A simple classical mapping of the spin-polarized quantum electron gas: distribution functions and local-field corrections.

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We use the now well known spin unpolarized exchange-correlation energy \( E_{xc} \) of the uniform electron gas as the basic “many-body” input to determine the temperature \( T_{q} \) of a classical Coulomb fluid having the same correlation energy as the quantum system. It is shown that the spin-polarized pair distribution functions (SPDFs) of the classical fluid at \( T_{q} \), obtained using the hyper-netted chain (HNC) equation are in excellent agreement with those of the \( T = 0 \) quantum fluid obtained by quantum Monte Carlo (QMC) simulations. These methods are computationally simple and easily applied to problems which are currently outside the scope of QMC. Results are presented for the SPDFs and the local-field corrections to the response functions of the electron fluid at zero and finite temperatures.

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The uniform interacting electron gas (UEG) is the “text-book” many-body problem of Fermi liquids in metals, plasmas or doped semiconductors. It provides a model exchange-correlation potential for the density-functional theory of inhomogeneous electronic systems. The random-phase-approximation (RPA) to the properties of the UEG provides an important “baseline” which could be reached by many techniques like quantum-linked cluster expansions, Green’s functions, or linearized equations of motion. RPA is a reasonable approximation when the “coupling parameter” \( \Gamma = (\text{potential energy})/(\text{kinetic energy}) \) of the Coulomb fluid is smaller than unity. The coupling parameter for the UEG at \( T=0 \) becomes identical with the mean sphere radius \( r_s \) per electron, i.e., for the 3-D case, \( \Gamma = r_s = (3/4\pi\pi)^{1/3} \). Here \( \pi \) is the electron number-density per atomic unit of volume.

Improving the RPA becomes nontrivial if the results of the theory are expected to satisfy sum rules and provide physically realistic pair-distribution functions (PDFs), i.e., \( g(r) \), of the quantum system. Diagrammatic methods look for resummations that conserve the sum rules, Ward identities etc. This line of research is followed in the work of Hubbard, Vosko, Langreth, Geldart et al. Calculations have been given by MacDonald et al. for the self-energy, while Green et al., Ichimaru et al. and Richardson et al. and many others have studied the response functions.

The equation of motion (EQM) method, when pushed beyond the RPA requires some “ansatz” to decouple or “close” the equations. Singwi et al. (SSTL) used a physically motivated classical analogy to close the EQM by introducing the electron-electron PDF determined self-consistently. There are also attempts to directly fit the response functions or the PDF’s to a form constrained to satisfy the sum rules and such requirements. In spite of a large effort, the calculation of the electron-gas \( g(r) \) by STLS and other methods at arbitrary spin-polarization \( (\zeta) \) and \( T \) is difficult. In fact, STLS gives negative PDFs for sufficiently large \( r_s \) in the metallic range and does not satisfy the compressibility sum rule. Even the attempts to directly fit the PDFs do not guarantee positive-definite a \( g(r) \) for important regimes of \( r_s \) and \( T \).

Another approach beyond RPA is to use a correlated wavefunction \( \psi = FD \) where \( F \) is a correlation factor and \( D \) is a Slater determinant. The best \( F \) is determined variationally. This approach leads to the Jackson-Feenberg energy functional which can be examined in several ways. The QMC techniques also use such a \( \psi \) and lead, e.g., to the variational Monte Carlo (VMC) method. Thus Ceperley and Alder provided \( E_{xc}(r_s) \) now available in several parametrized forms. The correlation energy given by various QMC methods agree with one another to about the same extent as with, say, the results from the Fermi-Hyper-Netted-Chain(FHNC) method. Thus, while it is easy to get good correlation energies, it is quite difficult to get other properties like the PDFs and local-field corrections (LFC) at arbitrary \( \zeta \), \( T \) and in the metallic range of \( r_s \).

