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Neutron Compton Scattering

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ABSTRACT

The development of suitable instrumentation at spallation neutron sources has opened the way for easier and better use of neutron Compton scattering to measure the ground state momentum distribution of nuclei in condensed matter. In the past, its main use has been in work on the He^4 aimed at measuring the condensate fraction. Recently, very good quality data has been obtained for several other systems, including H_2, and compared successfully with model calculations. This article aims to serve as an introduction to the potential use of neutron Compton scattering in biology, chemistry and physics.

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1. Introduction

Neutron Compton scattering is a direct probe of the momentum distribution of nuclei in condensed matter. For this purpose, it is an unrivalled experimental technique. Although the potential of neutron Compton scattering was recognized almost three decades ago, instrumentation suitable for accurate, routine investigations has been available for no more than a few years[2]. This circumstance reflects the need of a good beam of energetic neutrons in order to achieve the Compton limit of scattering; the energy scale is set by the characteristic vibrational energy of the scattering centre, which for the case of a proton in H_2 is 0.54 eV. Fortunately, spallation neutron sources now produce good 10 - 50 eV beams.

Let $\psi(\mathbf{R})$ be the real-space, ground state wave function of the scattering centre. The corresponding momentum density is,

$$\rho(\mathbf{q}) = (1/2\pi)^3 \left| \int d\mathbf{R}\, \psi(\mathbf{R}) \exp(-i\mathbf{q}.\mathbf{R}) \right|^2 . \qquad (1.1)$$

The Compton limit of the neutron scattering cross-section, often referred to as the Compton profile, is a particular integral of $\rho(\mathbf{q})$. The definition of the profile is provided in §3. For the moment, we address the data obtained by Mayers[3] for H_2, which is displayed in fig. (1). Included in fig. (1) are results from two plausible models. Because of the good quality of the experimental data one can judge that one model is superior to the other. In fact, Mayers argues that, the data show features due to the first observation of interference between the proton and neutron wave functions.

The original motivation to exploit neutron Compton scattering was the possible determination of the condensate fraction in He^4. Thirty years on, this work continues, on both pure He^4 and He^3 - He^4 mixtures[4]. Other recent experimental work includes a study of solid argon[5]. In all cases, the good quality of the data allows a meaningful confrontation with realistic model calculations.

Prior to giving more details about neutron Compton scattering, we pause to comment on related activities. Of course, the Compton effect was discovered through studies of photon scattering by electrons. Stuewer and Cooper[6] have prepared a history of the discovery, and its development as an experimental tool for the investigation of electron momentum densities. Work in this area is reviewed by Cooper[7]. In the past few years, magnetic Compton scattering has been shown to be a viable experimental probe of materials with a net magnetization[8,13]. Similar work should be possible with neutron beams[9], but it has not yet been demonstrated largely because the signals are very weak. Finally, we mention that the structure of nucleons, and the existence of quarks, has been revealed by performing with high-energy (~ 1 - 100 GeV) electrons or neutrinos the analogue of Compton scattering[10]. In the field of particle physics, Compton scattering from nucleons is usually referred to as deep inelastic scattering. The latter term is occasionally used in the context of neutron beam studies of condensed matter but, we choose to use the term neutron Compton

scattering. The reader might wish also to make contact with extensive experimental and theoretical work on nucleon momentum distributions in nuclei[11].

Concerning the rest of this article, the focus is on the basic nature of neutron Compton scattering from condensed matter, rather than a review of experimental and theoretical studies of particular materials. For one thing, Glyde[12] has reviewed the extensive work on liquid and solid helium, which is the only mature area of activity, while the few recent studies of other materials are found in the cited papers[2-5].

By way of an orientation to the more detailed description that follows, we now add a few remarks about the nature of the Compton limit of scattering. The direct relation that exists in this limit between the cross-section and the momentum density is achieved for sufficiently large incident energies and scattering vectors. A high incident energy and a modest energy transfer are required to make the scattering event almost instantaneous which, as we will demonstrate, is one key approximation. For this extreme condition, in the brief duration of the scattering event the scattering centre does not change its position very much, so its potential energy is also almost unchanged (assuming that the potential energy is a function only of position variables which means, for one thing, negligible relativistic corrections). Thus, in the energy conservation condition for the scattering event the potential energies before and after scattering cancel one another, to a good approximation, and the condition is the same as for free particles. However, the potential that binds the centre in the sample is manifest in the scattered signal in the guise of the governing transition matrix element which, in the Compton limit of scattering, is related to the momentum distribution for the ground state potential energy surface.

