Topics such as the thermal dyotropic H atom shift have also not yet been studied but will become possible with ESS intensity and instrumentation. This is an obvious example of the unique ability of the neutron to follow accurately the chemical changes which primarily involve proton measurements. We could expand this field to include examples of inorganometallic and biochemical systems as well as those already indicated which are relevant to the drug industry and fundamental organic chemistry.

Exotic hydrogen

There are very few confirmed examples in the field of co-ordinated molecular dihydrogen in high coordination transition metal polyhydrides $[ML_xH_y]$ and none yet which support conclusively the claims from solution NMR observations of fluxional η^2-$(H_2) \Leftrightarrow \eta^1$-(2H) behaviour. These complexes are not only of fundamental interest in coordination polyhedral geometries, but also in the field of homogeneous catalysis involving C-H activation and metathesis studies. More exotic possibilities include the possibility of adding H_2 *in situ* during an experiment and examining structural/coordination changes consequent upon changing concentrations, partial pressures, etc.

Weak inter- and intra-molecular agostic interactions

These interactions have been discovered relatively recently and cannot be quantified or even identified without recourse to neutron diffraction. The derived accurate molecular parameters are the sort of details which will change the basic bonding concepts appearing in the chemistry textbooks of the future in the same way as the myriad of diagrams in chemistry textbooks today would not be the same without the enormous efforts and consequent impact of chemical crystallography over the last 20-30 years.

It should also be noted that the ability of neutron diffraction to distinguish and identify neighbouring elements is still a *unique* property since the anomalous scattering at an absorption edge using synchrotron radiation up to now is not practically useful in the first and second row elements. There are numerous examples of non-stoichiometric materials whose properties depend on very small % impurities or dopants and with the realistic time scale experiments on ESS, their full characterisation becomes straightforward. In addition to the obvious and known mixed metal oxides, chemical catalytic systems are frequently found to be non-stoichiometric when studied at sufficiently high resolution.

As in the case of powder diffraction, it is impossible to predict the "newest" chemically interesting property-bearing molecule for 20 years hence, but the imagination and skill of the chemist will not fail to yield a 21st Century version of the ceramic superconductors, "Bucky Balls", clathrates, calixaranes or better organic/molecular semi- and super-conductors which together with the 'best' current drug are all enormously dependent on neutron diffraction for their characterisation. It is fair to say that with ESS we can confidently predict advances in structural chemistry comparable to those made with the advent of high field NMR, which have been tremendous.

2.4 Structural Biology

Neutron diffraction in biology addresses three areas, high resolution studies of small systems, low resolution work on large systems and fibre diffraction. Originally the emphasis was on hydrogen atom location, following the trend in small molecule crystallography, while more recently the concept of contrast variation employing H_2O/D_2O is being used to outline disordered parts of the system, which are not easily visible by X-ray analysis.

As for chemistry, there is currently a dearth of instrumentation available and only a limited number of studies have been done. This is also partly due to the limited neutron flux which imposes the use of very large single protein crystals or fibres. Most experiments in this field would use the Biological Molecule Diffractometer (BMX; SX-III), and from comparison with the experiments done at present it should be possible to bring the recording time down to the order of a week. However, this still assumes that the crystal size is around 1 mm^3, an obvious limitation. However, for the cases where neutron diffraction information is essential, the necessary large effort is likely to be invested to attempt to acquire crystals of a convenient size.

High resolution studies

In this regime, which aims at getting data to better than 2 Å d-spacing, one of the goals is of course to locate hydrogen atoms. A large number of the atoms can be exactly predicted from the X-ray structure, which is always a preamble to these studies, but the knowledge of the location of one or two hydrogen atoms can be crucial to the understanding of for example an enzyme mechanism. A good case of this was a study of trypsin, where the location of a hydrogen on a histidine at the active site allowed the choice between several proposed pathways. Hydrogen atoms in water and consequent information about the water network are also very much sought after, and an example of high resolution studies beyond 1 Å is the work on vitamin B_{12}, which is the first study to have been done both at room temperature and 15 K. Larger systems have also been studied, allowing estimates to be made of the fluidity of the solvent water. Along the same line of activity boundaries between protein and solvent is an area of high interest, with studies having been carried out to examine the contact zone between hydrophobic parts of the macromolecule and the adjacent water. The interactions between other solvent molecules have also been studied. Finally, the flexibility of proteins in crystals have been studied by H/D exchange and can be used to complement solution NMR studies on the same material.

In this field the past to some extent outlines the future, given how few successful studies there have been. When the instrument becomes available other systems can be studied with these techniques, either with larger unit cells or in more detail. In recent years molecular dynamics

analysis of biological systems has become an integral part of the toolkit used in understanding the behaviour of these macromolecules. These calculations also involve the solvent structure, and neutron diffraction results could be an important experimental input to this field. An interesting possibility is the opportunity for temperature variation studies, and relation of these to inelastic scattering studies, which have recently indicated a transition to occur at around 180K in myoglobin crystals. It would be of great interest to see whether this change can be observed in the structure of the water, which requires considerably higher performance diffractometers than are presently available.

Low resolution studies and fibres

In recent years there has been an explosive development of X-ray protein crystallography, yielding the three-dimensional structure of an increasing number of macromolecules of ever higher complexity. Many virus structures have been solved, membrane proteins have been investigated, and the structure of protein complexes occur more frequently. However, there remains a significant amount of information which cannot be obtained, especially on the disordered parts, where the overall scattering density is similar to that of the solvent.

In neutron scattering large contrasts can be obtained using specific deuteration or by simply changing the scattering density of the solvent using H_2O/D_2O variation. This has allowed, for example, the identification of the part of a photosynthetic reaction centre which is in contact with the membrane. Phospolipids have been located in a similar manner, for example inside the molecule of lipovitellin, where in this case the host molecule had a molecular weight of 300000 D. RNA/DNA is often also disordered, for example when it is found inside the virus capsid, and this has been used to locate these components inside a number of viruses including tomato bushy stunt virus. In most of these very large molecule studies the resolution has been between 10 and 15 Å, but very low resolution studies of very large systems can be extremely useful in *ab initio* structure analysis, for example in studies of crystals of ribosomes, where at present 30 Å data with different H_2O/D_2O contrasts are being exploited in attempts to start the structure determination.

Fibres can be studied with similar techniques, and again one of the obstacles is the production of sufficiently large samples. Nevertheless, it has been possible to outline to a resolution of about 4 Å the pockets of water bound to DNA, but it would of course be very valuable to go further. The value of an extended resolution range applies of course to other studies. It is generally clear, though, that this new area of low resolution crystallography will me more and more applicable as the complexity of the systems grow, and as more and more systems occur which are not completely crystallographically ordered. Eventually it might also be used to outline the assembly of complexes, where the individual components are known from high resolution studies.

In summary, for the whole field of structural biology, the work performed to date and in the future is seriously flux limited. This is one area in which the sheer flux advantage of ESS over existing facilities will be most noticeable.

2.5 Magnetism

The new ESS source also has some obvious advantages and offers major scientific opportunities in the area of magnetism. Quite apart from the obvious overall advantage of being able to work with smaller samples, it is possible to identify a number of topics where unique new experiments should become feasible. Many of the instrumentation considerations for magnetism are dealt with as part of other sections, and merely the scientific implications will be discussed here.

Small moments and diffuse spatial distribution

Although neutron scattering has played a vital role in gaining insight into high T_c superconductors, their magnetic properties remain difficult to study because of the low moment (S=1/2). Similar remarks could be made about molecular superconductors and organic magnets. Under these circumstances the enhanced flux of ESS coupled with the intrinsically low backgrounds available on a pulsed source will open up these fields to study.

Surface and thin layer magnetic structure

Work on magnetic structure of the surfaces of bulk crystals, as well as on epitaxial magnetic layers, is profoundly hampered by the inherently small number of moments that can be brought into the beam. Similar considerations apply to thin film epitaxial multilayers of potential technological importance. Again enhanced flux is a major advantage in enabling these areas to be explored.

Nuclear magnetism

Studies of nuclear magnetic order, necessarily carried out at very low temperatures, are hampered by beam heating when the beam is continuous. If the peak flux in the proposed source is great enough, data could be taken during a single pulse of neutrons, on a faster timescale than the transfer of energy to the sample. This would make such measurements feasible.

Time-dependent magnetic phenomena

Stroboscopic experiments using pulsed magnetic fields could be used to observe the motion of domain walls, for example. The whole area of stroboscopic measurements are naturally carried out on a pulsed source.

Magnetic structure in very high applied fields and/or pressures

The highest magnetic fields are only accessible in a pulsed mode (ultimately, as with the highest pressures, by implosion methods). Structural data under these constraints would have to be obtained by a 'single shot' or stroboscopic procedure, only feasible with the high peak flux of the new source.

Instrument design implications

- For magnetization density measurements, especially with rare earths, it is desirable to go to $\sin\theta/\lambda$ of at least 1.0 $Å^{-1}$, and preferably beyond, with good counting statistics.
- For isothermal separation of magnetic intensity scattered by antiferromagnetic powders by polarisation analysis, often the incident beam should be polarised to within say $\pm 5^{o}$ of **k**.
- High resolution studies of magnetic propagation vectors in powders are ideally carried out using long wavelength neutrons, from a hydrogen moderator for example.

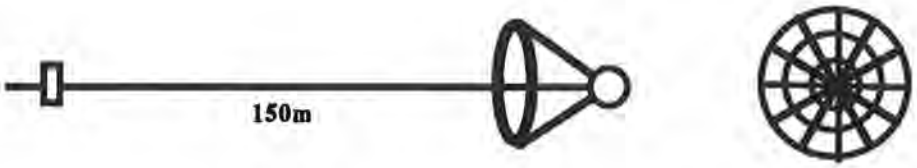
3. Instrumentation for the European Spallation Source

3.1 Powder Diffraction Instruments

Powder diffraction instrumentation has progressed to an advanced state on existing pulsed sources and reactors. This makes it possible to readily access the potential applications and feasibility of new instruments designed for use on an advanced source. Instrument performance can be scaled from existing instruments on IPNS, LANSCE, or ISIS. However, the sizeable increase in flux promises qualitatively new capabilities. The new instruments described in the following narrative and tables illustrate these new capabilities. We first present (Table I) the characteristics and performance of existing instruments at IPNS and ISIS in order to provide a benchmark for present capability. We then present (Table II) the characteristics and performance of new instruments proposed for the ESS. The performance parameters are estimated by scaling from existing instruments and should be viewed as an illustration of what could be achieved rather than a final design. Detailed designs have not as yet been attempted.

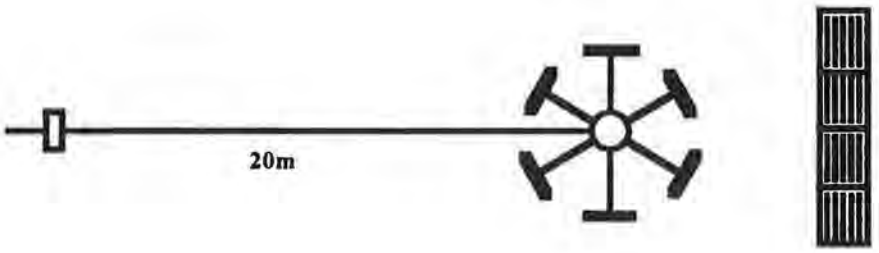
When final instrument designs are specified, it may be found that the same instrument can serve more than one of the functions listed below.

High Resolution Powder Diffractometer (POW-I)



Experience with the HRPD at ISIS has shown that high resolution can be advantageous for many experiments. It has been found that as the capabilities of the instrument have become clear, scientists have improved the quality of their samples and many of these now have intrinsic peak widths comparable to the instrumental resolution. In such cases, structure refinements of an unmatched quality can be achieved, and signal-to-noise ratios are improved enough to allow the study of some hydrogenous samples by powder techniques. Moreover, there is a growing interest in sample characteristics such as line broadening from strain, particle size, stacking faults, or twin boundaries that can be studied with such an instrument. The design goal for the HRPD resolution is $3x10^{-4}$ with a count rate sufficient to collect data over a wide range of d spacings in less than one hour. This performance is comparable to the best that is possible at synchrotron x-ray sources.

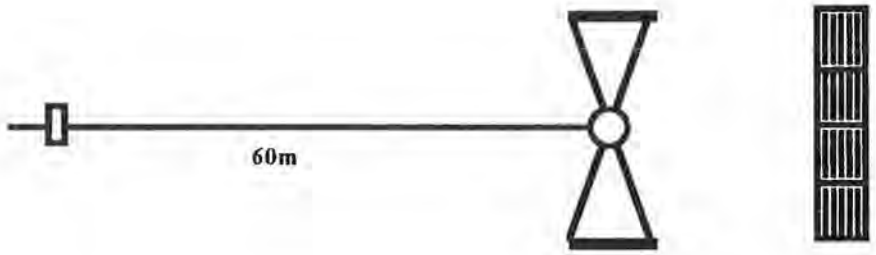
Structure Refinement Powder Diffractometer (POW-II)



As has been the experience on existing sources, the most common use of powder diffraction will probably continue to be structure refinement (by the Rietveld technique). The design goal for this instrument is a nominal resolution of $1x10^{-3}$ with a short data collection time, making it possible to accommodate a large number of samples in an automated way. Thus, samples as small as 100 mm^3 can be accommodated by using longer counting times. The high overall data

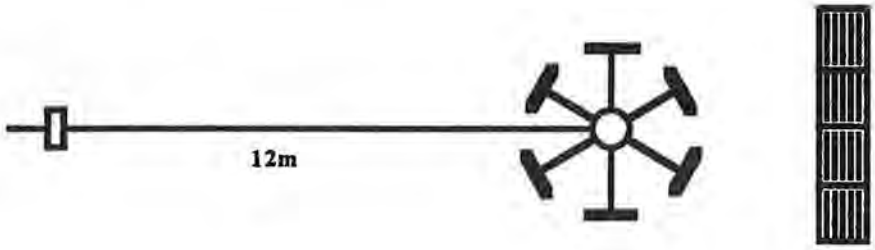
rate is achieved through the use of multiple scattering angles. With careful design, the variation of resolution with scattering angle can be matched to that best suited for structural refinement. Refinements will be based on partially overlapping data from the various scattering angles, providing a refinement that covers a wide Q range and is robust against wavelength-dependent systematic errors. The high count rate also allows the practical use of energy analysis or polarisation analysis.

Special Environment Powder Diffractometer (POW-III)



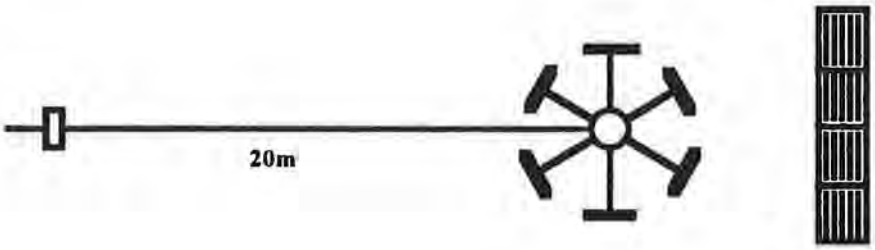
A unique characteristic of the time-of-flight technique is the ability to collect complete data at a single scattering angle. This ability is used to advantage in the study of samples in special environments such as high pressure, high temperature, or in situ chemical reaction vessels. For such studies, the scattering angle that provides the optimum collimation is 90°. The design goal for this instrument is resolution of $1x10^{-3}$ at 90° with counting times of less than one hour. This performance is quite difficult to achieve as it requires maintaining a small angular divergence of the incident and scattered beams. For this reason, such performance is not achieved on any existing source and the full flux of the ESS is needed. An incident flight path of about 60 m will be required. A guide tube will be employed, but, for the highest resolution, the angular divergence in the scattering plane must be restricted to 5 minutes by a Soller collimator. This incident-beam collimation can be relaxed or removed for higher count rates with lower resolution. Large detector areas on a rather long scattered flight path (5m) must be used. In this geometry, there is little degradation of the resolution if the vertical divergence is increased. Thus, this instrument could benefit significantly from improved guide-tube designs (e.g., supermirrors).

Time Resolved Powder Diffractometer (POW-IV)



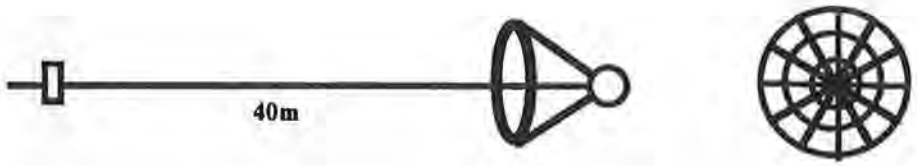
Many real-time experiments have been done on existing neutron sources. With the increase in flux afforded by the ESS, counting times of a few seconds are possible while maintaining resolution of $4x10^{-3}$ and statistics suitable for high-quality Rietveld refinement. Even shorter counting times can be achieved when larger samples are used or Rietveld-quality statistics are not required.

Small Sample Powder Diffractometer (POW-V)



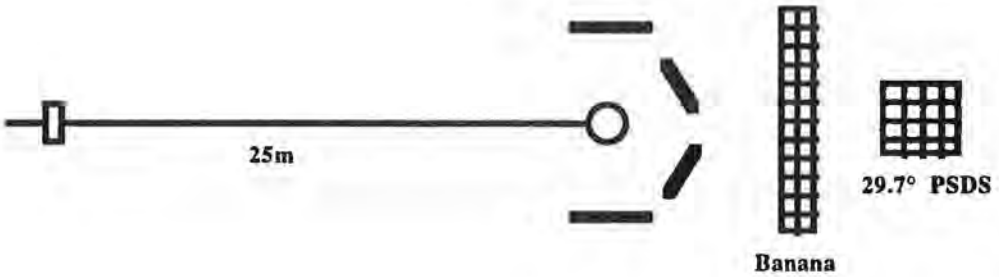
A large number of compounds of interest to the solid state chemistry community are available (initially at least) in only very small quantities. Superconducting Bucky Balls and organic superconductors are two well-known examples. This instrument offers the capability for obtaining high-quality data (i.e., suitable for Rietveld refinement) from 10 mm^3 samples in counting times of 1 hour while still maintaining resolution of $3x10^{-3}$. Clearly, even smaller samples (e.g., <1 mm^3) could be studied by extending the counting time.

Long-Wavelength Powder Diffractometer (POW-VI)



Recent experience has shown that it is straightforward to produce cold neutron beams at pulsed sources. This instrument will be optimised for the use of cold neutrons (wavelengths beyond 2Å) to study magnetic structures and nuclear structures with large unit cells over the range $2Å \leq d \leq 10Å$ while benefiting from the high resolution afforded by back scattering. In many cases the neutron wavelengths used will be beyond the Bragg cut-off of special sample containers (e.g., aluminium pressure vessels) making studies in special sample environments possible. This instrument will view a liquid hydrogen moderator via a cold-neutron guide tube. The instrument is designed for rather high count rates to accommodate the use of polarisation analysis and energy analysis techniques while still maintaining short overall running times. In addition to the obvious applications for magnetic systems, polarisation analysis will be used to reduce the incoherent scattering backgrounds from hydrogenous systems. Energy analysis will be used to reduce inelastic scattering backgrounds. This combination of experimental techniques is expected to make possible the study of whole new classes of materials (with relatively complex structures) by the Rietveld technique with powder samples. The count rate is fast enough for real-time studies in favourable cases (e.g., real-time studies for adsorption in zeolites).

Materials Science Diffractometer (POW-VII)



The power of the neutron in materials science is well established. The nature of neutron diffraction as a *bulk* probe enables the interior of a material to be examined non-destructively, a

feature not available with other scattering techniques. Neutrons are therefore ideally suited to the study of, for example, stress and strain (both macrostrain and microstrain), quantitative analysis of alloys and mixtures and the mapping out of pole figures in texture studies. It is vital in the study of real materials that sufficient flux is available to allow the scanning of real sizes (cm) at high resolution (typically to 1mm step scan across a material). However, it is also necessary to retain resolution to allow accurate peak positions and shapes to be determined. We therefore envisage an instrument of some 25m length on an ambient moderator with banana detectors at +/- 90°, yielding a resolution of some 2.5 10^{-3}. With a short wavelength range required (0.6-1.5Å typically) the instrument could be sited on a 100Hz target.

Energy analysis

One of the challenges that must be overcome as neutron powder diffraction pushes toward more complex structures is that of improving the signal-to-noise ratio. Increasing the resolution will partially achieve this goal, but in addition in many systems, a significant improvement can be gained by being able to discriminate elastic scattering from inelastic scattering. We propose that energy analysis techniques, probably based on the use of correlation choppers, should be developed as a standard technique that could be applied on any of the instruments.

Polarisation analysis

As more attention is given to the study of magnetic structures it is clearly important to develop routine polarisation analysis techniques for the time-of-flight instruments. Polarisation analysis is also expected to be useful for the reduction of incoherent scattering backgrounds from hydrogenous samples, allowing the study of many powder samples not previously possible.

Variable moderator poisoning

Many of the instruments in this list would benefit from the ability to modify the poisoning depth of the moderator to obtain the resolution best suited for the experiment being performed. For example, when an attempt is being made to solve an unknown structure, a sharper peak shape, achieved by poisoning the moderator to obtain a narrower pulse, may offer significant advantages. Clearly, such a feature could negatively impact other instruments viewing the same moderator unless those instruments used only the epithermal part of the spectrum.

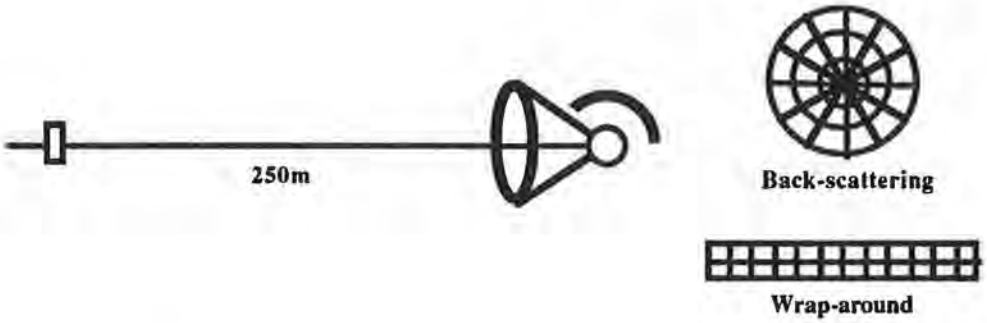
Variable repetition rate

Many of the instruments benefit from the ability to choose the repetition rate to suit the needs of the experiment. Higher repetition rates are used where data over a limited range of d spacing is desired.

3.2 Single crystal instruments

There were several areas identified in which the enhanced flux of ESS would allow very substantial advances in the areas of science we can approach in the field of single crystal diffraction. The instruments presented below exploit variously the pulse structure and time-of-flight techniques in reciprocal space surveying, the very high flux which will at last allow us full access to areas of chemistry and biology hitherto under exploited, and the ability to obtain very high resolution, both in real and reciprocal space.

Ultra-high resolution diffractometer (SX-I)



This instrument provides resolution of $1x10^{-4}$ for use in the study of such things as critical phenomena. It may be used for the study of single crystals or powders. The repetition rate is kept as high as possible (e.g., 50-100 Hz) because often in such studies, often a limited range of reciprocal space is of interest. This approach maintains the highest possible flux.

High resolution physics single crystal diffractometer (SX-II)

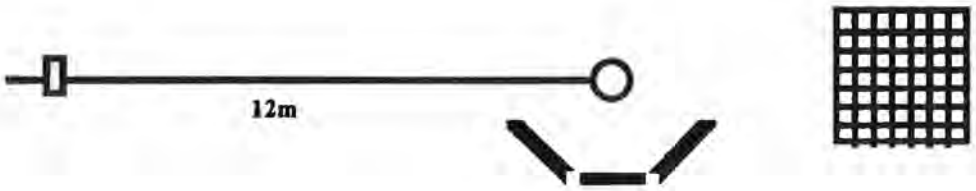
The high flux of useful epithermal neutrons available from the ESS will allow access to extremely high resolution - very high $\sin\theta/\lambda$ - data, and allow the detailed physics of the atomic

structure and thermal motions to be discerned. A single crystal diffractometer situated on this source will yield details on, for example, anharmonic thermal vibrations - and hence on interatomic potentials - which are beyond the scope of current instruments. The high flux of short wavelength neutrons is essential to obtain accurate information in the high resolution region where the scattering is intrinsically weak due to thermal and reflectivity effects.

The instrument would view an ambient moderator to maintain a high flux of warm and epithermal neutrons, and situated at some 15-20m from the moderator to maintain resolution. The detector array would not extend beyond around 150°, as the amount of information available above this is likely to be relatively low and the resolution is not required. This instrument could use a 100Hz target.

An instrument accessing the highest $\sin\theta/\lambda$ values (potentially up to 4 $Å^{-1}$) would enable us to study the following, for example: Anharmonicity and decoupling from other factors; Wavelength dependent effects; TDS; Phase diagram of single crystals; Precise thermal motion (cf X-rays); Order-disorder precursive to phase transitions; Huang scattering.

Chemistry single crystal diffractometer (SX-III)



It will be possible on ESS to collect high quality single crystal diffraction data from moderate sized unit cells, including most of inorganic and organic chemistry, on timescales as little as a day or so for moderate structures, allowing wide exploitation of the precise hydrogen and light atom location available with neutrons. This has potential applications, for example, in the monitoring of solid state reactions and polymorphism. For very small structures, one can envisage the use of the TOF Laue technique in revealing useful structural information on still shorter timescales, thus opening up the intriguing possibility of kinetic studies on single crystals in timescales of an hour or so.

The instrument would view a liquid methane or decoupled hydrogen moderator to achieve the required flux of neutrons in the wavelength range 0.5 - 1 Å which is most useful for this type of application. We would envisage studying materials with unit cell edges in the range up to 40Å, taking around a day or two for a full data set from crystals of volume as little as 0.1 mm^3

ideally. The modest resolution requirements mean a 12-15m instrument with large area PSDs to maximise data rate.

The chemistry instrument on ESS could be envisaged as studying the following among its applications: High precision H and "light" atom location; Solid state reactions and polymorphism; H_2 addition *in situ* and examination of changes; Long range order-disorder transitions using diffuse scattering; Non-destructive solid state chemical rearrangements; Co-ordinated molecular dihydrogen in high co-ordination TM polyhydrides; Non-stoichiometric materials - chemical catalytic systems; Weak inter- and intra- molecular agostic reactions.