In this letter we present a computationally simple method for calculating the PDFs and other derived quantities (e.g., response functions) of the UEG, given the unpolarized \( E_{xc} \) from whatever source. Since PDFs of classical fluids are easily obtainable via the Hyper-Netted-Chain (HNC) procedure, we model the electron gas as a classical fluid containing two species (up, down spins). We ask for the temperature \( T_q \) at which the excess free energy of a Coulomb fluid, obeying the classical HNC-integral equation, becomes equal to the correlation energy \( E_{xc} \) of the UEG at the same density and at \( T=0 \). The suffix \( q \) in \( T_q \) signifies that this temperature parametrizes the quantum many-body interactions in the UEG. Using the temperature \( T_q \) at each \( r_s \) we obtain \( g_{ij}(r) \), spin-
polarized correlation energies $E_c(r_s, \zeta)$, local-field corrections etc. While many interesting applications are possible, here we treat the 3-D spin-polarized UEG at zero and finite $T$.

The physical motivation for our approach comes from density functional theory (DFT) where the interacting electron gas is replaced by a noninteracting gas of Kohn-Sham (KS) particles whose wavefunction is a simple determinant. Here the philosophy is very different to the Feenberg approach which uses a correlated determinant for fermions. In DFT the many-body potential is replaced by a single-particle KS potential. Since the natural energy parameter of the classical ensemble is the temperature, we look for a temperature representation of the interactions.

Consider a fluid of mean density $\overline{\pi}$ containing two spin species with concentrations $x_i = \overline{\pi}_i/\overline{\pi}$. In the following we deal with the physical temperature $T$ of the UEG, while the temperature of the classical fluid $T_{cf}$ is $1/\beta$. Since the leading dependence of the energy on temperature is quadratic, we assume that $T_{cf} = \sqrt{(T^2 + T_0^2)}$. This is clearly valid for $T = 0$ and for very high $T$. This assumption will not be probed more deeply in this letter where the main effort is for $T = 0$. Our objective is to determine $T_q$, the temperature equivalent of the quantum correlations in the UEG at $T = 0$.

The pair-distribution functions for a classical fluid at an inverse temperature $\beta$ can be written as

$$g_{ij}(r) = \exp[\beta\phi_{ij}(r) - h_{ij}(r) - c_{ij}(r) + B_{ij}(r)] \quad (1)$$

Here $\phi_{ij}(r)$ is the pair-interaction potential between species $i,j$. For two electrons this is just the Coulomb potential $V_{cu}(r)$ if there is no exchange interaction. If the spins are parallel, the Pauli principle prevents them from occupying the same spatial orbital. Following the earlier work, notably by Lado,[4] we introduce a “Pauli potential”, $\mathcal{P}(r)$. Thus:

$$\phi_{ij}(r) = \mathcal{P}(r)\delta_{ij} + V_{cu}(r) \quad (2)$$

The Pauli potential $\mathcal{P}(r)$ will be discussed with the PDFs of the non-interacting UGE, i.e., $g^0_{ij}(r)$. The function $h(r) = g(r) - 1$: it is related to the structure factor $S(k)$ by a Fourier transform. The $c(r)$ is the “direct correlation function (DCR)” of the Ornstein-Zernike (OZ) equations.

$$h_{ij}(r) = c_{ij}(r) + \Sigma_s \pi_s \int dr' h_{i,s}(r-r')c_{s,j}(r) \quad (3)$$

The $B_{ij}(r)$ term in Eq. (3) is the “bridge” term arising from certain cluster interactions. If this is neglected Eqs. (1), (2) form a closed set providing the HNC approximation to the PDF of a classical fluid. Various studies have clarified the role of $B(r)$ and its treatment via “reference” HNC equations. $B(r)$ is important when the coupling constant $\Gamma$ exceeds, say, 20. The range of $\Gamma$ relevant to this work (e.g., $\Gamma \sim 4.5$ even for $r_s = 10$ ) is such that the HNC-approximation holds. The HNC-approximation suffers from a compressibility inconsistency (CI), i.e., the excess compressibility calculated from the small-$k$ limit of the short-ranged part of $c(k)$ does not agree with that obtained from the excess free energy. This CI can be corrected by choosing $B_{ij}(r)$ suitably.