The other facet of the Compton limit of scattering concerns the spatial scale of the scattering event. It is required that the scale matches the dimension of a scattering centre. In this limit, the signal contains no information on spatial correlations between the centres, i.e. the scattering vector is so large that the self and coherent scattering response functions are the same. Quite often, this is described as the condition for which the incoherent approximation to scattering is valid. For the special case of protons this requirement is not so obvious in the interpretation of data because in the scattered beam the coherent signal is weak compared to the incoherent signal.

2. Neutron Scattering Cross-Section

The basic quantity extracted from neutron beam scattering experiments is the partial differential cross-section which gives the fraction of neutrons of incident energy E scattered into an element of solid angle $d\Omega$ with an energy between E' and $E' + dE'$. A standard notation for the cross-section is,

$$(d^2\sigma / d\Omega dE'),$$

where the total cross-section, σ, has the dimension of area. Since neutron scattering is a weak process, the first Born approximation for scattering, equivalent to Fermi's Golden Rule, is adequate for the calculation of the cross-section. In this

approximation the incident and final neutron wave functions are simply plane waves with wave vectors $\mathbf{k}$ and $\mathbf{k}'$, respectively.

Let the scattering centres in the target sample be located at positions $\{\mathbf{R}_a\}$ where the integer index a takes all positive values up to a maximum N. The interaction between this array of centres and an incident particle at the position $\mathbf{r}$ is the sum over centres of individual interaction operators,

$$\sum_a V_a(\mathbf{r}-\mathbf{R}_a).$$

Because we utilize the Born approximation, the scattering amplitude contains the matrix element of the interaction formed with plane wave states, namely,

$$\int d\mathbf{r} \exp(-i\mathbf{k}'\cdot\mathbf{r}) \sum_a V_a(\mathbf{r}-\mathbf{R}_a) \exp(i\mathbf{k}\cdot\mathbf{r}) = \sum_a U_a(\mathbf{Q}) \exp(i\mathbf{Q}\cdot\mathbf{R}_a),$$

where the scattering vector $\mathbf{Q} = \mathbf{k} - \mathbf{k}'$ and,

$$U_a(\mathbf{Q}) = \int d\mathbf{r} \exp(i\mathbf{Q}\cdot\mathbf{r}) V_a(\mathbf{r}). \tag{2.1}$$

Here we have assumed that $V_a(\mathbf{r})$ is not a quantum mechanical operator that operates on the plane wave states. In consequence, the scattering amplitude is proportional to the spatial Fourier transform of the interaction.

In subsequent developments $U_a(\mathbf{Q},t)$ and $\mathbf{R}_a(t)$ denote time-dependent operators formed from the standard Heisenberg representation. Also on the subject of notation, $\langle ... \rangle$ denotes the thermal average of the enclosed quantity taken with respect to the states of the scattering centres.

The partial differential cross-section is,

$$\left(\frac{d^2\sigma}{d\Omega dE'}\right) = \frac{k'}{k}\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} dt \exp(-i\omega t) \tag{2.2}$$

$$* \sum_{a,b} \left\langle \exp(-i\mathbf{Q}\cdot\mathbf{R}_a) U_a^+(\mathbf{Q}) U_b(\mathbf{Q},t) \exp\{i\mathbf{Q}\cdot\mathbf{R}_b(t)\} \right\rangle.$$

Not displayed in this formula are the required averages with respect to the isotope distribution, orientation of the nuclear spins, defects and neutron polarization states[14].

In many applications, (2.2) can be simplified because there is negligible correlation between the interaction potentials $\{U_a(\mathbf{Q})\}$ and the position variables $\{\mathbf{R}_a\}$. In general, however, there is such a correlation induced by quantum mechanical forces but, it is negligible except for extreme sample environment conditions, such as those created in the study of quantum fluids and solids. We proceed on the assumption that,

the interaction potentials are uncorrelated with the scattering centre variables, so they are legitimately factored out of the thermal average in (2.2).

