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Ornstein-Zernike function and Coulombic correlation in the homogeneous electron liquid

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Adopting the original Ornstein-Zernike (OZ) definition of the direct correlation function $c(r)$, the present study deals with the deviation $\Delta(r)$ of $c(r)$ induced by Coulomb correlation in the homogeneous electron liquid beyond the OZ function $c_{FH}(r)$ for purely Fermi hole (FH) statistical correlations. It is first stressed that $\Delta(r)$ at large r is proportional to the Coulomb potential energy e^2/r, suitably scaled with the plasma frequency. Both **r** space and **k** space formulations are presented. In **k** space, direct numerical use is made of inequalities due to Kugler [Phys. Rev. A **1**, 1688 (1970)] by employing analytic representations of the pair correlations due to Gori-Giorgi *et al.* [Phys. Rev. B **61**, 7353 (2000)] as a function of the uniform electron density. Then, in **r** space, consideration is given to differential equations proposed by Dawson and March [Phys. Chem. Liq. **14**, 131 (1984)] and also in the recent study of Nagy and Amovilli [J. Chem. Phys. **121**, 6640 (2004)]. In both approaches, one-body potentials enter, into which Coulombic interelectronic repulsions are subsumed. Finally, Gaskell's [Proc. Phys. Soc. London **77**, 1182 (1961)] variational ground-state wave function is shown to be related to the OZ direct correlation function in **k** space.

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I. INTRODUCTION

In many-electron problems, the pair density, $n_2(\mathbf{r}_1,\mathbf{r}_2)$, say, is a crucially important quantity. In the homogeneous electron liquid, which is our sole focus here, $n_2(\mathbf{r}_1,\mathbf{r}_2)$ becomes translationally invariant, i.e., a function only of $r_{12}=|\mathbf{r}_1-\mathbf{r}_2|$. If we scale appropriately, we get then, instead of n_2, the pair correlation function $g(r)$, normalized such that $g(r)\to 1$ as r tends to infinity.

Instead of working directly with $g(r)-1=h(r)$, the latter being termed the total correlation function, we shall here follow the lead of Ornstein-Zernike (OZ),[1,2] who, in treating a classical liquid such as argon near its critical point, introduced the so-called direct correlation function $c(r)$ defined by the convolution relation

$$h(r)=c(r)+\rho_0\int c(|\mathbf{r}-\mathbf{r}'|)h(r')d\mathbf{r}'. \tag{1}$$

Though we are aware that in the ground state of the homogeneous quantal electron liquid we focus on here, other definitions than the classical OZ relation [Eq. (1)] are possible for $c(r)$, we shall here define $c(r)$ by Eq. (1), where ρ_0 is now the electron number density. However, $h(r)=g(r)-1$ and the structure factor $S(k)$ (Ref. 3) are related by Fourier transform, namely,

$$S(\mathbf{k})-1=\rho_0\int h(r)\exp(i\mathbf{k}\cdot\mathbf{r})d\mathbf{r}. \tag{2}$$

Using the convolution property of Eq. (1), it then follows that the Fourier transform of $c(r)$, say, $\tilde{c}(k)$, is related to the liquid structure factor by

$$\tilde{c}(k)=1-\frac{1}{S(k)}. \tag{3}$$

In classical liquid argon, already referred to above, in which the atoms interact via the pair potential $\phi(r)$, $c(r)\approx -\phi(r)/k_BT$ at large r in a model classical liquid, in equilibrium at temperature T. In the quantal electron liquid,

$$c(r)\approx\frac{e^2}{rE_{char}}\quad(r\to\infty) \tag{4}$$

where E_{char} is a (now quantal) characteristic energy, to be discussed further below, to replace the thermal energy k_BT in argon near its triple point. Thus, the OZ function $c(r)$ reflects the Coulomb energy e^2/r very directly at large r.

However, it has to be recognized that in the spin-compensated electron liquid with which we are concerned throughout, Fermi statistical correlations exist between parallel spin electrons. There is then a nontrivial $S(k)$ and hence, through Eq. (3), a direct correlation function $\tilde{c}(k)$ even when Coulomb interactions are "switched off." Denoting the Fermi hole by FH, it is well known that $S(k)$ has the form[2,3]

$$S_{FH}(k)=a_1k+a_3k^3\quad(k\leqslant 2k_f)=1\quad(k>2k_f), \tag{5}$$

where $a_1=3/4k_f$ and $a_3=-1/16k_f^3$, k_f being the Fermi wave number. This is related to the electron density ρ_0 by

$$\rho_0=\frac{k_f^3}{3\pi^2}. \tag{6}$$

From Eqs. (5) and (3), it follows immediately that $\tilde{c}_{FH}(k)=0$ for $k>2k_f$. Its form in **r** space has been given in the recent study of Nagy *et al.*[4] The definitive theory of $c(r)$ in the homogeneous electron liquid must recover this result $c_{FH}(r)$ as the elementary charge e is mathematically allowed to tend to zero. Therefore, in Fig. 1(a), we have plotted