Consider the non-interacting system at temperature $T$. The parallel-spin PDF, i.e., $g^0_{ii}(r,T)$, will be denoted by $g^0_T(r)$ for simplicity, since $g^0_{ij}(r,T)$, $i \neq j$ is unity. Consider the paramagnetic case $x_i = 0.5$. Denoting $(r_1 - r_2)$ by $r$, it is easy to show that

$$g^0_T(r) = \frac{2}{N^2} \sum_{k_1,k_2} n(k_1)n(k_2)[1 - e^{i(k_1-k_2)r}] \quad (4)$$

Here $n(k)$ is the Fermi occupation number at the temperature $T$. Eq. (3) reduces to:

$$g^0_T(r) = 1 - F^2_T(r) \quad (5)$$

$$F_T(r) = \int \frac{\sin(kr)}{kr} dk = \frac{\sin(kr)}{kr} \frac{2\pi^2}{k_F^2} \quad (6)$$

The Fermi momentum is denoted by $k_F$. Thus $g^0_T(r)$ can be obtained from the Fourier transform of the Fermi function. Then $c^0(r)$ can be evaluated from $g^0_T(r)$ using the OZ relations. The $T = 0$ case can be evaluated analytically[2]

Assuming that $g^0_{ii}(r)$ can be modeled by an HNC fluid with the pair interaction $\beta\mathcal{P}(r)$, dropping the indices, we have:

$$g^0(r) = \exp[-\beta\mathcal{P}(r) + h^0(r) - c^0(r)] \quad (7)$$

The k-space direct correlation function $c^0(k)$ at $T = 0$ decays as $4k_F/3k$ for small $k$, showing that the r-space form $c^0(r)$ is long ranged. The “Pauli potential” $\mathcal{P}(r)$ is given by

$$\beta\mathcal{P}(r) = -\log[g^0(r)] + h^0(r) - c^0(r) \quad (8)$$

We can determine only the product $\beta\mathcal{P}(r)$. The classical fluid “temperature” $1/\beta$ is still undefined and is not the thermodynamic temperature $T$. The Pauli potential is a universal function of $r_{kF}$ at $T = 0$. Plots of $\beta\mathcal{P}(r)$ and related functions are given in Fig. 1. It is long ranged and mimics the exclusion effects of Fermi statistics. As $T$ is increased, the Pauli potential restricts to about a thermal wavelength and becomes more hard-sphere like.

The next step is to use the full pair-potential $\phi_{ij}(r)$, Eq. (2), and solve the coupled HNC and OZ equations for the binary (up and down spins) interacting fluid. For the paramagnetic case, $\overline{\pi}_i = \overline{\pi}/2$, we have:

$$g_{ij}(r) = e^{-\beta\mathcal{P}(r)h_{ij}(r)+V_{cu}(r)} \quad (9)$$

$$h_{ij}(q) = \frac{E_T}{\beta} h_{ij}(r) \quad (10)$$

$$h_{11}(q) = c_{11}(q) + (\overline{\pi}/2)c_{11}(q)c_{11}(q) + c_{12}(q)h_{21}(q) \quad (11)$$

$$h_{12}(q) = c_{12}(q) + (\overline{\pi}/2)c_{11}(q)c_{12}(q) + c_{12}(q)h_{22}(q) \quad (12)$$

The Coulomb potential $V_{cu}(r)$ needs some discussion. For two point-charge electrons this is $1/r$. However, depending on the temperature $T$, an electron is localized.
to within a thermal wavelength. Thus, following Morita, and Minoo et al.\[22\] we use a “diffraction corrected” form:

\[ V_{cov}(r) = (1/r)[1 - e^{-r/\lambda_{th}}]; \quad \lambda_{th} = (2\pi m T_{c_f}). \]  \hspace{1cm} (13)

Here \( m \) is the reduced mass of the electron pair, i.e., \( m^*(r_s)/2 \text{ a.u.} \), where \( m^*(r_s) \) the effective electron mass. It is weakly \( r_s \) dependent, e.g., \( \sim 0.96 \) for \( r_s = 1 \). In this work we take \( m^*=1 \). The “diffraction correction” ensures the correct behaviour of \( g_{12}(r \sim 0) \) for all \( r_s \).

