X School of Neutron Scattering Francesco Paolo Ricci

Neutron scattering in the impulse approximation: Determination of the response function

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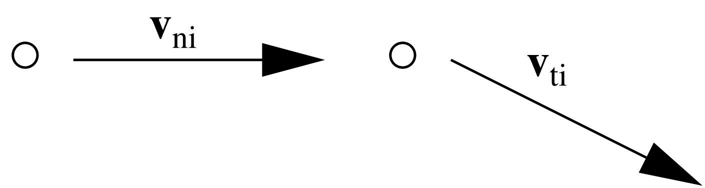
Neutron scattering in the Impulse Approximation (IA); scattering at high energy and wave vector transfer (Deep Inelastic Neutron Scattering, DINS) application to disordered systems such as quantum solids and liquids.

A classical analogue: an incident particle m_n and a target particle m_t=m_n with initial velocities $\mathbf{v}_{ni}$ and $\mathbf{v}_{ti}$



After the scattering m_n has velocity $\mathbf{v}_{nf}$, and we are able to measure only $\mathbf{v}_{ni}$, $\mathbf{v}_{nf}$

Apply conservation of momenta and kinetic energy

$$\begin{aligned} m_n = m_t &= m \\ \mathbf{v}_{ni} + \mathbf{v}_{ti} &= \mathbf{v}_{nf} + \mathbf{v}_{tf} \\ v_{ni}^2 + v_{ti}^2 &= v_{nf}^2 + v_{tf}^2 \end{aligned} \tag{1}$$

And defining

$$\begin{aligned} \Delta\mathbf{v} &= \mathbf{v}_{ni} - \mathbf{v}_{nf} \\ \omega &= v_{ni}^2 - v_{nf}^2 \end{aligned} \tag{2}$$

We obtain

$$\begin{aligned} \Delta\mathbf{v} + \mathbf{v}_{ti} &= \mathbf{v}_{tf} \\ v_{ti}^2 + \omega &= v_{tf}^2 \end{aligned} \tag{3}$$

Squaring the first equation and substituting into the second

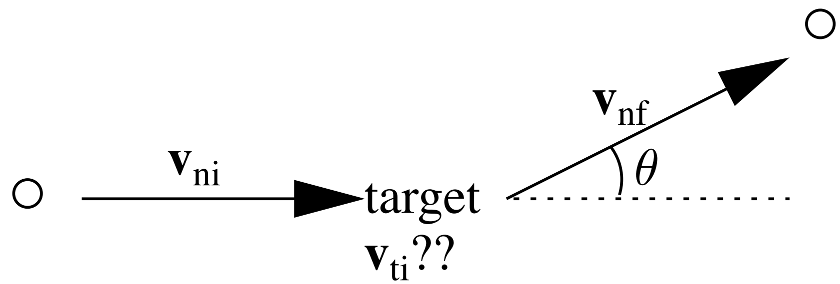
$$v_{tf}^2 = v_{ti}^2 + (\Delta v)^2 - 2\,\Delta\mathbf{v}\cdot\mathbf{v}_{ti} \tag{4}$$

$$v_{ti}^2 + \omega = v_{ti}^2 + (\Delta v)^2 - 2\,\Delta\mathbf{v}\cdot\mathbf{v}_{ti}$$

we have

$$\omega = (\Delta v)^2 - 2\,\Delta\mathbf{v}\cdot\mathbf{v}_{ti} = (\Delta v)^2 - 2\Delta v\, v_{ti}\cos\theta \tag{5}$$

where θ is the scattering angle!

