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IRIS & OSIRIS International Beamline Review - Report Presented to the Panel

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Foreword

The ISIS Facility at the STFC Rutherford Appleton Laboratory is a major international centre for neutron scattering and muon spectroscopy. To ensure that the instrumentation suite is kept at the forefront of capability, the ISIS Director has instituted a series of rolling reviews across the instrument suite, such that every instrument will be reviewed by an international panel every 10 years or when a major upgrade is required. Following an earlier review of the TOSCA spectrometer in 2013 (see *RAL-TR-2013-015*), this report contains the documentation presented to the *IRIS & OSIRIS International Beamline Review* panel in November 2014.

IRIS & OSIRIS International Beamline Review – Report Presented to the Panel

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Executive Summary

IRIS & *OSIRIS* have led the way in high-resolution neutron spectroscopy using cold neutrons at pulsed spallation sources. Their main strengths lie in the provision of high spectral resolution over a wide dynamic range, synchronous high-resolution diffraction, low instrument backgrounds, a versatile range of sample environments, and user-friendly operation. These instruments currently support a productive scientific community using quasielastic and low-energy inelastic neutron scattering techniques, as evidenced by an output of 200+ publications since 2008. For *IRIS* & *OSIRIS* to move forward in both a *UK* and international context, full integration of computational materials modelling into our science programme constitutes the highest priority at the present time and would require further investment in staff and high-performance computing infrastructure. Additional instrument developments on the horizon include an increase in energy resolution and flux on *OSIRIS* using a silicon analyser and a new primary spectrometer, as well as a step change in the scientific remit of *IRIS* so as to provide high-throughput spectroscopic capabilities for emerging research themes in materials chemistry, soft matter, and biology.

Technical specification

IRIS & *OSIRIS* are two high-resolution indirect-geometry quasielastic (*QENS*) and inelastic (*INS*) neutron-scattering spectrometers with long-wavelength diffraction capabilities. They share the *N6* beamline in *ISIS Target Station 1*, viewing a liquid-hydrogen moderator cooled to 25 K. Both instruments work on the principle that neutrons scattered by the sample are energy-analysed by means of Bragg scattering from a crystal-analyser array. As on other *ISIS* instruments, they use time-of-flight methods for energy analysis.

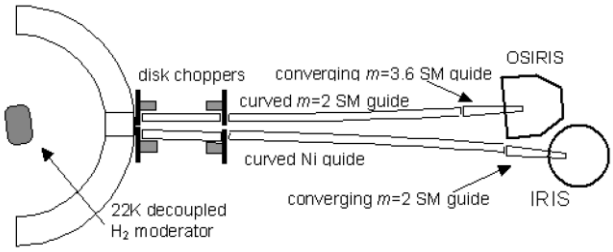

Figure 1. Schematic diagram of the IRIS & OSIRIS primary spectrometers.

As shown in ***Fig. 1***, neutrons are transported to *IRIS* & *OSIRIS* by curved neutron guides. *IRIS* is a day-one instrument operating since 1984 and still uses a natural-*Ni* guide terminated by a *Ni-Ti* (m=2) supermirror focusing guide. Over the years, it has seen significant upgrades to its analyser banks, vacuum tank, and detectors. *OSIRIS* has been operational since 1997 and uses an m=2 guide terminated by an m=3.6 focusing section in its primary spectrometer. Its original design and construction was supported by the *EPSRC* as well as *Spain, Sweden, India, and Switzerland.* Collaborative efforts with *Spain* continue to this day, particularly in instrument and technique development. In the early days, *OSIRIS* was used as a specialised diffractometer for long-d-spacing studies. In 2003, the instrument was equipped with a large-area, cooled pyrolytic graphite (*PG*) analyser, paving the way for its use in *QENS* and *INS* research. In 2013, its spectroscopic capabilities were upgraded via the provision of a retractable and cooled beryllium (*Be*) filter in the secondary spectrometer.

The *IRIS* & *OSIRIS* secondary spectrometers are illustrated in ***Figs. 2-3*** below. Both instruments are equipped with two disc choppers at *ca.* 6 and 10 m from the moderator so as to define the incident wavelength band and prevent frame overlap. Wavelength-band selection over the range 0.9−20 Å defines energy-transfer and d-spacing ranges for spectroscopic and diffraction studies, respectively. Both spectrometers use Bragg scattering from cooled *PG* crystals in so-called *near-backscattering* geometry to define a final energy of 1.84(7.38) meV for *PG002(004)* reflections, with a resolution of 17.5(54.5) μeV and 25(99) μeV on *IRIS* & *OSIRIS*, respectively. The primary setting in both instruments is *PG002* up to energy transfers of 20 meV in neutron-energy loss *via* the use of a movable and

cooled *Be* filter to suppress higher-order reflections. Neutrons are detected over the angular range 11-150°, leading to a momentum-transfer range Q = 0.2-1.8(0.4-3.6) Å^{-1} for *PG002*(*004*). *IRIS* is additionally equipped with a *Mica* analyser bank (muscovite or fluorinated) to achieve a higher energy resolution of 11 μeV (*006* reflection, final energy of 1.86 meV), 4.5 μeV (*004*, 0.83 meV), and 1.2 μeV (*002*, 0.21 meV) albeit at a much-reduced flux. Furthermore, it has a bank of 7 diffraction detectors at $2\theta \approx 170°$. In comparison, *OSIRIS* has a 1 m^2 diffraction bank (960 pixels) arranged in a ring around the incident beam at 2θ = 150-171°, with a resolution as high as $\Delta d/d = 2.5\times10^{-3}$. *OSIRIS* also has a larger *PG* analyser bank than *IRIS*. The larger analyser and a supermirror guide provide higher count rates on *OSIRIS* relative to *IRIS*, particularly at the shorter wavelengths. Recently, extensive *Monte Carlo* simulations (*RAL-TR-2013-008*) have provided detailed insights into the different contributions to the resolution and line shape of *OSIRIS*. These studies represent a solid starting point for a quantitative description of *QENS* and *INS* spectra across the momentum- and energy-transfer ranges of the instrument, as well as for the assessment of potential upgrade paths on both spectrometers.

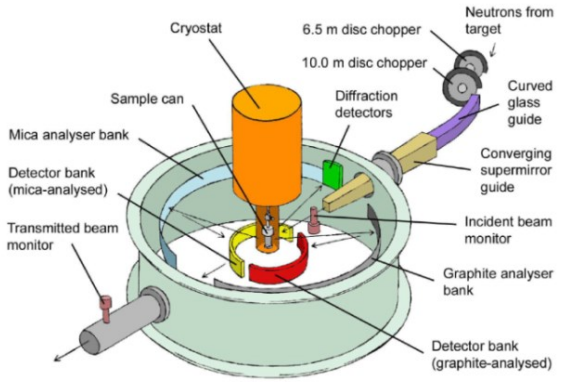

Figure 2. Schematic diagram of the IRIS secondary spectrometer.

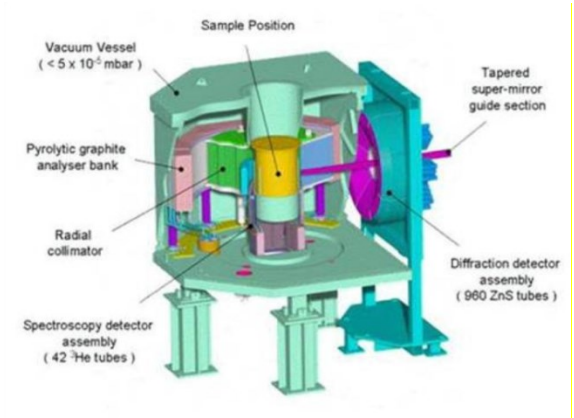

Figure 3. Schematic diagram of the OSIRIS secondary spectrometer.

IRIS & *OSIRIS* have been the backbone of cold-neutron spectroscopy at *ISIS* from the outset, providing high resolution, a flexible energy-transfer range, and good flux. They also provide simultaneous high-resolution diffraction capabilities. A similar spectral resolution can be achieved over a narrower Q-range on the newer *LET* spectrometer in *ISIS Target Station II*. *LET* can also achieve a higher energy resolution at complementary (smaller) Q-values, yet at the price of a reduced flux at these colder incident wavelengths. Envisaged as versatile instruments, *IRIS* & *OSIRIS* accept a wide range of sample environments (closed-cycle refrigerators, furnaces, high-pressure equipment, dilution fridges, gas handling manifolds, *etc*.) and samples (powders, films, liquids, single crystals). On *OSIRIS*, it is also possible to apply magnetic fields up to 7.5 T.

The scientific context of the instruments

The science programme on *IRIS* & *OSIRIS* spans a broad range of disciplines requiring access to low energy transfers with high spectral resolution and over a wide dynamic range – see ***Fig. 4***. Although both instruments have not been fully staffed for the most part of the past decade, they have been productive over their lifetimes: around 700 publications since *ISIS* started operations in 1985 and 200+ since 2008 (see also ***Annex I***).