For the particular case of scattering by nuclei, $U_a(\mathbf{Q}) = b_a$ where b_a is the scattering amplitude operator, i.e. the Fourier transform of the interaction potential is independent of the scattering vector, $\mathbf{Q}$. Referring to (2.2), we are faced with the task of evaluating the average of $b_a^* b_b$ with respect to the orientation of nuclear spins and the distribution of isotopes. Let us denote this average by a horizontal overbar. Clearly,

$$\overline{b_a^* b_b} = \overline{b_a^*}\ \overline{b_b} + \delta_{a,b}\left\{\overline{|b_a|^2} - \left|\overline{b_a}\right|^2\right\}, \tag{2.3}$$

and for a monatomic sample,

$$\overline{b_a^* b_b} = (1/4\pi)\{\sigma_c + \delta_{a,b}\sigma_i\}, \tag{2.4}$$

where σ_c and σ_i are the coherent and incoherent bound, single-atom cross-sections. For a hydrogenous sample the situation is even simpler, because the incoherent cross-section for a proton, σ_i = 79.8 barns, is large compared to cross-sections for all other isotopes. Hence, for a hydrogenous sample we can, to a good approximation, use,

$$\overline{b_a^* b_b} = (\sigma_i / 4\pi)\,\delta_{a,b},$$

if a and b refer to a proton, and completely neglect all other scattering centres.

The corresponding approximation to the cross-section for a hydrogenous sample is,

$$\frac{d^2\sigma}{d\Omega dE'} = \frac{k'}{k}\frac{\sigma_i}{4\pi} S_i(\mathbf{Q},\omega), \tag{2.4}$$

where the van Hove response function,

$$S_i(\mathbf{Q},\omega) = \frac{1}{2\pi\hbar}\int_{-\infty}^{\infty} dt\, \exp(-i\omega t) \sum_a Y_a(\mathbf{Q},t), \tag{2.5}$$

and the correlation function,

$$Y_a(\mathbf{Q},t) = \left\langle \exp(-i\mathbf{Q}\cdot\mathbf{R}_a)\, \exp\left\{i\mathbf{Q}\cdot\mathbf{R}_a(t)\right\}\right\rangle. \tag{2.6}$$

In (2.5) we have allowed for the possibility that there is more than one type of proton scattering centre.

3. Compton Limit of Scattering

The Compton limit of scattering is often referred to as the impulse approximation, because one uses a short-time expansion for the position variable in the correlation function $Y(\mathbf{Q},t)$, defined in (2.6). A large incident neutron energy, E, and relatively small energy transfer, $\hbar\omega$, provide the license for use of the short-time approximation $\mathbf{R}(t) = (\mathbf{R}(0) + t\mathbf{p}/M)$, where $\mathbf{p}$ is the momentum conjugate to $\mathbf{R} \equiv \mathbf{R}(0)$ and M is the mass of the scatterer.

After inserting the short-time approximation in $Y(\mathbf{Q},t)$ further progress can be made by using the operator identity,

$$e^A e^B = \exp\left\{A + B + \tfrac{1}{2}[A,B]\right\} ,$$

in which the commutator $[A,B]$ is not an operator. For the case in hand, $A = -i\mathbf{Q}\cdot\mathbf{R}$,

$$B = i\mathbf{Q}\cdot\mathbf{R} + (it/M)\mathbf{Q}\cdot\mathbf{p} ,$$

and the commutator of $\mathbf{R}$ and $\mathbf{p}$ is,

$$\left[\mathbf{R}_\alpha, \mathbf{p}_\beta\right] = i\hbar\delta_{\alpha,\beta} ,$$

where α, β label Cartesian components. Then one has obtained the Compton limit of the correlation function, namely,

$$Y(\mathbf{Q},t) \sim \Phi(\mathbf{Q},t) = \exp(itE_r/\hbar)\left\langle \exp(it\mathbf{Q}\cdot\mathbf{p}/M)\right\rangle , \tag{3.1}$$

in which the recoil energy of the scatterer $E_r = (\hbar Q)^2/2M$.

