Short-Range Correlations and Electron-Gas Response Functions

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The behavior of interacting electrons is dominated by the Coulomb repulsion in the limit of zero interparticle separation. Hence, in this limit the derivative of the two-particle correlation function is equal to the two-particle correlation function itself divided by the Bohr radius. Because correlation functions can be obtained from linear-response functions, the relation between the correlation function and its derivative can be used as a criterion for the validity of approximate linear-response functions. Some of the more familiar dielectric functions and spin susceptibilities are discussed in this context. For these response functions it is shown that the electric and magnetic response should approach the same ($q \to \infty$) asymptotic limit.

A gas of interacting electrons imbedded in a uniform positive background is an unsolved theoretical model of a real metal. A great deal of information on the properties of this model could be obtained from a knowledge of its linear-response functions.[1] The ground-state energy, plasmon dispersion relations, correlation functions, and the compressibility can all be obtained simply from the dielectric function.

The electron gas (at zero temperature) is characterized by one parameter—the density. For very high densities, a simple approximate self-consistent calculation of the dielectric and magnetic response, the random-phase approximation (RPA), is expected to give accurate results because the kinetic energy is dominant. For an electronic density corresponding to the conduction-electron density in real metals, corrections to the RPA are believed to be important. These "exchange and correlation" corrections cannot be calculated exactly, but a number of approximate expressions for these corrections have been suggested.[2-7] There are a number of checks on the validity of approximate response functions, including oscillator-strength sum rules and the compressibility sum rule.[8]

We will show that simple physical arguments on the form of the two-particle correlation functions can be used to obtain a criterion for the validity of linear-response functions. For simplicity the dielectric function will be discussed first and the generalization to the magnetic (spin) response function will follow.

As has been frequently emphasized, a dielectric function should yield a positive two-particle correlation function $g(r)$. Physically, $g(r)$ is proportional to the probability of finding two particles separated by a distance r. This function is normalized so that $g(r) \to 1$ for large r. We would like to point out here another condition which $g(r)$ should satisfy. This condition is

$$\left. \frac{\partial g(r)}{\partial r} \right|_{r=0} = \frac{1}{a_0} g(0) , \qquad (1)$$

where a_0 is the Bohr radius. This relation follows because when two electrons approach sufficiently close to each other, the "pair wave function" $\psi(r)$ and the pair correlation function which is proportional to $|\psi(r)|^2$ are determined by the solution to the two-particle Schrödinger equation

$$\left(-\frac{\hbar^2}{2\mu} \frac{\partial^2}{\partial r^2} - \frac{\hbar^2}{\mu r} \frac{\partial}{\partial r} + \frac{e^2}{r} \right) \psi(r) = \mathcal{E}\psi(r) . \qquad (2)$$

For simplicity, it is assumed that the two electrons are in a relative s state. Higher angular-momentum terms in $\psi(r)$ do not contribute to $g(r)$ or $g'(r)$ as r approaches zero. The symbol $\mathcal{E}$ represents a complicated operator, but as long as it is bounded it does not influence the results. The mass μ is $\frac{1}{2}$ the electron mass because r is the relative separation of the two electrons. Expanding $\psi(r)$ in a power series in r,

$$\psi(r) = a + br + \cdots , \qquad (3)$$

and equating terms diverging like $1/r$ as $r \to 0$ on the left-hand side of Eq. (2),

$$-(\hbar^2/\mu) b + e^2 a = 0 , \qquad (4)$$

leads to

$$\psi(r) \approx 1 + (1/2a_0) r , \qquad (5)$$

and since $g(r)$ is proportional to $\psi^*(r)\psi(r)$,

$$\left. \frac{\partial g(r)}{\partial r} \right|_{r=0} = \frac{1}{a_0} g(0) . \qquad (1)$$

A more formal discussion of this result is presented in the Appendix.