On *IRIS*, chemistry and materials science constitute *ca.* two thirds of the user programme, followed by physics (21%) and biology (13%) – *cf.* ***Fig. 4***. Soft condensed matter (including biological systems) and nanoporous materials have become increasingly important cross-cutting research themes in the past few years. In soft matter, relaxation phenomena in amorphous polymer systems (pure, blends, nanocomposites), foodstuffs, dental cements, surfactants, and lipids fall within the timescales probed by the spectrometer. Water dynamics is also a central theme in soft-matter and biological studies, as evidenced by a plethora of investigations on biomolecular function, from model systems like lysozyme to *in vivo* studies on *E-coli* cells.

In the area of nanoporous materials, a growing number of studies on *IRIS* have examined molecular diffusion and transport in layered materials of environmental importance, water in cements, or molecular uptake and storage in graphite-based nanostructured materials or metal-organic frameworks (*MOFs*). This work has been facilitated by the superb provision of gas-handling

equipment at *ISIS* and the complementarity of parallel studies on *TOSCA*, which accesses the elastic line as well as higher energy transfers.

OSIRIS has evolved differently relative to *IRIS*. During its first six years (1997-2003), *OSIRIS* operated solely as a diffractometer and this area of activity is still highly productive, although the allocated beam time for diffraction (both nuclear and magnetic) has dropped below 10% following the availability of *WISH* in *ISIS Target Station II*.

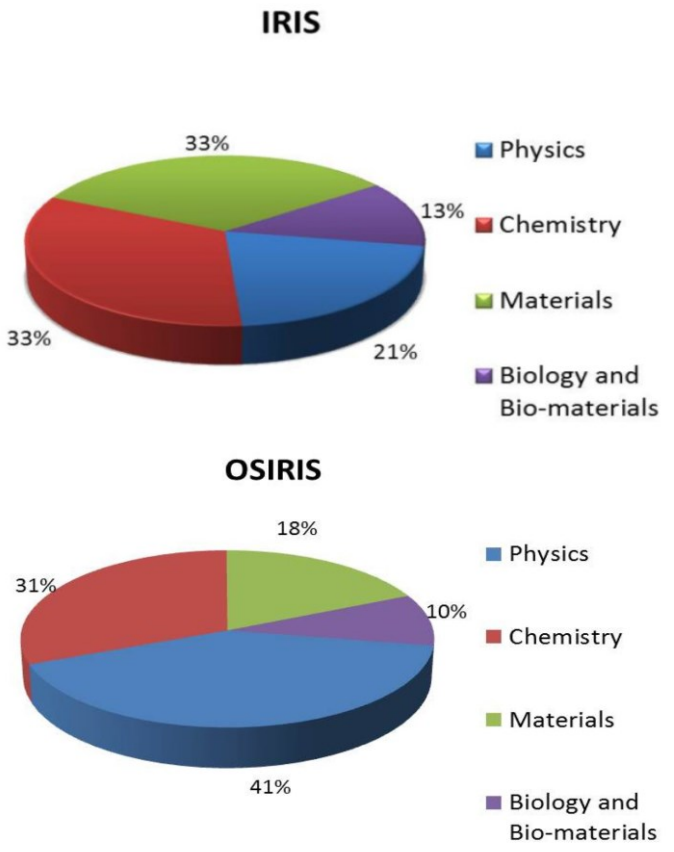

Figure 4. Usage of IRIS & OSIRIS by scientific discipline.

On *OSIRIS*, ~40% of the requested beam time comes from the condensed-matter-physics community, as the instrument provides unique capabilities for high-resolution cold-neutron spectroscopy to study single crystals at milliKelvin temperatures in the presence of applied magnetic fields. Recent studies in this area include the emergence of exotic quantum symmetries in low-dimensional magnets, as well as an increasing number of detailed line shape studies.

Chemistry and materials science constitute *ca.* 50% of the requested beam time on *OSIRIS*. *QENS* studies have ranged from molecular systems showing liquid-liquid phase transitions and room-temperature ionic liquids, to semiconductor alloys used in the manufacture of *DVD* discs. Energy-related research has focused on diffusion phenomena in thermoelectrics and ionic conductors, or in porous carbons for hydrogen storage. As on *IRIS*, recent (and growing) themes include the study of molecular transport in nanoporous materials for catalysis, work primarily carried out in collaboration with the *UK Catalysis Hub* at the *Harwell Research Complex*. An increasing number of these investigations have been accompanied by computational materials modelling, particularly those based on first-principles methodologies. This work has required extensive use of high-performance computing resources, including the *STFC*-funded *SCARF-RAL* cluster as well as external facilities such as *HECToR* and *ARCHER* in the *UK*, both available to us through the *EPSRC HPC Materials Chemistry Consortium*.

Xpress access was introduced on both *IRIS* & *OSIRIS* in the Summer of 2011. This access mode is primarily aimed at attracting new scientific communities, assessing feasibility, or completing experiments. The scheme has proven both popular and successful, with 60+ samples measured to date and 20+ new users to the technique.

The international context of the instruments

As a day-one *ISIS* instrument, *IRIS* was the first to provide routine access to low energy transfers with high resolution at a spallation source. At present, *OSIRIS* has superseded *IRIS* in most respects, owing to its higher flux and much-improved diffraction capabilities. The use of *PG* analysers limits the resolution of both instruments to around 20 μeV and, thus, the most recent spallation sources have chosen to use *Si* crystals (*111* reflection) to achieve a resolution of 2-3 μeV (*DNA* at *JPARC*, *Japan*; and *BASIS* at *SNS*, *USA*). *Si111* has also been the Bragg reflection of choice for backscattering spectrometers at reactor sources, affording a resolution down to the sub-μeV range and good flux, although with a limited energy-transfer range, typically up to 30 μeV (*IN16B* at *ILL*, *France*; *HFBS* at *NCNR-NIST*, *USA*; or *SPHERES* at *FRMII*, *Germany*). To circumvent this limitation, a time-of-flight option called *BATS* has been proposed for *IN16B*. At present, the high resolution afforded by these reactor-based backscattering instruments can only be approached with the *IRIS Mica* analysers, yet at the expense of prohibitively low detected fluxes, *i.e.,* two orders of magnitude lower than *PG*.

Using long incident wavelengths, spectral resolution as low as 5-10 μeV can also be achieved with direct-geometry spectrometers (*LET* at *ISIS*; *IN5* at *ILL*; *DCS* at *NCNR-NIST*; or *TOFTOF* at *FRMII*), although this choice is necessarily accompanied by a substantial reduction in flux and *Q*-range. At energy transfers up to 10-20 meV, indirect-geometry backscattering spectrometers like *IRIS* & *OSIRIS* provide an outstanding resolution (*ca.* 100 μeV at 15 meV) relative to direct-geometry spectrometers, only outperformed by single-detector-based, spin-echo techniques on three-axis spectrometers (*TRISP* at *FRMII* or *IN20-TASSE* at *ILL*). Further into the future, *ESS* in Sweden is currently assessing the case for *MIRACLES*, a high-flux and wide-dynamic-range backscattering spectrometer with a similar energy resolution to that provided by *BASIS* or *DNA* (*i.e.*, few μeV). At present, it is difficult to provide a robust appraisal of potential developments by *ESS* over the next 5-10 years, yet at the same time we can also see ample scope for joint collaborative efforts in the foreseeable future.

Scientific highlights

The tight resolution and wide dynamic range afforded by *IRIS* & *OSIRIS* at meV energy transfers have enabled a number of important studies of low-energy excitations in ***strongly correlated electron systems***. These include the elegant experiments on quantum criticality in an Ising chain by Coldea *et al.* [*Science* **327** 177 (2010)], or the emergence of spin-exciton Bose condensates in the superconductor $CeCoIn_5$ at high magnetic fields by Stock *et al.* [*Phys. Rev. Lett.* **109** 167207 (2012)]. In both cases, subtle spectral splittings and shifts were carefully monitored on *OSIRIS* as a function of external stimuli. Similar considerations apply to recent *IRIS* studies of frustration in nanomagnets by Baker *et al.* [*Phys. Rev. B* **86** 064405 (2012)], or of magnetic excitations in frustrated rare-earth magnets by Quintero-Castro *et al.* [*Phys. Rev. B* **86** 064203 (2012)]. Beyond spectral line positions, Tennant *et al.* [*Phys. Rev. B* **85** 014402 (2012)] have examined the effects of thermal fluctuations on the dynamical response of copper nitrate, a gapped quantum magnet. Such a task required a detailed analysis of spectral line shapes and their associated temperature dependence. From these data, it has been possible to differentiate between intra-band processes akin to Villain scattering in *1D* soliton systems versus one-magnon contributions.

At the ***nanoscale***, the formation of hydrogen-bond networks in ash from sugar-cane residuals (greener alternatives to traditional cements) has been investigated by Jacobsen *et al.* [*Sci. Rep.* **3** 2667 (2013)]. In this particular case, the *IRIS* data established that water motions occur within interlayer distances of 2-4 Å, and that the hydrogen atoms bound to the underlying matrix cannot diffuse freely, resulting in a decrease in water permeability as required for practical applications. Using a similar methodology, Bhowmik *et al.* [*Macromol.* **47** 304 (2014)] have examined the dynamical behaviour of poly(methyl) methacrylate soft nanoparticles dispersed in the ubiquitous polymer poly(ethylene oxide) using partially deuterated specimens. The α–methyl–group motions in *PMMA* seen on *IRIS* become faster than in the bulk, an effect which is explained in terms of plasticisation effects induced by the polymer-oxide chains.