There are several useful representations of the correlation function on the right-hand side of (3.1). First, we consider the representation quoted in the introduction in which the correlation function is expressed in terms of the momentum density $|\chi(\mathbf{q})|^2$, where $\chi(\mathbf{q})$ is the momentum representation of the real-space wave function of the scatterer, $\psi(\mathbf{R})$. For a ground state wave function, $\psi(\mathbf{R})$, the correlation function is,

$$\int d\mathbf{R}\, \psi^*(\mathbf{R}) \exp(it\mathbf{Q}\cdot\mathbf{p}/M)\psi(\mathbf{R}) = (1/2\pi)^3 \int d\mathbf{R} \int d\mathbf{q} \int d\mathbf{q}'\, \chi^*(\mathbf{q})\, \chi(\mathbf{q}') \exp(-i\mathbf{q}\cdot\mathbf{R})$$

$$\exp(it\mathbf{Q}\cdot\mathbf{p}/M) \exp(i\mathbf{q}'\cdot\mathbf{R}) = \int d\mathbf{q}\, |\chi(\mathbf{q})|^2 \exp(i\hbar t\mathbf{q}\cdot\mathbf{Q}/M). \tag{3.2a}$$

In reaching the last equality we have used the operator identity,

$$\exp(i\mathbf{n}\cdot\mathbf{p}/\hbar)\, f(\mathbf{R}) = f(\mathbf{R}+\mathbf{n}),$$

where $f(\mathbf{R})$ is some function of the position variable. The identity shows that the exponential operator translates **R** by a distance **n**. From (3.2) it follows that, in the Compton limit the response function is,

$$S_i(\mathbf{Q},\omega) \sim S_o(\mathbf{Q},\omega) = \int d\mathbf{q}\, |\chi(\mathbf{q})|^2\, \delta(\hbar\omega - E_r - \hbar^2\mathbf{q}\cdot\mathbf{Q}/M). \tag{3.3}$$

As often as not, this response function is expressed in terms of a wave vector,

$$y = M(\hbar\omega - E_r)/\hbar^2 Q. \tag{3.4}$$

(The choice of notation for this variable conforms with convention.) To this end, let the ζ - component of **q**, say, be parallel with the scattering vector, **Q**. The action of the delta function equates q_ζ to y, defined in (3.4). Hence, one arrives at the expression,

$$S_o(\mathbf{Q},\omega) = (M/\hbar^2 Q)J(y), \tag{3.5}$$

where the so-called Compton profile,

$$J(y) = \int dq_\xi \int dq_\eta\, |\chi(q_\xi, q_\eta, y)|^2. \tag{3.6}$$

For an isotropic momentum density, the double integral in (3.6) can be reduced to a single integral.

A second representation of the correlation function in $\Phi(\mathbf{Q},t)$ is obtained by use of the relation,

$$\hbar^3 \int d\mathbf{q}\, \delta(\mathbf{p} - \hbar\mathbf{q}) = 1,$$

where, as always, **p** is the momentum operator, while the integration variable, **q**, has the dimension of a wave vector. Applied to the correlation function defined in (3.1), the relation yields the representation,

$$\langle \exp(it\,\mathbf{Q}\cdot\mathbf{p}/M)\rangle = \int d\mathbf{q}\, \rho(\mathbf{q}) \exp(i\hbar t\mathbf{q}\cdot\mathbf{Q}/M), \tag{3.2b}$$

where the momentum density,

$$\rho(\mathbf{q}) = \langle \delta(\mathbf{q} - \mathbf{p}/\hbar)\rangle, \tag{3.7}$$

has the dimension of volume. The result (3.2b) can be viewed as the generalization of (3.2a) to finite temperatures. Of course, for the ground state $\rho(\mathbf{q}) = |\chi(\mathbf{q})|^2$. The more general expression for the Compton profile is,

$$J(y) = \int dq_\xi \int dq_\eta \, \rho(q_\xi, q_\eta, y), \tag{3.8}$$

where the ζ-component of the Cartesian axes ξ, η, ζ coincides with the scattering vector $\mathbf{Q}$, and the wave vector y is defined in (3.4).