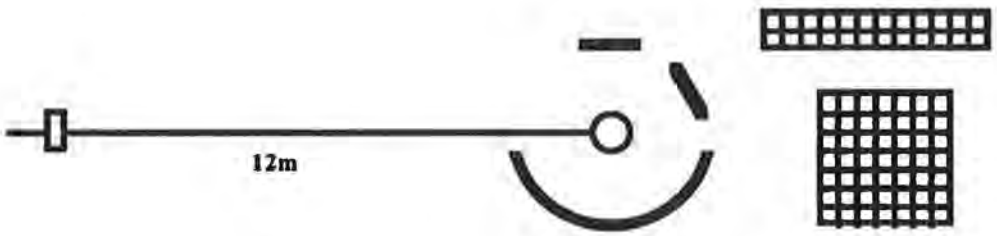
Biological molecule single crystal diffractometer (SX-IV)

Much of large molecule organic and modest size biological single crystal work could be accessible in data collection times of around a week on a cold moderator even from the small crystals which are likely to be available. The power of neutrons in determining solvent water, especially disordered water, and other aspects of protein structure is proven but applications to date have been limited due to the lack of appropriate instrumentation. The combination of high flux and pulse structure available on ESS, enabling the full exploitation of Laue techniques, are a powerful aid towards the expansion of this important field. In addition the availability of the high flux cold neutron beams at the ESS will enable the exciting field of high resolution neutron fibre diffraction to be further explored and allow its extension, for example, to real-time studies of conformation changes in DNA as a function of humidity and the precise correlation of these with changing water structure around the backbone.

This instrument would be on a short flight path to maximise flux, and the use of profile type fitting for intensity extraction from the very large area detectors we would require, again to reduce counting times. The application of the instrument would be in two distinct areas, high resolution studies of materials with cell edges in the range 50-100Å and low resolution work on systems of up to 1000Å cell edge, for which we would chop the <4Å neutrons and extend the frame into the longer wavelength region. Crystal sizes are assumed always to be limited to an absolute maximum of 1 mm^3.

Potential applications include: Access to different parts of proteins: H/D exchange; Water structure; DNA fibres - backbone water; Low resolution studies of partially ordered systems; Multienzyme complexes, viruses, lipoproteins - highlight parts using H/D substitution.

Diffuse scattering diffractometer (SX-V)



The time-of-flight Laue technique is ideal for the study of diffuse scattering (including magnetic diffuse) from single crystal samples, which often occurs in obscure regions of reciprocal space and are very conveniently accessed by the surveying nature of this technique. Such diffuse scattering is often relatively weak, especially at high Q values, and at present relatively long counting times are required for precise results to be obtained even if large single crystals are used. The ESS will allow diffuse scattering to be examined in detail using a TOF Laue instrument from both smaller single crystals (widening the range of systems amenable to study) and also on shorter time scales. For example, in the technologically important fast-ion conductors phase changes are frequently accompanied by the appearance of coherent diffuse scattering from defect clusters which are formed and destroyed as ionic mobility changes. Understanding the ionic mobility is obviously fundamental to an understanding of the mechanism of such high conductivity in these materials. Using the high flux from ESS and large single crystals and exploiting the wide range surveying capability mentioned above, the changes in the diffuse scattering pattern from such a material could be followed in periods of, say, several minutes, allowing greater understanding to be gained of the kinetics of the clusters.

The requirements for such an instrument are a wide Q-range coverage (0.03 - 2 Å) and the use of short wavelength neutrons to avoid the problems of phonon dispersion. In addition it is necessary to measure the whole pattern, for which large area PSDs and the Laue technique are ideal. The instrument could be sited on a liquid methane or decoupled hydrogen moderator, at some 12-15m given the relatively modest resolution requirements.

Some areas which would be conveniently studied on such an instrument include: order-disorder phase transitions e.g. oxygen in 214 superconductors; precursive effects prior to

phase transitions - phonon TDS close to Bragg peaks; coherent elastic diffuse scattering away from Bragg peaks, e.g. clusters in non-stoichiometric fast-ion conductors; pre-melting effects; Reverse Monte-Carlo inversion of whole pattern; short range magnetic order.

3.3. Sample Environment

The demands for advanced sample environment at the ESS require the development of new devices which can adequately match the foreseen performance of the source. The devices have to operate in a fast, reliable and reproducible way, offer stability and be user friendly. Only then one can exploit the ESS in an appropriate way and explore new and exciting areas in science. Besides the "classical" demands like high and low temperatures (mK to 3000° C), high pressures, magnetic and electrical fields and various combinations of these, the time structure of the source combined with the intensity allows for new experiments using time-resolved techniques.

High temperatures

Currently temperatures of 2000°C can be reached with furnaces using microwave or radiative heating. The latter have the advantage that the sample can be in air, vacuum or a variety of atmospheres thus enabling chemists to run high-temperature reactions on powder diffractometers. These furnaces can be used for single crystal and powder diffraction experiments with minor modifications due to the actual scattering geometry used. The opening angle is roughly 20°. Temperatures up to 3000° C may be reached by increasing the number of heating sources, by enhancing the reflectivity of the reflecting surfaces or by using arc lamps instead of halogen lamps. While arc lamps provide a much smaller illuminated volume (roughly $1mm^3$ compared to $1cm^3$), the ESS would allow reliable intensities to be measured from such small samples.

Low temperatures

Low temperature devices are at a very advanced stage with recent neutron scattering measurements at nano-Kelvin levels. These measurements are still extremely difficult, with demanding stability requirements. The speed with which they could be done on an ESS source and the fact that no sample movements are required to obtain integrated intensities would allow for more such studies. A dilution cryostat which can reach 200 mK without geometrical restrictions has also recently become available. A significant technological challenge for low temperature devices will be to couple them with other demanding sample environments such as magnetic or electrical fields. Ultimately the limits of low temperature devices will be set by beam heating, and a monochromatic beam may be required for extremely low temperatures.

High Pressure

The advantages of a high intensity pulsed source apply particularly to diffraction at high pressure. Since high pressure is always achieved at the cost of sample volume, a brighter source would allow the use of smaller samples and thus ensure the attainment of higher pressures. The pulsed nature of the beam makes it possible to work at a fixed scattering angle of 90°, where the optimum collimation can be achieved - the maximum illuminated sample volume seen by the detector with no view of illuminated containing materials. A fixed scattering geometry also allows a simple construction of the pressure cell, which helps in the achievement of higher pressures.

New pressure cell designs now make it possible to take samples of ~40mm^3 to 20 GPa (200kbar). These conditions give refinable patterns from reasonably strongly scattering atoms in simple structures in 8 hours. A 10- fold in detector solid angle (currently in progress at ISIS) will reduce this to ~1 hour. On a 30x brighter source it seems reasonable to aim for ~10mm^3 samples at 50 GPa.

For solving new structures, work on weaker scatterers and larger unit cells, there will be a continuing need for high pressure work on samples of around 0.5 - 1.0 cm^3. A goal of at least 10GPa seems reasonable for this sample volume.

At the other extreme, planning for the 21st century should aim eventually to get neutron diffraction to 2-3 megabar (200 -300 GPa) to study, for example, metallic hydrogen. This will be difficult, dangerous and exciting.

Similar goals can be set for single crystal diffraction. Here there is the exciting possibility that the 30x brighter source will make it possible to use a diamond anvil cell for "routine" work to 10 - 15 GPa, with good optical and angular access to the sample.

Possibilities for high temperature work under pressure are rather limited if the whole cell is heated - because this weakens the material of the cell. However, large opposed-anvil devices make it possible to include a furnace within the sample volume and then water cool the anvils to maintain the bulk of the cell at a low enough temperature. In this way sample temperatures of 2000K or so may be attained at 10 - 20 GPa. This is one of the established and well developed uses of so-called multi-anvil devices (MADs). It is not yet known to what extent the new opposed anvil design will take over the P-T field of MADs. Possibly some form of MAD may be required for the most extreme conditions. There are a many geophysical and mineralogical problems which could be tackled in this regime.

Cooling is restricted by the sheer thermal mass of the pressure cell. There is no excessive difficulty in taking a McWhan-type cell to 10K. However such cooling of presses weighing

several tons would clearly not be a practical proposition. The existing 200 ton press at ISIS weighs 50kg and this is perhaps on the boundary between the two regimes - it could be used if it could be cooled once, and then the pressure varied at low temperature. Hence conditions of 20GPa at 1.5K may be flexible. However, it would be more profitable to use the 30x flux gain to work with a smaller sample, and hence a smaller cell, than at present.

For single crystal work it should certainly be possible to work with quite a small cell on an ESS-type source - perhaps a standard diamond-anvil cell. In these circumstances, 20 GPa at 1.5K should then be quite achievable. In addition for single crystals, it will often be necessary to collect data over a wider range of 2theta than the $90° \pm 10°$ utilised in powder diffraction. This will require radial soller slits to collimate out the scattering from the containment materials. It would be advantageous also to make the sample containment from null-scattering material. Single crystal work to above 10GPa will also require the development of facilities for loading with liquefied gas (e.g. He) to act as a hydrostatic medium.

The pulsed nature of the source opens up the prospect of applying synchronised shock waves to small samples to reach more extreme pressure conditions and study shock - induced transitions.

Pressure is of great interest as a variable in the study of magnetic systems. Substantial new science would come from an ability to work to 10GPa and above coupled with low temperatures. ESS may make it possible, at least in some cases, to reach the required P-T in a cell small enough to simplify both T variation and the application of high magnetic fields. In larger devices, it may be necessary to incorporate magnets into the body of the pressure cell.

Chemical reaction cells

The core of chemical science is the study of reactions, but to date neutron fluxes have been inadequate to take structural snap-shots of reactions in progress in any but the most favourable cases. The high flux at ESS will enable diffraction data to be recorded as reactions proceed in the condensed phase, whether from solid-liquid or solid-gas reactants. Most important among the former are electrode reactions in electrochemical cells and batteries. Here the reaction takes place by migration of a proton or metal ion into the cathode as the cell discharges, e.g. MnO_2 in the Leclanche battery. The fixed geometry of the time of flight diffraction method will enable not only structural variation to be monitored, but also depth profiling of the intercalation reaction. The gas-solid interface is of overwhelming importance in catalysis which underpins much of the oil and polymer industries. Increasingly , the reactions being catalysed take place not on the surface of a monolithic solid, but in micropores of nanometric dimensions within the crystal lattice. At present much of the structural work on such catalysis is done with X-rays because of their intensity. Yet to locate hydrocarbons and other organic molecules within pores

(especially at low loadings) the contrast provided by H/D substitution in neutron diffraction makes the latter an inherently more desirable technique .

Light-induced structural changes

There are a series of technologically important materials proposed as future storage materials whose favourable properties are based on light induced structural changes. Such a structural change has been detected in a single crystal experiment on D9 at ILL and the metastable state could be preserved at low temperatures. Diffraction studies like this can only be done with neutrons since x-rays would immediately depopulate the optically excited state. Devices will have to be designed which allow measurements to be made at low temperatures with the possibility of continuous irradiation of the sample . There is also a great potential in studying the kinetics of optical pumping making use of the time structure of a spallation source or of the structural response to laser pulses.

Time-Resolved Studies

At present time-resolved studies on spallation sources and steady state reactors allow measurement of a medium resolution pattern in a couple of minutes. This has led to a tremendous increase in the understanding of chemical reactions, temperature or pressure induced phase transitions and kinetic processes. With the flux expected at the ESS it will be possible to probe phenomena occurring on an even faster time scale. There are two principal ways in which to approach ultra fast time-resolved studies :

Stroboscopic measurements:

Provided one is observing a reversible process the diffracted intensities are monitored against a variable delay time . Using D20 at the ILL it has been shown that in principle reversible processes can be followed down to 15μs. In addition neutron diffraction experiments have been performed in a pulsed magnetic field up to 16 Tesla, with measurements being taken every 2 s .

Single-shot measurements:

Many phenomena are non-reversible and the possibility of single shot experiments will open new and exciting fields. The structural responses to shock waves, pulsed magnetic and electric fields and laser pulses and the time dependency after such an event are genuinely new science. Test experiments have been performed which indicate that with the ESS a diffraction pattern might be obtained in a single pulse. Once again the high intensity of ESS makes exploitation of this field possible.

4 Summary

The ESS will provide an unparalleled opportunity to develop our understanding of the atomic structure of materials that range from high T_C superconductors, through zeolites and energy storage materials to macromolecules and proteins. The sheer breadth of the potential research is impressive covering scientific disciplines from physics and chemistry to engineering, geology and biology. However, the impact of the ESS with its increased brightness and its diversity of characteristics which permits optimisation of instrumentation for each discipline will lead to a wide ranging impact in both technologically relevant and academically challenging materials research. The need to understand the detailed atomic architecture underpins all materials research, and the nature of the interaction of the neutron with matter permits detailed quantitative comparison with theory. This will continue to be of indisputable importance in the future study of atomic and molecular structure. The massive increase in brightness that ESS will have over existing facilities makes it ideally matched to the diversity of materials research challenges which await us. ESS will be an invaluable part of the European materials research armoury for the 21st century.

	High Resolution Physics	Chemistry 20-30 Å	Biology Hi & Low Res 50-1000 Å	Materials Science	Diffuse Scattering
Range of $\sin\theta/\lambda$	to 2 $Å^{-1}$ or 4 $Å^{-1}$	to 1 $Å^{-1}$	to 0.4 $Å^{-1}$	to 0.8 $Å^{-1}$ 2.5 10^{-3} $\Delta d/d$	0.05 - 1 $Å^{-1}$
Range of λ of interest	0.1 - 1 Å	0.05 - 5 Å	1.5 - 6 Å	0.6 - 1.5 Å	0.5 - 6 Å
Image size (det resolution)	1 mm	1 mm	1 mm	Fine beam in & out	1 mm
Count times	hours	1-2 days 1 hr (kinetic)	1 week	Hours	hours
Moderator	Ambient	CH_4/DC-H_2	C-H_2	Ambient	CH_4/DC-H_2
Length L_1	15-20 m	12-15 m	7 m	25 m	12-15m
Angular	10-150°	30-160°	10-160°	90° bananas	10-50°
Coverage		small low angle	wraparound	1.5m x 5cm	60-120°
Magnetism	Yes	Yes	No	No	Yes
Diffuse	Yes	Yes	No	No	Yes
Frequency	high 100 Hz	50 Hz	50 Hz	100 Hz	50 Hz
Sec flight path L_2	0.3 - 1m	0.3 m	0.3 m	1m	0.3 - 1.5 m
Detector Resolution	2 mm	2.5 mm	1 mm	2 mm	2.5 mm
Detector area (one side)	0.5 m^2	0.4 m^2	> 1 m^2		(0.2+0.2) m^2
Detector Elements	1.5 10^5	> 10^5	> 10^6		> 10^5
Sample size (to max)	1-100 mm^3 (- 1000 mm^3)	0.1-1 mm^3 (-10 mm^3)	0.5-1 mm^3	1 mm^3	0.5 - 100 mm^3 (- 1000 mm^3)

Instrument	HRPD (Pow-I)	SRPD (Pow-II)	SEPD (Pow-III)	TRPD (Pow-IV)	SSPD (Pow-V)	LWPD (Pow-VI)	MSD (Pow-VII)	UHRD (Pow-VIII)
L_1 (m)	150	30	60	12	20	40	25	250
L_2 (m)	3	2.5, 2, 1	5	1.5, 1, 1	0.6	1.5	1.5	3
2θ	>160°	60°, 90°, >150°	90°	60°, 90°, >150°	60°, 90°, >150°	90°, >150°	70°, 90°, >150°	>170°
Moderator*	P	P	P	A	A	H_2	A	P
Guide	Yes	No	Yes	No	No	Yes	No	Yes
Frequency (Hz)	10	25-50	10-50	50	20-50	10-50	100	50-100
d - range Å	0.3 - 5	0.25 - 7	0.5 - 7	0.25 - 5	0.25 - 7	1 - 20		0.3 - 5
Δ d/d	3×10^{-4}	1.5×10^{-3}	10^{-3}	5×10^{-3}	2×10^{-3}	10^{-3}	2.5×10^{-3}	10^{-4}
ϕ	10^7	3×10^7	4×10^7	10^9	3×10^8	10^8	2×10^8	10^7
V (cm^3)	2.5	5	1	2	10^{-2} - 10^{-1}	2.5	10^{-3} - 1	
Ω	0.4	2	0.5	2	2	1.5	1	0.2
ϕVΩ	3×10^7	1.5×10^9	5×10^7	4×10^9	6×10^6 - 6×10^7	4×10^8	2×10^5 - 2×10^8	3×10^6
l	6 m	5 s	3 m	1 s	10 m - 2 h	10 s	20 s - 6 h	1 h

* P Poisoned Liquid Methane ; A Ambient Water; H_2 Poisoned Liquid Hydrogen

European Spallation Source

Report of the Liquids & Amorphous Materials Working Group

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Report of the Liquids & Amorphous Materials Working Group

1. Introduction

Disordered matter is ubiquitous in the natural and man-made worlds around us. It derives from the interplay between primarily structuring mechanisms, which are the short range forces between atoms and molecules, and the intrinsic disorder which arises from thermal excitations or by quenching a liquid or vapour at rate greater than the time required for an ordered structure to form by natural relaxation processes. This state of matter plays a fundamental role in many physical, chemical and biological processes and reactions and has therefore attracted a major effort to understand its microscopic properties over many decades. Oils and other liquids are used to lubricate and clean machinery, many chemical and almost all biological reactions occur in aqueous solution, disordered polymers and other amorphous structures form the basis for innumerable modern materials, the behaviour of molecules at interfaces is of crucial importance to many industries such as the paint and semiconductor industries; in all these cases the interactions and properties are determined ultimately by the fundamental inter-atomic forces which must be understood if new materials and applications are to be developed.

In the last 25 years or so neutron scattering has been at the forefront of developments in this field because it is able to obtain simultaneously vital *dynamic* (time dependent) and *structural* (time average) information on the microscopic arrangement of atoms. In addition, the ability to perform isotopic (and in the near future spin-specific) labelling means that the local environment at specific atomic species can be probed with remarkable accuracy and detail. For example, the local arrangement of atoms can be probed over a range of spacings from a tenth to hundreds of Angstroms with a resolution which is typically of order 0.1Å, while energy resolution extends from eV down to neV. As a result neutron scattering has solved a number of crucial questions concerning the disordered state of matter, for example the nature of the interatomic forces and many body correlations in simple liquids, the structure and dynamics of water molecules around dissolved ions in solution, the microscopic behaviour of supercooled liquids, structure of amorphous silicon and carbon hydrides which are being used to develop new high strength materials, the behaviour of liquids and gases adsorbed in porous materials and at surfaces, and the partial correlation functions in composite materials, which are vital for verifying the results of computer simulation and which also were an essential input for the theory of electrical conductivity in molten alloys. The power of the technique is further advanced by parallel developments in other fields, for example the use of computer simulation to test models of the interatomic forces.

The purpose of this report is to define a set of neutron scattering instruments for the European Spallation Source to do new science of the disordered state which is not possible on present instruments at existing sources. We have not attempted to design instruments in detail, but to write a set of specifications.

In addition to the new science discussed below, we also want to open up the field of $S(Q,\omega)$ measurements to become a *routine* condensed matter probe for chemists, physicists and biologists alike because the information obtained, with its Q dependence, complements many other techniques such as Raman scattering, infra-red, NMR and ultrasonic absorption

In some cases the proposals of other working groups may overlap with those made here. Irrespective of this overlap, we list our proposals for the sake of clarity and completeness. For quasielastic scattering the overlap was high and this subject was covered by a joint session with the High Resolution Spectroscopy group. Our requirements in that field have been included in their list of instruments.

2. New Science on the European Spallation Source

Brillouin Scattering

The development which may have the most far reaching impact at a new high intensity neutron source will be the ability to map out the dispersion of sound and other collective excitations in liquids and amorphous materials. Although some preliminary work has begun in the case of dense gases this work is *not* possible for most materials at existing facilities even though knowledge of these excitations is a crucial key to the theory of liquids. In addition the development of neutron Brillouin scattering, which should be compared with the past development of SANS in terms of number of applications, will have a major impact on many areas of physics and chemistry. This development has been anticipated for many years but has not been achievable at existing sources. Other subjects which can be studied by this method are:-

- dispersive excitations in glasses;
- anomalous sound modes in complex liquids;
- polymeric and large molecule liquid dynamics;
- molecular dynamics approaching critical points, e.g. phase separation and magnetic ordering;

- time dependent systems too fast for real time SANS investigations and small angle spin echo spectroscopy;
- large scale time dependent orientational correlations in molecular liquids;
- transition from hydrodynamic to kinetic behaviour in classical fluids;
- phonon density of states of predominantly incoherent scatterers with large Debye-Waller factors or anharmonicities.

Structure Determination

Diffraction measurements will be transformed by the new facility. This will occur because we will be able to increase the count rate over existing diffractometers by a factor of 100-1000, and also to broaden the dynamic range in Q available on a single diffractometer by up to two orders of magnitude (0.01 to 100 $Å^{-1}$) compared to existing facilities. This will lead to :

- unprecedented resolution of the shape of the first peak in g(r), especially for glasses, where at present extensive guesswork is used to discuss the nature of short-range order in glasses;
- direct information on the nature and extent of intermediate range order in dilute and low concentration mixtures;
- rapid determination of the g(r) and its *derivatives* with respect to temperature, pressure, composition, applied fields (magnetic or electric), isotope or location (x,y) co-ordinate in the sample;
- routine use of anomalous dispersion;
- precise mapping of diffuse scattering in crystalline materials: the measurement of diffuse scattering, whether thermally induced or static, would be a real growth area if a high count rate diffractometer were available, as crystallographers are increasingly realising that valuable information is contained in the scattering between Bragg peaks.

Important new results are urgently needed, for example, on the hydration of bases in DNA, the nature and structure of phase changes in n-component mixtures (where n>2), hydrophobic/hydrophilic effects in solution and S(Q)/g(r) measurements under large transients in pressure.

Contrast Variation

Isotope contrast variation with neutron diffraction has become an *essential* tool for investigating complex liquids and amorphous materials: the variation in scattering length with isotope for some elements is often large and even changes sign in a few cases, notably hydrogen. Anomalous dispersion with X-rays is used in a similar way to isotope substitution with neutrons, but in this case the changes of scattering length are often smaller (of order 20%) and in any case elements below sodium in the periodic table cannot be used for this purpose. Furthermore, the scattering lengths for neutrons are not Q dependent as they are for X-rays and electrons, so neutrons will always provide greater detail for selected elements than is possible by other methods.

The new high flux source will open up the number of isotopes that can be exploited. The scattering contrast is small when the cost of extraction is high (since then only small samples are available) or when the difference in scattering length with isotope is small (e.g. carbon isotopes). Such small contrasts, which are often beyond the capabilities of current spectrometers, will then be measurable on the high count rate instruments proposed here. The same high count rates would also enable us to use the isotope method to extract dynamic partial structure factors for the first time.

The possibilities for complementary anomalous dispersion studies at X-ray and neutron sources will become real at an advanced neutron source. Already work is underway on four component materials, and this will be extended to seven or eight component disordered materials, and so will be applicable to real technological materials.

Large Scale Correlations

The new source will also dramatically increase the range of momentum transfers accessible and this will have a major impact on studies of a broad range of materials. For example *collective excitations* (such as sound modes) frequently give rise to features at low Q (~0.01-0.1 $Å^{-1}$) which up to now *cannot* be measured. Concentration fluctuations in mixtures produce *intermediate range order* which appears as low Q peaks in the structure factor. The behaviour of such peaks with pressure, temperature and concentration is frequently anomalous and a very broad Q range is needed to separate them from the short range order peaks with which they tend to overlap at the lowest Q values of present diffractometers. The Q range available with neutrons is both broader and the data more reliable than is possible with X-rays, a feature which is relatively unimportant for crystalline work but is vital for glasses and molecular liquids. Another broad and expanding field is the use of diffraction to map out static and thermal disorder in crystalline materials such as arises from lattice parameter fluctuations. High count rates are needed to distinguish the weak diffuse scattering from the intense Bragg peaks.

Therefore given the possibility of a much enhanced neutron flux we have identified a number of neutron spectrometers and diffractometers which will give *qualitatively new* data on the microscopic properties of the disordered state and so challenge current theoretical thinking.

In the following sections we describe our main proposals and the new instruments (14 in total) are summarised in the two Tables.

3. Source Characteristics

For studies of liquids and amorphous materials it is important that the target/moderator configuration should be optimised to obtain the best flux. The primary requirement is to obtain a very broad Q range (0.01 to 100 $Å^{-1}$) at neutron energies significantly *above* those needed to cross the sound dispersion curve at a steep angle, in other words the incident neutron energy must be much greater than the energy of possible excitations, and we hope to achieve this for the first time on the ESS. This requirement is important for both inelastic and diffraction work. For the inelastic work it is necessary if collective modes at small Q are to be measured. It is also vital for diffraction measurements since if the path of integration in (Q,ω) space for the diffraction measurements crosses the dispersion curve at too shallow an angle it will give rise to one or more spurious peaks in S(Q) at small Q. This requirement will apply to high and low atomic weight materials alike.

Therefore the target/moderator must *emphasise* the 1/E part of the spectrum and keep the thermal peak at as low an energy as possible. This implies that methane or hydrogen moderators, depending on application, are required but the question of a 1/E moderator still needs to be explored and some research & development is needed here. In addition the technique of single pulse diffraction would require occasional single but intense proton pulses to be delivered to the target.

4. Instrumentation for the ESS

4.1 Inelastic Instruments

The Liquids & Amorphous Materials Working Group identified ten spectrometer types which should be built at the ESS. The full suite is specified in more detail in the accompanying Tables I and II.

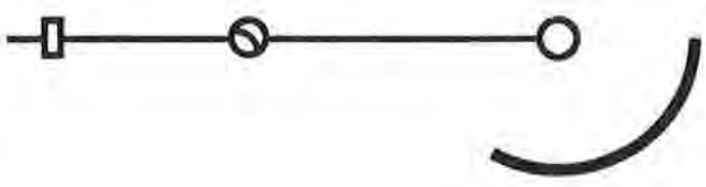
Neutron Brillouin Scattering

An outstanding question in the understanding of disordered materials concerns the propagation of collective atomic vibrations. Unlike crystalline materials where phonon dispersion curves can be mapped out around a number of reciprocal lattice points, there are no Brillouin zones in a disordered material so it is necessary to measure dynamics at all Q and ω. Moreover, the modes are usually heavily damped and disappear quickly with increasing Q. The study of the linewidth and shape gives key information on the physical processes leading to the decay of these collective vibrations.

To date there have been *no* measurements of the dispersion of sound with Q (except in the special case of dense gases where the velocity of sound is much lower than in solids and liquids) to study the transition from the known limiting hydrodynamic (Q = 0) behaviour to kinetic behaviour at finite Q. This lack of experimental information is preventing a comprehensive description of sound dispersion in liquids and is also beyond the range of present computer simulations.

