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Abstract

We show that the optical theorem – valid for neutron scattering in the approximation of first order local field correction – can be interpreted as a sum rule. Within this framework the connection of the Debye-Waller factor with diffusive anti-Stokes scattering channels becomes clear.

1 Introduction

In neutron and X-ray scattering, as well as in Mössbauer experiments, all the signals of coherence (e.g. Bragg-peaks) are diminished by a thermal form-factor, the Debye-Waller factor. This temperature dependent factor, however, is also effective on incoherent scattering processes. Indeed, the angular anisotropy of e.g. incoherent elastic neutron scattering is entirely determined by the Debye-Waller factor. At schools and workshops the experts are then often confronted with the obvious question: "Where do all these events for the case of higher temperature of the sample then go to?" The answer to this question is usually not even wrong, but accompanied by so much handwaving that it can not satisfy the audience.

A key for an unambigous answer to this question is the optical theorem (Bohr-Peierls-Placzek) which relates the total cross-section to the scattering amplitude for the foward direction. The optical theorem is a direct consequence of the unitarity of the S-matrix in scattering theory, that is it expresses the conservation of probability. Together with the causality condition, which leads to the Kramers-Kronig relation of the scattering amplitude, unitarity is a basic principle of formal scattering theory and leads to relationships which are exact and always true. On the other hand, there exist model approximations which - otherwise successful - violate these basic principles. A prominent example is the scattering cross-section obtained with the kinematical approximation and Born approximation, for the matrix element of the elementary scattering. This approach leads to the scattering functions, normally used to satisfactorily describe the experimental data.

The optical theorem can be expressed in the well known form [1]

$$\mathcal{I}m f(0) = \frac{k}{4\pi}\sigma_{tot} \tag{1}$$

with k being the momentum of the incident probing particle. $f(0)$ is the forward scattering amplitude and σ_{tot} the total cross-section. The Born approximation has an amplitude which is purely real in forward direction and hence cannot fulfill the optical theorem. In other words, it does not properly describe the diffractive shadow within the angular range

$$\vartheta \simeq \frac{1}{rk} \tag{2}$$

behind the scattering center (r is the size of the obstacle).

For neutron scattering the elementary interaction on nuclei is usually described by a Fermi-pseudopotential and is then for thermal energies just expressed by a scattering length. If we neglect nuclear absorption this number is purely real.

As a cure for the shortcoming of the Born approximation we may use for the elementary scattering amplitude the effective range approximation, which for a hard sphere becomes

$$f = -b + ib^2k \tag{3}$$

where b is the bound scattering length.

To achieve consistency we now have to include local field corrections as well, since they turn out to be of the same order of magnitude as the effective range term in (3). Within this framework we may explicitely formulate (1) consistently and use it as a sum rule.

The Optical Theorem

The scattered wave of a neutron scattering process with a transition $\alpha \to \alpha'$ in the sample can be written as

$$\psi_{\alpha\alpha'}(\vec{x}) = \delta_{\alpha\alpha'} e^{i\vec{k}\vec{x}} + < \vec{k}'\alpha' \mid F \mid \vec{k}\alpha > \frac{e^{ik'x}}{x} \tag{4}$$

with F as the matrix of the scattering amplitude and $\vec{k}\,'$ is the momentum of the scattered particle. The incident flux is

$$\mathcal{I} = \frac{\hbar k}{m} \tag{5}$$

and the scattered flux

$$\mathcal{I}' = \frac{\hbar k'}{m} \cdot \frac{1}{x^2} \mid < \vec{k}'\alpha' \mid F \mid \vec{k}\alpha > \mid^2 \tag{6}$$

The differential cross-section into the solid angle $d\Omega$ is then

$$d\sigma_{\alpha\alpha'} = \frac{\mathcal{I}'}{\mathcal{I}} x^2 d\Omega = \frac{k'}{k} \mid < \vec{k}'\alpha' \mid F \mid \vec{k}\alpha > \mid^2 d\Omega \tag{7}$$