In the study of ***molecular adsorption on surfaces and nanostructured materials***, Lovell *et al.* [*Phys. Rev. Lett.* **101** 126101 (2008)] have used *IRIS* and *TOSCA* to monitor *in situ* the uptake of H_2 in the graphite intercalate KC_{24}. From the high-resolution diffraction data collected on *IRIS*, it is seen that molecular adsorption leads to a significant expansion of the substrate, in agreement with first-principles predictions. These calculations can also account for the underlying symmetry of the H_2 orientational potential inferred from the *IRIS* and *TOSCA* data, provided that the molecular centre of mass is allowed to delocalise across three vicinal sites, a dominant and hitherto unexplored quantum-mechanical effect in porous materials. In a similar vein, Larese *et al.* [*Phys. Rev. Lett.* **101** 165302 (2008)] have studied the formation of H_2 films on MgO surfaces using *OSIRIS*, *TOSCA*, and first-principles modelling. The neutron diffraction data provide direct access to the reduced-dimensionality structures of these molecular films. At monolayer coverage, H_2 behaves like a quasiplanar rotor, and recovery of a bulk-like response requires 3-5 molecular layers. Beyond small molecules, Huang *et al.* [*Phys. Chem. Chem. Phys.* **11** 2869 (2009)] have looked at the docking of 2-methyl butane and *n*-pentane in the zeolite *MCM-22*. In combination with classical molecular dynamics (*MD*) simulations, they found that *n*-pentane moves more efficiently across zeolite supercages, an effect which is accompanied by significant internal molecular reorientations. These results explain at the microscopic level why monobranched alkanes like 2-methyl butane are

absorbed preferentially over their linear counterparts in these framework materials. Most recently, this line of work has been successfully extended by Yang *et al.* [*Nature Chem.* (accepted, 2014)] to the uptake of acetylene and ethylene in the nanoporous host *NOTT-300*. In combination with diffraction and computational modelling, *IRIS* and *TOSCA* data provide unambiguous spectroscopic signatures of cooperative-binding effects in mixtures of these two hydrocarbons, ultimately leading to a high selectivity and uptake capacity.

Investigations of ***advanced materials*** using *QENS* and diffraction on *IRIS* & *OSIRIS* have ranged from studies of ionic conductivity in the super-protonic conductor $H_3OSbTeO_6$ [Boysen *et al., J. Phys. Chem. Solids* **73** 808 (2012)], atomic and molecular transport in complex borohydrides [Remhof *et al., Chem. Phys.* **427** 18 (2013)] or the lithium nitrides [Bull *et al., Faraday Discuss.* **151** 263 (2011)] for hydrogen-storage applications, and the structure and underlying magnetism in layered iron-arsenide superconductors [Cortes-Gil *et al., Chem. Mater.* **23** 1009 (2011)]. Reliable first-principles calculations of materials properties using state-of-the-art methods have provided a robust link between experimental observables and materials properties and performance, as illustrated by recent work by Mukhopadhyay *et al.* on the above-room-temperature organic ferroelectric croconic acid [*Phys. Chem. Chem. Phys.* (in press, 2014)].

In the area of ***soft and biological materials***, Stadler *et al.* [*J. R. Soc. Interface* **8** 590 (2011)] have used *IRIS* to probe the *in-vivo* dynamics of haemoglobin in human erythrocytes. The observed crossover from global, short-time protein dynamics to long-time self-diffusion agrees well with hydrodynamic predictions. Moreover, higher water levels are seen to facilitate protein fluctuations in the picosecond regime compared to fully hydrated protein powders. At shorter length scales, Gerelli *et al.* [*Soft Matter* **7** 3929 (2011)] have embarked on a detailed survey of the conformational dynamics of lipid-based vesicles, evincing the presence of dynamical heterogeneities conforming to a quantitative model with three distinct hydrogen populations. The interaction of these vesicles with charged polysaccharides leads to a slowdown of relaxation phenomena associated with the head groups without significant effects on isomerization processes. At a molecular level, Magazù *et al.* [*J. Chem. Phys.* **132** 184512 (2010)] have used both *OSIRIS* & *IRIS* to examine the properties of mixtures of the glass-forming bioprotectants glycerol and trehalose. In conjunction with *MD* simulations, these experimental results show that relatively low glycerol concentrations give rise to the formation of a stronger hydrogen-bond network with a significant suppression of fast and localised diffusive motions.

Current status and staffing

Three members of staff are currently associated on a full-time basis to *IRIS* & *OSIRIS*, and both instruments have been operating at or below ¾ of their capacity since 2010. Recruitment of a fourth member of staff with experience in disordered matter and exposure to materials modelling would be necessary to return to full operational levels. Other staff and resource requirements to increase both capacity and capability on these instruments are described below in ***Future technical and scientific developments***.

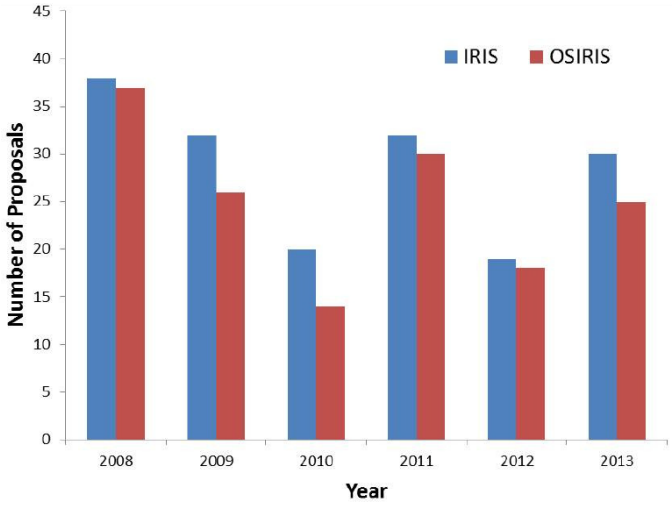

Figure 5. Beam time requests on IRIS & OSIRIS.

Statistics of use

As shown in more detail in ***Fig. 5***, there were a total of 321(209) proposals submitted(accepted) during the period 2008-2013, 171 on *IRIS* and 150 on *OSIRIS*. The total number of distinct principal investigators (*PIs*) was 161 from 23 countries.

Over this period, the *UK* user base across both instruments spans 79 distinct *PIs* across top-ranking higher-education institutions (*HEIs)* including *Cambridge*, *Edinburgh, Imperial*, *Oxford,* or *UCL* − see ***Fig. 6*** below and ***Table A2.1*** in ***Annex II***. Of this grand total, ~50% have been first-time users, primarily using

QENS techniques (~75%). On a per-instrument basis (~40 *UK PIs*), these figures compare favourably with a fully staffed and successful instrument like *TOSCA*, currently supporting 48 *UK PIs* (see *Table 1* in the *TOSCA International Beamline Review*). From these data, we envisage that a further increase in our user base would be possible with increased capacity (particularly staff effort) and a number of developments, as detailed in ***Future technical and scientific developments*** below.

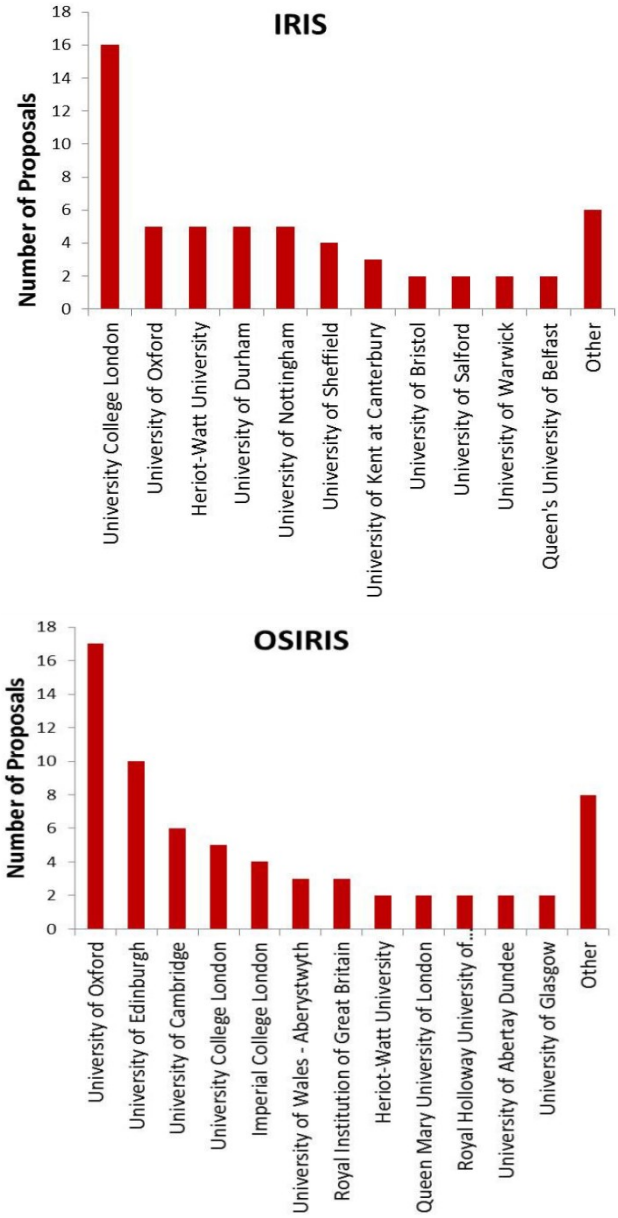

Figure 6. Number of proposals submitted by UK HEIs over the period 2008-2013. Additional data are given in Annex II.