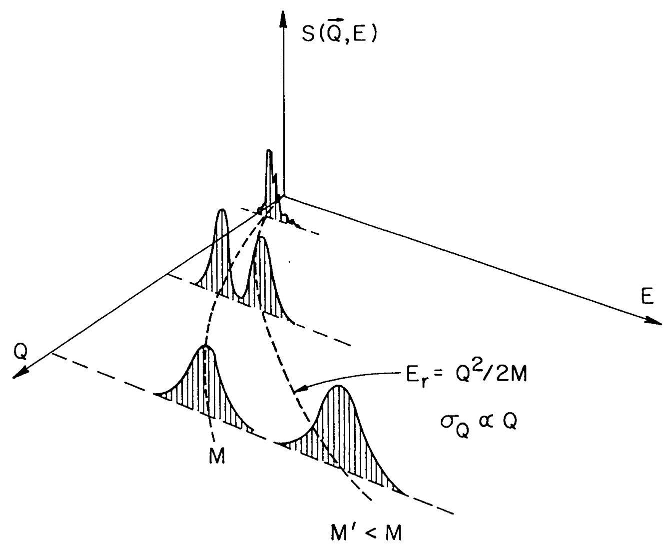


If we have point particles of (not necessarily) equal masses and we can measure the differences between initial and final velocities of the projectile (neutron?) and the deflection (scattering) angle we have access to the velocity of the target particle before being struck!

Focus on the short time behaviour of the correlation function

$$Y_{jj'}(\mathbf{Q},t) = \langle e^{-i\mathbf{Q}\cdot\mathbf{R}_j}\ e^{i\mathbf{Q}\cdot\mathbf{R}_{j'}(t)} \rangle \tag{6}$$

Relation between the measured scattering intensity and the longitudinal momentum distribution of the system under study: applications to the experimental investigations of the ground-state properties of condensed matter systems.



Dynamic structure factor "from collective to single particle" dynamics. Source: R. Simmons, in R. N. Silver (ed.) Proceedings of the 1984 Workshop on High-Energy Excitations in Condensed Matter, LA-10227-C (1984).

Neutron scattering: a probe of collective properties; when the magnitude of energy and wavevector increases, short-scale single-particle properties are probed.

The scattering occurs so rapidly, compared to the time-scales of atomic motion in the sample, that the measured response is rather simply related to the equilibrium momentum distribution of the atoms.

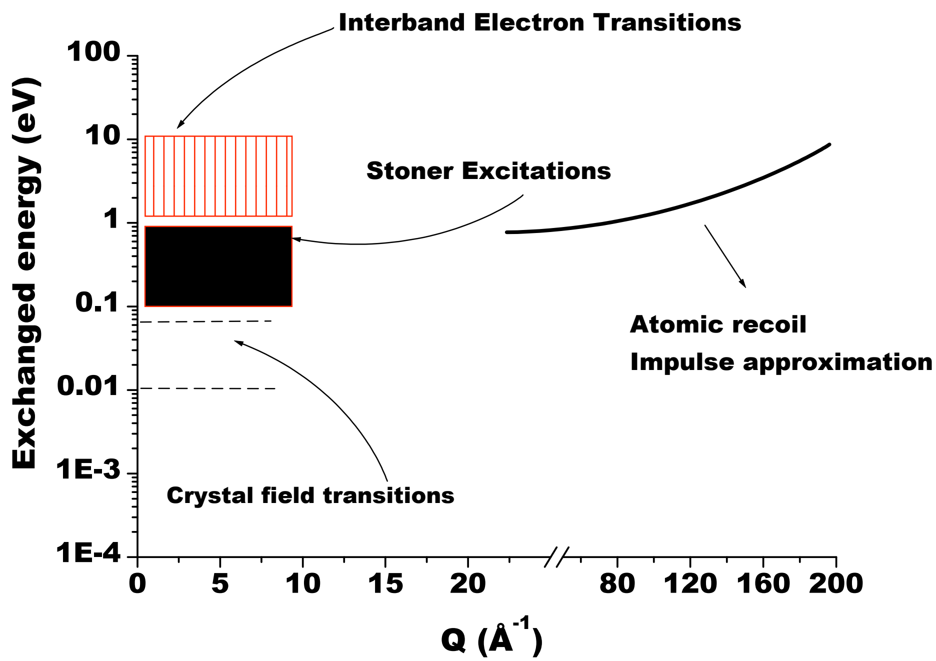
Momentum distributions are usually measured by scattering experiments in which the energy and momentum transferred are very high compared to the energies and momenta characteristic of ground-state properties and collective behaviour.

The scattering law may be related to the momentum distribution by invoking the Impulse Approximation (IA): a single particle of the system is struck by the scattering probe, and that this particle recoils freely from the collision.

