Quasi-Classical Treatment of Neutron Scattering*

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The classical limit of the neutron-scattering cross section for a general system is investigated. It is shown that the exact limit for the "self-function" is that for an ideal gas. An improved prescription, which utilizes classical quantities and which has been suggested earlier, is justified.

I. INTRODUCTION

It has been shown by Glauber and Van Hove[1] that the neutron-scattering cross section for an arbitrary system may be expressed in terms of a function $S(\mathbf{p},E)$ ($\mathbf{p},E$=momentum, energy transfer, respectively). Explicitly,

$$S(\mathbf{p},E)=\frac{N}{2\pi\hbar}\int d\mathbf{r}\int dt\,\exp\left[\frac{i}{\hbar}(\mathbf{p}\cdot\mathbf{r}-Et)\right]G(\mathbf{r},t), \quad (1)$$

and

$$G(\mathbf{r},t)=\int\frac{d\mathbf{p}}{(2\pi\hbar)^3}\exp\left(-\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)\times\mathrm{Tr}\left\{\frac{1}{N}\sum_{i,j=1}^{N}\rho\exp\left[-\frac{i}{\hbar}\mathbf{p}\cdot\mathbf{r}_i(0)\right]\times\exp\left[\frac{i}{\hbar}\mathbf{p}\cdot\mathbf{r}_j(t)\right]\right\}, \quad (2)$$

where $\mathbf{r}_j(t)$ is the Heisenberg position operator of scatterer j at time t and ρ is the density matrix of the scattering system (which contains N scatterers).

Vineyard[2] has suggested a "classical" approximation obtained from Eq. (2) by replacing the operators by corresponding classical variables. This approximation has, however, two unsatisfactory features:

(a) Recoil effects are inadequately treated in that the average energy loss is set equal to zero rather than the exact value $\mathbf{p}^2/2M$ (M=scatterer mass).

(b) As shown by Schofield,[3] detailed balance is not satisfied. Schofield has suggested a recipe to remedy this defect which Turner[4] has attempted to justify. This we feel is inadequate, however, since it uses "Weyl's rule" for Heisenberg operators—for which it does not generally hold—and because it attempts to expand a function in powers of $\hbar$ about an essential singularity.

The observation that S for an ideal gas is (in terms of the significant variables $\mathbf{p}$ and E) actually independent of $\hbar$ suggests that a well-defined classical limit for S exists which (1) does not suffer from the same difficulties as Vineyard's approximation, and (2) serves as a satisfactory zeroth approximation from which quantum corrections can be obtained by expansion in a power series in $\hbar$.

II. DERIVATION

This sequence of approximation has been obtained by introducing a Wigner representation.[5] Let

$$U_j(\mathbf{p},t)=\rho\exp\left(-\frac{i}{\hbar}\mathbf{p}\cdot\mathbf{r}_j(-t)\right), \quad (3)$$

where

$$\rho=\exp(-\beta H)/\mathrm{Tr}\exp(-\beta H), \quad (4)$$

and

$$\exp\left(-\frac{i}{\hbar}\mathbf{p}\cdot\mathbf{r}_j(-t)\right)=\exp\left(-\frac{iHt}{\hbar}\right)\exp\left(-i\frac{\mathbf{p}}{\hbar}\cdot\mathbf{r}_j(0)\right)\exp\left(\frac{iHt}{\hbar}\right).$$

Then Eq. (2) becomes

$$G(\mathbf{r},t)=\int\frac{d\mathbf{p}}{(2\pi\hbar)^3}\exp\left(-\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)\frac{1}{N}\times\sum_{i,j}\left\{\mathrm{Tr}\exp\left[\frac{i\mathbf{p}\cdot\mathbf{r}_i(0)}{\hbar}\right]U_j(\mathbf{p},t)\right\}. \quad (5)$$

Using a coordinate representation with

$$|\mathbf{R}\rangle\equiv|\mathbf{R}_1,\mathbf{R}_2,\cdots,\mathbf{R}_N\rangle, \quad (6)$$

we see that

$$\mathrm{Tr}\left\{\exp\left[\frac{i\mathbf{p}\cdot\mathbf{r}_i(0)}{\hbar}\right]U_j\right\}=\int d\mathbf{R}\exp\left(\frac{i}{\hbar}\mathbf{p}\cdot\mathbf{R}_i\right)\langle\mathbf{R}|U_j|\mathbf{R}\rangle. \quad (7)$$

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[1] L. Van Hove, Phys. Rev. **95**, 249 (1954); R. J. Glauber, *ibid.* **98**, 1692 (1955).
[2] G. H. Vineyard, Phys. Rev. **110**, 999 (1958).
[3] P. Schofield, Phys. Rev. Letters **4**, 239 (1960).
[4] R. E. Turner, Physica **27**, 260 (1961).
[5] See, e.g., J. H. Irving and R. W. Zwanzig, J. Chem. Phys. **19**, 1173 (1951).

