CERN group[16] measured scattering in carbon to the ground state and first few excited states, and compared directly with electron scattering results. The previous University of Washington experiment[17] was, like the present one, the summed elastic plus inelastic scattering in carbon. Most of the data was at momentum transfers below 200 MeV/c, and agreement with the low-resolution sum rule was obtained.

The present experiment is marginally able to distinguish some interesting features of nuclear models in light nuclei. Improved muon beams at high intensity accelerators should make possible some analysis of the energy and nature of the scattered particle, thereby placing the muon in a position more nearly competitive with the electron as a probe of medium and long-range nuclear structure. However, short-range correlations will require a better tool than the sum rule.

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Quasiclassical Theory of Neutron Scattering*

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A Wigner representation is used for expressing the thermal average occurring in the Van Hove formalism for slow-neutron scattering from macroscopic systems. For quadratic and lower-degree potentials, results in closed form may be obtained, and in general, an asymptotic series expansion in powers of $\hbar$ is still possible for the incoherent part of the differential cross section for quasiclassical systems. The lead term of this asymptotic expansion results in an expression relating the cross section to a four-dimensional Fourier inversion of the classical space-time distribution $G_s^c(\mathbf{r},t)$, and hence to the classical motions of the atoms in the scattering system. Correction terms of $O(\hbar^2)$ have been obtained explicitly and found to be small for systems at ordinary temperatures. It is shown that (at least to order $\hbar^2$) the results obey the constraint of detailed balance and satisfy the Placzek moments. It is also shown that because of the contact nature of the Fermi pseudopotential, the exact classical limit ($\hbar\rightarrow 0$) for any system is the ideal-gas result. In principle, the results can be extended to all orders of $\hbar^2$. No similar asymptotic expansion appears to exist, however, for the coherent cross section. The analysis is then used for deriving other existing prescriptions and for examining their implications and range of validity.

I. INTRODUCTION

A GENERAL approach to neutron scattering by arbitrary systems of atoms has been presented by Van Hove,[1] based on the Fermi pseudopotential approximation.[2] In this approach, the differential scattering cross section is expressed as a four-dimensional Fourier transform of a space-time correlation function $G(\mathbf{r},t)$. Such a formulation appears, then, as a natural time-dependent generalization of the Zernike-Prins "static approximation,"[3] in which the differential scattering cross section is given in terms of the well-known pair distribution function $g(\mathbf{r})$. In fact, the latter function is equal to the nondiagonal component of $G(\mathbf{r},t)$ evaluated at $t=0$. (Actually, the Van Hove approach is directly applicable to describing system response to any external probe. However, since the major application of the method has been in the field of neutron scattering, we shall discuss it in that context. Translation to other problems is, in most cases, immediate.)

Accurate calculations of $G(\mathbf{r},t)$ are possible only for systems where the many-particle Hamiltonian may be replaced by a sum of single-particle Hamiltonians. This is the case for dilute gases and simple crystals, for which the predicted angular and energy distributions of the scattered neutrons are, indeed, in good agreement with experiment. For dense fluids, on the other hand, the complexity of the atomic dynamics is much greater than in the above mentioned cases, and a calculation of $G(\mathbf{r},t)$, by reduction of the problem to a soluble one-body

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problem, necessitates highly simplifying assumptions in the specific dynamical models used.

There is, however, an alternative approach to the analysis of neutron scattering experiments based on the physical interpretation of the space-time correlation function $G(\mathbf{r},t)$ in the limit $\hbar \rightarrow 0$. In this limit, $G(\mathbf{r},t)$ represents the conditional probability density that given an atom at the origin at time $t=0$ there will be an atom (the same or another) at position $\mathbf{r}$ at time t. We shall denote the function by $G^c(\mathbf{r},t)$.

The plausibility of this approach is then subject to the existence of a relationship between this classical $G^c(\mathbf{r},t)$ and the differential scattering cross section. Establishing such a connection, however, is not as simple as might at first be assumed. Various semiempirical prescriptions have been proposed to this end. The most intuitive of them results simply from replacing $G(\mathbf{r},t)$ in the Van Hove formalism by the function $G^c(\mathbf{r},t)$. This, as observed by Vineyard,[4] corresponds to a development in which the neutron is treated quantum mechanically and the scatterer classically. It has the unsatisfactory features that recoil effects are inadequately treated in that the average energy loss is set equal to zero, and that, as observed by Schofield,[5] it violates the constraint of detailed balance. Schofield has suggested a recipe to remedy these defects in which he sets $G^c(\mathbf{r},t)$ equal to $G(\mathbf{r}, t+i\beta\hbar/2)$, where $\beta=1/k_BT$, instead of $G(\mathbf{r},t)$ and asserts its validity to first order in $\hbar$. (k_B=Boltzmann constant, T=absolute temperature.)

This assertion is not entirely correct, however, as may be seen from the fact that the prescription fails to yield the exact results for the ideal monatomic gas, for which the cross section is, in terms of the significant variables $\Delta\mathbf{p}$ and ϵ ($\Delta\mathbf{p}$, ϵ=momentum and energy transfer, respectively), actually independent of $\hbar$.

All of this is discussed in a previous paper,[6] where a connection is made between $G^c(\mathbf{r},t)$ and the differential scattering cross section, correct to lowest order in $\hbar$, by utilizing a Wigner representation[7,8a–8c] for the thermal average occurring in the expression for $G(\mathbf{r},t)$. The Wigner representation, which we review elsewhere,[9] results in the replacement of the conventional quantum average by a phase-space average, over a Wigner distribution, of the "Weyl[10] equivalent" of the operator present in the thermal average. The purpose of the present paper is to extend the results obtained in Ref. 6. In particular, it is shown that for the incoherent cross section, this "Weyl equivalent" admits an asymptotic series expansion in powers of $\hbar$ where the first contributing correction to the leading term is of order $\hbar^4$ for randomly oriented systems. By combining this expansion with an expansion[7,11] of the Wigner distribution in powers of $\hbar^2$, valid for ordinary temperatures, we obtain explicit corrections of order $\hbar^2$ to the "quasiclassical" approximation which we derive in Ref. 6. The approach here is particularly convenient in that it shows systematically how, in principle, higher-order corrections could be calculated.

It is further noted that because of the presence of an essential singularity in $G(\mathbf{r},t)$ the above indicated procedure may not be applied to the coherent component of the cross section, the exceptions being the cases of harmonic and lower degree potentials. This limitation is not considered too strong, however, because the interference scattering is quite insensitive to target dynamics.

The analysis is also used for deriving other existing prescriptions and to examine critically their implications and range of validity. Finally, numerical computations are presented in which the cross sections for some simple systems, as calculated by the Vineyard prescription, are compared with the results of this work.[12]

II. DENSITY FLUCTUATION CORRELATION FUNCTION IN THE WIGNER REPRESENTATION

In the Van Hove formalism, the differential cross section for scattering of slow neutrons by nuclei in an arbitrary macroscopic aggregate is expressed as the product of a function which depends only on the properties of the individual particles of the system with a spectral function which depends only on the dynamics of the scattering system.[1] The latter, usually denoted by $S(\Delta\mathbf{p},\epsilon)$ is defined as the time Fourier inverse of the density fluctuation correlation function $\chi(\Delta\mathbf{p},t)$[1,13]:

$$\chi(\Delta\mathbf{p},t)=N^{-1}\sum_{i,j=1}^{N}\mathrm{Tr}\left\{\rho\exp\left[-\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{R}_i\right]\times\exp\left[\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{R}_j(t)\right]\right\}. \quad (1)$$

(For the sake of simplicity we consider only monatomic and monoisotopic systems.) Here, ρ is the von Neumann[14] density matrix defined explicitly by

$$\rho=\frac{1}{S}\sum_{\nu=1}^{s}|\psi^\nu\rangle\langle\psi^\nu|, \quad (2)$$

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where $|\psi^\nu\rangle$ is the state vector of the νth system in the Gibbsian ensemble, and the summation is carried over all s systems of the ensemble. $\mathbf{R}_j(t)$ is the Heisenberg position operator for the jth particle at time

$$\mathbf{R}_j(t)=\exp[iH_st/\hbar]\mathbf{R}_j\exp[-iH_st/\hbar],$$

where H_s is the system Hamiltonian, and $\Delta\mathbf{p}=\hbar\boldsymbol{\kappa}$ is the momentum transfer to the scatterer. Then if $\partial^2\sigma/\partial\Omega\partial\epsilon$ represents the differential scattering cross section per atom, per unit solid angle and per unit interval of neutron energy transfer we have, in the absence of spin correlations,

$$(\partial^2\sigma/\partial\Omega\partial\epsilon)=(k/k_0)[a_{\text{inc}}{}^2S_s(\Delta\mathbf{p},\epsilon)+a_{\text{coh}}{}^2S(\Delta\mathbf{p},\epsilon)],\quad (3a)$$

where

$$S_s(\Delta\mathbf{p},\epsilon)=\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}dt\exp\left[-\frac{i\epsilon t}{\hbar}\right]\chi_s(\Delta\mathbf{p},t),$$
$$S(\Delta\mathbf{p},\epsilon)=\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}dt\exp\left[-\frac{i\epsilon t}{\hbar}\right]\chi(\Delta\mathbf{p},t),\quad (3b)$$

and $\chi_s(\Delta\mathbf{p},t)$ is the diagonal $(i=j)$ component of χ.

