evaluated by using the matrix elements given by Nilsson.

It is now known that for large deformations, positive quadrupole moments tend to predominate.[25] It is likely, therefore, that the sign of $\mu(Br^{76})$ is negative. Of the two configurations exhibiting a negative moment in Table IV, the configuration $(pn)=(p_{3/2}^-, f_{5/2}^-)$ seems most likely. A measurement of the magnetic moment of Se^{75} would shed light on this matter.

[25] B. R. Mottelson and S. G. Nilsson, Danske Videnskab. Selskab, Mat.-fys. Skrifter **1**, No. 8 (1958).

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Effects of Chemical Binding on Nuclear Recoil*

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The recoil of a chemically bound nucleus is considered for slow neutron scattering and for the resonant absorption of neutrons or gamma rays. The Doppler-broadened resonance line shape is derived in terms of the time-dependent self-correlation function describing the motion of a nucleus due to the interatomic forces. This explicitly relates the resonance line shape to the differential scattering cross section for slow neutrons in the Fermi pseudopotential approximation. Within this formulation an expansion for large nuclear recoil is naturally suggested. For the case of a crystal, this expansion can be directly related to the expansion associated with the central limit theorem of probability theory and can therefore be proved to be asymptotic in nature. The expansion parameter is $(K_{av}/R)^{\frac{1}{2}}$, where K_{av} is the average kinetic energy of a nucleus and R is the recoil energy for a free nucleus at rest. The leading term of the expansion is the weak binding limit originally obtained by Lamb. In this limit the Doppler-broadened line shape is the same as would obtain for an ideal monatomic gas of the same mass with an effective temperature $T'=(\frac{2}{3})K_{av}$. For noncrystalline systems, a similar expansion with the same leading term can be obtained by a rearrangement of the terms in an expansion used by Wick to study the slow neutron total cross section. The relation of the present expansion to Wick's expansion is discussed.

I. INTRODUCTION

FOR many nuclear processes a quantitative description requires the inclusion of the effects of chemical binding on the nuclear recoil. These processes include the scattering of neutrons with energies less than a few electron volts, the Doppler-broadening of neutron-absorption resonances, and the emission and resonant reabsorption of nuclear gamma rays. These processes can all be described in terms of the same formalism because the perturbation on the atomic system due to the nuclear interaction is accurately given in terms of the matrix element of a point interaction. For the neutron-scattering problem, this involves the familiar Fermi pseudopotential[1] approximation, the application of which has been extensively developed in the literature.[2–6] For the Doppler-broadening of neutron absorption resonances, the original derivation given by Lamb[7] contains the essential physical arguments, but is given only for crystalline materials. This derivation has been applied by Visscher to the study of the emission and resonant reabsorption of gamma rays[8] and neutrinos[9] in crystals. In Sec. II, we extend Lamb's derivation to arbitrary materials and show that the result can be expressed in terms of the same time-dependent self-correlation function that determines the incoherent slow neutron scattering.[4–6]

The explicit relationship between the resonance line shape and the slow neutron-scattering amplitude enables the application to the former problem of the formalism which has been extensively developed for the latter. In many cases the recoil momentum is sufficiently large that the details of the binding potential are not expected to be important. In this large recoil limit, the binding appears only insofar as it determines the average kinetic energy of the nucleus before the nuclear interaction. This limit was first studied by Lamb[7] in connection with the resonance line shape. More recently,

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[1] E. Fermi, Ricerca sci. **1**, 13 (1936).
[2] G. Placzek, Phys. Rev. **86**, 377 (1952).
[3] G. Placzek and L. Van Hove, Phys. Rev. **93**, 1207 (1954).
[4] G. C. Wick, Phys. Rev. **94**, 1228 (1954).
[5] A. C. Zemach and R. J. Glauber, Phys. Rev. **101**, 118, 129 (1956).
[6] L. Van Hove, Phys. Rev. **95**, 249 (1954).
[7] W. E. Lamb, Phys. Rev. **55**, 190 (1939).
[8] W. M. Visscher Ann. Phys. **9**, 194 (1960).
[9] W. M. Visscher, Phys. Rev. **116**, 1581 (1959).

Placzek[2] and Wick[4] have considered the large recoil limit for the slow neutron-scattering cross section by introducing expansions in powers of the effective collision time. They showed that the leading correction to the free-atom cross section at high energies is determined by the Doppler effect associated with the average kinetic energy of the bound nucleus. They also derived higher-order corrections that depend explicitly on the binding potential.

The expansions introduced by Placzek and Wick are not suitable, however, for the calculation of differential quantities, such as the energy-transfer cross section for slow neutrons, or the Doppler-broadened line shape for resonant absorption. An appropriate expansion for the calculation of these quantities is obtained in the present paper by a rearrangement of the terms in Wick's expansion. The properties of this new expansion are not easily studied for an arbitrary system. It is therefore useful to explicitly consider a system for which the appropriate time-dependent correlation function is exactly calculable.

We consider, in Sec. III, the case of a crystal with harmonic lattice vibrations. For this case the exact form of the appropriate time-dependent correlation function can be derived.[5,10] The nature of the final result is such that we can obtain an asymptotic expansion in inverse powers of the recoil momentum by direct application of the central limit theorem of probability theory.[11] The leading term in the expansion is the result that would apply for an ideal monatomic gas of unchanged mass, but with an effective temperature $T'=(\frac{2}{3})K_{\mathrm{av}}$, where K_{av} is the average kinetic energy per particle. This term is the "weak binding" limit obtained by Lamb. The higher terms in the expansion involve the same parameters of the lattice vibrations that appear in the work of Placzek[2] and of Wick.[4]

An alternative approach that is applicable for crystals is to first expand in terms of the number of phonons exchanged in a single interaction. The terms corresponding to zero or to one phonon interchange can be treated exactly and can yield detailed information about the distribution of lattice vibration frequencies in the crystal.[3,8] The central limit theorem can then be applied to the individual terms in the phonon expansion corresponding to the exchange of two or more phonons. In a paper by Sjölander,[12] this approach has been carried through in detail for the scattering of neutrons by crystals.