For the total differential cross-section we have to average over the initial states of the sample and to sum over the final states. Let p_α be the probability distribution of the population of the internal degrees of freedom for the sample. Then

$$\frac{d\sigma}{d\Omega d\epsilon'} = \frac{k'}{k} \sum_{\alpha,\alpha'} p_\alpha \, |< \vec{k}'\alpha' \mid F \mid \alpha\vec{k} >|^2 \, \delta(\omega - \omega_{\alpha,\alpha'}) \tag{8}$$

where $\hbar\omega_{\alpha,\alpha'} = \epsilon - \epsilon'$ is the energy transfer to the sample. For a sample in thermal equilibrium p_α is the canonical distribution

$$p_\alpha = e^{-\beta E_\alpha} / \sum_\alpha e^{-\beta E_\alpha} \tag{9}$$

with $\beta = 1/kT$ and T is the temperature of the sample.

Let us now derive the optical theorem directly from particle conservation for the scattering process,

$$\partial_t \rho + \vec{\nabla} \cdot \vec{\mathcal{I}} = -s \tag{10}$$

where

$\rho = \mid \psi \mid^2$ is the particle (neutron) density

and

$\vec{\mathcal{I}} = \mathcal{R}e(\psi^* \vec{v} \psi)$ is the corresponding current $\left(\vec{v} = \frac{\hbar}{im}\vec{\nabla}\right)$. In (10) s is the absorption rate.

Integration of (10) over a large volume V_3 around the scattering event gives the rate of change of the number of particles

$$\dot{N} = -\int_{\partial V_3} \vec{\mathcal{I}} d\vec{\sigma} - \int_{V_3} s d^3 x \tag{11}$$

where ∂V_3 designates the boundary of V_3. The absorption cross-section is defined by

$$\sigma_a = \frac{1}{\mathcal{I}} \int_{\partial V_3} \vec{\mathcal{I}}' d\vec{\sigma} = \sum_{\alpha\alpha'} \left(-\frac{x^2}{\mathcal{I}}\right) \int d\Omega \; \vec{x} \; \vec{\mathcal{I}}'_{\alpha\alpha'}. \tag{12}$$

Using (5) and (6) and the asymptotic wave (4) we carry out the integration and obtain

$$\sigma_\alpha = \frac{4\pi}{k} \sum_\alpha p_\alpha \mathcal{I}m < \vec{k}\alpha \mid F \mid \vec{k}\alpha > - \sum_{\alpha\alpha'} p_\alpha \left(\frac{k'}{k}\right) \int d\Omega \, |< \vec{k}'\alpha' \mid F \mid \vec{k}\alpha >|^2 \tag{13}$$

The second term to the left is the scattering cross-section. Hence, we have

$$\sigma_{tot} = \sigma_a + \sigma_s = \frac{4\pi}{k}\mathcal{I}m < \vec{k}\alpha \mid F \mid \vec{k}\alpha > . \tag{14}$$

On the right side appears the imaginary part of the forward amplitude for elastic scattering. This amplitude expresses the scattering in absolute coherence with the incident beam. For this quantity we shall write $f(0)$.

Since the neutron absorption in neutron scattering describes a nuclear property of the sample, we are in our context not interested in this effect. We hence assume the sample to be non-absorptive, that is $\sigma_a = 0$. The optical theorem has then the form

$$\frac{4\pi}{k}\mathcal{I}mf(0) = \sum_{\alpha,\alpha'} p_\alpha \left(\frac{k'}{k}\right) \int d\Omega \, |< \vec{k}\,'\alpha' \mid F \mid \vec{k}\alpha >|^2 \tag{15}$$

after integration of (8) over ω and Ω. Note that the amplitude F consists of an absolutely coherent part f and the part δF which describes diffuse scattering. The latter is proportional to the fluctuations of the scattering length

$$\delta b(\vec{x}) = b_c \delta\rho(\vec{x}) + \sum_j \delta b_j \delta(\vec{x} - \vec{X}_j) \tag{16}$$

b_c is the coherent scattering length which also determines f. In (16) $\delta\rho$ stands for the density fluctuations of the sample due to excitations of internal degrees of freedom, and δb_j for the "local" fluctuations of the scattering length of the individual scattering centers (spin, isospin). The first term of (16) is responsible for the ineleastic scattering with relative coherence; the second term describes the incoherent elastic and inelastic scattering phenomena. Two remarks are here in order:

1. Since there is no interference between these two contributions to the right hand side of (15), both are proportional to the squares of their corresponding scattering lengths.
2. All the terms on the right side of (15) contain a Debye-Waller form-factor $e^{-2W(\kappa)}$ and are accordingly diminished for large momentum transfer $\mid \vec{\kappa} \mid$, due to the thermal agitation in the sample; $W(\vec{\kappa} = 0) = 0$.

