Validity of the Convolution Approximation for the Van Hove $G(\mathbf{r},t)$ Function

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The validity of the convolution approximation is examined by expanding the spatial Fourier transforms of both the true and the approximate Van Hove $G(\mathbf{r},t)$ function in powers of the density. Only the lowest order terms in the expansions are explicitly calculated. However, comparison of the latter indicates that, for intermediate values of $\mathbf{k}$, terms which are retained in the approximation are of magnitude comparable to that of terms which are neglected. It is concluded, therefore, that the convolution approximation fails at low density for intermediate values of $\mathbf{k}$.

I. INTRODUCTION

As shown by Van Hove,[1] the cross section for the scattering of slow neutrons is related to the dynamical behavior of the scattering system through the relationship

$$d^2\sigma/d\Omega d\omega = (1/4\pi\hbar)(\kappa'/\kappa_0) \times[a_{\rm coh}{}^2 S_{\rm coh}(\mathbf{k},\omega)+a_{\rm inc}{}^2 S_{\rm inc}(\mathbf{k},\omega)], \quad (1.1)$$

where $S_{\rm coh}$ and $S_{\rm inc}$ (respectively, the coherent and incoherent scattering functions[2]) are functions dependent only upon the properties of the scattering medium. The latter have the definitions

$$\begin{Bmatrix} S_{\rm inc}(\mathbf{k},\omega) \\ S_{\rm coh}(\mathbf{k},\omega) \end{Bmatrix} \equiv \int_{-\infty}^{+\infty} dt \exp(-i\omega t) \begin{Bmatrix} \chi_s(\mathbf{k},t) \\ \chi(\mathbf{k},t) \end{Bmatrix}, \quad (1.2)$$

where, in turn, the "intermediate scattering functions" $\chi_s(\mathbf{k},t)$ and $\chi(\mathbf{k},t)$, defined as

$$\chi_s(\mathbf{k},t) \equiv (1/n)\sum_{j=1}^{n} \langle \exp[-i\mathbf{k}\cdot\mathbf{q}_j(0)] \times \exp[+i\mathbf{k}\cdot\mathbf{q}_j(t)]\rangle_T \quad (1.3a)$$

and

$$\chi(\mathbf{k},t) \equiv (1/n)\sum_{i=1}^{n}\sum_{j=1}^{n} \langle \exp[-i\mathbf{k}\cdot\mathbf{q}_i(0)] \times \exp[+i\mathbf{k}\cdot\mathbf{q}_j(t)]\rangle_T, \quad (1.3b)$$

are themselves spatial Fourier transforms of the "Van Hove G functions," viz.,

$$\begin{Bmatrix} \chi_s(\mathbf{k},t) \\ \chi_d(\mathbf{k},t) \equiv \chi - \chi_s \end{Bmatrix} = \int d\mathbf{r} \exp(i\mathbf{k}\cdot\mathbf{r}) \begin{Bmatrix} G_s(\mathbf{r},t) \\ G_d(\mathbf{r},t) \end{Bmatrix}. \quad (1.4)$$

In the classical limit, the G functions have a clear probabilistic meaning[1]:

$G_s(\mathbf{r},t)$ = probability density for finding a particle in the neighborhood of configuration phase point $\mathbf{r}$ at time t, given that at $t=0$, the *same* particle was located at $\mathbf{r}=0$; (1.5)

$G_d(\mathbf{r},t) \propto$ probability density for finding a particle in the neighborhood of $\mathbf{r}$ at time t, given that *another* particle was at $\mathbf{r}=0$ at $t=0$. (1.6)

However, except for some rather special cases, the problem of calculating the G functions seems far from having been resolved. Some success has been achieved in the calculation of $G_s(\mathbf{r},t)$. On the other hand, except for the lowest order terms in an expansion in powers of the time,[3] little is known about the exact kinetic form of $G_d(\mathbf{r},t)$. Indeed, the difficulties inherent in the calculation of the latter probability density are so great, gross approximations most probably need be applied.