Neutron scattering at energies above 1 eV is used to measure atomic momentum distribution n(p) in condensed matter, photon scattering at energies of tens of keV is used to measure electronic n(p) in condensed matter, electron scattering at GeV energies is used to measure n(p) of nucleons and inside nuclei.

. The information obtainable with Deep Inelastic Neutron Scattering (DINS)or neutron Compton scattering is to some extent complementary to that from diffraction experiments. The latter measures the Fourier transform of a time-averaged density; the former the instantaneous momentum density.

This is of particular interest if the motion of the atom of interest is well described by an effective (Born–Oppenheimer) potential, or adiabatic energy surface. An example is in molecular solids where a proton is bound in a heavy lattice at thermal energies (i.e. temperatures) well below the energy of the proton. In this case, the momentum distribution is the squared amplitude of the Fourier transform of the proton wave function, and from it, the potential energy function can be extracted (G. F. Reiter, J. Mayers and J. Noreland, *Phys. Rev.*, **B 65**, 104305 (2002).).



Schematics of the exchanged energy and wavevector range accessed by neutron scattering at eV energies.

The quantity which can be determined in a neutron scattering experiment is the partial differential cross section for the scattering of a beam of neutrons by a system of atomic nuclei. We will restrict our attention to the use of spectrometers operating at pulsed neutron sources, where the time-of-flight technique is used to determine the partial differential cross section.

If we consider a time channel of width Δ t centered at time t, then the rate at which counts are collected is (C. G. Windsor, Pulsed Neutron Scattering (Taylor and Francis, London) 1981):

$$C(t)\Delta t = I(E_0)\frac{dE_0}{dt}\Delta t N \frac{d^2\sigma}{d\Omega dE_1}\eta(E_1)\ \Delta\Omega\Delta E_1 \qquad (7)$$

where $I(E_0)\,\frac{dE_0}{dt}\,\Delta t$ is the number of incident neutrons per square centimeter, N is the number of scattering atoms, $\Delta\Omega$ is the detector solid angle, ΔE_1 the energy resolution of the analyser, $\eta(E_1)$ is the detector efficiency, and $\frac{d^2\sigma}{d\Omega dE_1}$ is the partial differential scattering cross section of the target system.

We will now focus on the behaviour of $\frac{d^2\sigma}{d\Omega dE_1}$ in the regime of high wavevector and energy transfers. The theory of "low energy" neutron scattering states(S. W. Lovesey, *Theory of Neutron Scattering from Condensed Matter,* 3rd ed. (Oxford University Press) 1987):

$$\frac{d^2\sigma}{d\Omega dE_1} = \frac{k_1}{k_0}\sigma_t S(\mathbf{Q},\omega) \tag{8}$$

$$S(\mathbf{Q},\omega) = \frac{1}{N\ 8\pi^2\hbar}\int_{-\infty}^{\infty} dt\, e^{-i\omega t}\sum_{jj'} Y_{jj'}(\mathbf{Q},t), \tag{9}$$

where $\mathbf{k}_0$ and $\mathbf{k}_1$ are the initial and final neutron wavevectors, and σ_t is the total scattering cross section:

$$4\pi(|\bar{b}|^2 + \delta_{jj'}(\overline{|b|^2} - |\bar{b}|^2)) \tag{10}$$

and $S(\mathbf{Q}, \omega)$ is the dynamic structure factor, where the correlation function is

$$Y_{jj'}(\mathbf{Q}, t) = \langle e^{-i\mathbf{Q}\cdot\mathbf{R}_j} e^{i\mathbf{Q}\cdot\mathbf{R}_{j'}(t)} \rangle \tag{11}$$

The operator $e^{-i\mathbf{Q}\cdot\mathbf{R}}$ couples the plane wave of the neutron with the position of the nucleus in the target system. The generic atom j has a position represented by the quantum mechanical operator $\mathbf{R}_j$. When the incident neutron energy is well in excess of the maximum energy available within the response spectrum of the target, the correlation function is approximated by its behavior at short times. The approximation involves a short time ($t \longrightarrow 0$) expansion of the atomic position operator :

$$\mathbf{R}_{j'}(t) = \mathbf{R}_{j'} + \frac{t}{M_{j'}}\mathbf{P}_{j'} \tag{12}$$

where $\mathbf{P}$ is the momentum of the struck nucleus of mass M.