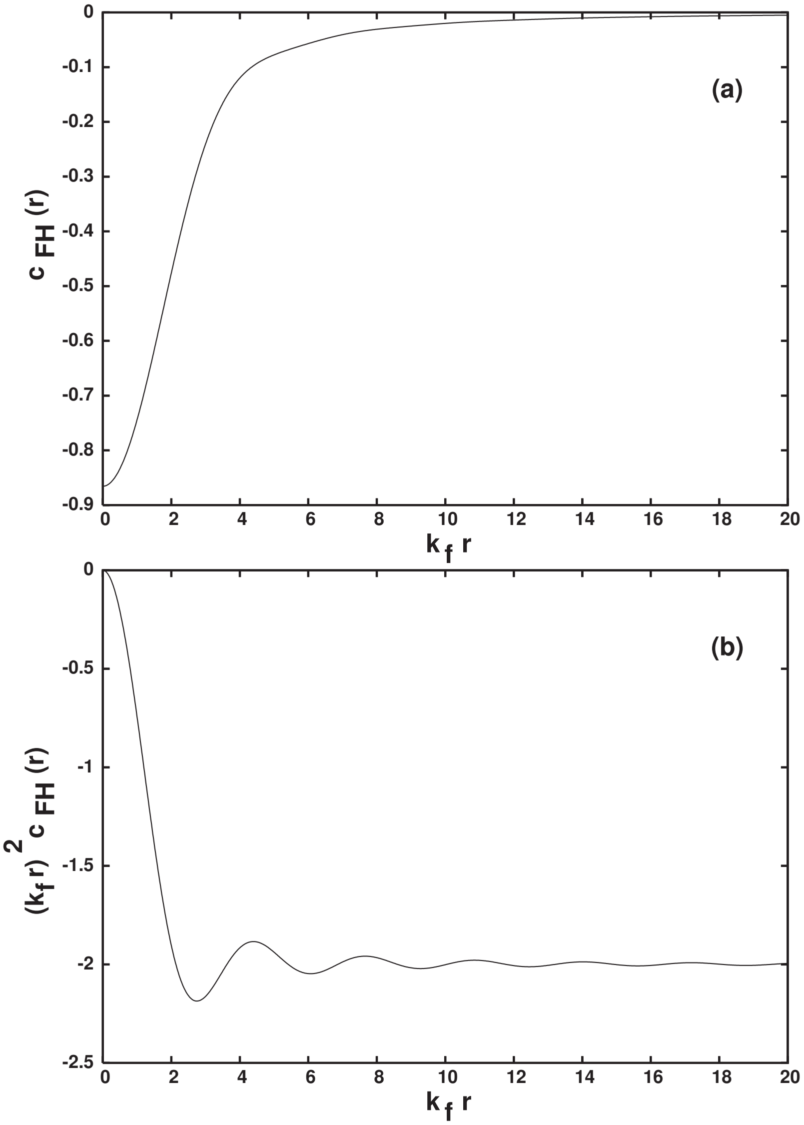

FIG. 1. (a) OZ direct correlation function $c_{FH}(r)$ obtained by a numerical Fourier transform of Eq. (3) when $S(k)$ has the Fermi hole form [Eq. (5)]. All quantities are in a.u. Note that $c_{FH}(r)$ is a universal function of $k_f r$ in this Fermi hole limit of the homogeneous electron liquid and that $c_{FH}(0)=-0.865\,58$. (b) Oscillatory behavior after multiplying $c_{FH}(r)$ by $(k_f r)^2$. It is important to stress how rapidly this plot approaches the limiting behavior $c_{FH} \propto 1/(k_f r)^2$ as $k_f r$ become large.

$c_{FH}(r)$ obtained by taking the Fourier transform of Eq. (3) when $S(k)$ has the Fermi hole form [Eq. (5)]. The important dimensionless independent variable in Fig. 1(a) is seen to be $k_f r$. The exact $c(r)$ including correlations (i.e., $r_s > 0$) does not tend smoothly to $c_{FH}(r)$ as $r_s = 0$ is a singular point. This is clear since the Fourier transform of $c(r, r_s)$, namely, $\tilde{c}(k)$ is proportional to $1/k^2$ as $k \to 0$ for $r_s > 0$, but $\tilde{c}_{FH}(k)$ goes to $1/k$ as $k \to 0$. This latter point is clear by combining Eq. (5) with the relation between $\tilde{c}$ and S in Eq. (3). What is remarkable to us is that $c_{FH}(r)$ is of a very simple form to graphical accuracy, tending at large r to an r^{-2} behavior. This follows analytically from the form $\tilde{c}_{FH} \propto 1/k$ as k tends to zero. This long range behavior, as will be demonstrated quite explicitly below, will be crucially altered by the addition of Coulombic correlations, with the direct correlation function $c(r)$ of the homogeneous electron liquid falling off as r^{-1}, i.e., having the form of the Coulomb repulsion e^2/r at sufficiently large r. In turn, this means that $\tilde{c}(k) \propto 1/k^2$ in the long wavelength limit due to the long range of the electron-electron repulsions in the model (jellium) under discussion throughout this article. We felt it of interest to show also in Fig. 1(b) a plot of $r^2 c_{FH}(r)$, which reveals some small underlying oscillatory behavior before tending to a known constant value as $r \to \infty$.

