Dielectric function of the uniform electron gas for large frequencies or wave vectors

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The dielectric function of a uniform electron gas is studied on the basis of the dynamical equations which govern the response of the system to a weak external field. The deviations from the random-phase approximation caused by exchange and correlation effects are incorporated in a local-field correction which is related to the response of a certain two-particle correlation function. Using the equation of motion for the correlation function we extract the exact behavior of the local-field correction for large wave vectors or high frequencies. The high-frequency result is identical to the one obtained from the third frequency moment. For large wave vectors we find that the local-field correction tends to $2[1-g(0)]/3$, $g(0)$ being the value of the pair distribution function at $r=0$. We also recover the result of Kimball, giving a relation between the pair distribution function and its radial derivative at $r=0$.

I. INTRODUCTION

The dielectric function of the uniform electron gas plays a central role in many theories of metallic properties, and much effort has gone into obtaining accurate numerical information about this function. It is well known that the conventional random-phase approximation is unsatisfactory at metallic densities because it treats in a proper way only the long-range part of the Coulomb interaction, neglecting the important short-range exchange and correlation effects. In recent years several attempts have been made to improve on the random-phase approximation, taking into account the fact that the exchange and correlation effects lead to a local depletion in the density around each electron. This makes that the effective field acting on an electron differs from the macroscopic mean field, and one refers to the difference as a local-field correction. The correction has been studied on the basis of Green's-function analysis and diagrammatic techniques by Hubbard,[1] Geldart and coworkers,[2–4] Toigo and Woodruff,[5] and others.[6–9] A different approach was introduced by Singwi *et al.*,[10] who expressed the local-field correction as a functional of the static pair distribution function. Several modified versions of this theory have subsequently been presented.[11–16]

Unfortunately, the results of the various theories can not be directly tested against experiments because one has to introduce additional approximations to take into account the interactions with the lattice. However, there are a number of exact sum rules which the true dielectric function has to satisfy, and these can be used to test whether a particular approximation is reasonable or not. The most important sum rules are the following[14,17,18]:

(a) The fluctuation-dissipation theorem leads to an expression for the static pair distribution function in terms of the dielectric function. Although the true pair distribution function is not known, a minimum requirement is that it should be positive.

(b) The compressibility sum rule gives a relation between the compressibility of the electron gas and the static dielectric function for small wave vectors. Fairly accurate numerical values for the compressibility can be obtained by means of the virial theorem.

(c) The first and third frequency moments give the leading terms in an expansion of the dielectric function in inverse powers of the frequency, and they contain information about the short-time response to an external disturbance.

It turns out that the sum rules impose quite severe restrictions on the possible approximations, and at present there exists no theory which satisfies all of them. The best compromise seems to be the recent theory by Vashishta and Singwi.[13]

None of the sum rules listed above gives information about the behavior of the dielectric function for large wave vectors and small frequencies, and in the literature there has been some controversy about the proper behavior in this limit.[4,6,7,19–22] We shall, in this paper, carry out a detailed study of the dielectric function when either the wave vector or the frequency is large, and in particular we shall obtain an exact expression for the local-field correction in the limit of large wave vectors. In the process we shall also obtain a general formulation of the problem which hopefully can be used as a starting point for improvements on previous theories. As will be discussed later on there is a great need for such improvements, because all the existing theories are based on rather crude approximations and serious questions concerning basic physical aspects of the problem remain unanswered.

The outline of the paper is as follows. In Sec. II we introduce the mathematical formalism and in Sec. III we discuss the definition of the local-field correction. This is expressed in terms of the linear response to an external disturbance of a certain two-particle correlation function. The equation of motion for the two-particle correlation function is studied in Sec. IV, and the physical meaning of the various terms is briefly discussed. In Sec. V we specialize to the case when either the frequency or the wave vector is very large. We show that in the limit of large frequencies and finite wave vectors we recover the results previously obtained through the third-moment sum rule.[16] In the opposite limit, when the frequency is finite but the wave vector tends to infinity, we obtain a simple relation between the local-field correction and the value of the pair distribution function at zero interparticle separation. Using the fluctuation-dissipation theorem we also show that the logarithmic derivative of the pair distribution at $r=0$ is equal to the inverse Bohr radius, in agreement with the result obtained by Kimball[23] through a completely different method. Section VI, finally, contains some concluding remarks about the validity of local-field theories, and we list a number of questions which need to be elucidated by future work in this field.

II. BASIC DEFINITIONS

We consider a system of N electrons in a uniform positive background perturbed by a weak external field. The Hamiltonian of the system is

$$H=\frac{\hbar^2}{2m}\sum_{\vec{k}\sigma} k^2 a^\dagger_{\vec{k}\sigma} a_{\vec{k}\sigma}+\frac{1}{2V}\sum_{\vec{q}} v(\vec{q})\sum_{\vec{k}\sigma}\sum_{\vec{k}'\sigma'} a^\dagger_{\vec{k}-\vec{q}/2,\sigma}\, a^\dagger_{\vec{k}'+\vec{q}/2,\sigma'}\, a_{\vec{k}'-\vec{q}/2,\sigma'}\, a_{\vec{k}+\vec{q}/2,\sigma}$$
$$+\frac{1}{V}\sum_{\vec{q}} \Phi^{\text{ext}}(-\vec{q},t)\sum_{\vec{k}\sigma} a^\dagger_{\vec{k}-\vec{q}/2,\sigma}\, a_{\vec{k}+\vec{q}/2,\sigma}\ , \tag{1}$$

where $a^\dagger_{\vec{k}\sigma}$ and $a_{\vec{k}\sigma}$ are creation and annihilation operators for an electron with momentum $\hbar\vec{k}$ and spin σ, V is the total volume of the system, and m is the mass of an electron. $\Phi^{\text{ext}}(\vec{q},t)$ is the spatial Fourier transform of the external potential, and $v(\vec{q})$ is the Fourier transform of the Coulomb potential, including the interaction with the uniform positive background,

$$v(\vec{q})=\begin{cases}4\pi e^2/\vec{q}^{\,2} & \text{if } q\neq 0\\ 0 & \text{if } q=0\ .\end{cases} \tag{2}$$