In Sec. IV we consider the development of an expansion for large momentum transfer applicable to noncrystalline materials. The expansion is obtained by making the same rearrangement of the terms in Wick's expansion that gives the result in Sec. III for the crystalline case. For a noncrystalline system, the expansion no longer gives an asymptotic expansion in inverse powers of the recoil momentum, but it continues to have the property that the leading terms of the expansion give the dominant contributions to the first few moments of the scattered energy distribution or of the absorption line shape. This feature makes the present method of interest in connection with such complicated phenomena as the scattering of slow neutrons by liquids. The present method is closely related to the moments method considered by de Gennes[13] in connection with the above-mentioned problem.

Finally, in Sec. V we discuss some applications of the techniques developed in the preceding sections. Numerical results for the Doppler-broadening of neutron absorption resonances are presented, as well as a discussion which makes it plausible that the weak binding limit of Lamb will be quite accurate for almost all cases of interest. The relevant formulas for the emission and resonant reabsorption of nuclear gamma rays are presented and discussed, but no numerical results are given. A discussion is given of the application of the present method to slow-neutron scattering problems. The relation of the present expansion to Wick's expansion for the total scattering cross section is discussed in Appendix A.

II. DERIVATION OF RESONANCE LINE SHAPE

We consider the resonant absorption of a neutron by a nucleus bound in an atomic system. The resonance line shape (normalized to unit area) summed over final atomic states and averaged over initial atomic states is given by[7,14]

$$W(E)=\frac{\Gamma}{2\pi}\sum_{b,a}\frac{\rho(a)|(b|\exp(i\mathbf{p}\cdot\mathbf{r})|a)|^2}{(E-E_0-\epsilon_b+\epsilon_a)^2+\frac{1}{4}\Gamma^2}, \qquad (1)$$

where E is the energy of the incident neutron or gamma ray; E_0 is the resonance energy; $\mathbf{p}$ is the recoil momentum to be taken up by the atomic system; $\mathbf{r}$ is the position coordinate of the nucleus; and $\rho(a)$ is the distribution of initial atomic states, $|a)$, with energy ϵ_a. The sum is over intermediate states, $|b)$, with energy ϵ_b. The sum over final atomic states has already been carried out by closure, and the dependence of the natural width, Γ, on the atomic state has been neglected. An identical expression applies to the resonant absorption of a gamma ray.[8]

In order to relate the resonance line shape to the neutron-scattering amplitude for the same atomic system, we introduce[7]

$$\delta(\omega-\epsilon_b+\epsilon_a)=\frac{1}{2\pi}\int_{-\infty}^{\infty}dt\,\exp[-it(\omega-\epsilon_b+\epsilon_a)], \qquad (2)$$

[10] F. Bloch, Z. Physik **74**, 295 (1932).
[11] H. Cramér, *Mathematical Methods of Statistics* (Princeton University Press, Princeton, New Jersey, 1946).
[12] A. Sjölander, Arkiv Fysik **14**, 315 (1958).
[13] P. G. de Gennes, Physica **25**, 825 (1959).
[14] A system of units will be used in which $\hbar=1$, and temperatures are given in energy units.

and rewrite (1) in the form

$$W(E)=\frac{\Gamma}{2\pi}\int_{-\infty}^{\infty}d\omega\, S(\mathbf{p},\omega)[(E-E_0-\omega)^2+\tfrac{1}{4}\Gamma^2]^{-1}. \quad (3)$$

The dependence of the line shape on the atomic system is contained in the function

$$S(\mathbf{p},\omega)=(2\pi)^{-1}\int_{-\infty}^{\infty}dt\, e^{-it\omega}\chi(\mathbf{p},t), \quad (4)$$

where

$$\chi(\mathbf{p},t)=\sum_{b,a}\rho(a)|(b|e^{i\mathbf{p}\cdot\mathbf{r}}|a)|^2e^{it(\epsilon_b-\epsilon_a)}. \quad (5)$$

We note that $S(\mathbf{p},\omega)$ is precisely the same function[6] that determines the noninterference slow neutron scattering in the Fermi pseudopotential approximation. The scattering of a neutron of energy E to energy E' through angle θ is given by

$$\sigma(E,E',\theta)=(\sigma_b/4\pi)(E'/E)^{\frac{1}{2}}S(\mathbf{K},E-E'), \quad (6)$$

where the momentum transfer K if given by $(K^2/2m)=[E+E'-2(EE')^{\frac{1}{2}}\cos\theta]$, and σ_b is the bound-atom cross section.

