Intermediate Scattering Function in Slow Neutron Scattering*

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The conditions under which the so-called intermediate scattering function occurring in the theory of slow neutron scattering and in the theory of the Mössbauer effect can be written in the form $\exp[-\kappa^2\gamma(t)]$ have been explicitly stated; the intermediate scattering function is then factorizable into two parts; the part referred to in the paper as the displacement part gives on Fourier transformation a real space-time function which, quite unambiguously, has the meaning usually attributed to the Van Hove $G_s(\mathbf{x},t)$ function; the other part arises out of the nucleus recoiling against the neutron. It is shown that for systems in thermal equilibrium, the recoil part can be expressed in terms of the displacement part. This relation enables one to take care of the recoil part in case a classical approximation is made for the dynamics of the scattering system.

I. FACTORIZATION OF THE INTERMEDIATE SCATTERING FUNCTION

IN the theory of slow neutron scattering when one is concerned with the incoherent part of the cross section, one has to evaluate average values of operators of the type

$$\phi(\boldsymbol{\kappa},t)=\exp(-iHt/\hbar)\exp(-i\kappa x)\times\exp(iHt/\hbar)\exp(+i\kappa x), \quad (1)$$

where $\hbar\boldsymbol{\kappa}$ is the momentum lost (initial-final value) by the neutron, κ is the magnitude of $\boldsymbol{\kappa}$, x is the position coordinate of the scattering nucleus in the direction of $\boldsymbol{\kappa}$. We shall further introduce the notation $\hbar\omega_R$ for E_R, the energy of recoil of the scattering nucleus of mass M, v for the component of its velocity operator in the direction of $\boldsymbol{\kappa}$. It is then easy to show (see, for example, Rahman, Singwi, and Sjölander[1]) that

$$\phi(\boldsymbol{\kappa},t)=\exp(i\omega_R t)A(\boldsymbol{\kappa},t), \quad (2)$$

$$dA(\boldsymbol{\kappa},t)/dt=i\kappa v(-t)A(\boldsymbol{\kappa},t), \quad (3)$$

where $v(-t)$ is the operator $\exp(-iHt/\hbar)v\exp(iHt/\hbar)$.

A formal solution of Eq. (3) is the Baker-Haussdorff formula and the reader is referred to Weiss and Maradudin[2] for details and references. Denoting $i\kappa v(-t)$ by $B(t)$ for short, and writing $C(\tau)$ for the integrand occurring in Eq. (6) below, we have

$$A(\boldsymbol{\kappa},t)=\exp(z_1+z_2+z_3+z_4+\cdots), \quad (4)$$

$$z_1=\int_0^t B(\tau)d\tau, \quad (5)$$

$$z_2=\frac{1}{2}\int_0^t\left[B(\tau),\int_0^\tau B(\sigma)d\sigma\right]d\tau=\frac{1}{2}\int_0^t C(\tau)d\tau, \quad (6)$$

$$z_3=\frac{1}{4}\int_0^t\left[B(\tau),\int_0^\tau C(\sigma)d\sigma\right]d\tau+\frac{1}{12}\int_0^t\left[C(\tau),\int_0^\tau B(\sigma)d\sigma\right]d\tau, \quad (7)$$

etc.

In this the z's have been given indices related to the number of times B occurs in the expression for that particular z. Since in our case B contains the parameter κ, z_n is in fact the term with $(\kappa)^n$ as factor. Further, the commutator $C(t)\equiv[B(t),z_1(t)]$ has been singled out in the above expressions to bring out the fact that all following terms, z_3, z_4, etc., are identically zero in the special case that $C(t)$ is a c number. In general, $C(t)$ is not a c number.

Introducing the notation $X(t)$ for $x(0)-x(-t)$, one can easily show that

$$z_1=i\kappa X(t), \quad (8)$$

$$z_2=-i\omega_R t+[x(-t),x(0)]. \quad (9)$$

Hence,

$$\phi(\boldsymbol{\kappa},t)=\exp\{i\kappa X(t)+[x(-t),x(0)]\kappa^2/2+z_3+\cdots\}, \quad (10)$$

and in case $C(\tau)$ is a c number

$$\phi(\boldsymbol{\kappa},t)=\exp[i\kappa X(t)]\exp\{[x(-t),x(0)]\kappa^2/2\}. \quad (11)$$

This result, of course, can be more directly seen from (1) itself. The elaboration indulged in above is meant to throw some light on the nature of the so-called Gaussian approximation to be mentioned later.