With this as background, the outline of this article is then as follows. In Sec. II, **k** space theory is considered. A formally exact treatment is first summarized, posed now in terms of the Coulomb correlation correction (CCC) $\Delta(r)$ defined by

$$\Delta(r) = c(r) - c_{FH}(r) \tag{7}$$

or its analogue in **k** space,

$$\tilde{\Delta}(k) = \tilde{c}(k) - \tilde{c}_{FH}(k). \quad (8)$$

Both $c_{FH}(r)$ and $\tilde{c}_{FH}(k)$ are known in exact analytic form, the latter from Eqs. (5) and (3) with the corresponding **r** space form given in Ref. 4 being collected for convenience in Appendix A. Then, as already mentioned, this formally exact theory can be turned into numerical results, but now in the form of an inequality for the CCC function $\tilde{\Delta}(k)$. Section III then considers analogous results in **r** space, with differential equations of the **r** space CCC function now being involved. Section IV constitutes a summary plus some proposals for further work. In an Appendix, Gaskell's ground-state variational wave function,[5] sometimes used as a valuable starting point for quantal computer simulation of the homogeneous electron liquid, is shown to be related to the CCC function $\tilde{\Delta}(k)$.

II. FORMAL k SPACE THEORY

A number of early workers directly considered **k** space theory aimed at relating the formally exact (but as yet unknown analytically) structure factor $S(k,r_s)$ to the Fermi hole result in Eq. (5), where the mean interelectronic spacing r_s is defined by

$$\rho_0 = \frac{3}{4\pi r_s^3}. \quad (9)$$

A convenient summary is given by Stoddart,[6] with earlier references collected there. In particular, Stoddart defines, in his Eq. (12),[6] a function $B(k)$ through

$$\left[\frac{k^2}{2S(k)}\right]^2 = \left[\frac{k^2}{2S_{FH}(k)}\right]^2 + 4\pi\rho_0\left[1 + \frac{k^2}{2\pi}B(k)\right]. \quad (10)$$

In terms of the CCC function $\tilde{\Delta}(k)$ introduced in Eqs. (7) and (8) above, we obtain the following for the relation between $B(k)$ and $\tilde{\Delta}(k)$ from Eqs. (10), (3), and (8):

$$\tilde{\Delta}(k)^2 + 2[\tilde{c}_{FH}(k) - 1]\tilde{\Delta}(k) = \frac{16\pi\rho_0}{k^4}\left[1 + \frac{k^2}{2\pi}B(k)\right]. \quad (11)$$

In the following section, we shall briefly summarize an approximate form for the Fourier transform $\mathcal{B}(r)$ of the function $B(k)$ entering Eq. (11). Here, however, we shall turn next to the pioneering work of Kugler,[7] who gave an inequality for the structure factor $S(k)$. Below, we discuss the implications of Kugler's work for the OZ function $\tilde{c}(k)$ and, hence, for the central CCC function $\tilde{\Delta}(k)$ of the present study.

A. Inequality involving the OZ function

Defining, following Kugler,[7] a frequency $\omega_0(k)$ by $\hbar k^2/m$ and the plasma frequency ω_p by the customary relation $\omega_p^2 = 4\pi e^2\rho_0/m$, and using Eq. (3), we can rewrite Kugler's original inequality for $S(k)$ (Ref. 7) in terms of the OZ function $\tilde{c}(k)$ as

$$\frac{k^4}{k_f^4}[1 - \tilde{c}(k)]^2 \leq \omega_p^2 + \frac{4\omega_0(k)}{\hbar}\langle KE\rangle + \omega_0^2(k) + I(k). \quad (12)$$

To complete the definitions needed in this inequality, we note that $\langle KE\rangle$ is the kinetic energy per electron. This quantity is plotted, for example, in Fig. 1 of the paper of Herman and March[8] over a wide range of r_s, making use of the virial theorem in the form

$$\langle KE\rangle + E = -r_s\frac{dE}{dr_s}. \quad (13)$$

For an accurate numerical work, $E(r_s)$ obtained in the quantal computer simulations of Ceperley and Alder[9] is well fitted by Perdew and Zunger[10] for $r_s < 20$. Finally, the definition of the remaining quantity $I(k)$, introduced originally by Kugler, in Eq. (12) is

$$I(k) = \frac{e^2}{m\pi}\int_0^\infty dq q^2[S(q) - 1]J(q,k), \quad (14)$$

with

$$J(k,q) = \frac{5}{6} - \frac{q^2}{k^2} + \frac{k}{8q}\left(\frac{q^2}{k^2} - 1\right)^2 \ln\left(\frac{q+k}{q-k}\right)^2, \quad (15)$$

which is a function only of q/k, positive for all values of its argument and monotonically decreasing with increasing q/k. We note for completeness that Eq. (12) involves $\tilde{c}(k)$ on the left-hand side and $S(k)$ via $I(k)$ on the right-hand side.

To obtain $I(k)$ and $\tilde{c}(k)$, we have used the model analytic results of Gori-Giorgi *et al.*[11] These workers have modeled analytically the electron static structure factor $S(k)$ and the corresponding pair correlation function $g(r)$. In addition to imposing known analytic constraints, the model presented in Ref. 11 accurately interpolates the extensive diffusion Monte Carlo data of Ortiz *et al.*[12] Therefore, we have utilized their model of $S(k)$ as a function of mean interelectronic spacing r_s to gain a quantitative understanding of the direct correlation $c(r)$, which is the main focus of the present study. In Fig. 2, we show $I(k,r_s)/k_f^3$ for $r_s=2$ and 5.