The derivative of $g(r)$ at $r = 0$ determines the large-q limit of the structure factor $S(q)$. Since $S(q)$ can be obtained from the dielectric function, this asymptotic limit and Eq. (1) can be used as a criterion for the validity of approximate dielec-

tric functions. The pair correlation function is given by

$$1 - g(r) = \frac{3}{2rq_F^3} \int_0^\infty q \sin qr \, [1 - S(q)] dq \; , \quad (6)$$

where q_F is the magnitude of the Fermi wave vector. When q is large, $1 - S(q)$ can be expanded in powers of $1/q^2$. The constant and $1/q^2$ terms must be zero if $g(r)$ is to be finite. The following equations define $s(q)$, a function which goes to zero like $1/q^6$ for large q:

$$s(q) = 1 - S(q) - \alpha/(1 + q^2)^2 \; , \quad (7)$$

where

$$\alpha = \lim \{ q^4 [1 - S(q)] \} \text{ as } q \to \infty \; . \quad (8)$$

Using Eq. (6), the pair correlation function is then

$$1 - g(r) = \frac{3\pi\alpha}{8q_F^3} e^{-r} + \frac{3}{2rq_F^3} \int_0^\infty q \sin qr \, s(q) dq \; . \quad (9)$$

The rapid convergence $s(q)$ means that $g'(r)$ can be obtained by explicitly differentiating the first term on the right-hand side of Eq. (9) and adding it to the second term, which can be obtained by differentiation within the integral sign. For $r = 0$, this second term is zero, and from Eqs. (8) and (9),

$$\left. \frac{\partial g(r)}{\partial r} \right|_{r=0} = \frac{3\pi}{8q_F^3} \lim \{ q^4 [1 - S(q)] \} \; . \quad (10)$$

From Eq. (1) we obtain the self-consistency relation between the large-q limit of $S(q)$ and $g(0)$:

$$g(0) = \frac{3\pi a_0}{8q_F^3} \lim_{q \to \infty} \{ q^4 [1 - S(q)] \} \; . \quad (11)$$

The following is a review of the dielectric function with emphasis on the general analytic structure. Approximate forms of this linear-response function and the implication of Eq. (11) will be discussed later. Neglecting electron-electron interactions, the polarization of an electron gas resulting from a small externally applied potential $V_0(q, \omega)$ can be calculated from perturbation theory. This polarizability is

$$\chi_0(q, \omega) = \frac{1}{N} \sum_{\vec{k}} \frac{f(\vec{k}) - f(\vec{k} + \vec{q})}{\epsilon_{\vec{k}+\vec{q}} - \epsilon_{\vec{k}} - \hbar\omega} \; , \quad (12)$$

where N is the number of electrons, $f(\vec{k})$ is a Fermi function for an electron with wave vector $\vec{k}$, and $\epsilon_{\vec{k}}$ is its energy. The electrostatic potential resulting from this polarization is $V_{pol} = v_q V_0 \chi_0$, where $v_q = 4\pi e^2/q^2$. Interactions among the electrons lead to further response of the electron gas to the potential produced by its own polarization V_{pol}. However, the electronic response to the induced potential is not simply $V_{pol} \chi_0$, because the electrons tend to avoid each other. Instead, the electrons can be viewed as responding to an effective potential

V_{eff} created by the polarization potential:

$$V_{eff}(q, \omega) = [1 - G(q, \omega)] V_{pol}(q, \omega) \; . \quad (13)$$

The induced potential is then given by

$$V_{pol} = -(V_0 + V_{eff}) v_q \chi_0 \; . \quad (14)$$

The dielectric function $\epsilon(q, \omega)$ is defined as the ratio of the externally applied potential to the total potential:

$$V_0/\epsilon = V_0 + V_{pol} \; , \quad (15)$$

which means that

$$\epsilon(q, \omega) = 1 + \frac{v_q \chi_0(q, \omega)}{1 - G(q, \omega) v_q \chi_0(q, \omega)} \; . \quad (16)$$

Calculating $G(q, \omega)$ is, of course, the major difficulty. In the random-phase approximation (RPA), $G(q, \omega) = 0$ and $\epsilon_{RPA}(q, \omega) = 1 + v_q \chi_0(q, \omega)$ because the electrons are assumed to respond in the same way to the potential produced by the induced polarization as they do to the external potential.