The Local Field Correction (Ewald Correction) [2]

As already mentioned in the introduction, the optical theorem is not fulfilled at the level of the Born approximation. Since the scattering amplitude enters linearly, higher orders of the scattering theory have to be included for $Imf(0)$ in order to achieve consistency with the optical theorem.

One way forward is the use of the effective range approximation (3) for the elementary scattering amplitude. For consistency reasons we have to include also the local field corrections up to first order in the scattering theory. The asymptotic wave is then given by

$$\psi(\vec{x}) = \lim_{|\vec{x}|\to\infty} \left(e^{i\vec{k}\vec{x}} + M \cdot \frac{e^{ik'x}}{x} \right) \tag{17}$$

with

$$M = f \int_{C_3} d^3x' \chi(\vec{x}\,') e^{-i\vec{k}\vec{x}\,'} \tag{18}$$

as scattering amplitude, including the local field corrections, and where χ is the local field. Its value at the position of a scattering center (i) is

$$\chi_i = e^{i\vec{k}\vec{x}_i} + \sum_{j\neq i} G(\vec{x}_i - \vec{x}_j) f_i \chi_j \tag{19}$$

where G is the Green's function for free propagation from scattering center (j) to (i), and f_j is the elementary scattering amplitude of the center (j). Within the approximation we are aiming for, we can cut the iteration of (19) after the first step (first order local field correction) and may, moreover, treat the problem in the thermo-dynamical limit:

$$\left.\begin{array}{l} N \to \infty \\ V \to \infty \end{array}\right\} \text{such that } \ \rho = N/V \to const. \tag{20}$$

For this case $\chi(\vec{x})$ can be written as a plane wave

$$\chi(\vec{x}) = c\psi(\vec{x}) = e^{i\vec{K}\vec{x}} \tag{21}$$

with a wave number

$$K^2 = k^2 + 4\pi c\rho f \tag{22}$$

and

$$c = (1-J)^{-1} \simeq (1+J)$$

$$J = \rho f \cdot \int d^3x e^{i\vec{k}\vec{x}} G(\vec{x})\{1-g(\vec{x})\} + O(b^3) \tag{23}$$

$g(\vec{x})$ is (within this approximation) the **static** correlation function for a homogenous medium. Dynamical effects would enter here in the next higher approximation. For the free propagator we write

$$G(\vec{x}) = \frac{1}{2\pi^2} \int d^3k' \frac{e^{i\vec{k}\vec{x}}}{k'^2 - i\epsilon - k^2} \tag{24}$$

Together with the coherent static structure factor

$$S_c(\vec{\kappa}) = 1 + \rho \int [g(r) - 1] e^{i\vec{\kappa}\vec{x}} d^3x \tag{25}$$

we obtain for J

$$J = \frac{b}{2\pi^2} \int d^3k' \frac{1 - S(\vec{\kappa})}{k'^2 - i\epsilon - k^2} \tag{26}$$

With $\vec{k}$ along the z-axis and $\vec{k}\,'$ in the direction (ϑ, ϕ) we can write

$$d^3k' = k'^2 dk' \sin\vartheta d\vartheta d\phi = \frac{mk'}{\hbar^2} d\Omega d\epsilon' \tag{27}$$

With the help of

$$\lim_{\epsilon \to 0} \frac{1}{\omega + i\epsilon} = P\left(\frac{1}{\omega}\right) - i\pi\delta(\omega) \tag{28}$$

we obtain finally for the imaginary part of J

$$\mathcal{I}mJ = \frac{kb}{4\pi} \int d\Omega [1 - S_c(\vec{\kappa})] \tag{29}$$

evaluated at $k = k'$. With (3), (18) and (29) we finally establish for the imaginary part of the forward scattering amplitude

$$\mathcal{I}mM(0) \simeq kb^2 \int S_c(\vec{\kappa}) d\Omega + O(b^3) \tag{30}$$

With (30) and (15) we may now express the optical theorem consistently up to orders $O(b^2)$. $\mathcal{I}mM(0)$ is the forward scattering amplitude including the first-order local field corrections.

