The Classical-Map Hyper-Netted-Chain (CHNC) technique for inhomogeneous electron systems. Application to quantum dots.

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The Classical-map Hyper-Netted-Chain (CHNC) technique is a simple method of calculating quantum pair-distribution functions, spin-dependent energies, etc., of strongly-interacting uniform systems. We present CHNC calculations of charge densities and energies of non-uniform systems, viz., quantum dots, and compare with quantum Monte Carlo and density-functional results. Results for up to 210 electrons are reported.

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The Hohenberg-Kohn and Mermin (HKM) theorems [1] of density-functional theory (DFT) assert that the one-body density \( n(\vec{r}) \) of an inhomogeneous system completely determines its physics. However, DFT uses the more laborious Kohn-Sham (K-S) approach [2] due to the lack of an accurate kinetic-energy functional [2]. The Kohn-Sham \( n(\vec{r}) \) of an electron system is:

\[
n(\vec{r}) = \sum_\nu |\phi_\nu(\vec{r})|^2 f_\nu(\epsilon_\nu/T)
\]

Here \( \phi_\nu \) are K-S eigenstates with “energies” \( \epsilon_\nu \), occupations factors \( f_\nu \) at the temperature \( T = 1/\beta \). Here \( \nu \) includes spin and other relevant quantum numbers. Quantum systems at high temperatures behave classically. Then \( n(r) \) is given by the Boltzmann form:

\[
n(r) = n_0(0) \exp(-\beta V_{KS}(r))
\]

where \( n_0(0) \) is a reference density, and \( V_{KS} \) is a classical Kohn-Sham potential. This suggests that the kinetic-energy functional may be side-stepped by (i) the use of an equivalent “classical-fluid” at a temperature \( T_{cf} \) for the quantum system whose actual physical temperature \( T \) may even be zero; (ii) use of effective classical pair-potentials to mimic quantum effects. In the following we suggest such a “classical map”, enabling a great simplification in quantum calculations of interacting electrons.

All quantum observables are mean values over suitable distribution functions, formed by averaging over most of the variables in the square of the many-body wavefunction. The most useful averaged quantities are the one-body and two-body distributions. We had already demonstrated a simple but accurate classical map for the interacting uniform electron fluid (UEF), by presenting explicit calculations of spin-polarized pair-distribution functions (PDFs) at zero and finite \( T \), for the 3D electron liquid [4, 5], the 2D-electron fluid [6, 7, 8, 9] and multi-valley systems [10]. Fermi-liquid parameters of thick-electron layers have also been determined via this classical-map technique [11]. It has been successfully applied to hot dense hydrogen and related systems [12, 13].

The method employs a classical Coulomb fluid whose PDFs are determined via classical statistical mechanics. The map using the Hyper-Netted-chain (HNC) method is named the CHNC. Molecular dynamics (MD) simulations may also be used [12], where it was called the CMMD. The temperature \( T_0 \) of the “equivalent classical Coulomb fluid” is chosen to reproduce the correlation energy of the original quantum fluid at \( T = 0 \). Then the PDFs of the classical fluid at \( T_{cf} = \sqrt{T^2 + T_0^2} \) are excellent approximations to the PDFs of the quantum fluid at \( T \). The so-obtained PDFs are then used in the standard way to obtain the energies and other properties of the system. Where possible, the accuracy of the CHNC results have been demonstrated by comparison with quantum Monte Carlo (QMC) or DFT results.