As a function of ω, the analytic properties of $G(q, \omega)$ are similar to those of $\epsilon(q, \omega)$; it is analytic in the upper half of the complex plane and is bounded as $\omega \to \infty$, and $G^*(q, -\omega^*) = G(q, \omega)$. This follows because both dielectric functions have these properties along with the additional property that $\epsilon(q, \omega) - 1 \neq 0$ for $\text{Im}(\omega) > 0$. We write $G(q, \omega)$ as

$$G(q, \omega) = -\left(\frac{1}{\epsilon(q, \omega) - 1} - \frac{1}{\epsilon_{RPA}(q, \omega) - 1} \right) \quad (17)$$

using Eq. (16) and the definition of ϵ_{RPA}. Since $\epsilon(q, \omega) - 1$ is analytic and nonzero for $\text{Im}(\omega) > 0$, its inverse must be analytic as well. $G(q, \omega)$ is then the difference of two analytic functions and so must be analytic in their common domain of analyticity—the upper half of the complex ω plane. For large ω, both dielectric functions can be written as a power series in $1/\omega$ and for both dielectric functions

$$\epsilon(q, \omega) - 1 = -\omega_p^2/\omega^2 + O(1/\omega^3) \; , \quad (18)$$

where ω_p is the plasma frequency. Substituting the expansion of Eq. (18) into Eq. (17), we have

$$G(q, \omega) = G^0(q) + g(q, \omega) \; , \quad (19)$$

where $G^0(q)$ is real and $g(q, \omega) \to 0$ for $\omega \to \infty$. These analytic properties of $G(q, \omega)$ allow a causal interpretation of the relationship between the polarization potential V_{pol} and the effective potential V_{eff} mentioned earlier.

Approximations are necessary in order to obtain actual values for the dielectric function. As was mentioned, $G(q, \omega)$ is assumed to be zero in the RPA. Most approximate forms for $G(q, \omega)$ which are numerically tractable assume that $G(q, \omega)$ is frequency independent: $G(q, \omega) \to G(q)$. The "optimum" $G(q)$ may not be the $G^0(q)$ of Eq. (19).

Perhaps the best-known approximations of this type are those of Singwi *et al.*,[2] in which V_{eff} and V_{pol} are related to each other through the correlation function $g(r)$. If it is assumed that $G(q, \omega)$ is independent of frequency, it is relatively easy to calculate $S(q)$ from the dielectric function, and the self-consistency criterion of Eq. (11) can be applied to find the asymptotic form of $G(q)$.

The structure factor is given as a frequency integral of the imaginary part of the inverse dielectric function

$$S(q) = \frac{-3q^2}{2e^2q_F^2} \int_0^\infty \text{Im}\left(\frac{1}{\epsilon(q, \omega)}\right) d\omega . \quad (20)$$

Only $\chi_0(g. \omega)$ has an imaginary part if $G(q)$ is frequency independent. For large q, $\chi_0(q, \omega)$ is complex only when

$$\frac{\hbar q^2}{2m} - \frac{\hbar q q_F}{2m} \leqslant \omega \leqslant \frac{\hbar q^2}{2m} + \frac{\hbar q q_F}{2m} ,$$

assuming $\epsilon_k = (\hbar k)^2/2m$. The condition that $q^2[1 - S(q)]$ approach zero for large q [see Eq. (6) and following comments] will be satisfied only if $G(q)$ does not diverge for large q, and if $G(q)$ does not diverge, the denominator of

$$\frac{1}{\epsilon(q, \omega)} = 1 - \frac{\chi_0 v_q}{1 + (1 - G)\chi_0 v_q}$$

can be expanded in a power series in $1/q^2$. To order $1/q^4$ the frequency integrals can be done, and we find

$$1 - S(q) = \frac{8 q_F^3}{3 \pi a_0 q^4} [1 - G(q)] + O\left(\frac{1}{q^6}\right) . \quad (21)$$

Combining Eqs. (11) and (21), the final form of the self-consistency condition is

$$g(0) = \lim[1 - G(q)] \text{ as } q \to \infty . \quad (22)$$

A realistic calculation of the magnetic susceptibility of an electron gas is more difficult than the corresponding calculation of the dielectric function because of the Lorentz force. We will discuss here only the effect of the magnetic field on the electron spin and ignore spin-orbit interactions. There is no clear proof that this "spin susceptibility" is a good approximation to the total magnetic response of interacting electrons, because the Landau–Peierls orbital diamagnetism has been neglected.