The most widely introduced of these is the "convolution approximation"[4] which asserts $G_d(\mathbf{r},t)$ to be dependent only upon the more easily calculated $G_s(\mathbf{r},t)$, viz.,

$$G_d(\mathbf{r},t) \approx c\int d\mathbf{r}' g_2(\mathbf{r}-\mathbf{r}')G_s(\mathbf{r}',t), \quad (1.7)$$

g_2 being the static pair correlation function. The primary purpose of the present investigation is to examine whether this approximation has any utility. Plausible thermodynamic arguments[5,6] already suggest that the approximation, when applied to $\chi_d(\mathbf{k},t)$, fails for small $\mathbf{k}$ (the hydrodynamic limit). In the following, by using detailed statistical mechanical arguments, we definitively show that there is *no* value of $\mathbf{k}$ for which the approximation can be properly exploited.[7]

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[1] L. Van Hove, Phys. Rev. **95**, 249 (1954).

[2] $a_{\rm coh}$ and $a_{\rm inc}$ are, respectively, the coherent and incoherent scattering lengths. κ_0 and κ' are the wave numbers of the neutron before and after scattering; $\mathbf{k}=\boldsymbol{\kappa}_0-\boldsymbol{\kappa}'$.

[3] P. G. DeGennes, Physica **25**, 825 (1959).

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

[5] P. G. DeGennes, in *Proceedings of the Symposium on Inelastic Scattering of Neutrons* (International Atomic Energy Agency, Vienna, 1960), p. 239.

[6] K. S. Singwi and A. Sjolander, Phys. Letters **9**, 120 (1964).

[7] Cf. also, A. Rahman, Phys. Rev. **136**, A405 (1964). The latter author has performed computer calculations for a model of liquid argon, and finds that the approximate G function decays at somewhat too rapid a rate.

For this purpose we utilize calculational procedures which have been developed by Prigogine, Balescu, Résibois, and others[8] to study the irreversible behavior of many-body systems. In Sec. II, these methods are employed to derive formal density expansions for the χ functions. The convolution approximation, itself, is investigated in Sec. III.

II. DEDUCTIONS FROM THE LIOUVILLE EQUATION

A. The Formalism due to Prigogine, Balescu, and Résibois

From Eqs. (1.4) and (1.6), the following relationship is obtained:

$$\chi_d(\mathbf{k},t)=(n-1)\int d\mathbf{X}_n \times\exp(+i\mathbf{k}\cdot\mathbf{q}_2)f_n(\mathbf{X}_n;t|\mathbf{q}_{1_0}=0), \quad (2.1)$$

where $f_n(\mathbf{X}_n;t|\mathbf{q}_{1_0})$ is the conditional probability density for finding the particles of an isolated n-particle equilibrium assembly to be in the neighborhood of Γ phase point $\mathbf{X}_n\equiv(\mathbf{q}_1,\mathbf{q}_2,\cdots,\mathbf{q}_n;\mathbf{p}_1,\cdots,\mathbf{p}_n)\equiv(\mathbf{Q},\mathbf{P})$ at time t, given that at $t=0$ a specified particle was located at $\mathbf{q}_{1_0}$. Consequently, we have

$$\chi_d(\mathbf{k}_2,t)=(n-1)\int d\mathbf{P}\,\rho(-\mathbf{k}_2;\mathbf{P},t), \quad (2.2)$$

where $\rho(\mathbf{k}_2)\equiv\rho(0,\mathbf{k}_2,0,\cdots)$ is one of the coefficients appearing in a generalized Fourier expansion of the n-particle probability density[8]:

$$f_n(\mathbf{X}_n;t|\cdot)=\Omega^{-n}\sum_{\mathbf{K}}\rho(\mathbf{K};\mathbf{P},t)e^{i\mathbf{K}\cdot\mathbf{Q}}. \quad (2.3)$$

[Terms appearing in the latter equation have the following definitions:

$$\mathbf{K}\equiv\{\mathbf{k}_1,\mathbf{k}_2,\cdots,\mathbf{k}_n\};\ \mathbf{K}\cdot\mathbf{Q}\equiv\{\mathbf{k}_1\cdot\mathbf{q}_1,\cdots,\mathbf{k}_n\cdot\mathbf{q}_n\}.]$$

The evolution of $f_n(\mathbf{X}_n;t|\cdot)$ may be obtained by solution of the Liouville equation, given an appropriate initial condition. Hence, Prigogine and co-workers[8] have demonstrated that the $\rho(\mathbf{K},\mathbf{P};t)$ may be related to initial coefficients by the following expansion:

$$\rho(\mathbf{K};\mathbf{P};t)=\mathcal{T}^{-1}\sum_{\mathbf{K}'}\sum_{\sigma}\Theta_\sigma(\mathbf{K},\mathbf{K}';z)\rho(\mathbf{K}';\mathbf{P},0). \quad (2.4)$$