The formalism that has been developed in the theory of slow neutron scattering[4–6] can therefore be applied to the calculation of the resonance line shape. In this formalism it is usually more convenient to work with the function $\chi(\mathbf{p},t)$, which is related to the line shape through

$$W(E)=(2\pi)^{-1}\int_{-\infty}^{\infty}dt\,\exp[i(E_0-E)t-\tfrac{1}{2}\Gamma|t|]\chi(\mathbf{p},t). \quad (7)$$

At this point, we note the relationship of our notation to that of other authors: $S(\mathbf{p},\omega)$ is the same function introduced by Van Hove,[6] and $\chi(\mathbf{p},t)$ is the spatial Fourier transform of Van Hove's time-dependent self-correlation function $G_s(\mathbf{r},t)$. It is equal to $\chi(\mathbf{p},-t)$ as defined by Zemach and Glauber,[5] which is equal to $(4\pi/\sigma_b)$ times the function $G(\mathbf{p},t)$ defined by Wick.[4]

By the introduction of the Hamiltonian, H, for the atomic system,[4,5] Eq. (5) for $\chi(\mathbf{p},t)$ can be rewritten in the convenient condensed form

$$\chi(\mathbf{p},t)=\sum_a\rho(a)(a|e^{-i\mathbf{p}\cdot\mathbf{r}}e^{iHt}e^{i\mathbf{p}\cdot\mathbf{r}}e^{-iHt}|a) \quad (8a)$$

$$=\sum_a\rho(a)(a|\exp[-i\mathbf{p}\cdot\mathbf{r}(0)]\exp[i\mathbf{p}\cdot\mathbf{r}(t)]|a). \quad (8b)$$

In general, $\chi(\mathbf{p},t)$ will depend on the relative orientation of the incident neutron or gamma ray and the molecular or crystalline axes. For a randomly oriented assembly of molecules or crystallites, the average of χ over orientation will depend only on the magnitude p of the recoil momentum.

For an ideal monatomic gas of mass M and temperature T, the use of (8a) enables a very simple derivation of[5]

$$\chi(p,t)=\exp[(p^2/2M)(it-Tt^2)], \quad (9)$$

and

$$S(p,\omega)=(M/2\pi Tp^2)^{\frac{1}{2}}\times\exp[-(M/2Tp^2)(\omega-p^2/2M)^2]. \quad (10)$$

Substitution of (9) into (7) or (10) into (3) gives the familiar Bethe-Placzek[15] form for the Doppler-broadened resonance line shape.

In general, exact analytic expression for $\chi(\mathbf{p},t)$ cannot be given except in the operator form of Eq. (8). From (8b) it appears plausible that an expansion of $S(\mathbf{p},\omega)$ for large p can be obtained from the behavior of $\chi(\mathbf{p},t)$ for small t. In the special case that $\mathbf{r}(t)$ can be written as a sum of harmonic oscillator coordinates, $\chi(\mathbf{p},t)$ can be exactly determined, and the behavior for large recoil explicitly studied. In the next section we examine the application to the harmonic lattice vibrations of a solid. A related expansion is developed for arbitrary systems in Sec. IV.

III. ASYMPTOTIC EXPANSION FOR LATTICE VIBRATIONS

In the approximation of an infinite phonon lifetime the lattice vibrations of a solid can be represented by an $\mathbf{r}(t)$ which is a sum of simple harmonic oscillator coordinates. By the use of standard field theoretical techniques plus a theorem due to Bloch[10] for the averaging over a thermal equilibrium distribution of initial states, an exact expression for $\chi(\mathbf{p},t)$ can be derived[5,6] for this case. This expression will, in general, depend on the orientation of the incident neutron or gamma ray with respect to the crystal axes. For randomly oriented polycrystalline material, an average over this orientation must be performed. In this paper we will consider only the case of a Bravais lattice, for which $\chi(\mathbf{p},t)$ is independent of orientation, and is given by

$$\chi(p,t)=\exp\{R[\gamma(t)-\gamma(0)]\}, \quad (11)$$

where

$$R=(p^2/2M), \quad (12)$$

and

$$\gamma(t)=\int_0^{\omega_m}[\coth(\zeta/2T)\cos\zeta t+i\sin\zeta t]f(\zeta)\zeta^{-1}d\zeta, \quad (13)$$

in which M is the nuclear mass, ω_m is the maximum vibrational frequency, and $f(\zeta)$ is the distribution of vibrational frequencies normalized to unity. The familiar expansion in the number of phonons exchanged in a single interaction is obtained by extracting the Debye-Waller factor, $\exp[-R\gamma(0)]$, and expanding the remaining factor in powers of $R\gamma(t)$. The first few terms in this expansion are sensitive to the details of the frequency distribution. In a range where $R\gamma(0)$ is of order unity, or smaller, these terms will dominate, and

[15] H. Bethe and G. Placzek, Phys. Rev. **51**, 462 (1937).

detailed information about the frequency distribution of the lattice vibrations can be obtained from the experiment. This has been discussed in some detail for the case of neutron scattering by Placzek and Van Hove,[3] and for the case of gamma-ray absorption by Visscher.[8] It has been shown by Sjölander[12] that the terms corresponding to emission of two or more phonons can be accurately calculated by means of an expansion based on the central limit theorem of probability theory.[11] This simplification of the multiphonon terms greatly simplifies the analysis of experiments in which multiphonon corrections are significant but not dominant.

In the present paper we apply the central limit theorem to obtain an asymptotic expansion for large recoil without first making the expansion in the number of phonons exchanged. The resulting expansion is useful only when the multiphonon terms are dominant, so that the details of the frequency distribution are not important. We expect that for large values of R, the Fourier transform of $\chi(\mathbf{p},t)$ will be determined by the behavior of $\chi(\mathbf{p},t)$ for small values of t. Expanding $\gamma(t)$ about $t=0$, we obtain

$$\gamma(t)-\gamma(0)=\sum_{\nu=1}^{\infty}\frac{x_\nu}{\nu!}(it)^\nu, \tag{14}$$

where

$$x_{2\nu-1}=\int_0^{\omega_m}\zeta^{2\nu-2}f(\zeta)d\zeta, \tag{15}$$

and

$$x_{2\nu}=\int_0^{\omega_m}\zeta^{2\nu-1}\coth(\zeta/2T)f(\zeta)d\zeta. \tag{16}$$

In particular,

$$x_1=1,$$
$$x_2=(4/3)K_{\mathrm{av}}\equiv 2T', \tag{17}$$

where K_{av} is the average kinetic energy per particle associated with the lattice vibrations. In the limit that ω_m goes to zero, T' approaches T and the terms in (14) for $\nu\geq 3$ vanish, giving the proper ideal gas limit (9) for $\chi(p,t)$.