Here we apply the CHNC to a typical inhomogeneous systems, viz., electrons trapped in 2D parabolic potentials (quantum dots). In standard calculations, if \( N_e \) electrons are in the “external potential”, a suitable basis set of \( N_b \) functions, with \( N_b \) significantly larger than \( N_e \), is selected. A Hartree-Fock (single-determinant) calculation is followed by a configuration-interaction (CI) expansion in Slater determinants. The complexity of the problem grows factorially with \( N_b \). It is the electron-electron interactions, which make the problem prohibitive. In CHNC or CMMD, we treat many-body effects classically (i.e., an \( O(0) \) approach), while the non-interacting Hamiltonian \( H_0 \) is treated exactly. Here we summarize the salient features of the classical-map technique: (i) Assignment of a classical-fluid temperature \( T_{cf} \) to the electron system. (ii) Replacement of the Coulomb-interaction operator \( 1/r \) by a classical “diffraction potential” \( v_c(r) = \{1 - \exp(-k_{th}r)\}/r \) which accounts for the thermal de Broglie length \( 1/k_{th} \) of the electron at \( T_{cf} \). (iii) Ensuring that the non-interacting electron PDFs with spin polarization \( \zeta \), viz., \( g^\zeta(r, T, \zeta) \) are correctly recovered if the Coulomb interaction is switched off. Thus a “Pauli exclusion potential” \( P(\vec{r}) \) is used to exactly reproduce the Fermi hole [14] in \( g^\zeta(r, T, \zeta) \). We apply CHNC to parabolic quantum dots to show that their interacting inhomogeneous charge densities and energies can be readily calculated via CHNC, for arbitrary \( N_e \).

Quantum dots– Electrons trapped in parabolic potentials are found in Fermion traps and in the quantum dots.
of nanotechnology\cite{15}. Such quantum dots have been studied extensively by several methods\cite{16,17}, including QMC\cite{18}. Using atomic units ($|e| = \hbar = m_e = 1$), the Hamiltonian operator $H = H_0 + H_{\text{int}}$ for electrons subject to a potential $u(\vec{r})$ is given by

$$H_0 = \sum_i \left[ \frac{\nabla_i^2}{2} + u_i(\vec{r}_i) \right], \quad H_{\text{int}} = \frac{1}{2} \sum_{i \neq j} \frac{1}{r_{ij}}$$  \hspace{1cm} (3)

The classical map converts the Hamiltonian operator to the classical Hamiltonian $H_c$.

$$H_c = \sum_i \left[ \frac{\vec{p}_i^2}{2m_e} + u_c(\vec{r}_i) \right] + \frac{1}{2} \sum_{i \neq j} \left[ v_c(\vec{r}_{ij}) + P_{s_i s_j}(\vec{r}_{ij}) \right]$$  \hspace{1cm} (4)

Here $\vec{p}_i$ is the momentum of the $i$-th electron, with spin $s_i$. The Coulomb-interaction operator $1/\vec{r}$ is replaced by the well-known diffraction-corrected classical form $v_c(\vec{r})$. The operator defining the parabolic confinement in the dot is $u(\vec{r}) = \omega_0^2 \vec{r}^2/2$. It maps to the classical function $u_c(\vec{r})$, constructed so that the non-interacting density $n^0(\vec{r})$ is recovered from $u_c(\vec{r})$ as a classical distribution. The potentials $u(\vec{r})$ and $u_c(\vec{r})$ differ because the quantum system is sensitive to the boundary conditions imposed on the eigenstates of $H_0$.

Thus the essential input to the classical mapping of inhomogeneous systems is the non-interacting density $n^0(\vec{r})$. Here we suppress vector notation (unless needed for clarity) and consider circular quantum dots. Given $n^0(\vec{r})$, we seek the classical potential which generated it. As this involves the inversion of an inhomogeneous HNC-type equation, we proceed indirectly. If the presence of each electron did not affect any other electron, the corresponding classical potential $u_c(\vec{r})$ is:

$$n^0(\vec{r}) \equiv n^0(0) \exp\{-\beta u_c(\vec{r})\}$$  \hspace{1cm} (5)

This equation determines the product $\beta u_c(\vec{r})$, and not separately the potential $u_c(\vec{r})$, or an inverse temperature $\beta$. The reference zero of all potentials will be referred to the center of the dot. The classical potential $u_c(x)$ depends on $N_e$ even though the electrons are non-interacting. In effect, they have developed indirect steric interactions via the confining potential.