In the above expression, $\mathcal{T}^{-1}$ is the inverse Laplace transform, viz.,[9]

$$\mathcal{T}^{-1}\cdots\equiv\frac{1}{2\pi i}\oint_C dz\exp(-izt),$$

the matrix elements are given by

$$\Theta_0(\mathbf{K},\mathbf{K}';z)\equiv(\mathbf{K}\cdot\mathbf{V}-z)^{-1}\prod_{i=1}^{n}\delta^{kr}(\mathbf{k}_i-\mathbf{k}_i'), \quad (2.5)$$

$$\Theta_\sigma(\mathbf{K},\mathbf{K}';z)\equiv\sum_{\mathbf{K}_1}\sum_{\mathbf{K}_2}\cdots\sum_{\mathbf{K}_{\sigma-1}} \times[(\mathbf{K}\cdot\mathbf{V}-z)^{-1}\langle\mathbf{K}|L_1|\mathbf{K}_1\rangle \times(\mathbf{K}_1\cdot\mathbf{V}-z)^{-1}\langle\mathbf{K}_1|L_1|\mathbf{K}_2\rangle\times\cdots \times\langle\mathbf{K}_{\sigma-1}|L_1|\mathbf{K}'\rangle(\mathbf{K}'\cdot\mathbf{V}-z)^{-1}], \quad \sigma\geq1; \quad (2.6)$$

$\mathbf{V}$ is defined as $\mathbf{V}\equiv(\mathbf{p}_1/m_1,\cdots,\mathbf{p}_n/m_n)$ and, upon assuming pairwise spherically symmetric interactions between the particles constituting the assembly, $\langle\mathbf{K}|L_1|\mathbf{K}'\rangle$ has the form

$$\langle\mathbf{K}|L_1|\mathbf{K}'\rangle\equiv-\sum_{i<j}\sum\Omega^{-1}\tilde{\Phi}_{ij}(|\mathbf{k}_i-\mathbf{k}_i'|)[(\mathbf{k}_i-\mathbf{k}_i')\cdot\mathbf{D}_{ij} \times\delta^{kr}(\mathbf{k}_i+\mathbf{k}_j-\mathbf{k}_i'-\mathbf{k}_j')\prod_{l\neq i,j}\delta^{kr}(\mathbf{k}_l-\mathbf{k}_l')]. \quad (2.7)$$

$\mathbf{D}_{ij}$ is defined as $\mathbf{D}_{ij}\equiv\partial/\partial\mathbf{p}_i-\partial/\partial\mathbf{p}_j$, Ω is the volume of the system, and $\tilde{\Phi}_{ij}(k)$ is defined to be the spatial Fourier transform of the interparticle potential $\Phi_{ij}(r)$, viz.,[10]

$$\tilde{\Phi}_{ij}(k)\equiv\int d\mathbf{r}\exp(i\mathbf{k}\cdot\mathbf{r})\Phi_{ij}(r). \quad (2.8)$$

We shall later take the limit $\Omega\to\infty$, for which the $\mathbf{K}$ summations appearing in Eq. (2.6) shall be replaced by continuous integrals:

$$\sum_{\mathbf{k}_i}\cdots\to(8\pi^3\Omega)^{-1}\int d\mathbf{k}_1\cdots. \quad (2.9)$$

B. Matrix Elements at $t=0$

Assuming that the stationary n-particle probability density of the scattering assembly is canonical, the initial condition for the n-particle *conditional* probability density is given by[11]

$$f_n(\mathbf{X}_n;0|q_{1_0}=0) =\Omega Z_n^{-1}(\beta)\exp[-\beta\Phi(\mathbf{Q})]\rho_0^{eq}(\mathbf{P})\delta^3(\mathbf{q}_1), \quad (2.10)$$

where

$$\Omega^{-1}Z_n(\beta)\equiv\Omega^{-1}\int d\mathbf{Q}\exp[-\beta\Phi(\mathbf{Q})] \quad (2.11)$$

is the partition function for the assembly, and ρ_0^{eq} is defined by

$$\rho_0^{eq}(\mathbf{P})\equiv\prod_{i=1}^{n}[(2\pi m\beta^{-1})^{-3/2}\exp(-\beta p_i^2/2m)]. \quad (2.12)$$

[8] For a review, see I. Prigogine, in *Non-Equilibrium Statistical Mechanics* (Interscience Publishers, Inc., New York, 1962); P. Résibois, in *Physics of Many Particle Systems*, edited by E. Meeron (Gordon and Breach Science Publishers, Inc., New York, 1966).