It can be expressed by the static correlation function

$$S_c(\vec{\kappa}) = \int_{-\infty}^{+\infty} S_c(\vec{\kappa}, \omega) d\omega \sim <|\, \delta\rho_{\vec{\kappa}} \,|^2>_T \tag{31}$$

which is the canonical average at temperature T of the square of the density fluctuations

$$\delta\rho_{\vec{K}} = \rho_{\vec{K}} - <\rho_{\vec{K}}> \tag{32}$$

We know that $<(\delta\rho)^2>$ is related to the isothermal compressibility of the sample or the thermal expansion parameters (α) and the specific heat C_v, C_p. [3].

$$<(\delta\rho)^2> = \frac{\rho^2}{v\beta}\kappa_T = k\rho^2 \frac{T^2\alpha^2}{C_p - C_v}. \tag{33}$$

with (v is the specific volume)

$$\kappa_T = -\frac{1}{v}(\partial_p v)_T \quad \text{and} \quad \alpha = \frac{1}{v}(\partial_T v)_p$$

From the third Law of Thermodynamics (W. Nernst) we have for the entropy

$$\lim_{T\to 0} S(T,v) = 0 \tag{34}$$

From this we conclude

$$\lim_{T\to 0} \alpha = 0 \quad \text{and} \quad \lim_{T\to 0} \frac{C_p - C_v}{T} = 0 \tag{35}$$

From (33) we recognize that the fluctuations disappear as $O(\alpha^2)$ with vanishing temperature. In spite of a "classical" approximation invoked by (31) we may nevertheless safely assume that

$$<(\delta\rho)^2>_{T=0} \quad \leq \quad <(\delta\rho)^2>_T \tag{36}$$

and by writing

$$B(T) = \int <|\,\delta\rho_\kappa\,|^2>_T \, d\Omega \tag{37}$$

$$\boxed{B(T) \geq B(0)} \tag{38}$$

The optical theorem (15) consistent up to orders $O(b^2)$ including local field correlations (replacement of $f(0)$ by $M(0)$ can then be written as [1]

[1]The assumptions of a hard sphere model in (3) and $\sigma_a = 0$ are somewhat arbitrary. However both concern nuclear properties and are hence independent of temperature. In the subtraction leading to (43) these possible constants on the left side of (39)/(40) cancel.

$$kb^2B(0) = \sum_{\alpha'} k'(\alpha')A(|\ \vec{\kappa}\ |, \alpha_0 \to \alpha')e^{-2W_0(\vec{\kappa})} \tag{39}$$

for $T = 0$, and

$$kb^2B(T) = \sum_{\alpha\alpha'} p_\alpha k'(\alpha')A(|\ \vec{\kappa}\ |, \alpha_0 \to \alpha')e^{-2W(\vec{\kappa})} \tag{40}$$

for $T \neq 0$, with

$$A = \int d\Omega\ |< \vec{k}\ '\alpha' \mid F \mid \vec{k}\alpha >|^2 \tag{41}$$

For convenience we have separated off the Debye-Waller factor. In (39)/(40) α_0 designates the ground state of the sample. The p_α's give the population probabilities at thermal equilibrium

$$p_\alpha = \frac{e^{-\beta E_\alpha}}{\sum_\alpha e^{-\beta E_\alpha}} \quad , \quad \sum_\alpha p_\alpha = 1 \tag{42}$$

