Simple Classical Mapping of the Spin-Polarized Quantum Electron Gas: Distribution Functions and Local-Field Corrections

M. W. C. Dharma-wardana* and F. Perrot

National Research Council, Ottawa, Canada K1A 0R6
and Centre d'Etudes de Bruyères le Châtel, P.O. Box 12, 91689 Bruyères le Châtel, France

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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 hypernetted chain 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 beyond QMC simulations. Results are presented for the SPDFs and the local-field corrections to the response functions of the electron fluid at $T = 0$ and finite T.

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The uniform interacting electron gas (UEG) is the "textbook" many-body problem of Fermi liquids in metals, plasmas, or doped semiconductors [1]. It provides a model exchange-correlation potential for density functional theory (DFT) applications [2]. 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" $\Gamma =$ (potential energy)/(kinetic energy) of the Coulomb fluid is smaller than unity. The Γ for the UEG at $T = 0$ is the mean sphere radius r_s per electron; i.e., for the 3D case, $\Gamma = r_s = (3/4\pi\overline{n})^{1/3}$, with $\overline{n}$ the number density per a.u.

Improving the RPA is nontrivial if the theory is 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 approach is followed in the work of Hubbard, Vosko, and Langreth, and Geldart and Taylor (GT) [1,3]. MacDonald *et al.* [4] have considered the self-energy, while others have studied the response functions [5].

The equations of motion beyond the RPA require an "ansatz" to "close" the equations. Singwi *et al.* (STLS) [6] used a physically motivated closure by introducing the electron PDF determined self-consistently. In spite of a large effort, the calculation of the electron gas $g(r)$ at arbitrary spin polarization ζ, r_s, and T is unsolved. In fact, STLS, Ichimaru *et al.* [5], give negative PDFs for sufficiently large r_s in the metallic range. Even the attempts to directly fit the PDFs and response functions to sum rules, etc., seem to fail for important regimes of r_s and T [7]. However, the *static* local-field correction (LFC) has been fitted to quantum Monte Carlo (QMC) data and sum rules by Farid *et al.* (FHER) [8].

Another approach uses a trial wave function $\psi = FD$ where F is a correlation factor and D is a Slater determinant. This typically leads to the Feenberg energy functionals which are handled in several ways [9–11]. QMC techniques also use such a ψ and lead, e.g., to the variational Monte Carlo (VMC) method [12–14]. The QMC estimate of the exchange-correlation energy, $E_{\mathrm{xc}}(r_s)$, is now available in several parametrized forms [15,16]. While it is easy to get good $E_{\mathrm{xc}}(r_s)$, the opposite is true for other properties like the PDFS and LFCs.

In this Letter we present a computationally simple, conceptually novel method for calculating the PDFs and other properties (e.g., response functions) of the UEG, given the unpolarized E_c from *whatever* source (e.g., STLS, QMC, etc.). The PDFs of classical fluids are easily obtainable from classical Monte Carlo or via the hypernetted chain (HNC) procedure [17]. We consider a *classical* Coulomb fluid with two species (up, down spins). We ask for the temperature T_q at which the PDFs of a Coulomb fluid, obeying the *classical* HNC-integral equation, yield the E_{xc} of the UEG at the same density and at $T = 0$. The suffix q in T_q signifies that this temperature reflects 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 $E_c(r_s, \zeta)$, LFCs, etc. While many interesting applications are possible, here we treat the 3D spin-polarized UEG at zero and finite T.

The physical motivation for our method comes from DFT where interacting electrons are replaced by noninteracting Kohn-Sham (KS) particles whose wave function is a simple determinant. This is very different to the Feenberg approach which uses a correlated $\psi = FD$ [10,11]. In DFT the many-body potential is replaced by a one-body KS potential, V_{KS}. Since the natural energy parameter of the classical ensemble is the temperature, we look for a temperature mapping of the V_{KS}.

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

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

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

Here $\phi_{ij}(r)$ is the pair potential between the species i, j. For two electrons this is just the Coulomb potential $V_{\mathrm{Cou}}(r)$. If the spins are parallel, the Pauli principle prevents them from occupying the same spatial orbital. Following the earlier work, notably by Lado [18], we also introduce a "Pauli potential," $\mathcal{P}(r)$. Thus $\phi_{ij}(r)$ becomes $\mathcal{P}(r)\delta_{ij} + V_{\mathrm{Cou}}(r)$. The Pauli potential $\mathcal{P}(r)$ will be discussed with the PDFs of the noninteracting UEG, i.e., $g_{ij}^0(r)$. 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 (DCF)" of the Ornstein-Zernike (OZ) equations.

$$h_{ij}(r) = c_{ij}(r) + \Sigma_s \bar{n}_s \int d\mathbf{r}' h_{i,s}(|\mathbf{r} - \mathbf{r}'|) c_{s,j}(\mathbf{r}'). \tag{2}$$

The $B_{ij}(r)$ term in Eq. (1) is the "bridge" term arising from certain cluster interactions. If this is neglected, Eqs. (1) and (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 [19]. $B(r)$ is important when the coupling constant Γ exceeds, say, 20. The range of Γ 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 including a suitable bridge term.