The function $G(\mathbf{r},t)$ discussed in the previous section is simply

$$G(\mathbf{r},t)=\frac{1}{(2\pi)^3}\int d^3\kappa\exp[-i\boldsymbol{\kappa}\cdot\mathbf{r}]\chi(\hbar\boldsymbol{\kappa},t).\quad (3c)$$

In order to evaluate the thermal average appearing in Eq. (1), we make use of the following results of Ref. 9:

The ensemble expectation value of a quantum-mechanical operator[15] $O(\mathbf{P},\mathbf{R},t)$,

$$\langle O(\mathbf{P},\mathbf{R},t)\rangle_T\equiv\text{Tr}[\rho O(\mathbf{P},\mathbf{R},t)],$$

can be written as[16]

$$\langle O(\mathbf{P},\mathbf{R}\cdot t)\rangle_T\equiv\int\int d\mathbf{p}d\mathbf{q}\,\rho_w(\mathbf{p},\mathbf{q})O_w(\mathbf{p},\mathbf{q},t),\quad (4)$$

where ρ_w is the Wigner distribution function

$$\rho_w(\mathbf{p},\mathbf{q})=\left(\frac{1}{2\pi\hbar}\right)^{3N}\int d\mathbf{z}\exp\left[\frac{i}{\hbar}\mathbf{z}\cdot\mathbf{p}\right]\langle\mathbf{q}-\tfrac{1}{2}\mathbf{z}|\rho|\mathbf{q}+\tfrac{1}{2}\mathbf{z}\rangle\quad (5)$$

and $O_w(\mathbf{p},\mathbf{q},t)$ is the Weyl equivalent of $O(\mathbf{P},\mathbf{R},t)$. We write W: $O(\mathbf{P},\mathbf{R},t)\leftrightarrow O_w(\mathbf{p},\mathbf{q},t)$. Specifically O_w is found from

$$O_w(\mathbf{p},\mathbf{q},t)=\left(\frac{1}{2\pi\hbar}\right)^{3N}\int\int d\mathbf{x}d\mathbf{y}\exp\left[\frac{i}{\hbar}(\mathbf{x}\cdot\mathbf{p}+\mathbf{y}\cdot\mathbf{q})\right]$$
$$\times\text{Tr}\left\{O(\mathbf{P},\mathbf{R},t)\exp\left[-\frac{i}{\hbar}(\mathbf{x}\cdot\mathbf{P}+\mathbf{y}\cdot\mathbf{R})\right]\right\}$$

and leads to:

$$(1)\ W:\quad O_1(\mathbf{P},\mathbf{R},t)O_2(\mathbf{P},\mathbf{R},t)\leftrightarrow O_{1w}e^{(\hbar/2i)\Lambda}O_{2w},\quad (6a)$$

where Λ is the Poisson bracket operator

$$\Lambda=(\overleftarrow{\nabla}_p\cdot\overrightarrow{\nabla}_q-\overleftarrow{\nabla}_q\cdot\overrightarrow{\nabla}_p);$$

$$(2)\ W:\quad O(\mathbf{R})\leftrightarrow O_w(\mathbf{q})=O(\mathbf{q});\quad (6b)$$

$$(3)\ W:\quad O(\mathbf{P})\leftrightarrow O_w(\mathbf{p})=O(\mathbf{p});\quad (6c)$$

(4) When $O(\mathbf{P},\mathbf{R},t)$ denotes the Heisenberg operator

$$O(\mathbf{P},\mathbf{R},t)=\exp[(i/\hbar)Ht]O(\mathbf{P},\mathbf{R})\exp[-(i/\hbar)Ht],$$

then

$$W:\quad O(\mathbf{P},\mathbf{R},t)\leftrightarrow O_w(\mathbf{p},\mathbf{q},t)$$
$$=\exp[(2t/\hbar)H_w\sin(\tfrac{1}{2}\hbar\Lambda)]O_w(\mathbf{p},\mathbf{q}).\quad (6d)$$

All this is explained in more detail in Ref. 9. Applying it now to Eq. (1) we find

$$\chi(\Delta\mathbf{p},\hbar\tau)=N^{-1}\sum_{i,j=1}^{N}\int\int d\mathbf{p}d\mathbf{q}\ \rho_w(\mathbf{p},\mathbf{q})$$
$$\times\left\{\exp\left[-\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{q}_i\right]\exp\left(\frac{\hbar}{2i}\Lambda\right)\Omega_w(\mathbf{p},\mathbf{q},\tau)\right\},\quad (7)$$

where

$$t=\hbar\tau,$$

$$\Omega_w(\mathbf{p},\mathbf{q},\tau)=\exp[2\tau H_w\sin(\tfrac{1}{2}\hbar\Lambda)]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]$$

and

$$H_w(\mathbf{p},\mathbf{q})=H(\mathbf{p},\mathbf{q})=(p^2/2M)+V(\mathbf{q}).$$

Or, since

$$\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_i]\exp\left(\frac{\hbar}{2i}\Lambda\right)$$
$$=\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_i]\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\nabla_{p_i}]\quad (8)$$

then

$$\chi(\Delta\mathbf{p},\hbar\tau)=N^{-1}\sum_{i,j=1}^{N}\int\int d\mathbf{p}d\mathbf{q}\,\rho_w(\mathbf{p},\mathbf{q})$$
$$\times\exp\left[-\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{q}_i\right]\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\nabla_{p_i}]\Omega_w(\mathbf{p},\mathbf{q},\tau).\quad (9)$$

It is interesting to note that if V is quadratic or of lower degree in $\mathbf{q}$, the operator

$$H_w\Lambda^{2m+1}=0\quad\text{for}\quad m\geq 1$$

and Eq. (9) reduces to

$$\chi(\Delta\mathbf{p},\hbar\tau)=N^{-1}\sum_{i,j=1}^{N}\int\int d\mathbf{p}d\mathbf{q}\,\rho_w(\mathbf{p},\mathbf{q})\exp\left[-\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{q}_i\right]$$
$$\times\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\nabla_{p_i}]\exp\left[\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)\right],\quad (10)$$

where use has been made of the Taylor series expansion

[15] $\mathbf{P}$ and $\mathbf{R}$ represent $3N$-dimensional quantum mechanical momentum and position operators corresponding to an N-particle system.

[16] $\mathbf{q}$ and $\mathbf{p}$ are $3N$-dimensional c numbers which satisfy Hamilton's equations of motion, and thus may be interpreted as classical dynamical variables. However, the same variables subindexed refer specifically to the particle denoted by the index.

property

$$\exp[\hbar\tau H_w\Lambda]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]=\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]. \quad (11)$$

In these cases, the expression for ρ_w may be obtained in closed form and the calculation of $\chi(\Delta\mathbf{p},\hbar\tau)$ is straightforward.

A. Asymptotic Expansion of $\chi(\Delta\mathbf{p},\hbar\tau)$

Except for the special cases mentioned previously, a solution in closed form of Eq. (9) is impossible for the following reasons:

(1) The operator $\exp[2\tau H_w\sin(\frac{1}{2}\hbar\Lambda)]$ acting on $\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]$ yields an infinite series; and

(2) $\rho_w(\mathbf{p},\mathbf{q})$ cannot be obtained in closed form although, as mentioned previously, for a "quasiclassical" system a series expansion in powers of $\hbar^2$ is possible. A similar expansion in powers of $\hbar$ for the rest of the integrand in Eq. (9) is not possible, however, because it contains an essential singularity at the point $\hbar=0$.

All of these considerations lead us to attempt, then, an asymptotic expansion[17] for $\Omega_w(\mathbf{p},\mathbf{q},\tau)$. The term $\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}]$ is retained *in toto*. To determine the form of this asymptotic expansion, we note that

$$\exp[2\tau H_w\sin(\tfrac{1}{2}\hbar\Lambda)]=\exp[\tau(\hbar H_w\Lambda+A)], \quad (12a)$$

where

$$A=-2\sum_{n=1}^{\infty}\frac{(-1)^n}{(2n+1)!}\left(\frac{\hbar}{2}\right)^{2n+1}(\nabla_q V\cdot\overrightarrow{\nabla}_p)(\overleftarrow{\nabla}_q\cdot\overrightarrow{\nabla}_p)^{2n}. \quad (12b)$$

Moreover, making use of the identity (A1) and Eqs. (A2) and (A3) as proven in Appendix A, we get

$$\exp[\tau(\hbar H_w\Lambda+A)]=e^{\hbar\tau H_w\Lambda}e^{\tau A}\Gamma(\tau) \quad (13)$$

and

$$\partial\Gamma(\tau)/\partial\tau=\Upsilon(\tau)\Gamma(\tau),\quad \Gamma(\tau=0)=1,$$

where

$$\Upsilon(\tau)=e^{-\tau A}\sum_{n=1}^{\infty}\frac{(-1)^n}{n!}\tau^n[\hbar H_w\Lambda,A]_n e^{\tau A}. \quad (14)$$

Equation (14) may be integrated formally, leading to the integral equation

$$\Gamma(\tau)=1+\int_0^\tau\Upsilon(\tau')\Gamma(\tau')d\tau'$$

which is readily solved by Picard's process of successive approximations, yielding

$$\Gamma(\tau)=1+\sum_{m=1}^{\infty}\int_0^\tau\Upsilon(\tau_1)d\tau_1\int_0^{\tau_1}\Upsilon(\tau_2)d\tau_2\cdots\times\int_0^{\tau_{m-1}}\Upsilon(\tau_m)d\tau_m,\quad(\tau_0=\tau). \quad (15)$$

Observing, however, that each term in the sum in Eq. (15), when operating on $\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}]$, generates an infinite series in powers of $\hbar$, of which the lowest is $\hbar^0$, we can write

$$\Omega_w(\mathbf{p},\mathbf{q},\tau)=\exp[\tau(\hbar H_w\Lambda+A)]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]=f(\tau)[1+\hbar F_1(\tau)+\hbar^2F_2(\tau)+\cdots], \quad (16)$$

where

$$f(\tau)=\exp[\hbar\tau H_w\Lambda]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j] \quad (17)$$

and

$$f(0)=\Omega_w(\mathbf{p},\mathbf{q},0)=\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]$$
$$F_n(0)=0,\quad \text{for } n>0. \quad (18)$$