The confining potential defines a length scale $\ell_0 = \sqrt{\hbar/(m \omega_0)}$. We use the effective mass $m^* = 0.067$ and the dielectric constant $\kappa = 12.4$ typical of GaAs. These define effective atomic units (a.u.) with a Hartree energy of $me^4/(\hbar^2\kappa) \approx 11$ meV, and a Bohr radius $a_B = \hbar^2 \kappa/(me^2) \approx 9.79$ nm. In Fig.1(a) we show the $n^0(\vec{r})$ and $\beta u_c(\vec{r})$ for a 20-electron circular quantum dot, $N_e = 20$, with $\omega_0 = 3.33$ meV\cite{15}. The non-interacting density $N^0(\vec{r})$ shows shell-filling effects. In Fig.1(a) we show a smoothed charge density $n^\alpha(\vec{r})$ as well. The density difference $n^0(\vec{r}) - n^\alpha(\vec{r})$ integrates to zero, and may be though of as the result of a steric interaction $U_{\text{str}}(\vec{r})$ arising from the packing of classical particles into the parabolic trap. In the quantum system it arises from the boundary conditions on $H_0$. As many shells are filled with increasing $N_e$, and as Coulomb interactions come on, this effect becomes unimportant, as seen in the interacting-$n(\vec{r})$ for the dot with $N_e=210$, in Fig.1(d).

The electron-distribution $n_\alpha(\vec{r})$, spin $\alpha$, in the dot defines a dot-electron PDF by, e.g., $n_\alpha(r) = n_\alpha(0)g_{d\alpha}(r)$. This in turn depends on the electron-electron PDFs $g_{ee}$, i.e., explicitly, $g_{\alpha\alpha}(\vec{r},\vec{r}')$. Evaluating this coupled set is complex even within classical mechanics. Unlike in uniform systems, these PDF’s depend on $\vec{r}, \vec{r}'$. While a classical-map-MD approach is the best option, we show

![Diagram](image)
that simple approximations to the CHNC integral equations lead to surprisingly accurate results.

Our simplified approach is based on constructing the density \( n_{da}(r) = n_q(0)g_{da}(r) \) while the e-e PDFs \( g_{a,a'}(\vec{r}, \vec{r}') \) are evaluated from an average-density approximation. This saves us from solving a set of coupled HNC equations. The approximations proposed are: (i) replacing the e-e PDFs \( g_{a,a'}(\vec{r}, \vec{r}') \) by the PDFs of a uniform slab (USB) of average density, \( \bar{n} \), (ii) determining the USB density, viz., \( \bar{n} \) from \( n(r) \) using Eq. (3.4) given there, (iii) using \( \bar{n} \) and the equations of Ref. 6 to determine \( T_q \) and other UEF parameters needed for the CHNC, (iv) calculating the charge density \( n_{da}(r) \) in the quantum dot via a simplified HNC-like classical integral equation inclusive of a bridge term \( B_{de}(r) \). For brevity of presentation, we use a spin-unpolarized system (\( \zeta = 0 \)), and write \( g_{de}(r) \) for the PDF defining the charge density in the dot. The basic CHNC equation for the electron density in the inhomogeneous system is:

\[
n(r) = n(0) \exp[-bU_c(r) + V_{m-b}] \tag{6}
\]

This is a Boltzmann distribution for the pair-potential \( u_c(r) \) inclusive of its many-body correction \( V_{m-b} \). This includes mean-field and correlation effects. The classical mean-field term is just a Poisson potential. We also need the correlation potentials beyond mean-field from classical mechanics. Hence we rewrite \( V_{m-b} \) in terms of the Nodal function \( N(r) \) and the bridge function \( B(r) \) of HNC theory, remembering that “exchange” already appears in the classical map as an effective pair-potential (“Pauli potential”) between like-signs. The “bridge” diagram brings in irreducible (three-body and higher) packing effects beyond simple HNC.

\[
V_{m-b} = N(r) + B_{de}(r) \tag{7}
\]

\[
N(r) = \beta [V_p(r) + V_x(r) + V_c(r)] \tag{8}
\]

\[
V_p(r) = \int n(r')d\vec{r}'/|\vec{r} - \vec{r}'| \tag{9}
\]

\[
V_x(r) = \int n(r')d\vec{r}'P(\vec{r}, \vec{r}') \tag{10}
\]

\[
V_c(r) = \int n(r')d\vec{r}' \left[ \log\{g_{ee}(\vec{r}, \vec{r}')\} - h_{ee}(\vec{r}, \vec{r}') \right] \tag{11}
\]