In solving the above equations for a given \( r_s \), and at \( T=0 \), we have \( T_{c_f}=T_q \). A trial \( T_q \) is adjust to obtain an \( E_c(T_q) \) equal to the known paramagnetic \( E_c(r_s) \) at each \( r_s \).

\[ E_c(T_q) = \int_0^1 d\lambda \int r^2[(h_{11}(r, \lambda) + h_{12}(r, \lambda))]\tau^2 dr \] \hspace{1cm} (14)

The resulting “quantum” temperatures \( T_q \) could be fitted to the form:

\[ T_q/E_F = 1.0/(a + b\sqrt{r_s} + cr_s) \] \hspace{1cm} (15)

The QMC results for \( E_c \) obtained from different QM methods differ, e.g., by \( \sim 6\% \) at \( r_s=1 \). We used the most-recent Ortiz-Ballone \( E_c \) data for the paramagnetic UEG from VMC and DMC\[22\]. The small difference in \( E_c \) in these two simulations lead to slightly different fits. The fit coefficients are, for DMC, \( a=1.594, b=-0.3160 \) and \( c=0.0240 \), while for VMC \( a=1.3251, b=-0.1779 \) and \( c=0.08 \). Eight values of \( r_s \), viz., \( r_s = 1 – 6, 8, 10 \), were used in the fit to \( T_q \). At \( r_s = 1 \) and 10, \( T_q/E_F \) goes from 0.768 to 1.198. As \( r_s \) tends to 0, \( g(r) \) tends to \( g^0(r) \). The UEG as \( r_s \) goes to 0 goes to a high-density fluid interacting via the Pauli potential. It is beyond the HNC-approximation.

For any given \( r_s \), knowing the \( T_q \) from the spin-unpolarized case, we can obtain \( g_{12}(r) \) and \( E_{xc}(r_s, \zeta, T) \) at arbitrary, spin-polarization \( \zeta \) by solving the coupled HNC equations. Unlike many of the standard theories of electron fluids, the PDF's obtained from the HNC-procedure are guaranteed to be positive at all \( r_s \). In Fig. 2 we show typical results for \( g_{12}(r) \) and compare them with those of QM-simulations. Our results are in excellent agreement with the DMC results. The reported \( g_{12}(r) \) do not go to zero for \( r=0 \). The depletion hole for \( g_{12}(r = 0) \) from DMC are in agreement with ours and are deeper than those from VMC\[22\]. The difference in VMC and DMC, even at \( r_s = 1 \), Fig. 2(a), is a warning that the even when \( E_c \) are in close agreement, other properties may have significant errors. In Fig. 2(c) we show the paramagnetic distribution functions at \( r_s = 1 \) and 10 together with those of the DMC simulations.

At present there is no reliable finite-temperature microscopic theory to compare with the \( g(r, T) \) obtained by our HNC approach. The theory of Tanaka and Ichimaru\[22\] does better than STLS and is comprehensive. However, like in STLS, the \( g(r) \) becomes negative for some values of \( r \) even at \( r_s = 5 \). Figure 2 (d) give the \( g(r) \) at \( r_s = 5, T = 2T_F \) obtained from HNC. Unlike in SLTS or Tanaka and Ichimaru, the \( g_{ij}(r) \) is positive definite, as expected. Finite-temperature systems will be discussed more fully in a future publication as they are relevant to doped-semiconductors and hot plasmas?\[22\].