4. Properties of the Compton Response Function

From the definition (3.3),

$$\hbar \int_{-\infty}^{\infty} d\omega \, S_o(\mathbf{Q}, \omega) = \int d\mathbf{q} \rho(\mathbf{q}) = 1. \tag{4.1}$$

A second sum-rule of interest is,

$$\hbar^3 \int_{-\infty}^{\infty} d\omega \, \omega^2 S_o(\mathbf{Q}, \omega) = E_r(E_r + \tfrac{4}{3} < T >), \tag{4.2}$$

where $<T>$ is the average kinetic energy. The particularly simple form of the left-hand side of (4.2) applies for an isotropic momentum density.

An isotropic harmonic oscillator potential is a model of genuine interest for which the response function has a simple closed form. The key result is[14],

$$\langle \exp(i\mathbf{A} \cdot \mathbf{p}) \rangle = \exp\left\{ -\tfrac{1}{2} A^2 \langle p_\xi^2 \rangle \right\}, \tag{4.3}$$

to which can be added,

$$\langle T \rangle = \left(3 \langle p_\xi^2 \rangle / 2M \right). \tag{4.4}$$

To evaluate $\Phi(\mathbf{Q},t)$ with (4.3) one takes,

$$\mathbf{A} = (t\mathbf{Q} / M).$$

The result is,

$$\Phi(\mathbf{Q}, t) = \exp\left\{ (itE_r / \hbar)(1 + \tfrac{2it}{3\hbar} < T >) \right\}, \tag{4.5}$$

and the corresponding response function is,

$$S_o(Q,\omega) = (M/\hbar^2 Q)J(y), \tag{4.6}$$

with,

$$J(y) = (l/\sqrt{\pi})\exp\left\{-(ly)^2\right\}, \tag{4.7}$$

and the length l satisfies,

$$l^2 = (3\hbar^2/4M<T>).$$

Note that the integral of $J(y)$ over the interval $-\infty \le y \le \infty$ is equal to one, in accord with the definition (3.6). The result (4.6) is illustrated in fig. (2).

The second example illustrated in fig. (2) is a perfect, degenerate Fermi fluid. The latter is defined by a momentum density,

$$\rho(\mathbf{q}) = 1\,;\, q < p_f$$

and,

$$\rho(\mathbf{q}) = 0\,;\, q > p_f,$$

where the Fermi wave vector p_f is related to the particle number density n_o through $p_f^3 = 3\pi^2 n_o$. The corresponding Compton response function is readily shown to be,

$$S_o(Q,\omega) = (3M/\hbar^2 4p_f Q)(1-(y/p_f)^2), \tag{4.8}$$

for $|y| < p_f$, and zero elsewhere. Here, and in (4.6), the wave vector y is defined by (3.4). However, some care must be exercised in the application of these results because of the influence on the response function of quantum mechanical exchange forces, and spin-dependent potentials. The latter are not present in a perfect quantum fluid, and hence the corresponding spin density and particle density response functions are the same, apart from a constant.

The particle density response function for a perfect quantum fluid is,

$$S(\mathbf{Q},\omega) = (g/N)\sum_{\mathbf{q}} \delta\left\{\hbar\omega + \varepsilon_{\mathbf{q}} - \varepsilon_{\mathbf{Q}+\mathbf{q}}\right\} f_{\mathbf{q}}(1 - f_{\mathbf{q}+\mathbf{Q}}), \tag{4.9}$$

where g is the spin degeneracy factor, and $\varepsilon_{\mathbf{q}}$ is the energy of a particle with wave vector $\mathbf{q}$. The result (4.9) applies to both Bose and Fermi fluids but, of course, one must employ the appropriate distribution function, $f_{\mathbf{q}}$;

$$f_{\mathbf{q}} = \left\{\exp\left\{(\varepsilon_{\mathbf{q}} - \mu)/k_B T\right\} \pm 1\right\}^{-1},$$

where the lower (upper) sign applies for Bose (Fermi) particles, and the chemical potential, μ, is determined by,

$$N = g\sum_{\mathbf{q}} f_{\mathbf{q}} \quad .$$

It is interesting to observe the behaviour of (4.9) with increasing values of Q. If $\varepsilon_{\mathbf{q}}$ increases with increasing $\mathbf{q}$ then, for a sufficiently large Q and a given $\mathbf{q}$,

$$f_{\mathbf{q}+\mathbf{Q}} << 1 .$$

In this case, the response function (4.9) approaches the Compton limit (3.3).

