

# Phase Diagram of the Low-Density Two-Dimensional Homogeneous Electron Gas

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We have used quantum Monte Carlo (QMC) methods to calculate the zero-temperature phase diagram of the two-dimensional homogeneous electron gas, achieving higher accuracy than previous studies. We find a transition from a paramagnetic Fermi fluid to a triangular Wigner crystal at density parameter  $r_s = 33(1)$  a.u. Our results show that a fully spin-polarized fluid is never stable, in contradiction with earlier work. We have also searched for a recently proposed “hybrid” phase [1], but have not found it.

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The two-dimensional (2D) homogeneous electron gas (HEG) is one of the most important model systems in condensed-matter physics. It is our basic model of the conduction electrons in layered semiconductor devices, and consists of a set of electrons moving in 2D in a uniform, inert, neutralizing background. At high densities the HEG exists in a Fermi fluid phase, but at low densities it forms a Wigner crystal to minimize the electrostatic repulsion between the electrons [2]. Classical 2D Wigner crystals can be produced in the laboratory by spraying electrons onto droplets of liquid He [3], and quantum Wigner crystals can be formed at the interface between two semiconductors [4]. As well as being objects of fundamental scientific interest, it is hoped that 2D Wigner crystals may be of use in quantum computing devices [5].

At high densities the fluid ground state is spin-unpolarized (the paramagnetic fluid), but at low densities this is unstable to a spin-polarized phase (the ferromagnetic fluid). There is some experimental evidence suggesting that a ferromagnetic fluid is stable at densities between the paramagnetic fluid and crystal phases [6], although these data may not be conclusive. Previous theoretical studies [7, 8] have reported a region of stability for the ferromagnetic fluid. However, the energy differences between the various phases are small, and highly accurate calculations are required to resolve them. Another issue of recent interest has been the possible existence of intermediate phases in the vicinity of the fluid–crystal transition [1, 9–11]. In this paper we address the issues of the stability of the ferromagnetic fluid, the fluid–crystal transition, and the possibility of a “hybrid” phase of the type proposed by Falakshahi and Waintal [1].

We have performed quantum Monte Carlo (QMC) [12, 13] calculations for the 2D fluid and Wigner crystal phases, achieving much smaller statistical error bars than previous QMC studies [7, 8, 14]. We used the variational and diffusion quantum Monte Carlo (VMC and DMC) methods as implemented in the CASINO code [15]. DMC is the most accurate method available for studying quantum many-body systems such as electron

gases. Fermionic symmetry is imposed via the fixed-node approximation [16], in which the nodal surface is constrained to equal that of a trial wave function. We used extremely flexible Slater-Jastrow-backflow [17] wave functions for the fluid phases, and we optimized the crystal wave functions by directly minimizing the DMC energy. Our fixed-node DMC energies are therefore more accurate than those of earlier calculations. Finally, we have dealt with finite-size effects in a more sophisticated fashion than previous studies [18].

It is well-established that the triangular crystal has a considerably lower energy than competing lattices in the density range of interest. A triangular lattice was therefore used in all our crystal calculations. Antiferromagnetism is frustrated on a triangular lattice, so we have restricted our attention to ferromagnetic crystals. The electrons are spatially localized around fixed lattice sites in the Wigner crystals studied in this work, breaking the translational symmetry of the Hamiltonian. However, the energy difference per particle between the true, homogeneous ground state and the broken-symmetry phase vanishes in the limit of large system size [19].

In our trial wave functions we used the Jastrow factor proposed in Ref. [20]. Plane-wave orbitals  $\exp(i\mathbf{G} \cdot \mathbf{r})$  were used in the Slater wave function in our fluid calculations, while identical Gaussian orbitals  $\exp(-Cr^2)$  centered on lattice sites were used in our crystal calculations. In all our fluid calculations and some of our crystal calculations, the Slater wave function was evaluated at quasiparticle coordinates defined by the backflow transformation described in Ref. [17]. The backflow functions for antiparallel spins are much larger than those for parallel spins, which are already kept apart by Pauli exclusion. Backflow is much less important in the crystal because the localization of the orbitals already keeps the electrons apart. For example, at  $r_s = 30$  a.u. [21] we find that backflow lowers the DMC energy of the paramagnetic fluid by a significant amount [0.000036(3) a.u. per electron], but the energy of the ferromagnetic Wigner crystal is lowered by an insignificant amount [0.0000010(4) a.u. per electron].

The free parameters in the Jastrow factor and back-

flow function were optimized within VMC by minimizing the variance of the energy [22, 23]. Time-step bias was removed from our final DMC energies by linear extrapolation to zero time step [24]. A target population of at least 1500 configurations was used in our production runs, making population-control bias negligible [24]. (Population-control bias has previously been found to be a significant problem at low densities [19].)