Although in Appendix B we give a numerical example of the use of Eq. (12) for a high density value $r_s=0.1$, for which $\langle KE\rangle$ can be obtained analytically from Eq. (13), we want to emphasize here that while, in form (12), the right-hand side gives a useful estimate of $k^4[1-\tilde{c}(k)]^2$ over the whole range of k, the example in Appendix B demonstrates conclusively that if we subtract k^4/k_f^4 from both sides of Eq. (12), the resulting inequality is only useful at small k and is therefore quite limited in practical applications. In particular, inequality (12) is not useful in the interesting limit of large k.

In contrast, we can return to the result for $B(k)$ introduced in Eq. (10) in the following section in which we consider **r** space theory.

III. r SPACE THEORY OF OZ FUNCTION

To link **r** space theory with the **k** space presentation of Sec. II, let us begin with the Vashista and Singwi relation[13]

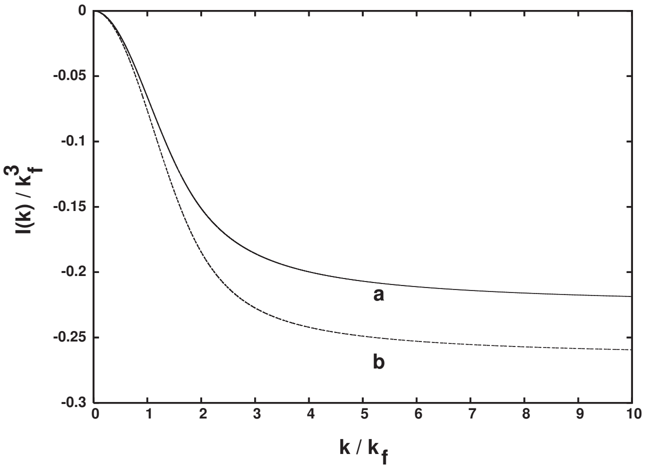

FIG. 2. Function $I(k,r_s)$ defined in Eq. (14), which enters inequality (12) for the OZ function $\tilde{c}(k)$, for (a) $r_s=2$ and (b) $r_s=5$. All quantities are in a.u.

of the Fourier transform $\mathcal{B}(r)$ of $B(k)$ introduced in Eqs. (10) and (11). This is written by Stoddart[6] in the form

$$\nabla\mathcal{B}(r) = [\nabla\phi(r)]\left[1 + a\rho_0\frac{\partial}{\partial\rho_0}\right][g(r,\rho_0) - 1], \qquad (16)$$

where a is an adjustable parameter in the Vashista-Singwi treatment. In Eq. (16), $\phi(r)$ is the Coulombic repulsion in the form $4\pi/r$, while $g(r)$ is the pair function in which the density dependence ρ_0 is displayed.

To extract $B(k)$, the Fourier transform of $\mathcal{B}(r)$, from Eq. (16), let us form $\nabla^2\mathcal{B}$ as

$$\nabla^2\mathcal{B} = \left[h + a\rho_0\frac{\partial h}{\partial\rho_0}\right]\nabla^2\left(\frac{4\pi}{r}\right) + \left[1 + a\rho_0\frac{\partial}{\partial\rho_0}\right]\nabla h \cdot \nabla\left(\frac{4\pi}{r}\right), \qquad (17)$$

where $h=g-1$ is the total correlation function introduced above. Then, in Fourier transform, we find

$$k^2B(k) = \frac{16\pi^2\rho_0}{k_f^3}\left[1 + a\rho_0\frac{\partial}{\partial\rho_0}\right]h(0,\rho_0) + \frac{16\pi^2\rho_0}{k_f^3} \times\left[1 + a\rho_0\frac{\partial}{\partial\rho_0}\right]\int_0^\infty dr\frac{\sin kr}{kr}\frac{\partial h}{\partial r}, \qquad (18)$$

which is convenient to rewrite in the form

$$k^2B(k) = \frac{16\pi^2\rho_0}{k_f^3}\left[1 + a\rho_0\frac{\partial}{\partial\rho_0}\right]\int_0^\infty dr\left[\frac{\sin kr}{kr} - 1\right]\frac{\partial h}{\partial r}. \qquad (19)$$

In Fig. 3, we compare the formally exact $k^2B(k)$ function obtained from Eq. (9) and calculated by means of the fitted $S(k)$ of Gori-Giorgi *et al.*[11] with the function coming from Eq. (19).

By way of example, we show plots for $r_s=2$ and 5 and with a in the range ±1. Though the agreement is, at best, semiquantitative, we wish to stress here that the merit of the Vashista and Singwi approach is clear by analogy with a classical statistical mechanical theory of liquid argon. There, the density dependence of $g(r)$ [or, equivalently, $h(r)$] is connected with integrals over the three-body correlation function. Clearly, obtaining $B(k)$ in Eq. (9) from the known quantum mechanical density matrix hierarchy (see, for example, Dawson and March[14]) must involve higher order density matrices and, in particular, the third order form.