In terms of sponsorship, about 40% of all proposals submitted by *UK PIs* on *IRIS* and *OSIRIS* have been supported by *EPSRC* grants and ~5% involved *Programme Access*. The remainder either do not state a funding source or are supported by *STFC* via the *Direct Access* route. A number of academic *PIs* have direct links or are supported by industry, including *General Motors* (gas storage in *MOFs*), *AWE* (*H/D* separation), or *Cella Energy* and *Toyota* (hydrogen storage).

At an international level, ***Fig. 7*** and ***Annex II*** show that the user base on *IRIS* (51% *UK*) spans a wider geographical area than *OSIRIS* (65% *UK*). Comparatively, *OSIRIS* received more proposals from the *USA*. Over 80% of proposals across both instruments come from the *European* user base.

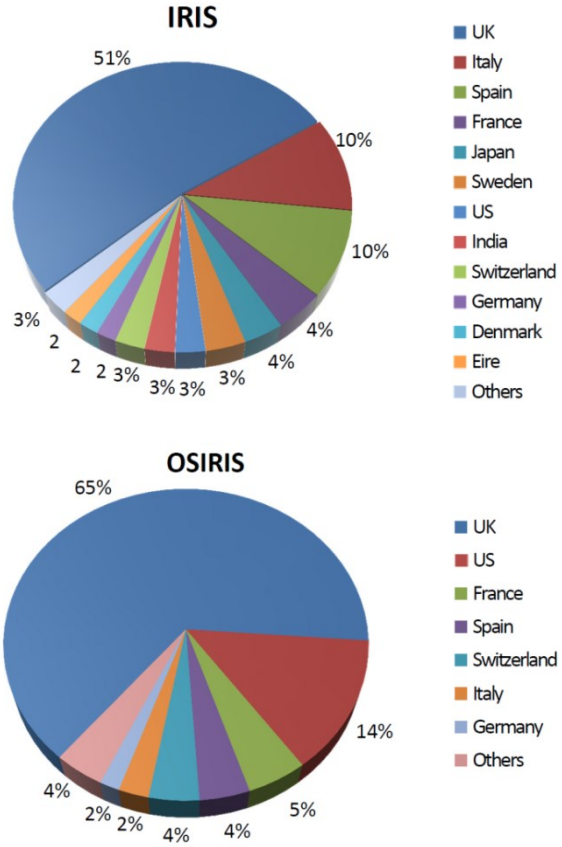

Figure 7. Proposals by nationality over the period 2008-2013. Additional data are given in Annex II.

IRIS & *OSIRIS* proposals are reviewed across several *ISIS Facility Access Panels* (*FAPs*): 52% on *FAP5* (*Molecular Spectroscopy*); 25% on *FAP4* (*Excitations*); 15% on *FAP2* (*Liquids*); and 8% on *FAP1* (*Diffraction*). *QENS* research maps almost entirely onto *FAP5* and *FAP2* (67%), whereas *FAP4* and *FAP1* primarily deal with magnetic excitations (25%) and diffraction (8%), respectively − see ***Fig. A2.2*** in ***Annex II***. Following the *OSIRIS Be*-filter upgrade, the majority of beam time requests have been scheduled interchangeably across both instruments except for experiments requiring

applied magnetic fields (*OSIRIS*) or the *Mica* analyser (a rare occurrence on *IRIS*). Since the addition of *LET* to the *ISIS User Programme,* about 20% of *LET* beam time has been awarded by *FAP5* and *FAP2* to *QENS* research. For these science areas, a dramatic increase in demand for *LET* above these levels (*i.e.,* >30%) is not expected in the foreseeable future.

As shown in ***Fig. A2.3*** in ***Annex II***, the average number of days requested on *IRIS* is around 6. The average for *OSIRIS* is also 6 days/experiment, yet this instrument features a broader underlying distribution reflecting a wider range of scientific requirements. The lowest end of the distribution corresponds to diffraction experiments, whereas the upper end is dominated by single-crystal spectroscopy. *QENS* experiments typically fall between these two extremes.

Xpress access to both *IRIS* & *OSIRIS* has been a successful venture. Around 2 days of *Xpress* time are allocated on each instrument per cycle and demand continues to be on the rise − over the past year, *Xpress* has supported 15 distinct research groups, ~60% from the *UK* and ~50% new to the technique. Since its inception, around 50% of *Xpress* runs have resulted in subsequent full beam time applications across the board, from biological systems to magnetic materials. The remaining beam time has been used to attract new users (30%), or to complete experimental campaigns required for the submission of theses or papers (20%). Recent *IRIS Xpress* data to determine the Mott gap in CoO have led directly to publication [R.A. Cowley *et al.*, *Phys. Rev. B* **88** 205117 (2013)].

IRIS & *OSIRIS* are productive instruments, with a total of 207 publications from 2008 to date (see ***Annex I***). The overall ratio of *refereed-publication-to-accepted-proposal* over the period 2008-2013 is 0.72 and 0.58 for *IRIS* & *OSIRIS*, respectively. The average value across both instruments amounts to 0.64, a figure which rises to 0.79 if theses and reports are also taken into account in the calculation. In terms of the training of early-career scientists, over 25 theses have resulted from work on the instruments from 2008 and a one-day *QENS practical* is run annually on *OSIRIS* as part of the *ISIS Neutron Training Course*. *IRIS* & *OSIRIS* data from elastic-window scans have also been earmarked to be part of the forthcoming *neutron Dynamics Data Bank* (*nDDB*).

Future technical and scientific developments

Data from low-energy neutron spectroscopy experiments (*QENS* in particular) have been traditionally interpreted using a limited number of phenomenological and heuristic models. To date, the use of computational materials modelling along with *IRIS* & *OSIRIS* data has been restricted to a small subset of (quite favourable) situations, as illustrated earlier in this report. Advances in materials modelling methodologies and computational power over the past decade offer a number of largely untapped opportunities to facilitate the analysis and subsequent interpretation of experimental data. Some progress has already been made in this direction with the recruitment in 2012 of a new member of staff with expertise in materials modelling.

Given the physico-chemical phenomena amenable to investigation on *IRIS* & *OSIRIS*, direct recourse to time-dependent techniques becomes necessary, *i.e.*, *MD*. In this case, atomic trajectories obtained from *MD* simulations are Fourier transformed for direct comparison with measured scattering functions. These time-dependent simulation tools are also of direct relevance to the analysis of neutron data at higher energy transfers. To date, the computation of vibrational or recoil neutron spectra has relied almost entirely on time-independent (phonon) calculations in periodic media. The use of time-dependent methods represents the natural starting point to overcome these limitations, as recently highlighted in the *TOSCA International Beamline Review* and the final *recommendations* of the review panel. We also anticipate that the use of first-principles calculations in conjunction with low-energy neutron data represents an important and growing area to address a number of scientific challenges, particularly in the study of disordered systems in materials chemistry, nanomaterials, and catalysis. As opposed to traditional force-field-based approaches, first-principles calculations give access to the electron density and underlying density of states, thereby offering an unprecedented level of chemical insight which, nonetheless, requires detailed experimental scrutiny. In this respect, low-energy neutron spectroscopy could play a pivotal role in the foreseeable future, as access to μeV spectral resolution over atomic and molecular length and time scales remains the preeminent domain of neutron-based techniques.

In the light of the above, our highest priority in the short and medium terms involves the development and subsequent deployment of a software framework for the calculation of neutron observables from time-dependent computer simulations and a systematic comparison with low-energy spectroscopic data. The progressive integration of this toolkit within the open-source *MANTID project* (a process which has already started in close collaboration with *Imperial College*) will facilitate wider dissemination for general use. Further engagement with the user community will be pursued primarily *via* training activities and scientific collaborations. Although emphasis will be placed on first-principles methods, we note that these tools can also facilitate the interpretation of force-field-based *MD* simulations, currently required for the investigation of large systems (>500 atoms per unit cell, our present limit for first-principles *MD*). To implement this workplan, a full-time scientist embedded within the *IRIS-OSIRIS* team and with extensive expertise in first-principles modelling would be required. We emphasize that this staff requirement comes *in addition* to the fourth permanent member of staff presently needed to return to full operational levels on the two instruments (see ***Current status and staffing*** above). In terms of infrastructure, a medium upgrade of the *SCARF-RAL* supercomputing facility with a dedicated 2k-core cluster (£750k) would provide the requisite level of computing power to tackle up to ten systems per year using first-principles methods. In the medium to long terms, new collaborative models will be explored with other institutions beyond *ISIS* such as the *STFC Scientific Computing Deparment* and external academic partners, with a view to further increases in both capacity and capability.