First, let us consider a few special cases. In all three cases (11) will apply.

1. Freely Moving Particle

For a particle moving with velocity v, $X(t)=vt$ and, therefore,

$$\phi(\boldsymbol{\kappa},t)=\exp(i\kappa vt)\exp(i\omega_R t). \quad (12)$$

This is quite a familiar result. Here we would like to point out that it is the first term $\exp(i\kappa vt)$ which contains the velocity of the particle whereas the second is independent of v and contains only the mass of the scattering nucleus. It is obvious that the second arises from the recoil of the scattering nucleus and has nothing to do with its initial state of motion.

2. Harmonic Oscillator

If the oscillator frequency is $\omega_{\rm osc}$, we get

$$\phi(\boldsymbol{\kappa},t)=\exp[i\kappa X_{\rm osc}(t)]\exp(i\omega_R\sin\omega_{\rm osc}t/\omega_{\rm osc}). \quad (13)$$

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[1] A. Rahman, K. S. Singwi, and A. Sjölander, Phys. Rev. **126**, 986 (1962).

[2] G. H. Weiss and A. A. Maradudin, J. Math. Phys. **3**, 771 (1962).

The displacement operator $X_{\rm osc}(t)$ is

$$X_{\rm osc}(t)=x(0)(1-\cos\omega_{\rm osc}t)+v(0)\sin\omega_{\rm osc}t/\omega_{\rm osc},$$

and, hence, depends on the initial state of motion through $x(0)$ and $v(0)$. On the other hand, the term in ω_R is independent of $x(0)$ and $v(0)$.

3. Vineyard-Langevin Equation

In the equation considered by Vineyard[3] one gets

$$\phi(\kappa,t)=\exp[i\kappa X_L(t)]\times\exp\{i\omega_R[1-\exp(-\beta|t|)]/\beta\}, \quad (14)$$

where β is the damping parameter. Here again the same remarks as for (12) and (13) are applicable. $X_L(t)$ is given in Vineyard's paper.[3]

The purpose of showing the three Eqs. (12), (13), and (14) is to bring out the fact that *before* one has started to average $\phi(\kappa,t)$ to get the so-called intermediate scattering function $F_s(\kappa,t)$, in the notation of reference 1, certain terms involving ω_R get separated out; these terms always start with $\exp(i\omega_R t)$ for small times but their eventual form depends on the nature of the binding of the recoiling nucleus. Furthermore, these terms are independent of the initial state of motion of the scattering nucleus.

We shall, therefore, refer to the part of $\phi(\kappa,t)$ arising from the commutator $[x(-t), x(0)]$ in (11) as the recoil part and $\exp[i\kappa X(t)]$ as the displacement part. This separation has a deeper meaning in the sense that the scattering is determined in part by the intrinsic motion of the scattering nuclei bringing into play only the wave aspect of the neutron wave front; the scattering is also affected by the fact that the neutron carries a mass and, hence, through the requirements of momentum conservation gives a recoil to the scattering nucleus, irrespective of the state of motion of the nucleus.

To clarify the meaning of the separation mentioned above, one can consider the Fourier transform with respect to κ of $\exp[i\kappa X(t)]$ alone; one then gets $\delta(\mathbf{x}-\mathbf{X}(t))$. Through this transform one can define a new function $M_s(\mathbf{x},t)$ which has an unambiguous meaning of the kind envisaged for $G_s(\mathbf{x},t)$ by Van Hove. Let

$$M_s(\mathbf{x},t)=\langle\delta(\mathbf{x}-\mathbf{X}(t))\rangle, \quad (15)$$

where $\langle\cdots\rangle$ implies the usual averaging process.[4] In fact, taking the average of the displacement part $\langle\exp[i\kappa X(t)]\rangle$ before taking its transform, one finds that the average is a real function since all odd powers of the displacement should average out to zero. Hence, quite generally, $M_s(\mathbf{x},t)$ has the probabilistic interpretation usually attributed to $G_s(\mathbf{x},t)$; the Van Hove function is complex and, hence, one encounters difficulties in attributing to it an interpretation in terms of a probability; this is obviated in dealing with $M_s(\mathbf{x},t)$.