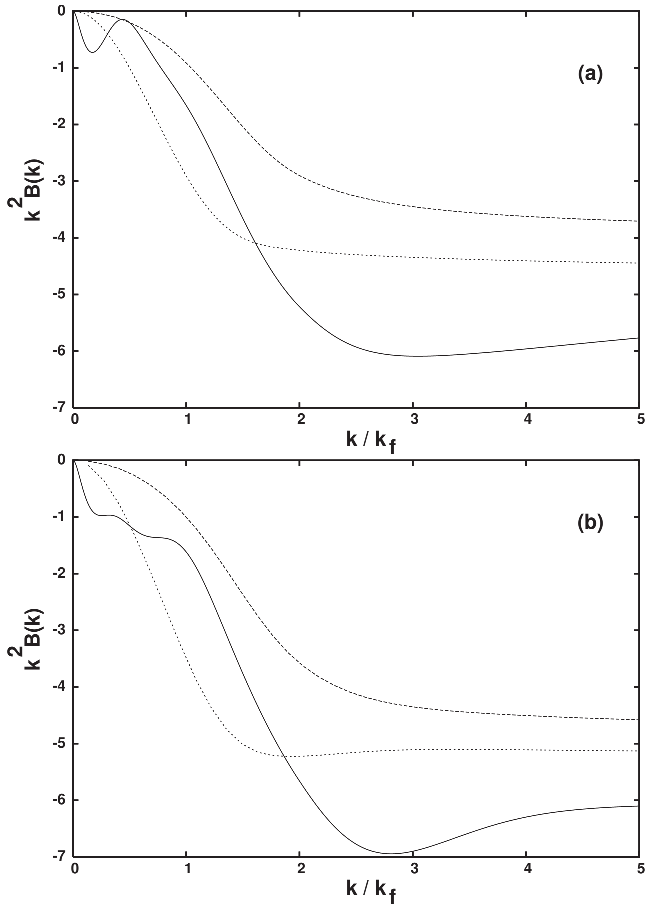

FIG. 3. Shape of function $k^2B(k)$ in the formally exact **k** space theory set out in Eq. (10) (full line), (a) for $r_s=2$ and (b) $r_s=5$. In the same plots, the exact function is compared with the Fourier transform of $\nabla^2\mathcal{B}(r)$, obtained from the Vashista and Singwi (Ref. 13) approach embodied in Eq. (19) (dashed lines) with $a\pm1$. The density dependence of $g(r,\rho_0)$ involved in that equation is calculated from the fit given by Gori-Giorgi *et al.* (Ref. 11). All quantities are in a.u.

A. Dawson-March differential equation

Dawson and March[15] used the explicit form of the Fermi hole to write

$$g_{FH}(r) = 1 - \frac{9}{2}\left[\frac{j_1(k_f r)}{k_f r}\right]^2, \qquad (20)$$

where $j_1(x)$ denotes the first order spherical Bessel function defined by $j_1(x)=[\sin x - x\cos x]/x^2$. These authors then demonstrate that Eq. (20) can be rewritten as

$$g_{FH}(r) = 1 - \frac{3\hbar^2}{m\rho_0 r^2}\sigma_{10}(r,E_F), \tag{21}$$

where $\sigma_{10}=(\partial n_{10}/\partial E)_{E_F}$ is the free electron local density of states of the p-component $(l=1)$ at the Fermi energy E_F.

The earlier work of March and Murray[16] allows a differential equation to be written, which determines $\sigma_1(r,E)$, the generalization of the free electron quantity σ_{10} entering Eq. (21), in the presence of a central potential energy $V(r)$. Their equation reads

$$\frac{1}{8}\frac{\partial^3}{\partial r^3}(r^2\sigma_1) + \left(E - V - \frac{1}{r}\right)\frac{\partial}{\partial r}(r^2\sigma_1) - \frac{1}{2}\left(\frac{\partial V}{\partial r}\right)r^2\sigma_1 = 0. \tag{22}$$

Schinner[17] has subsequently reported a numerical solution of the above Dawson-March model, using, as the authors proposed, a self-consistent field determination of $V(r)$. The pair function $g(r)$ remains positive even for the low density corresponding to $r_s=10$. However, $g(0,r_s)$ is not good, being too large when compared, for example, with the useful Overhauser form,[18]

$$g(0,r_s) = \frac{32}{(8+3r_s)^2}. \tag{23}$$

Employing again the result of Gori-Giorgi *et al.*,[11] one can use Eq. (22) as a route to determine $V(r)$ via a first order differential equation. However, we turn instead to a formally exact **r** space theory given recently by Nagy and Amovilli,[19] as this, though nonlinear compared with the Dawson and March[15] linear theory of $g(r)$, is again in the form of an **r** space theory, but now for the "pair function amplitude" $\sqrt{g(r)}$, given a one-body potential subsuming electronic Coulombic correlations. Their differential equation is

$$\frac{\partial^2}{\partial r^2}(r\sqrt{g}) + [\epsilon - v_{eff}(r)]r\sqrt{g} = 0. \tag{24}$$