To measure these excitations an instrument with the main detection capability at very low scattering angles (typically 0.2 degrees), and which uses high incident energies, is required. This implies fairly long primary and scattered flight paths are needed. To cover the full range of (Q,ω) space we would need three spectrometers of this type with incident energies of 2-30, 20-100 and 80-1000 meV. The spectrometer in the thermal neutron range will have polarisation analysis as an option.

High Resolution Instrument



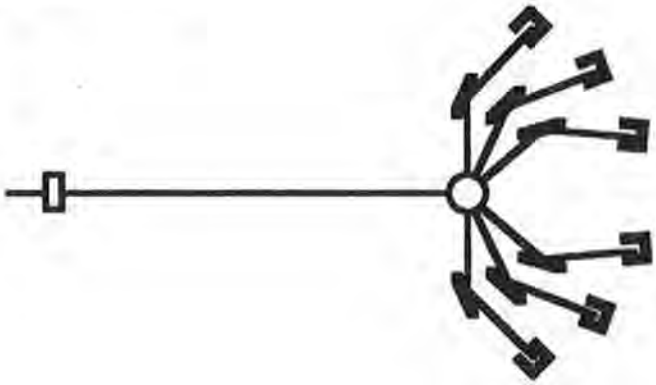
The second instrument is primarily directed at obtaining a detailed and very accurate measure of the dynamic structure factor $S(Q,\omega)$ of liquids and solids in a sufficiently large range of energy and momentum transfers ($Q > 2$ Å^{-1}) and very good peak to background ratio (>2000). Here a resolution $\Delta E_0/E_0 = 0.4\%$ is aimed for and this is *significantly better* than that available at present. The range of applications is very wide: e.g. quantitative determination of the $S(Q,\omega)$ of amorphous solids in a large energy and Q region down to very low energies, where two level states are to be expected; measurement of the phonon density of states of polycrystalline samples with very high accuracy; determination of *partial* $S(Q,\omega)$'s. Similarly the interaction of torsional motions in molecular solids would be measurable by this technique as well as magnetic excitations. At least 2 spectrometers of this type are absolutely necessary; one for incident energies 2-20 meV and another for 20-250 meV.

High Productivity Instrument

The primary goal is to develop for the first time a high productivity $S(Q,\omega)$ instrument for incident energies between 20 and 250 meV . This has already been done for diffraction measurements, but at present dynamic structure factor determinations take at least two days per sample or state condition on *any* existing source. The new instrument will enable these measurements to become routine so that trends with state conditions, such as pressure, temperature, isotope or chemical composition can be mapped out readily. Such information will be vital to a comprehensive understanding of *liquid dynamics* via $S(Q,\omega)$, particularly for molecular systems and solution chemistry, and *density of states of atomic vibrations in amorphous materials* and also of many polycrystals. These measurements are done already with, for example, Raman scattering, Brillouin light scattering and magnetic resonance, but neutrons can provide *complementary* dynamical information as a function of the important momentum transfer variable, independent of energy dependent coupling functions, and also for metallic samples.

The requirement is for at least one (and possibly several with a range of specifications) MARI-type of instrument, although resolution could be slightly relaxed, compared to MARI.

Inverted Geometry Instrument



For the investigation of very small samples or samples under extreme conditions (temperatures up to 3000K, pressures up to 0.2 Mbar) a very high intensity spectrometer is required. As the accessible scattering angles are restricted by the small windows in the sample environment (furnaces, high pressure cells) an inverted geometry instrument with relaxed energy and momentum transfer resolution could be a reasonable choice for the spectrometer. The incident energies in the white beam would be determined by time-of-flight techniques and the scattered neutrons could be analysed by a filter, a crystal analyser or some other method.

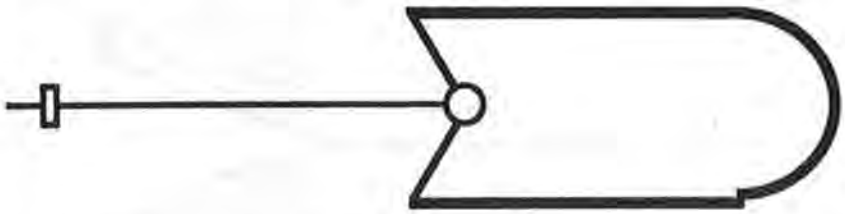
Such an instrument will enable, for the first time, the investigation of the *dynamics* of new materials in small quantities or under extreme conditions, or even time dependent dynamic experiments, not imaginable on present neutron sources. Polarisation analysis is needed as an optional capability.

High Energy Spectrometer

A spectrometer with direct geometry and very high incident energies (1 - 20 eV) is needed for investigations of momentum distributions in quantum liquids and other disordered matter for the study of mass effects in isotope substitution, for the application of anomalous scattering in the measurement of partial $S(Q,\omega)$, and for the measurement of neutron induced chemical reactions.

4.2 Total Scattering Instruments

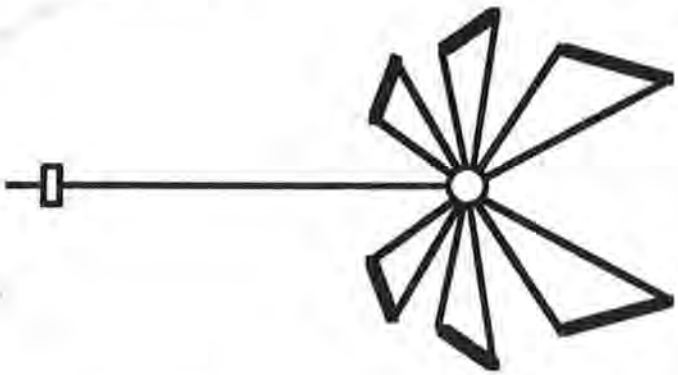
High Resolution Diffractometer



A SANDALS type of instrument with better resolution and lower accessible Q values could be a reasonable solution for this type of instrument. It would obtain S(Q) with high accuracy especially at small Q values and at the same time it would be able to exploit the small recoil corrections which are accessible for light atoms at small scattering angles only (0.3° - 10°).

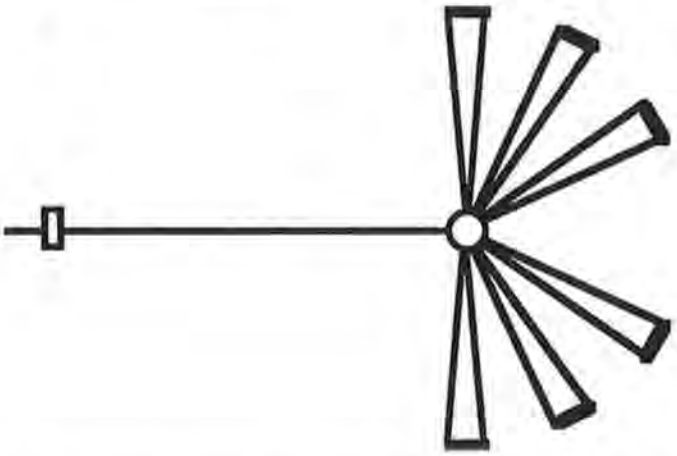
The requirements to achieve this involve a rather long flight path (25 m) and low repetition rate (10 - 20 Hz) in order to achieve the desired angular divergence. Since the resolution of this instrument is dominated by the geometrical constraints, the resolution is not greatly sensitive to the *neutron pulse width* which can be broadened to some extent if this will permit an enhanced 1/E spectrum.

High turnaround diffractometer



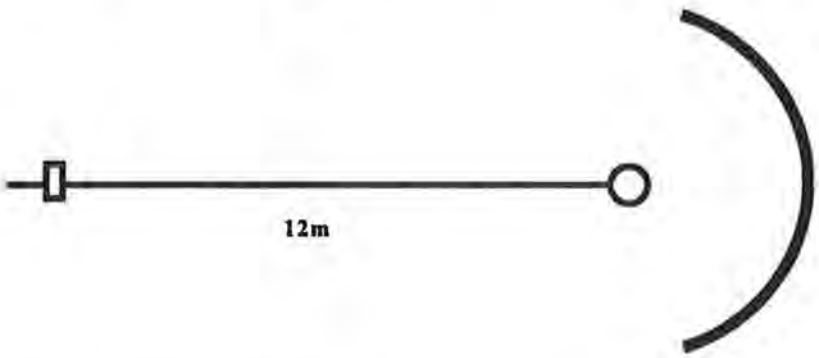
In many cases, and especially at the beginning of the investigation of new disordered materials, a high precision is not necessary but a large number of samples with different compositions or in different thermodynamic states have to be investigated. Such studies require a diffractometer with relaxed resolution but high intensity and a large accessible Q range. Extremely fast data reduction and transformation to g(r) would be made using data from this diffractometer. A very important application would be kinetic (time dependent) structure investigations which are essential for mostly unstable samples (supercooled liquids, glassy materials) to study phase transitions or chemical reactions.

Special environment diffractometer



For the investigation of samples under special environment conditions a diffractometer is needed which eliminates the background from complicated sample environment equipment (such as specialised furnaces or pressure cells) with a highly collimated primary and secondary flight paths utilising 90° scattering geometry. The study of partially oriented samples such as *aligned polymers* under stress or *liquid crystals* will also require a special diffractometer which can measure the structure in two directions simultaneously.

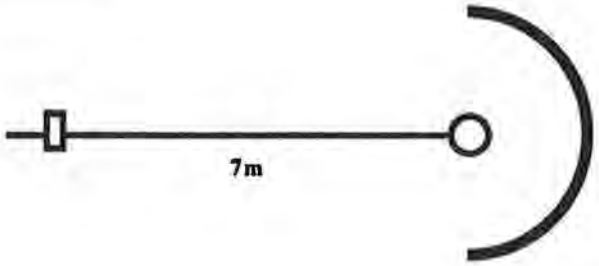
Anomalous neutron scattering diffractometer



A very powerful method to change the scattering cross-section in one and the same sample is the use of anomalous diffraction near the neutron resonance of one of the elements in the sample. In contrast to anomalous X-ray scattering, which will be developed at the third generation synchrotron sources, in neutron anomalous scattering the change in the scattering cross-sections are considerably larger. In order to open up this field higher fluxes of epithermal neutrons than available now are required and so is ideally suited to the new source. It would enable the measurement of partial structure factors associated with one of the elements in the same experiment (each time channel in the time-of-flight spectrum representing one complete diffraction measurement with a given set of values for the neutron scattering lengths).

The important characteristic of this instrument is that the angular coverage of the detectors should be continuous out to about $2\theta = 50°$

Single pulse diffractometer



In many cases one would like to investigate the structure of samples under extreme transients in pressure (shock waves) or magnetic and electric pulses. Such experiments could be done either by synchronising the sample treatment with the incident pulses (if the sample survives) or else one has to obtain all the diffraction information in the one single pulse. The latter would be a specialised application requiring occasional single intense pulses to be delivered to one of the targets.

5 Technical Developments Required

5.1 Detectors

The specification for detectors is stringent and will require some major research and development to determine whether the required attributes can be achieved in the time scale of developing a new source. Specific requirements include:

- large area (up to 2π steradians)
- low cost
- detection thickness 10mm to 15mm - not more
- quiet (500 cts/hr/m2)
- low dead time, perhaps 10-100ns
- high efficiency at 1eV
- very stable 0.01%
- negligible γ-ray sensitivity.

5.2 Sample environment

We would expect to see a dedicated sample environment facility and professional expertise for obtaining high pressure (up to 200 kbar) and temperature (3000K), both in low background

containment. In addition, construction materials must have weak small angle scattering. The requirement to generate pulsed sample environments was also foreseen.

5.3 Polarisation analysis

Polarisation analysis will be needed in a number of experiments (e.g. magnetic glasses, including metallic glasses, and for incoherent/coherent separation) and so would be an integral requirement for at least two of the proposed instruments and perhaps an option for some of the others.

5.4 Data visualisation

Present day liquids and amorphous materials research is hampered by the time taken (typically many days of effort) to obtain a real space image (static or dynamic) of the material under investigation. However computers are continuously being improved and some initial attempts have been made via simulation techniques to both speed up and simplify the process of data visualisation. By the time the new instruments come on line it would be entirely reasonable to expect that much of the data analysis could be performed in real time. This can only happen however with a substantial development effort, which is probably best carried out in association with existing facilities where real data is being accumulated.

6 Summary

Throughout the 3 days spent in session the liquids and amorphous materials working group expressed considerable enthusiasm for the idea of a new high flux pulsed neutron source, and among many proposals were unanimous on the need to instigate neutron Brillouin scattering as a routine technique and the requirement to achieve rapid turnaround diffraction measurements, so that changes as a function of thermodynamic state can be mapped out readily over a wide range of conditions. Progress towards doing this on existing sources already showed that the proposed source would enable measurements to be taken quickly so that the measurements can be repeated readily to check for consistency, provided the appropriate data analysis and visualisation methods are in place . This would increase both the reliability of the method and open it up to a broad range of interests beyond those of neutron scattering experts.

To achieve this goal some areas of research and development were identified, particularly in detector development, sample environment equipment and data analysis software. The ability to perform wide energy band polarisation analysis was also seen to be important, particularly for the inelastic scattering methods. If any way could be found to improve still further on the

neutron flux that could be generated then this would be a useful asset as well. Various requirements were discussed including the possibility of an "all low temperature moderator target" and a "specialised eV region target".

TABLE I Liquids and Amorphous Inelastic Spectrometers (* = polarization analysis option)

	Name	Type	Moderator	Repetition Rate (Hz)	Angular Range	Special Requirements
	Neutron Brillouin Scattering	Chopper			0.2° - 20°	S/B > 2000
1	E_0 = 2 - 30 meV		H_2	20		Area Detector (2 m^2)
2	*E_0 = 20 - 100 meV		CH_4	50		Resolution 2 cm
3	E_0 = 80 - 1000 meV		CH_4	100		Off centre hole for beam
	High Resolution Spectrometer	Chopper			2° - 120°	$\Delta E/E$ = 0.4%
4	E_0 = 2 - 20 meV		H_2	20		S/B > 2000
5	E_0 = 20 - 250 meV		CH_4	50		
	*Phonon Factory	Chopper	CH_4	100	2° - 120°	$\Delta E/E$ = 3%
6	E_0 = 20 - 250 meV					Automatic Sample Changer
	*High Intensity Crystal Analyser	Inverted Geometry	CH_4	50	30°, 60°, 90°	$\Delta E/E$ = 5%
7	E_0 =					S/B > 2000, High T and P
	High Energy Spectrometer	To be determined	High ϕ	200	0.2° - 120o	Low angle detector as in 1 - 3
8	E_0 = 1 - 20 eV		in 1/E region			

TABLE II Liquids and Amorphous Total Scattering Spectrometers

	Name	Type	Moderator	Repetition Rate	Angular Range	Special Requirements
1	High Resolution Diffractometer	SANDALS	CH_4	20	0.1° - 120°	Azimuthal sensitivity
	Q = 0.01 - 100 A^{-1}					Two instruments may be needed
2	High Intensity Diffractometer	Total Scattering	CH_4	50	2° - 120°	Multiple survey measuments
	Q = 0.1 - 100 A^{-1}				(continuous)	Real time measurements
						High activity samples
3	Special Environment Diffractometer	Total Scattering	CH_4	50	30°, 60°, 90°	Beam sizes down to 1 mm^2
	Q = 0.5 - 50 A^{-1}					Very high P, T
4	Anomalous Dispersion Diffractometer	Total Scattering	High ϕ	50	0.1° - 50°	Continuous range of detector
			at 1/E			angles
5	Single Pulse Diffractometer	Transmission +		few/day	0° - 90°	Giant single pulses
		diffraction				2π azimuthal

European Spallation Source

Report of the Large Scale Structures Working Group

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Report of the Large Scale Structures Working Group

1 Introduction

Small angle neutron scattering, SANS, is one of the most widely used neutron scattering techniques, and provides information on the structure of a wide range of systems such as polymers, colloids, alloys and biological materials in the scale range of 20-6000 Å. (References 1 and 2 provide an excellent summary of recent work in the field).

In neutron reflection, the optical properties of the neutron are used to probe the structure of surfaces and interfaces. Thus it is still very much an emerging technique, but in its spectacular growth to date there have been applications in a wide range of fields including surface chemistry, solid films and surface magnetism (3,4).

It is clear that there is now a close relation between SANS and neutron reflection. The properties of many of the systems studied by SANS are dominated by interfaces within the structure. An understanding of the interface can often come from a reflection experiment and provide the important basic information for an explanation of the total structure as determined by SANS. There is therefore considerable complementarity between the two techniques.

1.1 Small Angle Neutron Scattering

The Field Now

The present Q range of a typical experiment is about 10^{-3} - 0.3 $Å^{-1}$ and is suitable for a wide range of complex systems with dimensions in the range 20-6000Å. The unique feature of SANS is that contrast variation may be used to isolate the different structural features of a complex system, e.g. an isolated polymer molecule in a matrix, or the interface between a colloidal particle and its dispersion medium. Examples of current fields are :

- Polymers (melt, solution, composites)
- Amphiphilic systems and aggregation and self assembly (5)
- Colloids - interfacial structure, particle interactions (6)
- Liquid crystal polymers
- Magnetic colloids, ferrofluids (7)

- Biological systems - aggregation problems
- Alloys - defects, clustering, aggregation
- Spin glasses

Many of the above are closely related to technologically important systems, which is reflected in the extensive use of SANS by industrial groups. There has been a convergence of technological and fundamental interest in complex systems and SANS has contributed substantially to understanding, for example, the morphology of amphiphilic systems and polymer conformation. Furthermore, it is clear that SANS will remain an important tool in continuing to improve the understanding of such systems.

The Future

The following developments are envisaged :

- Continued demand at the level of recent years on the types of system listed above.
- A move to more complex systems as the base of our fundamental understanding is broadened, e.g. an increasing tendency to examine anisotropy (8,9).
- A need to extend measurements to higher spatial resolution (i.e. higher Q), e.g. pendant groups on liquid crystal polymers, the intermediate and short range structure of a polymer molecule (for comparison with more advanced predictions), defects in crystalline structures.
- A need to study larger spatial dimensions overlapping with light scattering techniques, e.g. stiffer polymer units in new generation materials with larger correlation lengths, large anisotropy in amphiphilic aggregates. An illustration of the difficulty with light scattering is that refractive index matching in a colloid switches off the Van der Waals interaction, totally changing the nature of the system.
- An increased interest in kinetic studies made possible by increased flux, e.g. the relaxation of polymeric structures during extrusion,
- The use of real space resolution, to study e.g. stress in alloys, magnetic domains, dissipative structures. This is a development which will be made possible by increased flux and improved focusing technology.
- A need to increase the "information space" of an experiment.. In both biological and chemical systems the effects of changes in parameters such as temperature, concentration, and pH are often the most interesting features.

1.2 Reflection

It is not worth separating the present status of the field from future developments because the field is so new and there is a huge potential for growth. As there is a dearth of suitable techniques in this area, neutron reflectometry has rapidly moved to occupy a unique and important position. We make further comments on this in the next section.

Fields where a significant number of measurements have already been made are :

- Adsorption of amphiphiles (10) and polymers (11) at the air/liquid interface
- Insoluble monolayers
- Adsorption at the solid/liquid interface (12)
- Multilayers and thin solid films (13)
- Polymer films and interfaces (miscibility of blends, surface enrichment, kinetics) (14)
- Magnetism (moment profiles in thin films) (15)

All of the above are of considerable technological interest. This is shown by strong industrial demand and involvement in these experiments. Amongst the relevant industrial areas are detergents, photographic film production, magneto-optical recording media and adhesion.

The above examples are far from having achieved maturity. They require substantially more beam time than is available and their investigation will therefore continue well into the future. However, there are additional important areas with a large growth potential. They are :

- Adsorption at the liquid/liquid interface (16)
- Electrochemical interfaces (17)
- Pure liquid surfaces (interfacial thickness near critical temperature, molecular orientation)
- Buried interfaces
- *In situ* ultra high vacuum experiments studying for example, epitaxial growth
- Interactions between different components at more complex interfaces
- Dynamic processes (e.g. flowing liquid surfaces)

The new source will give further possibilities all arising from increase flux. They are :

- Kinematic experiments, e.g. polymer interdiffusion
- Better spatial resolution made possible by reducing the background and extending the Q range.
- More speculative experiments, e.g. off specular reflection, grazing incidence diffraction, and surface inelastic scattering.

1.3 Frequency-domain Experiments

Neutron spin echo on reactor sources has been used successfully to look at polymer motion (reptation) (18,19) and particle fluctuations (20). There is clearly a potentially very interesting field in the study of the "molecular" rheology of complex systems because it is the route to a fundamental understanding of the technologically important macroscopic rheology. The lack of good experimental information has meant that only in one or two areas have there been theoretical developments of significance. As discussed in the context of SANS, the possibilities of contrast variation offer very interesting opportunities in the range normally associated with dynamic light scattering.

1.4 Comparison of Reflection with Other Techniques

The unusual position of neutron reflection makes it important to compare with other surface techniques. X-rays can cover the same Q-range and offer higher flux. However, in specular reflection the inability to manipulate the refractive index limits the possibility of using X-rays to disentangle the elements of a complex interface. In such cases neutron reflection will be more generally applicable. There are many cases, especially in solid films, where X-ray reflection will be superior. For in-plane diffraction (21), X-rays will be supreme except for magnetic structures and the occasional special system.

Tunnelling and atomic force microscopy probe the peripheral layer of atoms, give little depth information and are at best qualitative. They are very effective for the study of roughness and may therefore be complementary to off-specular reflection experiments.

Second harmonic and sum frequency generation are very new techniques. They give information about the orientation of groups in the interface where there is preferential orientation. They give no other spatial information and will be complementary to neutron reflection in some cases.

Fluorescence techniques and nuclear reaction analysis have very low resolution at present. and other techniques that could be used in this area are analytical and destructive.

2 Technique Developments

2.1 Small Angle Neutron Scattering

The current thrust to investigate more complex materials/systems under more sophisticated sample environments has implications for the SANS instrumentation. There is a strong need to make measurements over a wider Q range, and much of recent instrumental development has been directed towards that aim (an aspect particularly suited to pulsed sources). There is undoubtedly a need for lower Q (to overlap with light scattering), and the proposed source characteristics will be particularly beneficial for this. The need to extend to higher Q poses other problems which the increased flux of ESS will be help to overcome.

There are complications in the sample dependent background, common to reactors and pulsed sources, which limit the efficacy of SANS, especially at higher Q. These result from inelastic and incoherent scattering. They reduce the signal to noise ratio and introduce uncertainty into the absolute scaling. The high flux of the new source allows the possibility of solving this problem by means of polarisation analysis and Drabkin filters (22).

2.2 Reflection

As previously stated, neutron reflection is still very much an emerging technique, and as such the current instrumentation, which has developed in parallel on both pulsed sources and reactors, is first generation. However, the proposed source offers distinct advantages in increased flux and greater emphasis on cold neutrons, in addition to the advantages of the white beam time of flight method. The proposed instrumentation in neutron reflection capitalises upon these advantages, and will offer unrivalled opportunities for the future in this area of great importance.

Improvement of the spatial resolution by extending measurements to higher Q is limited entirely by the sample dependent background. This can be solved exactly as for small angle scattering above, but is easier to implement.

A pulsed source offers a distinct advantage for reflection from liquid samples by offering a wide Q range at fixed geometry. (23).

In some areas of study there are definite advantages in reducing the illuminated area of surface, e.g. the solid/liquid interface, where greater homogeneity of surface usually results from measuring over a smaller area. For buried interfaces, e.g. liquid/liquid, there is a parallel advantage in reducing the attenuation by decreasing the length of the sample. A combination of high flux and focusing can be used to solve this problem.

Possible ways of specifically utilising the pulse structure are not clear to us at present. One might imagine pulsing some kind of molecular rearrangement at an interface and this would have consequences for the instrument design.

3 Instrumentation for the European Spallation Source

3.1 Small Angle Neutron Scattering.

The instrumentation for SANS and neutron reflection are summarised in **Table 1** and described in more detail in the following text.

We identify five instrument types :

a) Flagship SANS

The purpose of this instrument is to do SANS over a wide range of Q on isotropic or anisotropic systems with the possibility of more accurate absolute scaling than hitherto possible. This would cover the Q range 10^{-3}-1Å^{-1} with a $\Delta Q/Q$ of the order of 10%. The Q range would be achieved by using a set of multidetectors at fixed distances from the sample. Incoherent background would be eliminated by polarisation analysis and elastic scattering selected by a Drabkin filter. The instrument would view a liquid H_2 moderator in flux-trap geometry operating to 10 Hz. The total flight path would be in the region of 20-40m. A backscattering multidetector might also be included. A T_0 chopper and/or a bender would be incorporated in the incident beam.

The main R&D problems are the dynamic range of the detectors (presumed to be based on the ANS solid state detector development), the polarisation analysis of the white beam, and the use of Drabkin filters for energy discrimination.

b) Very low Q/anisotropic samples

The purpose of this instrument is to study anisotropic samples into a Q range which overlaps with light scattering.

It would cover a Q range of 0.01 to 10^{-4} Å-1 with a resolution $\Delta Q/Q$ of 10%. Many of the basic features would be as outlined above, except that focusing would be used to achieve the required Q_{min}. This requires major development in the area of focusing mirrors and so leaves uncertainty in parameters such as flight path.

c) Very low Q/Isotropic samples

This is similar to (b), except that the restriction to isotropic samples permits the use of a double crystal diffractometer to achieve a Q range of 10^{-3}-10^{-5} $Å^{-1}$. This instrument takes advantage of the large mean flux, but makes no use of the pulse structure and is based on a well proven design.

d) High Flux

The purpose of having a high flux instrument is primarily for kinetic studies. The basic design would be as (a), but would have a smaller Q range of 0.01-5$Å^{-1}$, with similar resolution, the higher flux would be achieved by using a shorter sample detector distance and possibly viewing a coupled H_2 moderator at 50 Hz.

e) Small Sample Size

There is an important need for examining small regions of a sample. This can only be achieved by strong focusing typically to areas of the order of 1 mm^2. This is anticipated to be a major development which could consist of capillary microguide focusing onto the sample, following by capillary microguides to a multidetector. A further gain might be possible by viewing more than one part of the sample simultaneously. This instrument depends heavily on the development of focusing devices.

3.2 Reflection

Main Instruments

There are only two conflicts in reflectometry, between high and low resolution and between vertical and horizontal sample geometry. Low resolution is for experiments that require high flux for higher Q measurements, e.g. the thinner monolayers occurring at a liquid surface. Samples that require particular geometries are draining liquids, solid/liquid interface (both vertical) and liquid samples (horizontal).