Consider the noninteracting system at temperature T, with $x_i = 0.5$ for the paramagnetic case. The parallel-spin PDF, i.e., $g_{ii}^0(r, T)$, will be denoted by $g_T^0(r)$ for simplicity, since $g_{ij}^0(r, T)$, $i \neq j$ is unity. Denoting $(\mathbf{r}_1 - \mathbf{r}_2)$ by $\mathbf{r}$, it is easy to show that

$$g_T^0(\mathbf{r}) = \frac{2}{N^2} \Sigma_{\mathbf{k}_1,\mathbf{k}_2} n(k_1) n(k_2) [1 - e^{i(\mathbf{k}_1 - \mathbf{k}_2)\cdot\mathbf{r}}]. \tag{3}$$

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

$$g_T^0(r) = 1 - F_T^2(r), \tag{4}$$

$$F_T(r) = (6\pi^2/k_F^3) \int n(k) \frac{\sin(kr)}{r} \frac{k\,dk}{2\pi^2}. \tag{5}$$

Here k_F is the Fermi momentum. Thus $g_T^0(r)$ is obtained from the Fourier transform of the Fermi function. Then $c^0(r)$ can be evaluated from $g_T^0(r)$ using the OZ relations. The $T = 0$ case can be evaluated analytically [18].

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

$$g^0(r) = \exp[-\beta\mathcal{P}(r) + h^0(r) - c^0(r)]. \tag{6}$$

The k-space DCF, i.e., $c^0(k)$, decays as $4k_F/3k$ for small k and for $T = 0$, showing that the r-space form $c^0(r)$ is long ranged. The Pauli potential $\mathcal{P}(r)$ is given by

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

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 rk_F at $T = 0$. It is long ranged and mimics the exclusion effects of Fermi statistics. At finite T its range is about a thermal wavelength and is increasingly hard-sphere-like. Plots of $\beta\mathcal{P}(r)$ and related functions are given in Fig. 1.

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

$$g_{ij}(r) = e^{-\beta[\mathcal{P}(r)\delta_{ij} + V_{\mathrm{Cou}}(r)] + h_{ij}(r) - c_{ij}(r)}, \tag{8}$$

$$h_{ij}(q) \xrightarrow{\mathrm{FT}} h_{ij}(r), \tag{9}$$

$$h_{11}(q) = c_{11}(q) + (\bar{n}/2)[c_{11}(q)h_{11}(q) + c_{12}(q)h_{21}(q)], \tag{10}$$

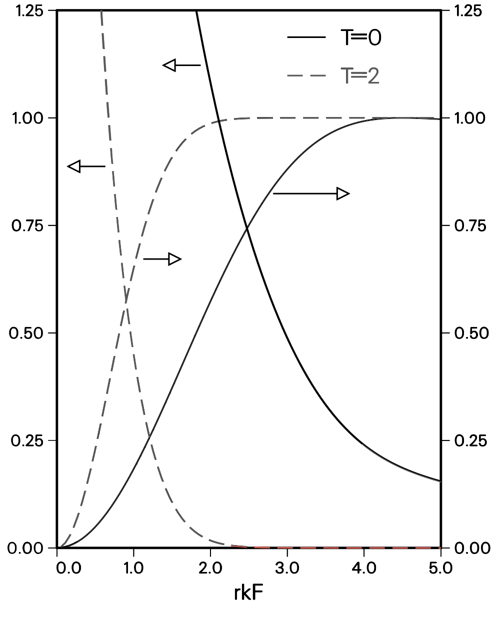

FIG. 1. The Pauli potential, Eq. (8), and the noninteracting PDF, $g_{11}^0(r)$ at $T/E_F = 0$ and 2 (dashed lines). They are universal functions of rk_F, where k_F is the Fermi wave vector. The arrows refer the curves to the axes.

$$h_{12}(q) = c_{12}(q) + (\overline{n}/2)[c_{11}(q)h_{12}(q) + c_{12}(q)h_{22}(q)], \tag{11}$$

The Coulomb potential $V_{\text{Cou}}(r)$ 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 Minoo *et al.* [20], we use a "diffraction corrected" form:

$$V_{\text{Cou}}(r) = (1/r)[1 - e^{-r/\lambda_{th}}]; \quad \lambda_{th} = (2\pi\overline{m}T_{cf})^{-1/2}. \tag{12}$$