In order to develop an expansion valid for large values of R, we note that the Fourier transform of (11) is formally identical to the probability distribution of the sum of (R/T') identically distributed random variables. The central limit theorem of probability theory tells us that this distribution approaches a Gaussian with corrections of order $R^{-\frac{1}{2}}$. A systematic procedure exists for obtaining an expansion in powers of $R^{-\frac{1}{2}}$, which can be proved to be asymptotic in nature.[11] Following Cramer, we write

$$\chi(p,t)=\exp[R(it-T't^2)]\exp\left[R\sum_{\nu=3}^{\infty}\frac{x_\nu}{\nu!}(it)^\nu\right], \tag{18}$$

and then expand the second exponential in (18) in powers of (it). In order to group terms according to powers of $R^{-\frac{1}{2}}$, we introduce the dimensionless variable $z^2=2RT't^2$. The values of z which contribute to the Fourier transform of $\chi(\mathbf{p},t)$ will be of order unity. The Fourier transform of each term in the expansion can easily be carried out by noting that

$$\int_{-\infty}^{\infty}e^{-izx}(iz)^\nu\exp(-z^2/2)dz = (2\pi)^{\frac{1}{2}}H_\nu(x)\exp(-x^2/2), \tag{19}$$

where

$$H_n(x)=(-1)^n\exp(x^2/2)\frac{d^n}{dx^n}[\exp(-x^2/2)] \tag{20}$$

is the nth Hermite polynomial.

The resulting expression for $S(p,\omega)$ is

$$S(p,\omega)=(\pi\Delta^2)^{-\frac{1}{2}}\exp(-\tfrac{1}{2}\epsilon^2)\left\{1+\frac{1}{12\sqrt{2}}\left(\frac{T'}{R}\right)^{\frac{1}{2}}\frac{x_3}{T'^2}H_3(\epsilon)+\frac{T'}{R}\left[\frac{1}{96}\frac{x_4}{T'^3}H_4(\epsilon)+\frac{1}{576}\left(\frac{x_3}{T'^2}\right)^2H_6(\epsilon)\right]+O((T'/R)^{\frac{3}{2}})\right\}, \tag{21}$$

where

$$\Delta=2(RT')^{\frac{1}{2}} \tag{22}$$

is the Doppler width, and

$$\epsilon=\sqrt{2}(\omega-R)/\Delta. \tag{23}$$

The leading term in (21) is the result for an ideal gas of mass M and temperature T'. The "weak binding" limit first obtained by Lamb[7] corresponds to retaining only this term. The correction terms are seen to vanish both in the limit of large recoil and in the limit of weak binding for arbitrary recoil. Since the expansion is known to be asymptotic, the error in truncating at any term can be estimated. The higher order terms in (21) can be computed in a straightforward manner for any assumed frequency distribution.

For arbitrary crystal structure, we must replace (11) by

$$\chi(\mathbf{p},t)=\exp\{R\mathbf{u}\cdot[\mathfrak{M}(t)-\mathfrak{M}(0)]\cdot\mathbf{u}\}, \tag{24}$$

where $\mathbf{u}$ is a unit vector in the direction of $\mathbf{p}$, and $\mathfrak{M}(t)$ is a tensor depending on the orientation of the incident neutron with respect to the crystal axes. The expansion corresponding to (21) can still be carried out, but T', x_3, x_4, etc., will depend on orientation. An average over orientation of such an expression is cumbersome. In the next section we discuss a closely related expansion which can be applied to noncrystalline systems and which gives a fairly simple result for crystalline systems of complicated structure.

IV. APPROXIMATION FOR ARBITRARY SYSTEMS

We would like to develop an expansion of the form (21) for arbitrary systems. Since $\chi(\mathbf{p},t)$ does not in general depend exponentially on R, the expansion will

not yield an asymptotic series in powers of $R^{-\frac{1}{2}}$. We consider first the expansion developed by Wick[4] for the average of $\chi(\mathbf{p},t)$ over orientations.

$$\chi(R,t)=\langle\chi(\mathbf{p},t)\rangle=e^{iRt}\sum_{n=0}^{\infty}\frac{s_n}{n!}(it)^n. \tag{25}$$

The quantities s_n can be related[4] to parameters introduced by Placzek[2]:

$$s_0=1,\quad s_1=0,\quad s_2=(\tfrac{4}{3})RK_{\rm av},\quad s_3=RB_{\rm av},$$
$$s_4=(16/5)R^2(K^2)_{\rm av}+2RC_{\rm av},\ \cdots,$$

where $K_{\rm av}$ and $(K^2)_{\rm av}$ are the mean and mean square kinetic energy of the nucleus averaged over initial states. $B_{\rm av}$ and $C_{\rm av}$ depend explicitly on the binding potential. For the case of a crystal, $B_{\rm av}=x_3$ and $C_{\rm av}=(\frac{1}{2})x_4$.