Low resolution instruments would view a coupled H_2 moderator, high resolution a decoupled H_2 moderator. All instruments would incorporate a To chopper, polarisation analysis and a Drabkin filter. There is some uncertainty in the optimal repetition rate. 10 Hz will certainly be satisfactory and we should know within a few months (by doing tests on CRISP) whether the frame overlap neutrons can be utilised so that 50 Hz would become preferable. All machines would use 2-D multidetectors and have the possibility of incorporating focusing. The horizontal geometry machines would preferably have collimation with a variable tilt over the range 0-1.5°.

The combination of vertical and horizontal geometry and low and high resolution gives four basic instruments suitable for all the fields of study anticipated earlier. The Q range would be 10^{-3}-0.5Å^{-1} with a reflectivity range down to 10^{-7}.

Reflection: Evanescent Wave

In-plane neutron diffraction with a grazing incident beam is unlikely to be competitive with X-rays, except in magnetism and other special cases. There is a proven instrument EVA for this type of work at the ILL. It would be desirable in a complete suite of instruments to have one on the ESS. The white beam method may offer some advantages.

Reflection: Off-Specular

Strong off-specular effects have been observed from solid surfaces (24). They are obviously related to lateral inhomogeneity and are not in general understood. The combination of this lack of understanding with the extremely asymmetric resolution function in reflection makes it impossible at present to provide a specification for an off-specular reflectometer. However, it is our feeling that this is likely to be an important and interesting area.

Frequency-domain Instrument

The interesting Q and ω ranges are 10^{-1}-10^{-4} Å^{-1} or less and 2 neV to 10^{-2} neV or less. This would involve a major development of the time-of-flight neutron spin echo method (25), and further specification is impossible at this stage. However, this area will be discussed in more detail in the section on high resolution inelastic studies.

3.3 Summary of Development Areas

The instrument suite specified for SANS and neutron reflection indicates a number of areas where important developments are required. The dynamic range of multidetectors is a major requirement for SANS and reflection; the ANS development of solid state detectors looks a particularly promising route for SANS.

Efficient polarisation of white neutron beams (2-20Å) will require further developments of polarising supermirrors, and optical components/assemblies. For the incorporation of polarisation analysis and for the use of Drabkin filters energy discriminators over wide wavelength ranges in both SANS and reflection will be required. There will be a need in both SANS and reflection for the development of focusing devices/mirrors, and capillary microguides (predominantly for SANS).

The extreme Q and ω range requested for dynamic studies will require a major development of the time of flight neutron spin echo method.

4 Summary

The field of "soft condensed matter" is an increasingly important area of research activity, and neutron reflection and SANS are techniques which will play a central role in its wider understanding. Research areas, of relevance to both techniques and which will extend well into the next two decades, have been identified. The instrumentation, and its necessary developments to achieve this research and to fully exploit the vast potential of the proposed source, have been identified.

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INSTRUMENTATION for LARGE SCALE STRUCTURES

	Moderator	Hz	FP (m)	Features
Wide Q Range Flagship SANS	DH	10	20-40	PA, Drabkin
Very Low Q Anisotropic Sans	CH	10	40	
Very Low Q Isotropic SANS (Double Xtal)	CH	50	15	Double Xtal
High Flux SANS	CH	50	15	
Small Sample SANS	DH	10	20-40	
High Resolution Reflectometer (Horizontal)	DH	10	15-20	PA, Drabkin
High Resolution Reflectometer (Vertical)	DH	10	15-20	
High Intensity Reflectometer (Horizontal)	CH	10	15	
High Intensity Reflectometer (Vertical)	CH	10	15	
Evanescent Wave Reflectometer	DH	10	15-20	
Off-Specular Reflectometer	DH	10	15-20	

European Spallation Source

Report of the Excitations Working Group

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Report of the Excitations Working Group

1. Introduction

Over the last thirty years, neutron spectroscopy has transformed our understanding of *solids and liquids*. The wavelength and energy of thermal neutrons are comparable to the characteristic length and energy scales in condensed matter. The Born approximation is well satisfied and the scattering cross-section can be determined simultaneously as a function of both the energy (ε) and wavevector ($\mathbf{Q}$) transfer from the neutron to the sample. This cross-section is then directly proportional on an atomic scale to the nuclear and magnetic correlation functions of the sample. This explains why neutron scattering has been of such fundamental importance in the investigation of both atomic motion and magnetic dynamics. The technique is especially powerful when applied to single-crystal samples, in which wavevector transfers correspond to points in the reciprocal space of the sample, since it makes possible the tracking of coherent excitation processes throughout the Brillouin zone. Complete phonon dispersion relations are now routinely measured in complex crystal structures such as molecular solids and layered perovskites. Such measurements have elucidated the mechanisms of structural phase transitions (e.g. soft modes, martensitic transitions), the nature of electron-phonon couplings in superconductors and the dynamics of quantum fluids. Neutron scattering has revealed for example coherent spin waves in transition metal magnets, diffusive spin fluctuations in weak itinerant magnets, RKKY exchange couplings in rare earth metals, antiferromagnetic correlations in heavy fermion compounds and energy gaps in the spin excitations of high-temperature superconductors. Neutron scattering provides direct measurement of the wavevector dependence of the exchange coupling in magnetic systems and is a valuable test of statistical physics theories in the realm of critical phenomena, low-dimensional magnets and incommensurable systems.

Neutron scattering has not yet inpinged on some important areas of solid state physics. These include mid-infrared physics, the study of organic metals and semiconductor science. We will show below that it is our view that with the ESS neutron scattering will make major contributions to many such new areas as well as to the traditional topics.

The success of single-crystal neutron spectroscopy is largely due to the development of the *triple-axis spectrometer (TAS)*. The principles of its design have remained unchanged since it was first used by Brockhouse in 1955 to measure phonon dispersion relations. However, major improvements in the associated technology, such as focussing monochromators,

Rutherford Soller collimators, "Tanzboden" techniques, polarization analysis and fully automated scans and data storage have greatly enlarged the experimental possibilities. Every reactor neutron centre of moderate flux has at least one TAS, and usually several; the high flux reactor at the ILL has six, differing from each other only in their optimized energy range and their special auxiliary equipment (*e.g.* polarized neutron optics).

The *flexibility* of the spectrometer is its major advantage. The TAS allows the step-by-step measurement of discrete points in $(\mathbf{Q},\varepsilon)$ space, so that scans may be made either as a function of $\mathbf{Q}$, *i.e.* constant-ε scans, or as a function of ε, *i.e.* constant-$\mathbf{Q}$ scans. Since the resolution can be tailored by selecting the (fixed) incident or scattered energies and/or the collimation, the experimenter has considerable control in optimizing the scans for the problem under investigation, whether it is low-energy critical scattering or high-energy spin waves. Attempts to modify the design, for example by installing a multi-angle analysing system, which could measure a range of energy transfers simultaneously, have not been successful because of increased backgrounds with little effective gain in useful signal. At reactor centres at least, the TAS is still unchallenged in single-crystal neutron spectroscopy. It is the view of the Excitations Working Group that the Next Generation Pulsed Source will provide an opportunity to make significant improvements in the methods of performing single-crystal neutron spectroscopy. Features which will contribute to this development are discussed below. However, we also believe that there will still be a place for TAS type instruments on a source with the characteristics of the ESS.

- *Time-of-flight spectrometers* employing multi-detectors provide a much greater volume of data. The TAS technique is optimized for the "surgical strike" which homes in on a small region of $(\mathbf{Q},\varepsilon)$-space to the exclusion of the remainder. Although there are some problems for which such an approach may be justified (*e.g.* critical scattering), the inherent danger is that other important features of the scattering will be overlooked. For example, in "soft mode"-induced phase transitions, there may be significant modifications of the phonon spectrum away from the ordering wavevector itself. Until recently, it has been assumed that the problems of analysing such large data arrays would be prohibitive, but the dramatic improvements in both computational power and graphical displays have weakened the force of this argument. It is therefore timely to begin a software development programme to explore practical techniques to manage the huge data output.

- There is increasing interest in systems which do not have well defined dispersion surfaces but where strong interactions *broaden* the excitation spectrum. By definition, this entails measuring the scattering function $S(\mathbf{Q},\varepsilon)$ over a wide $(\mathbf{Q},\varepsilon)$-range. This leads to two measuring criteria.

- The scattered intensity must be reliably normalized over the entire range.

- The background needs to be well-established over the range of the signal. The techniques of normalizing spectra from TOF-spectrometers are already more developed than on TAS instruments and backgrounds, which are normally slowly varying in Q, may be estimated more easily on "broad survey" spectrometers by extrapolation from regions of low signal.

- The *sharp pulse structure* of the ESS makes high energy resolution relatively easy to achieve. Current TOF spectrometers at ISIS achieve $\Delta E_i/E_i \sim 1.5\%$ routinely (incident energy E_i) and even better resolutions are anticipated on ESS through the use of longer flight paths. In contrast, the TAS has rather modest resolution, typically ~ 5% $\Delta E_i/E_i$, and significant improvements through tighter collimation or by employing backscattering geometry are only possible with a substantial loss of intensity. However, the flexibility of the TAS often permits excellent energy resolution by the use of phase space focussing.

- There is an ever present danger of *spurious scattering* processes contaminating the "true" signal in TAS scans. These usually arise from higher-order Bragg scattering from the monochromator and/or analyser combined with incoherent scattering off the sample. Although crystal monochromators and/or analysers may be used in TOF machines, the problem of spurious processes is considerably reduced because they are either measured in different regions of the time frame, eliminated by intermediate choppers, or identified by their time stamp.

- The *intrinsic backgrounds* of spectrometers on pulsed sources are significantly better than on reactor sources. This is because the fast neutrons, which are most difficult to shield, are not produced between the pulses when the lower energy neutrons are being measured. Furthermore, fixed geometry TOF machines have fully evacuated flight paths, eliminating air scattering and allowing much smaller scattering angles to be used.

- In *low-dimensional samples*, large gains in effective solid angle are possible by exploiting the higher symmetry of $S(\mathbf{Q},\varepsilon)$ when, for example, the incident neutron wavevector k_i is aligned parallel to the appropriate symmetry axis. This allows many detectors measuring equivalent regions of $(\mathbf{Q},\varepsilon)$-space to be grouped together. Neutron spectroscopy on polycrystalline materials is already well-established on pulsed sources and the advantages, over similar reactor spectrometers, of high resolution, low background and high energy transfers have already been demonstrated.

- Pulsed sources are characterized by an intense flux of *epithermal neutrons* i.e. with energies in the range of a few hundred meV to a few eV. This has already led to significant extensions in the energy range of both polycrystalline and single crystal

neutron spectroscopy. Intermultiplet transitions in rare earth metals have been measured with energy transfers of nearly 2 eV while the spin wave excitations in iron have been measured to the zone boundary at about 500 meV. More generally, neutrons will access phenomena which have hitherto been the sole realm of optical studies confined to $Q < 0.001 Å^{-1}$ and of electron spectroscopy which is primarily surface sensitive.

Pulsed source neutron single-crystal spectroscopy is still in its infancy, but experiments so far performed at ISIS have already shown that the potential advantages over reactor techniques summarized above will be realized on the ESS.

In this introduction, we have concentrated on aspects of single crystal measurements as these types of experiment provide the most detailed information and present one of the biggest challenges to pulsed sources. Inelastic scattering experiments on polycrystalline samples and isotropic systems such as quantum fluids are also important and may be performed with relative ease on a pulsed source. Studies of dispersionless modes, of $S(Q,\varepsilon)$ and $\chi''(Q,\varepsilon)$ over an extended dynamical range often provide important information which cannot be obtained in any other way. The Working Group concluded that the potential impact that the ESS would have in these areas was also substantial.

In section 2, we discuss some of the new scientific fields that may be opened up by the increase in source brightness. Section 3 discusses the general features of spectrometers based on current experience at ISIS and other pulsed neutron sources while section 4 outlines the required specifications of an instrument suite designed to meet the new scientific challenges. In section 5, the R&D requirement raised by these designs are summarised.

2. New Science on the ESS

It is anticipated that the flux of spectrometers exploiting the tight pulses of the ESS will be a factor of 30 greater than on ISIS. Similarly, because of the time structure, we expect gains in experimental efficiency ranging up to two orders of magnitude with respect to the current ILL instruments. That such an increase will inevitably widen the scope of science achievable with neutrons can be seen by extrapolation from the already vigorous scientific programmes on existing sources. The extra intensity should allow :

- the measurements of *weaker cross sections* than the present limit of about 1 mb. This will enable measurements of, for example, electronic transitions and very dilute samples.

- the use of *smaller samples*. The recent history of materials science has shown that neutron investigations of new compounds are vital before large quantities are available (*e.g.* high-T_c superconductors, C_{60}).

- *parametric studies e.g.* as a function of temperature, magnetic field, alloy composition *etc.* Such investigations can transform the value of the neutron spectra but are often impossible given current intensity limitations.

- *polarization analysis.* The flux penalty of polarization analysis is usually too great for it to be used in general even though it eliminates ambiguity in separating magnetic and nuclear scattering. In the past, the intensity loss has been compensated by a severe reduction in the energy resolution. On the ESS, with its very bright pulses, such compromises will not be necessary as long as efficient beam polarizers are available. A development programme to achieve this goal is urgently called for.

- significant improvement of the *resolution* in both energy andwavevector. This will extend the range of investigations of lineshapes, interactions between excitations, and so on.

- the time structure of the neutron beam with *pulsed sample environments* to be exploited and to perform *time dependent* studies.

Principal among the classical inelastic scattering techniques is the study of well-defined low energy *propagating excitations* in crystalline materials. Whether spin waves or phonons, their behaviour affords a more complete picture of interatomic forces than any other technique. It is impossible to overestimate the continuing importance of these traditional studies. As long as new materials with unusual/interesting properties continue to be synthesized, such inquiries will remain essential techniques for their characterization and study in the same way as NMR is in chemistry. Recent examples include high-T_c superconductors, the fullerenes (*e.g.* C_{60}) and improved permanent magnets (*e.g.* $Nd_2Fe_{14}B$). The common theme among all these materials is their structural complexity. These and other developments within the past decade teach us that we must be prepared to understand the detailed microscopic behaviour of materials with hundreds of atoms in a unit cell. This structural complexity is expressed in the excitation spectra as an increased number of degrees of freedom and a high density of energy levels in $(\mathbf{Q},\varepsilon)$ space, which presents a challenge to neutron spectroscopy.

Neutron spectroscopy has become a standard tool for investigating the microscopic properties of solids and liquids, and will remain so in future. It provides unique information and is indispensable for our understanding of new materials as well as many body physics. Thus the scientific programme on the ESS will be guided by developments in materials science and physics that cannot be predicted at present. Nevertheless, in the following sections, *some*

areas that are likely to benefit especially from the enhanced flux , the improved resolution and the time structure of the neutron beam of the ESS are highlighted to give an indication of the new fields that may be opened up. Obviously not all possible topics are considered below. Also history has demonstrated that predicitions of this kind are always incomplete and usually inaccurate.

2.1 High Intensity Applications

2.1.1 Electronic Excitations

The neutron cross section is proportional to the dynamic magnetic susceptibility $\chi''(\mathbf{Q},\varepsilon)$ which is the fundamental function describing the spatial and temporal correlations among the magnetic moments of electrons. As a probe of the *electronic structure*, it is complementary to X-ray spectroscopy (photo-emission and inverse photo-emission) which measures the charge, as opposed to the spin, susceptibility of the electrons. However, neutron scattering has the advantage that the perturbation to the system is much weaker. This complementarity has not yet been realized in practice because the energy scale has been beyond the reach of conventional neutron spectrometers. The studies of intermultiplet excitations in rare-earth metals by neutron spectroscopy have shown that the accessible energy range may be expanded to several eV. The extra intensity of the ESS combined with the substantial flux of epithermal neutrons should help to fulfil this early promise. While magnetic X-ray scattering holds great promise in the field of diffraction, its cross-section is sufficiently small to make high-resolution inelastic measurements prohibitively difficult, even with the next generation of synchrotron sources.

We present here two important areas in which neutrons may be used to probe the electronic structure.

Strongly Correlated Electron Systems

One of the central problems in condensed matter physics is to understand strongly correlated electron systems. These are systems in which the electrons cannot be adequately described either by localized atomic models, because of strong hybridization or narrow-band formation, or by one-electron band theory, because the *intra-atomic correlations* are too strong. Examples include the transition metal magnets such as Fe, Ni, Cr and Mn, heavy fermion and intermediate valence systems, actinide compounds and high-T_c materials. Theoretical progress has depended on the development of many-body theories based on the Hubbard or Anderson Hamiltonians which are the simplest models capable of describing the basic interactions giving rise to the complex magnetic phenomena observed in these materials. The measurements of $\chi''(\mathbf{Q},\varepsilon)$ over a wide dynamical range presents a number of serious challenges.

- In most cases, single crystals should be studied because of the vector dependence on Q. Polycrystalline studies have given valuable information where the dynamics are dominated by single-ion fluctuations, such as in intermediate valence compounds. However, in most compounds, interatomic correlations must be taken into account even where sharp propagating excitations are not observed.

- The response may well be weak in both ε and Q. This is typical in actinide compounds where well-defined excitations are only observed in small regions of the Brillouin zone. In the remainder, the magnetic response is strongly broadened by hybridization of the 5f-electrons. Another example is the well-known problem of determining the excitation spectrum of transition metals at very high energy where the spin waves interact with single-particle interactions, the "Stoner" excitations, which extend over most of the zone.

- Experimentally, it is extremely important to be able to distinguish the electronic from the vibronic excitations - to do this reliably requires polarization analysis of the scattered neutrons.

A start has been made in these measurements on most of the systems discussed above, but the information and particularly the range of frequency transfer, is extremely limited. A source such as the ESS will allow substantial further progress to be made from increased interaction with theories of the electronic structure. As the measurements are performed and analysed, calculations can be used to establish areas of the (Q,ε) phase space that are of particular interest. The subsequent increase for our understanding of phenomena such as that of high-T_c superconductivity and the critical electronic parameters in the model Hamiltonians are likely to be dramatic.

Single particle excitations

A challenge for high energy transfer neutron scattering is the observation of *single particle excitations* e.g. of electrons across the bandgap in a semiconductor. This would be a very powerful tool for the experimental determination of energy band structures. Although some information on the conduction and valence band structure can be gained by inverse photoemission and angle resolved photoemission spectroscopy, with neutrons unique information on the Q-dependence of the scattering can be obtained.

Calculations of the total magnetic scattering cross-section for electronic excitations have shown the difficulty of such an experiment. The interaction of the neutrons with the electrons is the familiar magnetic one in which the cross section contains spin and orbital parts. In the case of Si and Ge, the band gap is of the order of 1 eV, but the scattering cross section intensity remains negligible until the energy transfer is about 3 eV. To obtain useful information on the

bandstructure it is essential to be able to measure the cross sections up to values of 7 or 8 eV. As a working hypothesis one needs to study energy transfers from about 3 to 8 times the band gap. The increased flux of the ESS might make it possible to measure these very small cross sections.

In most semiconductors the energy gap ranges from 400 meV to 3 eV and above. However, the group of narrow gap semiconductors and a number of solid solutions based upon them have energy gaps in the order of 100meV, an energy range which is easily accessible with a chopper spectrometer on a pulsed source. We may expect progress on such materials using existing pulsed source instruments in preparation for the more difficult experiments possible with the ESS.

2.1.2 Surface Excitations

Magneto-optic thin films containing mixtures of rare-earth and transition metal ions, such as Tb-Fe-Co, are becoming increasingly important in technological applications. These films are used in erasable data-recording disc systems with extremely high storage capacities. Many magneto-optic materials have been investigated, but basic questions about the atomic and magnetic structures and the spin dynamics remain unanswered. Neutron scattering is playing an essential role in these investigations. Reflectometry can provide valuable information on the structure perpendicular to the surface normal, and by the use of polarized beam techniques, can give unique information on the magnetization density. Laser-annealing is being used to improve the lifetime and magnetic properties of these magnetic thin films with neutron measurements before and after the annealing process; future work is planned in which the magnetization will be studied during the anneal. Very high intensity sources are needed for this work. With a source such as the ESS, studies of the *spin dynamics* can be envisaged, leading to a knowledge of the microscopic magnetic interactions. This basic information will lead to much improved performance of those important data-storage devices.

Inelastic surface experiments are bound to be hard but might be possible if a large surface and glancing angle are used. Probably the easiest experiments are measurements of the critical scattering associated with both magnetic and/or structural transitions. Different behaviour and exponents are predicted according to whether the surface favours ordering or not. The theoretical progress is considerable in this area but only very few experiments have been performed. Systems with antiferromagnetic ordering at the surface and ferromagnetic ordering in the bulk should likewise give interesting results. Another area of interest are *phonons localized at interfaces*. They play important roles in semiconductors but there is very little direct experimental information.

2.1.3 Excitations in low-dimensional systems

In dimensions lower than three, detailed theoretical treatment of collective excitations is quite often possible. Concepts untractable in three dimensions can be treated in 2D or 1D. If the models used in the theory can be realized for experimental investigations then a very deep understanding of such collective excitations is possible. Examples where progress has been obtained are the verfication of exact calculations for the 2D Ising model, the discovery of solitons in classical 1D magnets and a new quantum ground state in S=1 chains.

It is clear that such investigations will continue to improve further our understanding of collective phenomena in many body systems. In particular we expect the study of quantum effects to remain a lively field as theory and computer simulation advance. Continuing the exceptionally successful interplay between theory and experiment should yield explanations of the behaviour of frustrated quantum spin models in two dimensions, the Haldane groundstate and the excitations in nonlinear systems. Progress in these fields could form the basis of a better understanding of 3D-systems and unresolved problems like the S=1/2 Heisenberg antiferromagnet on the fcc lattice.

2.2 High Resolution Applications

The extra intensity of ESS can be used to improve the resolution already achieved in the present generation of pulsed neutron spectrometers. This will make it possible amongst other things to separate closely lying dispersion branches, the study of linewidths, either of phonons in order to monitor anharmonicity and electron-phonon couplings, or of crystal field excitations which can probe the local magnetic susceptibility.

2.2.1 Hard Magnetic Materials

Strong permanent (*"hard"*) magnets play an essential part in recent technologically important applications as well as having fascinating magnetic properties. The aim is to understand the magnetic properties on a microscopic scale in order to guide the development of improved magnets in terms of energy density, remanent fields, transition temperatures *etc.* Such improvements are currently achieved by trial and error. $Nd_2Fe_{14}B$ is one of the best available hard magnets with a complicated structure comprising 64 magnetic ions, Nd and Fe, and a transition temperature of 500 K.

The very recently discovered compound $Sm_2Fe_{17}N_x$ (x~2-3) is an extremely promising alternative to the $Nd_2Fe_{14}B$ compound for permanent magnet applications. The high uniaxial magneto-crystalline anisotropy and the technologically very useful high Curie temperature (up to 750K) are very promising starting points for the development of new generation of

permanent magnets. The rather large increase in T_c in the nitride compounds is most likely due to an increase of the Fe-Fe exchange interaction.

The *only* way to determine the relevant microscopic parameters is to measure the dispersion of the magnetic excitations and to compare the results with a spin wave calculation which then leads, in the case of $Nd_2Fe_{14}B$, to values of anisotropy energies of the Nd, the exchange energies between Nd and Nd, Nd and Fe and the different Fe-Fe exchange energies. Due to the large number of modes the measurements have to be performed with very good resolution, preferably employing many detectors simultaneously. Furthermore, the high exchange constants responsible for the high transition temperatures necessitate very energtic neutrons. Such investigations are only just at the edge of feasibility. An essential part of such studies is the separation between the magnons and phonons which have comparable energies and equally complicated dispersion surfaces. This is usually done by measuring in different Brillouin zones with a single crystal. The most direct way of separating magnons from phonons would be the use of polarized neutrons and polarization analysis.

Research on hard magnetic materials forms part of the European Community's CEAM (Concerted European Action on Magnets) programme for the development of advanced magnetic materials.

2.2.2 Quantum Fluids

Much of our knowledge of condensed matter is based on the understanding of interacting Bose and Fermi systems. These theories can best be tested by experiments on *two-dimensional electron systems* and the *two helium fluids ^3He and ^4He*. The latter are both superfluids and one of the most crucial tests of any microsopic theory of these systems is the extent to which the theories can explain neutron scattering data. Examples of such experiments which require more refined techniques are:

- The measurement of the spin dependent cross section in ^3He and ^3He-^4He mixtures. The interacting Fermi liquid theory is essential to the theories of high-T_c superconductors and heavy fermions and one of the simplest magnetic systems is ^3He. These experiments require a polarized beam spectrometer to separate spin-flip scattering from that due to density fluctuations (zero-sound collective modes and quasiparticle-quasihole continuum excitations).

- An experiment requiring very high intensity and good resolution is to study the change in the scattering from ^3He when it becomes superfluid at about 2mK. In principle, this would provide a unique measure of the energy gap and of the order parameter in the superfluid phases. This is clearly an extremely difficult experiment combining extremes of neutron technique and sample environment.

- The measurement of the high Q recoil scattering provides information about the Bose-Einstein (for ^{4}He) and the Fermi-Dirac (for ^{3}He) momentum distributions as well as providing a crucial test for the impulse approximation. At present, the resolution is not good enough and the wavevector transfer not large enough to provide definitive answers, although the development of the electron-volt spectrometer (eVS at ISIS) encourages optimism for future developments at the ESS.

- There has been a great deal of interest in the states of the two-dimensional electron system in a magnetic field because of the discovery of the quantum Hall effect and possibly of the Wigner solid. In principle, neutron scattering can provide information about the structure and excitations of these novel electron phases but the experiments are very difficult because of the small number of electrons involved. The high intensity of the ESS employing polarized beam techniques may make these experiments possible if the semiconductor technology also advances to the point where multilayers of these states can be produced.

2.2.3 Magnetic ions as paramagnetic probes

It is possible to dope dilute quantities of paramagnetic ions, typically rare earth ions, to act as *weakly interacting probes* of the host lattice magnetism. The linewidths of crystal field excitations are proportional to the local dynamic susceptibility at the crystal field energy, in an analogous way to the $1/T_1$ of NMR but at much larger frequency. This susceptibility may be studied as a function of temperature, magnetic field or pressure to give information on the evolution of the host's electronic structure under these conditions. In metallic systems, for example, χ'' is proportional to the electronic density-of-states, so the development of electronic energy gaps with magnetic or superconducting instabilities can be investigated. The high-intensity neutron beams of the ESS may also be used to probe the electronic structure of dilute rare earth ions in which the f-shell is unstable because of extreme local environment effects. For instance, praseodymium doped into palladium by ion implantation becomes intermediate valent although it has a stable f^2 configuration in normal compounds . Inelastic spectrometers on the ESS should be able to monitor the processes by which the f-electron becomes delocalized through their intra-atomic excitation spectra, that is by studying the intermultiplet transitions.