The external perturbation leads to variations in space and time of the density of electrons, and we write

$$n(\vec{r},t)=n+\bar{n}(\vec{r},t)\ , \tag{3}$$

where $n=N/V$ is the mean density in the absence of any external forces and $\bar{n}(\vec{r},t)$ is the induced density. Throughout this paper we shall use the convention that quantities which represent deviations from equilibrium are denoted by a bar over the symbol. According to linear response theory we have after Fourier transformation with respect to $\vec{r}$ and t

$$\bar{n}(\vec{q},\omega)=\chi(\vec{q},\omega)\Phi^{\text{ext}}(\vec{q},\omega)\ , \tag{4}$$

where the response function $\chi(\vec{q},\omega)$ is related to the dielectric function $\epsilon(\vec{q},\omega)$ through

$$\frac{1}{\epsilon(\vec{q},\omega)}=1+v(\vec{q})\chi(\vec{q},\omega)\ . \tag{5}$$

The response function can be expressed in terms of the equilibrium density-density correlation function, and it can, in principle, be calculated through the conventional diagrammatic technique.[1-4,6-9] The disadvantage with this method is that for metallic densities all the diagrams are essentially of the same order of magnitude, and it is not obvious how simple physical aspects, such as the existence of a correlation hole, enter in the theory. The alternative approach which will be adopted here is to study directly the dynamical equations governing the response of the system to the external field. For this purpose we write the local density of electrons as

$$n(\vec{r},t)=\sum_{\vec{k}\sigma} f^{(1)}_{\vec{k}\sigma}(\vec{r},t)\ , \tag{6}$$

where $f^{(1)}_{\vec{k}\sigma}(\vec{r},t)$ is the quantum-mechanical analogue of the classical phase-space distribution function. The Fourier transform of $f^{(1)}_{\vec{k}\sigma}(\vec{r},t)$ with respect to $\vec{r}$ is given by

$$f^{(1)}_{\vec{k}\sigma}(\vec{q},t)=\langle a^\dagger_{\vec{k}-\vec{q}/2,\sigma}(t)\, a_{\vec{k}+\vec{q}/2,\sigma}(t)\rangle\ , \tag{7}$$

and in the presence of an external potential we write

$$f^{(1)}_{\vec{k}\sigma}(\vec{q},t)=\delta^{\,0}_{\vec{q}}\, n_{\vec{k}\sigma}+\bar{f}^{(1)}_{\vec{k}\sigma}(\vec{q},t)\ , \tag{8}$$

where the first term is the equilibrium part, $n_{\vec{k}\sigma}$ being the occupation number for electrons with momentum $\hbar\vec{k}$ and spin σ. We shall also introduce a two-particle distribution function $f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{r},\vec{r}\,';t)$ which in the classical case gives the probability to find a particle with momentum $\hbar\vec{k}$ at the position $\vec{r}$

if there is at the same time t another particle with momentum $\hbar\vec{k}'$ at the position $\vec{r}'$. The Fourier transform of this function with respect to $\vec{r}$ and $\vec{r}'$ is defined through

$$f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}\,';t)+f^{(1)}_{\vec{k}\sigma}(\vec{q},t)f^{(1)}_{\vec{k}'\sigma'}(\vec{q}\,',t)$$
$$=\langle a^{\dagger}_{\vec{k}-\vec{q}/2,\sigma}(t)\,a^{\dagger}_{\vec{k}'-\vec{q}\,'/2,\sigma'}(t)\,a_{\vec{k}'+\vec{q}\,'/2,\sigma'}(t)\,a_{\vec{k}+\vec{q}/2,\sigma}(t)\rangle\,, \tag{9}$$

where the second term on the left-hand side represents the uncorrelated part of the two-particle distribution function. In the quantum-mechanical case the function $f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}\,';t)$ contains both exchange and Coulomb correlations, and in particular it contains information about the local depletion in density around each particle. In analogy with Eq. (8) we write

$$f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}\,';t)=\delta^{\vec{0}}_{\vec{q}+\vec{q}\,'}f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q})+\bar{f}^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}\,';t)\,, \tag{10}$$

where the first term is the equilibrium part. Summing this over all momenta and spin one obtains the static structure factor $S(\vec{q})$ which is essentially the Fourier transform of the static pair distribution function:

$$\frac{1}{N}\sum_{\vec{k}\sigma}\sum_{\vec{k}'\sigma'}f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q})=S(\vec{q})-1\,, \tag{11}$$

$$g(\vec{r})-1=\frac{1}{N}\sum_{\vec{q}}e^{i\vec{q}\cdot\vec{r}}[S(\vec{q})-1]\,. \tag{12}$$

By ordering the creation operators to the left of the annihilation operators in Eq. (9) we have taken away the self-part of the correlation function, and this is the reason why $S(\vec{q})-1$ rather than $S(\vec{q})$ appears in Eq. (11).

III. LOCAL-FIELD CORRECTION

Using the Hamiltonian given in Eq. (1) it is straightforward to obtain the equation of motion for the one-particle distribution function $f^{(1)}_{\vec{k}\sigma}(\vec{q},t)$. Keeping only terms linear in the external field we get after Fourier transformation with respect to t

$$[\hbar\omega-(\hbar^2/m)\vec{k}\cdot\vec{q}]\bar{f}^{(1)}_{\vec{k}\sigma}(\vec{q},\omega)=\frac{1}{V}[n_{\vec{k}-\vec{q}/2,\sigma}-n_{\vec{k}+\vec{q}/2,\sigma}][\Phi^{\text{ext}}(\vec{q},\omega)+v(\vec{q})\bar{n}(\vec{q},\omega)]$$
$$+\frac{1}{V}\sum_{\vec{q}\,'}v(\vec{q}\,')\sum_{\vec{k}'\sigma'}[\bar{f}^{(2)}_{\vec{k}-\vec{q}\,'/2,\sigma;\vec{k}'\sigma'}(\vec{q}-\vec{q}\,',\vec{q}\,';\omega)-\bar{f}^{(2)}_{\vec{k}+\vec{q}\,'/2,\sigma;\vec{k}'\sigma'}(\vec{q}-\vec{q}\,',\vec{q}\,';\omega)]\,. \tag{13}$$

The term $v(\vec{q})\bar{n}(\vec{q},\omega)$ which appears together with the external field is the Hartree mean field, and it arises from the uncorrelated part of the two-particle distribution function. The effects of exchange and Coulomb correlations enter through the last term which contains the induced change of the correlated part of the two-particle distribution function. Neglecting this term for a moment, we obtain after summation over $\vec{k}$ and σ

$$\bar{n}(\vec{q},\omega)=\chi^0(\vec{q},\omega)[\Phi^{\text{ext}}(\vec{q},\omega)+v(\vec{q})\bar{n}(\vec{q},\omega)]\,, \tag{14}$$

where

$$\chi^0(\vec{q},\omega)=\frac{1}{V}\sum_{\vec{k}\sigma}\frac{n_{\vec{k}-\vec{q}/2,\sigma}-n_{\vec{k}+\vec{q}/2,\sigma}}{\hbar\omega-(\hbar^2/m)\vec{k}\cdot\vec{q}}\,. \tag{15}$$