The Wigner representation is introduced by Fourier transformation. Thus, let

$$f_j(\mathbf{P},\mathbf{R},t;\,\mathbf{p})=\frac{1}{(2\pi\hbar)^{3N}}\int d^N\mathbf{R}'\exp\left(-\frac{i\mathbf{P}\cdot\mathbf{R}'}{\hbar}\right)\times\langle\mathbf{R}+\tfrac{1}{2}\mathbf{R}'|U_j(\mathbf{p},t)|\mathbf{R}-\tfrac{1}{2}\mathbf{R}'\rangle, \quad (8)$$

where the symbols $\mathbf{P}$ and $\mathbf{R}$ represent the set of classical momenta and position vectors $\{\mathbf{P}_i\}$, $\{\mathbf{R}_i\}$; $i=1,2,\cdots,N$. Then

$$G(\mathbf{r},t)=\int\frac{d\mathbf{p}}{(2\pi\hbar)^3}\exp\left(-\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)\frac{1}{N}\sum_{i,j}\int d^N\mathbf{R}\,d^N\mathbf{P}\times\exp\left(\frac{i\mathbf{p}\cdot\mathbf{R}_i}{\hbar}\right)f_j(\mathbf{P},\mathbf{R},t;\,\mathbf{p}). \quad (9)$$

As

$$i\hbar\partial U_j/\partial t=[H,U_j], \quad (10)$$

it readily follows that the f_j satisfy the equations

$$\left\{\frac{\partial}{\partial t}-\frac{2}{\hbar}\sin\left[\frac{\hbar}{2}\sum_{i=1}^{N}\left(\frac{\partial}{\partial P_{if}}\frac{\partial}{\partial R_{iH}}-\frac{\partial}{\partial R_{if}}\frac{\partial}{\partial P_{iH}}\right)\right]H(\mathbf{P},\mathbf{R})\right\}\times f_j(\mathbf{P},\mathbf{R},t;\,\mathbf{p})=0, \quad (11)$$

with the initial conditions

$$f_j(\mathbf{P},\mathbf{R},0;\,\mathbf{p})=\exp(-i\mathbf{p}\cdot\mathbf{R}_j/\hbar)\times\exp(-\mathbf{p}\cdot\nabla_{\mathbf{P}j}/2)\rho_w(\mathbf{P},\mathbf{R}). \quad (12)$$

Here ρ_w is the Wigner distribution function[6] and the subscripts H and f indicate which functions are to be differentiated.

Equation (11) is, to terms of order $\hbar^2$, the classical Liouville equation. To the same order ρ_w is the Maxwell-Boltzmann distribution. Then to lowest order we obtain G in terms of the classical solutions of the classical equations of motion as:

$$G(\mathbf{r},t)\equiv\int\frac{d\mathbf{p}}{(2\pi\hbar)^3}\exp\left(-\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)\exp\left(-\frac{\beta\mathbf{p}^2}{8M}\right)\times\left\langle\frac{1}{N}\sum_{i,j}\exp\left\{\frac{i\mathbf{p}}{\hbar}\cdot[\mathbf{R}_i(t)-R_j(0)]\right\}\times\exp\left[\frac{\beta\mathbf{p}\cdot\mathbf{P}_j(0)}{2M}\right]\right\rangle_{\mathrm{TC}}. \quad (13)$$

where $\langle\ \rangle_{\mathrm{TC}}$ denotes the classical thermal average. The integral over $\mathbf{p}$ can be performed, yielding

$$G(\mathbf{r},t)=\frac{1}{N}\sum_{i,j}\langle(2M/\pi\hbar^2\beta)^{\frac{3}{2}}\exp(-2M\mathbf{s}^2/\beta\hbar^2)\rangle_{\mathrm{TC}}, \quad (14)$$

where

$$\mathbf{s}=\mathbf{r}+\mathbf{R}_j(0)-\mathbf{R}_i(t)+i\hbar\mathbf{P}_j(0)\beta/2M. \quad (15)$$

[6] H. Weyl, *The Theory of Groups and Quantum Mechanics* (Dover Publications, New York, 1950), p. 275; H. J. Groenwald, Physica **12**, 405 (1946).