For *IRIS* and *OSIRIS* to move forward in an internationally changing neutron-scattering landscape, instrument upgrades are mandatory. As detailed below, they build upon our experience with instrument concepts like *FIRES* or *ELF* (see ***Annex III***), as well as ongoing developments (see *RAL-TR-2013-008* and recent publications in ***Annex I***). From an instrumentation viewpoint, the primary objective of these developments is to attain a neutronic performance in terms of dynamic range and flux comparable to the most recent near-backscattering instruments at other high-flux spallation sources around the globe (*BASIS* and *DNA*). Scientifically, these developments are aimed at both supporting and strengthening our current user base (see ***Statistics of use*** above), in addition to community expansion. As stressed earlier, computational modelling underpins these instrument initiatives in a fundamental manner.

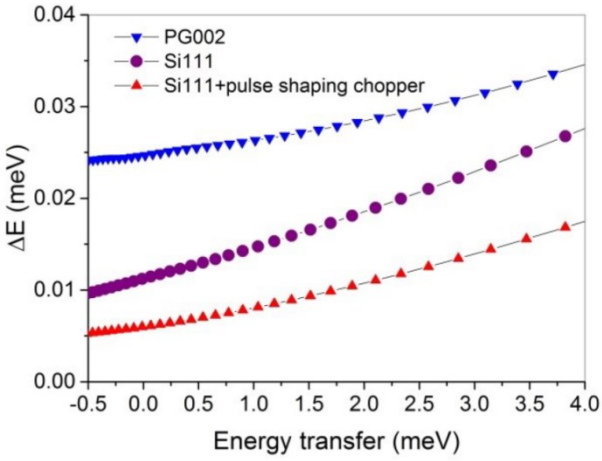

Figure 8. Calculated energy resolution for the present PG002 configuration on OSIRIS, and comparison with Si111 with and without a pulse-shaping chopper.

In the case of *OSIRIS*, its secondary spectrometer has room to house a *Si111* analyser to increase its spectral resolution and dynamic range (£600k). In particular, access to longer relaxation times would, for example, provide new opportunities to strengthen our science programme in ionic and proton conduction in complex media, or molecular transport in porous materials and catalysts of industrial relevance, which quite often suffer from present limits. With a *Si111* analyser, an immediate increase in energy resolution by a factor of 2.5 compared to *PG002* (see ***Fig. 8***) would give access to correlation times of hundreds of picoseconds. Likewise, spectroscopic studies on molecular and magnetic systems (>90% of current demand across both instruments) would certainly profit from an increase in spectral resolution up to 10-20 meV in energy transfer, a feature that also trascends present capabilities on *LET* or *TOSCA*. Combined with the use of position-sensitive detectors, it would also provide four-dimensional $S(\mathbf{Q},\omega)$ data sets with high resolution for single-crystal studies, currently amounting to *ca.* 25% of current demand across both instruments. All in all, this development would equip *ISIS* with a unique cold-neutron spectrometer capable of simultaneous spectroscopic measurements with high (*Si*) and intermediate resolution (*PG*), in addition to synchronous high-resolution diffraction. Further ahead, the upgraded secondary spectrometer could be combined with a refurbished primary spectrometer utilising a fast pulse-shaping

chopper so as to reach an energy resolution of ~5 μeV over the same *Q*-range as *PG002* (see ***Fig. 8***). Taking into account an estimated gain factor for the H_2 moderator of *ca.* 5 following the *ISIS Target Station 1 Upgrade Project*, a pulse-shaping chopper together with an optimised focusing guide (total of £3M) would lead to an order-of-magnitude increase in flux with much-improved resolution. To summarise, the level of investment required to upgrade *OSIRIS* (total of *ca.* £4M) would serve to maintain and expand our user base in the foreseeable future. As such, it represents significantly better value-for-money relative to the cost of a new instrument like *BASIS* at the *SNS* or *DNA* at *JPARC*.

The long-term future and sustainability of *IRIS* calls for a more radical rethink of its remit beyond the current *status quo*, with a view to both strengthening and expanding the user base into new research areas in materials chemistry, soft matter, and biology. Building upon the success of the *Xpress* system, *IRIS* is currently in a position to become the platform of choice for high-throughput *QENS* research *via* the further automatisation and standarisation of experimental and data-analysis protocols. This development would eventually turn *IRIS* into the primary gateway to *QENS* research at *ISIS* by enabling rapid parametric surveys with high resolution and over a wide *Q*-range – a preamble to more detailed experimental campaigns on *OSIRIS* or *LET*, as well as a link to emerging initiatives worldwide such as the *nDDB* database for bio-oriented research. In this manner, *IRIS* would serve as a first port of call for new research groups, including their requisite training in neutron-scattering techniques. From the point of view of outreach to new scientific communities, this high-throughput approach to *QENS* research would provide spatially and temporally resolved maps of the dynamical response of complex materials as a function of temperature or other external stimuli, in some ways reminiscent of (more conventional) calorimetric techniques. Science drivers underpinning this strategy include detailed spectroscopic surveys of compositional phase diagrams of complex hydrides, ionic conductors, or nanoporous materials for gas and energy storage; systematic studies of the properties of soft materials such as polymer nanocomposites or room-temperature ionic liquids across well-defined chemical series (*e.g.*, acidity, hydrogen-bond strength, *etc.*); or the enticing prospects of the screening of pharmaceutical drugs on the basis of their relaxation behaviour. In the short term, these tasks require a number of developments in sample environment (*e.g.,* humidity chambers, pressurised liquids, corrosive environments), as well as the implementation of complementary *in situ* techniques (*e.g.,* calorimetry). Some of these developments such as the design and construction of a sample changer or seamless operation over the temperature range 4-600K using an automated cryofurnace are well underway. An additional £300k would be required over the next three years to support these efforts, with the aim of increasing the number of systems studied on *IRIS* by a factor of *ca.* 10.

The above short-term workplan for *IRIS* also provides a starting point for larger instrument developments in the longer term. This task follows naturally from ongoing neutronics developments on *OSIRIS*. For example, an *IRIS* guide upgrade (which could happen synchronously with *OSIRIS*) in combination with, say, the use of a *Si311* analyser would extend the *Q*-range up to *ca.* 4 $Å^{-1}$ with a higher resolution (~10 μeV) than presently achievable at *ISIS* (~50 μeV with *PG004*) – see ***Annex III***. In the study of complex systems, this feature becomes of paramount importance so as to attain an accurate description of the geometry of motions, as well as in the separation of fast *vs.* slow modes, observables also amenable to calculation using *MD* simulations. Other possibilities include the simultaneous implementation of spectroscopic and small-angle-scattering measurements, to establish dynamics-structure-property relationships via the correlation of *QENS* observables with structural information, particularly at the mesoscale. As such, this development would represent a novel platform to engage with the large-scale-structures community, a collective which has primarily relied on the use of structural neutron techniques to date.

Table 1 below summarises the proposed developments and associated timelines presented in this section. These plans assume the staffing levels requested earlier in the document.

Table 1. Proposed developments for IRIS & OSIRIS and associated timelines.

Development	Year					
	2015	2016	2017	2018	2019	2020
Modelling Phase I	Staff recruitment with modelling expertise	SCARF upgrade & software development	Software and method development	Deployment & training		
Modelling Phase II				Extension to other HPC initiatives	Extension to other HPC initiatives	Deployment & training
OSIRIS Phase I	Feasibility & design of new secondary	Secondary upgrade (Si111)	Secondary upgrade (Si111)	Start of operations with upgraded OSIRIS		
OSIRIS Phase II			Feasibility & design of primary	Primary upgrade: guide + pulse shaping chopper	Primary upgrade: guide + pulse shaping chopper	Start of operations with upgraded OSIRIS
IRIS Phase I	High-throughput QENS: sample environment	High-throughput QENS: sample environment, simultaneous techniques	High-throughput QENS: simultaneous techniques			
IRIS Phase II		Baselining & feasibility in context of TS1 upgrade	Feasibility & design of primary & secondary	Primary upgrade: guide + pulse shaping chopper	Primary upgrade: guide + pulse shaping chopper	Secondary upgrade: e.g., Si311, QENS-SANS

Annex I

IRIS Publications 2008-2014

2014 (last update 15 October 2014)

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Annex I (Continued)

OSIRIS Publications 2008-2014

2014 (last update 15 October 2014)

1. D. Vaknin and F. Demmel. Magnetic spectra in the tri-diminished-icosahedron $\{Fe_9\}$ nanocluster by inelastic neutron scattering. *Physical Review B (Rapid Communications)* **89** 180411 (2014).

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74. D. Barreca *et al.* Stabilization effects of kosmotrope systems on ornithine carbamoyltransferase. *International Journal of Biological Macromolecules* **45** 120 (2009).