Note that this expression includes the first correction term to the static approximation employed to describe elastic scattering (the second correction term is $1/2(\mathbf{F}/M)t^2)$. The basis of the approximation relies on the assumption that if the time of passage of the neutrons through the sample is short compared to the characteristic time scales of the samples, only the short-time properties are relevant. Now

$$Y_{jj'}(\mathbf{Q},t) = \langle e^{-i\mathbf{Q}\cdot\mathbf{R}_j}\ e^{\left(i\mathbf{Q}\cdot\mathbf{R}_{j'}+\frac{it}{M_{j'}}\mathbf{Q}\cdot\mathbf{P}_{j'}\right)}\rangle \qquad (13)$$

Remember that

$$[R_{\alpha j'}, P_{\beta j}] = i\hbar\, \delta_{jj'}\delta_{\alpha\beta} \tag{14}$$

and making use of the operator identity:

$$e^{A+B} = e^A e^B e^{\,{}^1\!/\!{}_2[A,B]}, \tag{15}$$

which holds when $[A, B]$ commutes with both A and B, then the exponentials in eq. 13 can be written

$$e^{-i\mathbf{Q}\cdot\mathbf{R}_j}\; e^{(i\mathbf{Q}\cdot\mathbf{R}_{j'}+\frac{it}{M_{j'}}\mathbf{Q}\cdot\mathbf{P}_{j'})} = \tag{16}$$

$$= e^{-i\mathbf{Q}\cdot\mathbf{R}_j+i\mathbf{Q}\cdot\mathbf{R}_{j'}+\frac{it}{M_{j'}}\mathbf{Q}\cdot\mathbf{P}_{j'}+\,{}^1\!/\!{}_2[\mathbf{Q}\cdot\mathbf{R}_j,(\mathbf{Q}\cdot\mathbf{R}_{j'}+\frac{t}{M_{j'}}\mathbf{Q}\cdot\mathbf{P}_{j'})]} \tag{17}$$

The commutator in the above equation involves position operators at the same time (=0), and commutation following eq.12.

The result is:

$$Y_{jj'}(\mathbf{Q},t) = e^{\frac{i\hbar tQ^2}{2M_j}\delta_{jj'}} \langle e^{i\mathbf{Q}(\mathbf{R}_{j'}-\mathbf{R}_j)+\frac{it}{M_{j'}}\mathbf{Q}\cdot\mathbf{P}_{j'}} \rangle \tag{18}$$

If we are interested in large Q behavior and considering that the spatial scale of the scattering event is given by $1/Q$, we can assume that correlations between the positions of different nuclei are absent and the incoherent approximation holds (as shown in the previous lectures). The exponentials containing position operators of different nuclei in the above correlation function will oscillate rapidly from atom to atom and cancel out on average.

$$Y_j(\mathbf{Q},t) = e^{\frac{i\hbar tQ^2}{2M_j}} \langle e^{\frac{it}{M_j}\mathbf{Q}\cdot\mathbf{P}_j} \rangle \tag{19}$$

$$S(\mathbf{Q},\omega) = \frac{1}{N\ 8\pi^2\hbar} \sum_j \int_{-\infty}^{\infty} dt\ e^{\left(-i\omega t + \frac{i\hbar t Q^2}{2M_j}\right)} \langle e^{\frac{it}{M_j}\mathbf{Q}\cdot\mathbf{P}_j} \rangle \tag{20}$$

The most important application of 20 is to observe the behavior of the correlation function in terms of momentum states (i.e. to measure probability distribution functions of atomic momentum in condensed matter). If we label these states by wave vectors $\mathbf{q}$,

$$\mathbf{P}|\mathbf{q}\rangle = \hbar\mathbf{q}|\mathbf{q}\rangle \tag{21}$$

we obtain:

$$\langle e^{\frac{it}{M_j}\mathbf{Q}\cdot\mathbf{P}_j}\rangle = \sum_{\mathbf{q}} n_{\mathbf{q}}\, e^{\frac{i\hbar t}{M_j}\mathbf{Q}\cdot\mathbf{q}} \tag{22}$$