We use now (38) in order to compare (39) with (40) and obtain

$$\boxed{\frac{\sum_{\alpha,\alpha'} p_\alpha k'(\alpha')A(|\ \vec{\kappa}\ |), \alpha \to \alpha')}{\sum'_\alpha k'(\alpha')A(|\ \vec{\kappa}\ |, \alpha_0 \to \alpha')} \geq \frac{e^{-2W_0(\vec{\kappa})}}{e^{-2W(\vec{\kappa})}} \geq 1 \qquad \forall \vec{\kappa}} \tag{43}$$

where the second inequality follows from

$$e^{-2W(\vec{\kappa})} \leq e^{-2W_0(\vec{\kappa})} \qquad \forall \vec{\kappa} \tag{44}$$

These inequalities show how the diminuation of the cross-sections by the Debye-Waller factor has to be compensated by the more extensive sum over the initial states of the sample α, that is by the opening of diffusive anti-Stokes channels. Note that this sum rule is valid for every individual momentum transfer $|\ \vec{\kappa}\ |$.

This becomes fairly obvious with the help of Figure 1, which shows the kinematical region of access in momentum- and energy-transfer of the scattering function for a neutron with incident energy E_0. For a constant momentum transfer $|\ \vec{\kappa}\ |$ and a sample at "zero" temperature the events are restricted to the domain of positive energy transfer, that is transfer from the neutron to the sample. For higher temperature of the sample these events are reduced by the Debye-Waller-factor. Instead the domain with negative energy transfer (from the sample to the neutron) is now populated.

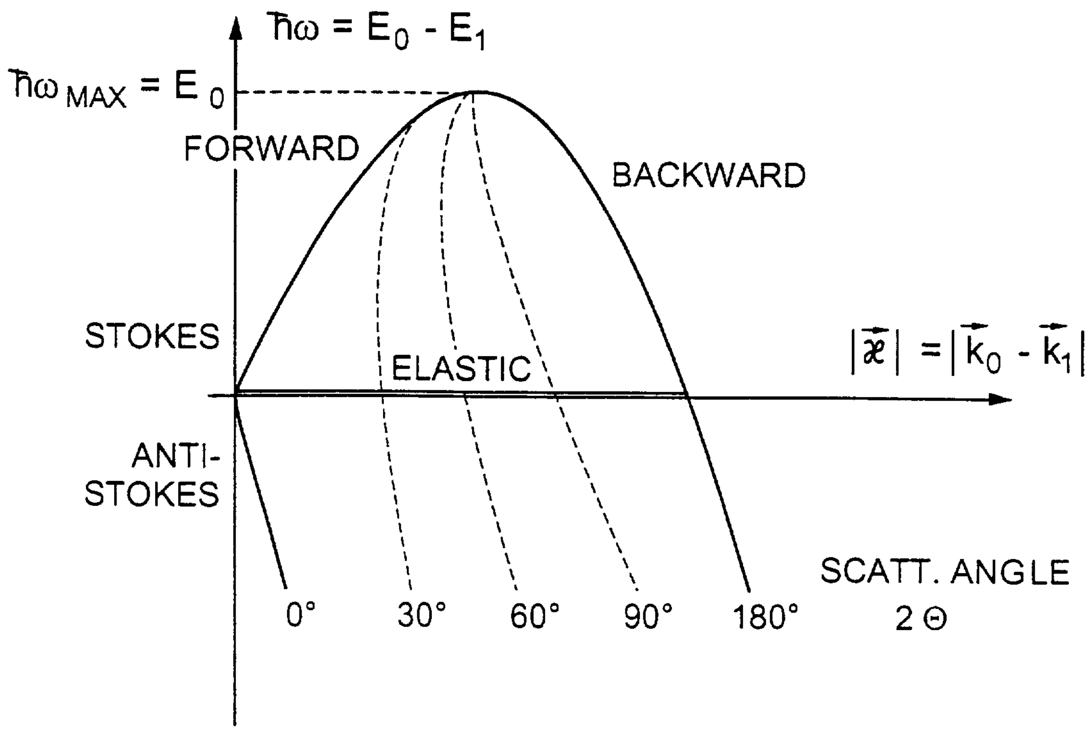

Figure 1: Kinematical domain in energy - and momentum transfer for neutron scattering with incident beam energy E_0. The integration over $d\Omega$ in (37) and (39)/(40) is along a vertical $|\vec{\kappa}| = const.$ over all angles accessible.

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