Within this approximation one should for consistency use the free-electron occupation number, and $\chi^0(\vec{q},\omega)$ is then the Lindhard response function for the noninteracting electron gas. Equation (14) is the random-phase approximation and it means that the electrons respond as free particles to the external field plus the Hartree mean field. In theories which go beyond the random-phase approximation one argues that the presence of a correlation hole around each particle modifies the Hartree mean field, and one writes the effective mean field acting on an electron as

$$\Phi^{\text{eff}}(\vec{q},\omega)=\Phi^{\text{ext}}(\vec{q},\omega)+v(\vec{q})[1-G(\vec{q},\omega)]\bar{n}(\vec{q},\omega)\,, \tag{16}$$

where the function $G(\vec{q},\omega)$ represents the local-field correction. Explicit expressions for $G(\vec{q},\omega)$ can be obtained by making specific assumptions about the shape and the dynamics of the correlation hole.[10–13] In most theories one makes the additional assumption that the electrons respond as free particles to the effective field, which leads to

$$\bar{n}(\vec{q},\omega)=\chi^0(\vec{q},\omega)\{\Phi^{\text{ext}}(\vec{q},\omega)+v(\vec{q})[1-G(\vec{q},\omega)]\bar{n}(\vec{q},\omega)\}\,. \tag{17}$$

On physical grounds we should certainly expect some modifications also in $\chi^0(\vec{q},\omega)$, arising for instance from self-energy corrections. However, in order to make contact with the existing theories, we shall keep Eq. (17) in its present form, and we shall look upon it as the definition of the unknown function $G(\vec{q},\omega)$. This means that we include in the local-field correction certain terms which would more naturally belong in a modified free-particle response function, and we shall discuss the origin of these terms at a later stage. For the sake of clarity it should be pointed out that the free-particle response function is defined through Eq. (15), where $n_{\vec{k}\sigma}$ are the exact occupation numbers for the interacting system, and hence $\chi^0(\vec{q},\omega)$ deviates somewhat from the Lindhard free-electron response function. It is obvious that even if each individual electron responds as a free particle, the total response must still depend on the true distribution of particles in the system.

Using Eq. (13) together with the definitions in Eqs. (15) and (17) it is straightforward to obtain the following exact expression for $G(\vec{q},\omega)$:

$$G(\vec{q},\omega)=-[v(\vec{q})\chi^0(\vec{q},\omega)\bar{n}(\vec{q},\omega)]^{-1}\times\frac{1}{V}\sum_{\vec{q}'}v(\vec{q}')\sum_{\vec{k}\sigma}\left(\frac{1}{\hbar\omega-(\hbar^2/m)(\vec{k}+\frac{1}{2}\vec{q}')\cdot\vec{q}}-\frac{1}{\hbar\omega-(\hbar^2/m)(\vec{k}-\frac{1}{2}\vec{q}')\cdot\vec{q}}\right)\sum_{\vec{k}'\sigma'}\bar{f}^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q}-\vec{q}',\vec{q}';\omega)\ . \tag{18}$$

In order to obtain explicit results we have to make approximations on $\bar{f}^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}';\omega)$. The most common procedure is to factorize the two-particle distribution function into a product involving one-particle distribution functions and the static pair distribution function.[5,10-13] Unfortunately, it is difficult to justify such approximations on a rigorous basis, particularly if one is interested in the dynamical aspects which determine the frequency dependence of $G(\vec{q},\omega)$. For this reason we shall carry the exact formalism one step further and study the equation of motion for the two-particle distribution function in the presence of the external field.

IV. TWO-PARTICLE DISTRIBUTION FUNCTION

Using the definitions in Eqs. (9) and (10) together with the Hamiltonian in Eq. (1) we can derive the equation of motion for $\bar{f}^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}';\omega)$. After some tedious but straightforward transformations we obtain

$$\begin{aligned}[\hbar\omega-(\hbar^2/m)\vec{k}\cdot\vec{q}-(\hbar^2/m)\vec{k}'\cdot\vec{q}']\bar{f}^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}';\omega)&=F^{ext}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}';\omega)+F^{ext}_{\vec{k}'\sigma';\vec{k}\sigma}(\vec{q}',\vec{q};\omega)\\&\quad+F^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}';\omega)+F^{m}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}';\omega)\\&\quad+F^{m}_{\vec{k}'\sigma';\vec{k}\sigma}(\vec{q}',\vec{q};\omega)\ ,\end{aligned}\tag{19}$$

where the terms on the right-hand side are associated with different aspects of the interactions in the system. In order to discuss the physical meaning of the various terms we make the classical interpretation of $f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}';t)$ as giving the equal-time correlation between two particles with momenta $\hbar\vec{k}$ and $\hbar\vec{k}'$, and we refer to the two particles as "the first particle" and "the second particle," respectively. The term $F^{ext}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}';\omega)$ arises from the action of the external force on the first particle in the presence of the second particle. Similarly, $F^{ext}_{\vec{k}'\sigma';\vec{k}\sigma}(\vec{q}',\vec{q};\omega)$ gives the effect of the external force acting on the second particle in the presence of the first one. The explicit form of $F^{ext}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}';\omega)$ is

$$\begin{aligned}F^{ext}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}';\omega)&=\frac{1}{V}[f^{(2)}_{\vec{k}-(\vec{q}+\vec{q}')/2,\sigma;\vec{k}'\sigma'}(-\vec{q}')-f^{(2)}_{\vec{k}+(\vec{q}+\vec{q}')/2,\sigma;\vec{k}'\sigma'}(-\vec{q}')]\\&\quad\times\Phi^{ext}(\vec{q}+\vec{q}';\omega)\ ,\end{aligned}\tag{20}$$

where the equilibrium part of the two-particle correlation function appears. The term $F^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}';\omega)$ in Eq. (19) arises from the mutual interaction between the first and the second particle, and thus it contains all the effects associated with the two-body problem. It is given by