In both these examples, the extra intensity of ESS is required to allow only very dilute quantities of the probe ions to be used. This is essential to minimize interactions between the paramagnetic ions leading to exchange broadening. Resolution is clearly important in linewidth studies, but may also be important in enhancing the signal-to-noise of high energy electronic transitions which will be superimposed on large multiple-scattering backgrounds.

2.2.4 Phase transitions

The importance of neutron and x-ray scattering measurements in this field can be gauged from the fact that a phase transition is simply the *macroscopic manifestation of a change in a correlation function at the atomic scale*. Neutron and x-ray scattering are the only techniques which can measure directly the Fourier transforms of these microscopic correlation functions. From measurements of the static correlation functions (integrated over energy), it is possible to extract the spatial correlations as a function of an applied variable. The limitation to such measurements is the wavevector resolution which in the case of neutrons is of order $0.001Å^{-1}$. A future neutron diffractometer should be capable of a resolution of at least $0.0001Å^{-1}$. Such a resolution is available from synchrotron sources and many exciting experiments on structural phase transitions will be carried out with x-rays in the future. However for materials where light elements and magnetic phenomena are involved neutrons will remain the scattering probe of choice. Future experiments will certainly involve both neutron and x-ray measurements on the same materials, in which case it is absolutely necessary that the measurements are carried out with comparable resolutions. The improved resolution must be available for neutron wavevectors of up to at least 5 $Å^{-1}$ if not higher.

Critical dynamics studies

Inelastic neutron scattering is the only well-developed method for studying the *temporal* as well as the *spatial* correlations associated with a phase transition where the bare length scale is an interatomic distance. As the transition temperature is approached the characteristic times describing the correlation functions become very long. This effect, known as "critical slowing down", manifests itself as a temperature dependent narrowing of the width of the inelastic spectrum. Studies of the critical dynamics of phase transitions have been severely limited because instruments with the appropriate resolution are not available. Although it is currently possible with backscattering spectrometers to obtain μeV resolution this can only be done by sacrifying wavevector resolution. For critical dynamics measurements a spectrometer with energy resolution of μeV order and a wavevector resolution of ~0.001 $Å^{-1}$ is required. Even though the energy resolution may be relaxed compared to that of a backscattering spectrometer the Q resolution requirements will mean that a reasonable count rate could only be achieved with a substantial increase in effective flux.

Electron-phonon couplings

With very few exceptions the study of propagating excitations by inelastic neutron scattering has been carried out with instrumental resolution insufficient to measure *intrinsic excitation linewidths*. The powerful, high energy resolution backscattering and spin-echo techniques, while admirably suited to the study of dispersionless, quasi-elastic scattering have been of little

use in the study of phonon and magnon linewidths. Yet such studies are potentially highly rewarding. Consider the effect of superconductivity on the linewidth of phonons. In a superconductor, the effect of the conduction electrons on the phonon linewidth disappears below T_c if the phonon energy is below that of the superconducting gap. The phonon does not have enough energy to break the electron pairing and the phonon linewidth narrows at that point. These measurements provide unique information about the electron-phonon coupling for individual phonons. Such detailed knowledge of high-temperature superconductors would considerably advance our understanding of these materials. However, because of present resolution limitations, only very strongly coupled superconductors like niobium, and Nb_3Sn have been studied by this method, and even for those materials there remain many unanswered questions. The problem is very challenging because the resolution of the spectrometer must be manipulated to decouple the energy and wavevector along a particular trajectory. It seems that this can be done to a limited extent by combining the TAS and spin-echo principles in a single instrument, but different and perhaps more flexible ways of manipulating $(\mathbf{Q},\varepsilon)$ space are possible with pulsed beams of neutrons. An instrument capable of excellent energy resolution with relatively good momentum resolution, at meV energy transfers, that could be focussed over a range of excitation velocities would revolutionize the study of excitation lifetimes.

Anharmonicity effects

As is well known the density-density response function measured by means of neutron inelastic scattering is characterized by the presence of delta-function features only in ideally harmonic systems. Any *anharmonic interaction* on an atom with its neighbours produces a change in the inelastic response, which broaden and shift the phonon or magnetic peak according to the nature of the interaction. Until now most of the lattice dynamics studies have been performed employing triple-axis spectroscopy and therefore only a limited number of line-shape studies have been done, as point by point measurement precludes the possibility of analysing a large energy range in a reasonable time. The considerable increase in intensity and signal-to-background ratio, together with an improved resolution will open the possibility of analysing the tails of the phonon response function as well as multi-phonon contributions, thus allowing the quantitative study of the phonon self-energy and other aspects of the phonon interactions. Studies of phonon line-shapes deserve much more attention in view of their importance from both a fundamental and an application point of view providing a direct tool to detect the presence of some anomalous behaviour, even when it is not yet fully developed. These considerations apply of course also in the case of magnetic excitations.

2.3 Time dependent measurements and extreme environments

The time structure of the neutron beam has great potential for time resolved studies. These can be divided into measurements of relaxation processes after the sample has been subjected to an external perturbation and experiments under extreme sample environment which can be maintained only for a very short time.

2.3.1. Time dependent investigations

For continuous phase transitions equilibrium is regained very rapidly after a change in an effective variable such as temperature. However, for materials which undergo first order phase transitions there can be significant time delays when the material is quenched through the phase transition before the order associated with the new phase is established. To characterise the behaviour associated with the intermediate states data must be collected in *"time-slices"* as one phase evolves into another phase. These time slices must be small compared to the time taken for one phase to evolve into the other, but long enough to accumulate reasonable counting statistics. In practise this means that only phenomena which take many hours can be studied at present. A difficulty with such measurements on a reactor based diffractometer or triple axis spectrometer is that if a spectrum is measured point by point every data point is taken at a different stage in the evolution. At a pulsed source, with the use of time of flight techniques, a whole spectrum is measured for every pulse. However, at the present pulsed sources the flux is such that a large number of pulses are required to give a reasonable signal. With a time-integrated flux equal to that of the ILL reactor, complete spectra could be taken in the time needed to measure a single data point at the reactor source. Substantially shorter time slices would be possible and phenomena on the scale of a few minutes or less could be studied. In addition to diffraction measurements, inelastic experiments, studying the changes in the phonon, spinwave, crystal-field excitations during the evolution between the two phases would be become feasible.

2.3.2 Extreme environments

The frontiers of physics are generally defined by extreme conditions, which in condensed matter physics correspond to extreme sample environments. Examples include high pressures, very low and very high temperatures and high magnetic fields. Here we will only highlight the case of intense magnetic fields.

Pulsed magnetic fields

Considering the magnetic cross-section and the ability to penetrate support structures and thermal shields, neutrons are well suited for probing high field effects. Currently the maximum steady state magnetic field available on a sample in a neutron scattering experiment

is about 10 Tesla. Superconducting solenoid magnets with fields up to 20T have been constructed, but the usual need to have a split-coil geometry for beam access implies much lower fields at the sample position. In order to reach significantly greater magnetic fields, pulsed techniques will be required. The combination of a *repeating pulsed magnetic field* and a pulsed beam neutron spectrometer has obvious advantages over pulsed fields on a steady-state neutron source. Because it is naturally suited for time-of-flight measurements, which do not require movement of the spectrometer components, especially the sample table to perform scans in (Q-ε) space, the ESS will provide a unique opportunity to construct a special purpose spectrometer for high-field research.

A first generation pulsed magnet has already been tested at the KEK pulsed neutron source. This system can generate a 20 Tesla field every 2 seconds. The field shape is sinusoidal with a pulse duration time of about 1ms. We anticipate that significant advances in maximum field and repetition rate are possible. This requires a more efficient design to extract the heat produced by the field pulse from the magnet. We can also envisage hybrid systems containing a combination of steady state and pulsed field coils. Pulse shaping to produce a constant field for a limited duration (i.e. flat-top rather than sinusoidal pulse), is also possible. We propose a design target of 40 to 50 Tesla at 2Hz. Assuming that this repetition rate can be achieved, and that the ESS flux is 30 times more intense than ISIS, then the flux available for pulsed field experiments at the ESS would be greater than that currently available at ISIS for steady field experiments.

The prospect of experiments in fields up to about 50T opens up many exciting possibilities. We envisage both elastic studies of magnetic structures and subsequently, inelastic studies of spin-waves, crystal field excitations, intermultiplet transitions and excitations across electronic band gaps. Provision should be made for experiments on both single crystal and polycrystalline samples. Because of the complexity of the equipment (electronics, cooling systems etc.) and special safety features, we propose that a spectrometer be specifically dedicated and designed to accommodate the pulsed field apparatus. It is also possible to envisage future developments of pulsed magnets to reach fields of perhaps 100 Tesla, using flux compression techniques. Experiments would probably be confined to structural studies in this case.

With the development of magnet technology over the last decades, many interesting phenomena, ranging from the quantum Hall effect to metamagnetic behaviour in heavy fermion systems, have been discovered in the 10-25T range. We give two examples of experiments which require fields in excess of 20 Tesla.

Study of metamagnetic transitions in heavy fermion systems

Many heavy fermion materials exhibit a metamagnetic phase transition with a rapid increase in magnetization above a critical field. In the classic heavy fermion system UPt_3, this transition occurs at about 20 Tesla. The origin of the transition is not properly understood, but may arise from quenching of the Kondo effect by the large magnetic field. Neutron scattering studies (elastic, quasielastic and inelastic scattering) of the magnetic correlations, spin fluctuations and excitations are essential to understand the origin of this phenomenon.

Field-induced phase transitions in rare-earth metals, alloys and compounds

Singlet ground state systems may be forced to become magnetically ordered in sufficiently large magnetic fields. For example, the element praseodymium may be driven to an incommensurable antiferromagnetic state by the application of an estimated 22 Tesla magnetic field. Detailed experimental studies of the excitations in Pr have exposed the inadequacy of simple RPA theory, leading to advances in the theory of magnetic excitations in f-electron systems. The theory makes specific predictions about the nature of the excitations in an incommensurate phase which is important to test.

3 Instrumentation for the ESS

Pulsed source spectrometers constructed so far may be classified in two broad categories. In the first, commonly known as *direct geometry spectrometers (DGS)*, the incident energy is defined by a monochromating device, either a chopper or a crystal monochromator, and the scattered energy is determined by the subsequent neutron time-of-flight. In the second category are the *indirect geometry spectrometers (IGS)* which comprise the crystal analyser spectrometers. Here a white neutron beam is incident on the sample and the scattered energy is analysed by an array of crystal analysers each diffracting into separate detectors. These two types of spectrometer have been employed for both single-crystal and polycrystalline spectroscopy. A third category is likely to be important on the ESS because of the enhanced time-averaged flux on the closely-coupled unpoisoned moderators. This is the *triple-axis spectrometer* (TAS) familiar from reactor sources. It is argued in the this section that such spectrometers will still have an important role at the ESS for specialized applications where extreme flexibility of control over the neutron scattering phase space is essential. Although we concentrate in this report on traditional phase space manipulators such as monochromator/analyser crystals, more sophisticated systems will become important in the future and the TAS is the ideal testbed for incorporating them.

3.1 Direct Geometry Spectrometers

Direct geometry spectrometers that are presently in operation are all optimized for polycrystalline neutron spectroscopy but have also proved to be of value on single crystal samples, particularly at high energy transfers. The principle is to produce a monochromatic pulse on the sample, either by phasing a fast Fermi chopper to the proton pulse or by installing crystal monochromators. Neutrons scattered into detectors at an angle ϕ are measured as a function of their time-of-flight or equivalently of the final wavevectork_f. Each detector therefore measures along a parabola in $(\mathbf{Q},\varepsilon)$-space. By employing a multi-detector array, the scattered intensity on a paraboloid surface is mapped out. If a chopper is employed the incident energy resolution is optimized by matching the chopper burst time to the intrinsic pulse width of the source while the scattered energy resolution is dependent on the secondary flight paths. Resolutions of 1% ($\Delta\varepsilon/E_i$) are achieved for incident energies ranging from 10 meV to 2 eV and these could be improved by extending the primary and secondary flight paths. For higher energies, it would be necessary to spin the Fermi choppers faster than the current limit of 600 Hz. For high intensity applications, it is possible to degrade the resolution by increasing the chopper slit widths (i.e. increasing the burst time) and shortening the flight paths. For very low incident energies, below 50 meV, it is more efficient to use a crystal monochromator especially when focussing techniques are used.

The effectiveness of such spectrometers for polycrystalline spectroscopy has been amply demonstrated by the chopper spectrometers on pulsed neutron sources already. They are extremely flexible. The incident energy can be varied over a very wide range simply by adjusting the chopper phase. Above energy transfers of 10 meV, their resolution is better than can be achieved by any other technique currently in use and means that measured spectra cover a very wide dynamic range. The extra flux of the ESS should allow the resolution to be improved even further. Experience at ISIS has shown that, even at high energy transfers, high scattering angles are required. Even for magnetic scattering, which falls rapidly with Q, the high angle data is required for reliable determinations of the nuclear scattering. There is also considerable interest in extending the Q-range to investigate the higher multipolar contributions to the magnetic cross sections. A second lesson is that it is important that the secondary flight path is constant for all detectors. Although many cross sections broaden with Q, e.g. as multi-phonon contributions grow, it is necessary to have constant resolution with scattering angle in order to track the Q-dependence reliably.

The use of DGS for single crystal spectroscopy is still in the early stages of development. A proposal for a spectrometer at ISIS dedicated to such studies is currently under consideration and this will be a vital testbed for the technique. Nevertheless, early experience on HET and MARI is very promising. In a single crystal, the paraboloid scattering surface in $(\mathbf{Q},\varepsilon)$ space

must be aligned with respect to the crystal axes. Provided the detector coverage is complete, it is usually possible to align the $(\mathbf{Q},\varepsilon)$ trajectory of an individual detector close to a symmetry direction, and passing through the Brillouin zone centre or other region of interest. A peak is seen where the trajectory cuts through a branch of the sample's dispersion relations. The spectra from adjacent detectors are usually summed to improve the statistics. This reduces the transverse $\mathbf{Q}$ resolution but because of the plane of symmetry along the direction of $\mathbf{k_f}$ the loss of energy resolution is minimized. Generally, the paraboloid cuts across several Brillouin zones and intersections can occur at several points, depending on the energy scale of the excitations in relation to the incident energy. Further points on the dispersion relation can be obtained by changing the incident energy, with small rotations of the crystal, and it is possible to construct constant energy scans from neighbouring detectors and build up contours of scattering intensity in $(\mathbf{Q},\varepsilon)$-planes.

When the system is one- or two-dimensional, the experimental method can be modified to take advantage of the higher symmetry. With a two-dimensional crystal, if the third dimension is perpendicular to $\mathbf{k_f}$ then many detectors can be summed to improve statistics. An alternative arrangement is to align the third dimension along $\mathbf{k_f}$ in the chosen detector. The component of $\mathbf{Q}$ perpendicular to this direction is fixed, and because the excitations are independent of the component along $\mathbf{k_f}$ the scan is equivalent to the constant-Q scan of the TAS. Nearby detectors also satisfy the constant-Q condition, so the dispersion relation can be mapped to the zone boundary by successively examining the intensity in adjacent detectors. With one-dimensional systems, the method is to align the one-dimensional axis along $\mathbf{k_i}$. All the detectors at low angles both within the scattering plane and at other azimuthal angles can be summed without greatly degrading the $\mathbf{Q}$-resolution. Large gains in solid-angle are therefore possible in low-dimensional systems.

It is clear that major developments in computational techniques are required to improve the interface between the experimentalist and the data so that the experiment can be efficiently managed and the information obtained from DGS spectra effectively extracted.

3.2 Crystal Analyser Spectrometers

These instruments utilize in general a white beam on the sample. The energy and direction of the scattered beam are measured with an analyser crystal. By determining the total time of flight in the detector the energy of the incident neutron and thus the energy transfer at the sample can be calculated. The detector thus records data along a parabolic trajectory in $(\mathbf{Q},\varepsilon)$-space during a time frame. The most successful instrument of this kind to date is the multi-analyser spectrometer e.g. PRISMA at ISIS whereas the rotating analyser technique is still in its development stage.

3.2.1 The Multi-analyser Spectrometer

This instrument employs a large number of analyser crystals each equipped with its own detector. By keeping the component of the wavevector perpendicular to $\mathbf{k}_i$ constant [$Q_\perp = (\pi/d)\ (\sin\phi/\sin\theta_A)$] all detectors record data along the same high-symmetry direction in reciprocal space covering simultaneously a large area in ($\mathbf{Q}$,ε)-space. Such an instrument is ideally suited for overview measurements of dispersion curves over a wide range in $\mathbf{Q}$ and ε. Measurements of the dependence of the excitation spectrum on external parameters such as temperature can be performed very quickly and provide insight in the change of behaviour of not only one particular mode but of the whole excitation spectrum.

The drawback of this technique is the limited flexibility in performing an inelastic scan. Traditional constant-Q or constant energy scans are not directly possible; however, subsequent treatment of the data by suitable software can produce this information. Another disadvantage is the limited influence of the experimenter on the resolution conditions of the scan. The resolution is largely determined by the lattice parameters of the system under investigation and focussing methods cannot be applied. This problem can be alleviated to some extent by providing a range of crystal analyser sets with different d-spacings. The use of a background chopper together with a flexible wavelength band selector reduces the background considerably on instruments of this kind.

With a broadband polarizing filter in the incident beam, polarization analysis can easily be achieved with the (conventional) analyser crystals replaced by Heusler crystals. The design of the instruments should therefore take into account the requirement that no magnetic materials be employed. In the case of an instrument optimized for very small energy transfers polarizing supermirrors could be used in the scattered beam.

3.2.2 The Rotating Analyser Spectrometer

This instrument makes use of a rotating analyser which is accelerated nonuniformly during a time frame. The rate of acceleration is variable and is determined by the type of scan and the sample parameters. The time variation of the analyser scattering angle provides another degree of freedom for the choice of inelastic scans and, for example, in contrast to the multi-analyser spectrometer a constant-energy scan and also a pure longitudinal scan are possible. The acceleration rate of the analyser crystal decreases with the square of the frame length. If the instrument were located on a target with a lower pulse frequency or employed variable speed frame overlap choppers, more time could be spent on on-line adjustment and better precision/resolution could be achieved. Such an instrument would be optimized for small energy transfers. A reduced acceleration rate of the motor would also allow the use of larger

(and non-cylindrical) analyser crystals such as pyrolytic graphite and mica which would give more intensity and better resolution, respectively.

This concept is still under development and it is too early to make a final judgement on its performance in comparison to other instruments. It might also be possible to change the analyser d-spacing as a function of TOF by using crystals which exhibit a piezo-electric effect. R&D is necessary to determine if suitable crystals exist and if their piezoelectric response is large and fast enough to be of practical use.

3.2.3 Triple-Axis Spectrometers

Traditionally, the instrument best suited to the study of magnons and phonons is the triple-axis spectrometer which was invented precisely for this task. Its distinguishing characteristic is the ability to isolate one particular region of (Q,ε) for study. This is a virtue, particularly for complex systems, making it possible to study special points in reciprocal space that simplify the spectra and serve to anchor the modelling and interpretation of the data. Of course, the sequential mode of data collection, well suited to continuous neutron sources, is slow and (at least with a pulsed source) inefficient.

A TAS instrument at the ESS would employ the traditional elements of a triple-axis spectrometer, a monochromator and an analyser crystal together with the possibility to rotate the sample and change the angles of the secondary spectrometer. An inelastic scan would then be performed in the standard way by rotating the sample and changing the scattering angle, together with the required variations of the incident or final energy. Thus the classic scan types constant energy and constant-Q - will be achieved. The instrument will profit from the high average flux of the ESS and will be at least as powerful as a high intensity TAS on a reactor (such as IN8 at ILL). Moreover, there are several advantages over these instruments due to the time structure of the incident neutron beam. The problem of dealing with higher order contamination in the incident beam can be eliminated to a large extent by installing before the monochromator a chopper with a variable and rather long burst time ("sloppy" chopper) without having to restrict the choice of the incident neutron energy as is normally done to allow the use of higher order filters. The ability to distinguish higher order neutrons by time of flight makes this instrument also a prime candidate for the use of polarized neutrons avoiding completely the nonpolarised second order from a Heusler polarizer.

As the neutron beam has a time structure there is the additional possiblity of time-of-flight analysis by the detector and to employ not only a single detector element but even a two dimensional position-sensitive detector. Together with specially developed software (as for the DGS instruments) it should be possible to extract further valuable information from a scan. The sloppy chopper in front of the monochromator (with a continuously variable energy range,

such as developed for HET at ISIS), will also reduce incoherent scattering from the monochromator. Together with the possibility of time-gating the detector i.e. making it only sensitive in a small part of the time frame, this will further cut down the neutronic background. This means that the effective flux is enhanced further and will thus enable the investigation of smaller crystals and/or weaker signals than is currently possible.

Only with further experience will it be possible to fully assess the practical gains to be afforded by the use of TOF spectroscopy for the study of crystal excitations. For this reason it will undoubtedly be prudent to propose several TAS instruments at the ESS. But even the unconditional supporters of this technique admit that this represents an inelegant and probably transitional approach for a pulsed source and that more appropriate choices of instruments will emerge. In this respect the ongoing experience at ISIS and other pulsed sources will be invaluable.

3.3 Special Environments

3.3.1. High magnetic field spectrometer

We propose a dedicated spectrometer built around a hybrid system involving a Bitter magnet with a pulsed field insert for achieving 25T fields at the sample. This option, making use only of present technology, already achieves fields a factor of 2.5 above the highest fields available, but not routinely reached at the ILL today. It will probably possible, even before the ESS is built, to meet the more ambitious targets (≥40T) set in section 2.3.2. The neutron scattering spectrometer will be a direct geometry chopper instrument with a relatively long secondary flight path, which will make good momentum resolution available when the instrument is operated as a diffractometer without the Fermi chopper. It should be located on the 50 Hz source, for exploitation with maximum efficiency in the steady state (Bitter coil) mode.

3.3.2 High pressure spectrometer

Inelastic neutron scattering studies under high pressure have been very limited and restricted to rather modest pressures of 30kbar for polycrystalline and 5kbar for single crystal studies. This is due to the contradictory requirements of large sample volumes and large forces to produce high pressures. Recent developments of an anvil pressure cell for diffraction experiments have pushed the limit beyond 150kbar and with an intense source such as the ESS it should become feasible to perform inelastic measurements. Examples of high pressure investigations include the lattice dynamics of materials of geological importance and the study of novel phases , both structural and magnetic, induced under pressure. We believe that the best way to study inelastic scattering at high pressure is by the use of a dedicated instrument. The reasons for this choice are very similar to the case of high magnetic fields. The constraints on scattering geometry

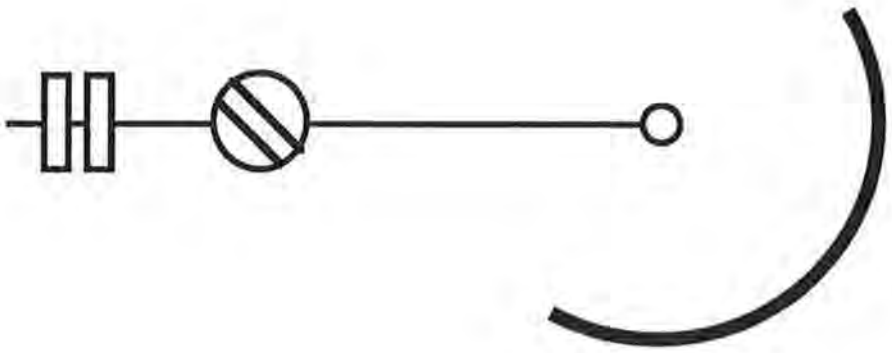
imposed by the complex and bulky equipment are such that the neutron spectrometer and pressure apparatus should be designed together as a single dedicated instrument.