We optimized the Gaussian exponent  $C$  in the crystal orbitals by minimizing the fixed-node DMC energy. At  $r_s = 40$  a.u., the DMC-optimized exponent ( $C_{\text{DMC}} \approx 0.0003$  a.u.) is smaller than the exponent obtained by minimizing the VMC energy ( $C_{\text{VMC}} \approx 0.0006$  a.u. with our Jastrow factor), which is, in turn, substantially smaller than the exponent within either Hartree or Hartree-Fock theory ( $C_{\text{H}} = 0.0019$  a.u.) [25]. The DMC energy depends much less sensitively than the VMC energy on the value of  $C$  [24]. However, the energy difference between DMC results obtained with  $C = C_{\text{DMC}}$  and  $C = C_{\text{VMC}}$  is statistically significant, while the fixed-node error that results from using  $C = C_{\text{H}}$  is very substantial. We find that the DMC-optimized exponents of 64-electron crystals at different densities are given by  $C_{\text{DMC}} = 0.071 r_s^{-3/2}$  and that the DMC-optimized exponents for 16-, 64-, and 196-electron crystals at  $r_s = 40$  a.u. are very similar. We have therefore used  $C = C_{\text{DMC}}$  at all densities and system sizes.

Simulations were performed with up to 162 electrons for the paramagnetic fluid, 109 electrons for the fully spin-polarized (ferromagnetic) fluid, and 196 electrons for the crystal [24]. We eliminated single-particle finite-size effects from the fluid energies by twist averaging [26]. Every so often during VMC or DMC simulations, an offset to the grid of  $\mathbf{G}$  vectors was chosen at random in the first Brillouin zone of the simulation cell, the lowest-energy plane-wave orbitals were selected, and a short period of re-equilibration was carried out. The particle number was kept fixed, so that we worked in the canonical ensemble. We have recently demonstrated that the finite-size errors in the energy per particle of a 2D HEG resulting from the compression of the exchange-correlation hole into the simulation cell and the neglect of long-ranged correlations in the kinetic energy fall off as  $N^{-5/4}$  [18]. We therefore extrapolated our fluid energies to infinite system size by fitting our data at each density to  $E_N = E_\infty - c/N^{5/4}$ , where  $E_N$  is the energy per electron of the  $N$ -electron system and  $c$  and  $E_\infty$  are fitting parameters [24]. This differs from the form of bias that has been incorrectly assumed in previous studies of the 2D HEG [7, 8, 14].

Our DMC results for the Wigner crystal are given in Table I. Our Wigner-crystal energy data were fitted to the first five terms in the low-density expansion of the crystal energy ( $E = c_1 r_s^{-1} + c_{3/2} r_s^{-3/2} + c_2 r_s^{-2} + c_{5/2} r_s^{-5/2} + c_3 r_s^{-3}$ ) [27]. The first term is the

$r_s$ (a.u.)	DMC energy (a.u. / elec.)		
	Crystal	Para. fluid	Ferro. fluid
15	-0.059 647 8(4)	...	...
20	-0.046 186 5(8)	-0.046 267(3)	-0.046 213(3)
25	-0.037 726 3(5)	-0.037 767(1)	-0.037 740(2)
30	-0.031 910 6(8)	-0.031 923(1)	-0.031 913(1)
35	-0.027 666 5(5)	-0.027 660(1)	-0.027 657(1)
40	-0.024 428 7(4)	-0.024 411(2)	-0.024 416(1)
45	-0.021 878 2(7)	...	...
50	-0.019 814 2(4)	...	...

TABLE I: DMC energy as a function of density parameter  $r_s$  for a ferromagnetic triangular Wigner crystal, a paramagnetic Fermi fluid, and a fully ferromagnetic Fermi fluid. All results have been extrapolated to zero time step and infinite system size.

Madelung energy of the static lattice, while the second is the quasiharmonic zero-point phonon energy. The corresponding coefficients can be determined analytically:  $c_1 = -1.106103$  and  $c_{3/2} = 0.814$  [28]. The remaining three coefficients were determined by fitting to our QMC data. We find that  $c_2 = 0.1349362$ ,  $c_{5/2} = -1.364242$ , and  $c_3 = 3.524405$ . Our DMC results for the paramagnetic and ferromagnetic fluids are also given in Table I. We fitted our fluid energy data to the parameterization of the correlation energy suggested by Rapisarda and Senatore [Eq. (28) of Ref. 8]. For the paramagnetic fluid we find  $a_0 = -0.1451806$  a.u.,  $a_1 = 7.721501$ ,  $a_2 = 0.1080078$ , and  $a_3 = 5.091923$ , where  $a_0$ ,  $a_1$ ,  $a_2$ , and  $a_3$  are the free parameters in Rapisarda and Senatore's fitting form for the correlation energy [8]. For our ferromagnetic data we find  $a_0 = -0.2909102$  a.u.,  $a_1 = -0.6243836$ ,  $a_2 = 1.656628$ , and  $a_3 = 3.791685$ .