$$\begin{aligned}F^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}';\omega)&=\frac{1}{V}\sum_{\vec{q}''}v(\vec{q}'')[\bar{f}^{(2)}_{\vec{k}+\vec{q}''/2,\sigma;\vec{k}'-\vec{q}''/2,\sigma'}(\vec{q}+\vec{q}'',\vec{q}'-\vec{q}'';\omega)-\bar{f}^{(2)}_{\vec{k}-\vec{q}''/2,\sigma;\vec{k}'+\vec{q}''/2,\sigma'}(\vec{q}+\vec{q}'',\vec{q}'-\vec{q}'';\omega)]\\&\quad+\frac{1}{V}v(\vec{q})[n_{\vec{k}-\vec{q}/2,\sigma}\bar{f}^{(1)}_{\vec{k}'+\vec{q}/2,\sigma'}(\vec{q}+\vec{q}',\omega)-n_{\vec{k}+\vec{q}/2,\sigma}\bar{f}^{(1)}_{\vec{k}'-\vec{q}/2,\sigma'}(\vec{q}+\vec{q}',\omega)]\\&\quad+\frac{1}{V}v(\vec{q}')[n_{\vec{k}'-\vec{q}'/2,\sigma'}\bar{f}^{(1)}_{\vec{k}+\vec{q}'/2,\sigma}(\vec{q}+\vec{q}',\omega)-n_{\vec{k}'+\vec{q}'/2,\sigma'}\bar{f}^{(1)}_{\vec{k}-\vec{q}'/2,\sigma}(\vec{q}+\vec{q}',\omega)]\ .\end{aligned}\tag{21}$$

The last two terms in Eq. (19) contain the many-body aspects of the interaction, and they lead to a screening of the external field and the two-body interaction. The term $F^{m}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}';\omega)$ arises from the interaction between the first particle and all the surrounding particles in the presence of the second particle, and similarly $F^{m}_{\vec{k}'\sigma';\vec{k}\sigma}(\vec{q}',\vec{q};\omega)$ arises from the interaction of the second particle with the surrounding ones in the presence of the first particle. These terms contain three-particle correlation functions, and in analogy with Eq. (9) we make a decomposition into cumulants, writing

$$\begin{aligned}&f^{(3)}_{\vec{k}\sigma;\vec{k}'\sigma';\vec{k}''\sigma''}(\vec{q},\vec{q}',\vec{q}'';t)+f^{(1)}_{\vec{k}\sigma}(\vec{q},t)f^{(2}_{\vec{k}'\sigma';\vec{k}''\sigma''}(\vec{q}',\vec{q}'';t)\\&\quad+f^{(1)}_{\vec{k}'\sigma'}(\vec{q}',t)f^{(2)}_{\vec{k}\sigma;\vec{k}''\sigma''}(\vec{q},\vec{q}'';t)+f^{(1)}_{\vec{k}''\sigma''}(\vec{q}'',t)f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}';t)+f^{(1)}_{\vec{k}\sigma}(\vec{q},t)f^{(1)}_{\vec{k}'\sigma'}(\vec{q}',t)f^{(1)}_{\vec{k}''\sigma''}(\vec{q}'',t)\end{aligned}$$

$$=\langle a^{\dagger}_{\vec{k}-\vec{q}/2,\sigma}(t)\, a^{\dagger}_{\vec{k}'-\vec{q}'/2,\sigma'}(t)\, a^{\dagger}_{\vec{k}''-\vec{q}''/2,\sigma''}(t)\, a_{\vec{k}''+\vec{q}''/2,\sigma''}(t)\, a_{\vec{k}'+\vec{q}'/2,\sigma'}(t)\, a_{\vec{k}+\vec{q}/2,\sigma}(t)\rangle \;. \tag{22}$$

With this definition, the term representing the many-body forces on the first particle takes the form

$$\begin{aligned} F^{m}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}\,';\omega) = {} & \frac{1}{V}[f^{(2)}_{\vec{k}-(\vec{q}+\vec{q}\,')/2,\sigma;\vec{k}'\sigma'}(-\vec{q}\,') - f^{(2)}_{\vec{k}+(\vec{q}+\vec{q}\,')/2,\sigma;\vec{k}'\sigma'}(-\vec{q}\,')]\, v(\vec{q}+\vec{q}\,')\bar{n}(\vec{q}+\vec{q}\,';\omega) \\ & + \frac{1}{V} v(\vec{q}) \sum_{\vec{k}''\sigma''} f^{(2)}_{\vec{k}''\sigma'';\vec{k}'\sigma'}(-\vec{q}\,')[\bar{f}^{(1)}_{\vec{k}+\vec{q}\,'/2,\sigma}(\vec{q}+\vec{q}\,';\omega) - \bar{f}^{(1)}_{\vec{k}-\vec{q}\,'/2,\sigma}(\vec{q}+\vec{q}\,';\omega)] \\ & + \frac{1}{V} v(\vec{q})[n_{\vec{k}-\vec{q}/2,\sigma} - n_{\vec{k}+\vec{q}/2,\sigma}] \sum_{\vec{k}''\sigma''} \bar{f}^{(2)}_{\vec{k}''\sigma'';\vec{k}'\sigma'}(\vec{q},\vec{q}\,';\omega) \\ & + \frac{1}{V}\sum_{\vec{q}\,''} v(\vec{q}\,'') \sum_{\vec{k}''\sigma''} [\bar{f}^{(3)}_{\vec{k}-\vec{q}\,''/2,\sigma;\vec{k}'\sigma';\vec{k}''\sigma''}(\vec{q}-\vec{q}\,'',\vec{q}\,',\vec{q}\,'';\omega) - \bar{f}^{(3)}_{\vec{k}+\vec{q}\,''/2,\sigma;\vec{k}'\sigma';\vec{k}''\sigma''}(\vec{q}-\vec{q}\,'',\vec{q}\,',\vec{q}\,'';\omega)]. \end{aligned} \tag{23}$$

Here, the first term contains the Hartree mean field, and it can simply be added to Eq. (20), leading to a screening of the external field. The following two terms in the above equation contain correlation functions describing the charge distribution around the second particle, and they represent the screening of the Coulomb potential from this particle. The terms considered so far would give the true force acting on the first particle if it had been an infinitesimal test charge which does not itself disturb the system. However, it is in reality surrounded by a correlation hole which modifies both the Hartree mean field and the screened field from the second particle. These modifications are described by the irreducible three-particle correlations appearing in the last term of Eq. (23).