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Annex II

Supplementary Data

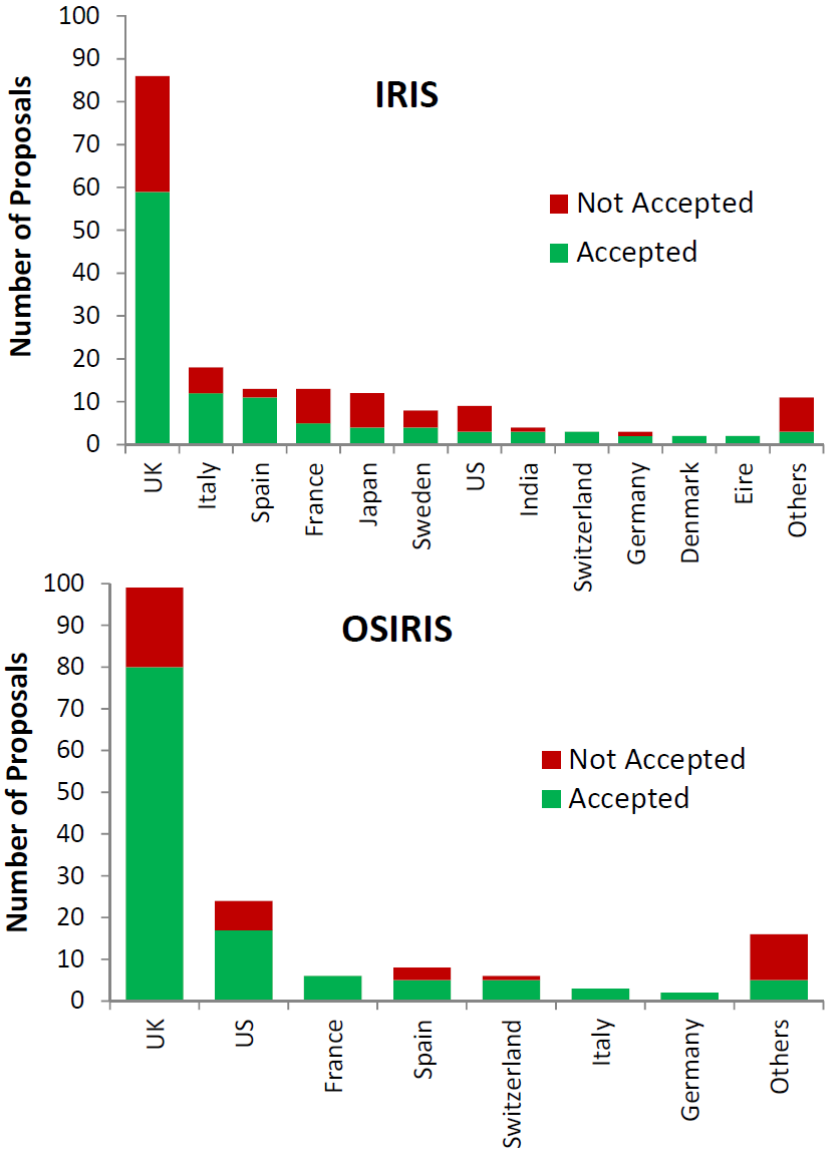

Figure A2.1. Proposals by nationality during 2008-2013.

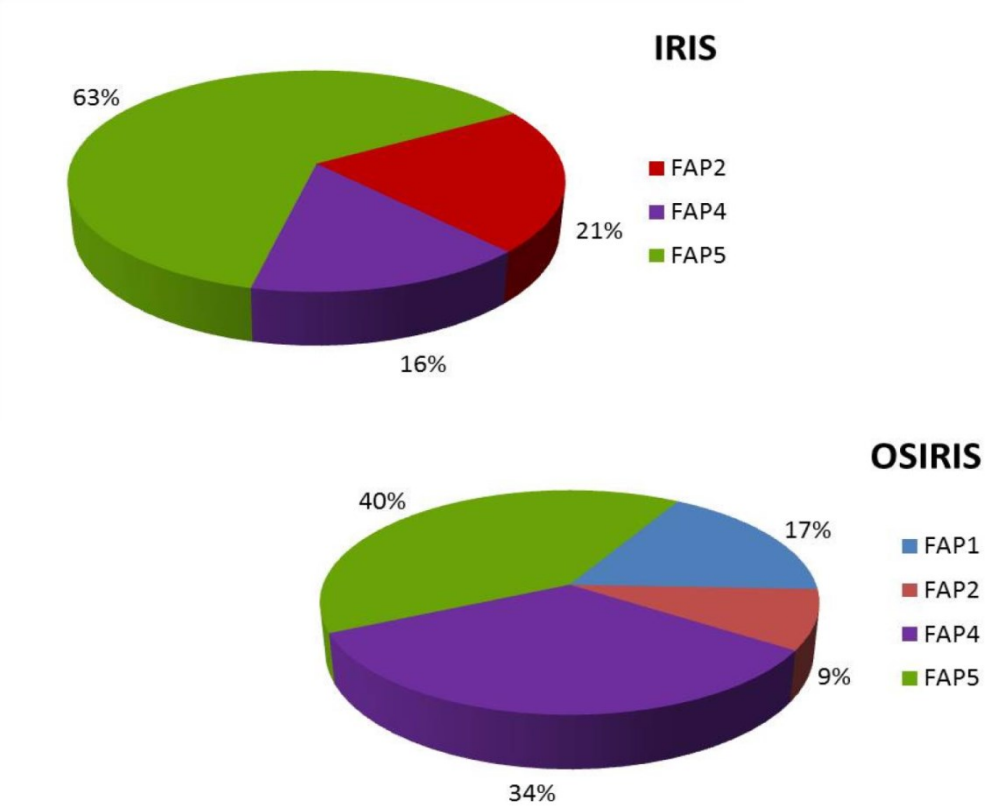

Figure A2.2. Allocation of IRIS & OSIRIS proposals to FAPs during 2008-2013.

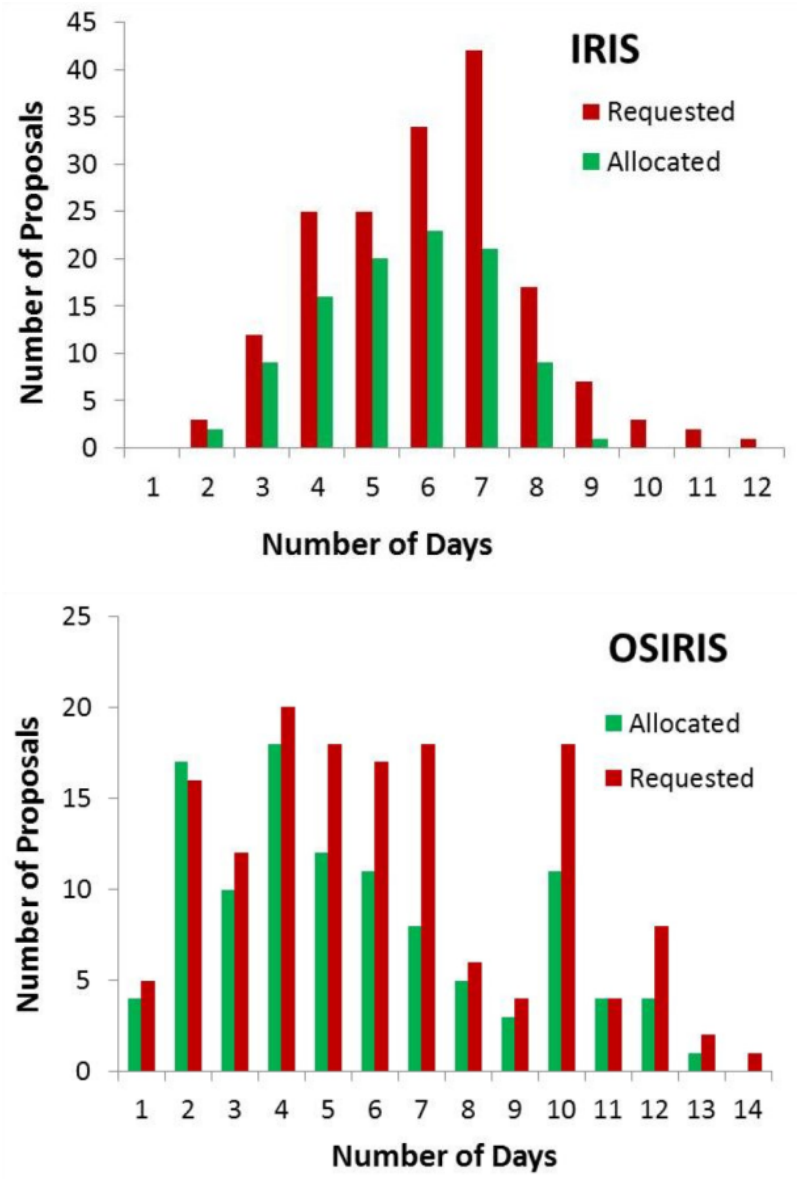

Figure A2.3. Histograms of number of proposals as a function of requested and allocated days.