[9] The (clockwise) contour C is formed by a line drawn in the upper half of the complex plane, closed by a large semicircle drawn through the lower half.

[10] ...the assumption (usually tacit) being that such a procedure is valid even if, as is the case for any realistic intermolecular potential with a hard repulsive core, the Fourier transform of $\Phi(r)$ does not exist. In the present investigation, these expansion procedures are used primarily for convenient bookkeeping and a resummation is later performed; consequently, the Fourier decomposition is probably legitimate in this case. However, in general, one must guard against possible improprieties.

[11] R. Nossal, Phys. Rev. 135, A1579 (1964).

Consequently, from Eq. (2.3), it may readily be shown that the initial value coefficients are given by

$$\rho(\{\mathbf{k}_1,0,\cdots,\mathbf{k}_{i_1},\mathbf{k}_{i_2},\cdots\mathbf{k}_{i_j},\cdots 0\};\mathbf{P},0) = \Omega^{-j}\tilde{g}_j(\mathbf{k}_{i_1},\cdots,\mathbf{k}_{i_j}|\rho_0^{eq}(\mathbf{P}), \quad (2.13)$$

where j is equal to the number of nonzero wave vectors (other than $\mathbf{k}_1$). In the above expression, the mth order equilibrium correlation functions are defined by

$$g_m(\xi_2,\cdots,\xi_m)\equiv\Omega^m Z_n^{-1}\int d\xi_{m+1}\cdots d\xi_n \times\exp[-\beta\Phi(\xi_2,\cdots,\xi_n)], \quad (2.14)$$
$$[\xi_l\equiv\mathbf{q}_l-\mathbf{q}_1].$$

C. Density Expansion for Scattering Functions

Let us now obtain the terms of a density expansion for $\chi_d(\mathbf{k};t)$ from the corresponding terms of $\rho(-\mathbf{k}_2;\mathbf{P},t)$. Using the initial coefficients given by Eq. (2.13), the contributory terms can be obtained from Eq. (2.4) as follows[12]:

There are two contributions to the zeroth-order term. One is that which corresponds to the evolution of the associated probability density functions in the absence of collisions and is given by the $\sigma=0$ term of series Eq. (2.4). Designating this term as $\chi_d^{0|1}(\mathbf{k}_2,t)$, it can be readily shown from Eqs. (2.5) and (2.13) that

$$\chi_d^{0|1}(\mathbf{k}_2,t)=c\exp(-k_2^2t^2|2m\beta)\tilde{g}_2(k_2). \quad (2.15)$$

[To obtain Eq. (2.15), the limit $\Omega\to\infty$, $n\to\infty$, $n/\Omega=c$ has been applied (c is the average density of particles in the system).]

The other contribution to the zeroth-order term of χ_d^0 (i.e., a term proportional to c^1) is found, *a posteriori*, to consist of all terms in Eq. (2.4) describing events such that the designated particle (particle No. 2) interacts *only* with the particle for which the initial position is specified (particle No. 1):

$$\chi_d^{0|2}(\mathbf{k}_2,t)=(n-1)\int d\mathbf{P}\,\mathcal{T}^{-1} \times\{\sum_{\mathbf{K}'}\sum_{\sigma=1}^{\infty}\Theta_\sigma^{2,1}(0,-\mathbf{k}_2,0,\cdots;\mathbf{K}',z)\rho(\mathbf{K}';\mathbf{P},0)\}. \quad (2.16)$$

The terms[13] comprising Eq. (2.16) are evaluated according to Eqs. (2.6)–(2.8); the calculations are lengthy but otherwise without difficulty. One finds

$$\chi_d^{0|2}(\mathbf{k}_2,t) = -\frac{c}{8^3\pi}\int d\mathbf{P}\int_0^t d\tau\int_{-\infty}^{+\infty}d\mathbf{q}\;e^{i\mathbf{v}_2\cdot\mathbf{k}_2(t-\tau)}\left(i\frac{\partial}{\partial\mathbf{q}}\Phi(q)\right)\cdot\mathbf{D}_{2,1} \times G^{0|2}(\tau,\mathbf{k}_2,\mathbf{v}_1,\mathbf{v}_2,\mathbf{q})\rho_0^{eq}(\mathbf{P}), \quad (2.17)$$