4 Proposed Instrument Suite

4.1 Direct Geometry Instruments

1) Medium energy, single crystal TOF spectrometer, measures large slices of (Q,ω)

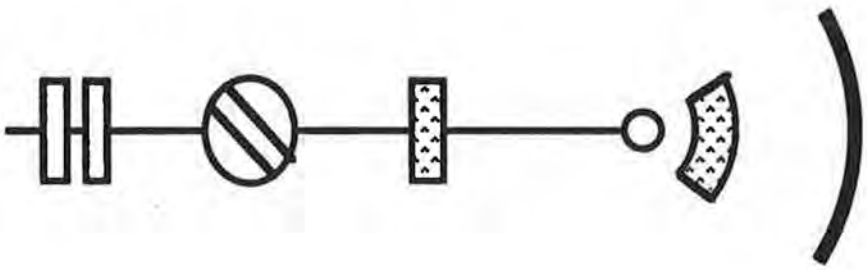
E_o : 20 - 400 meV
$\Delta E/E \sim 0.5\%$
$\Delta Q/Q \sim 1\%$
L_i : 10 - 15m; L_f : 4m
Detectors: horizontal: -30° to +120°, vertical: ± 30°, pixel resolution ~ 1cm
50Hz source
H_2O or CH_4 moderator, decoupled/poisened - sharp pulse
Background, bandpath and Fermichopper
Optional collimator in incident beam



2) Medium energy, single crystal TOF spectrometer, measures large slices of (Q,ω), but with polarisation analysis

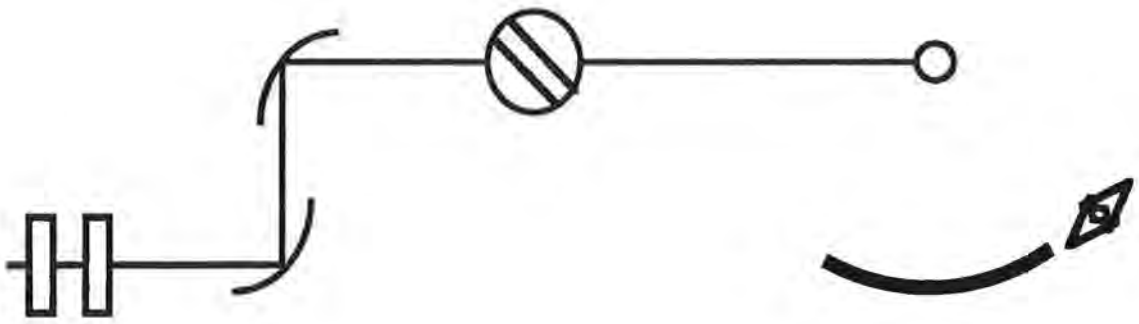
Same as 1) but with polariser for a medium to high energy monochromatic neutron beam, white beam polariser in scattered beam

Detectors: horizontal: ± 30° , vertical: ± 30°, pixel resolution ~ 1cm



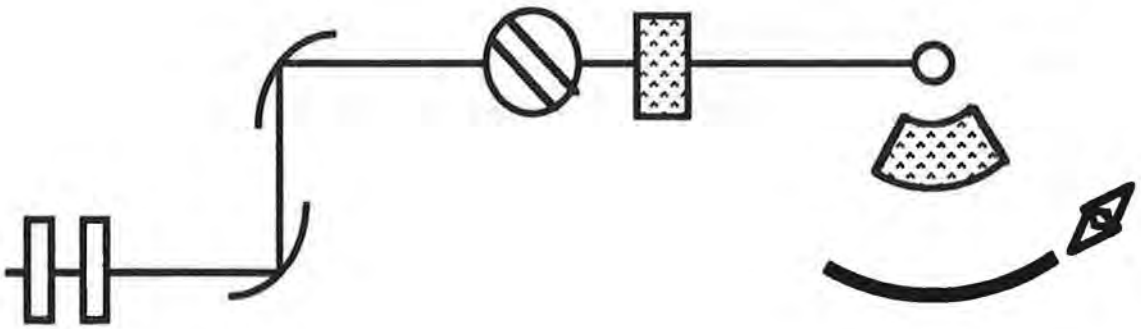
3) Low energy, single crystal TOF spectrometer, low energy excitations, dynamics of phase transitions

E_o : 2 - 20meV
$\Delta E/E \sim 0.5\%$
$\Delta Q/Q \sim 1\%$
L_i : 10 - 15m
L_f : 4m
Detectors: horizontal: ± 30°, vertical: ± 30°, pixel resolution ~ 1cm,
detector bank can be rotated around sample position from 0° to 135°
25 - 50Hz (50Hz) source
H_2 moderator, decoupled/poisened - sharp pulse
Background and bandpath chopper
2 possible versions: focussing monochromator in incident beam or
focussing beam optics with Fermi chopper in incident beam



4) Low energy, single crystal TOF spectrometer, low energy excitations, dynamics of phase transitions, with polarisation analysis

as 3) but with possibility for polarization analysis;
Heusler monochromator in incident beam, white beam polarizer in scattered beam



5) High energy, single crystal TOF spectrometer, very high energy excitations and n(p) measurements

E_o : 200 - 5000 meV

$\Delta E/E \sim 1\%$, or better

$\Delta Q/Q \sim 0.1\%$

L_i : 10 -15m

2 Detector banks: 1) L_f : variable from 4m to 10m,

horizontal: $\pm 15°$ (at 4m) , vertical: $\pm 30°$, pixel resolution ~ 1cm

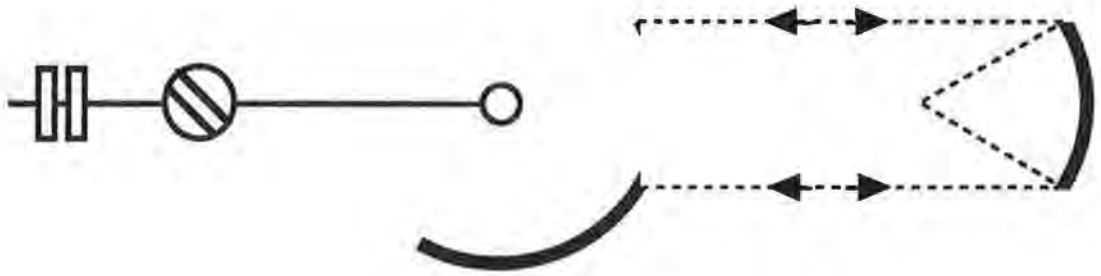
2) L_f at 4m,

horizontal: +15° to +135°, vertical: $\pm 30°$, pixel resolution ~ 1cm

100Hz (50Hz) source

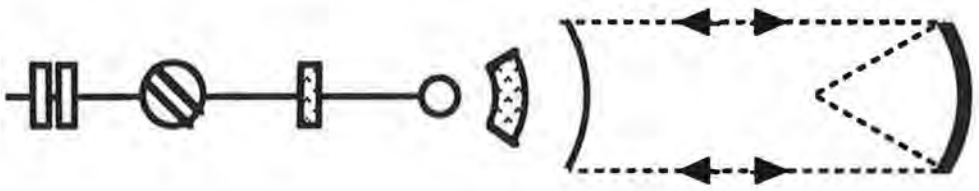
H_2O moderator, decoupled/poisened - sharp pulse

very fast background and Fermi chopper



6) High energy, single crystal TOF spectrometer, very high energy excitations and n(p) measurements, with polarisation option

as 5) but with the possibility of polarisation analysis in the small angle detector bank; polariser for very high energy monochromatic neutrons in incident beam, white beam polarisation analyser in scattered beam.



7) to 12) as 1) to 6) but optimized for polycrystalline/powder samples, i.e. degraded detector pixel resolution

4.2 Crystal Analyser TOF Spectrometer

13) Medium energy, single crystal spectrometer, overview measurements of dispersion curves

$h\omega \sim 10meV - 100meV$

$\Delta h\omega/h\omega \sim 5\%$

$\Delta Q/Q \sim 2\%$

100Hz (50Hz) source

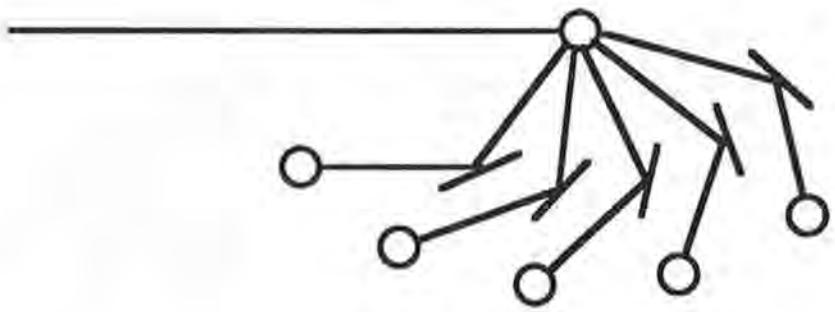
H_2O or CH_4 moderator, decoupled/poisoned - sharp pulse

background and bandpath chopper

vertical linear position sensitive detector

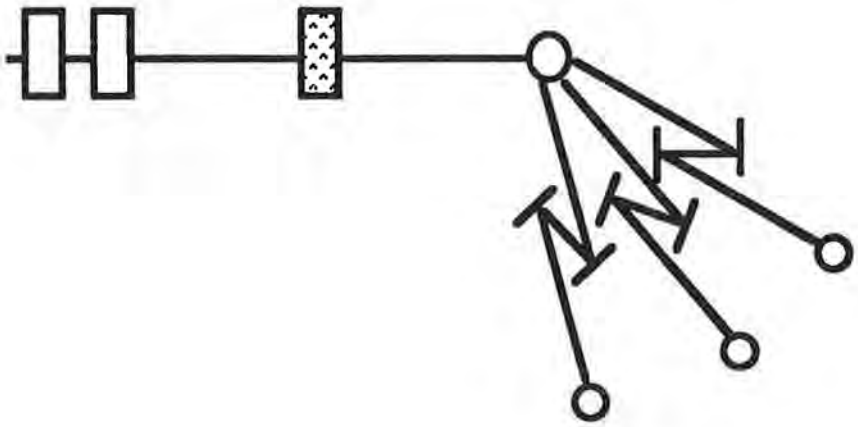
range of crystal analyser (Be, PG, Cu, Ge)

polarisation option with white beam polarizer in incident beam and Heusler analysers;



14) Low energy, single crystal spectrometer,
overview measurements of low energy dispersion curves with good resolution over an extended Q-range

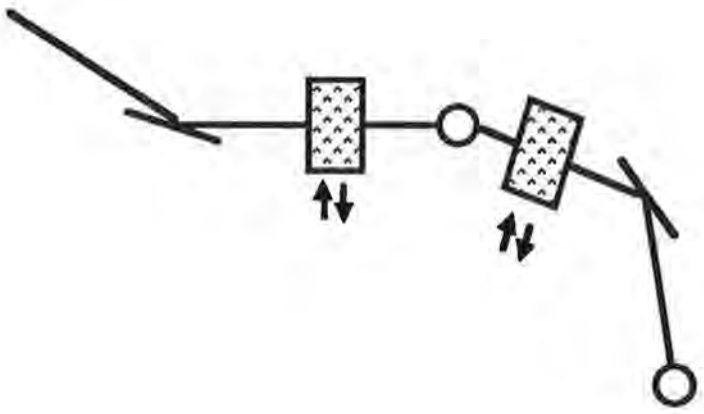
$\hbar\omega < 10meV$
$\Delta\hbar\omega/\hbar\omega \sim 5\%$
$\Delta Q/Q \sim 2\%$
10Hz source
H_2 moderator, decoupled/poisoned - sharp pulse
background and bandpath chopper
vertical linear position sensitive detector
double analyser system (PG, Mica)
polarisation option with white beam polarizer in incident beam and Heusler analysers or supermirrors as polarizers in scattered beam;



4.3 Triple Axis Spectrometers

15) Medium energy, single crystal TAS, "surgical strike" measurements

$\hbar\omega$ up to 100 meV
$\Delta\hbar\omega/\hbar\omega \sim 5\%$
$\Delta Q/Q \sim 1 - 2\%$
50Hz source or greater
H_2O moderator, *coupled (flux) !!!!*
background and bandpath chopper
vertical linear position sensitive detector
a range of crystal monochromators and analyser (Be,PG, Cu, Ge) with the possibility of focussing
Heusler monochromator and analysers;
possibility to use phase space manipulators;
possibility to use Spinecho system;
possibility to use classic cryopad;
possibility of gating the detector;



16) Low energy TAS
"surgical strike" measurements

$\hbar\omega < 5meV$
$\Delta\hbar\omega/\hbar\omega \sim 5\%$
$\Delta Q/Q \sim 1\%$
50Hz source
H_2 moderator, coupled (flux)
background and bandpath chopper
vertical linear position sensitive detector
PG and Mica crystal monochromators and analyser,possibility of focussing
Heusler monochromator and analysers;
possibility to use phase space manipulators;
possibility to use Spinecho system;
possibility to use classic cryopad;
possibility of gating the detector;

4.4 Special Environment Instrument

17) Special Instrument for High Magnetic Field Studies (DC and pulsed)
as 2) to be used for elastic and inelastic experiments

18) Special Instrument for High Pressure Studies
as 2) to be used for elastic and inelastic experiments

4.5 Development Instruments

19) Rotating Analyser Spectrometer, medium energy, single crystal studies

$\hbar\omega$ 10meV to 100meV
$\Delta\hbar\omega/\hbar\omega \sim 5\%$
$\Delta Q/Q \sim 2\%$
50Hz source
H_2O or CH_4 moderator, decoupled/poisoned - sharp pulse
background and bandpath chopper
position sensitive detector
a range of crystal analyser (Be, Cu, Ge)
white beam polarizer in incident beam, Heusler analysers

20) Critical Scattering Instrument with VERY high resolution in both $\hbar\omega$ and Q, for finite values of wavevector and energy transfer, measurements of critical scattering, lineshapes

$\Delta E \sim \mu eV$
$\Delta Q \sim mA^{-1}$

Several options:

- High resolution PRISMA-type instrument with long L_i (~100m) and analysers in backscattering configuration
- An IRIS-type spectrometer for large wavevector and large energy transfers

Other development instruments include :

- a spectrometer to measure the inelastic scattering from surfaces and thin layers/multilayers
- an inelastic neutron spin-echo spectrometer
- an inelastic cryopad device

5. Research and Development

Although the complete report from the ESS Expert Meeting will contain a section devoted to the R&D work identified by all the groups, for the sake of completeness of this report we have summarized in the following paragraphs the areas of R&D which are of particular relevance for this group.

5.1 Detectors

Detectors have not only to be improved in their overall performance but they have also to be made cheaper as the cost of large He-3 detectors is prohibitive. The performance of an instrument, in particular the direct geometry spectrometers, is greatly enhanced by the size of the angular coverage by detectors (typically 1 steradian solid angle currently requires hundreds of individual detectors). On the technical development side we have identified the following requirements:

- to obtain sufficient resolution in Q, in particular for single crystal experiments, the pixel size of the detectors has to be small (~1 cm^2).
- the detectors have to be efficient for high energy neutrons (up to 5eV).
- the dead-time has to be as small as possible; a typical Bragg peak intensity is 10^7 cps, within a 100μs time window.
- inelastic scattering signals are inherently small; so it is important so that there should be minimal detector cross talk between neighbouring elements, some of which may be counting intense Bragg peaks.

5.2 Polarization Analysis

It is extremely useful for magnetic neutron scattering to be able to perform fully polarized experiments. Practically all of the instruments proposed by this group include a polarization option. We require :

- white beam polarizers for the incident and the scattered beam, in the latter case with a wide (30°) angular acceptance.
- high reflectivity Heusler alloy monochromators and analysers with an extended energy range. IGS and TAS instruments would particularly benefit from such developments.

- spin flipper devices which operate equally well at higher energies. Otherwise alternatives to existing devices have to be developed.
- it would be most efficient to perform the polarisation analysis with the detector itself. The use of ^{3}He detectors appears to be an ideal starting position as a white beam polariser employing ^{3}He is already under development.

5.3 Choppers

For inelastic spectroscopy at very high energies very fast Fermi choppers and gamma flash suppressing choppers have to be developed.

5.4 Software

The success, in particular of DGS for single crystal studies, will depend crucially on the development of user friendly software not only for managing an experiment but also for evaluating the results. Although the new instruments will provide us with nearly complete information over a very wide range of $(\mathbf{Q},\varepsilon)$, software is required to visualize the results, to allow for construction of particular scan types through specific reciprocal lattice zones and for search "expeditions" through reciprocal space. We consider this to be a particularly important development without which the efficient exploitation of these instruments will be prejudiced. Computing technology is advancing at a pace which suggests that the manipulation of the very large data sets (several Mbytes) will not pose a technical problem.

5.5 Phase Space Manipulators

These devices are of particular importance to the CAS and the TAS groups. They would enable us e.g. to manipulate the resolution ellipsoid and thus would make it possible to obtain a focussing configuration which cannot otherwise be achieved. An example of such a device is the Drabkin flipper which permits adjustement of the principal axis of the resolution ellipsoid.

5.6 Beam optics

High reflectivity mirrors and guides which function at high energies would greatly improve beam delivery to the sample, especially for chopper machines which do not have intrinsic focussing elements such as monochromators. Obviously, the effort in this area will have clear benefits for neutron polarisation.

6. Conclusion

In the above we have summarised the scientific case for inelastic scattering instruments, to study elementary excitations in condensed matter, at the ESS pulsed neutron source. We have identified four classes of instruments, plus a number of "development instruments", which will allow a very significant extension of experimental capabilities in this area. In each instrument class we have established the major instrument parameters for a set of spectrometers to cover appropriate ranges in wavevector and energy. During our deliberations we have also identified a range of R&D items which are of particular importance for the development of instruments relevant to the research interests of our group.

The study of phase transitions and excitations has historically driven the development of the techniques of thermal neutron scattering. The set of instruments described in this report will allow exciting advances in our field and will maintain neutron inelastic scattering at the forefront of scientific endeavour well into the 21st century.

Acknowledgement:

We are very grateful for the advice of many of our colleagues and in particular for the help of A.T.Boothroyd, M.Hagen, D.McK. Paul, T.G.Perring, R.Pynn, D.S.Sivia and A.D.Taylor.

European Spallation Source

Report of the High Resolution & Molecular Spectroscopy Working Group

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Report of the High Resolution & Molecular Spectroscopy Working Group

1 Introduction

It is generally accepted that neutron scattering as a technique is intensity limited. High resolution neutron spectroscopy necessarily represents the extreme of such limitations; yet, in spite of the almost prohibitively low count rates of high resolution measurements there has been a continual quest to push instrumental methods beyond these limits. This endeavour has allowed a glimpse into future realms of science which currently are not readily accessible to a large community of users.

The realisation of the ESS would advance this work in two significant directions. It would allow the application of the highest resolutions to the most complex scientific problems that cannot be tackled at the present time; and it would provide a unique opportunity for the development of instruments which would operate with an order of magnitude more precision than any existing world-wide, to explore realms of neutron science presently only dreamt of.

The high resolution and molecular spectroscopy working group has addressed three major topics:

- novel scientific applications,
- necessary neutronic techniques to exploit effectively the ESS;
- state of the art instruments to allow new science to be carried out.

2 New Science on the European Spallation Source

2.1 Quasielastic Scattering from Large Molecules

There are two types of motion that are of interest when attempting to understand the dynamics of polymers: the local rotation of segments of molecules (or even whole molecules) about a longitudinal axis; and diffusive motion of whole molecules or their internal modes.

The former is generally best studied in ω-space via both the rotational frequencies and, very importantly, the EISF (for which the technical problems are very similar to those encountered

with small molecules). The latter are much better studied in the time domain by using neutron spin echo techniques.

It is easy to think of experiments that will specifically require the pulsed nature of the proposed source since there is an enormous range of experiments that have not yet been contemplated, but that would be feasible with the high flux of the ESS. In order to study motions which are characteristic of the large molecule, the spatial range needs to be long with respect to the chemical structure. For stiff liquid crystal polymers this could mean $Q<.01$ Å^{-1} while even for flexible polymers $Q<0.1$ Å^{-1} is essential. At present, polymer melts far above the glass transition temperature can be studied for a few carefully chosen, highly flexible substances. Resolution improvements, even within the current Q-range (~ 0.01 to 0.1Å^{-1}), of about one order of magnitude would allow us to study the technologically important stiffer polymers.

If the Q-range is to be extended downwards, it should be remembered that the inverse correlation time, Ω, varies as Q^2 for rigid samples and by as much as Q^4 for internal modes of polymeric molecules. Thus, in order to extend the range of Q, energy resolutions must also be improved.

If all the technical challenges in adapting Neutron Spin Echo to a pulsed source (which does appear possible) were to be solved, an NSE instrument could be built with $0.01<Q<1$ Å^{-1} and an energy resolution $\Delta E \sim 100$ peV *together with* an intense flux, what would one look at ? There would be interest in looking at the motion and, more interestingly, at the anisotropy of motion, of a molecule in a strongly stretched sample, both cross-linked and free. Equally, the motion of a liquid crystal polymer with the mesogens in the main chain would change as one cooled through an isotropic to nematic transition. If this were done in a magnetic field the motion would be expected to be anisotropic. Similarly, the motion of the flexible polymer backbone is severely hindered in the unusual class of materials, liquid crystal polymers, where the mesogenic phase is formed by the side chains. Little work has been done on this important class of polymer.

There is considerable interest in the *conformation* of polymer molecules in various confined spaces, for example trapped in pores or which are attached by one end to a surface. Practically these latter are used as steric stabilisers of colloidal systems, such as paints. No-one has yet contemplated trying to observe how the confinement and constraint in these situations affects the polymer dynamics.

2.2 Non-Equilibrium Molecular Dynamics

The vast area of study of the dynamical properties of fluids under extreme non-equilibrium conditions has only begun to be explored using computer simulation and some coarse theoretical approaches. There is, as yet, a paucity of experimental work.

By the very virtue of being a pulsed source, the ESS will enable the possibilities of applying short but intense pulses synchronously with the pulse repetition frequency (or one every few pulses, depending upon the particular relaxation to be explored). Such a facility will enable the measurement of the properties of molecular materials subjected to the following kinds of stress:-

AC Electrical fields (up to10^6 Vcm^{-1}). A number of precursor phenomena has been found in optical studies of molecular fluids below the dielectric breakdown field. Their microscopic correlations have been extensively studied on computer Molecular Dynamics simulations, but such results, based on simple model potentials which are probably unrealistic at these field-strengths, remain to be validated by experiments.

Shock-waves in viscous media. Similarly a rather large number of results based upon hydrodynamic approaches has already appeared, and their microscopic counterparts explored by means of computer simulation. Very strong dynamic orientational correlations have been suggested but only preliminary results are known from light scattering.

Very viscous fluids under shearing stresses. A substantial theoretical effort has been devoted to the study of these phenomena. From neutron scattering some preliminary results have been obtained by SANS on suspensions of large bodies (i.e. latex spheres) where clear distortions of S(Q) have been observed. The interest in the *dynamics* of such phenomena is readily apparent, though no results have yet been obtained from neutron scattering.

Dynamic correlations in Kerr media. QENS is specially suited for the investigation of the dynamic correlations in isotropic Kerr substances, and a large number of molecular liquids such as CS_2 or benzene have large Kerr constants. The dynamics of such systems, when they are irradiated by moderately intense laser fields (~1 W/cm^2), have also been investigated by molecular dynamics calculations. This is particularly exciting where the strongest deviations from equilibrium dynamics have been predicted.

2.3 Recoil Molecular Spectroscopy

This technique has only recently become available in a prototype form with the advent of epithermal neutron fluxes from pulsed sources. Recoil molecular spectroscopy examines the

nuclear momentum distribution of atoms in a molecule, thus allowing the ground state wavefunction of each nucleus in the molecule to be directly observed.

The present state of the art addresses the proton self interference in potential double minima such as occur in hydrogen bonds. Predicted about four years ago, subtle differences between hydrogen and deuterium have been recently observed. They are however not fully understood, but could be using the proposed ESS instrumentation. A new source with much improved intensity would look beyond this to resolve the isotopes of other chemically common nuclei. This technique would take advantage of specialised isotopic labelling techniques, e.g. ^{13}C, to examine *individual* nuclear momenta in molecules.

2.4 Liquid and Molecular Crystals

The quasielastic spectrometers at ILL have provided fundamental information on the molecular dynamics of liquid crystals, but the limitations of energy resolution, flux and access have prevented a major impact being made on some of the more complex current problems. For instance energy resolutions now available are not good enough to observe the most important molecular motion which is the end-over-end tumbling of the molecule. For simple thermotropic nematic phases a detailed knowledge of this motion would require an energy resolution in the neV range. This is essential to a full understanding of the dielectric properties of liquid crystals that are exploited in many of the technological applications of nematics.

There are now many more types of liquid crystal with both potential and real applications. Chiral phases are likely to be an important source of applications in the future. Surface stabilised ferroelectric liquid crystal displays are just beginning to be marketed but our very limited understanding of the smectic-c phase at a molecular level suggests that there is considerable scope for improvement. For instance, smectic-c material design would be greatly assisted if a good model could be established to relate the rotation of the molecular dipole moment to features of the molecular structure such as distance from the chiral centre. Selective deuteration could be used to distinguish motion of different parts of the molecule. An energy resolution of 100 neV and a Q-range of 0.1 to 1.0 A^{-1} with high intensities would allow considerable progress to be made.

Another exciting new class of mesogenic materials are the liquid crystal polymers. Here the complex molecular motion would need a wide range of energy transfer and momentum transfer. Motion of the backbone would require the highest possible resolution (<100 neV) whilst more local motions would probably require less stringent resolutions.

2.5 Density of States Measurements at High Resolution

On ISIS, using the present crystal analyser spectrometer TFXA, it is possible to produce spectra with good statistics for hydrogen-containing samples and those consisting solely of light atoms, with a resolution of ~ 2% ($\Delta E/E$) over energy transfers from several meV up to several hundred meV (where Debye-Waller factor and multiphonon effects begin to dominate). For strongly bound molecules (such as ice), one measures an amplitude-weighted density of states with very sharp cut-offs corresponding to zone edge frequencies. For weakly bound molecules, the internal modes appear as sharp peaks. An important case involves the hydrogen atom in hydrogen bonds where measurements demonstrate that the dynamics can be understood in terms of a fixed potential well. The technique also provides precise spectra for matrix isolation and defect mode cases. The major advantage of the neutron technique, with respect to all other scattering spectroscopies, is that one can calculate the vibrational eigenvectors directly from the spectra for a given set of force constants. Well-developed computer programs (PHONON and CLIMAX) already exist which perform this procedure yielding unique information where single crystal samples are not available.

The CASSANDRA instrument, which is proposed for ESS, would considerably enhance this technique. The attainable resolution ($\Delta E/E$) would be 3×10^{-3} and the enhanced count rate would allow sets of temperature and pressure dependent spectra to be measured in a reasonable time with good statistics. A diffraction detector on the spectrometer would allow a simultaneous determination of cell parameters. When this performance is coupled to a sufficiently powerful data analysis package, the advantage of neutron scattering over all other spectroscopic techniques will be clear.

2.6 Tunnelling Spectroscopy

The use of the high intensity of the ESS.

Tunnelling spectroscopy will profit in various ways from a next generation neutron source. The obvious development is to exploit the increased flux. This allows experiments on considerably more complex and much more dilute samples. At high pressure, $p \leq 20$ kbar, only very small sample volumes are possible. The pressure dependence of rotational tunnelling combined with high precision diffraction gives access to the fundamental inter-molecular interactions. Sometimes, due to the difficulty of preparation, even at standard environmental conditions only small quantities of sample material are available, such as is the case for biological materials. In special surroundings the concentration of the tunnelling molecules is low. In matrix isolation spectroscopy the almost-free molecule is studied and the low concentration limit must be extended to concentrations $\leq 10^{-4}$ to be free from intermolecular

interaction effects. This is more than an order of magnitude beyond the concentrations able to be investigated at the present time.

Similarly, molecules enclosed as clathrates or cryptates, in layered compounds such as intercalates, or on surfaces with large coherent area, represent very weak scatterers. A desirable manipulation of such systems by partial deuteration is beyond the present possibilities because of the low cross-sections of deuterium. Weaker scatterers than hydrogen (the simplest extrapolation being deuterated systems) will, on the ESS, become generally accessible to the method.

The tunnel splitting represents the energy difference of the split ground states of the various spin symmetry species of a quantum rotor. The decay into the lowest ground state is slow at low temperature. The relevant spin conversion times τ_c can range from fractions of a second to years. They must be determined in real time experiments, especially for the more complex ground states of three-dimensional rotors such as methane where a number of different spin conversion times are present. To cover a wide range of τ_c it must be possible to record a spectrum in 5 minutes. This sensitivity might also allow the kinetics of phase transitions to be followed by looking at the temporal evolution of tunnel spectra.

The use of the high resolution of the ESS.

Intensity can also be traded for improved energy resolution. Improved resolutions, over wider energy transfer ranges, would extend tunnelling spectroscopy to more strongly hindered rotors and thus create a significant overlap with indirect methods of tunnelling spectroscopy, such as NMR. Apart from this, and arguably much more important, rotors with smaller rotational constants (synonymous with smaller splittings at similar strengths of the rotational potential) such as N_2, become accessible to the method.