In Eq. (19) the particles with momenta $\vec{k}$ and $\vec{k}'$ enter in a completely symmetric way. However, when $\bar{f}^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q}-\vec{q}\,',\vec{q}\,';\omega)$ is inserted into Eq. (13) we see that $\vec{k}$ labels the particle we are studying whereas $\vec{k}'$ labels one of the surrounding particles which make up the correlation hole. Hence, the terms in Eq. (19) which represent the forces on the second particle are associated with the dynamics of the correlation hole, whereas the forces on the first particle determine its motion inside the correlation hole. One may argue that a proper theory should include in the local-field correction $G(\vec{q},\omega)$ only terms associated with the dynamics of the correlation hole, whereas terms associated with the motion of the particle itself should appear in a modified free-particle response function. As was pointed out by Goodman and Sjölander, who studied the magnetic response, it is essential to take into account the motion of the particle relative to its correlation hole in order to get proper results for high frequencies.[24] Equation (19) may provide a basis for further clarification of these aspects.

V. LIMIT OF LARGE q OR ω

If the external disturbance varies rapidly in time the response of the system is determined by the motion of the particles for short times. This is essentially a free-particle behavior since it takes a certain time before the motion of a particle is changed by the forces from other particles. Similarly, the response to a disturbance which varies rapidly in space is also determined by the free-particle behavior, because a particle travels a certain distance before it is affected by the presence of other particles. This means that for large values of q or ω the terms on the left-hand side of Eq. (13) dominate over the term containing the Coulomb interaction on the right-hand side. Thus we get

$$\bar{f}^{(1)}_{\vec{k}\sigma}(\vec{q},\omega) = \frac{1}{V}\frac{n_{\vec{k}-\vec{q}/2,\sigma} - n_{\vec{k}+\vec{q}/2,\sigma}}{\hbar\omega - (\hbar^2/m)\vec{k}\cdot\vec{q}}\,\Phi^{\rm ext}(\vec{q},\omega)\;, \tag{24}$$

and after summation over $\vec{k}$ and σ this leads to

$$\begin{aligned} \bar{n}(\vec{q},\omega) &= \chi^0(\vec{q},\omega)\Phi^{\rm ext}(\vec{q},\omega) \\ &= \frac{1}{V}\sum_{\vec{k}\sigma}\left(\frac{1}{\hbar\omega - (\hbar^2/2m)\vec{q}^{\,2} - (\hbar^2/m)\vec{k}\cdot\vec{q}} - \frac{1}{\hbar\omega + (\hbar^2/2m)\vec{q}^{\,2} - (\hbar^2/m)\vec{k}\cdot\vec{q}}\right) n_{\vec{k}\sigma}\Phi^{\rm ext}(\vec{q},\omega)\;, \end{aligned} \tag{25}$$

where we have made a trivial change of the summation variable. In the denominators we can here neglect the term $(\hbar^2/m)\vec{k}\cdot\vec{q}$, and hence we obtain

$$\chi(\vec{q},\omega) = \chi^0(\vec{q},\omega) = \frac{n\hbar^2}{m}q^2\frac{1}{(\hbar\omega)^2 - (\hbar^2q^2/2m)^2}\;. \tag{26}$$

This formula is valid for frequencies and wave vectors such that

$$\left|\hbar\omega \pm \frac{\hbar^2q^2}{2m}\right| \gg \epsilon_F\;, \tag{27}$$

i.e., for points in the (q,ω) plane well outside the region of particle-hole excitations.

In order to get the leading correction to Eq. (26) we have to take into account the last term in Eq. (13) which leads to the local-field correction given in Eq. (18). The two-particle correlation function, appearing there, is governed by Eq. (19), where we should now replace $\vec{q}$ by $\vec{q}-\vec{q}\,'$. Using the same argument as before we conclude that the terms proportional to ω and q, and associated with the free-particle behavior, dominate over all the terms arising from the Coulomb interaction. Neglecting the latter we get

$$\bar{f}^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q}-\vec{q}\,',\vec{q}\,';\omega)=\frac{1}{\hbar\omega-(\hbar^2/m)\vec{k}\cdot(\vec{q}-\vec{q}\,')-(\hbar^2/m)\vec{k}'\cdot\vec{q}\,'}\times[\bar{F}^{\rm ext}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q}-\vec{q}\,',\vec{q}\,';\omega)+\bar{F}^{\rm ext}_{\vec{k}'\sigma';\vec{k}\sigma}(\vec{q}\,',\vec{q}-\vec{q}\,';\omega)]\,. \tag{28}$$

Inserting this into Eq. (18) and using the explicit expression for $\bar{F}^{\rm ext}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q}-\vec{q}\,',\vec{q}\,';\omega)$ given in Eq. (20), we obtain after some simple transformations