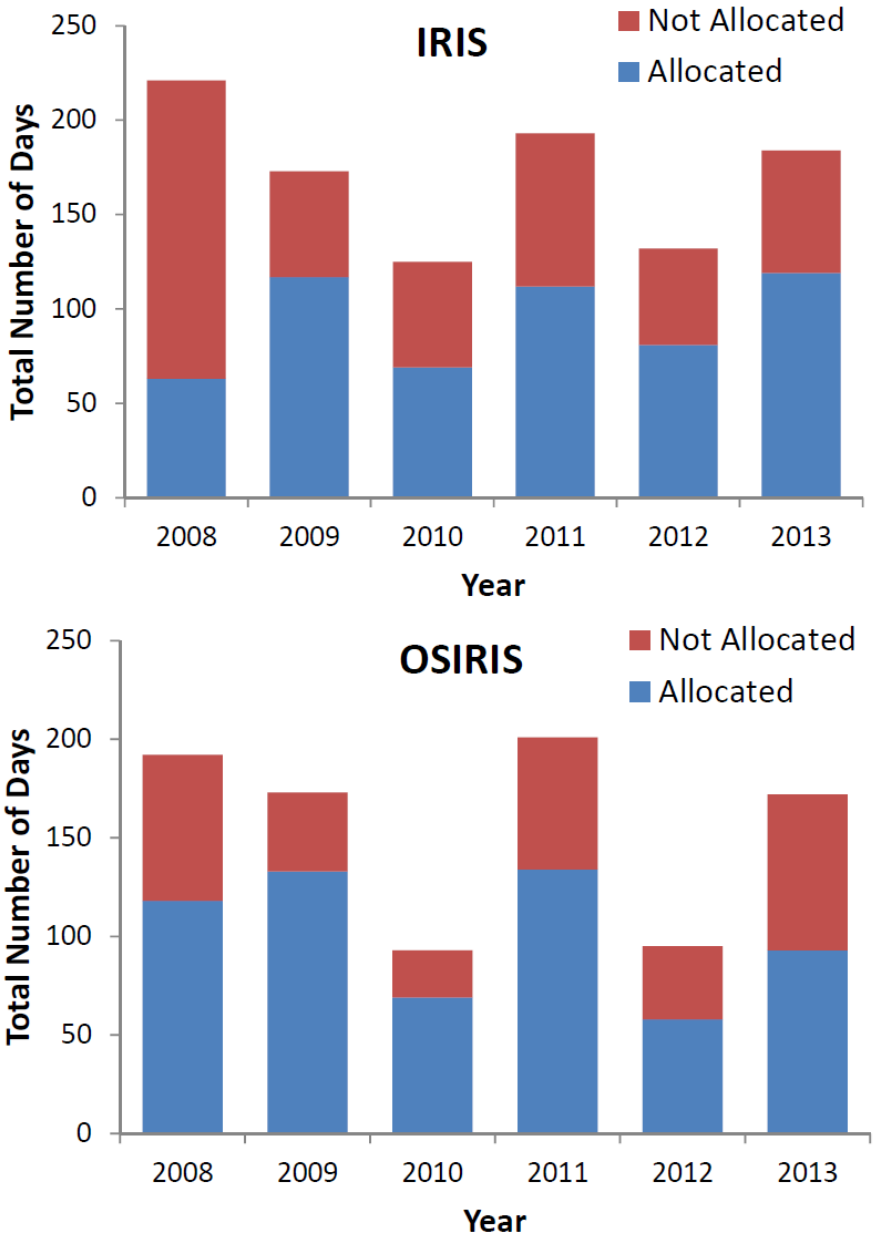

Figure A2.4. Beam time allocation on IRIS & OSIRIS during 2008-2013.

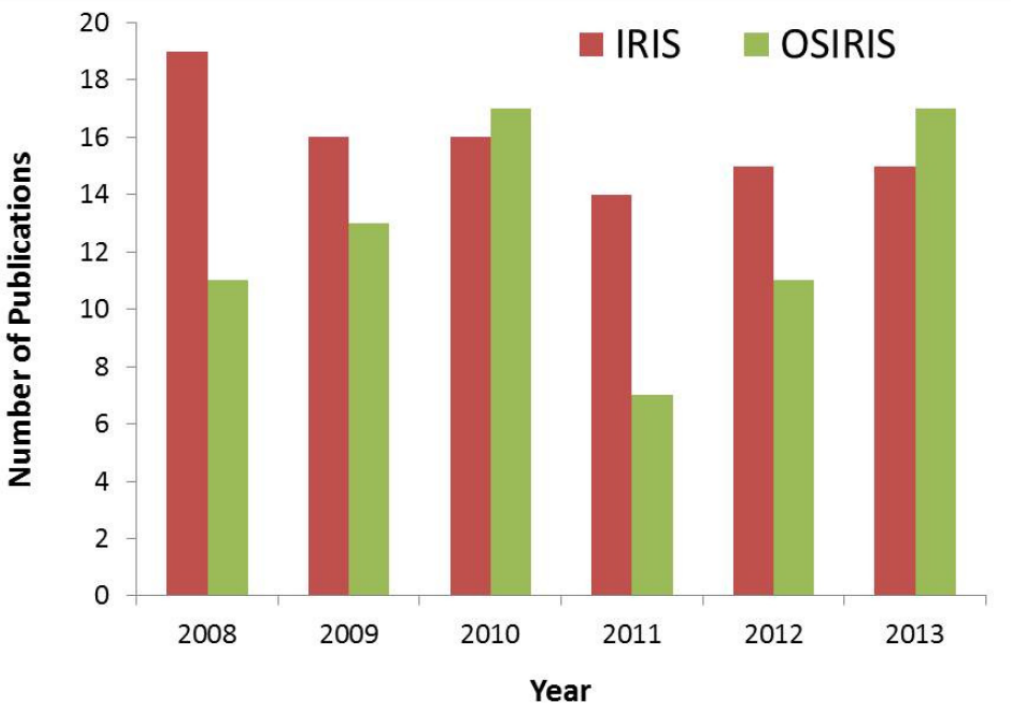

Figure A2.5. Publications from IRIS & OSIRIS during 2008-2013.

Table A2.1. UK PIs of IRIS & OSIRIS (2008-2013).

	User Number	Submitted Proposals
1	462	12
2	6590	9
3	9624	8
4	9524	7
5	15821	6
6	13551	5
7	4481	5
8	10632	5
9	1487	5
10	13373	4
11	1787	4
12	10540	4
13	7463	4
14	6515	4
15	1817	4
16	109	4
17	4381	4
18	148	3
19	5072	3
20	11235	3
21	4167	3
22	871	3
23	1039511	3
24	5519	3
25	5356	3
26	35	2
27	2915	2
28	8513	2
29	1047	2
30	8494	2
31	5287	2
32	11149	2
33	4866	2
34	1030055	2
35	10362	2
36	5925	2
37	1305	2
38	3644	2
39	416	2
40	9779	2

41	15442	2
42	36	2
43	8578	2
44	298	2
45	952	1
46	1043951	1
47	6727	1
48	8049	1
49	16093	1
50	155	1
51	8927	1
52	8547	1
53	6553	1
54	2406	1
55	882	1
56	244	1
57	569	1
58	8266	1
59	3462	1
60	15400	1
61	8242	1
62	14045	1
63	7581	1
64	1019393	1
65	17864	1
66	10157	1
67	14263	1
68	1214	1
69	1043699	1
70	1039672	1
71	1018	1
72	1043954	1
73	15499	1
74	188	1
75	455	1
76	335	1
77	1030057	1
78	9078	1
79	5876	1

Table A2.2. USA (2008-2013).

	Institution	Submitted proposals (*IRIS* & *OSIRIS*)
1	University of Tennessee	7
2	Oak Ridge National Laboratory	5
3	Iowa State University	5
4	MIT	3
5	NIST	2
6	University of Delaware	1
7	University of California − San Diego	1
8	University of Syracuse	1
9	Rutgers University	1
10	Ohio State University	1
11	Los Alamos National Laboratory	1
12	Lawrence Berkeley National Laboratory	1
13	Eastern Washington University	1
14	Clemson University	1
15	Carnegie Institution of Washington	1
16	Brookhaven National Laboratory	1
17	Argonne National Laboratory	1

Table A2.3. Spain (2008-2013).

	Institution	**Submitted proposals (*IRIS & OSIRIS*)**
1	Universidad del País Vasco (UPV-EHU)	6
2	Universidad Politécnica de Cartagena	3
3	Universidad Autónoma de Madrid	3
4	Instituto de Ciencia & Tecnología de Polímeros	3
5	Universidad Complutense de Madrid	2
7	CSIC	2
8	Universitat Politécnica de Catalunya	1
9	Universidad de Almería	1
10	Donostia International Physics Centre	1

Table A2.4. Italy (2008-2013).

	Institution	**Submitted proposals (*IRIS & OSIRIS*)**
1	Università di Roma − Tor Vergata	6
2	Università di Messina	5
3	Università di Milano - Bicocca	3
4	CNR	2
5	Università di Milano	2
6	Università di Parma	2
7	Università di Roma Tre	1

Table A2.5. France (2008-2013).

	Institution	Submitted proposals (*IRIS* & *OSIRIS*)
1	Institut Laue-Langevin	7
2	Université Denis Diderot - Paris 7	4
3	Université de Rennes 1	3
4	CEA-Saclay	2
5	CNR INFM at ILL	1
6	Université de Montpellier II	1
7	Université d'Orléans − CNRS	1
8	Université Joseph-Fourier Grenoble	1

Table A2.6. Japan (2008-2013).

	Institution	Submitted proposals (*IRIS* & *OSIRIS*)
1	Tokyo University	7
2	KEK - High Energy Accelerator Research Organization	2
3	Ochanomizu University	2
4	Kyushu University	2
5	Ochanomizu University	1
6	Toyota Central Research & Development Laboratory	1
7	Keio University	1

Table A2.7. Switzerland (2008-2013).

	Institution	**Submitted proposals (*IRIS* & *OSIRIS*)**
1	ETH Zurich and Paul Scherrer Institut	6
2	EMPA − Materials Science and Technology	2
3	Ecole Polytechnique Federale de Lausanne	1

Table A2.8. Germany (2008-2013).