[3] G. H. Vineyard, Phys. Rev. **110**, 999 (1958).

[4] The interference counterpart of $M_s(x,t)$ can similarly be defined but through a convolution of two δ functions.

The conclusion, thus, is that we have two alternative ways of formulating the problem of scattering. One can write the scattering in terms of a simple four-dimensional transform of *one* function, the Van Hove $G_s(\mathbf{x},t)$ function. However, $G_s(\mathbf{x},t)$ *must* be a complex function.

The other alternative is to break up the intermediate scattering function, which is the average of $\phi(\kappa,t)$ into a product of two parts, one arising from the displacement of the scattering nuclei irrespective of their encounter with the neutron, the other from their recoil against the neutron. One can then identify, through the first part, a space-time function $M_s(\mathbf{x},t)$ which contributes to but does not completely determine the scattering of the neutrons.

The advantage of sacrificing the formal simplicity of introducing the $G_s(\mathbf{x},t)$ function of Van Hove is apparent when one tries to make a classical approximation for the dynamics of the scattering system.

When both the neutron and the scattering system are considered as classical particles, one can solve the scattering problem completely without invoking the coordinates of the scattering particles but only their velocities.

However, since the whole purpose of slowing down the neutrons is to emphasize their de Broglie wavelength relative to the distances between the scattering nuclei, it is obvious that this point of view is of little value or consequence.

One would, therefore, welcome the possibility of treating the neutron as a wave particle and at the same time the dynamics of the scattering nuclei classically. It will be shown below that, at least in the so-called Gaussian approximation, this can be done correctly, provided the recoil part of $\phi(\kappa,t)$ is first taken care of through the commutator $[x(0),x(t)]$. This point of view is already implicit in the work of Rahman *et al.*[1] but the reformulation given below throws more light on the problem.

II. THE GAUSSIAN APPROXIMATION

From (10) it is seen that for the intermediate scattering function $F_s(\kappa,t)$ to be of the form $\exp[-\kappa^2\gamma(t)]$ one has two conditions:

$$[x(0), x(-t)] \text{ is a } c \text{ number},$$
$$\langle\exp[i\kappa X(t)]\rangle \text{ is expressible as } \exp[-\kappa^2\langle X^2(t)\rangle/2]. \quad (16)$$

Thus, in the Gaussian approximation the displacement part of $F_s(\kappa,t)$ is clearly the transform of a Gaussian-shaped $M_s(\mathbf{x},t)$; the recoil part is $\exp\{-[x(0), x(-t)]\kappa^2/2\}$ and does not need averaging since $[\ ]$ is a c number.

The displacement part gives rise to an unambiguous, real, width function $\gamma_D(t)$ say[5]; we define $2\gamma_D(t)$ as the mean square displacement $\langle X^2(t)\rangle$; it is the width of the Gaussian $M_s(x,t)$.

[5] The suffixes D and R are used for describing the connection with the displacement and the recoil part, respectively.

The explicit expressions for $\gamma_D(t)$ in the three cases cited above can be easily written down. In all cases it is a real function, with value 0 at $t=0$ and starting off as t^2 for short times. Notice that a width function defined naturally and generally through $\exp[i\kappa X(t)]$ does *not* have a nonzero value at $t=0$.

The central problem is to take care of the recoil part of $F_s(\kappa,t)$ while making a classical approximation for the displacement part.

This can be done for systems in thermal equilibrium by use of the fluctuation-dissipation theorem.