We find a greater energy difference between the energies of the paramagnetic and ferromagnetic Fermi fluids at  $r_s = 20$  than Attacalite *et al.* [14], which results in somewhat smaller values of the spin susceptibility.

The ground-state energies of the different phases of the 2D HEG are plotted against  $r_s$  in Fig. 1. Unlike previous QMC studies, our statistical error bars are sufficiently small that we can resolve the energy difference between the ferromagnetic and paramagnetic fluids. We can also identify the crystallization density with much greater precision. Previous studies found crystallization to occur at  $r_s = 37(5)$  a.u. [7] and  $r_s = 34(4)$  a.u. [8]. We find the crystallization density to be  $r_s = 33(1)$  a.u., but the transition is from the paramagnetic fluid, not the ferromagnetic fluid as found in the previous studies. Our calculations locate the density at which the paramagnetic fluid becomes unstable to the ferromagnetic fluid at  $r_s = 36(1)$  a.u., but at this density the Wigner crystal is more stable than either fluid phase, so we do not find a region of stability for the ferromagnetic fluid. We

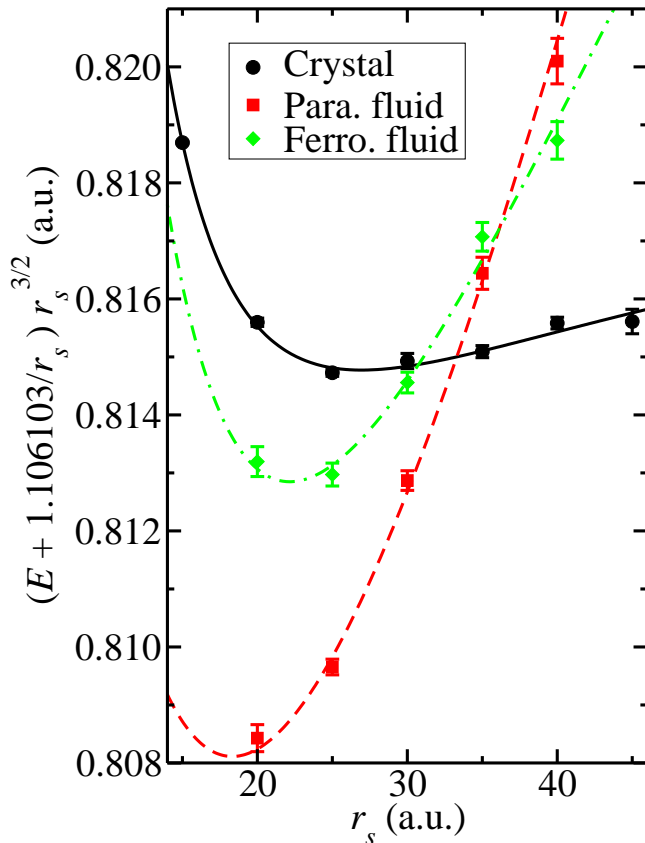


FIG. 1: (Color online) DMC energy as a function of density parameter  $r_s$  for a ferromagnetic triangular Wigner crystal, a paramagnetic Fermi fluid, and a fully ferromagnetic Fermi fluid. In each case the Madelung energy of a triangular crystal has been subtracted from the energy and the result has been multiplied by  $r_s^{3/2}$  to highlight the differences between the curves.

have looked for a region of stability for a partially spin-polarized fluid by calculating the energy of a fluid of spin polarization  $\zeta = 2/5$  at  $r_s = 35$  a.u. The DMC energy, extrapolated to zero time step and infinite size, is  $-0.027659(2)$  a.u. per electron, which is not significantly less than the energies of the paramagnetic and ferromagnetic fluids, but is significantly higher than the crystal energy (see Table I). It is therefore unlikely that a region of stability exists for a partially spin-polarized fluid.