	Institution	**Submitted proposals (*IRIS* & *OSIRIS*)**
1	Helmholtz Zentrum Berlin für Materialien und Energie	3
2	Universität Freiburg	1
3	Forschungszentrum Jülich	1

Annex III

ELF Preliminary Proposal – July 2013

ELF_Prelim_proposal_Jul13.pdf – Page 1

ELF

… a workhorse spectrometer dedicated to the study of single particle motions via elastic window measurement

Contact: *Victoria Garcia Sakai & Mark Telling*

Aim

An instrument is needed at ISIS dedicated to providing ***high energy resolution and wide Q-range measurements at high flux*** for the isolation, and characterisation, of motions at the nanoscale, with an emphasis on biological systems. Such capability is lacking in the current ISIS instrument suite yet is paramount for linking two scientific communities; soft matter/biology experimentalists and simulators.

The new instrument should provide:

(1) high energy resolution (< 8ueV) AND high momentum transfer coverage (> 2Å^{-1})
(2) high flux for high experimental throughput/statistics, especially for small samples
(3) routine measurement of the elastic incoherent structure factor (EISF) via elastic intensity mapping as a function of temperature

Such capacity will bridge an existing gap in S(Q,ω) at ISIS. While the highest resolution spectrometers at ISIS (IRIS or LET) do afford resolutions of ~ 10 μeV (fwhm) with reasonable flux, they do so at the expense of momentum transfer; the Q-range limited to Q_{max} ~ 1.8Å^{-1}. The only spectrometer in the world currently able to access a wider Q-regime with high energy resolution is the significantly oversubscribed CRG-IN13 instrument at the ILL, which provides 8μeV (fwhm) and 0.2 < Q < 4.9.

Science Drivers

Dynamical processes occurring in the 100 ps to few ns regime are precursors to the larger amplitude modes in macromolecules; such as those in proteins which lead to function. Routine high energy resolution measurement at ISIS, however, limits the user to temporal ranges up to ~ 150 ps. Consequently, there is a clear need for the high energy resolution proposed. Furthermore, to distinguish between different motions in complex macromolecular systems, which typically co-exist, a wide Q-range is essential. This need is exemplified in Fig. 1 where it can be seen that the extended Q-range proposed is paramount if different single particle motions (jump reorientation, rotational and translational diffusions, etc...) are to be clearly distinguished from experimentally determined EISFs. At present, such differentiation can be speculative at best with the Q-range currently accessible at the highest energy resolution; not to mention greatly dependent upon measurement time! ELF aims to resolve this.

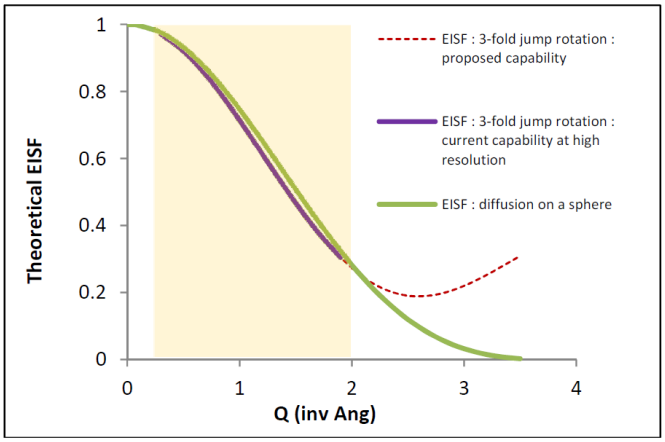

Fig.1. Differentiating between different modes of motion by extending momentum transfer

Measurement on ELF will be optimised predominately for the well-established elastic fixed window scan; data being collected as a function of temperature and/or other external parameter. Of course, complementary Quasi-Elastic measurement will also be supported. Such measurement will enable the dynamical characterisation of complex systems at the nanoscale and over relevant ps-ns timescales. The results will have immediate impact in many scientific areas of interest in the life sciences (biophysics, biomaterials, healthcare and medicine). In addition, with high flux and beam focussing, the instrument proposed will also be able to accommodate reduced (sometimes also costly) sample quantities; an all too frequent pre-requisite for the study of, for example, proteins of medical relevance, deuterated or small neutron-cross section materials, advanced polymers for photovoltaics, catalysts, organometallics and cellular membranes and tissues, amongst others. It will also provide a means for rapid comparative studies.

In the field of biology and medicine, the importance of dynamical and geometric data measured from such a neutron spectrometer cannot be under stated. Such data underpins current understanding at a molecular level, as demonstrated by the number of publications (>300) in refereed journals over the last few decades. Of these publications, one third focusses on the analysis of elastic window measurement and about half also include quasi-elastic spectra. All measurements, however, are performed primarily on neutron backscattering spectrometers of the type proposed. The scope of these publications ranges from the general physics of protein/solvent dynamics to the study of biologically relevant dynamics-function relationships in living cells (a developing area of research). Furthermore, such data is adding to the neutron Dynamics Data Bank (*n*DDB), currently being set-up in collaboration with the ILL. This initiative aims to make neutron dynamical data widely accessible to the community and to complement the structural Protein Data Bank. It should be mentioned that the initiative has been extremely well received by theoretical/computation communities who rely on experimental neutron data to validate and improve force field models.

Of course, the elastic fixed window mode of operation of ELF, will be a useful dynamical characterisation tool at the nanoscale for transitions in any system, not limited to biologicals.

Realisation: technical details

The proposed spectrometer will be an inverted geometry machine in perfect backscattering using, most likely, Si311 analyser crystals Bragg reflecting at λ=3.3Å. We envisage two options:

(1) to use the existing IRIS port and recycle existing beam line infrastructure, or

(2) build a longer spectrometer (ca. 100m)

The former should be a vastly cheaper option. In both cases we plan to redesign the secondary spectrometer vessel; this is of paramount importance if we utilise the exiting IRIS infrastructure since the present tank is over 20 years old and prone to vacuum leaks which hamper efficient operation. To achieve high resolution, we would request a neutron pulse width of 10μs at 3.3Å. While perhaps challenging from a neutronics /moderator perspective, such a pulse width would be possible using a chopper arrangement. Alternatively, for an elongated instrument (2), the pulse width could be relaxed to 40 μs. Considering the proposed analyser type we anticipate energy resolutions of ~ 5ueV (fwhm, IRIS infrastructure, Δt_{mod}= 10 μs) or ~ 7ueV (fwhm, 100m, Δt_{mod} = 40 μs). Based on our experience developing the IRIS and OSIRIS instruments we believe we can expect the following flux gain factors over those presently afforded on IRIS at 3.3 Å:

a) A long overdue guide upgrade will increase the flux at 3.3Å by a minimum of x 8 (upgrade to m=2 minimum, gain based on comparison of IRIS/OSIRIS guides @ 3.3A)
b) increasing the analyser area will increase the detected flux by a factor of at least x 2
c) considering estimates of potential moderator flux gains compared to the original H_2 design we believe peak flux improvements to be in the order of x 10

Of course, should a chopper arrangement be needed to construct the necessary pulse profile then we envisage a factor of 6 loss in flux at 3.3Å. Additionally, Si(311) is a much poorer analyser material than graphite in terms of reflectivity to the order of x8.

Nonetheless, we envisage a minimum gain in detected flux at the elastic line of x 3. NB: if Δt_{mod}= 10 μs is achievable from the moderator redesign then the gain factor would exceed x 30!

It is worth mentioning that for elastic intensity measurement vs. temperature, the rate of data collection will be dominated by the ramp rate programmed by the user rather than data collection time. However, based on our conservative gain factor, vastly superior statistics per temperature point will be collected. This will circumvent the need to group detectors when reducing the data and ensure access to the full extended Q-range afforded by the instrument. Of course, finer temperature steps could be programmed which would allow more detailed scans to be collected than presently achieved.

In terms of future planning, we would ideally like to position the instrument on a moderator that also yields significant flux at ~ 6.6Å. By doing so we could exploit other analyser

materials; for example Si(111) crystals would yield a resolution of ~1ueV (fwhm) and 0.2 < Q < 1.8. This would be a vast improvement on the current IRIS mica set-up which affords similar resolution but has a severely limited Q-range and minimal flux; the analyser crystals Bragg reflecting λ ~ 20Å. Redesign of the secondary spectrometer vessel would also allow the option to install position sensitive detectors for those samples with preferential orientations (such as in-plane and out-of-plane features in cell membranes).

Finally, dedicated sample environment capabilities will be developed in-line with the instrument design. As well as the standard CCR, new equipment will include humidity chambers and high pressure, electric field and extreme temperature capabilities for, for example, the study of extromophile proteins.

Community

We believe this instrument will be of extreme interest to the soft condensed matter and biological community. As stated, results will benefit both experimental and computation fraternities and help facilitate future collaboration - a necessary endeavour. At these early stages of the proposal no research teams have been contacted. However, the meeting between neutron scientists and simulators that took place to kick-off the nDDB initiative drew over 70 interested international and UK participants. Of course, we also envisage support from our current user base, especially those who have supported past endeavours on IRIS and OSIRIS such as the beryllium filter upgrade project. Finally, the construction of this instrument will present new opportunity to target, via focussed outreach, UK communities for whom neutrons present a new or alternative method of measurement. We believe that they will embrace the simple and rapid dynamical characterisation that ELF will provide; one immediate sign of such endorsement being the successful QENS Xpress programme.