We first write

$$F_s(\kappa,t)=e^{-\kappa^2\gamma_D(t)}e^{-i\kappa^2\gamma_R(t)}. \tag{17}$$

$\gamma_D(t)$ has already been defined; it is a real quantity and so is $\gamma_R(t)$.

$$\begin{aligned}\gamma_R(t)&=(1/2i)[x(0),x(-t)],\\ \gamma_D(t)&=\tfrac{1}{2}\langle X^2(t)\rangle.\end{aligned} \tag{18}$$

It is easy to see that

$$\begin{aligned}\ddot{\gamma}_R&=(1/2i)[v(0),v(t)],\\ \ddot{\gamma}_D&=\tfrac{1}{2}\langle v(0)v(t)+v(t)v(0)\rangle,\\ \ddot{\gamma}_D+i\ddot{\gamma}_R&=\langle v(0)v(t)\rangle.\end{aligned} \tag{19}$$

Now one can use the fluctuation-dissipation theorem and get the well-known relation, with $\beta=\hbar/2k_BT$ and $D\equiv d/dt$,

$$\ddot{\gamma}_R=-(\tan\beta D)\ddot{\gamma}_D. \tag{20}$$

Hence,

$$\gamma_R=-(\tan\beta D)\gamma_D. \tag{21}$$

The constants of integration can be shown to vanish.[6] (21), in fact, directly follows from the requirement that $\gamma(t+i\beta)$ is real.

Hence, in the Gaussian approximation

$$\begin{aligned}F_s(\kappa,t)&=\exp[-\kappa^2\gamma(t)],\\ \gamma(t)&=[1-i(\tan\beta D)]\times\tfrac{1}{2}\langle X^2(t)\rangle.\end{aligned} \tag{22}$$

The last equation can be written alternatively as

$$\gamma(t)=\exp(-i\beta D)\{\sec(\beta D)\times\tfrac{1}{2}\langle X^2(t)\rangle\}. \tag{23}$$

Thus, $\rho(t)$, defined as $\gamma(t+i\beta)$ is given by

$$\rho(t)=\sec(\beta D)\times\tfrac{1}{2}\langle X^2(t)\rangle. \tag{24}$$

[6] The proof involves the fact that correlations like $\langle v(0),x(t)\rangle$ also obey the relation: imaginary part $=-\tan\beta D$ (real part).

It is easy to see that this corresponds exactly to the equation for $\gamma(t+i\beta)$ given by Rahman *et al.*,[1] which is

$$\rho(0)+\int_0^t(t-u)(\sec\beta D)\,\mathrm{Re}\langle v(0)v(u)\rangle du. \tag{25}$$

Expressions (24) and (25) can be shown to be equivalent. The fact that the scattering cross section can be expressed so simply in terms of the mean square displacement is masked by the form (25) in which $\rho(t)$ was given. A prescription to take care of the recoil terms while calculating $\langle X^2(t)\rangle$ classically now appears from (24).

We have, using (24) and expanding $\sec(\beta D)$ in powers of βD,

$$\begin{aligned}F_s(\kappa,t+i\beta)&=\exp[-\kappa^2\rho(t)]\\ &=\exp[-\kappa^2\gamma_D-(ME_R/4k_B^2T^2)\ddot{\gamma}_D\\ &\qquad+E_R\times O(\hbar^2)+\cdots].\end{aligned} \tag{26}$$

Now using classical values for γ_D and $\ddot{\gamma}_D$ one gets

$$\begin{aligned}F_s(\kappa,t+i\beta)\simeq\exp(&-\kappa^2\langle X^2(t)\rangle_{\mathrm{cl}}/2\\ &-ME_R\langle v(0)v(t)\rangle_{\mathrm{cl}}/4k_B^2T^2).\end{aligned} \tag{27}$$

To get this classical $F_s(\kappa,t+i\beta)$ one should, thus, take

$$\begin{aligned}\rho(t)=&\int_0^t(t-u)\langle v(0)v(u)\rangle_{\mathrm{cl}}du\\ &+(\hbar^2/8k_B^2T^2)\langle v(0)v(t)\rangle_{\mathrm{cl}}.\end{aligned} \tag{28}$$

The second term in (28) contains a time dependence, through $\langle v(0)v(t)\rangle$, and this time dependence is usually neglected by putting $\langle v^2\rangle$ instead. It is easy to see that this term is anyhow of *no* practical importance in the interpretation of the experimental data available on slow neutrons scattered from liquids. Prescription (26) or (27) for using the classical velocity correlation is now seen to be equivalent to calculating the displacement part of the intermediate scattering function classically and then using it to take care of the recoil part.

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