The terms in the expansion (16) may be evaluated by substituting this equation into the expression

$$(\partial\Omega_w/\partial t)(\mathbf{p},\mathbf{q},t)=(2/\hbar)H_w\sin(\tfrac{1}{2}\hbar\Lambda)\Omega_w(\mathbf{p},\mathbf{q},t) \quad (19)$$

which describes the time evolution of the Weyl equivalent of a Heisenberg operator, thus obtaining

$$f(\tau)[\hbar\dot{F}_1(\tau)+\hbar^2\dot{F}_2(\tau)+\cdots]-f(\tau)\hbar H_w\Lambda[1+\hbar F_1(\tau)+\cdots]-\sum_{m=1}^{\infty}\frac{(-1)^m(\hbar)^{2m+1}H_w\Lambda^{2m+1}}{4^m(2m+1)!}\{f(\tau)[1+\hbar F_1(\tau)+\hbar^2F_2(\tau)+\cdots]\}=0, \quad (20)$$

where

$$\dot{F}_n=\partial F_n/\partial\tau.$$

Grouping terms with equal powers of $\hbar$, by explicitly taking into account that

$$-\hbar^{2m+1}H_w\Lambda^{2m+1}[f(\tau)F_n(\tau)]=\hbar^{2m+1}(\nabla_qV\cdot\overrightarrow{\nabla}_p)(\overleftarrow{\nabla}_q\cdot\overrightarrow{\nabla}_p)^{2m}\{F_n(\tau)\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j+(i\tau/M)\Delta\mathbf{p}\cdot\mathbf{p}_j-(i\hbar\tau^2/2M)\Delta\mathbf{p}\cdot\nabla_{q_j}V-(i\hbar^2\tau^3/6M^2)(\mathbf{p}\cdot\nabla_q)(\Delta\mathbf{p}\cdot\nabla_{q_j}V)+\cdots]\}=O(\hbar^{2m+1}) \quad (21)$$

yields the following set of differential equations for the first three terms in the expansion (16):

$$\begin{aligned}&\dot{F}_1(\tau)-H_w\Lambda F_0(\tau)=0,\\&\dot{F}_2(\tau)-H_w\Lambda F_1(\tau)=0,\\&\dot{F}_3(\tau)-H_w\Lambda F_2(\tau)-(1/24)(i\tau/M)^3(\Delta\mathbf{p}\cdot\nabla_{q_j})^2(\Delta\mathbf{p}\cdot\nabla_{q_j}V)=0.\end{aligned} \quad (22)$$

[17] An approach somewhat akin to the asymptotic expansion of the WKB method.

The solutions to these equations, with the initial conditions given by Eqs. (18), are

$$F_1(\tau)=F_2(\tau)=0$$
$$F_3(\tau)=-(i\tau^4/96M^3)(\Delta\mathbf{p}\cdot\nabla_{q_j})^2(\Delta\mathbf{p}\cdot\nabla_{q_j}V). \quad (23)$$

Hence, Eq. (16) may be expressed as

$$\Omega_w(\mathbf{p},\mathbf{q},\tau)=\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)][1-(i\tau^4\hbar^3/96M^3)(\Delta\mathbf{p}\cdot\nabla_{q_j})^2(\Delta\mathbf{p}\cdot\nabla_{q_j}V)+O(\hbar^4)], \quad (24)$$

and inserting this result into Eq. (9) yields

$$\chi(\Delta\mathbf{p},\hbar\tau)=N^{-1}\sum_{i,j}\langle\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_i]\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\nabla_{p_j}]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]$$
$$\times[1-(i\hbar^3\tau^4/96)(\Delta\mathbf{p}\cdot\nabla_{q_j})^2(\Delta\mathbf{p}\cdot\nabla_{q_j}V)+O(\hbar^4)]\rangle_{TW}, \quad (25)$$

where $\langle\ \rangle_{TW}$ denotes the phase-space average over ρ_w. Equation (25) is the desired asymptotic expansion. As shown below, the contribution of $O(\hbar^0)$ from the term containing $F_3(\tau)$ vanishes for randomly oriented systems. Retaining only the leading term in the asymptotic expansion, which still contains $\hbar$, and using the $\hbar^0$ term of ρ_w gives what we call the "quasiclassical" approximation.[6] The first correction is of $O(\hbar^2)$ and comes from the $\hbar^2$ term of ρ_w. The next term is of $O(\hbar^4)$; one contribution comes from the $\hbar^4$ term in ρ_w; another comes from F_3 and F_4. However, we consider only terms as high as $\hbar^2$.

Note that Eq. (25) still contains an essential singularity which, for the diagonal component of χ, is only apparent since in this case $i=j$; the term $e^{(-i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j}$ is canceled out by the first term resulting from a power series expansion of $\mathbf{q}_j(\hbar\tau)$. It can be shown that only in this case is it possible to have a power series expansion for χ. Hence, the following discussion will be restricted to direct scattering.

B. Randomly Oriented Systems

If in the term involving $F_3(\tau)$ of Eq. (25) we write

$$\Delta\mathbf{p}\cdot\nabla_{q_j}=\mu|\Delta\mathbf{p}|(\partial/\partial q_j),$$

where

$$\mu\equiv\Delta\mathbf{p}\cdot\mathbf{q}_j/|\Delta\mathbf{p}||\mathbf{q}_j|,$$

then

$$\langle\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\nabla_{p_j}]F_3\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]\rangle_{TW}$$
$$=-\frac{i\tau^4}{96M^3}\left\langle\mu^3|\Delta\mathbf{p}|^3\frac{\partial^3V}{\partial q_j^3}\exp\left[\frac{i\tau}{M}\Delta\mathbf{p}\cdot\mathbf{p}_j\right]\right\rangle_{TW}\exp\left[\frac{i\tau\Delta p^2}{2M}\right]+O(\hbar). \quad (26)$$

Moreover, for a randomly oriented system, $\chi(\Delta\mathbf{p},\hbar\tau)$ can depend only on the magnitude of $\Delta\mathbf{p}$. Consequently,

$$\chi(\Delta\mathbf{p},\hbar\tau)=\chi(|\Delta\mathbf{p}|,\hbar\tau)\equiv(1/4\pi)\int\chi(|\Delta\mathbf{p}|,\hbar\tau)d\Omega_{\Delta p}.$$

It follows readily from this that the first term on the right of (26) vanishes, and (25) yields

$$\chi_s(\Delta\mathbf{p},\hbar\tau)=\langle\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\nabla_{p_j}]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]\rangle_{TW}+O(\hbar^4). \quad (27)$$

Introducing now the expansion[7]

$$\rho_w=f_N{}^c(1+\hbar^2A_2+\hbar^4A_4+\cdots) \quad (28)$$

of the Wigner distribution function into Eq. (27) results in

$$\chi_s(\Delta\mathbf{p},\hbar\tau)=\langle\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\nabla_{p_j}]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]\rangle_{TC}$$
$$+\hbar^2\langle\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]A_2\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\nabla_{p_j}]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]\rangle_{TC}+O(\hbar^4). \quad (29)$$

Here the phase-space average is performed with respect to the classical joint canonical distribution function $f_N{}^c$, and $\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\nabla_{p_j}]\mathbf{q}_j(t)$ is the vector position of the jth particle at time t, subject to an impulse at $t=0$ of the force

$$\mathbf{F}_{\text{imp}}=\tfrac{1}{2}\Delta\mathbf{p}\delta(t).$$

The above result is extremely useful because the corrections of order $\hbar^2$ to the "quasiclassical" limit come only from ρ_w. Note that this derivation is quite self-consistent in that corrections of $O(\hbar^4)$ and higher could be obtained in principle by straightforward extension of the manipulations carried out so far. The analysis becomes laborious, but at least the procedure is well defined. However, if correction terms of $O(\hbar^4)$ to our approximation are important (as is the case for systems near absolute zero[11]), this approach is likely to be poor anyway. Also, note that by partial integration of the first term in (29), $\exp[\frac{1}{2}\Delta\mathbf{p}\cdot\overleftrightarrow{\nabla}_{p_j}]$ can be made to operate on f_N^c. As will be seen below, this results in a particularly convenient form for relating the quasiclassical approximation to the classical singlet particle space-time correlation function.

C. Corrections of $O(\hbar^2)$ to the "Quasiclassical" Approximation

As previously observed, the second term in Eq. (29) gives quantum-mechanical corrections to our "quasiclassical" approximation and contains all powers of $\hbar$, the lowest being of $O(\hbar^2)$. Note, however, that retaining terms of order higher than $\hbar^2$ is senseless, since these terms were neglected in the expansions of both Ω_w and ρ_w. Thus, expanding $\mathbf{q}_j(\hbar\tau)$ in this term in a Maclaurin series and ignoring terms beyond $O(\hbar)$ yields

$$\hbar^2\langle\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]A_2\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\boldsymbol{\nabla}_{p_j}]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]\rangle_{TC}=\exp[i\tau\Delta p^2/2M]\langle\hbar^2A_2\exp[(i\tau/M)\Delta\mathbf{p}\cdot\mathbf{p}_j]\rangle_{TC}$$
$$-(i\hbar^3\tau^2/2M)\exp[i\tau\Delta p^2/2M]\langle(\Delta\mathbf{p}\cdot\boldsymbol{\nabla}_{q_j}V)A_2\exp[(i\tau/M)\Delta\mathbf{p}\cdot\mathbf{p}_j]\rangle_{TC}+O(\hbar^4).$$

Again, for a randomly oriented system, the term of $O(\hbar^3)$ vanishes and

$$\hbar^2\langle\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]A_2\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\boldsymbol{\nabla}_{p_j}]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]\rangle_{TC}$$
$$=\exp[i\tau\Delta p^2/2M]\langle\hbar^2A_2\exp[(i\tau/M)\Delta\mathbf{p}\cdot\mathbf{p}_j]\rangle_{TC}+O(\hbar^4),$$
$$=\exp[i\tau\Delta p^2/2M]\langle(1+\hbar^2A_2+\cdots)\exp[(i\tau/M)\Delta\mathbf{p}\cdot\mathbf{p}_j]\rangle_{TC}-\exp[i\tau\Delta p^2/2M]\langle\exp[(i\tau/M)\Delta\mathbf{p}\cdot\mathbf{p}_j]\rangle_{TC}+O(\hbar^4),$$
$$=\exp[i\tau\Delta p^2/2M]\int\int f_1(\mathbf{p}_j,\mathbf{q}_j)\exp[(i\tau/M)\Delta\mathbf{p}\cdot\mathbf{p}_j]d^3p_jd^3q_j-\exp[i\tau\Delta p^2/2M]\langle\exp[(i\tau/M)\Delta\mathbf{p}\cdot\mathbf{p}_j]\rangle_{TC}+O(\hbar^4),\quad(30)$$

where

$$f_1(\mathbf{p}_j,\mathbf{q}_j)=\left(\frac{\beta}{2\pi M}\right)^{3/2}\exp\left[-\frac{\beta p_j^2}{2M}\right]\left[n_1(\mathbf{q}_j)+\frac{\hbar^2\beta^2}{24M}\left(\frac{\beta p_j^2}{3M}-1\right)(N-1)\int n_2^c(\mathbf{q}_j,\mathbf{r}+\mathbf{q}_j)\nabla_r^2\phi(\mathbf{r})d^3r+\cdots\right]$$

is the singlet specific distribution function evaluated in Ref. 11.