5. Criteria for the Compton Limit

The Compton limit of scattering emerges when the scattering event is almost instantaneous, for then a short-time expansion of the position variable is valid. To assess the constraints on the scattering experiment required to realize this extreme limit one can appeal to a systematic expansion in time of the exponential operator in the correlation function, $Y_a(\mathbf{Q},t)$, defined in (2.6). Given such an expansion in t one can enquire as to when the linear term dominates the quadratic term. When this situation prevails there is license to neglect all but the linear term in the expansion. On estimating the duration of the scattering event by $t = \hbar/E$, where E is the incident neutron energy, the linear term dominates the quadratic term in t when[19],

$$QE >> 2|\mathbf{F}| , \qquad (5.1)$$

where $\mathbf{F}$ is the force on the scattering centre, i.e. if Φ is the potential energy,

$$\mathbf{F} = -\nabla\Phi \quad .$$

Note that (5.1) is, for a given Φ, a condition on the product of the incident energy and the magnitude of the scattering vector.

Let us rewrite (5.1) in a form appropriate for harmonic or nearly harmonic potentials. If the characteristic vibrational energy is ε_o, expressed in units of eV, and a_o is the Bohr radius, the condition (5.1) reads,

$$a_o QE >> (M/m)\varepsilon_o^2 \times 10^2 \quad , \qquad (5.2)$$

with E in eV, and m the mass of a neutron.

There is a large body of published work on the subject of corrections to the Compton limit of scattering[12, 15-18]. In this work it is customary to write the correlation function as,

$$Y(\mathbf{Q},t) = \Phi(\mathbf{Q},t)R(\mathbf{Q},t)\,. \qquad (5.3)$$

The function $R(\mathbf{Q},t)$ accounts for the shortcomings in $\Phi(\mathbf{Q},t)$ to describe the scattering event. By definition, in the Compton limit $R(\mathbf{Q},t) = 1$ for all permitted values of the arguments.

6. Concluding Remarks

To a large extent, the goal of this article is to raise the awareness of researchers in chemistry, biology, and physics to the potential value of neutron Compton scattering as a probe of the ground state energy surface of nuclei in condensed matter. Because of the exceptionally large cross-section for neutron scattering by protons, the technique is ideally suited for the study of hydrogenous materials. However, to date this field of work is much in its infancy when compared to almost three decades of effort on He^4. Hopefully, recent successful work on argon and H_2, for example, will encourage researchers to use neutron Compton scattering to investigate a broader range of materials in conjunction with various (extreme) sample environments.