The expansion (25) was applied by Wick to the calculation of the total scattering cross section for slow neutrons as an expansion in inverse powers of the neutron energy. Wick's expansion is not suitable for the calculation of differential quantities such as the resonance line shape because the resulting expansion of $S(R,\omega)$ is in terms of singular functions. The expansion of the preceding section was in terms of nonsingular functions because of the appearance of the Doppler term $(-RT't^2)$, as the argument of an exponential. A natural generalization of our result for crystalline systems can be obtained by multiplying the right-hand side of (25) by

$$\exp(-\tfrac{1}{2}s_2t^2)\sum_{n=0}^{\infty}\frac{1}{n!}(\tfrac{1}{2}s_2t^2)^n$$

to give

$$\chi(R,t)=\exp[R(it-T't^2)]\left(1+\sum_{n=3}^{\infty}\frac{r_n}{n!}(it)^n\right), \tag{26}$$

where

$$T'=\tfrac{2}{3}K_{\rm av},\quad r_3=s_3=RB_{\rm av},$$
$$r_4=s_4-3s_2^2=2RC_{\rm av}+(16/5)[(K^2)_{\rm av}-(5/3)(K_{\rm av})^2]R^2. \tag{27}$$

The corresponding expansion for $S(R,\omega)$ is

$$S(R,\omega)=(\pi\Delta^2)^{-\frac{1}{2}}\exp(-\tfrac{1}{2}\epsilon^2)\times\left\{1+\frac{1}{12\sqrt{2}}\left(\frac{T'}{R}\right)^{\frac{1}{2}}\left(\frac{B_{\rm av}}{T'^2}\right)H_3(\epsilon)+\frac{1}{48}\left(\frac{T'}{R}\right)\left(\frac{C_{\rm av}}{T'^3}\right)H_4(\epsilon)+\frac{1}{8}\left[\frac{4}{15}\frac{(K^2)_{\rm av}}{T'^2}-1\right]H_4(\epsilon)+\cdots\right\}, \tag{28}$$

where Δ and ϵ are defined by (22) and (23).

Comparing (21) and (28) through terms containing $H_4(\epsilon)$, we see that (28) contains a term in $H_4(\epsilon)$ which is of order unity. The coefficient of this term vanishes for a Bravais lattice and for an arbitrary crystal structure for a fixed orientation. There will be similar terms of order unity for all the even Hermite polynomials in (28) and terms of order $R^{-\frac{1}{2}}$ for all the odd Hermite polynomials. It is therefore clear that (28) is not an asymptotic expansion in powers of $R^{-\frac{1}{2}}$ for a general system. For many quantities of physical interest, however, the contributions arising from Hermite polynomials of high order will be small, so the expansion (28) is useful. This is best illustrated by the specific applications considered in the next section and in Appendix A.

V. APPLICATIONS

Doppler-Broadening of Neutron Absorption Resonances

The present expansion can be applied with greatest accuracy to the line shape for a neutron absorption resonance in a crystal. This is true because of the appreciable magnitude of the natural width of the resonance compared with the Doppler width. The line shape in the wings of the resonance is determined primarily by the natural width. It is in the wings, however, that the higher Hermite polynomial terms would be important. Near the center of the line the Doppler-broadening has its greatest effect and is likely to be given accurately by the weak binding limit of Lamb, which constitutes the leading term of the present expansion. In addition, the natural width is roughly independent of incident neutron energy for a purely absorbing resonance, whereas the Doppler width increases with increasing energy. The resonances for which the Doppler-broadening is largest will therefore correspond to large recoil and consequently to small coefficients of the Hermite polynomial correction terms.

We will consider a numerical example. The line shape is given by a convolution (3) of (21) with the natural line shape. The resulting expression, which has been programmed for computation with a digital computer,[16] is

$$W(E)=(\Gamma/2\pi)\phi(\xi,x), \tag{29}$$

where

$$\xi=(\Gamma/\Delta),\quad x=(2/\Gamma)(E-E_0-R),$$

Δ is given by (22), and

$$\phi(\xi,x)=\phi_0(\xi,x)+\sum_{n=3}A_n\phi_n(\xi,x). \tag{30}$$

The functions $\phi_n(\xi,x)$ are defined by

$$\phi_n(\xi,x)=(\xi/2\pi^{\frac{1}{2}})\int_{-\infty}^{\infty}dy\,(1+y^2)^{-1}\times\exp\left\{-\frac{\xi^2}{4}(x-y)^2\right\}H_n\left(\frac{\xi(x-y)}{\sqrt{2}}\right), \tag{31}$$

and $\phi_0(\xi,x)$ is the usual Bethe-Placzek line shape $\psi(\xi,x)$.

[16] G. Kuncir (private communication).

TABLE I. Peak cross section (arbitrary units) as a function of temperature for the 1.26-ev resonance in Rh^{103}.

T(°K)	T'(°K)	Peak cross section: Free atom	Lamb's approximation	Present approximation
0	142	1.000	0.957	0.959
75	153	0.976	0.954	0.956
150	198	0.954	0.942	0.945
300	324	0.918	0.913	0.917
600	612	0.861	0.860	0.864

The coefficients, A_n, for a Debye crystal are conveniently written by defining the functions

$$F(\Theta/2T)=(T'/T), \tag{32}$$

and

$$H(\Theta/2T)=(5C_{\rm av}/3T\Theta^2), \tag{33}$$

where Θ is the Debye temperature. In terms of these functions,

$$\begin{aligned} A_3&=\frac{1}{20\sqrt{2}}\frac{\Theta^2}{T^{\frac{3}{2}}R^{\frac{1}{2}}F^{\frac{3}{2}}},\\ A_4&=\frac{1}{80}\frac{\Theta^2 H}{RTF^2},\\ A_5&=\frac{1}{1120\sqrt{2}}\frac{\Theta^4}{R^{\frac{3}{2}}T^{\frac{5}{2}}F^{\frac{5}{2}}},\\ A_6&=\frac{1}{1600}\frac{\Theta^4}{T^3RF^3}. \end{aligned} \tag{34}$$

The functions F and H both go to one for high temperature. Their detailed properties are discussed in Appendix B. As a numerical example, we will consider the 1.26-ev resonance in rhodium. The Doppler-broadening of this resonance as a function of temperature has been studied experimentally by Landon.[17] The low energy of this resonance makes the inherent convergence of the Hermite polynomial expansion rather poor. This is compensated, however, by the small ratio of Doppler width to natural width, so that the effects on the line shape due to crystalline binding are rather small. The most sensitive parameter is the peak cross section. In Table I we give the peak cross section as a function of temperature. A Debye temperature of 378°K has been assumed.