Substituting this formula into (30) and performing the indicated operations yields

$$\hbar^2\langle\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]A_2\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\boldsymbol{\nabla}_{p_j}]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]\rangle_{TC}$$
$$=-\frac{\beta}{2}(N-1)\left(\frac{\hbar\tau\Delta p}{6M}\right)^2\exp\left[\frac{i\tau\Delta p^2}{2M}\right]\exp\left[-\frac{\tau^2\Delta p^2}{2M\beta}\right]\int\int n_2(\mathbf{q}_j,\mathbf{r}+\mathbf{q}_j)\nabla_r^2\phi(\mathbf{r})d^3rd^3q_j+O(\hbar^4),\quad(31)$$

where the classical specific doublet density distribution function n_2^c has been replaced to first approximation by the actual doublet density distribution function n_2 after noting that

$$n_2=n_2^c+O(\hbar^2).$$

It is conventional to rewrite this quantity according to[18]

$$(N-1)n_2(\mathbf{q}_j,\mathbf{r}+\mathbf{q}_j)d^3rd^3q_j=(N-1)n(\mathbf{q}_j)n_2(\mathbf{q}_j|\mathbf{r}+\mathbf{q}_j)d^3rd^3q_j,\quad(32)$$

where

$$(N-1)n_2(\mathbf{q}_j|\mathbf{r}+\mathbf{q}_j)d^3r$$

is the probability of finding a second unspecified particle in d^3r about $\mathbf{r}$ given that the jth particle is in $\mathbf{q}_j$ and $n(\mathbf{q}_j)d^3q_j$ is the probability of finding the jth particle in d^3q_j about $\mathbf{q}_j$. In a fluid, n_2 can depend only on $|\mathbf{r}+\mathbf{q}_j-\mathbf{q}_j|=r$ and Eq. (32) simplifies to

$$(N-1)n_2(r)d^3rd^3q_j=n(q_j)g(r)d^3rd^3q_j,\quad(33)$$

where $g(r)$ is just the familiar radial distribution function obtained experimentally from x-ray scattering. Substituting this expression into (31) and integrating over $\mathbf{q}_j$ results in

$$\hbar^2\langle\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]A_2\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\boldsymbol{\nabla}_{p_j}]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]\rangle_{TC}$$
$$=-\frac{\beta}{2}\left(\frac{\hbar\tau\Delta p}{6M}\right)^2\exp\left[\frac{i\tau\Delta p^2}{2M}\right]\exp\left[-\frac{\tau^2\Delta p^2}{2M\beta}\right]\int g(r)\nabla_r^2\phi(\mathbf{r})d^3r+O(\hbar^4).\quad(34)$$

[18] T. R. Hill, *Statistical Mechanics* (McGraw-Hill Book Company, Inc., New York, 1956).

Thus, the density fluctuation correlation function is given as

$$\chi_s(\Delta\mathbf{p},\hbar\tau)=\langle\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\nabla_{p_j}]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]\rangle_{TC}$$
$$-\frac{\beta}{2}\left(\frac{\hbar\tau\Delta p}{6M}\right)^2\exp\left[\frac{i\tau\Delta p^2}{2M}\right]\exp\left[-\frac{\tau^2\Delta p^2}{2M\beta}\right]\int g(r)\nabla_r^2\phi(\mathbf{r})d^3r+O(\hbar^4). \quad (35)$$

D. Placzek Moments[19]

A consistency check on the above results is provided by the so-called Placzek moments defined by

$$\langle\epsilon_s\rangle_n=(-i)^n\frac{d^n}{d\tau^n}\chi_s(\Delta\mathbf{p},\hbar\tau)\big|_{\tau=0}. \quad (36)$$

Substitution of Eq. (35) into this expression yields

$$\langle\epsilon_s\rangle_0=\chi_s(\Delta\mathbf{p},0)$$

and

$$\langle\epsilon_s\rangle_n=-(-i)^{n+1}\hbar^{n-1}\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\left\langle\exp\left[\frac{\beta}{2M}\Delta\mathbf{p}\cdot\mathbf{p}_j\right]\exp\left[-\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{q}_j\right]\right.$$
$$\left.\times\left[\frac{1}{M}\mathbf{p}_j\cdot\nabla_{q_j}-\nabla V\cdot\nabla_p\right]^{n-1}\left[\frac{1}{M}\mathbf{p}_j\cdot\Delta\mathbf{p}\right]\exp\left[\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{q}_j\right]\right\rangle_{TC}-\frac{\beta}{2}(-i)^n\left(\frac{\hbar\Delta p}{6M}\right)^2\int g(r)\nabla_r^2\phi(\mathbf{r})d^3r$$
$$\times\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\frac{d^n}{d\tau^n}\left\{\tau^2\exp\left[-\left(\tau-\frac{i\beta}{2}\right)^2\frac{\Delta p^2}{2M\beta}\right]\right\}\bigg|_{\tau=0}+O(\hbar^3),\quad\text{for}\quad n\geq 1. \quad (37)$$

The first few moments may be evaluated by tedious but straightforward application of these equations, and are given by

$$\begin{aligned}
\langle\epsilon_s\rangle_0&=1,\\
\langle\epsilon_s\rangle_1&=\Delta p^2/2M,\\
\langle\epsilon_s\rangle_2&=\left\{\frac{2}{3M}\Delta p^2\left[\frac{3}{2\beta}+\frac{\hbar^2\beta}{24M}\langle\nabla_{q_j}^2V\rangle_{TC}+O(\hbar^4)\right]+\frac{\Delta p^4}{4M^2}\right\}=\left\{\frac{2}{3M}\Delta p^2\langle K\rangle+\frac{\Delta p^4}{4M^2}\right\}+O(\hbar^4),\\
\langle\epsilon_s\rangle_3&=\left\{\frac{\Delta p^4}{M^2}\langle K\rangle+\frac{\Delta p^6}{8M^3}+\frac{\hbar^2\Delta p^2}{6M^2}\langle\nabla^2V\rangle\right\}+O(\hbar^4),\\
\langle\epsilon_s\rangle_4&=\left\{\frac{4\Delta p^4}{5M^2}\langle K^2\rangle+\frac{\Delta p^6}{M^3}\langle K\rangle+\frac{\Delta p^8}{16M^4}+\frac{\hbar^2}{3}\frac{\Delta p^2}{M^2}\langle|\nabla V|^2\rangle+\frac{\hbar^2}{3}\frac{\Delta p^4}{M^3}\langle\nabla^2V\rangle\right\}+O(\hbar^4).
\end{aligned} \quad (38)$$

(K here denotes the kinetic energy of the scattering atom). Equations (38) are indeed correct to $O(\hbar^2)$, as may be seen by comparison with Placzek's results.[19]

E. Time-Displaced Pair Distribution Formulation

In order to obtain further information from Eq. (35) on the atomic motions of the scattering system, one may resort to specific dynamical models leading to a soluble Hamiltonian. From these models, values for the angular and energy distribution of the scattered neutrons can be predicted, and these predictions are then subjected to experimental test.

However, as indicated in Sec. I, there is an alternate approach which does not require any assumptions at this point on the dynamics of the scattering system. It is based on the physical interpretation of the classical limit ($\hbar\to 0$) of Van Hove's space-time correlation function $G_s(\mathbf{r},t)$, and the possibility of establishing a relationship, if only approximate, between the direct scattering cross section and this "classical" $G_s^c(\mathbf{r},t)$.

One such relationship was suggested by Vineyard,[4] who proposed that the classical limit of the direct scattering differential cross section could be obtained by substituting $G_s^c(\mathbf{r},t)$ for $G_s(\mathbf{r},t)$ in

$$\begin{aligned}
S_s(\Delta\mathbf{p},\epsilon)&\\
&=\frac{1}{2\pi\hbar}\int_{-\infty}^{\infty}dt\,e^{-i\epsilon t/\hbar}\chi_s(\Delta\mathbf{p},t),\\
&=\frac{1}{2\pi\hbar}\int d\mathbf{r}\int_{-\infty}^{\infty}dt\,\exp\left[\frac{i}{\hbar}(\Delta\mathbf{p}\cdot\mathbf{r}-\epsilon t)\right]G_s(\mathbf{r},t).
\end{aligned} \quad (39)$$

[19] G. Placzek, Phys. Rev. 86, 377 (1952).