Here $\overline{m}$ is the reduced mass of the electron pair, i.e., $m^*(r_s)/2$ a.u., where $m^*(r_s)$ is the electron effective mass. It is weakly r_s dependent, e.g., ~ 0.96 for $r_s = 1$. In this work we take $m^* = 1$. The "diffraction correction" ensures the correct behavior 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 $T_{cf} = T_q$. A trial T_q is adjusted to obtain an $E_c(T_q)$ equal to the known *paramagnetic* $E_c(r_s)$ at each r_s, via a coupling constant integration.

$$E_{\text{xc}}(T_q) = \int_0^1 \frac{d\lambda}{2} \int \frac{4\pi r^2\,dr}{r}[h_{11}(r,\lambda) + h_{12}(r,\lambda)] \tag{13}$$

(E_x alone is obtained if λ is fixed at 0). The resulting "quantum" temperatures T_q could be fitted to the form

$$T_q/E_F = 1.0/(a + b\sqrt{r_s} + cr_s). \tag{14}$$

The results for E_c from different QMC 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 from diffusion Monte Carlo (DMC) [13]. The difference in E_c in VMC and DMC leads 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 $c = 0.0$. Eight values of r_s, viz., $r_s = 1$–6, 8, and 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 $r_s \to 0$, $g(r)$ tends to $g^0(r)$. The UEG as $r_s \to 0$ goes to a high-density fluid interacting via the Pauli potential.

For any given r_s, given the T_q from the paramagnetic case, we can obtain $g_{ij}(r)$ and $E_{\text{xc}}(r_s, \zeta, T)$ [21], at *arbitrary* spin polarization ζ by solving the coupled HNC equations. Unlike in many theories of electron fluids, the PDFs obtained from the HNC procedure are positive definite at all r_s. In Fig. 2 we show typical results for $g_{ij}(r)$ and comparisons with QMC simulations. Our results are in excellent agreement with the DMC results. The depletion hole for $g_{12}(r = 0)$ from DMC are in close agreement and are deeper than those from VMC [13]. The difference in VMC and DMC, even at $r_s = 1$, Fig. 2(a), signals that even when the E_c are in agreement, other properties may have bigger errors. Figure 2(c) displays our paramagnetic PDFs and those of DMC at $r_s = 1$ and 10.

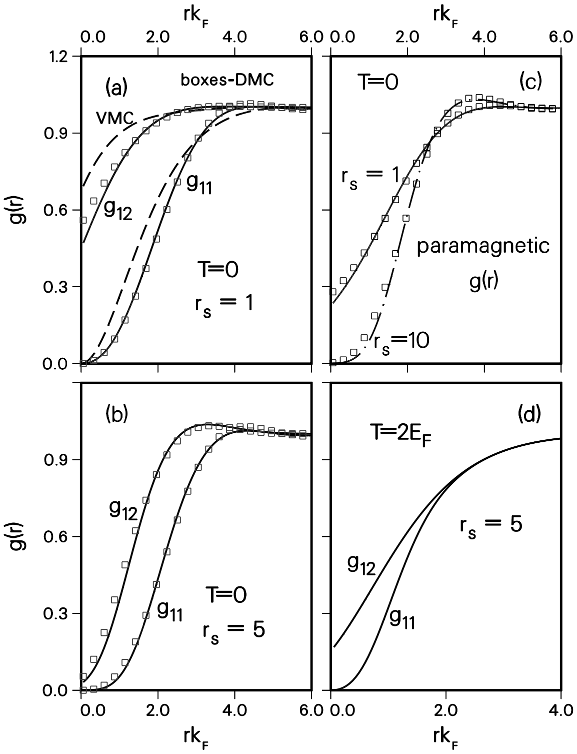

FIG. 2. (a) Here the HNC $g(r)$ are compared with VMC and DMC simulation results: the interacting PDFs $g_{11}(r)$ and $g_{12}(r)$ at $r_s = 1$ are shown. Solid lines: HNC; boxes: DMC; dashed line: VMC. (b) $r_s = 5$, DMC, and HNC. In (c) the paramagnetic $g(r)$ at $r_s = 1$ and $r_s = 10$, $T = 0$ are compared with DMC. (d) Finite temperature PDFs (HNC) for $T/E_F = 2$, $r_s = 5$.

At present there is no reliable finite-temperature microscopic theory to compare with the $g(r, \zeta, T)$ obtained by our HNC approach. The theory of Tanaka and Ichimaru [5] improves on STLS and is more comprehensive. However, like in STLS, the $g(r)$ has unphysical regions for r_s near 5. Figure 2(d) gives the $g(r)$ at $r_s = 5$, $T = 2T_F$ obtained from HNC. 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 $E_c(r_s, \zeta, T)$ at any ζ. The QMC results for $E_c(r_s, \zeta)$ at $T = 0$ agree with ours, since our $g_{ij}(r)$ agree with those from MC. For example, at $r_s = 10$, the spin-polarized $-E_c$ is as follows: Ceperley-Alder, 0.0209 Ry; Ortiz-Ballone, 0.0206 Ry; our method, 0.0201 Ry; Lantto (Fermi-HNC), 0.0186 Ry; Kallio and Piilo, 0.0171 Ry [11].

