Toward Strong Interactions in Circular Quantum Dots: Correlation Induced Inhomogeneity

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Physical properties of the “electron gas” – in which conduction electrons interact via Coulomb forces but the ionic potential is neglected – change dramatically depending on the balance between the strength of the kinetic energy and the Coulomb repulsion. The limiting cases are well understood. For weak interactions (high density), the system behaves as a Fermi liquid, with delocalized electrons. In contrast, in the strongly interacting limit (low density), the electrons localize and become ordered in a Wigner crystal phase. The physics at intermediate densities is phenomenally rich and remains a subject of fundamental research.5–11 Here we study the intermediate density electron gas confined to a circular quantum dot. By using accurate quantum Monte Carlo techniques,6 we show that the correlation induced by increasing interaction strength smoothly causes, first, ring structure and, then, angular modulation, without any signature of a sharp transition or even a crossover in this density regime.

Quantum dots – a nanoscale island containing a puddle of electrons – provide a highly tunable and simple setting to study the effects of large Coulomb interaction. They introduce level quantization and quantum interference in a controlled way. As one can in principle make them in the very low density regime for which correlation effects are strong, it is only a matter of time before such dots are studied experimentally.

We therefore consider a model quantum dot consisting of electrons moving in a two dimensional (2D) plane, with kinetic energy (−∑iq∇iq2), and interacting with each other by long-range Coulomb repulsion (∑ij|riri−1|). All energies are expressed in atomic units, defined by ℏ2/ε = m∗ = 1 (with electronic charge e, effective mass m∗, and dielectric constant ε). The electrons are confined by an external quadratic potential Vext(r) = ω2r2. The spring constant ω controls the density and is varied between 3.0 and 0.0075. The ratio between the strength of the Coulomb interaction and the kinetic energy is usually characterized by the interaction parameter rs ≡ (πn)−1/2, with n the density of electrons. For our confined system in which n(r) varies, we define rs in the same way using the mean density ̄n ≡ ∫ n2(r)dr/N. We have studied this system with up to N = 20 electrons; for the range of ω considered, rs is between 0.4 and 17.7.

In the bulk 2D electron gas, numerical work suggests a direct phase transition between the Fermi liquid and Wigner crystal states in two dimensions with a critical rs ≈ 37. On the other hand, experiments (which include, of course, disorder and residual effects of the ions) on the 2D electron gas observe more complex behavior, including evidence for a metal-insulator transition.

Circular quantum dots have been studied previously using a variety of methods, yielding a surprisingly inconclusive scenario. Many studies have used mean field methods, such as density functional theory or Hartree-Fock. These typically predict charge or spin density wave order even for modest rs (unless the symmetry is restored after the fact – see Ref.11 which raises questions about their accuracy. Exact diagonalization calculations are free of the mean-field approximation but are restricted to small N and rs. Path integral quantum Monte Carlo (PIMC) has also been applied: One study found a crossover from Fermi liquid to “Wigner molecule” at a single rs ≈ 4 – a value significantly smaller than the 2D bulk rs. Another, using different criteria, found a two-stage transition for rs significantly larger than rs. Although PIMC treats interactions accurately, it has its own systematic and statistical problems: it becomes inefficient as T → 0 and it generates a thermal average of states with different L and S quantum numbers, preserving only Sz symmetry.

To avoid these various difficulties and so clarify the scenario, we have carried out a study using the variational and diffusion Monte Carlo techniques, which we used previously to study both circular16,17 and irregular18 dots at rs ~ 2. This method is free of the problems of PIMC but is approximate in that a “fixed-node” error is made. We believe the latter is small for the range of parameters studied here, as described in the “methods” section below. Using well optimized trial wave functions, the statistical and systematic errors of DMC are much smaller than the statistical errors of the published PIMC results; a detailed comparison to PIMC and exact diagonalization results is presented in the supplementary material.

Results for the electron density, n(r), are shown in Fig. 1. There is a dramatic change in n(r) upon increasing interaction strength: For weak interactions [panel (a)], the density is rather homogeneous; the small modulation seen is caused by shell effects in the orbitals of the mean-field problem. In contrast, large rs induces strong radial modulation in n(r) [panel (b)], resulting in the formation of rings. Because the ground state has L = 0 (this is a closed shell configuration in the non-interacting limit), no angular modulation of the density can occur. Interestingly, for rs > 10 the number of rings for each N is the same as that seen in the classical limit (rs → ∞), e.g. three rings for N = 20.