. This can be inserted in 20:

$$S(\mathbf{Q},\omega) = \frac{1}{N\ 8\pi^2\hbar}\sum_j \int_{-\infty}^{\infty} dt\ e^{(-i\omega t + \frac{i\hbar t Q^2}{2M_j})}\langle e^{\frac{it}{M_j}\mathbf{Q}\cdot\mathbf{P}_j}\rangle = \tag{23}$$

$$= \frac{1}{N\ 8\pi^2\hbar} \sum_j \int_{-\infty}^{\infty} dt\ e^{(-i\omega t + \frac{i\hbar t Q^2}{2M_j})} \sum_{\mathbf{q}} n_{\mathbf{q}}\ e^{\frac{i\hbar t}{M_j}\mathbf{Q}\cdot\mathbf{q}} \tag{24}$$

$$= \frac{1}{8\pi^2\hbar} \sum_{\mathbf{q}} n_{\mathbf{q}} \int_{-\infty}^{\infty} dt\ e^{-it(\omega - \frac{\hbar\, Q^2}{2M} - \frac{\hbar}{M}\mathbf{Q}\cdot\mathbf{q})} \tag{25}$$

which results in:

$$S(\mathbf{Q},\omega) = \frac{1}{4\pi\hbar} \sum_{\mathbf{q}} n_{\mathbf{q}}\ \delta(\omega - \frac{\hbar\, Q^2}{2M} - \frac{\hbar}{M}\mathbf{Q}\cdot\mathbf{q}) \tag{26}$$

or

$$S(\mathbf{Q},\omega)_{IA} = \frac{1}{4\pi\hbar} \int_{-\infty}^{\infty} d\mathbf{q}\ n(\mathbf{q})\ \delta(\omega - \frac{\hbar\, Q^2}{2M} - \frac{\hbar}{M}\mathbf{Q}\cdot\mathbf{q}) \tag{27}$$

.

Now

$$\hbar\omega_R = \frac{\hbar^2 Q^2}{2M} \qquad (28)$$

is the free -atom recoil energy and having re-defined

$$n_{\mathbf{q}} = n(\mathbf{q}) = \langle \delta(\mathbf{q} - \mathbf{P}/\hbar) \rangle \qquad (29)$$

. Physical implications and related instrumental techniques Equation 27 is the form used to describe the dynamic structure function in terms of momentum distribution in the Impulse approximation framework. This description implies that, when in the IA regime (we will discuss briefly "when"), the scattering intensity, eq.7, measured in the experiment can be used to derive the single-particle momentum distribution of the atoms constituting the sample under study (27).

It is important to note that, although it is a single particle response, the momentum distribution is a property of the many-body system of all the atoms and their interactions. These "billiard ball" experiments have relevance in the description of the condensed system. If we consider a single atom at rest, then the scattering will be represented by $\delta(\omega - \frac{\hbar Q^2}{2M})$, i.e. a peak in the dynamic structure factor at the recoil energy

$$\hbar\omega_R = \frac{\hbar^2 Q^2}{2M} \tag{30}$$

(see Fig. 1) . Not all the target atoms will be at rest, and a probability distribution function (pdf) of atomic momentum will weight the peak of $S(\mathbf{Q}, \omega)$ to account for the spread of atomic momenta.

The determination of the extent and the nature of this "spread" is the objective of neutron scattering measurements in the Impulse Approximation, a technique known as Deep Inelastic Neutron Scattering (DINS). If we are measuring a system of integer-spin atoms at low temperature, we should be able to observe a fraction of atoms at zero-momentum (Bose condensate); if we measure a system of half-integer spin atoms at low temperature we should be able to observe an upper limit, corresponding to the radius of the Fermi sphere, in the distribution of atomic momenta (the maximum allowed value would be $|\mathbf{q}| = k_F$).

If we have a proton bound in a heavy lattice, the momentum distribution is the squared amplitude of the Fourier transform of the proton wave function, from which the potential energy function can be derived. This would allow to construct a database of Born-Oppenheimer potentials for this class of systems (G. F. Reiter, J. Mayers and J. Noreland, *Phys. Rev.*, **B 65**, 104305 (2002)).