Work actually in progress[20] on the potential v_{eff} in this equation will be published elsewhere. Here, we limit ourselves to show in Fig. 4 the plot of the function

$$\phi(r) = \frac{1}{r\sqrt{g}}\frac{\partial^2}{\partial r^2}(r\sqrt{g}), \tag{25}$$

which gives the numerical form of $v_{eff}(r)$, for $r_s=2$ and 5, to within an additive constant. A referee has requested a brief discussion of the physical meaning of $v_{eff}(r)$ in Eq. (24). A hierarchy of density matrix equations, which is the quantal generalization of the Bogoliubov-Born-Green-Kirkword-Yvon hierarchy of classical statistical mechanics of fluids, shows that to exactly evaluate the pair function $g(r)$, which is the diagonal of the two-particle density matrix in the uniform electron fluid under discussion, one must know the three-particle density matrix. Thus, any exact theory of $v_{eff}(r)$ seems presently beyond the scope of available methods (we can liken the situation to that of using, for example, the approach of Vignale and Singwi[21]). Of course, we can only make quantitative progress on $v_{eff}(r)$ here by utilizing the modeling of $g(r)$ by Gori-Giorgi *et al.*[11]. In the light of the above status of $v_{eff}(r)$, it remains of interest to establish a relation, albeit approximate, between $v_{eff}(r)$ and the OZ function $c(r)$. The motivation here is that, again in classical statistical mechanics, the approximation $-k_BTc(r)$ is a useful effective potential, with k_BT now being the thermal energy.

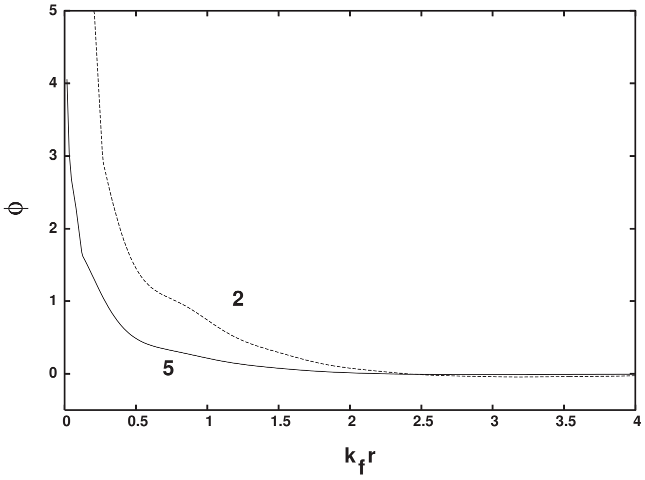

FIG. 4. Plot of function ϕ defined in Eq. (25) against $k_f r$ for $r_s=2$ (curve 2) and 5 (curve 5). ϕ is the effective potential, to within an additive constant, which gives the pair function amplitude in the Nagy and Amovilli approach (Ref. 19). All quantities are in a.u.

B. Form of OZ function $c(r,r_s)$

To conclude this section on **r** space theory, it is natural to calculate this function $c(r,r_s)$ from the fits of Gori-Giorgi *et al.*[11] for $S(k)=1/[1-\tilde{c}(k)]$. In this way, we have constructed $c(r,r_s)$ for $r_s=0.1$, 2, and 5 and have added the Fermi hole form [see Fig. 1(a)] for comparison in Fig. 5(a). In Fig. 5(b), we have plotted $(k_f r)^2 c(r)$, which diverges like the negative of $k_f r$ for large r, since $c(r) \propto e^2/r$ in this limit.

We have also studied the r_s dependence of $c(0,r_s)$. A simple fit of our numerical results in the range $0 \leqslant r_s \leqslant 20$ a.u. gives the result

$$c(0,r_s) \approx \frac{c_{FH}(0) - 1.45 r_s^{1/2} - 2.71 r_s}{1 + 1.83 r_s^{1/2}}, \tag{26}$$

which complements the Overhauser form [Eq. (23)] of $g(0,r_s)$.

Returning to the $c(r,r_s)$ plot in Fig. 5(a), it is known that at large $k_f r$,

$$c(r,r_s) \propto \frac{e^2}{r\hbar\omega_p} \propto \frac{r_s^{3/2}}{r} \propto \frac{r_s^{1/2}}{k_f r}, \tag{27}$$

with $k_f r$ being the independent variable in that plot.

IV. SUMMARY AND FUTURE DIRECTIONS

We have focused here on the CCC to the OZ Fermi hole function $c_{FH}(r)$ in **r** space or in momentum space $\tilde{c}_{FH}(k)$. In