The \( T_q \) determined from the unpolarized \( E_c \) is used to calculate \( E_{xc}(r_s, \zeta, T) \) at arbitrary \( \zeta \). The QMC results for \( E_{xc}(r_s, \zeta, T) \) at \( T=0 \) agree with ours, as expected from the agreement of our \( g_{ij}(r) \) with those from MC. For example, at \( r_s = 10 \), the spin-polarized \( E_c \) from Ceperly-Alder is -0.0209 Ry, our HNC procedure gives -0.0211 Ry, Lantto-FHNC gives -0.0186, while Kallio and Pilo report a value of -0.0171 Ry\[22\].

In the theory of the UEG the response \( \chi(k, \omega) \) of the interacting system is written in terms of the noninteracting response function \( \chi^0(k, \omega) \) and a LFC denoted by \( G(k, \omega) \). Considerable effort has been spent in obtaining even the static approximation \( G(k) \) for the LFC. Thus the main thrust of STLS, Ichimaru and Utsumi\[21\], Geldart and Taylor\[22\] and others has been to provide the \( G(k) \) as a function of \( r_s \). In the static case, for a one-component fluid we have:

\[ -V_k G(k) = V_k/\chi(k) - 1/\chi^0(k) \] \hspace{1cm} (16)

where \( V_k = 4\pi/k^2 \). For a classical fluid we also have

\[ S_{ij}(k) = -(1/\beta)\chi_{ij}(k)/(n_i n_j)^{1/2} \] \hspace{1cm} (17)

Hence the LFCs can be expressed in terms of \( S_{ij}(k) \) and compactly in terms of the short-ranged k-space direct correlation functions \( \tilde{c}_{ij}(k) \). For the paramagnetic case:

\[ -V_k G(k) = \{\tilde{c}_{11}(k) + \tilde{c}_{12}(k) - \tilde{c}_{ij}^0(k)\}/2\beta. \] \hspace{1cm} (18)

We display the \( T=0 \) LFC for \( r_s = 5 \) and compare it with the results of QMC and other theories in Fig.3. In HNC, and in standard SLTS, the \( k = 0 \) limit is overestimated due to the IC. The Vashista-Singwi version of SLTS, as well as UI have parametrizations which externally impose the k=0 and \( k \to \infty \) sum rules. The latter sum rule states that \( G(k) \to 1-g(0) \) for large k. The \( G(k) \) from QMC does not seem to follow this and is close to the second-order perturbation calculation of Geldart and Taylor\[22\]. We have ascertained numerically that our HNC generated \( G(k) \) correctly recovers the \( k \to \infty \) sum rule very well.

In conclusion, we have presented a simple classical mapping of a quantum Fermi liquid of arbitrary spin-polarization and temperature \( T \) and shown that it quantitatively recovers the QMC pair-distribution functions. The only parameter is a temperature mapping of the correlation energy. It seems to be sufficient to map the static properties of the UEG. Using the method, we have examined spin-dependent correlation energies, pair-correlation functions at zero and finite T, as well as the local-field correction to the response functions. The method clearly has potential applications to Bose fluids, 2-D electron and other systems.
FIG. 1. The non-interacting PDF, \(g_0^0(r)\), and the Pauli potential, Eq. 8, at \(T/E_F=0\) and 2 (dashed lines). They are universal functions of \(rk_F\), where \(k_F\) is the Fermi wavevector.

FIG. 2. The interacting PDFs \(g_{11}(r)\) and \(g_{12}(r)\) at \(r_s=1\). Solid lines-HNC, boxes-DMC (a), VMC (dashes), and DMC. Panel (b), \(r_s=5\), DMC and HNC (c), (d) are for \(T/E_F=2\). In (c) The paramagnetic \(g(r)\) at \(r_s=1\) and 10 at \(T=0\) compared with the DMC simulations.

FIG. 3. The local-field correction \(G(k)\), Eq. 18, to the static response at \(T=0\) and \(r_s=5\). Results from the HNC, QMC, Geldart and Taylor (GT), Vashista and Singwi (VS), and Utsumi and Ichimaru (UI) are shown.
$r_s = 5$

The diagram shows the relationship between $G(k)$ and $k/k_F$ for various models, including GT, QMC, VS, hnc, and UI. The figure is labeled as 'fig.3'.