In general, for condensed matter systems at low temperature ($0.1K < T < 30K$) the quantum zero-point motion results in an additional broadening of the recoil peak $\langle q_0^2 \rangle = \frac{2M}{\hbar^2} \langle E_K \rangle_0 > \frac{2M}{\hbar^2}\, k_B\, T$ where $\langle E_K \rangle_0$ is the zero-point single particle mean kinetic energy. This broadening is the signature of quantum effects in fluid and solid systems.

We will now briefly return to Eq.(12) to discuss the kinematic $(\mathbf{Q}, \omega)$ range of validity of the Impulse Approximation. The corrective term to Eq.(12) is related to the time derivative of the atomic momentum, i.e. the force exerted on the struck nucleus by the surrounding atoms:

$$\mathbf{R}(t) = \mathbf{R} + \frac{t}{M}\mathbf{P} + {}^{1}\!/_{2}(\mathbf{F}/M)t^2 + .. \tag{31}$$

where

$$\mathbf{F} = d\mathbf{P}/dt \tag{32}$$

. Now Eq.(12) is valid if

$$\langle (d\mathbf{P}/dt \cdot \mathbf{Q}) \rangle \tau \ll \langle (\mathbf{P} \cdot \mathbf{Q}) \rangle \tag{33}$$

, i.e the change in the atomic momentum due to the forces acting on the struck atom during the scattering process, $\langle d\mathbf{P}/dt \rangle \tau$, is negligible.

In eq. 33 τ is the interaction (scattering) time, which can be defined as the time over which $Y(\mathbf{Q},t)$ contributes to $S(\mathbf{Q},\omega)$, or alternatively, by the time-scale for the decay of $Y(\mathbf{Q},t)$ to zero. This quantity equals $1/\Delta\omega$ where $\Delta\omega$ is the width of $S(\mathbf{Q},\omega)$. This definition gives $\tau \simeq \frac{M}{Q\langle\mathbf{P}^2\rangle^{1/2}}$ where $\langle\mathbf{P}^2\rangle^{1/2}$ is the RMS momentum. For example, in quantum fluids and solids (liquid and solid helium) typically $\langle\mathbf{P}^2/\hbar^2\rangle^{1/2} \simeq 10$ nm^{-1}, while Q is above 1000 nm^{-1} for neutron scattering at the eV energies, validating the short time expansion in the assumption of finite interatomic forces (this means $\tau \simeq 1/Q$).

The above considerations are crude estimates of the validity of the IA as it has been demonstrated that for Q values up to 300 nm^{-1} corrections have to be applied. Rigorous descriptions of the range of validity of the IA are present in the condensed matter, nuclear and sub-nuclear physics literature (R. N. Silver and P. E. Sokol (eds) *Momentum Distributions* (Plenum Press, New York, 1989)).

Dynamic structure factor of an isotropic system in momentum space: longitudinal Compton profile.

In eq.(27), the dynamic structure factor in the IA limit, the scattering is no longer a function of the energy and wavevector transfer separately. Let us now consider the case of isotropic systems, such as a liquid or a polycrystalline solid. Then the probability distribution function of atomic momenta is isotropic.

NOTE in order to comply with the notation of present literature we will denote the atomic momentum probability distribution function by momentum distribution and will use the notation $\mathbf{p} \doteq \mathbf{q}$, so that eq.(27) becomes

$$S(\mathbf{Q},\omega)_{IA} = \frac{1}{4\pi\hbar}\int_{-\infty}^{\infty} d\mathbf{p}\, n(\mathbf{p})\, \delta(\omega - \frac{\hbar\, Q^2}{2M} - \frac{\hbar}{M}\mathbf{Q}\cdot\mathbf{p}) \quad (34)$$

having re-defined

$$n(\mathbf{p}) = \langle \delta(\mathbf{p} - \mathbf{P}/\hbar) \rangle \quad (35)$$

. Note that for isotropic systems The momentum (wavevector) distribution is $n(p)$.