When only the interactions of the spins with the magnetic field are considered, the calculation of the susceptibility is closely analogous to the dielectric calculation. An infinitesimal external magnetic field $B(q, \omega)$ is directed along the z axis and produces a spin-dependent potential. Spin-up electrons are subject to an external potential $V_{0+}(q, \omega) = \mu_e B(q, \omega)$. ($\mu_e$ is the magnetic moment of the electron.) Spin-down electrons are sub-

jected to the same potential with the opposite sign. Neglecting interactions, the magnetic polarization of the electron gas is related to the external potential by the same polarizability $[\chi_0(q, \omega)]$ which determined the dielectric response. The resulting magnetization is

$$M(q, \omega) = \mu_e^2 \chi_0(q, \omega) B(q, \omega) . \quad (23)$$

When electron-electron interactions are included, this magnetization leads to an effective magnetic field B_{eff}. The effective field arises because spin-up electrons respond differently to the (spatial) polarization of spin-up electrons than they do to polarization of spin-down electrons. When calculating the dielectric response it was not necessary to distinguish the effects of parallel and antiparallel electrons on each other. However, the function $G(q, \omega)$ could have been written as

$$G(q, \omega) = \tfrac{1}{2}[G_p(q, \omega) + G_a(q, \omega)], \quad (24)$$

where the subscripts (p for parallel and a for antiparallel) denote the relative spin orientations of the electrons producing the effective potential and the electrons experiencing that potential. The functions G_p and G_a need not be the same. The distinction between G_a and G_p becomes important when calculating the magnetic response because the induced effective magnetic field is proportional to the difference between these two functions[9]:

$$B_{eff} = \tilde{G} M / \mu_e^2 , \quad (25)$$

where

$$\tilde{G} = \tfrac{1}{2}(G_p - G_a) . \quad (26)$$

The total magnetization is

$$M = \mu_e^2 \chi_0(B + B_{eff}) , \quad (27)$$

which leads to

$$\chi = \mu_e^2 \frac{\chi_0}{1 - \tilde{G} v_q \chi_0} . \quad (28)$$

As was the case for the dielectric function, the real difficulty lies in obtaining an appropriate form for $\tilde{G}(q, \omega)$. This function has the same analytic properties as $G(q, \omega)$ and can also be viewed as a causal response function.

In order to relate the magnetic spin susceptibility to the pair correlation function, the spin directions must be explicitly noted. The function $g(r)$ defined earlier is actually the sum of pair correlations between electrons with parallel and antiparallel spins:

$$g(r) = \tfrac{1}{2}[g_p(r) + g_a(r)] . \quad (29)$$

Because of the Pauli exclusion principle, $g_p(r)$ and $\partial g_p(r)/\partial r$ vanish as r approaches zero and $g(r) \to \tfrac{1}{2}g_a(r)$. A Fourier transform of the magnetic structure factor $\tilde{S}(q)$ yields the two-particle spin

correlation function

$$\tilde{g}(r) = \frac{3}{2rq_F^3} \int_0^\infty q \sin qr \, [\tilde{S}(q) - 1] dq , \quad (30)$$

where

$$\tilde{g}(r) = \tfrac{1}{2}[g_p(r) - g_a(r)] , \quad (31)$$

and the magnetic structure factor can be obtained from a frequency integral of the imaginary part of the spin susceptibility:

$$\tilde{S}(q) = \frac{6\pi}{\mu_e^2 q_F^2} \int_0^\infty \mathrm{Im}[\chi(q, \omega)] d\omega . \quad (32)$$