That this approximation is unsatisfactory may be seen from the fact that in obtaining $G_s^c(\mathbf{r},t)$ by setting $\hbar=0$ in $G_s(\mathbf{r},t)$ zero momentum transfer is implied (since $\boldsymbol{\kappa}=\Delta\mathbf{p}/\hbar$ and $\boldsymbol{\kappa}$ is kept finite).[6] This may be best illustrated by considering an ideal gas for which

$$G_s(\mathbf{r},t)=[M\beta/2\pi t(t-i\beta\hbar)]^{3/2} \times\exp[-Mr^2\beta/2t(t-i\beta\hbar)] \quad (40)$$

and

$$S_s(\Delta\mathbf{p},\epsilon)=\exp\left(\frac{\beta\epsilon}{2}\right)\exp\left(-\frac{\beta\Delta p^2}{8M}\right)\left(\frac{\beta M}{2\pi\Delta p^2}\right)^{1/2} \times\exp\left[-\frac{M\epsilon^2\beta}{2\Delta p^2}\right]. \quad (41)$$

Although in this case the cross section is entirely classical (in terms of the significant variables $\Delta\mathbf{p}$ and ϵ), the Fourier transform of $G_s^c(\mathbf{r},t)$ obtained by taking the limit $\hbar\to 0$ in Eq. (40) yields

$$S_s^c(\Delta\mathbf{p},\epsilon)=(\beta M/2\pi\Delta p^2)^{1/2}\exp[-M\epsilon^2\beta/2\Delta p^2]; \quad (42)$$

i.e.,

$$\lim_{\hbar\to 0} S_s(\Delta\mathbf{p},\epsilon)\neq S_s^c(\Delta\mathbf{p},\epsilon). \quad (43)$$

Additional evidence of the inacceptability of Vineyard's approximation is provided by observing that the symmetry condition

$$G_s^c(\mathbf{r},t)=G_s^c(-\mathbf{r},-t) \quad (44)$$

implies that the scattering function calculated from $G_s^c(\mathbf{r},t)$ will obey the relation

$$S_s^c(\Delta\mathbf{p},\epsilon)=S_s^c(-\Delta\mathbf{p},-\epsilon), \quad (45)$$

thus violating (as shown by Schofield[5]) the constraint

$$S_s(\Delta\mathbf{p},\epsilon)=e^{\beta\epsilon}S_s(-\Delta\mathbf{p},-\epsilon), \quad (46)$$

as well as the Placzek moments. Nonetheless, an improved prescription which relates the cross section to $G_s^c(\mathbf{r},t)$ and does not suffer from the above mentioned difficulties may be obtained. To this end, we integrate (35) by parts to obtain

$$\chi_s(\Delta\mathbf{p},\hbar\tau)=\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\left\langle\exp\left\{\frac{i}{\hbar}\Delta\mathbf{p}\cdot\left[\mathbf{q}_j(\hbar\tau)-\mathbf{q}_j-\frac{i\beta\hbar\mathbf{p}_j}{2M}\right]\right\}\right\rangle_{TC}$$
$$-\frac{\beta}{2}\left(\frac{\hbar\tau\Delta p}{6M}\right)^2\exp\left[\frac{i\tau\Delta p^2}{2M}\right]\exp\left[-\frac{\tau^2\Delta p^2}{2M\beta}\right]\int g(r)\nabla_r^2\phi(\mathbf{r})d^3r+O(\hbar^4). \quad (47)$$

Furthermore,

$$\exp\left[-\frac{i}{\hbar}\Delta\mathbf{p}\cdot\left(\mathbf{q}_j+\frac{i\beta\hbar}{2M}\mathbf{p}_j\right)\right]=\exp\left[-\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{q}_j\left(\frac{i\beta\hbar}{2}\right)\right]$$
$$\times\left[1+\frac{i\beta^2\hbar}{8M}\Delta\mathbf{p}\cdot\nabla_{q_j}V-\frac{\beta^4\hbar^2}{128M^2}(\Delta\mathbf{p}\cdot\nabla_{q_j}V)^2-\frac{\beta^3\hbar^2}{48M^2}(\mathbf{p}\cdot\nabla_q)(\Delta\mathbf{p}\cdot\nabla_{q_j}V)+O(\hbar^3)\right] \quad (48)$$

and substituting this expression into (47) results in

$$\chi_s(\Delta\mathbf{p},\hbar\tau)=\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\left\langle\exp\left\{\frac{i}{\hbar}\Delta\mathbf{p}\cdot\left[\mathbf{q}_j(\hbar\tau)-\mathbf{q}_j\left(\frac{i\beta\hbar}{2}\right)\right]\right\}\right\rangle_{TC}$$
$$+\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\frac{i\beta^2\hbar}{8M}\left\langle\Delta\mathbf{p}\cdot\nabla_{q_j}V\exp\left[\frac{i}{M}\left(\tau-\frac{i\beta}{2}\right)\Delta\mathbf{p}\cdot\mathbf{p}_j\right]\right\rangle_{TC}$$
$$+\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\left(\tau^2+\frac{\beta^2}{8}\right)\frac{\beta^2\hbar^2}{16M^2}\left\langle(\Delta\mathbf{p}\cdot\nabla_{q_j}V)^2\exp\left[\frac{i}{M}\left(\tau-\frac{i\beta}{2}\right)\Delta\mathbf{p}\cdot\mathbf{p}_j\right]\right\rangle_{TC}$$
$$-\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\frac{\beta^3\hbar^2}{48M^2}\left\langle(\mathbf{p}\cdot\nabla_q)(\Delta\mathbf{p}\cdot\nabla_{q_j}V)\exp\left[\frac{i}{M}\left(\tau-\frac{i\beta}{2}\right)\Delta\mathbf{p}\cdot\mathbf{p}_j\right]\right\rangle_{TC}$$
$$-\frac{\beta}{2}\left(\frac{\hbar\tau\Delta p}{6M}\right)^2\exp\left[\frac{i\tau\Delta p^2}{2M}\right]\exp\left[-\frac{\tau^2\Delta p^2}{2M\beta}\right]\int g(r)\nabla_r^2\phi(\mathbf{r})d^3r+O(\hbar^3). \quad (49)$$

By the same argument used to justify Eq. (27), it can be shown that for a randomly oriented system the second term on the right of (49) and terms of $O(\hbar^3)$ will vanish. The third, fourth, and fifth terms can be combined into one since the mean values involved are connected by the relation

$$\langle|\nabla_{q_j}V|^2\rangle_{TC}=\frac{1}{\beta}\langle\nabla_{q_j}{}^2V\rangle_{TC} \tag{50}$$

which readily follows from applying Green's theorem to the identity

$$\frac{1}{Z}\int\int d\mathbf{q}d\mathbf{p}\nabla_{q_j}\cdot(e^{-\beta H}\nabla_{q_j}V)=\frac{1}{Z}\int\int d\mathbf{p}d\mathbf{q}(\nabla_{q_j}V\cdot\nabla_{q_j}e^{-\beta H})+\frac{1}{Z}\int\int d\mathbf{p}d\mathbf{q}(e^{-\beta H}\nabla_{q_j}{}^2V). \tag{51}$$

Thus,

$$\chi_s(\Delta\mathbf{p},\hbar\tau)=\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\left\langle\exp\left\{\frac{i}{\hbar}\Delta\mathbf{p}\cdot\left[\mathbf{q}_j(\hbar\tau)-\mathbf{q}_j\left(\frac{i\beta\hbar}{2}\right)\right]\right\}\right\rangle_{TC}$$
$$+\left(\frac{\tau^2}{\beta}-i\tau-\frac{\beta}{8}\right)\left(\frac{\hbar\beta\Delta p}{12M}\right)^2\exp\left[-\left(\tau-\frac{i\beta}{2}\right)^2\frac{\Delta p^2}{2M\beta}\right]\int g(r)\nabla_r{}^2\phi(\mathbf{r})d^3r+O(\hbar^4). \tag{52}$$

Moreover, due to time translational invariance

$$\left\langle\exp\left\{\frac{i}{\hbar}\Delta\mathbf{p}\cdot\left[\mathbf{q}_j(\hbar\tau)-\mathbf{q}_j\left(\frac{i\beta\hbar}{2}\right)\right]\right\}\right\rangle_{TC}=\left\langle\exp\left\{\frac{i}{\hbar}\Delta\mathbf{p}\cdot\left[\mathbf{q}_j\left(\hbar\tau-\frac{i\beta\hbar}{2}\right)-\mathbf{q}_j\right]\right\}\right\rangle_{TC} \tag{53}$$

and after a simple transformation of variables, Eq. (52) becomes

$$\chi_s\left(\Delta\mathbf{p},\,\hbar\tau+\frac{i\beta\hbar}{2}\right)=\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\left\{\chi_s{}^c(\Delta\mathbf{p},\hbar\tau)+\left(\frac{\tau^2}{\beta}+\frac{\beta}{8}\right)\left(\frac{\hbar\beta\Delta p}{12M}\right)^2\exp\left[-\frac{\tau^2\Delta p^2}{2M\beta}\right]\int g(r)\nabla_r{}^2\phi(\mathbf{r})d^3r\right\}+O(\hbar^4), \tag{54}$$

where

$$\chi_s{}^c(\Delta\mathbf{p},\hbar\tau)=\left\langle\exp\left\{\frac{i}{\hbar}\Delta\mathbf{p}\cdot[\mathbf{q}_j(\hbar\tau)-\mathbf{q}_j]\right\}\right\rangle_{TC}. \tag{55}$$

It is interesting to note at this point that the essential singularity in (55) is only apparent and disappears, as indicated previously, when expanding $\mathbf{q}_j(\hbar\tau)$ in a Maclaurin series. Furthermore, multiplying both sides of (54) by $(1/2\pi)\exp(-i\epsilon\tau)d\tau$, integrating over all values of τ, and making use of the relation

$$e^{-\beta\epsilon/2}S(\Delta\mathbf{p},\epsilon)=\frac{1}{2\pi}\int_{-\infty}^{\infty}d\tau\, e^{-i\tau\epsilon}\chi\left(\Delta\mathbf{p},\,\hbar\tau+\frac{i\beta\hbar}{2}\right) \tag{56}$$

yields

$$S_s(\Delta\mathbf{p},\epsilon)=\exp\left[\frac{\beta\epsilon}{2}\right]\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\frac{1}{2\pi}\int d\mathbf{r}\exp\left[\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{r}\right]\int_{-\infty}^{\infty}d\tau\, e^{-i\epsilon\tau}G_s{}^c(\mathbf{r},\hbar\tau)$$
$$+\exp\left[\frac{\beta\epsilon}{2}\right]\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\left(\frac{\hbar\beta}{12}\right)^2\left(\frac{\beta}{2\pi M\Delta p^2}\right)^{1/2}\left(1-\frac{\epsilon^2M\beta}{\Delta p^2}+\frac{\Delta p^2\beta}{8M}\right)\exp\left[-\frac{M\beta\epsilon^2}{2\Delta p^2}\right]\int g(r)\nabla_r{}^2\phi(\mathbf{r})d^3r+O(\hbar^4). \tag{57}$$