We note that the dominant corrections are given by Lamb's result even in this low recoil case. A more interesting comparison, considering the improved resolution of today's neutron spectrometers, would be for the detailed line shape of a more severely Doppler-broadened resonance. The authors welcome suggestions from experimentalists measuring neutron resonance parameters for specific cases to calculate.

[17] H. Landon, Phys. Rev. **94**, 1215 (1954).

Emission and Absorption of Gamma Rays

The line shape for resonant emission or absorption of gamma rays can be much more sensitive to the details of chemical binding than for the case of neutron absorption, since the natural width of the gamma-ray line is negligible compared with the Doppler width. The recoil energy R is given by $E_0^2/2Mc^2$, which is small (compared to T) for a 10-kev gamma ray, and large for a 300-kev gamma ray. For sufficiently small R, the possibility exists for studying the details of the frequency distribution of lattice vibrations in a crystal.[8] The relevant experiment will be to measure the nuclear reabsorption of a gamma ray as a function of the Doppler shift of the emitted gamma ray with respect to the absorber.

Consider the emitter to be moving at a velocity v toward the absorber, and thus Doppler shifting the energy of the emitted gamma ray by an amount $s=(v/c)E_0$. The probability of resonant reabsorption as a function of s is given by

$$\begin{aligned} W'(s)&=\int W_a(E)W_e(E-s)dE\\ &=(2\pi)^{-1}\int_{-\infty}^{\infty}dt\,\exp[-\Gamma|t|-ist]\\ &\qquad\times\chi_a(R,t)\chi_e(R,t), \end{aligned} \tag{35}$$

where the subscripts a and e on the χ functions refer to the state of chemical binding of absorber and emitter, respectively.

For small R and for a crystalline system, the appropriate procedure is to expand χ_a and χ_e in terms of the number of phonons exchanged.[8] For large R, the average number of phonons emitted will be large and the appropriate expansion is that of (26). Carrying out this expansion for each of the functions in (35), we obtain a line shape $W'(s)$ of the form (3) with

$$(E_0-E)\rightarrow s \quad \Gamma\rightarrow 2\Gamma,$$

and $S(R,\omega)$ given by an expression of the form (28) with

$$\epsilon=(\omega-2R)\sqrt{2}/\Delta_0.$$

The parameters T', $B_{\rm av}$, $C_{\rm av}$, and $(K^2)_{\rm av}$ in (28) are to be replaced by the sum of the values for the emitter and the absorber. The appropriate Doppler width becomes $\Delta_0=2[R(T_e'+T_a')]^{\frac{1}{2}}$. Terms involving Hermite polynomials of the sixth and higher orders will have coefficients containing products of emitter and absorber parameters.

The above results are of interest in the context of Visscher's suggestion that gamma rays can be used to study effects of chemical binding on the nuclear motion.[8] A particularly interesting case is that of liquids; the physics is essentially the same as for slow neutron inelastic scattering, as discussed in the following.

Slow Neutron Scattering

The essential feature of the expansion (28) is that it characterizes the differential energy-transfer cross section (6) in terms of a few parameters of the nuclear motion in the interatomic force field. This characterization is not of great interest for a system of known dynamical properties, such as a crystal, because more detailed information on the lattice vibrations can be obtained[3] by using appropriately chosen experimental conditions. The expansion (21) is of some practical interest in the crystalline case, however, since it is useful in describing neutron moderation by a crystal for neutron emergies greater than the Debye temperature. For the neutron moderation problem, we do not expect the details of the lattice vibrations to be of great importance.

For a system of poorly known dynamical properties, the expansion (28) is of somewhat greater interest since it defines in an approximate way what parameters of the dynamics of the atomic motion are likely to be determined from slow neutron inelastic scattering. The developments of the present work have been limited to the case of incoherent scattering, but they already show some interesting features.

It is instructive to compare the present results with the work of de Gennes.[13] He considered the calculation of the second and fourth energy-transfer moments of $S(\mathbf{K}, E'-E)$ for fixed momentum transfer $\mathbf{K}$. De Gennes' considerations were restricted to a classical liquid in the limit of large mass where recoil can be neglected. He considered this case for coherent scattering, as well as incoherent scattering, but we will be concerned only with the latter. De Gennes found that the second moment of $S(\mathbf{K}, E'-E)$ is given by $2RT$, and the fourth moment by $[12R^2T^2+2RC_{\rm av}]$. It is interesting to note the differences in the present treatment in which quantum mechanical effects and recoil are included.

First, we note that the parameters of the atomic motion appear directly in an expansion for the differential energy-transfer cross section so that the restriction to fixed momentum transfer at a given scattering angle is not present. This restriction appears only in the interpretation in terms of moments and is avoided by the Hermite polynomial expansion. Second, the free-atom recoil is explicitly included in the definition of ϵ in (28). Third, the temperature T is replaced by T'; a correction due to quantum mechanical zero-point motion. The third moment term in $B_{\rm av}$ also arises from zero-point motion. Finally, we have an additional fourth moment term involving $(K^2)_{\rm av}$. This term vanishes for a classical system in thermal equilibrium. Its order of magnitude for a real liquids is hard to estimate.