Great progress is expected from exploiting a much larger accessible energy transfer range with an almost unchanging, high energy resolution. A number of systems showing complex coupled tunnelling spectra have been uncovered so far but the spectra are often not fully resolved. Inequivalent rotors in large unit cells, low site symmetries in the case of three-dimensional rotors, or rotor-rotor coupling can be the cause of such complex features. Coupling might be present between pairs only, or extended over a large number of rotors leading to collective tunnelling. This field has just opened up and will benefit greatly from upgraded facilities.

The improved energy resolution would allow, in principle, the tunnel splitting of excited librational states to be resolved. Apart from a handful of rare exceptions librational branches usually show dispersion. Thus the quantum spin structure of librational bands in phonon dispersion curves might only be observable using deuterated single crystals with rather good Q

resolution, which is beyond today's possibilities but feasible with the 30 times source flux increase of the ESS.

Often materials investigated by tunnelling spectroscopy are prepared *in situ* in the neutron beam. The simultaneous observation of diffraction patterns and inelastic spectra, possible on inverted geometry TOF spectrometers on pulsed sources, represents a qualitative improvement in the information obtainable (see §8 below)..

2.7 Liquids and Glasses

An important area of research is the understanding of the liquid-glass transition but the instrument requirements are stringent. An instrument with very good resolution (~ 0.1 μeV) over a wide energy transfer range and an extended Q-range ($Q \leq 2$ Å^{-1}) is needed. Suitable materials for these studies are high viscosity molecular liquids such as bromopropane.

Solid state proton conductors are likely to be of increasing importance; the number density of protons in such materials can be quite low, therefore needing the high intensity of the ESS.

A further broad field of interest is the dynamics of hydration in aqueous solutions to which electrolyte salts have been added. These salts strongly affect the local hydration structure, and thus the macroscopic properties, of these systems. The resolution range needed is $0.1\ \mu eV < \Delta E < 50\ \mu eV$ over a Q range of $0.1 < Q < 1.5$ Å.

Diffusive motions in molten salts are poorly understood, especially in those salts which, in the solid phase, have a large ionic conductivity such as silver bromide and solid state proton conductors. To study these systems the isotope substitution technique has to be used. In addition to the instrumental characteristics stated above these measurements are also extremely intensity demanding since cross sections are low and absorption is often high.

2.8 Simultaneous Spectroscopy and Diffraction

It is often stated that neutrons can measure both structural and dynamical properties of matter, however this has only very recently been carried out simultaneously, using pulsed source instruments.

High resolution time-of-flight neutron diffraction patterns and inelastic scattering spectra taken on the IRIS instrument from caesium intercalated graphite after adsorbing H_2 at 110K and then cooling to 4 K, shows the power of simultaneous measurements of high resolution diffraction and inelastic scattering The data are shown in Figure 1 where the spectacular change in tunnelling spectrum with hydrogen content is evident. However the change in c-axis lattice

spacing is, unexpectedly, very gradual even at the very high diffraction resolution used. This system behaves in quite a different way to the $C_{28}Cs(NH_3)_x$ intercalate, which was the first material to be studied in this manner, there being little sign of the step-like behaviour seen for the ammonia system. This result points clearly to changes in the in-plane packing, rather than the c-axis component of the crystal field at the adsorption sites, as the cause of the changes in the tunnelling spectrum. Furthermore there is no evidence in the diffraction patterns for the coexistence of different multiply-staged intercalated phases.

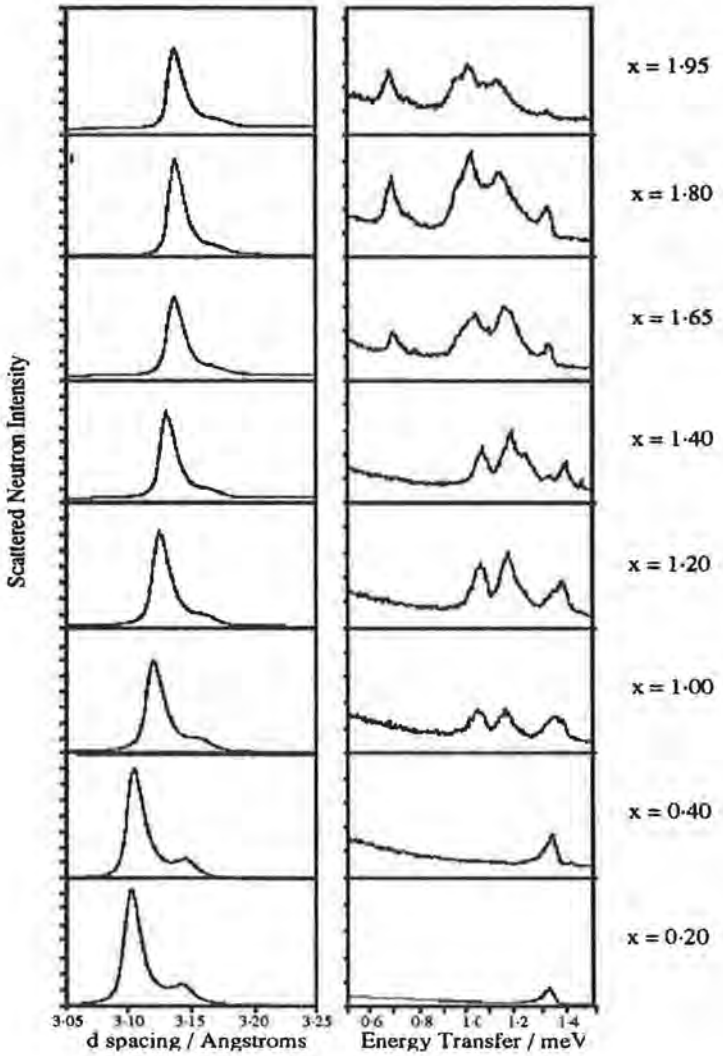

Figure 1. Neutron diffraction (a) and high resolution neutron inelastic scattering (b) from $C_{28}Cs(H_2)_x$.

These two experiments have revealed the power of simultaneous diffraction and spectroscopy to understand the correlations between dynamical effects and structural evolution in such uniquely prepared materials. Often these materials are necessarily prepared in-beam and can not be reproducibly re-created when attempting to make consecutive measurements on different instruments. Simultaneous measurements of structure and dynamics is therefore invaluable. Whilst this technique will

undoubtedly be developed on present-day sources, the ESS holds out the possibility of designing purpose-built instruments with remarkable power, combining the best available performances in both spectroscopy and diffractometry in one instrument. Future scientific applications will include the study of matrix isolated species such as ammonia in nitrogen, and other non-equilibrium systems such as solid-state batteries during discharge. Furthermore simultaneous diffraction can act as a powerful diagnostic tool to ascertain the crystallographic phase of a sample under spectroscopic study, such as a liquid crystal material with many phases closely separated in temperature, and the study of supercooled liquids.

2.9 Liquid Alloys and Fused Salts

Observation of inter-diffusion of species in liquid alloys such as NaCl in Na, can show either a critical point for phase separation (critical narrowing) or a tendency to immiscibility, is beyond the capability of current instrumentation. Samples need isotopic substitution to be able to weight appropriate fluctuations, and measurements must be made at low Q and with high resolution (10 $mÅ^{-1}$, 100 neV) to be sure of a chance of success.

Interdiffusion of different species in liquid alloys can show phase separation or immiscibility, but not always. The same conditions for observing this type of critical narrowing also apply to some types of magnetic critical scattering. The ability to measure with high intensity and high resolution, leading to general use of isotope differences comparable to work done now for H/D, is likely to have considerable importance for general studies of multicomponent liquids and fast ion conductors. There is also interest in distingusihing single particle and correlated motions; this has been done with polarisation analysis but isotopes are sometimes a possibililty.

Investigations into the diffusion mechanism in the technologically important superionic conductors and in the increasingly complex systems such as fast ion-conducting glasses can just be attempted with present neutron scattering scattering instrumentation but inadequately. Polymer electrolytes will need good resolution (10 μeV) and high *intensity* at intermediate Q values for their stochastic behaviour to be understood. In addition Q variation is an absolute necessity to study the dynamics of salt fusion and the accompanying structural changes. Isotopically-enriched samples are essential for the successful outcome of these studies, which of course implies that the samples will be small.

2.10 Technology of Hydrogen in Solids

Neutron scattering techniques are a useful tool in technological problems involving hydrogen in metals and in other solids. Possible topics to be addressed are

- Low concentrations of hydrogen in steels and in other structural materials
- Behaviour of tritium in fusion reactor materials
- Proton behaviour in batteries (rechargeable metal hydride batteries)
- Development of improved proton conducting materials
- Location of hydrogen in semiconductors.

Perhaps the most sensitive way of measuring small amounts of hydrogen in materials is by inelastic neutron scattering because of the large cross section and the small mass. When the system is such that all the protons have a fixed vibrational frequency (the Einstein oscillator) separate from the lattice modes of the material, the peak can be identified using a high sensitivity, high resolution instrument such as CASSANDRA. The area of the observed peak compared with the band modes is an accurate measure of the H concentration.

In the development of metal hydride batteries, a major unsolved problem is that the intermetallic hydride (e.g. $LaNi_5H$) gradually degrades as a result of hydrogen cycling. This degradation is best measured by observing the inelastic scattering from the charged hydride. If trapped in degraded sites it gives a new peak in the spectrum. Again high sensitivity and resolution is necessary to distinguish the subtle spectral changes.

Hydrogen permeation membranes are important in many industrial processes such as the removal of hydrogen from reversible reactions. and in fuel cells and heat pumps. Palladium alloys, plated onto porous tubes, and ceramic proton conductors such as $SrCeO3(Yb)_{0.05}$ are being actively developed. Inelastic Neutron Scattering can be used to identify hydrogen site distributions whilst Quasielastic Neutron Scattering is a sensitive way of investigating microscopic diffusion mechanisms. These processes frequently involve trapping of hydrogen atoms which shows up as a combination of broader Lorentzians under the elastic line. Understanding the mechanisms of trapping will certainly require QENS studies over a wide range of temperatures and therefore will require access to instruments able to detect diffusional processes over many decades of time. This implies a suite of instruments with a variety of resolutions.

2.11 Dynamics of Complex Systems

The term *Complex System* is used to refer to condensed matter in a broad sense not possessing a highly ordered structure and with a high degree of disorder. Spin glasses were the first examples widely described under this appellation, which also implies that there is no known

unique ground state. Other examples are structural glasses, gels, and certain polymer and biological samples. Some of these structures can be characterised as fractals, i.e. they are self-similar at increasing magnifications.

The dynamical behaviour of these complex system is, rather naturally, also complex and unusual. This applies both to the density of states (DOS) and to the relaxation phenomena. Thus the DOS in fractal systems reveals several regimes in the vibrational spectrum, with several cross-overs between propagating modes (such as long wavelength sound waves) and localised modes, the proposed "fractons". The structural relaxation behaviour in the time domain for systems such as spin glasses and structural glasses also deviates from the usual exponential Debye behaviour. Unconventional time dependencies such as power laws or stretched exponentials can be tentatively identified, while there is no accepted theoretical framework to describe these processes, in spite of a large theoretical activity in the past decade. Nevertheless several general, more or less phenomenological approaches (such as the mode coupling theory of generalised hydrodynamic models) have produced some general mathematical predictions, for example scaling relationships of various kinds.

In the experimental study of the dynamics of these complex systems neutrons have a special role to play, since they can explore both the space and the time domain simultaneously.

Instrumental requirements are:

- High resolution, since many of these processes are slow, or become very slow on cooling. The complex character is precisely related to the "slow motion" properties, the local atomic vibrations being rather similar to those incrystals and liquids.
- A wide dynamic range in energy/time, in order to follow the complex line shape in its full extent, not just attempting to deduce a few components with known or assumed shapes and widths. This range cannot be successfully exploited without high resolution.
- Good statistics, and thus high flux, in order to verify the mathematical properties of unknown lineshapes when comparing to assumed functional forms.

Neutron scattering results have to be used in combination with data obtained from other techniques such as dielectric relaxation, time dependent specific heat, light scattering, and NMR, which cannot explore the momentum space domain (0.1 to 5 $Å^{-1}$ desirable for neutrons) but which can nevertheless provide highly accurate calibration points.

2.12 Biomolecular Applications

Biological macromolecules exhibit an extraordinary structural and functional diversity; the study of the interrelationship of structure, dynamics and function at the molecular level is nowadays a central subject of research in molecular biology. Biomolecules are normally functionally active only in aqueous environments (water content above 20-30%), therefore interactions with water are of fundamental interest at all levels of hydration. At the molecular level these considerations are reflected in a rich spectrum of low-frequency fluctuations and collective processes including the following:

- Soliton and dispersive (phonon-like) modes in the biopolymer backbone and in the closely associated first shells of hydration.
- Dynamics of side groups which, at the biopolymer-water interface, may be largely controlled by solvent interactions.
- Motions of massive parts of a molecule (domains) relative to each other. These are thought to play an important role in determining enzymatic activities by modifying the interactions with the solvent at the active sites of enzymes.
- Rotational and translational degrees of freedom of hydrate water which is markedly modified with respect to bulk water.

High Q-resolution and high energy-resolution data coupled to the enormous progress of biochemical selective labelling techniques (H/D substitutions), will be the basic instruments and techniques needed to arrive at a self-consistent description of the molecular mechanisms responsible for the functional behaviour of biomolecules.

2.13 Spin Labelling in High Resolution Spectroscopy - A Gedanken Experiment

Isotopic substitution, and in particular selective H/D exchange, has proved a most effective method for simplifying and assigning the incoherent inelastic scattering from molecules of moderate complexity and from polymers. Dynamically polarised nuclei at low temperatures would produce a sample with enhanced cross-sections which is today feasible following the pioneering work of Stuhrmann in Geesthacht. A somewhat futuristic extension of this method, made possible with the fluxes of the ESS, would be to use dynamic nuclear polarisation at low temperature to produce a largely coherently scattering sample of molecular and polymeric solids (including biopolymers) and then to selectively depolarise individual proton groups, which might be identified by their dipolar and possibly chemically shifted, split resonances. Data

would be taken synchronously as a function of time after the depolarisation burst, to correct for relaxation effects.

This procedure would create strong incoherent scattering at the depolarised protons whose contributions, in different molecular modes, would then become apparent.

2.14 Quantum Fluids: Helium

Liquid ^{4}He has been studied by neutrons since 1958 and there are still, in 1992, significant experiments to be performed. Because it is *the* fundamental quantum liquid of basic interest, it is conceivable that a precise systematic mapping of $S(Q,\omega)$ as a function of pressure and temperature from 200 mK through T_λ up to 5 K should be contemplated on future ESS instruments. Ideal for this study would be an indirect geometry (TOF-crystal analyser) instrument which would allow $S(Q,\omega)$ and its integrals to be measured in one run. Calibration of Q and ω *must* be better than one part in a thousand (i.e. *much better than is nowadays available*). Q ranges from 0.1Å^{-1} to 4Å^{-1} and energy transfers $\hbar\omega$ from 100 μeV and extending up to 10 meV with a resolution (at $\hbar\omega$=0) of 50 μeV would be necessary with adequately high signal and sufficiently low backgrounds.

At very high energies, recoil scattering (deep inelastic or Compton scattering) is governed essentially by the momentum distribution n(p) of helium atoms. This applies to both isotopes. The condensate fraction n_0 is still an issue of debate and research which will surely yield to the power of ESS instrumentation.

Liquid ^{3}He exhibits two dynamical modes: a spin fluctuation (paramagnon) mode and the collective zero sound. A dream experiment would utilise polarisation analysis (not yet possible on present instruments) to distinguish both modes and - more importantly - their mixing. Technically, energy transfers $\hbar\omega$<3 meV with a resolution of 100 μeV on a direct geometry TOF instrument are needed (the white beam of an indirect geometry instrument, otherwise perfectly acceptable, would heat the sample too much). Excellent background and contrast are essential. ^{3}He exhibits an extremely low scattering probability of 10^{-4} and represents one of the most difficult measurements possible in neutron scattering.

^{3}He-^{4}He mixtures. ^{3}He, diluted in superfluid ^{4}He, is a model Fermi gas with an adjustable Fermi momentum. The Fermion excitation has been studied by inelastic neutron scattering at $0.8<Q<1.7$Å^{-1}. There is great interest in extending these studies to significantly lower Q values, say to 0.3Å^{-1}. A series of measurements at different concentrations and temperatures is required in order to retrieve information about the interaction between the ^{3}He-quasiparticles. A direct geometry instrument allowing an energy transfer $\hbar\omega$ from zero to 1 meV and a resolution δE = 20 μeV would be necessary for this study.

2.15 Electron - Phonon Interactions

With the exception of an extremely small number of well-known examples, the lifetime of elementary excitations and thus their interactions cannot be determined by conventional neutron scattering techniques. These excitations are definitively inaccessible to other methods, with only the coarsest energy ranges slowly becoming explorable by x-rays, but here the resolution limitations are very substantial. Compared to the resolutions achieved up to now with neutron techniques in the study of elementary excitations (15μeV at best) an improvement of two orders of magnitude is required to open up a new field of investigation of phonon interactions as a general tool. Electron-phonon interactions are of particular importance because of their role in superconductivity and other electrical and magnetic properties of metals and semiconductors.

The application of Neutron Spin Echo techniques in combination with a conventional method of studying elementary excitations (where NSE is used as a magnifying glass to explore within the resolution spot of the conventional spectrometer) has been demonstrated to have the required capability for this work. The advent of the Neutron Resonance Spin Echo variant offers a feasible and manageable technical solution. Nevertheless, the method remains complex and requires the best possible fluxes, since the addition of the NSE option to an ordinary inelastic machine results in some 30-100 times loss in counting rate. (Actually, a brute force resolution improvement, if it were technically possible, would result in an intensity loss of between 10^4 and 10^6) Backscattering techniques now allow 1 μeV resolution studies of excitations without dispersion ($\delta\omega/\delta q=0$), but this is a special case. Improved intensity is at a premium in these studies.

2.16 Experiments To Come

Phonon-phonon and electron-phonon interactions which occur because of the connection between electrical properties and crystal dynamics require resolutions of better than 1 μeV at selected Q,ω points. Generalised NSE (or the NRSE technique) combined with a chopper machine to pre-select the Q,ω spot is a possible means of tackling this subject.

Protein dynamics showing fluctuations in the ternary structure (large subunits ~ 10-50Å) are related to the functioning of proteins. Their study requires an improved absolute resolution at small Q's using very long wavelength NSE with $\lambda>10$Å.

Relaxation after an external trigger such as a laser flash allows kinematic studies of the structure and dynamics of many materials. In this case improved flux is an absolute necessity.

Study of diffusion and the structural relaxations in solids and liquids on short length scales, require Q's up to 10Å^{-1} and NSE on a thermal or hot beam, pointing to the need for improved hot neutron fluxes.

Dynamics of complex systems such as undercooled liquids, high viscosity systems (paints, gels) etc. show non-conventional lineshapes and unusual scaling properties. For these studies, the highest resolution and intensity combinations are needed, providing good statistics for non-Lorentzian lineshapes (NSE, or backscattering of cold neutrons).

2.17 Diffraction with Elastic Filtering

Thermal diffuse scattering is a well-known contaminant effect in crystallographic experiments, and it can be more or less satisfactorily handled for ordinary, high Debye-temperature solids. This is not the case with so-called "soft matter", e.g. ordered polymer structures or protein crystals. A separation of the "strictly" elastic contributions from the inelastic ones in SANS, diffuse scattering and diffraction not only improves the quality of the structural information sought, but also provides information on the dynamics, e.g. via the determination of the Debye-Waller factors.

Elastic filtering can be done by any inelastic scattering technique and surprisingly often the highest possible resolution is required in order to separate out inelasticity. The Rayleigh scattering of Mossbauer radiation method will provide a tool for synchrotron radiation to do this with a 10^{-9} eV resolution. However, light elements, primarily H and D to C, N & O cannot be distinguished or traced with synchrotron radiation, thus the implementation of an elastic filtering technique is equally important for neutrons.

2.18 Fundamental Physics and UCN at ESS

Many of the activities of the Nuclear Physics College of the ILL are recognised as being some of the most successful work of the Institut. In the planning for ESS arrangements for similar work to be performed at a future spallation source must be catered for. The construction of ESS represents an opportunity to produce a uniquely intense source of ultra-cold neutrons which would transform the field by an amount even greater than the transformation achieved by the UCN source at ILL.

Such a UCN source on the ESS would allow the present most successful applications of UCN (the neutron electric dipole moment and the ß-decay lifetime of the neutron) to be performed at much higher accuracies and would open up a new range of measurements and instruments. These include angular correlations in neutron ß-decay (polarised UCN offer an extremely clean

way of carrying out such experiments), UCN interferometers with a broad range of applications ranging from the neutron charge determination to general relativistic effects, the search for a gravitational dipole moment, and operation of a UCN microscope of much higher resolution than has been obtained to date.

Coupled with the further development of UCN inelastic and quasielastic scattering as a tool for studying condensed matter, (quasielastic energy exchanges with a vibrating surface of the order of 10 peV/bounce have already been observed) these points constitute a very strong case for the inclusion of a strong UCN programme at ESS.

Other applications of ESS to fundamental physics should not be overlooked The use of ESS as an intense muon and neutrino source offer many possibilities for new experiments.

The neutron has several unique properties for fundamental physics investigations. It exhibits all fundamental interactions: strong, weak, electromagnetic and gravitational interaction. All these interactions cause measurable effects when investigated by dedicated high precision instruments. Much progress has been achieved in the field of *neutron interferometry* which most clearly demonstrates the wave-particle dualism of quantum mechanics. This method has been used to achieve extremely high energy and momentum resolutions ($\Delta E \sim 10^{-19}$ eV, $\Delta Q \sim 10^{-7}$Å^{-1}) and is therefore a candidate for future extremely high resolution instruments. Many other fundamental phenomena have been demonstrated by this method :

- 4π symmetry of spinors
- macroscopic spin superposition
- Aharonov-Casher effect (interaction with an electrical field)
- magnetic Josephson effect
- scalar Aharonov-Bohm effect
- neutron gravitational effects
- neutron Fizeau-effects

Many other phenomena are under investigation (Berry-phase, delayed choice experiments, tidal force experiments etc.). Besides these investigations in the field of fundamental physics, the interferometric technique will become applicable to condensed matter research too (inhomogeneities, phase topography for example). The extremely high sensitivity can be further increased as longer perfect crystals and long wavelength interferometers come into operation. Many achievements of neutron interferometry are included in modern text books of physics and their epistemological aspects can not be stressed too strongly.

Modern neutron optics will develop further and will contribute to our knowledge of nature and will be incorporated into advanced neutron instrumentation. Steps towards *neutron quantum optics* have been made in the last few years but many aspects are still open for exploration,

such as: coherent wave mixing (possibly also with electromagnetic waves), squeezing and antisqueezing phenomena, higher order coherence experiments and contributions to the quantum mechanical measurement theory.

With the availability of a high intensity pulsed neutron source such as ESS, the direct measurement of the *neutron-neutron interaction* becomes feasible. This would be of central interest to answer the question of charge symmetry and/or charge independence of hadronic interactions. Pulsed sources are extremely well-suited for such investigations because this effect depends on the square of the available peak flux. The spin dependence of the neutron-neutron interaction is of special interest.

In addition to the search for a static electric dipole moment as discussed above, the neutron exhibits an *induced electric dipole moment* whose more exact measurement is of central importance for testing modern quantum chromodynamics theories. Neutron-laser interactions will play a central role towards that end.

Many other topics will come up because the neutron is indeed itself an interesting object for basic investigations due to the variety of its fundamental interactions, its wave-particle character and its availability in an energy range from 10^{-8} to 10^{8} eV.

Thus fundamental physics with neutrons is an exciting, promising and active field of research which will gain both qualitatively and quantitatively from the availability of higher intensity neutron sources.

3 Techniques for the European Spallation Source

Polarisation Analysis

Polarisation analysis is important for the identification of magnetic and spin incoherent scattering and is a fundamental prerequisite of the Neutron Spin Echo spectrometer.

It is not difficult to polarise a neutron beam in a guide by the use of a "polarising cavity" type device. The polarising cavity has the advantage that it does not modify the beam direction compared with the unpolarised case. An existing instrument could thus be converted from an unpolarised mode to a polarised mode by simply replacing one guide element with this cavity. With existing supermirror benders this method of polarising can be considered as fully satisfactory for $\lambda>2.5$Å. Below this wavelength supermirrors can still be used with some intensity losses. Much is to be expected from current developments in polarised ^{3}He filters.

The analysis of the scattered beam is more of a problem, in the quantitative sense. In order to cover a large detector solid angle large amounts of supermirrors are needed (or large area ^{3}He filters would be required). The production of the necessary amounts of supermirrors is technically possible, but very expensive. However as more and more laboratories, and even commercial companies, enter the field, substantial supermirror production capabilities could be available within a few years if properly encouraged. (D7 at ILL is presently the only operational instrument. It uses 32 supermirror benders of about 1° x 2° solid angle analysers).

The number of supermirrors needed is inversely proportional to the neutron wavelength. Thus at λ=1Å it would be prohibitive to cover the same kind of solid angle that could be envisaged at say λ=5Å. (For large samples even more supermirrors are required). Therefore it is not reasonable at present to build polarisation analysers with wavelength bands too large (such as 1 to 20Å).

Note that on crystal analyser instruments such as IRIS the use of polarising analyser crystals (e.g. Heusler alloys) offers a reasonable solution. Good quality Heuslers are not impossible to make, but currently manufacturers cannot provide acceptable reproducibility of production as yet.

The idea of polarisation sensitive detectors using dynamically polarised He^3 is a sensational prospect for future generations of neutron scatterers.

FOTOF - Frame Overlap Time-of-Flight

The idea of FOTOF is to operate a time of flight system but deliberately allowing a large amount of frame overlap. In this way it is possible to reconstruct the time structure of the source pulse at a large distance from the moderator(~ 700 m for 2 x 10^3 m/sec (2Å), ~ 70 m for 200 m/sec (20Å) neutrons). The advantage of such a system is similar to the NSE technique: it allows us to work with an incident spectral width ($\Delta E/E \sim 20\%$) which is much wider than the energy resolution. The system can be used for quasi- and inelastic scattering and is complementary to NSE in that it gives results in the energy transfer domain rather than the time domain.