$$\begin{aligned}
v(\vec{q})\chi^0(\vec{q},\omega)G(\vec{q},\omega)\bar{n}(\vec{q},\omega)={}&-\frac{1}{V^2}\sum_{\vec{q}\,'}v(\vec{q}\,')\sum_{\vec{k}\sigma}\sum_{\vec{k}'\sigma'}\Bigg\{\left[\hbar\omega-\frac{\hbar^2}{2m}q^2-\frac{\hbar^2}{m}(\vec{k}-\tfrac{1}{2}\vec{q}\,')\cdot\vec{q}+\frac{\hbar^2}{m}(\vec{k}-\vec{k}')\cdot\vec{q}\,'\right]^{-1}\\
&\times\left[\left(\hbar\omega-\frac{\hbar^2}{2m}q^2-\frac{\hbar^2}{m}\vec{k}\cdot\vec{q}-\frac{\hbar^2}{2m}\vec{q}\cdot\vec{q}\,'\right)^{-1}-\left(\hbar\omega-\frac{\hbar^2}{2m}q^2-\frac{\hbar^2}{m}\vec{k}\cdot\vec{q}+\frac{\hbar^2}{2m}\vec{q}\cdot\vec{q}\,'\right)^{-1}\right]\\
&-\left[\hbar\omega+\frac{\hbar^2}{2m}q^2-\frac{\hbar^2}{m}(\vec{k}-\tfrac{1}{2}\vec{q}\,')\cdot\vec{q}+\frac{\hbar^2}{m}(\vec{k}-\vec{k}')\cdot\vec{q}\,'\right]^{-1}\left[\left(\hbar\omega+\frac{\hbar^2}{2m}q^2-\frac{\hbar^2}{m}\vec{k}\cdot\vec{q}-\frac{\hbar^2}{2m}\vec{q}\,'\cdot\vec{q}\right)^{-1}\right.\\
&\left.-\left(\hbar\omega+\frac{\hbar^2}{2m}q^2-\frac{\hbar^2}{m}\vec{k}\cdot\vec{q}+\frac{\hbar^2}{2m}\vec{q}\,'\cdot\vec{q}\right)^{-1}\right]\Bigg\}f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(-\vec{q}\,')\Phi^{\rm ext}(\vec{q},\omega)\\
&-\frac{1}{V^2}\sum_{\vec{q}\,'}v(\vec{q}+\vec{q}\,')\sum_{\vec{k}\sigma}\sum_{\vec{k}'\sigma'}\Bigg\{\left[\hbar\omega-\frac{\hbar^2}{2m}q^2-\frac{\hbar^2}{m}(\vec{k}+\tfrac{1}{2}\vec{q}\,')\cdot\vec{q}\right]^{-1}\\
&-\left[\hbar\omega+\frac{\hbar^2}{2m}q^2-\frac{\hbar^2}{m}(\vec{k}-\tfrac{1}{2}\vec{q}\,')\cdot\vec{q}\right]^{-1}\Bigg\}\Bigg\{\left[\hbar\omega-\frac{\hbar^2}{2m}q^2-\frac{\hbar^2}{m}(\vec{k}'+\tfrac{1}{2}\vec{q}\,')\cdot\vec{q}+\frac{\hbar^2}{m}(\vec{k}-\vec{k}')\cdot\vec{q}\,'\right]^{-1}\\
&-\left[\hbar\omega+\frac{\hbar^2}{2m}q^2-\frac{\hbar^2}{m}(\vec{k}'-\tfrac{1}{2}\vec{q}\,')\cdot\vec{q}+\frac{\hbar^2}{m}(\vec{k}-\vec{k}')\cdot\vec{q}\,'\right]^{-1}\Bigg\}f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(-\vec{q}\,')\Phi^{\rm ext}(\vec{q},\omega)\,.
\end{aligned} \tag{29}$$

For consistency we should only keep the leading contribution of the above expression in the limit of large ω or large q. This leads to considerable simplifications, and as shown in the Appendix we obtain for frequencies and wave vectors satisfying the condition (27)

$$G(\vec{q},\omega)=\alpha(\vec{q},\omega)\frac{1}{N}\sum_{\vec{q}\,'}\frac{(\vec{q}\cdot\vec{q}\,')^2}{q^4}\frac{v(\vec{q}\,')}{v(\vec{q})}[S(\vec{q}\,')-1]-\frac{1}{N}\sum_{\vec{q}\,'}\left(\frac{\vec{q}\cdot(\vec{q}+\vec{q}\,')}{q^2}\right)^2\frac{v(\vec{q}+\vec{q}\,')}{v(\vec{q})}[S(\vec{q}\,')-1]\,, \tag{30}$$

where

$$\alpha(\vec{q},\omega)+\frac{1}{2}\left(\frac{[\hbar\omega+(\hbar^2/2m)q^2]^2}{[\hbar\omega-(\hbar^2/2m)q^2]^2}+\frac{[\hbar\omega-(\hbar^2/2m)q^2]^2}{[\hbar\omega+(\hbar^2/2m)q^2]^2}\right). \tag{31}$$

This result is valid for an arbitrary potential $v(\vec{q})$, provided only that the sums over $\vec{q}\,'$ are convergent in the absolute sense. Several approximative theories lead to expressions for the local-field correction containing integrals over the static structure factor similar to those in the above equation.[10–16] However, in general this comes about because one has explicitly introduced the static pair distribution function through an approximative factorization of the nonequilibrium two-particle distribution function. No such approximation has been made in the present treatment. We note that the second term in Eq. (30) is independent of the frequency, and it can be traced back to the term $\bar{F}_{\vec{k}'\sigma';\vec{k}\sigma}(\vec{q},\vec{q}\,';\omega)$ in Eq. (19). As was discussed previously, it is associated with the dynamics of the correlation hole, and it represents a proper local-field correction. The first term in Eq. (30) arises from the term $\bar{F}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q},\vec{q}\,';\omega)$ in Eq. (19) and it has to do with the motion of a particle inside its correlation hole. We note that this term has a drastic frequency dependence with a strong peak around the free-particle energy $(\hbar^2/2m)q^2$.

Specializing Eq. (30) to the case when q is finite but ω tends to infinity we get

$$\lim_{\omega\to\infty}G(\vec{q},\omega)=\frac{1}{N}\sum_{\vec{q}\,'}\left\{\left(\frac{\vec{q}\cdot\vec{q}\,'}{q^2}\right)^2\frac{v(\vec{q}\,')}{v(\vec{q})}-\left[\frac{\vec{q}\cdot(\vec{q}+\vec{q}\,')}{q^2}\right]^2\frac{v(\vec{q}+\vec{q}\,')}{v(\vec{q})}\right\}$$

$$\times[S(\vec{q}\,')-1]\,, \tag{32}$$

and it can easily be verified that the same expression is obtained by using the third-moment sum rule. In particular, for a Coulomb potential we have

$$\lim_{\omega\to\infty} G(\vec{q},\omega) = \frac{1}{N}\sum_{\vec{q}\,'}\left\{\frac{(\vec{q}\cdot\vec{q}\,')^2}{q^2q'^2} - \frac{[\vec{q}\cdot(\vec{q}+\vec{q}\,')]^2}{q^2(\vec{q}+\vec{q}\,')^2}\right\} \times[S(\vec{q}\,')-1]\,, \tag{33}$$

which is the form used for numerical calculations by Pathak and Vashishta.[16] In the opposite limit, when ω is finite but q tends to infinity, we get

$$\lim_{q\to\infty} G(\vec{q},\omega) = \frac{1}{N}\sum_{\vec{q}\,'}\frac{(\vec{q}\cdot\vec{q}\,')^2}{q^4}\frac{v(\vec{q}\,')}{v(\vec{q})}[S(\vec{q}\,')-1]+1-g(0) \tag{34}$$

where we have used Eq. (12) to bring in the value of the pair distribution function at $r=0$. The above equation can be compared with the relation

$$\lim_{q\to\infty} G(\vec{q},\omega)=1-g(0)\,, \tag{35}$$

derived by Shaw[19] within the framework of an approximative theory by Singwi *et al.*[10] Equation (35) has sometimes been referred to as an exact condition on the local-field correction, but Eq. (34) shows that this is true only if we exclude from the local field the term associated with the motion of a particle inside its correlation hole.