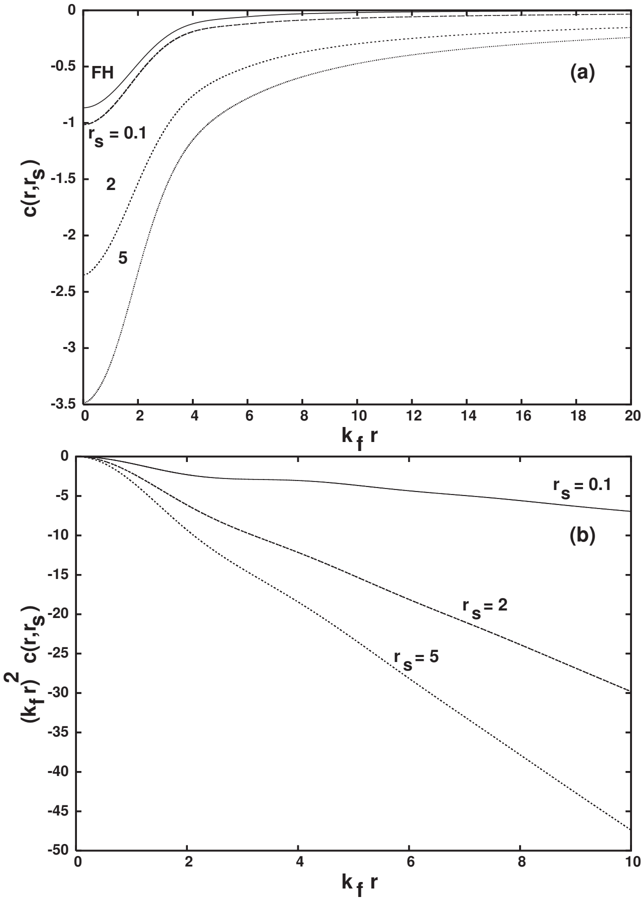

FIG. 5. OZ direct correlation function $c(r,r_s)$ (a) plotted as a function of $k_f r$ for r_s=0.1, 2, and 5. The Fermi hole form is added for comparison (uppermost curve). In (b), the OZ direct correlation function is multiplied by $(k_f r)^2$ for three different densities corresponding to r_s=0.1, 2, and 5. Note that $2 \leqslant r_s \leqslant 5$ is the range of the simpler metal densities (e.g., Al and Cs). All quantities are in a.u. One sees the simplicity of the OZ function and especially how quickly it approaches its large r form determined by Coulombic interaction e^2/r.

k space, we have employed the analytic fits of Gori-Giorgi *et al.*[11] for $S(k)$ to make use of Kugler's inequality, but now written to reflect the OZ function. The CCC function $\Delta(r)$ is proportional to e^2/r at large r and, thus, directly reflects the Coulombic interelectronic repulsion.

While the **k** space theory, at present, appears to be the more direct approach, it does not ensure that the pair correlation function $g(r) > 0$: an obvious physical requirement. The current applications of the Dawson and March differential equation [Eq. (22)] give $g(0) > 0$, but too large to agree with the Overhauser fit.[18] Any definitive **r** space theory should embody Overhauser's proposal at least approximately.

Finally, some attention has been paid to the formally exact Nagy and Amovilli nonlinear equation for $g(r)$,[19] and the one-body potential $v_{eff}(r)$ subsuming correlations which it contains has been extracted for sample values of r_s in Fig. 4 to within an additive constant.

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APPENDIX A: FERMI HOLE FORM OF OZ DIRECT CORRELATION FUNCTION

In a very recent work, Nagy *et al.*[4] considered a homogeneous Fermi liquid with a model repulsive inverse square law interparticle potential energy. In the course of their study, the Fermi hole form of the direct correlation function $c_{FH}(r)$ was obtained analytically. Since $c(r)$ is the heart of this article, we first summarize the closed form of $c_{FH}(r)$ below.

In terms of the Fermi wave number k_f, the result for the OZ function representing exactly the Fermi hole is

$$c_{FH}(r) = \frac{3}{2k_f r}\left[\frac{\sin(2k_f r) - 2k_f r\cos(2k_f r)}{(k_f r)^2} - \frac{4}{3k_f r}\mathcal{I}_{FH}(r,k_f)\right]. \tag{A1}$$

The function $\mathcal{I}_{FH}(r,k_f)$ in the work of Nagy *et al.* is given in terms of two contributions. However, here we have calculated the function $\mathcal{I}_{FH}(r,k_f)$ directly from the numerical results for $c_{FH}(r)$.

It is also worth noting here that Nagy *et al.*[4] proposed an approximate form of $c_{FH}(r)$ in which the term $\mathcal{I}_{FH}(r,k_f)$ in Eq. (A1) is replaced by

$$\mathcal{I}_{FH}(r,k_f) \approx 1 - \cos(2k_f r), \tag{A2}$$

which is in good agreement with the large r behavior of $\mathcal{I}_{FH}(r,k_f)$.

APPENDIX B: ILLUSTRATION OF INEQUALITY (12) INVOLVING OZ FUNCTION $\tilde{c}(k)$ FOR A HIGH DENSITY HOMOGENEOUS ELECTRON LIQUID

As mentioned in the main text, let us remove the terms of $O(k^4)$ in inequality (12) by subtracting k^4/k_f^4 from both sides. Then, we find the following in a.u.:

$$\left(\frac{k^4}{k_f^4}\right)[\tilde{c}(k)^2 - 2\tilde{c}(k)] \leqslant \frac{1}{k_f^4}\left[\frac{3}{r_s^3} + 4k^2\langle KE\rangle + I(k,r_s)\right], \tag{B1}$$

where the first term on the right-hand side of Eq. (B1) denotes the square of the plasma frequency ω_p in a.u.

We take a high density example r_s=0.1 to illustrate form (B1). Then, the kinetic energy $\langle KE\rangle$ reads as follows, using the analytic Hartree-Fock energy plus the Gellmann-Bruckner correlation energy to insert the $E(r_s)$ in the virial theorem [Eq. (13)]:

$$\langle KE\rangle = \frac{1.105}{r_s^2} + \frac{0.916}{r_s} - A\ln r_s - B + A, \tag{B2}$$

where $A=(1-\ln 2)/\pi^2$ and $B=-0.0469$.