The limit of this formula as $\hbar\rightarrow 0$ is the Vineyard result, i.e.,

$$\lim_{\hbar\rightarrow 0}G(\mathbf{r},t)=\frac{1}{N}\sum_{i,j}\langle\delta[\mathbf{r}+\mathbf{R}_j(0)-\mathbf{R}_i(t)]\rangle_{\mathrm{TC}}. \quad (16)$$

From Eqs. (1) and (13) we find

$$S(\mathbf{p},E)=\frac{N}{2\pi\hbar}\int_{-\infty}^{\infty}dt\exp\left(-\frac{iEt}{\hbar}\right)\exp\left(-\frac{\beta\mathbf{p}^2}{8M}\right)\times\frac{1}{N}\sum_{i,j=1}^{N}\left\langle\exp\left\{\frac{i\mathbf{p}}{\hbar}\cdot[(\mathbf{R}_j(t)-\mathbf{R}_i(0)]\right\}\times\exp\left[\frac{\beta\mathbf{p}}{2M}\cdot\mathbf{P}_j(0)\right]\right\rangle_{\mathrm{TC}}. \quad (17)$$

Connection with Schofield's conjecture[3] is made by noting that through terms which vanish with $\hbar$ the argument of the exponential to be averaged in (17) is

$$(i/\hbar)\mathbf{p}\cdot[\mathbf{R}_j(t)-\mathbf{R}_i(i\hbar\beta/2)]. \quad (18)$$

Utilizing time translational invariance we can then write Eq. (17) as

$$S(\mathbf{p},E)=\frac{N}{2\pi\hbar}\int_{-\infty}^{\infty}dt\exp\left(-\frac{iEt}{\hbar}\right)\exp\left(-\frac{\beta\mathbf{p}^2}{8M}\right)\times\frac{1}{N}\sum_{i,j}\left\langle\exp\left\{\frac{i\mathbf{p}}{\hbar}\left[\mathbf{R}_j\left(t-\frac{i\hbar\beta}{2}\right)-\mathbf{R}_i(0)\right]\right\}\right\rangle_{\mathrm{TC}}. \quad (19)$$

Except for the factor $\exp(-\beta\mathbf{p}^2/8M)$, this is Schofield's result.

Thus[7]

$$S(\mathbf{p},E)=\exp\left(\frac{\beta E}{2}\right)\exp\left(-\frac{\beta\mathbf{p}^2}{8M}\right)S(\mathbf{p},E)_V, \quad (19a)$$

where $S(\mathbf{p},E)_V$ is related, through Eq. (1), to Vineyard's approximation for $G(\mathbf{r},t)$; i.e., $S(\mathbf{p},E)_V$ is $N/2\pi\hbar$ times the four dimensional Fourier transform of Vineyard's "classical" approximation to $G(\mathbf{r},t)$, [Eq. (16)].

III. DISCUSSION

The essential point here is that Eq. (19), which is in practice as simple as Vineyard's approximation, does not imply zero momentum transfer and does satisfy the requirement of detailed balance. [It may be noted that the rigorous classical limit here of the "self terms" ($i=j$) is exactly the correct ideal gas result]. The difference with respect to Vineyard is just that we have kept $\mathbf{p}$ and E finite and second passed to the limit $\hbar\rightarrow 0$ in Eq. (1)—not passing to the limit in Eq. (2) and then inserting the result in Eq. (1).

[7] This form has been suggested by K. S. Singwi and A. Sjolander, Phys. Rev. **120**, 1093 (1960); See also P. Schofield, *Proceedings of the Symposium on Slow Neutron Scattering, Vienna, 1960* [International Atomic Energy Agency (to be published)].

IV. ALTERNATE TREATMENT

Another derivation of the above result, more along the lines of Turner's work,[4] begins with the "intermediate" scattering function

$$\chi(\mathbf{p},t)=\int \exp\left(\frac{i\mathbf{p}\cdot\mathbf{r}}{\hbar}\right)G(\mathbf{r},t)d\mathbf{r}$$

$$\equiv N^{-1}\,\mathrm{Tr}\{\rho\sum_{i,j}\psi_{ji}\},\tag{20}$$

where

$$\psi_{ji}=\exp\left[-\frac{i\mathbf{p}\cdot\mathbf{r}_j(0)}{\hbar}\right]\exp\left[\frac{i\mathbf{p}\cdot\mathbf{r}_i(t)}{\hbar}\right].\tag{21}$$