Instrumental Characteristics

Source frequency	Pulse width	λ Range	$\hbar\omega$	δE
4-500 Hz	20-25 μs	2-20Å	QE → 100 μeV	0.1 → 1 μeV

UCN Scattering

Maier Leibnitz showed that for a given size of resolution element at a given small value of Q,ω and for constant ΔQ,Δω one gains intensity by going to lower neutron energy, if one can use the entire allowed phase space. Currently an extension of the NESSIE spectrometer at ILL which uses this principle is under construction at Kyoto. A Neutron Resonance Spin Echo spectrometer using UCN which is coupled to a super-thermal (non-Liouville's theorem limited) cold source promises to open up significant new scientific fields. This prototype spectrometer could extend neutron scattering to higher resolution than has hitherto been achieved. NESSIE has been used to detect quasielastic scattering from a liquid surface which exhibits 10 peV/bounce - an unprecedented achievement.

Instrumental Characteristics

Source frequency	Pulse width	λ source	hω	δE
10-50 Hz	wide	6-20Å	QE → 100 μeV	0.1→ 1 meV

Neutron Resonance Spin Echo

NRSE or zero-field spin echo, is similar to NSE except that the magnetic fields which determine the resolution are confined to small geometrical regions (~5cm) instead of being applied over flight paths several metres long. In this manner the collection of scattered neutrons over large solid angles should be easier. This mechanism amplifies the effectiveness of the magnetic field by a factor of 4 and has much more flexibility than the standard techniques.

Because of its well-defined magnetic field boundaries the NRSE system offers undisputed advantages for studies of phonons with finite group velocities. The studies of phonon linewidths, which can give important new information on the electron-phonon interaction in superconductors, promises to be one of the most important areas of new physics in the future. In principle the NRSE method, when installed on a classical inelastic spectrometer, would allow the resolution ellipsoid to be aligned along the dispersion curve by adjusting the magnetic field boundaries obliquely along the incident and scattered beams. The NRSE method, with its two separated and well-defined rotatable magnetic field boundaries offers an undisputed opportunity for these measurements

Bunching of Polarised Neutrons

The bunching of neutrons serves to increase the intensity of TOF instruments operating in direct geometry. A well known example in which bunching is used is the IN6 spectrometer at the ILL which focuses neutrons of different velocities onto the detector in such a way that the intensity is increased without degrading the resolution. A technically very simple method of bunching polarised neutrons can be realised with the so called Drabkin resonator. First of all this device is used to rotate the spin of a well defined velocity band by π which is subsequently selected or separated by a polarised neutron analyser. All other neutrons are removed. If the two magnetic fields of the Drabkin resonator are changed in time in such a way that the focusing condition $n(t) = L_0/(t_0-t)$ is accomplished, then a wavelength band of about 10% in width can be focused generating an effective increase in count rate of at least a factor of ten. This was recently shown for a very short focusing length of 3.3 metre with a Drabkin resonator having a resolution of 1%. One drawback in this demonstration experiment was the inherent background below the focused line, which could be reduced to a level less than 10^{-3} by using more effective polarisers.

Neutron Magnetic Resonance Bunching System

Inside a resonance flipper, Zeeman energy can be exchanged between the neutron and the resonator system if the resonance conditions for spin inversion are fulfilled. The kinetic energy changes at the entrance and at the exit of the guide field B_0. It acts non-destructively on the wave function and therefore it represents a coherent optical component. Owing to the smallness of the Zeeman energy ($2\,\mu B_0 \sim 0.5\,\mu eV$), a multi-stage system will be needed with high flipping efficiencies and low depolarisation rates. To add the energy shift of the individual stages static flippers or a field inversion have to be applied between the stages. The spatial dispersion of the pulsed beams can be used to influence neutrons of different velocities differently by a time-dependent operation of the neutron magnetic resonance system.

The conditions for an effective use of such a device are strongly relaxed if the spatial separation is made by properly arranged backscattering crystals kept at different temperatures (similar to the MUSIC spectrometer principle).

Using the silicon (111) reflection in backscattering geometry feasible instrument parameters can be obtained for a primary flight path of 10m with an 8 degreetemperature difference between the crystals. By means of this method a quasi-continuous monochromatic beam can be produced whose intensity is n-times the intensity of neutrons in that energy interval in the original pulse. It provides a possibility of producing a stationary monochromatic and polarised beam with the luminosity of the peak intensity of the pulse. The whole system can be installed

inside and around a neutron guide tube. Various other possibilities arise if a multiple element crystal system and a proper energy transfer are also applied to the re-reflected neutrons.

An even more effective method exists if travelling magnetic potentials are applied synchronously with the neutron pulses. In this case the phase space density can be preserved from the source up to the sample position in the longitudinal direction also. This can result in gain factors up to 50.

Data Analysis and Visualisation

Probabilistic Bayesian Spectral Analysis, often referred to as Maximum Entropy, is just beginning to be applied to neutron scattering data. There is no doubt that data analysis methods in neutron work have traditionally been very primitive and some conservative in outlook, and particularly so when set beside the long accepted techniques used in other disciplines such as radio and optical astronomy and medical tomography.

Bayesian spectral analysis works on data in an objective manner aiming towards the physics, rather than by proposing subjective physical models and comparing these with the data. When as much reliable prior knowledge of the problem as possible is fed into the analysis, without prejudicing the objectivity, Bayesian analysis is a very powerful method of extracting the essential underlying images in neutron scattering patterns. This field will undoubtedly be facilitated by the continuing growth in the power of computers and the eventual elimination of out-of-date attitudes to admit powerful mathematical manipulation of data . An investment in the development of such techniques and their subsequent acceptance by the community will pay back handsome scientific dividends when properly applied at the ESS.

We cannot stress too strongly at this stage the contribution that advanced computational techniques will have on the use (and also design) of instruments in 10 years time. As well as the principle of Bayesian analysis there will also be direct modelling of the physical system which is refined using the data obtained. Work towards use of inelastic scattering data, as well as structural data, in forming an overall picture of structure and dynamics has already started. In a sense this would be a completely general extension of the methodology of CLIMAX and PHONON.

High Resolution Molecular Spectroscopy & CLIMAX

Climax is a normal coordinate analysis program used to extract the nuclear eigenvectors from molecular vibrational spectra. The program utilises the Bright-Wilson formulation and Valence

Forces, both familiar concepts to chemists. Infra red and Raman spectroscopic data are also able to be incorporated into the calculation, making a self-consistent approach possible.

The observed neutron band positions and intensities are fitted, using a Reitveld-type method. The vibrational fundamentals, their first overtones, and all combination modes are included in the calculation. The effects of external vibrational modes (phonons) are also included, although rather simplistically at the present time. Further progress is being made to model phonon spectra by the PHONON package. This approach is recognised as being the best available analysis program for small symmetric molecules, and pure solids (e.g. NH_4 salts) or absorbates (e.g. C_6H_6) are equally well represented.

Within the next few years the following improvements are anticipated.

- Inclusion of low symmetry, larger molecules such as biomolecules.
- Sophisticated analysis of multiphonon events.
- Treatment of data recorded along Q,ω cuts.

For the analysis of data from the ESS there is clearly a need to handle the dispersion of internal vibrations. A significant advantage is expected from the application of parallel processing methods to this approach.

Illumination of Neutron Guides on Pulsed Sources

For full illumination of neutron guides, it is necessary that the entrance of the guide is adequately close to the moderator face such that the cone of acceptance of the guide is covered completely by the moderator face. As the neutron wavelength increases, this cone expands and full illumination is then more difficult to achieve. Similarly the promise held out by guide coatings of significantly higher critical angle than nickel or nickel[58], such as supermirrors, can only be utilised on pulsed sources by installing the entrance apertures of the guides even closer to the moderator. Therefore moderators, which are relatively small on pulsed sources, must be optimised for face area without significantly degrading the surface flux.

We propose that neutron guides, fabricated from radiation hard thin nickel nose sections, should be sited in bunches as at ILL and not in individual beam pipes as at ISIS. Experience on ISIS has demonstrated that curved guides can adequately filter out the very fast neutrons and γ-rays from the spallation process so efficiently that 0.5 mm cadmium is quite sufficient to close the beam.

The geometrical bunching of the guides could consist of a 3 by 3 matrix of guides with beams being taken to 3 different experimental levels separated vertically by 2 to 2.5 metres. The guide entrance should be a *maximum* of 50 cm from the moderator face if complete matching is to be achieved with 20Å neutrons. For very cold neutrons (100Å and longer), which may be required for fundamental neutron physics measurements, the ability to site the guide even nearer to the face of the moderator should **not** be precluded.

CHINTZ - A chopper inelastic spectrometer

Specification

Resolution 5 μeV to 20 μeV variable
Multichopper monochromatic instrument
Energy transfer range 5 μeV to 2 meV
High Q with good Q resolution
Poisoned H_2 moderator
Optimum source frequency 100 Hz

Description

In order to tackle complex problems in QENS, high precision data at high momentum transfers with the ability to eliminate Bragg contamination, is an essential requirement. The Q resolution must therefore be very fine so that detectors containing coherent signal can be disregarded. Models for diffusional behaviour only become distinguishable in these high-Q regimes.

Quasi-elastic scattering using CHINTZ

The study of quasi-elastic scattering from atoms diffusing in a lattice gas have suffered from the limited Q range that can be reached with adequate resolution, using IN5/IN6/IN10. However the sharp resolution function of IN5 has considerable advantage when it comes to extracting more than one Lorentzian from the quasielastic line. This has produced unsurpassed data on incoherent H diffusion but it is now very important to extend the method to coherent QENS. For deuterium, one has to separate incoherent and coherent scattering and this has been done for NbD using spin analysis on D7 where the energy resolution unfortunately is only just good enough. This experiment successfully demonstrated the effect of $d\mu/dc$ (the rate of change of chemical potential gradient with concentration) on the chemical diffusion coefficient and also the fact that the broadening varies as $\Delta E_{c\text{-}e}S(Q)$ where $\Delta E_{c\text{-}e}$ is the corresponding Chudley Elliott broadening, and S(Q) is the diffuse scattering due to D-D

correlations. Correlation effects also influence the coherent broadening differently from the incoherent case. With improved resolution many other problems would be tractable.

CHINTZ will be ideal for single crystal QENS measurements as well as powders. Sharp resolution will be ideal for extracting a number of Lorentzian components from a complex spectrum as is found in samples with more than one site per unit cell. It would be possible to extend these measurements to superionic conductors and to non-stoichiometric oxides where the mobile species is diffusing through a static lattice. The detection of 10 μeV broadening will enable us to be sensitive to diffusion coefficients down to 10^{-7} cm^2/s. The quasielastic cross section is proportional to c(1-c) where c is the fraction of filled sites. If the static lattice is periodic, its scattering will appear on Bragg peaks, which can easily be removed if each detector represents a small increment in Q. For atoms heavier than hydrogen, modest heating will normally be necessary to establish observable broadening. Good resolution will also allow separation of the wings of the quasielastic line from the acoustic phonon scattering.

VIRUS - a very high resolution crystal analyser spectrometer

Specification

Resolution 0.1 to 0.2 μeV
Incident flight path 50m
Energy transfer range 0-100 μeV
Mica crystal analyser in exact backscattering λ_a = 19.8Å
Poisoned H_2 moderator
Optimum source frequency 50 Hz

Description

The effective use of 20Å neutrons on the present IRIS spectrometer, providing a resolution of 1μeV with count rates equivalent to IN10 at ILL, has recently been shown. It is therefore demonstrated that copious fluxes of cold neutrons can be generated on pulsed sources which rival the cold fluxes on reactors. The IRIS concept can now be optimised on the ESS by viewing a poisoned liquid H_2 moderator with a sharp pulse, and placing the mica analyser in exact backscattering. At 50 metres from such a moderator the incident and scattered resolutions would be matched, providing a resolution of 150 neV overall. The ESS instrument would have an energy transfer range of 100 μeV and an intensity an order of magnitude higher than IN10 even at this fine resolution, which would represent the highest direct resolution ever achieved by any neutron spectrometer.

The MUSIC Backscattering Instrument

One of the main limitations in high resolution backscattering spectroscopy is intensity, even with instruments at the highest flux sources. The situation becomes much worse if a backscattering spectrometer were to be used *in the conventional mode* at a pulsed spallation source. Such an instrument would be active only during the pulse duration Δt and it would be inactive during the rest of the pulse repetition time. Such an unpromising performance can be improved by replacing the Doppler drive of the reactor instrument with its single monochromator, by a more sophisticated multiple monochromator on a pulsed source. All other components of the reactor backscattering instrument could remain unchanged. In order to make full use of the peak flux of the pulsed source, the monochromator would consist of about 30 crystals, each with the same backscattering orientation but having slightly different lattice spacings *and* being mounted with a specified distance between each succeeding crystal, which would therefore reflect a slightly different wavelength. Due to the path length difference the different energy neutrons would arrive at the sample at different times. Putting this into numbers, one obtains for polished silicon crystals with $\Delta d/d \sim 2.10^{-5}$ and on a pulsed source with a moderator pulse length of 300 μs, the distance between the crystals, Δl, is found to be ~10cm. The necessary difference in lattice spacing can be obtained either by thermal expansion or, better, with mixed $Si_{1-x}Ge_x$ crystals. The atomic concentration x of germanium from one crystal to the next has to be changed by $\Delta x=0.0005$ in order to shift the reflection curve by one FWHM. With the given numbers the modified backscattering instrument is active all the time, if the repetition rate of the pulsed source is 50Hz.

When using 30 crystals, the dynamical range is also 30. This range can be extended to a larger value if mixed crystals of $Si_{1-x}Ge_x$ with $x>0.015$ can be produced.

Cold Neutron Moderators

There are two options for moderator material in ESS that are certain to provide suitable service: room temperature water and liquid hydrogen. Their suitability stems in part from the absence of permanent radiation damage effects, in that radiolytic decomposition products of water (hydrogen, oxygen, hydroxyl radicals, hydrogen peroxide, etc.) recombine into the original material, perhaps outside the irradiated volume. Decomposition products of liquid hydrogen recombine locally, but the low temperature form, parahydrogen, reverts to the normal ortho-para mix upon recombination with consequent effects on the thermalisation properties. As liquids, they can be circulated to remove the internally generated heat and the heat generated in their containers. At high internal power densities, solid moderators need to be circulated as suspensions in cryogenic coolants, supported on high conductivity metal substrates or otherwise innovatively cooled due to their low thermal conductivities.

Liquid hydrogen has a rather low proton density, about 70% that of water; it remains liquid to 15 K. Supercritical hydrogen exists above about 40 K and 5 atmospheres, has desirable heat transport properties and shows no physical distinction between liquid and gas phase (and therefore no bubbles). Thus liquid hydrogen offers low temperatures, convenient heat transport and stability against radiation damage and void accumulation and for these reasons is the default choice for the cold moderators of ESS.

As a consequence of its low proton density, the pulse widths from moderators of liquid hydrogen are broader than in more dense materials, about 5 microseconds at 1 eV and about 300 microseconds infinite medium decay time of thermalized neutrons. The "thermal" neutron energy distribution is only roughly maxwellian because of the abrupt dip in the total scattering cross section of parahydrogen below 15 meV. The best-fitted Maxwellian always has a temperature significantly above the physical temperature of liquid hydrogen because, when the neutron energy falls below 15 meV, neutron energy loss by rotational level excitation becomes impossible; only liquid-state broadened translational modes remain as means for absorbing energy. Liquid hydrogen moderators are subject to gradual change as the spontaneous ortho-para conversion progresses.

The negative features notwithstanding, liquid hydrogen moderators serve very satisfactorily in existing cold sources in both reactors (where liquid deuterium is sometimes the material of choice) and pulsed sources.

Moderators may be decoupled from the reflector or not. In the latter, coupled, case the highest overall intensity is produced, but the pulses are broader than in the former, at short wavelengths broadened by the entry of neutrons slowing down (with broader pulses) in the heavier reflector, at long wavelengths by the entry of neutrons from the long-lived population of (warm) thermal neutrons (subsequently re-thermalised) from the reflector. The slowing of pulses appears both as a lengthened tail and as a broadened rising edge. Decoupled moderators provide shorter pulses.

The width of the tail in the maxwellian region can be reduced by making the moderator smaller; this can be accomplished by diminishing the actual dimensions or by introducing heterogeneous poisons parallel to and beneath the viewed surface. Usually one thinks of poisoning decoupled moderators, but the effects of poisoning coupled moderators should also be investigated. Little data exists on the effect of poisoning liquid hydrogen, and in view of its potential desirability, a database should be developed.

The intensity of beams emerging from moderators can be enhanced by providing grooves on the viewed surface, by a factor of about 1.5. The benefit is accompanied by the appearance of structure on the rising edge which, however, can be removed at about 10% sacrifice in

intensity by shielding the tips of the grooves. The decay time of the tail is increased by the presence of grooves.

An attractive prospect which needs to be investigated, still within the context of liquid hydrogen, is H_2-D_2 (and HD as a consequence of radiolysis and recombination) mixtures. Such mixtures would have a total scattering cross section different from H_2 since its thermalisation properties include the influence of the smaller spacing of rotational levels in HD and D_2.

Solid hydrogen, which has not been tested as a pulsed source moderator, should be investigated; one could imagine a moderator of solid hydrogen cooled by liquid helium.

A "not-so-cold" moderator is liquid methane (CH_4), which solidifies at about 90 K. Its proton density is about equal to that of water and its inelastic scattering properties are superior to those of water and liquid hydrogen because of the presence of nearly-free, closely-spaced, and richly-plentiful rotational levels. Radiation damage effects in liquid methane lead to the production of hydrogen and heavier hydrocarbon polymers; their slow accumulation leads to clogging of plumbing and heat transport components of liquid methane systems. It may be possible to control this by providing sufficient overpressure to maintain hydrogen in solution (thus inhibiting the formation of heavy materials), or by continuous repurification. A way might be found periodically to replace obstructed components if the rate of accumulation of offending materials is not too great.

The best material for cold moderators, from the point of view of high intensity, short pulses and low spectral temperatures is solid methane, or possibly a mixture of $CH_{4-x}D_x$. Slightly less advantageous would be H_2O ice. Both these materials however have low thermal conductivity and exhibit unstable thermal behaviour during irradiation which would be very difficult to manage at the power density and radiation damage rate of ESS. If a way could be found to circulate finely divided solid methane or water ice in, say, liquid hydrogen, some of the advantages of solid methane might accrue to ESS.

Moderators consisting of radiation-stable materials with high proton densities (such as metal hydrides) which are themselves poor choices as cold moderator material because of the very tight binding of the principle scatterers (protons) might advantageously be incorporated into heterogeneous moderators cooled by liquid hydrogen and in which the viewed layer is liquid hydrogen.

These prospects should be evaluated in the process of ESS concept development.

Δt_{mod} Reduction Methods

Pulse widths and intensities can be adjusted by design in pulsed source moderators by several methods. Nature however usually prevails:- decreased intensity usually attends decreased pulse width. Two sets of factors can be considered: size of moderator, poisoning and decoupling options; and materials choices.

The pulse at short wavelengths depends mostly on the proton density of the moderator material. Allowing the reflector to communicate with the moderator increases the intensity available but broadens the moderated neutron pulses. A decoupler can be placed between the reflector and the moderator. The decoupling energy is chosen so as to preserve the width intrinsic to the moderator medium below the chosen decoupling energy (adjusted by choice of boron thickness, use of cadmium or gadolinium, etc. with different resonance energies). Usually the decoupling energy is chosen to be just above the energy at which the Maxwellian in the reflector diminishes to insignificance. This transmits as much as is allowable of the slowing-down distribution from the reflector, which is fairly narrow, but excludes the long-lived Maxwellian from also returning from the reflector. A very significant intensity decrease at all energies below the decoupling energy accompanies the decoupling, but a very significant pulse narrowing can be accomplished. Communication of the moderator with the reflector also broadens the rising edge of the moderated pulse.

Adjusting the size of the moderator affects the lifetime of the maxwellian population in the moderator; the smaller the moderator, the shorter the lifetime and the lower the intensity. The size effect can be accomplished by placing a sheet of material beneath the viewed surface of the moderator which is opaque to low energy neutrons and transparent to fast neutrons. Then the moderator looks small to low energy neutrons but big to higher energy neutrons which slow down with greater efficiency in large moderators than in small ones. This heterogeneous poisoning method is the standard way of adjusting the pulse length. Most of the loss in intensity comes about due to the shortened tail of the pulse at low energies, the peak intensity is lowered but not as strongly affected. The rise time is hardly changed at all.

Material with high proton density provides sharp pulses at "epithermal" energies (above the maxwellian range) and provides a sharp rising edge. Materials with good thermalising power (rich spectrum of low energy modes which can be excited by neutrons losing energy) provide sharply rising pulses.

Homogeneous absorption narrows the pulses at all energies, but decreases the intensity more severely than does accomplishing the same effect by heterogeneous poisoning.

The most obvious variable parameter is the moderator temperature. The lower the moderator temperature, the lower the energy at which the long-lived Maxwellian ("storage") component of the population becomes significant in relation to the short-lived slowing-down component. Thus the range of energies in which the short pulses, characteristic of the slowing-down distribution, dominate the time distribution, can be extended to lower energies by cooling. "Moving the Maxwellian out of the way" in this manner benefits the resolution of instruments that use the intermediate range of energies, and is a motive for cooling independent of the desire for increased intensity of low energy neutrons. The Maxwellian neutrons are lost in the intermediate range of energies and appear at lower energies, and the rise time and the peak intensity tend to remain unchanged at each energy in the intermediate energy range. Accompanying cooling in most materials is a desirable increase in proton density.

The methods above apply to the moderators themselves and their surroundings, and thus affect all the beams extracted from their surface.

The long tail associated with the Maxwellian could possibly be removed from pulses by devices in the neutron beam. The obvious idea of a chopper is only applicable when it is desired to monochromate also, since it is impractical to place a chopper close to the source, because of radiation damage. In any case a tail-cutting chopper only operates as such when it is close to the moderator before the neutron pulse has been able to disperse. Even a monochromating chopper is ineffective in tail-cutting: it succeeds only in inverting the pulse shape in time.

A rotating single crystal filter, placed some distance from the moderator and arranged to revolve so that its angle satisfies the Bragg condition at all wavelengths of interest, can remove neutrons on either side of the maximum of the pulse, and might therefore be provided to control the width of the pulses by tracking along the ridge of the pulse as a function of energy. Such a filter would have to revolve non-uniformly in time but this can be achieved within tolerable limits and should be investigated as a means for controlling the width of pulses in individual beams. The phasing of such a "pulse trimmer" could possibly be adjusted to enable the resolution and intensity of an instrument downstream to be varied.

4 Recommendations for R & D

To be able to implement the instruments identified by the group, and to carry out the science described above it is essential that in the intervening period before the source is funded and built that a number of specifically identified R & D projects be studied.

It is also necessary that those many people in Europe with vast expertise in reactor-based instrumentation should be supported to allow them to take the fullest advantage of the learning opportunities presented by the pulsed sources currently operating in the world, and notably ISIS.

The high resolution and molecular spectroscopy group have identified the following R & D projects:-

1. Investigating the feasibility of NSE techniques on pulsed sources.
2. Initial investigations into polarisation analysis methods on inverted geometry spectrometers.
3. Investigating the feasibility of bunching techniques.
4. The use of liquid hydrogen as a higher order suppression filter for neutron energies >14meV.
5. Investigating dynamic methods of polarising nuclei in samples.
6. The realisation of synchronously applied pulsed fields, shock waves, and intense laser bursts.
7. Investigating the use of solid hydrogen moderators.
8. Creating a database of information on poisoning of liquid hydrogen moderators.
9. The use of hydrogenous samples exhibiting low-lying tunnelling excitations (such as methane hydrates) as moderator materials for very cold neutrons.
10. The development of UCN moderators.
11. The development of rotating crystal pulse tail-cutters.
12. Development of mica and synthetic multilayers (such as Langmuir-Blodgett films) as long wavelength analysers
13. An investigation of radiation hard neutron guides to be placed very close to the moderator faces.
14. An investment into statistical methods of data analysis and fast data visualisation techniques.
15. The development of mixed crystal analysers of $Si_{1-x}Ge_x$ with $x>0.015$.

16. The development of Heusler alloy crystals with reproducible quality.

17. Encouragement of transfer of supermirror technology to pulsed source laboratories.

5 Instrumentation for the European Spallation Source

A summary of instruments required on the ESS to implement the broad programme of scientific work outlined above is given in Table 1.

6 Conclusions

The high resolution and molecular spectroscopy group strongly believes that we are on the verge of a new era in neutron spectroscopy similar to that which was initiated with the advent of the ILL. Building upon the solid advances at ILL and the success of the instruments at ISIS, the technological methods have been matured and the scientific climate is now ripe to capitalise upon the significant instrumental improvements which the ESS will usher in. There is now a large reservoir of scientific expertise available with the energy, vision and expertise to construct ESS and keep Europe as world leader, thereby creating a first-class centre for neutron scattering which will be pre-eminent until the middle of the 21st century.

Table 1. Instruments proposed for ESS. High Resolution and Molecular Spectroscopy Group.

Instrument	ν (Hz)	Δt (ms)	λ (Å)	δE (meV)	ħω (meV)	Q (A^{-1})	Features
1.Musical	50	300	6-6.5	0.1	3	0.01min	CH
2.Virus	50	60	15-25	0.1-0.2	100	0.02min	DH
3.NSE*1	15	>300	5-20	0.0005	10	10^{-4}	CH
4.NSE*2	50	25-100	1-4	0.5	10-10meV	0.01min	CH
5.Cassandra	15	10-80	0.8-5	0.25%	100meV	n/a	Str Guide CH
6.Chintz*1	100	100	3-15	5 ≡ 3A	2meV	0.1-3	DH
7.Chintz*2	50	300	1-7	20	15	0.2-4	CH
8.Opera	100	100	5-7	0.1	1	0.2-2	DH
9.Bunching	100	300	6.28	50	500	0.15-1.8	DH
10.Pyrrhus	50	120	4-8	15	2 meV	0.2-1.8	PA: CH
11.Iris	50	120	2-30	1-50	10meV	0.05-3.6	CH
12.NRSE	25	150	1-20	0.1	1 meV	0.01min	CH
13.Fotof	400	25	1-6	<1	n/a	n/a	CH
14.Diffractospec	50	120	2.3-20	$8/3\ 10^{-4}$	2 meV	0.2-1.8	CH
15.UCN	10	400	10-250	neV	1	10^{-6}	CH
16.FundamPhys	10	150	4-25	n/a	n/a	n/a	CH
17.Recoil	100	25	0.1-2	50meV	0.01-10eV	20-300	CH4
18.HiIntCrysAnal	50	250	4-8	25	2	0.2-1.8	CH
19.Interferometer	25	100	4-10	10^{-6}	1	10^{-3}	CH
20.Chrysalis	50	25	0.2-8	0.7%	1 - 500meV	n/a	CH4