As was done for the dielectric function, we examine the implications of the standard approximation in which $\tilde{G}(q, \omega)$ is assumed to be frequency independent, $\tilde{G}(q, \omega) \to \tilde{G}(q)$. If this approximation is made, the large-q limit of $\tilde{S}(q)$ can be obtained from an expansion of $\chi(q, \omega)$. This leads to

$$1 - \tilde{S}(q) = - \frac{8 q_F^3}{3 \pi a_0 q^4} \tilde{G}(q) + O\left(\frac{1}{q^6}\right) . \quad (33)$$

As in the dielectric case, the asymptotic form of $\tilde{S}(q)$ determines the short-range limit of the spin correlation function:

$$\tilde{g}(0) = \frac{3\pi a_0}{8 q_F^2} \lim_{q \to \infty} \{q^4 [1 - \tilde{S}(q)]\} , \quad (34)$$

so that

$$\tilde{g}(0) = - \lim [\tilde{G}(q)] \text{ as } q \to \infty . \quad (35)$$

Combining Eq. (35) with Eq. (22), and considering parallel and antiparallel spins separately, we have

$$g_p(0) = 1 - G_p(\infty), \quad g_a(0) = 1 - G_a(\infty) . \quad (36)$$

The restriction that $g_p(0) = 0$ means that

$$G_p(\infty) = 1$$

and $\qquad\qquad\qquad\qquad\qquad\qquad\quad (37)$

$$G_a(\infty) = 1 - 2g(0) .$$

Finally, we note that the assumption that $G(q, \omega)$ and $\tilde{G}(q, \omega)$ are frequency independent implies that χ/μ_e^2 and $(\epsilon - 1)/v_q$ approach an identical large-q limit which has an intuitively appealing form,

$$\left(\frac{\chi/\mu_e^2}{(\epsilon - 1)/v_q}\right) \to \frac{\chi_0}{1 - g_0(0)v_q\chi_0} . \quad (38)$$

The large-q "exchange enhancement" is given simply in terms of the zero-range two-particle correlations.

Shaw[10] has derived the relation between $g(0)$ and $G(\infty)$ [Eq. (22)] by utilizing an approximate dielectric function proposed by Singwi et al.[2] Our Eq. (37) is a generalization of Shaw's result. Over-hauser[6] used this asymptotic condition in his simplified dielectric function. Vashista and Singwi[11] have argued that since Shaw's derivation was based

on a particular approximate dielectric function, his results would not be true in general. We have shown here that Eq. (22) should be satisfied by any approximate dielectric function for which $G(q, \omega)$ is assumed to be independent of ω if $g(r)$ is to have the right properties at $r = 0$.

Of those approximate dielectric functions which ignore the frequency dependence of $G(q, \omega)$ (most notably the RPA, the Hubbard approximation, and the various approximations of Singwi et al.), only the first two approximations of Singwi et al. satisfy the condition of Eq. (22). Although we consider this point to be in favor of these two approximations, it should be remembered that these schemes have some other difficulties. In particular, $g(r)$ itself is negative for small r and moderate or low electron densities.

In recent times there has been some controversy over the merit of various response functions. Clearly the assumption that $G(q, \omega)$ and $\tilde{G}(q, \omega)$ are frequency independent will eventually lead to inconsistencies. One such inconsistency can be seen by noting that the large-q limit of $G(q, \omega = 0)$ has been calculated by Geldart and Taylor[12] and Rajagopal.[3] This quantity is equal to $\tfrac{1}{3}$. On the other hand, if $G(q, \omega)$ is assumed to be frequency independent, then our self-consistency condition, Eq. (21), requires that $G(q \to \infty) = 1 - g(0)$, which is always between $\tfrac{1}{2}$ and 1 and is never $\tfrac{1}{3}$.

Observable quantities such as the plasmon dispersion relations are influenced by the frequency dependence of $G(q, \omega)$ and calculations of this frequency-dependent effect using a variational form of dielectric function show that it is not small.[13] We speculate that in the future our knowledge of response functions will progress significantly only if the frequency dependence of $G(q, \omega)$ and $\tilde{G}(q, \omega)$ is accepted as an unpleasant necessity.