Let

$$\Phi_{ji}=\exp\left(-\frac{it\mathbf{p}^2}{2M\hbar}\right)\exp\{-i\boldsymbol{\kappa}\cdot[\mathbf{r}_j(0)-\mathbf{r}_i(0)]\}\psi_{ji},\tag{22}$$

where

$$\boldsymbol{\kappa}=\mathbf{p}/\hbar.\tag{23}$$

Wick[8] has obtained the expansion

$$\Phi_{ji}=\sum_{n=0}^{\infty}\frac{1}{n!}\left(\frac{it}{\hbar}\right)^n g_n(\mathbf{p}).\tag{24}$$

Here the $g_n(\mathbf{p})$ satisfy the recursion relations

$$g_{n+1}=g_nL+ng_{n-1}[H,L]+\tfrac{1}{2}(n)(n-1)g_{n-2}[H,[H,L]]+\cdots\tag{25}$$

where

$$L=\mathbf{p}\cdot\mathbf{P}_j(0)/M\quad\text{and}\quad g_0=1.\tag{26}$$

[$\mathbf{P}_j(t)$ = Heisenberg momentum operator conjugate to $\mathbf{r}_j(t)$.]

Having expressed Φ_{ji} in the form of Eq. (24), we can now apply Weyl's rule[6] term by term. The resulting function Φ_{ji}^c is one which, averaged with respect to the Wigner distribution function, yields a result equal to the average of Φ_{ji} with respect to the canonical distribution. In particular the average of Φ_{ji}^c with respect to the Maxwell distribution is equal to the canonical average of Φ_{ji} up to terms of order $\hbar^2$.

We find that

$$\Phi_{ji}^c=\sum_{n=0}^{\infty}\frac{1}{n!}\left(\frac{it}{\hbar}\right)^n g_n^c(\mathbf{p}),\tag{27}$$

where the $g_n^c(\mathbf{p})$ obey the recursion relations

$$g_{n+1}^c=g_n^cL+\frac{n\hbar}{i}g_{n-1}^c\{L,H\}+\frac{(n)(n-1)}{2}\left(\frac{\hbar}{i}\right)^2 g_{n-1}^c\times\exp[\tfrac{1}{2}\hbar\boldsymbol{\kappa}\cdot\nabla_{\mathbf{P}_j(0)}]\{\{L,H\},H\}+\cdots+O(\hbar^2).\tag{28}$$

Here $g_0^c=1$, and { } are Poisson brackets.

It is readily verified that the series of Eq. (27) and the recursion relations of Eq. (28) is just the expansion of the classical function

$$\Phi_{ji}^c=\exp\left[-\frac{i}{\hbar}\mathbf{p}\cdot\mathbf{R}_j(0)\right]\exp\left[-\frac{i\mathbf{p}^2t}{2M\hbar}\right]\times\exp\left[\frac{\mathbf{p}}{2}\cdot\nabla_{\mathbf{P}_j(0)}\right]\exp\left[\frac{i}{\hbar}\mathbf{p}\cdot\mathbf{R}_j(t)\right].\tag{29}$$

Applying Weyl's rule again to obtain ψ_{ji}^c from Φ_{ji}^c, we find that

$$\psi_{ji}^c=\exp\left[-\frac{i}{\hbar}\mathbf{p}\cdot\mathbf{R}_i(0)\right]\exp\left[\frac{\mathbf{p}}{2}\cdot\nabla_{\mathbf{P}_i(0)})\right]\times\exp\left[\frac{i\mathbf{p}}{\hbar}\cdot\mathbf{R}_j(t)\right]+O(\hbar^2).\tag{30}$$

Thus, to terms of order $\hbar$,

$$\chi(\mathbf{p},t)=\frac{1}{N}\sum_{j,i}\langle\psi_{ji}^c\rangle_{\mathrm{TC}}.\tag{31}$$

Integration by parts shows this result to be identical with Eq. (19) for $S(\mathbf{p},E)$.

Turner's result[4] can be obtained from Eq. (30) by expanding the operator $\exp[\mathbf{p}\cdot\nabla_{\mathbf{P}_i(0)}/2]$ in a formal Taylor series and retaining only the first two terms in the expansion.

It is perhaps interesting to note that for the "self case" ($j=i$) the Fourier *transform* of the function defined in Eq. (31) is just the correlation function for a particle to be at $\mathbf{r}$ at time t if it were at $\mathbf{r}=0$ at $t=0$ and received an impulse of $\mathbf{p}/2$ then.

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[8] G. C. Wick, Phys. Rev. 94, 1228 (1954).