SUMMARY

We have considered three applications of our extension of Wick's short-collision-time method[4] for calculating slow neutron scattering. For the Doppler-broadening of neutron absorption resonances, the essential result is a justification of an earlier approximation introduced by Lamb.[7] For the scattering of slow neutrons or for the resonant emission and reabsorption of nuclear gamma rays, the method is applicable in situations where the recoil is sufficiently large that the effects of chemical binding can be given in terms of a few parameters. The determination of these parameters is of particular interest for liquids where the dynamics of nuclear motion is poorly known. The method presented here may be useful in analyzing experiments on slow neutron inelastic scattering or the broadening of low-energy gamma-ray absorption lines in liquids.

APPENDIX A. SHORT-COLLISION-TIME APPROXIMATION

The "short-collision-time" approximation was first introduced by Wick,[4] who used it to express the total neutron cross section as a series in inverse powers of the neutron energy. (That this series contains only the contributions from collision processes of short duration was rigorously shown to be true for an isotropic oscillator in its ground state.) The developments of the previous section constitute an extension of the short-collision-time approximation to the calculation of the differential properties of neutron scattering. It should be possible, then, to obtain from the expansion (26) the contribution of "short" duration processes to the integral properties of the scattering. In particular, we should be able to obtain the result obtained by Wick for the total cross section, namely,

$$\sigma(E)=\sigma_b\left(\frac{M}{M+1}\right)^2\left[1+\frac{1}{2}\frac{T'}{ME}-\frac{1}{32}\left(\frac{M+1}{M}\right)^2\frac{C_{\rm av}}{ME^3}+O(E^{-4})\right]. \quad \text{(A1)}$$

For the calculation of $\sigma(E)$, one is first tempted to insert (28) into (6) and integrate over energy and angle. The first two terms in (A1) are obtained correctly in this manner, and the contributions from the $B_{\rm av}$ and $(K^2)_{\rm av}$ terms are found to be zero. The term in $C_{\rm av}$, however, gives only half the contribution in (A1) and diverges logarithmically for small momentum transfer. If we wish to include terms of order $1/R$, or higher, in (28), it is necessary to introduce a minimum recoil R_0 and to use (28) only for $R>R_0$. The form of $\sigma(E,E',\theta)$ for $R<R_0$ can be estimated on physical grounds for high incident energy. By such a procedure we cannot choose R_0 so that the error in $\sigma(E)$ is exponentially small. The direct integration of our expansion (28) to obtain the total cross section is therefore not a very useful procedure.

An alternative, and more successful, procedure is to

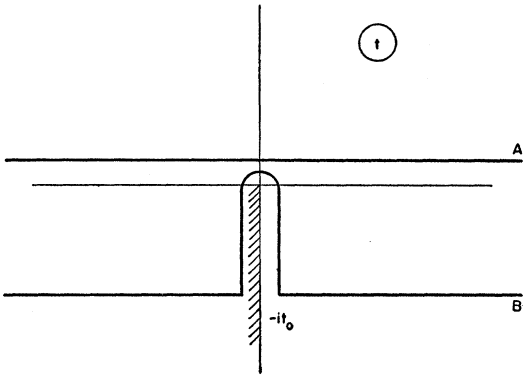

FIG. 1. Contour for t integration in Eq. (A2).

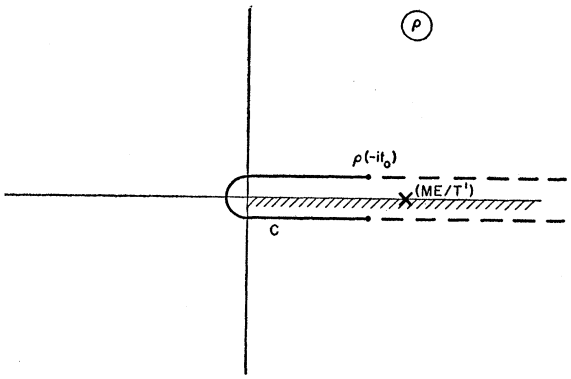

FIG. 2. Contour for ρ integration in Eq. (A4).

make more direct use of the short-collision-time nature of the expansion (26) rather than the large recoil nature of (28). Since our expansion is only valid for short times, we must eliminate the long-time contributions to the cross section on the basis of the behavior of $\chi(\mathbf{K},t)$ for physical systems before making the short-time expansion.

The cross section, $\sigma(E)$, is given by

$$\sigma(E)=\frac{\sigma_b}{8\pi^2k}\int_{-\infty}^{\infty}dt\int d^3\mathbf{K}\,e^{i(E'-E)t}\chi(\mathbf{K},t), \quad \text{(A2)}$$

where $k=(2mE)^{\frac{1}{2}}$ is the incident momentum, and

$$E'-E=(2m)^{-1}[K^2+2\mathbf{k}\cdot\mathbf{K}].$$

Here, we have assumed that the $\mathbf{K}$ and t integrations can be exchanged. As pointed out by Wick, this interchange is permissible if we first attribute to t a small positive imaginary part. This corresponds to the contour A in Fig. 1.

In order to eliminate the long-time contributions to the cross section, we deform the contour from A to B in Fig. 1. For a harmonic system with $\chi(\mathbf{K},t)$ given by (11), it has been shown[4] that the contributions from the horizontal portion of B are of the order of $\exp(-E/T')$, if t_0 is greater than or equal to (M/T'). (This is true except for the special case of an isotropic oscillator, with $M=1$, considered by Wick.) It is probable that the contribution from the horizontal parts of B will be equally small for a more general system, but this has not been shown.