The nodal term \( N(r) \) has been decomposed into a Poisson potential \( V_p(r) \), an “exchange potential” \( V_x(r) \) arising from the Pauli exclusion potential \( P(\vec{r}, \vec{r}') \), and a classical correlation potential \( V_c(r) \). This is a standard analysis based on the Ornstein-Zernike equation, and was already discussed in Ref. 21. The expression for \( V_c(r) \) is Eq. (3.4) given there, and \( h_{ee}(\vec{r}, \vec{r}') \) is the total correlation function. \( V_p \) has been written with the Coulomb potential \( 1/r \) rather than with the diffraction correction (DC), since we found that the numerical effects of DC are negligible for the \( g_{de}(r) \) of the quantum dots studied here (n.b. the DC is needed in the CHNC evaluation of the \( g_{ee}(r) \) at the uniform-slab density \( \bar{n} \)).

We approximate \( g_{ee}(\vec{r}, \vec{r}') \) by \( g_{ee}(|\vec{r} - \vec{r}'|) \) of the uniform slab at density \( \bar{n} \). The \( \bar{n} \) in our quantum dots are found to have Wigner-Seitz radii \( r_s \sim 1 \) and hence the Coulomb correlation potential \( V_c(r) \) is small, while the Pauli exclusion effect is large. Here \( P(\vec{r}, \vec{r}') \) is replaced by \( P(|\vec{r} - \vec{r}'|) \) at the uniform density \( \bar{n} \). This is a universal function of \( x = r/r_s \). It can be fitted to \( \{-2a\log(x) + b\}/(1 + cx) \), or to the simpler form \( a_1/(1 + a_2x) \), with \( a_1 = 10.1186, a_2 = 3.69352 \), for 2D systems at \( T = 0 \). Replacing \( g_{ee}(\vec{r}, \vec{r}') \) by its uniform-density value leads to the question of the appropriate form for \( n(\vec{r}') \) which now has to play the role of a \( n(\vec{r}) \). The usual simple choices (e.g., \( n(\vec{r}') \rightarrow (n(\vec{r}) + n(\vec{r}'))/2 \), or even \( n(\vec{r}) \) give similar results, but the replacement \( n(\vec{r}') \rightarrow \bar{n} \), i.e., completely by a uniform slab, is too drastic.

The results obtained from the self-consistent solution of Eq. (6) are shown in Fig. 11 Panel (a) shows the input potential \( \beta u_c(r) \) based on the non-interacting density \( n^0(r) \). This \( n^0(r) \) gets modified by the interactions. The average density \( \bar{n} \), the corresponding inverse temperature \( \beta \), potentials \( V_p, V_x \) etc., were calculated from \( n(r) \) in each iteration, with the total number of electrons fixed to \( N_e \). In panel (b) the self-consistent \( n(r) \) obtained as we successively add Poisson (curve with boxes), exchange-correlation (dashed red curve), and Bridge corrections (solid blue curve) is found to converge to the benchmark results from QMC and DFT (solid green circles). Panels (c), (d) shows the full CHNC results and QMC and/or DFT results for \( N_e = 6, 12, 20, 30 \). These have been done using the quantum temperature \( T_q \) defined by Eq. 5 of Ref. 20, which assigns \( a_q \) to a given \( r_s \). For high electron densities when \( r_s < 1 \), the Bulut-Tanatar(BT) map seems to be more accurate. Calculations for \( N_e = 110, 210 \) etc, using a BT-type map are given in panel (d), but no published QMC or DFT results are available.