It has been argued that the transition from a 2D Fermi fluid to a Wigner crystal cannot be first order, because, at the transition density, it is energetically favorable to create boundaries between macroscopically separated regions of fluid and crystal [9]. The energy of the Bose fluid is substantially lower than that of the Fermi fluid in the vicinity of the crystallization density in 2D, unlike 3D [29]. There is therefore more scope for interesting phase behavior in 2D. Various intermediate phases have been proposed in the literature, such as a hexatic phase with orientational but not translational order [10], a supersolid

phase [11], or a microemulsion phase [9]. Falakshahi and Waintal [1] have suggested that a “hybrid” phase is stable in the vicinity of the transition density from a ferromagnetic fluid to a Wigner crystal. (We have found that a paramagnetic fluid is stable at this density; however, we restrict our attention to ferromagnetic HEGs to investigate the proposed hybrid phase.) The hybrid phase has the same symmetry as the Wigner crystal, but has partially delocalized orbitals.

Falakshahi and Waintal generated hybrid orbitals for the ferromagnetic  $N$ -electron system by solving the Schrödinger equation for a single, positively charged particle moving in a lattice of negative charges placed at the  $N$  Wigner-crystal lattice sites. If the charge of the test particle is zero then the orbitals for the system are plane waves, i.e., fluid orbitals are obtained. If the charge is large then localized (crystal-like) orbitals are obtained. As the charge of the test particle is increased from zero, there must come a point at which a band gap opens up between the  $N$  and  $(N + 1)$ th states. The set of orbitals obtained at the point at which the gap opens correspond to the hybrid phase. We have tried to find such a hybrid phase by optimizing orbitals of the form

$$\phi(\mathbf{r}) = A \exp(-Cr^2) + \sum_P c_P \sum_{\mathbf{G} \in P} \cos(\mathbf{G} \cdot \mathbf{r}) \quad (1)$$

centered on the crystal lattice sites, where  $P$  denotes a star of symmetry-equivalent simulation-cell reciprocal-lattice vectors. The coefficients  $\{c_P\}$  and  $C$  are optimizable parameters. Equation (1) is a general expansion of Wannier-like orbitals. It can therefore describe the crystal and hybrid phases, but not the fluid, since the latter corresponds to a partially filled band.

Our QMC results for ferromagnetic HEGs at  $r_s = 30$  a.u. obtained with different forms of orbital are shown in Table II. At the VMC level it is possible to lower the energy by optimizing the plane-wave coefficients in Eq. (1), apparently suggesting that Gaussian crystal orbitals are nonoptimal. This does not carry over to DMC, however. In fact it is more important to use Gaussian exponents optimized within DMC than it is to use either plane-wave expansions in the orbitals or backflow functions, suggesting that the fixed-node errors in our crystal DMC energies are very small. We have searched for the hybrid phase by optimizing wave functions with orbitals of the form given in Eq. (1) using different starting points, but have not been able to lower the DMC energy. While this does not prove the nonexistence of the hybrid phase, as searching for minima in a high-dimensional space is well-known to be a very difficult problem [30], the fact that our extensive searches have been unable to find it strongly suggests that it may be a numerical artifact in Falakshahi and Waintal’s work [1].

In summary, we have studied the zero-temperature phase behavior of the 2D HEG using QMC. We find a transition from a paramagnetic fluid to a triangular

Method	Orbitals	Backflow	Energy (a.u. / elec.)
VMC	DMC-opt. Gaussian	No	-0.031 840 13(3)
VMC	VMC-opt. Gaussian	No	-0.031 850 15(9)
VMC	Gaussian+plane-wave	No	-0.031 852 97(7)
VMC	Gaussian+plane-wave	Yes	-0.031 871 3(1)
DMC	VMC-opt. Gaussian	No	-0.031 916 5(3)
DMC	Gaussian+plane-wave	No	-0.031 917 9(3)
DMC	Gaussian+plane-wave	Yes	-0.031 918 0(3)
DMC	DMC-opt. Gaussian	No	-0.031 919 7(2)

TABLE II: VMC and DMC energies obtained using different orbitals for a ferromagnetic 121-electron HEG of density parameter  $r_s = 30$  a.u.: crystal (Gaussian) orbitals, in which the Gaussian exponent has been optimized within VMC or DMC and “hybrid” (Gaussian+plane-wave) orbitals of the form given in Eq. (1). The coefficients for the first 20 stars of  $\mathbf{G}$  vectors were optimized within VMC in the Gaussian+plane-wave orbitals. In each case the orbitals are centered on the lattice sites of a triangular crystal.

Wigner crystal at  $r_s = 33(1)$  a.u. We find no region of stability for a ferromagnetic fluid, although we find that the paramagnetic fluid is unstable to the ferromagnetic one at  $r_s = 36(1)$  a.u. We find no evidence for the existence of hybrid phases of the type suggested by Falakshahi and Waintal [1], but we cannot, of course, rule out the existence of other types of intermediate phase.

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