Including this term and specializing to a Coulomb potential we obtain from Eq. (34)

$$\lim_{q\to\infty} G(\vec{q},\omega)=\tfrac{2}{3}[1-g(0)]\,. \tag{36}$$

In particular, in the Hartree-Fock approximation we have $g(0)=\frac{1}{2}$ and hence

$$\lim_{q\to\infty} G^{\mathrm{HF}}(\vec{q},\omega)=\tfrac{1}{3}\,, \tag{37}$$

in agreement with the result derived by Geldart and Taylor.[4,20] In general, the value of $g(0)$ must lie between 0 and $\frac{1}{2}$ which means that

$$\tfrac{1}{3}<\lim_{q\to\infty} G(\vec{q},\omega)<\tfrac{2}{3}\,. \tag{38}$$

It is perhaps worth noting that Eq. (36) is identical with what we obtain by specializing the high-frequency result in Eq. (33) to large wave vectors. The mathematical reason for this coincidence is that the function $\alpha(\vec{q},\omega)$, given in Eq. (31), tends to unity both for large wave vectors and large frequencies.

Using Eq. (17) we can write the response function $\chi(\vec{q},\omega)$ as

$$\chi(\vec{q},\omega)=\frac{\chi^0(\vec{q},\omega)}{1-v(\vec{q})[1-G(\vec{q},\omega)]\chi^0(\vec{q},\omega)}\,. \tag{39}$$

For large values of q the term proportional to $v(\vec{q})$ in the denominator is small, and hence we can write

$$\chi(\vec{q},\omega)=\chi^0(\vec{q},\omega)\,\{1+v(\vec{q})[1-G(\vec{q},\omega)]\chi^0(\vec{q},\omega)\}\,. \tag{40}$$

This expression can be used to study the behavior of the static structure factor for large $\vec{q}$ vectors. Using the fluctuation-dissipation theorem,

$$S(\vec{q})=-\frac{\hbar}{\pi n}\int_0^\infty \mathrm{Im}\,\chi(\vec{q},\omega)\,d\omega\,, \tag{41}$$

together with the asymptotic expressions for $\chi^0(\vec{q},\omega)$ and $G(\vec{q},\omega)$, given in Eqs. (26) and (30), we obtain for a Coulomb potential

$$S(\vec{q})-1=-\frac{8\pi e^2 mn}{\hbar^2 q^4}g(0)\,. \tag{42}$$

On the other hand, it follows from the general properties of Fourier transforms that

$$\lim_{q\to\infty} q^4[S(\vec{q})-1]=-8\pi n g'(0)\,, \tag{43}$$

where $g'(0)$ is the radial derivative of $g(\vec{r})$ at $r=0$. Comparing Eqs. (42) and (43) we conclude that

$$\frac{g'(0)}{g(0)}=\frac{me^2}{\hbar^2}=\frac{1}{a}\,, \tag{44}$$

where a is the Bohr radius. This exact relation between the slope of the pair distribution function and its value at $r=0$ has previously been derived by Kimball using a completely different method.[23] His argument was that when two electrons are very close to each other their mutual interaction dominates over the interactions with the surrounding particles and hence one needs only solve the two-body problem. In the present treatment we have used no such arguments, but we claim that our expression for $G(\vec{q},\omega)$ is exact for large q or ω and hence that the two-body aspects of the problem are automatically included.

VI. CONCLUDING REMARKS

The idea of a local-field correction has proved to be very fruitful, and numerical calculations have led to significant improvements over the random-phase approximation. However, there is still a considerable lack of understanding of various basic physical aspects. First of all, the very concept of an effective field described by some function $G(\vec{q},\omega)$ is questionable because electrons with different momenta have different surroundings, and hence the force acting on an electron depends on its momentum. Secondly, granting that the concept is physically meaningful, there is still the question of how an electron responds to the effective field. In particular, one needs a better understanding of the motion of an electron relative to the surrounding correlation hole. This aspect of the problem is essential for the frequency dependence of the response function, as was made obvious by Goodman and Sjölander in the magnetic case.[24] Most theories have been based on the assumption that $G(\vec{q},\omega)$ is independent of frequency, but there is no good justification for this assumption. For instance, Eq. (30) shows that for large $\vec{q}$ vectors there is a very drastic frequency dependence around the free-particle energy. For smaller $\vec{q}$ vectors the frequency dependence is presumably less drastic so that $G(\vec{q},\omega)$ varies smoothly over the region of particle-hole excitations, but it may still be essential.

In order to clarify the problems discussed above it is essential to have as a starting point a rigorous formulation where different physical aspects enter in a clear-cut way. It is our hope that Eq. (19) which governs the response of the two-particle correlations to an external field can be used for this purpose. Although it is a very complicated equation, the physical meaning of the various terms is quite clear. In order to obtain explicit results it is certainly necessary to make drastic approximations, but hopefully one should at least be able to gain a better understanding of the limitations and the merits of different approximative theories.

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APPENDIX: MATHEMATICAL DETAIL

We shall here discuss in some detail the steps leading from Eq. (29) to Eq. (30). We start by expanding all the terms on the right-hand side of Eq. (29) in inverse powers of $[\hbar\omega \pm (\hbar^2/2m)q^2]$. For instance, we have

$$\left[\hbar\omega - \frac{\hbar^2}{2m}q^2 - \frac{\hbar^2}{m}(\vec{k}+\tfrac{1}{2}\vec{q}\,')\cdot\vec{q}\right]^{-1}$$

$$= \left[\hbar\omega - \frac{\hbar^2}{2m}q^2\right]^{-1} + \left[\hbar\omega - \frac{\hbar^2}{2m}q^2\right]^{-2}\frac{\hbar^2}{m}(\vec{k}+\tfrac{1}{2}\vec{q}\,')\cdot\vec{q} + \left[\hbar\omega - \frac{\hbar^2}{2m}q^2\right]^{-3}\left[\frac{\hbar^2}{m}(\vec{k}+\tfrac{1}{2}\vec{q}\,')\cdot\vec{q}\right]^2 + \cdots , \qquad (A1)$$

and similar expansions can be made of all the other terms. For finite values of k and q', the above expansion is obviously rapidly convergent when $q\to\infty$ or $\omega\to\infty$. However, k and q' are integration variables which can be arbitrarily large, and the question arises whether one can interchange the order between the limiting procedure and the integration. There is clearly no difficulty if all the terms in the expansion lead to convergent integrals, but because the terms contain successively higher powers of the integration variables it may well happen that divergencies appear at some step in the expansion. To clarify this point, let us first consider the simple one-dimensional integral

$$I(z) = \int_0^\infty \frac{1}{z-x+i\epsilon} f(x)\,dx , \qquad (A2)$$

which has a structure somewhat similar to the integrals in Eq. (27). Expanding in inverse powers of z we obtain

$$I(z) = \frac{1}{z}\int_0^\infty f(x)\,dx + \frac{1}{z^2}\int_0^\infty x f(x)\,dx + \cdots . \qquad (A3)$$