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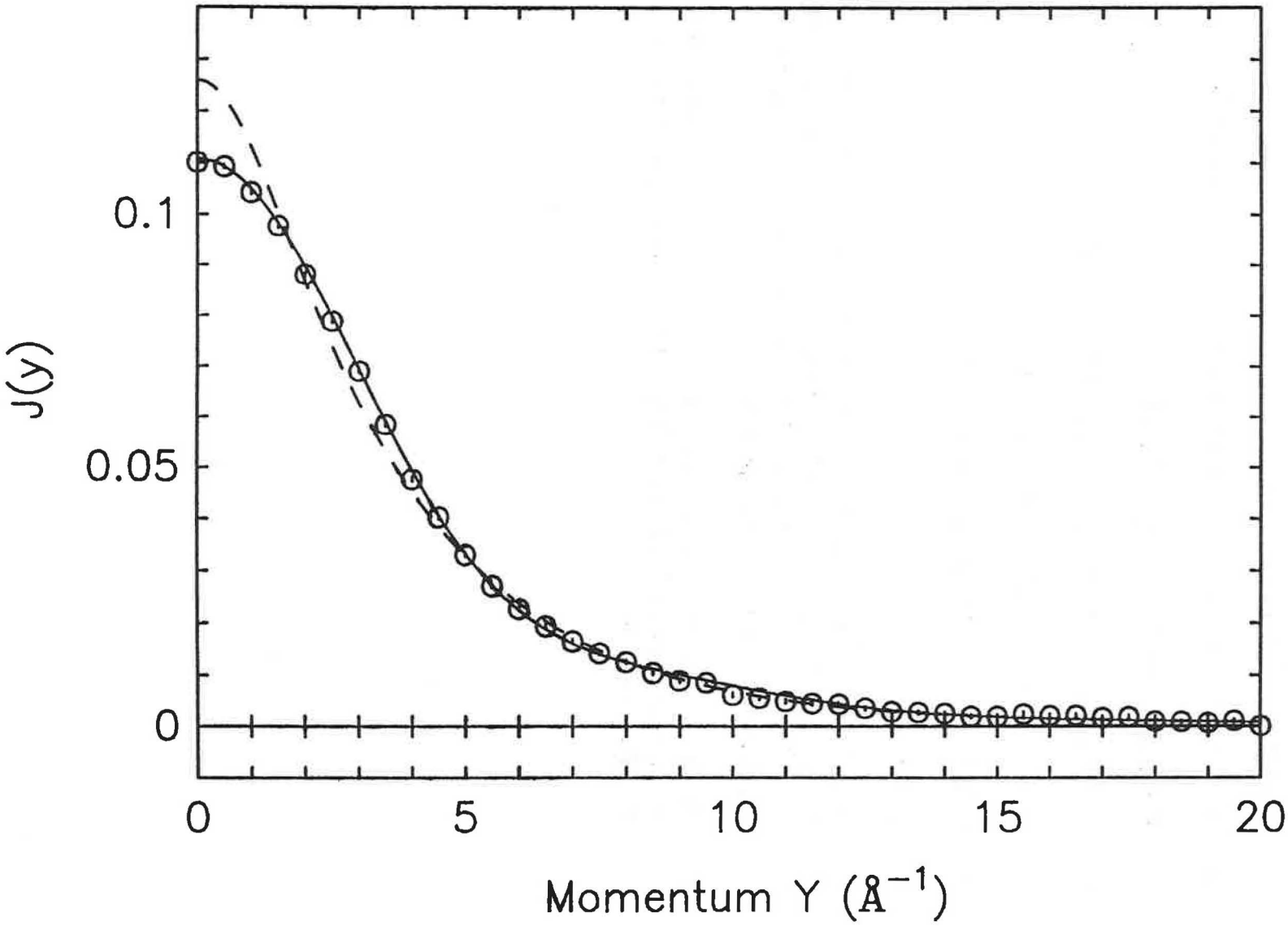
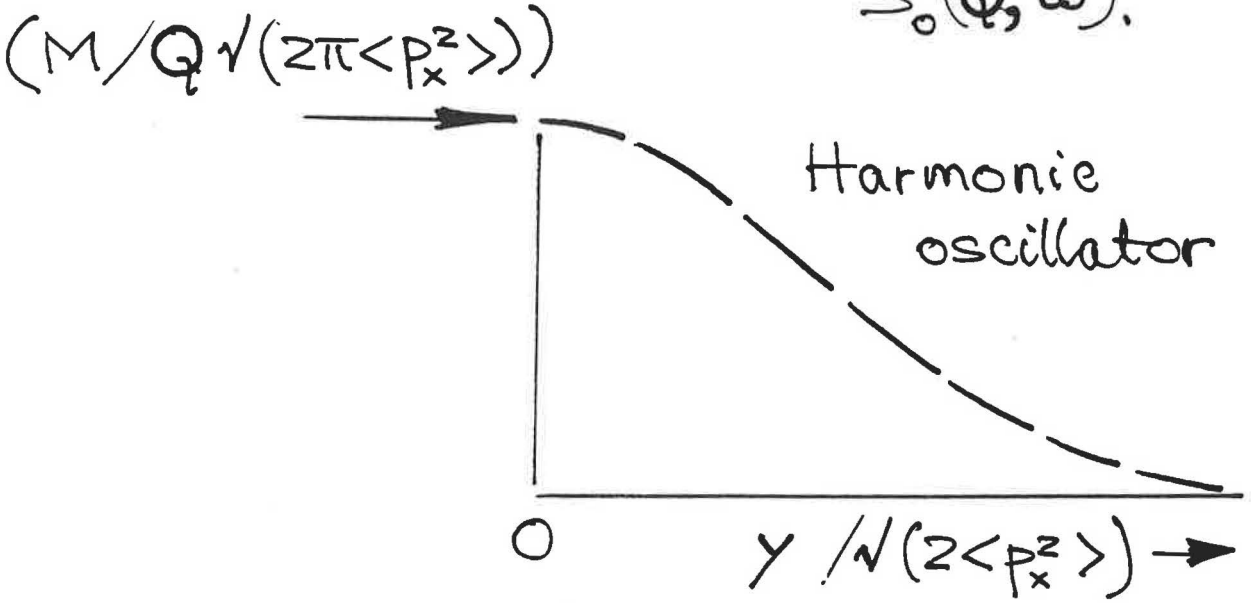
Figure Caption