We thus obtain explicitly the factor $\exp(\beta\epsilon/2)$ essential to satisfy the condition of detailed balance. Equation (57) is our sought for connection between $S_s(\Delta\mathbf{p},\epsilon)$ and $G_s{}^c(\mathbf{r},t)$. The first term in this expression is the form suggested by Singwi and Sjölander,[20] who speculated that it might be correct because it works exactly for the ideal gas. It differs from the Vineyard prescription by the factor

$$\exp[\beta\epsilon/2]\exp[-\beta\Delta p^2/8M].$$

The order of magnitude of this correction is illustrated in Figs. 1–4, where a comparison is made between the direct neutron differential scattering cross section for some simple systems, as calculated by the Vineyard prescription, and the cross section obtained from the first term in (57).[12] The differences are seen to be significant, particularly for high incident energy, large momentum transfers and light scatterers. This, of course, is due to the form of the missing factor.

[20] K. S. Singwi and A. Sjölander, Phys. Rev. **120**, 1093 (1960).

For sufficiently high temperatures the second term in (57) is negligible and may be expressed in terms of the deviation of the free energy from its classical value. Accordingly, we get the following alternate result:

$$S_s(\Delta\mathbf{p},\epsilon)=\exp\left[\frac{\beta\epsilon}{2}\right]\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\left\{S_s{}^c(\Delta\mathbf{p},\epsilon)+\frac{\beta}{6}\left(\frac{\beta M}{2\pi\Delta p^2}\right)^{1/2}(F-F_c)\left(1-\frac{\epsilon^2M\beta}{\Delta p^2}+\frac{\Delta p^2\beta}{8M}\right)\exp\left[-\frac{M\beta\epsilon^2}{2\Delta p^2}\right]\right\}+O(\hbar^4). \quad (58)$$

Detailed Balance and Placzek Moments. Because of the symmetry condition [Eq. (44)] on $G_s{}^c(\mathbf{r},t)$, $\exp[-\beta\epsilon/2]\times S_s(\Delta\mathbf{p},\epsilon)$ in (57) is invariant [at least to $O(\hbar^2)$] when interchanging the initial and final states of the neutron, i.e.,

$$\exp[-\beta\epsilon/2]S_s(\Delta\mathbf{p},\epsilon)=\exp[\beta\epsilon/2]S_s(-\Delta\mathbf{p},-\epsilon). \quad (59)$$

Consequently, the condition (46) and its corollary, the principle of detailed balance, are satisfied. Furthermore, as previously indicated, Eq. (57) was derived essentially by adding a given quantity to the first term in (35) and subtracting the same quantity from the second term. Therefore, Eqs. (57) and (58) will also satisfy the Placzek moments [Eqs. (38)] to $O(\hbar^2)$.

F. Other Prescriptions

In the light of the above analysis, it is possible to critically examine various other "prescriptions."

(*1*) *Schofield's prescription.* From the observation that the time correlation function $F(\mathbf{r},t)$, defined by

$$F(\mathbf{r},t)=\left(\frac{1}{2\pi\hbar}\right)^3\int d\Delta\mathbf{p}\,\exp\left[-\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{r}\right]\chi_s\left(\Delta\mathbf{p},\,\hbar\tau+\frac{i\beta\hbar}{2}\right) \quad (60)$$

is real and that its double Fourier transform satisfies the condition (46), Schofield[5] suggested that this function

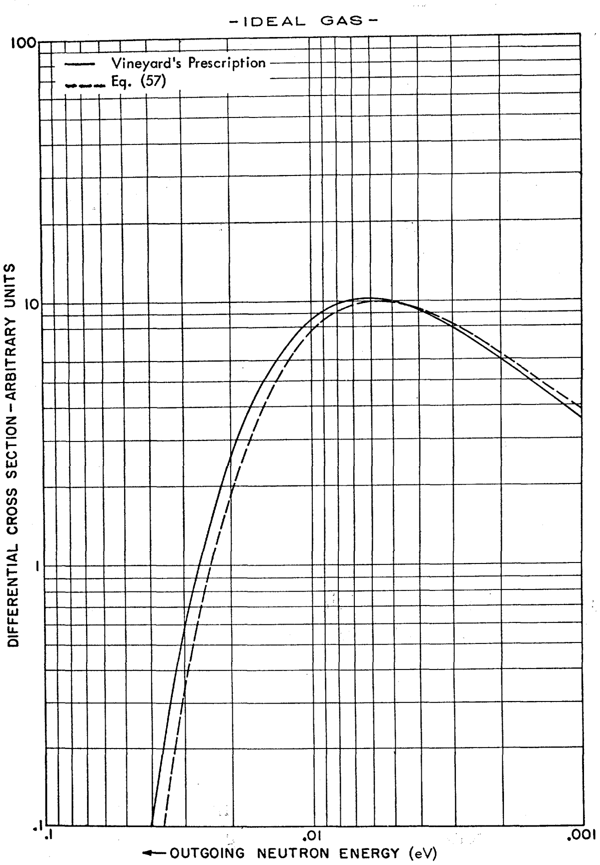

FIG. 1. Differential scattering cross section versus outgoing neutron energy for neutrons of incident energy 5×10^{-3} eV scattered at 90° by an ideal gas of mass 18 at 295°K.

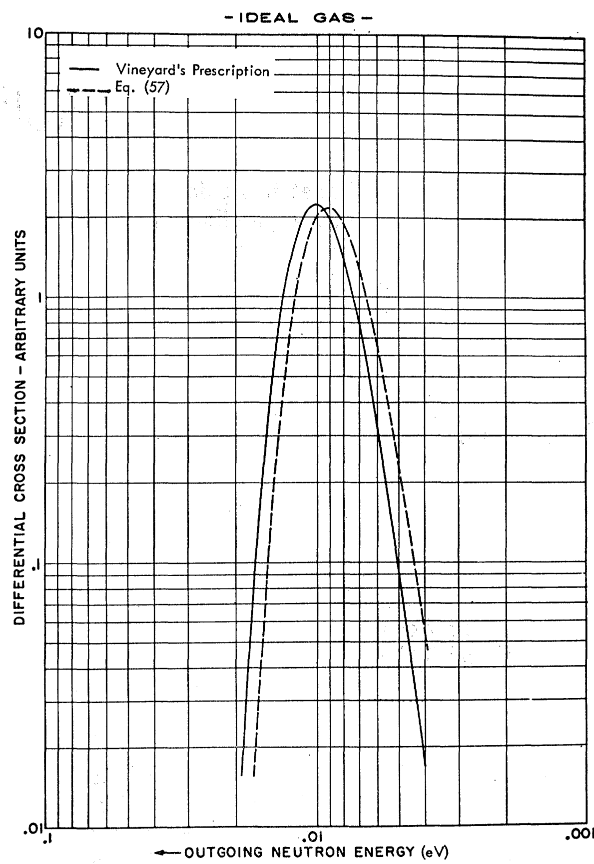

FIG. 2. Differential scattering cross section versus outgoing neutron energy for neutrons of incident energy 0.1 eV scattered at 90° by an ideal gas of mass 18 at 295°K.

be made equal to $G_s^c(\mathbf{r},t)$. This leads to an expression for $S_s(\Delta\mathbf{p},\epsilon)$ which, with the exception of the factor $\exp[-\beta\Delta p^2/8M]$, is equal to the first term in (57). Therefore, this approximation will be valid only for small momentum transfers and heavy scatterers.

(2) Letting $S_s(\Delta\mathbf{p},\epsilon)\approx S_s^c[\Delta\mathbf{p},\ \epsilon-(\Delta p^2/2M)]$.[21] Since (57) is an asymptotic expansion, clearly it will not be unique. In fact, if instead of integrating

$$\langle\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\nabla_{pj}]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]\rangle_{TC} \tag{61}$$

by parts in (35), $\mathbf{q}_j(\hbar\tau)$ is formally expanded in a Taylor series and is operated on by $\exp[\frac{1}{2}\Delta\mathbf{p}\cdot\nabla_{pj}]$, it can be shown that

$$\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\nabla_{pj}]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]=\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]\exp[i(\Delta p)^2\tau/2M]$$
$$\times\exp[-(i\hbar^2\tau^3/12M^2)(\Delta\mathbf{p}\cdot\nabla_q)(\Delta\mathbf{p}\cdot\nabla_{qj}V)][1+J(\tau,\mathbf{q},\Delta\mathbf{p})], \tag{62}$$

where $J(\tau,\mathbf{q},\Delta\mathbf{p})$ is of $O(\hbar^4)$. In this case, the thermal average (61) becomes