We therefore choose $t_0=(M/T')$ and evaluate the integral around the loop in B by use of the expansion (26). By transforming from $\mathbf{K}$ to

$$\mathbf{q}=\mathbf{K}+\frac{M}{M+1+iT't}\mathbf{k},$$

carrying out the q integration, and introducing the new independent variable

$$\rho=iEt\Big/\left(\frac{M+1}{M}+\frac{T'}{ME}iEt\right), \quad \text{(A3)}$$

the resulting expression becomes

$$\sigma(E)=\sigma_b\left(\frac{M}{M+1}\right)^2\frac{1}{4\pi^{\frac{1}{2}}}\int_C\rho^{-\frac{3}{2}}e^{-\rho}I(\rho)d\rho, \quad \text{(A4)}$$

where

$$I(\rho)=1-\frac{T'}{ME}\rho-\frac{1}{12}\frac{B_{\rm av}}{ME^2}\frac{M+1}{M}\rho^2(3-2\rho)$$
$$-\frac{1}{24}\left(\frac{M+1}{M}\right)^2\frac{C_{\rm av}}{ME^3}\rho^3(3-2\rho)\left(1-\frac{T'\rho}{ME}\right)^{-1}$$
$$+\frac{1}{30}\left((K^2)_{\rm av}-\frac{5}{3}K_{\rm av}^2\right)\frac{1}{M^2E^2}\rho^2\left(1-\frac{T'\rho}{ME}\right)$$
$$\times(15-20\rho+4\rho^2)+O(E^{-4}). \quad \text{(A5)}$$

The contour C in the complex ρ plane is shown in Fig. 2. The branch cut for the integrand runs along the positive real axis starting from the branch point of $\rho^{-\frac{3}{2}}$ at the origin. We note the occurrence of a pole at $\rho=ME/T'$ in the terms in the integral, which are greater than second order in E^{-1}.

The integral in (A4) can be conveniently carried out if we first extend the contour C in Fig. 1 to $+\infty$ on both sides of the real axis. The contribution from the horizontal path between $\rho(-it_0)$ or $\rho^*(-it_0)$ and infinity vanishes as $\exp(-ME/T')$ if t_0 is chosen to be $\gtrsim(M/T')$. For example, the error in the term involving $C_{\rm av}$ is of order

$$\left(\frac{M+1}{M}\right)^2\frac{C_{\rm av}}{ME^3}\left(\frac{ME}{T'}\right)^{\frac{5}{2}}$$
$$\times\exp(-ME/T')\ln\left[1-\frac{T'\rho(-it_0)}{ME}\right]. \quad \text{(A6)}$$

This is the first term in (A5) that would give difficulty if t_0 were to approach infinity. Higher order terms will have an error containing powers of

$$[1-(T'\rho(-it_0)/ME)]^{-1},$$

but these errors will still be proportional to

$$\exp(-ME/T').$$

The integral around our extended contour is conveniently carried out by use of the integral representation of the Γ function.[18] The result of integrating (A4) in this way is (A1). The short-collision-time contribution to the cross section is therefore in agreement with Wick.

Because of the deformation from contour A to contour B in Fig. 1, there are no difficulties with long collision time. The time t_0 is not required to approach infinity, but must only be $\gtrsim(M/T')$. This corresponds to

$$\left[1-\frac{T'\rho(-it_0)}{ME}\right]=M/(M+\tfrac{1}{2}), \tag{A7}$$

so the exponentially small error in (A1) is not appreciably increased by factors containing inverse powers of (A7).

APPENDIX B. T' AND C_{av} FOR A DEBYE CRYSTAL

We found it convenient to define two functions $F(\Theta/2T)$ and $H(\Theta/2T)$ by

$$\tfrac{1}{2}x_2=T'=TF(\Theta/2T), \tag{B1}$$

$$\tfrac{1}{2}x_4=C_{av}=\tfrac{3}{5}T\Theta^2H(\Theta/2T). \tag{B2}$$

From the definitions (15) and (16), we have

$$F(x)=(3/x^3)\int_0^x y^3\coth y\,dy, \tag{B3}$$

$$H(x)=(5/x^5)\int_0^x y^5\coth y\,dy. \tag{B4}$$

For small x, we have[2]

$$F(x)=1+\tfrac{1}{5}x^2+O(x^4), \tag{B5}$$

$$H(x)=1+(5/21)x^2+O(x^4). \tag{B6}$$

and for large x

$$F(x)=\frac{3x}{4}\left(1+\frac{\pi^4}{30x^4}\right), \tag{B7}$$

$$H(x)=\frac{5x}{6}\left(1+\frac{\pi^6}{42x^6}\right), \tag{B8}$$

giving as the zero-temperature limits, $T'\to(\tfrac{3}{8})\Theta$ and $C_{av}\to(\tfrac{1}{4})\Theta$.[3] The behavior for intermediate x was studied numerically on a digital computer,[19] and it was found that the high-temperature form (B5) can be used for $x\lesssim 2$ and the low-temperature form (B7) for $x\gtrsim 2$. The maximum error is for $x=2[T=(\tfrac{1}{4})\Theta]$, where the exact value of F is 1.68 and both (B5) and (B7) give a value of 1.80.

Similarly, for C_{av} the maximum error occurs at $x=2.22$, where the correct value of H is 1.97, and both (B6) and (B8) give 2.21.

[18] P. Morse and R. H. Feshbach, *Methods of Theoretical Physics* (McGraw-Hill Book Company, New York, 1953), p. 431.

[19] Joan Bell (private communication).