It is an easy mathematical exercise to prove that the first term in this expansion is the dominant term for large values of z, provided that the integral $\int_0^\infty f(x)\,dx$ is nonzero and convergent in the absolute sense. If the integral $\int_0^\infty xf(x)\,dx$, appearing in the next-order term, happens to be divergent it means only that the leading correction has a nonanalytic behavior. Returning now to Eq. (29) we observe that the terms on the right-hand side can be reduced to a sum of integrals of the form given in Eq. (A2), except that the integral over x is replaced by three-dimensional integrals over $\vec{k}$, $\vec{k}'$, and $\vec{q}\,'$. This is not expected to be an essential difference, and we conclude that we can freely use expansions of the type indicated in Eq. (A1) as long as the integrals appearing in the final answer are convergent in the absolute sense.

Collecting all terms of lowest non-vanishing order in $[h\omega \pm (h^2/2m)q^2]^{-1}$ on the right side of Eq. (29) we obtain

$$v(\vec{q})\chi^0(\vec{q},\omega)G(\vec{q},\omega)\bar{n}(\vec{q},\omega) = \frac{1}{2}\left(\frac{\hbar^2}{m}\right)^2\left[\left(\hbar\omega - \frac{\hbar^2}{2m}q^2\right)^{-4} + \left(\hbar\omega + \frac{\hbar^2}{2m}q^2\right)^{-4}\right]$$

$$\times \frac{1}{V^2}\sum_{\vec{q}'} v(\vec{q}')(\vec{q}\cdot\vec{q}')^2 \sum_{\vec{k}\sigma}\sum_{\vec{k}'\sigma'} f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(-\vec{q}')\Phi^{\text{ext}}(\vec{q},\omega)$$

$$-3\left(\frac{\hbar^2}{m}\right)^2\left[\left(\hbar\omega-\frac{\hbar^2}{2m}q^2\right)^{-4}-\left(\hbar\omega+\frac{\hbar^2}{2m}q^2\right)^{-4}\right]$$

$$\times \frac{1}{V^2}\sum_{\vec{q}'} v(\vec{q}')\vec{q}\cdot\vec{q}' \sum_{\vec{k}\sigma}\sum_{\vec{k}'\sigma'} \vec{k}\cdot\vec{q} f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(-\vec{q}')\Phi^{\text{ext}}(\vec{q},\omega)$$

$$-\left(\frac{\hbar^2}{m}\right)^2\left[(\hbar\omega)^2-\left(\frac{\hbar^2}{2m}q^2\right)^2\right]^{-1}$$

$$\times \frac{1}{V^2}\sum_{\vec{q}'} v(\vec{q}+\vec{q}')[\vec{q}\cdot(\vec{q}+\vec{q}')]^2 \sum_{\vec{k}\sigma}\sum_{\vec{k}'\sigma'} f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(-\vec{q}')\Phi^{\text{ext}}(\vec{q},\omega)\ . \qquad \text{(A4)}$$

In the derivation we have made use of the obvious symmetry relation:

$$f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(-\vec{q}') = f^{(2)}_{-\vec{k}\sigma;-\vec{k}'\sigma'}(\vec{q}')\ , \qquad \text{(A5)}$$

which makes that certain terms vanish identically. Furthermore, the second term in Eq. (A4) vanishes because

$$\sum_{\vec{k}\sigma}\sum_{\vec{k}'\sigma'} \vec{k}\cdot\vec{q} f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(-\vec{q}') = 0\ . \qquad \text{(A6)}$$

This can be proved as follows. We first observe that the symmetry requirement makes that only the longitudinal part can be nonzero:

$$\sum_{\vec{k}\sigma}\sum_{\vec{k}'\sigma'} \vec{k}\cdot\vec{q} f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(-\vec{q}') = \frac{\vec{q}\cdot\vec{q}'}{q'^2}\sum_{\vec{k}\sigma}\sum_{\vec{k}'\sigma'} \vec{k}\cdot\vec{q}' f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(-\vec{q}')\ . \qquad \text{(A7)}$$

Secondly, from the conservation of particle number it follows that

$$i\hbar\left(\frac{d}{dt}\langle\rho(\vec{q}',t)\rho^\dagger(\vec{q}',0)\rangle\right)_{t=0} = \frac{\hbar^2}{m}\sum_{\vec{k}\sigma}\sum_{\vec{k}'\sigma'} \vec{k}\cdot\vec{q}'\langle a^\dagger_{\vec{k}-\vec{q}'/2,\sigma} a_{\vec{k}+\vec{q}'/2,\sigma} a^\dagger_{\vec{k}'+\vec{q}'/2,\sigma'} a_{\vec{k}'-\vec{q}'/2,\sigma'}\rangle\ , \qquad \text{(A8)}$$

where the expectation value on the right-hand side differs from $f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(\vec{q}')$ only through the ordering of the operators [cf. Eqs. (9) and (10)]. Using the commutation rules we get

$$\frac{\hbar^2}{m}\sum_{\vec{k}\sigma}\sum_{\vec{k}'\sigma'} \vec{k}\cdot\vec{q}' f^{(2)}_{\vec{k}\sigma;\vec{k}'\sigma'}(-\vec{q}') = \frac{N\hbar^2}{2m}q'^2 - i\hbar\left(\frac{d}{dt}\langle\rho(\vec{q}',t)\rho^\dagger(\vec{q}',0)\rangle\right)_{t=0} \qquad \text{(A9)}$$

which is zero because of the first-moment sum rule.[17,18]

It is now straightforward to obtain Eq. (30) from Eq. (29), using the asymptotic form for $\chi^0(\vec{q},\omega)$ and $\chi(\vec{q},\omega)$, given in Eq. (26), and the expression for the static structure factor, given in Eq. (11).

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