$$\langle\exp[-(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j]\exp[\tfrac{1}{2}\Delta\mathbf{p}\cdot\nabla_{pj}]\exp[(i/\hbar)\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)]\rangle_{TC}$$
$$=\exp\left[\frac{i\Delta p^2\tau}{2M}\right]\left\langle\exp\left[-\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{q}_j\right]\exp\left[\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)\right]\right\rangle_{TC}-\frac{i\hbar^2\tau^3}{12M^2}\exp\left[\frac{i\Delta p^2\tau}{2M}\right]$$
$$\times\left\langle\exp\left[-\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{q}_j\right]\exp\left[\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)\right](\Delta\mathbf{p}\cdot\nabla_q)(\Delta\mathbf{p}\cdot\nabla_{qj}V)\right\rangle_{TC}+O(\hbar^4)$$
$$=\exp\left[\frac{i\Delta p^2\tau}{2M}\right]\left\langle\exp\left[-\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{q}_j\right]\exp\left[\frac{i}{\hbar}\Delta\mathbf{p}\cdot\mathbf{q}_j(\hbar\tau)\right]\right\rangle_{TC}$$
$$-\frac{i\hbar^2\tau^3\Delta p^2}{36M^2}\exp\left[\frac{i\Delta p^2\tau}{2M}\right]\exp\left[-\frac{\tau^2\Delta p^2}{2M\beta}\right]\int g(r)\nabla_r^2\phi(\mathbf{r})d^3r+O(\hbar^4) \tag{63}$$

and

$$S_s(\Delta\mathbf{p},\epsilon)=S_s^c\left(\Delta\mathbf{p},\ \epsilon-\frac{\Delta p^2}{2M}\right)-4\left(\frac{\hbar\beta}{12}\right)^2\left(\frac{\beta}{2\pi M\Delta p^2}\right)^{1/2}\left\{\frac{\epsilon M}{\Delta p^2}\left[3-\frac{M\beta}{\Delta p^2}\left(\epsilon-\frac{\Delta p^2}{2M}\right)^2\right]-1\right\}$$
$$\times\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\exp\left[\frac{\beta\epsilon}{2}\right]\exp\left[-\frac{\epsilon^2M\beta}{2\Delta p^2}\right]\int g(r)\nabla_r^2\phi(\mathbf{r})d^3r+O(\hbar^4). \tag{64}$$

Although Eq. (64) is also correct to $(\hbar^2)$ and satisfies the Placzek moments to this order, it differs from Eq. (57) in that, because of the way the terms are grouped, it does not satisfy the condition (46). This makes (57) preferable.

(3) y^2 *Time Approximation.* Based on the fact that Schofield's prescription does not satisfy the zeroth Placzek moment, it was suggested by Egelstaff[22] and Schofield[23] that, for an isotropic system, $\chi_s(\Delta\mathbf{p},t)$ may be obtained from $\chi_s^c(\Delta\mathbf{p},t)$ by replacing t^2 by $y^2=t-i\hbar t\beta$. In order to establish connection between this recipe and the quasiclassical approximation, note that for a randomly oriented system, $\chi_s^c(\Delta\mathbf{p},t)$ is real and is given by

$$\chi_s^c(\Delta\mathbf{p},t)=\chi_s^c(|\Delta\mathbf{p}|,t)=\langle\exp[(i\Delta p/\hbar)(z_j(t)-z_j)]\rangle_{TC}$$
$$=\left\langle\cos\left[\frac{\Delta p}{\hbar}(z_j(t)-z_j)\right]\right\rangle_{TC}=\sum_{n=0}^{\infty}\frac{(-1)^n}{(2n)!}\left(\frac{\Delta p}{\hbar}\right)^{2n}\langle[z_j(t)-z_j]^{2n}\rangle_{TC}, \tag{65}$$

where z_j is the component of $\mathbf{q}_j$ along the direction of $\Delta\mathbf{p}$, and $\Delta\mathbf{p}$ is chosen along the z axis. Hence, the formal expansion in powers of Δp^2,

$$\ln\left\{\chi_s^c(\Delta\mathbf{p},t)+\left(\frac{\tau^2}{\beta}+\frac{\beta}{8}\right)\left(\frac{\hbar\beta\Delta p}{12M}\right)^2\exp\left[-\frac{\tau^2\Delta p^2}{2M\beta}\right]\langle\nabla_j^2V\rangle\right\}=\sum_{n=0}^{\infty}\left(\frac{\Delta p^2}{\hbar^2}\right)^n\frac{C_n(t)}{n!}, \tag{66}$$

[21] M. Nelkin, in *Proceedings of the Symposium on Inelastic Scattering of Neutrons in Solids and Liquids* (International Atomic Energy Agency, Vienna, 1960), p. 3.

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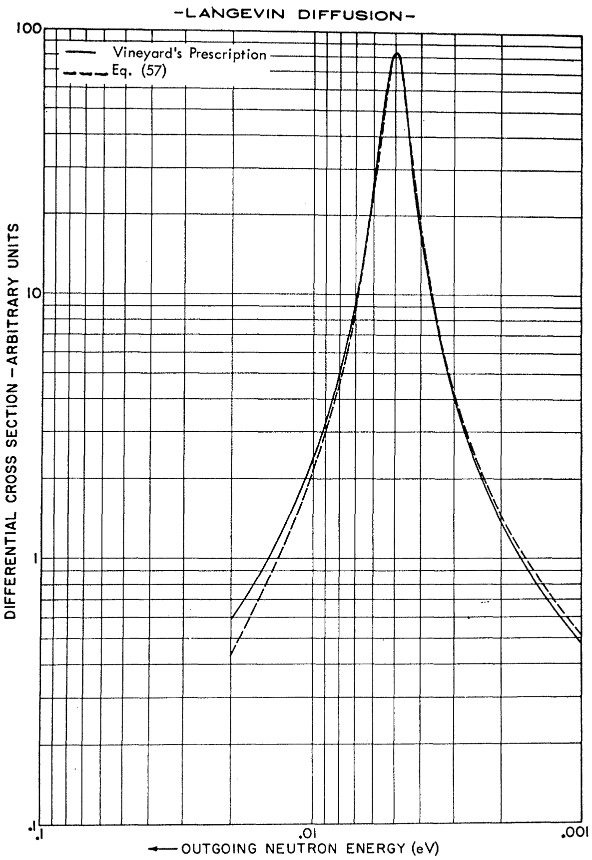

FIG. 3. Differential scattering cross section versus outgoing neutron energy for neutrons of incident energy 5×10^{-3} eV scattered at 90° by a system of particles of mass 18 diffusing according to the Langevin model at 295°K.

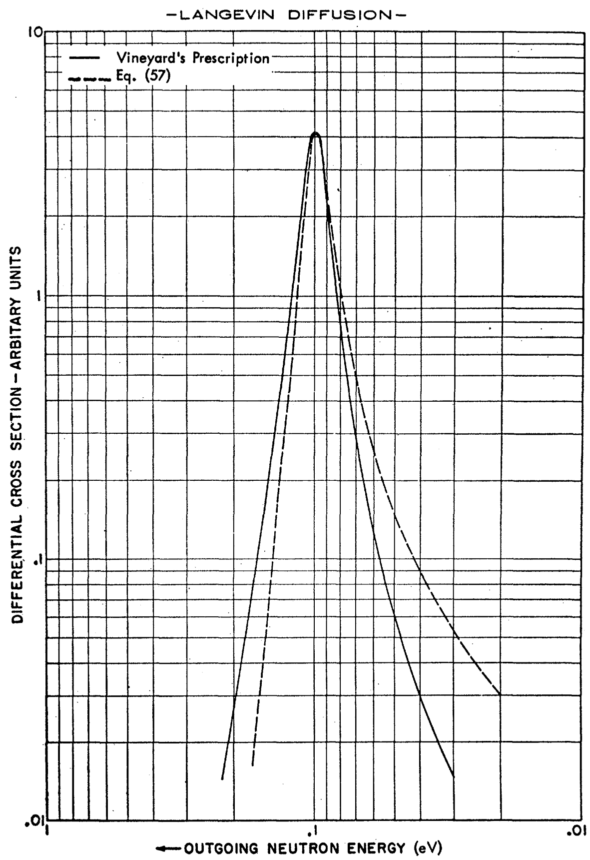

FIG. 4. Differential scattering cross section versus outgoing neutron energy for neutrons of incident energy 0.1 eV scattered at 90° by a system of particles of mass 18 diffusing according to the Langevin model at 295°K.

is justified. The coefficients $C_n(t)$ may be evaluated from (65) by noting that

$$C_n(t)=\hbar^{2n}\frac{d^n}{d(\Delta p^2)^n}\ln\left\{\chi_s{}^c(\Delta\mathbf{p},t)+\left(\frac{\tau^2}{\beta}+\frac{\beta}{8}\right)\left(\frac{\hbar\beta\Delta p}{12M}\right)^2\exp\left[-\frac{\tau^2\Delta p^2}{2M\beta}\right]\langle\nabla_j{}^2V\rangle\right\}\Bigg|_{\Delta p^2=0} \tag{67}$$

yielding

$$C_0(t)=0$$

and

$$C_1(t)=-\frac{1}{2}\left[\frac{t^2}{M\beta}-\frac{1}{36\beta M^2}\left(t^2+\frac{\hbar^2\beta^2}{4}\right)^2\langle\nabla_j{}^2V\rangle_{TC}+\frac{1}{360}\frac{t^6}{\beta M^3}\left\langle\left(\frac{\partial^2V}{\partial z\partial z_j}\right)^2\right\rangle_{TC}+O(t^8)\right] \tag{68}$$

for the first two terms. Consequently, Eq. (54) becomes

$$\ln\chi_s\left(\Delta\mathbf{p},\,\hbar\tau+\frac{i\beta\hbar}{2}\right)=-\frac{\Delta p^2}{2\hbar^2\beta M}\left[\hbar^2\left(\tau^2+\frac{\beta^2}{4}\right)-\frac{\hbar^4}{36M}\left(\tau^2+\frac{\beta^2}{4}\right)^2\langle\nabla_j{}^2V\rangle_{TC}\right.$$
$$\left.+\frac{\hbar^6}{360M^2}\left(\tau^2+\frac{\beta^2}{4}\right)^3\left\langle\left(\frac{\partial^2V}{\partial z\partial z_j}\right)^2\right\rangle_{TC}+O(\tau^2\hbar^6)\right]+O(\Delta p^4) \tag{69}$$

or

$$\ln\chi_s(\Delta\mathbf{p},\hbar\tau)=-\frac{\Delta p^2}{2\hbar^2\beta M}\left[y^2-\frac{1}{36M}y^4\langle\nabla_j{}^2V\rangle_{TC}+\frac{y^6}{360M^2}\left\langle\left(\frac{\partial^2V}{\partial z\partial z_j}\right)^2\right\rangle_{TC}+O(\tau^2\hbar^6)\right]+O(\Delta p^4). \tag{70}$$

The coefficient of Δp^2 on the right of the above equation is, except for correction terms of $O(\hbar^2 t^2)$, indeed equal to the coefficient of Δp^2 in the series expansion in powers of Δp^2 of $\ln\chi_s^c(|\Delta\mathbf{p}|,y)$.