with $G^{0|2}(\mathbf{q},\cdots,\tau)$ given by the solution of

$$G^{0|2}(\mathbf{q},\tau)=\exp[i(\mathbf{v}_2\cdot\mathbf{k}_2-i\mathbf{v}_{2,1}\cdot\partial/\partial\mathbf{q})\tau]\;e^{i\mathbf{k}_2\cdot\mathbf{q}}\tilde{g}_2(\mathbf{q}) +\frac{1}{8\pi^3}\int_0^\tau d\tau_1\exp i(\mathbf{v}_2\cdot\mathbf{k}_2-i\mathbf{v}_{2,1}\cdot\partial/\partial\mathbf{q})(\tau-\tau_1) \times(i\partial/\partial\mathbf{q}\Phi(q)\cdot\mathbf{D}_{2,1})G^{0|2}(\tau_1,\mathbf{q}). \quad (2.18)$$

In Eqs. (2.17) and (2.18), we have designated $\mathbf{v}_2\equiv\mathbf{P}_2/m$; $\mathbf{v}_{2,1}\equiv\mathbf{v}_2-\mathbf{v}_1$.

Note that $\chi_d^{0|1}$ and $\chi_d^{0|2}$ are the only terms constituting the zeroth contribution to a density expansion of χ_d; terms of Eq. (2.4) for which more than two designated particles take part in an evolution history must contribute at higher order, since every summation over particles introduces powers of the density c.

In a similar manner, a formal density expansion can be obtained for the "self" part of the intermediate scattering function. One finds

$$\chi_s(\mathbf{k},t)=\chi_s^{(0)}+\chi_s^{(1)}+0(c^2)\cdots, \quad (2.19)$$

where

$$\chi_s^{(0)}(\mathbf{k},t)=\exp(-k^2t^2/2m\beta), \quad (2.20)$$

and $\chi_s^{(1)}$ provided by an equation identical to Eq. (2.17), except that $G^{0|2}$ of the latter must be replaced by $G^{(1)}$ given by solution of the following integral equation:

$$G^{(1)}(\tau,\mathbf{k},v_1,v_2,\mathbf{q})=\exp[i(\mathbf{v}_i\cdot\mathbf{k}-i\mathbf{v}_{2,1}\partial/\partial\mathbf{q})\tau]\;\tilde{g}_2(\mathbf{q}) +\frac{1}{8\pi^3}\int_0^\tau d\tau_1\exp i(\mathbf{v}_2\cdot\mathbf{k}-i\mathbf{v}_{2,1}\partial/\partial\mathbf{q})(\tau-\tau_1) \times(i\partial/\partial\mathbf{q}\Phi(q)\cdot\mathbf{D}_{2,1})G^{(1)}(\tau_1,\mathbf{q}). \quad (2.21)$$

Density expansions for $\chi_s(\mathbf{k},t)$ have already been investigated by Mazo and Zemach,[14] the first few terms of a time expansion of Eqs. (2.17) and (2.21) having been obtained in their studies. Primary interest here, however, is the demonstration of the similarity between $\chi_s^{(1)}$ and $\chi_d^{0|2}$. Comparing Eqs. (2.17) and (2.18) with Eqs. (2.17) and (2.21), it is seen that these terms are of the same order in c and have the same form. The only difference between the two expressions appears through the inhomogeneous term of the integral equations.

III. CONVOLUTION APPROXIMATION

Expressed in terms of Fourier transforms, the convolution approximation [cf. Eq. (1.7)] takes the particularly simple form

$$\chi_d(\mathbf{k},t)\simeq\chi_d(\mathbf{k},0)\chi_s(\mathbf{k},t)=c\tilde{g}_2(\mathbf{k})\chi_s(\mathbf{k},t). \quad (3.1)$$

The cross section $\chi_d(\mathbf{k},t)$ is never measured by itself but always appears along with $\chi_s(\mathbf{k},t)$. Indeed [cf. Eq. (1.1)], the coherent scattering cross section is defined as being proportional to the full χ function, i.e.,

$$\frac{d^2\sigma_{coh}}{d\Omega d\omega}\propto\int dt\,\exp(-i\omega t)\chi(\mathbf{k},t),$$

[12] In virtue of the momentum integration demanded by Eq. (2.2), not all terms constituting the series given by Eq. (2.4) are contributory to $\chi_d(\mathbf{k};t)$.