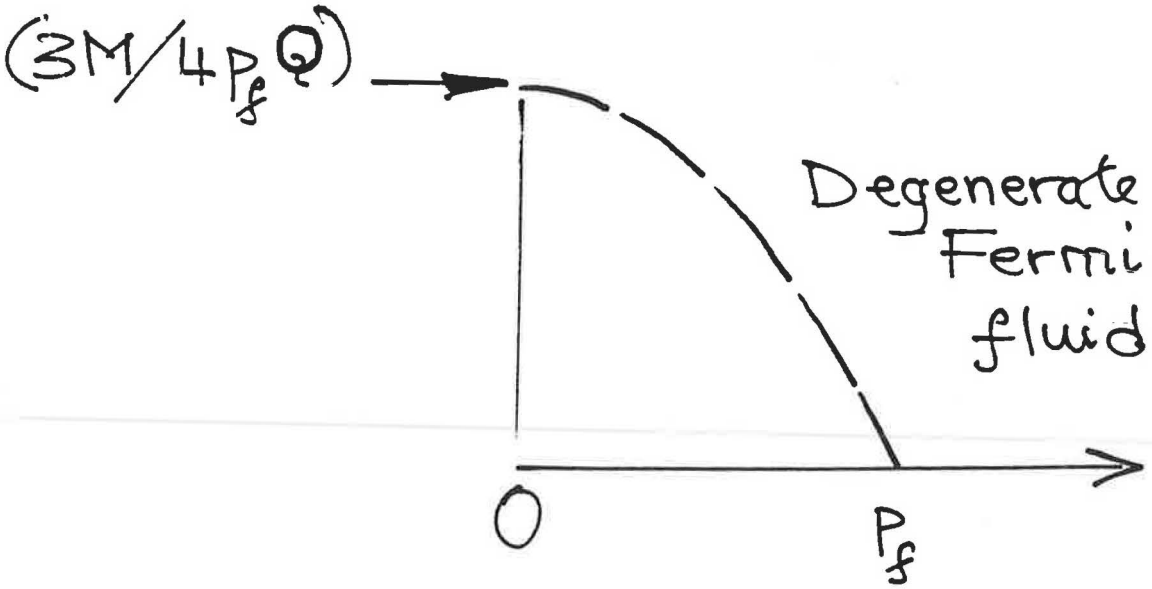
1. Data for the Compton profile of parahydrogen. There are no significant differences between data collected for samples at 4K and 20K (the solid and liquid phases respectively). The solid line is derived from a model wave function, incorporating previous spectroscopic data[3].

2. The Compton response functions are shown for an isotropic harmonic oscillator, and a perfect degenerate Fermi fluid. The corresponding analytic expressions are (4.6), (4.7) and (4.8).


J(y)
0.1
0.05
0
0
5
10
15
20
Momentum Y (Å^{-1})

Compton Limit of the Neutron Scattering Response Function $S_o(Q, \omega)$.





$$y = \{M(\omega - E_r)/Q\}.$$