The response $\chi(k, \omega)$ of the interacting UEG is usually written in terms of a reference $\chi^0(k, \omega)$ and an LFC denoted by $G(k, \omega)$. The simplest static form, $G(k)$, differs from $G(k, 0)$, especially at large k. The main thrust of STLS [6], GT [3], UI (Utsumi and Ichimaru) [23] and others has been to provide the $G(k)$ as a function of r_s at $T = 0$. Even though UI begins as a basic theory, it actually relies on fit parameters constraining the $G(k)$ to fits to Monte Carlo E_{xc} and derivatives, Yasuhara's $g(0)$, etc.,

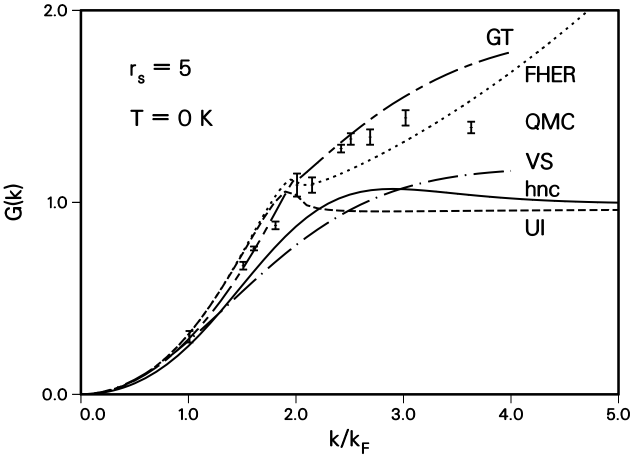

FIG. 3. The local-field correction $G(k)$, Eq. (16), to the static response at $T = 0$ and $r_s = 5$. Results shown are as follows: HNC; Utsumi and Ichimaru (UI) [23]; Vashista and Singwi (VS) [6]; QMC [14]; Farid *et al.* (FHER) [8]; and Geldart and Taylor (GT) [3]. QMC and FHER explicitly base their LFC on the Lindhard χ^0 while some of the others are explicitly or implicitly based on a χ_I^0 (see text).

i.e., quantities outside UI theory. FHER is a fitted form using over two dozen parameters. The only parameter of the present model is T_q. It provides approximate $G(k)$ comparable to STLS and UI, involves only a few algebraic steps, and holds for finite T as well. Thus consider the simplest LFC, viz., $G(k)$, for a one-component fluid.

$$V_{\mathrm{Cou}}(k)G(k) = V_{\mathrm{Cou}}(k) + 1/\chi(k) - 1/\chi^0(k). \quad (15)$$

For a classical fluid, $\chi(k)$ can be expressed in terms of $\beta S(k)$. Hence, for the paramagnetic case,

$$V_{\mathrm{Cou}}(k)G(k) = V_{\mathrm{Cou}}(k) - (T/n)[1/S(k) - 1/S^0(k)]. \quad (16)$$

In these expressions the $\chi^0(k)$ and $S^0(k)$ are based on a Slater determinant, while the Lindhard function is applicable to the Hartree case. We display in Fig. 3 the $T = 0$ LFC for $r_s = 5$, and the LFCs of other methods. The LFCs based on a χ_I^0 calculated with the interacting density distribution tend to a constant for large k, viz., $G(k,0) \rightarrow 2[1 - g(0)]/3$, while $G(k) \rightarrow 1 - g(0)$. Although the theory of UI is based on a χ_I^0, in practice the Lindhard χ^0 is used. The fitted $G(k,0)$ of FHER is built to behave like k^2 at large k, being an LFC based on the Lindhard form [24]. It can be shown analytically that the HNC-LFC tends to $1 - g(0)$ for large k, as required. Thus it is seen that the approximate forms, QMC-LFC, as well as the HNC-LFC, are in general agreement.

In conclusion, we have presented a simple classical mapping of a quantum-Fermi liquid for any spin polarization and temperature T and shown that it *quantitatively* recovers the quantum Monte Carlo PDFs at $T = 0$. The only parameter is a temperature mapping of the E_{xc}. We have examined spin-dependent correlation energies, PDFs at zero and finite T, as well as a simple approximation to the LFC of the electron response at arbitrary T. The method has potential applications to nonlocal E_{xc} in DFT, 2D electrons, Bose systems, and extensions to dynamical models of quantum fluids.

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*Electronic address: chandre@cm1.phy.nrc.ca

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