[13] $\Theta^{2,1}$ is defined as in Eqs. (2.6)–(2.9), except that the (i,j) summation in Eq. (2.7) is suppressed (we designate $i=2$, $j=1$).

[14] R. M. Mazo and A. C. Zemach, Phys. Rev. **109**, 1564 (1958).

where

$$\chi=\chi_s+\chi_d.$$

It is immediately determined from Eqs. (2.15), (2.17), (2.19), and (3.1) that

$$\chi_d^{\text{actual}}-\chi_d^{\text{approx}}=c\Delta_1(\mathbf{k},t)+o(c^2)\cdots \tag{3.2}$$

with $c\Delta_1$, the first-order term neglected when the approximation is applied, being defined by

$$c\Delta_1(\mathbf{k},t)=\chi_d^{0|2}(\mathbf{k},t). \tag{3.3}$$

Suppose, however, that one did use the convolution approximation to calculate $\chi(\mathbf{k},t)$. Doing so would provide

$$\chi^{\text{approx}}(\mathbf{k},t)=\chi_s+\chi_d^{\text{approx}}=\chi_s^{(0)}+(\chi_s^{(1)}+\chi_d^{0|1})+o(c^2). \tag{3.4}$$

But it is now evident that for small and intermediate $\mathbf{k}$, this procedure is in error; as seen from Eqs. (3.3), (2.18), and (2.21), the neglected term is of the same order of magnitude as the $\chi_s^{(1)}$ term which is retained and, in fact, tends closer and closer to the latter as $\mathbf{k}\to 0$.

At first glance, the approximation seems to be somewhat better founded for large $\mathbf{k}$. This is true because the inhomogeneous g_2 term appearing in Eq. (2.18) becomes so heavily modulated by the rapidly oscillating function $\exp(i\mathbf{q}\cdot\mathbf{k}_2)$ that integration over $\exp(i\mathbf{q}\cdot\mathbf{k}_2)g_2(q)$ in Eqs. (2.17) and (2.18) presumably yields a very small number.

Thus, for example, a first iteration of Eqs. (2.17) and (2.21) provides a term proportional to a system constant η defined as

$$\eta\equiv\int d\mathbf{q}\; g_2(q)\nabla^2\Phi(q); \tag{3.5}$$

the corresponding term arising from Eqs. (2.17)–(2.18) is proportional to a modified constant η', defined as

$$\eta'\equiv\int d\mathbf{q}\exp(i\mathbf{k}\cdot\mathbf{q})g_2(q)\nabla^2\Phi(q). \tag{3.6}$$

Suppose that the intermolecular potential is Lennard-Jones or similar type, with parameters σ_r and σ_a characterizing the *widths* of the repulsive core and "attractive well" terms. We can ensure that $\eta'\ll\eta$ by requiring that $\mathbf{k}\cdot\sigma_r$, $\mathbf{k}\cdot\sigma_a\gg 1$.

But, as shown by DeGennes,[3] for $\mathbf{k}\cdot\sigma_r\geq 2$, $\chi(\mathbf{k},t)$ is well represented by an expression calculated from an ideal-gas model. Consequently, although the convolution approximation is valid in this range, it is seen to be unnecessary.

IV. REMARKS

The contribution of this paper is explicit demonstration of the failure of the convolution approximation for intermediate $\mathbf{k}$ and low density. The reader must be cautioned that the analysis is not applicable to an investigation of the validity of $\chi(\mathbf{k},t)$ for very small $\mathbf{k}$. Small momentum transfer implies small energy transfer and, as is evident from Eq. (1.2), at small ω a scattering experiment probes the long-time behavior of $\chi(\mathbf{k},t)$. However, when $\mathbf{k}$ is small the latter is provided by a hydrodynamic description and, in order to obtain the correct hydrodynamic behavior, a summation over all powers of the density is required.

In summary, the first term of an exact density expansion of $\chi(\mathbf{k},t)$ has been compared with the corresponding expansion term of the convolution approximate scattering function. It has been found that for intermediate $\mathbf{k}$ the unmodified convolution approximation is inconsistent because terms of importance which are retained in the convolution approximation are of the same order as similar terms which are neglected. It is only at large $\mathbf{k}$ that the neglected terms are unimportant, but in such a case $\chi(\mathbf{k},t)$ is adequately represented by an ideal-gas calculation and the convolution approximation is superfluous.

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