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APPENDIX

In order to justify the results from Eq. (2), we write the total wave function with the coordinates for two of the electrons written in center-of-mass coordinates:

$$\psi = \psi(r, R, r_3, r_4, \dots, r_n) ,$$

$$r_1 = R + \tfrac{1}{2}r , \quad r_2 = R - \tfrac{1}{2}r .$$

In terms of the same coordinates, the total Hamiltonian is

$$H = H_0 + H_1 ,$$

$$H_0 = - \frac{\hbar^2}{2\mu} \nabla_r^2 + \frac{e^2}{r} ,$$

$$H_1 = -\frac{\hbar^2}{4m}\nabla_R^2 - \frac{\hbar^2}{2m}\sum_{i=3}^{n}\nabla_{r_i}^2$$

$$+ e^2\sum_{i=3}^{n}\left(\frac{1}{|r_i - (R + \frac{1}{2}r)|} + \frac{1}{|r_i - (R - \frac{1}{2}r)|}\right)$$

$$+ e^2\sum_{i>j=3}^{n}\frac{1}{|r_i - r_j|} \,.$$

A density matrix $\rho(r, r')$ is defined as

$$\rho(r, r') = \int \psi^*(r, R, r_3, \ldots, r_n)$$

$$\times \psi(r', R, r_3, \ldots, r_n) d^3R \prod_{i=3}^{n} d^3r_i$$

and the correlation function is

$$g(r) = \rho(r, r) \,.$$

Since

$$H\psi = E\psi \,,$$

$$\int (\psi^* H\psi) d^3R \prod_{i=3}^{n} d^3r_i \equiv H(r')\rho(r, r') = E\rho(r, r') \,,$$

$$H_0(r')\rho(r, r') = [E - H_1(r')]\rho(r, r') \,.$$

If $H_1(r')\rho(r, r')$ does not diverge as r and r' go to zero, the divergent terms on the left-hand side of the above equation can be equated, and Eq. (1) will be obtained. It suffices to show that individual terms from the Hamiltonian do not diverage:

$$\lim_{r,r'\to 0}\int \psi^*(r, R, r_3, \ldots, r_n)\frac{e^2}{|r_3 - (R \pm \frac{1}{2}r)|}$$

$$\times \psi(r', R, r_3, \ldots, r_n) d^3R \prod_{i=3}^{n} d^3r_i$$

$$= \int g(r_1, r_2, r_3)\frac{e^2}{|r_2 - r_3|} d^3r_3 \,.$$

This term is finite because the three-particle correlation function $g(r_1, r_2, r_3)$ does not diverge. $g(r_1, r_2, r_3)$ is proportional to finding particles simultaneously at the points r_1, r_2, and r_3:

$$\lim_{r,r'\to 0}\int \psi^*(r, R, r_3, \ldots, r_n)\frac{e^2}{|r_3 - r_4|}$$

$$\times \psi(r', R, r_3, \ldots, r_n) d^3R \prod_{i=3}^{n} d^3r_i$$

$$= \int g(r_1, r_2, r_3, r_4)\frac{e^2}{|r_3 - r_4|} d^3r_3 d^3r_4 \,.$$

This term is also finite because the four-particle correlation function is finite. The kinetic-energy terms also should not diverge. The integrals

$$\lim_{r,r'\to 0}\int \psi^*(r, R, r_3, \ldots, r_n)\left(\frac{-\hbar^2\nabla_R^2}{4m}\right)$$

$$\times \psi(r', R, r_3, \ldots, r_n) d^3R \prod_{i=3}^{n} d^3r_i$$

and

$$\lim_{r,r'\to 0}\int \psi^*(r, R, r_3, \ldots, r_n)\left(-\frac{\hbar^2}{2m}\nabla_{r_3}^2\right)$$

$$\times \psi(r', R, r_3, \ldots, r_n) d^3R \prod_{i=3}^{n} d^3r_i$$

are finite if the mean center-of-mass kinetic energy of two particles remains finite when they are close together, and if the mean kinetic energy of a third particle remains finite when two other particles are close together. On physical grounds we are convinced that these conditions are satisfied.

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