Note, however, that the above argument does not constitute a proof of the validity of the y^2 approximation. This would require showing a one to one correspondence [at least to terms of $O(\hbar^2)$] between the coefficients of all powers of Δp^2 in the series for $\ln\chi_s(|\Delta\mathbf{p}|,t)$ and $\ln\chi_s^c(|\Delta\mathbf{p}|,y)$. Such a proof is lacking and even if the y^2 approximation were valid it would be at best correct to the same order as the first term in Eq. (54). These facts make Eq. (54) a superior choice.

(4) The Classical Limit of $S_s(\Delta\mathbf{p},\epsilon)$. In order to investigate the limiting behavior of $S_s(\Delta\mathbf{p},\epsilon)$ as $\hbar\rightarrow 0$, it is convenient to expand $\chi_s^c(\Delta\mathbf{p},\hbar\tau)$ in Eq. (55) in a power series in $\hbar\tau$. Thus,

$$\chi_s^c(\Delta\mathbf{p},\hbar\tau)=\left\langle\exp\left[\frac{i\tau}{M}\Delta\mathbf{p}\cdot\mathbf{p}_j\right]\right\rangle_{TC}-\frac{i\hbar\tau^2}{2M}\left\langle\Delta\mathbf{p}\cdot\nabla_{q_j}V\exp\left[\frac{i\tau}{M}\Delta\mathbf{p}\cdot\mathbf{p}_j\right]\right\rangle_{TC}$$
$$-\frac{i\hbar^2\tau^3}{6M^2}\left\langle(\mathbf{p}\cdot\nabla_q)(\Delta\mathbf{p}\cdot\nabla_{q_j}V)\exp\left[\frac{i\tau}{M}\Delta\mathbf{p}\cdot\mathbf{p}_j\right]\right\rangle_{TC}-\frac{\hbar^2\tau^4}{8M^2}\left\langle(\Delta\mathbf{p}\cdot\nabla_{q_j}V)^2\exp\left[\frac{i\tau}{M}\Delta\mathbf{p}\cdot\mathbf{p}_j\right]\right\rangle_{TC}+O(\hbar^3). \quad (71)$$

Performing the indicated thermal averages gives

$$\chi_s^c(\Delta\mathbf{p},\hbar\tau)=\exp\left[-\frac{\tau^2\Delta p^2}{2M\beta}\right]+\frac{\hbar^2\tau^4\Delta p^2}{72\beta M^2}\exp\left[-\frac{\tau^2\Delta p^2}{2M\beta}\right]\int g(r)\nabla_r^2\phi(\mathbf{r})d^3r+O(\hbar^4). \quad (72)$$

Substituting this result into Eq. (57) yields

$$S_s(\Delta\mathbf{p},\epsilon)=\left(\frac{M\beta}{2\pi\Delta p^2}\right)^{1/2}\exp\left[\frac{\beta\epsilon}{2}\right]\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\exp\left[-\frac{\beta M\epsilon^2}{2\Delta p^2}\right]$$
$$\times\left\{1+\left[\frac{M}{2\beta\Delta p^2}H_4(E)-\frac{1}{4}\left(H_2(E)-\frac{\Delta p^2\beta}{8M}\right)\right]\frac{\hbar^2\beta^2}{36M}\int g(r)\nabla_r^2\phi(\mathbf{r})d^3r\right\}+O(\hbar^4), \quad (73)$$

where

$$E=(\epsilon^2M\beta/\Delta p^2)^{1/2},\quad H_2(E)=E^2-1,\quad H_4(E)=E^4-6E^2+3. \quad (74)$$

From Eq. (74) it immediately follows that

$$\lim_{\hbar\rightarrow 0}S_s(\Delta\mathbf{p},\epsilon)=\left(\frac{M\beta}{2\pi\Delta p^2}\right)^{1/2}\exp\left[\frac{\beta\epsilon}{2}\right]\exp\left[-\frac{\beta\Delta p^2}{8M}\right]\exp\left[-\frac{\beta M\epsilon^2}{2\Delta p^2}\right]. \quad (75)$$

That is, the exact classical limit of any system, defined in this way, is the ideal-gas result. This is physically understandable since classically the neutron-nuclear collision is instantaneous; thus the neutron never samples the potential which binds the scattering system (since the Fermi pseudopotential is a contact potential). In fact, since the quantum mechanical corrections in (73) contain the factor β, the idealization to a monatomic gas is not far from reality at sufficiently high temperatures. The rapidity of convergence to this asymptotic behavior is determined by the factor $(M/\beta\Delta p^2)H_4(E)$, and therefore increases with increasing momentum transfers.

APPENDIX A

In this appendix we prove the identity[24]

$$\phi=e^{\alpha(A+B)}=e^{\alpha B}e^{\alpha A}\Gamma(\alpha), \quad (A1)$$

[24] R. K. Osborn (private communication).

where A, B are arbitrary operators independent of α, and $\Gamma(\alpha)$ is defined by the differential equation

$$\frac{\partial\Gamma(\alpha)}{\partial\alpha}=e^{-\alpha A}\sum_{n=1}^{\infty}\frac{(-1)^n\alpha^n}{n!}[B,A]_n e^{\alpha A}\Gamma(\alpha) \quad (A2)$$

and the initial condition

$$\Gamma(\alpha=0)=1. \quad (A3)$$

To this end, we differentiate (A1) with respect to α to get

$$Ae^{\alpha B}e^{\alpha A}\Gamma(\alpha)=e^{\alpha B}Ae^{\alpha A}\Gamma(\alpha)+e^{\alpha B}e^{\alpha A}(\partial\Gamma/\partial\alpha). \quad (A4)$$

Multiplying both sides of (A4) by $e^{-\alpha A}e^{-\alpha B}$, and noting that

$$e^{-\alpha B}Ae^{\alpha B}=\sum_{n=0}^{\infty}\frac{(-1)^n}{n!}[B,A]_n, \quad (A5)$$

where

$$[B,A]_n=[B,[B,A]_{n-1}]$$

and

$$[B,A]_0=A\,,$$

readily yields Eq. (A2). Q. E. D.

For the case that A and B each commute with the commutator $[A,B]$, Eq. (A2) simplifies to

$$\partial\Gamma/\partial\alpha=-\alpha[B,A]\Gamma$$

or

$$\Gamma=\exp\{-\tfrac{1}{2}\alpha^2[B,A]\}\Gamma(\alpha=0)=\exp\{-\tfrac{1}{2}\alpha^2[B,A]\}\,,$$

i.e.,

$$e^{(A+B)}=e^Be^A\exp\{\tfrac{1}{2}[A,B]\}\,. \qquad \text{(A6)}$$

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Polarization of Protons Elastically Scattered by Oxygen*

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Measurements of the polarization of protons elastically scattered by oxygen have been made between 2 and 12 MeV by double scattering using gas targets. Scattering by helium at 46° was used as the polarization analyzer for most of the work. In addition, four angular distributions of the cross section were measured between 2.5 and 3.8 MeV. The experimental results show that away from the known sharp resonances at 2.66 and 3.47 MeV the polarization changes slowly with energy between 2 and 5 MeV. At 3 MeV the polarization is negative at forward angles and positive at back angles, the observed extrema being -0.14 ± 0.02 at 45° and 0.43 ± 0.03 at 115°. At higher energies the pronounced resonance structure causes rapid fluctuations of the polarization with energy. Angular distributions of the polarization were measured at eleven energies away from sharp resonances. At 10.7 MeV the observed extrema in the polarization are -0.88 ± 0.04 at 50°, 0.92 ± 0.04 at 65°. and -0.83 ± 0.03 at 133°. A phase shift analysis of the polarization and cross section was made between 2 and 5 MeV and at four energies between 5 and 7 MeV. The phase shifts obtained differ from those of Salisbury and Richards by less than 10 deg. The measured angular distributions of the polarization between 8 and 12 MeV are also compared with the predictions of the optical model. New results of the polarization for p-α scattering and p-C scattering between 2 and 4 MeV are also reported.

1. INTRODUCTION

PREVIOUS studies of the elastic scattering of protons by oxygen consisted primarily of measurements of the differential cross section as a function of energy.[1–5] At Wisconsin excitation curves of the cross section have been measured for several angles. Eppling[2] made measurements up to 4.6 MeV, Salisbury *et al.*[3] from 4.2 to 8.6 MeV, and Hardie *et al.*[4] from 8.5 to 13 MeV. In addition, Hardie measured angular distributions at thirteen energies between 4.8 and 13 MeV. Phase-shift analyses of cross-section data have been reported between 2.5 and 5.2 MeV by Harris *et al.*[5] and between 2 and 7.6 MeV by Salisbury and Richards.[6]

The polarization can, in principle, be calculated from the phase shifts, but these calculations are seldom reliable since the polarization and the cross section depend on the phase shifts in different ways. For this reason measurements of the polarization provide an independent test of the validity of the phase shifts obtained by fitting cross-section data and can be used in conjunction with cross-section data for a more accurate determination of the phase shifts. Indeed, at higher energies where there are more parameters to be determined, cross-section data alone is not sufficient to determine the phase shifts unambiguously.

Previous measurements of the polarization for proton-oxygen scattering are not very extensive. Early measurements by Sorokin *et al.*[7] near 2.7 MeV and Al-Jeboori

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