Now, since ω and $\mathbf{Q}$ are closely related in the IA scattering regime it is useful to introduce a new variable y which couples wavevector and energy transfer (G. B.West, *Phys. Rep.*, **18**, 263 (1975).):

$$y = \frac{M}{\hbar^2 \, Q}(\hbar\omega - \hbar\omega_R) \tag{36}$$

and, considering that

$$\hbar\omega = \frac{\hbar^2(\mathbf{p}+\mathbf{Q})^2}{2M} - \frac{\hbar^2 \, \mathbf{p}^2}{2M} \tag{37}$$

, then y just represents the component of atomic wavevector along the scattering direction (i.e. $y = \mathbf{p} \cdot \widehat{\mathbf{Q}}$).

Now considering the delta function in eq. (34):

$$\delta(\omega - \frac{\hbar\, Q^2}{2M} - \frac{\hbar}{M}\mathbf{p}\cdot\mathbf{Q}) = \delta(\frac{\hbar\, Q}{M}\, y - \frac{\hbar}{M}\mathbf{p}\cdot\mathbf{Q}) \tag{38}$$

we have

$$\delta[\frac{\hbar\, Q}{M}(y - \mathbf{p}\cdot\widehat{\mathbf{Q}})] \tag{39}$$

. Using $\delta(ax) = \frac{\delta(x)}{|a|}$ we obtain

$$\frac{M}{\hbar Q}\,\delta(y - \mathbf{p}\cdot\widehat{\mathbf{Q}}) \tag{40}$$

. If we are again considering an isotropic system we can choose the z-axis to be along the scattering vector, then $\mathbf{p}\cdot\widehat{\mathbf{Q}} = p_z = y$

and we obtain:

$$S(\mathbf{Q},\omega)_{IA} = \frac{1}{4\pi\hbar}\int_{-\infty}^{\infty} d\mathbf{p}\, n(\mathbf{p})\, \frac{M}{\hbar Q}\,\delta(y - p_z) = \tag{41}$$

$$= \frac{M}{4\pi\hbar^2\, Q}\int_{-\infty}^{\infty} dp_x dp_y\, n(p_x, p_y, y) \tag{42}$$

. We have to perform an integration over p_x and p_y. We can define $\mathbf{p}_\perp = (p_x, p_y)$, therefore using polar coordinates $dp_x dp_y = p_\perp dp_\perp d\theta$.

We have

$$\frac{M}{4\pi\hbar^2 Q}\int_0^{2\pi} d\theta \int_0^{\infty} dp_\perp\, p_\perp\, n(p_x, p_y, y) \tag{43}$$

. Now if $n(\mathbf{p}) = n(p)$ we have

$$p = \sqrt{p_x^2 + p_y^2 + y^2} = \sqrt{p_\perp^2 + y^2} \geq |y| \tag{44}$$

, then Eq. (43) , being $p\, dp = p_\perp\, dp_\perp$ becomes

$$\frac{M}{4\pi\hbar^2 Q} 2\pi \int_{|y|}^{\infty} dp\, p\, n(p) \tag{45}$$

where the lower limit of integration recalls the fact that, from Eqs.(44,43), $p \geq |y|$.

Now if we define a new function

$$J(y) = \frac{2\pi\hbar^2 Q}{M} S(\mathbf{Q}, \omega)_{IA} \tag{46}$$

, then J(y) will be maximum in zero

$$J(0) = \int_0^{\infty} dp\, p\, n(p) \tag{47}$$

, and, in general

$$J(y) = \int_{|y|}^{\infty} dp\, p\, n(p) \tag{48}$$

.Note that $y = 0$ corresponds to the centroid of the recoil peak. The above equation is the important result which is used in neutron scattering within the IA which relates the dynamic structure factor to the momentum distribution of an isotropic system along one direction.

$J(y)$ is called Neutron Compton Profile (NCP) or Longitudinal Compton Profile (similarly to photon-electron scattering and electron-nucleon scattering techniques), and represents the probability distribution function of atomic momentum along $\widehat{\mathbf{Q}}$ or z, i.e. n(y). From the general moment relations for the incoherent response functions, it is possible to derive the single-atom mean kinetic energy, $\langle E_K \rangle$:

$$\langle E_K \rangle = \frac{3\hbar}{2\ M} \int_{-\infty}^{\infty} y^2\ J(y) dy \tag{49}$$

where, by definition of probability distribution function $\int_{-\infty}^{\infty} J(y) dy = 1$ and zero first moment.