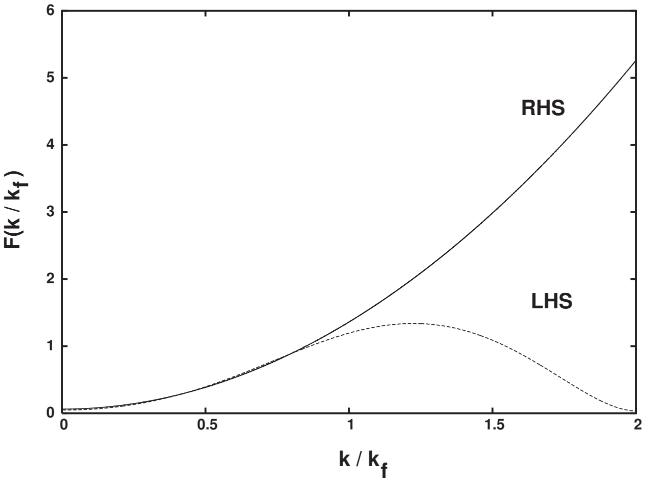

FIG. 6. Kugler's inequality from Eq. (B1). All quantities are in a.u.

Figure 6 shows both sides of inequality (B1), and it is quite clear that this is only useful at long wavelengths, though, of course, the Kugler inequality holds for all k values.

APPENDIX C: GASKELL'S VARIATIONAL WAVE FUNCTION FOR THE HOMOGENEOUS ELECTRON LIQUID AND ITS RELATION TO THE OZ FUNCTION

Gaskell wrote a ground-state wave function Ψ for a homogeneous electron liquid in terms of density fluctuations $\rho_{\mathbf{k}}$ (Ref. 5) defined by

$$\rho_{\mathbf{k}} = \sum_{i=1}^{N} \exp(-i\mathbf{k}\cdot\mathbf{r}_i), \tag{C1}$$

where the vectors $\mathbf{r}_i$ denote electronic positions. With D denoting a Slater determinant of plane waves, Gaskell's wave function has the form

$$\Psi = D \exp\left[-\sum_{\mathbf{k}} d(\mathbf{k})\rho_{\mathbf{k}}\rho_{\mathbf{k}}^{*}\right]. \tag{C2}$$

Gaskell then determined the function $d(\mathbf{k})$ entering Eq. (C2) variationally.

In the high density limit corresponding to small interelectronic separation r_s, and for a long wavelength, Gaskell then obtained $d(\mathbf{k})$ in the form

$$d(\mathbf{k}) \approx \left[\frac{4}{3\pi}\left(\frac{4}{9\pi}\right)^{1/3}\right]^{1/2} \frac{k_f^2 r_s^{1/2}}{2k^2}. \tag{C3}$$

However, as indicated in the main text, the OZ function $\tilde{c}(k)$ in the same long wavelength limit behaves as

$$\tilde{c}(k) \approx \frac{4\pi e^2}{k^2 \frac{1}{2}\hbar\omega_p}, \tag{C4}$$

where $(1/2)\hbar\omega_p$ denotes the zero-point energy of the plasmon. By comparison of Eqs. (C3) and (C4), one recognizes—using the result that the product $k_f r_s$ is a pure number—that in this long wavelength limit (at least) $d(\mathbf{k})$ is proportional to the OZ function $\tilde{c}(k)$.

Returning briefly to the Fermi hole pair function and writing $\eta = k/k_f$ and $x = k_f r$, we have

$$g_{FH}(x) - 1 = -\frac{3}{2}\int_0^\infty \eta^2 j_0(\eta x)[1 - S_{FH}(\eta)]d\eta, \tag{C5}$$

where $j_0(x) = \sin x/x$. Using Eq. (3) again in the Fermi hole limit, corresponding to $d(\mathbf{k}) = 0$ in the Gaskell wave function [Eq. (C2)], we immediately find an alternative expression for Eq. (C5) in terms of the OZ function as

$$g_{FH}(x) - 1 = -\frac{3}{2}\int_0^\infty \eta^2 j_0(\eta x)\left[1 - \frac{1}{1 - \tilde{c}_{FH}(\eta)}\right]d\eta. \tag{C6}$$

Introducing the Coulombic correlations reflected through $d(\mathbf{k})$, one has an approximate pair function of the form

$$g(x) - 1 = -\frac{3}{2}\int_0^\infty \eta^2 j_0(\eta x)\left[1 - \frac{1}{1 - \tilde{c}_{FH}(\eta) + 4d(\eta)}\right]d\eta, \tag{C7}$$

where $d(\eta)$ depends, of course, on the density ρ_0 or, equivalently, r_s.

In short, the main conclusion of this appendix is that $d(k)$ in the Gaskell wave function is intimately related to the OZ function. It would be interesting if quantum Monte Carlo calculations starting from the wave function [Eq. (C2)] could derive (of course, approximately) the pair function $g(r)$ and, hence, relate $d(k)$ directly to $\tilde{c}(k)$ beyond the long wavelength limit discussed above.

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