An oscillatory structure in \( g_{ee}(r) \) occurs even in uniform fluids, when interactions are important, and is well understood. The \( g_{ee}(r) \) of the uniform fluid at strong coupling could be accurately recovered on including bridge contributions \( B_{ee}(r) \) to the HNC, as shown in Ref. 21. There the particle-packing theory of the hard-sphere fluid could be used, since the PDF is not too sensitive to the details of the bridge interaction. In the classical map of the quantum dot, packing effects are dominated by the steric crowding effect of the confining potential. This steric potential \( U_{str} \) is already available to us in the charge distribution \( n^0(r) \) of \( H_0 \). We assume that the difference between the smoothed distribution \( n^0(r) \), and \( n^0(r) \), Fig 11(a), corresponds to the effect of \( U_{str}(r) \). Then, converting a charge distribution into a potential \( \beta U_{str} \), we write

\[
\beta U_{str}(r) = -\log\{n^0(r)/n^0(0)\} - \log\{n^0_0(r)/n^0_0(0)\}
\]

\[
B_{de}(r) = \gamma (\beta/\beta_0)^2 \xi \gamma V_x(r/\xi), \quad \xi = r_s/\xi_0 \tag{12}
\]

The equation for \( B_{de}(r) \) reflects the rescaling of the
TABLE I: The Exchange-Correlation and kinetic energies evaluated from the densities (Fig. 1), i.e., CHNC \( n(r) \), and from the DFT \( n(r) \) of Ref. [17]. \( N_e \) is the number of electrons. The energy unit is \( \hbar \omega_0 \).

| \( N_e \) | \( E_{\text{kin,CHNC}} \) | \( E_{\text{kin,DFT}} \) | \( -E_{\text{xc,CHNC}} \) | \( -E_{\text{xc,DFT}} \) |
|---|---|---|---|---|
| 6 | 2.317 | 2.415 | 7.491 | 7.638 |
| 12 | 5.981 | 5.897 | 16.97 | 16.80 |
| 20 | 11.55 | 11.47 | 30.20 | 30.07 |
| 30 | 21.30 | 19.51 | 49.90 | 48.78 |
| 110 | 111.2 | – | 213.1 | – |
| 210 | 257.4 | – | 445.7 | – |

uniform slab density \( \bar{n}^0 \) to \( \bar{n} \) due to interactions, changing the scales of the parameters \( \beta^0, r_s^0 \), etc., to \( \beta, r_s \) etc., of the final self-consistent density, thus rescaling the steric potential of the non-interacting system. The numerical factor \( \gamma \) is set to 1.5. This simple model avoids the complex microscopic calculation of a bridge correction, and is seen to be justified \textit{a posteriori}. It does not appeal to any parameterizations outside the problem. In fig. 1(c) we show comparisons of the CHNC \( n(r) \) for \( N_e = 6,12,20 \), with QMC results. DFT results for \( N_e = 20 \) are shown in fig. 1(b). Panel (d) shows the CHNC results for \( N_e = 30, 110 \) and 220 electrons. In the last two cases we do not have microscopic calculations for comparison with CHNC. The calculation of the interacting density \( n(r) \) for arbitrary \( N_e \), at finite temperatures, finite values of \( \zeta \) or finite magnetic fields pose no additional difficulty in CHNC.

The charge distributions of CHNC can now be used for the total energy \( E \), which involves the confinement energy \( E_c \), the Poisson energy \( E_{\text{poi}} \), \( E_{\text{xc}} \), and \( E_{\text{kin}} \). The exchange-correlation and kinetic contributions are the quantum mechanically sensitive, “difficult” terms. The simplest approach is found to be adequate for 2D quantum dots. That is, we use the LDA (local-density approximation), with the known 2D exchange-correlation energy functionals [22]. The success of the LDA for the 2D kinetic energy has been noted by van Zyl et al. [24], and also by Koivisito et al. [2]. A comparison of our xc-energies with those from DFT are given in Table I. In conclusion, we have presented classical-map calculations for a 2D inhomogeneous system of interacting electrons, viz., quantum dots, which are in good agreement with microscopic calculations where available. This method requires no basis sets, no evaluation of matrix elements etc. It is an order-zero, viz., \( \mathcal{O}(0) \) approach independent of the number of electrons. Similar applications to atomic systems, (“naturally occurring quantum dots”), are clearly feasible. The uniform-slab approximation, and the need to model the bridge-diagram corrections to the CHNC equations may be avoided by resorting to a classical molecular-dynamics implementation of the method [22]. The author thanks Mario Gattobigio for providing the DFT \( n(r) \) and energies of Ref [17].

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