We find that the formation of rings and the increase in their sharpness is very smooth, without any sign of a threshold. This is shown quantitatively in Fig. 1(d) by using the fractional peak height (FPH, defined in Fig. 1(c)) of the outer ring...
FIG. 1: Electron density, $n(r)$, for the ground state of a $N = 20$ circular quantum dot ($L = 0$, $S = 0$). The extrapolated quantum Monte Carlo (QMC) estimator is used. (a) High density: $r_s \approx 0.4$ ($\omega = 3.0$). (b) Low density: $r_s \approx 15$ ($\omega = 0.01$). Note the dramatic change in electron density upon decreasing the density: the electron-electron correlation caused by stronger interactions at low density produces sharp rings. The 3 ring structure agrees with that seen in electron correlation caused purely by Coulomb repulsion (correlation hole), while for like spins the antisymmetry of the wave function plays an important role (exchange hole). For small $r_s$, correlation is weak, so the hole in $g_{\sigma,-\sigma}$ is much smaller than that in $g_{\sigma,\sigma}$. As $r_s$ increases, the correlation hole grows bigger, becoming of the same size as the exchange hole around $r_s \approx 4-5$. Results for the pair density in the circular quantum dot are shown in Fig. 2 for an up-electron fixed on the outer ring. Top: $g_{\uparrow\uparrow}(r_0; r)$ (left) and $g_{\uparrow\downarrow}(r_0; r)$ (right) for the $N = 20$ ground state ($L = 0$, $S = 0$) with $r_s \approx 15$ ($\omega = 0.01$, $r_0 = (57,0)$). Short-range order develops near the fixed electron, indicating “incipient” Wigner localization but not true long-range order. Bottom: Evolution of angular oscillations along the outer ring with $r_s$ and $N$. The top trace shows $0.65 \times g_T$ for $r_s \approx 6$, $N = 20$: though strong radial modulation has already appeared, leading to “ring formation”, there is almost no angular modulation. The middle trace is $g_T$ for $r_s \approx 15$, $N = 20$: clear angular structure is present, though compared to the ring modulation it is weak and short-range. Spin-resolved angular structure is also shown here; note the peculiar bump at $\theta = \pi$. The bottom trace is $g_T$ for $r_s \approx 16$, $N = 6$ ($L = 0$, $S = 0$): for small $N$, angular modulation is clearly stronger. (The y-axis is shifted and scaled for $N = 20$ for clarity.)
and $N$ is illustrated in the bottom panel of Fig. 2. Comparing the top two traces, for $r_s \approx 6$ and $N = 20$, we see that $g_T$ is almost featureless even for an $r_s$ substantially bigger than 1, while short-range oscillations have set in by our larger $r_s$. The weakness of these oscillations suggests that electrons remain more or less delocalized along the ring for $N = 20$ up to the largest $r_s$ studied. An intriguing feature of the spin-resolved pair densities shown is the bump at $\theta = \pi$: $g_{\uparrow\downarrow}$ decreases while $g_{\uparrow\uparrow}$ increases compared to the average value. This feature is present for all $r_s \geq 4$ and tends to grow with increasing interaction strength; we have no explanation for it at this time. Turning now to smaller $N$, we find that two rings are present for $N = 6$ at large $r_s$: the outer one has 5 electrons while the remaining electron is at the center. The lower trace in Fig. 2 shows that individual electrons are better localized for small $N$, a behavior that we find holds quite generally.

We next turn our attention to the addition energy, $\Delta^2 E(N) = E_G(N+1) + E_G(N-1) - 2E_G(N)$. This is experimentally accessible as the spacing between conductance peaks in a Coulomb blockade transport measurement, and is given by the charging energy in the simplest model of a quantum dot.\footnote{We next turn our attention to the addition energy, $\Delta^2 E(N) = E_G(N+1) + E_G(N-1) - 2E_G(N)$. This is experimentally accessible as the spacing between conductance peaks in a Coulomb blockade transport measurement, and is given by the charging energy in the simplest model of a quantum dot.} Our results for $\Delta^2 E(N)$ (normalized by $\omega$) for different interaction strengths are shown in Fig. 3 ($r_s$ for fixed $\phi$ varies slightly with $N$). For $r_s \approx 2$, $\Delta^2 E(N)$ is similar to previous studies\footnote{We next turn our attention to the addition energy, $\Delta^2 E(N) = E_G(N+1) + E_G(N-1) - 2E_G(N)$. This is experimentally accessible as the spacing between conductance peaks in a Coulomb blockade transport measurement, and is given by the charging energy in the simplest model of a quantum dot.} and consistent with weakly interacting physics: non-interacting “shell effects” produce strong peaks for closed shell configurations ($N = 6, 12, 20$). At larger $r_s$, the peaks weaken considerably, reducing mesoscopic fluctuations in $\Delta^2 E$. For similar $r_s$, shell effects are more strongly affected for small $N$, while their remnant persists for large $N$. For comparison, we plot the addition energy in the classical limit obtained from the ground state energies in Ref. 19.\footnote{We next turn our attention to the addition energy, $\Delta^2 E(N) = E_G(N+1) + E_G(N-1) - 2E_G(N)$. This is experimentally accessible as the spacing between conductance peaks in a Coulomb blockade transport measurement, and is given by the charging energy in the simplest model of a quantum dot.} The remarkable similarity to our quantum result for small $N$ at the largest $r_s$ is strong evidence for electron localization in that regime. For large $N$ and $r_s$, however, our results differ from the classical ones in that we still have a weak remnant of shell effects.

Strong correlations can shuffle the energy ordering of different quantum states at fixed $N$. However, for $\omega > 0.01$, the ground state remains consistent with Hund’s first rule (except for $N = 3$). For smaller $\omega$, we see a trend toward violation of this rule, primarily for small $N$ (which are in general more affected by strong correlations), as in the following example. For $N = 9$, the Hund’s rule ground state has $(L, S) = (3/2, 1)$. We find that for $\omega = 0.01$, the highly polarized state $(0, 7/2)$ becomes degenerate with the usual ground state (within our numerical accuracy). (All other $(L, S)$ states lie higher in energy.) Note that $S = 7/2$ requires promotion between non-interacting shells, and so lies much higher in energy in the weakly interacting limit. At large $r_s$, this difference is overcome by the gain in interaction energy. The pair density $g_T$ for both these $N = 9$ states is shown in Fig. 3. The more polarized state is clearly more localized; we find this is generally the case, as expected since exchange keeps the electrons apart.

In conclusion, we have investigated signatures of increasing electron-electron correlation in a parabolic circular quantum dot as the interaction strength increases. The scenario which emerges here is significantly different from the bulk: the cross-over between the delocalized and strongly inhomogeneous regimes appears to be completely smooth – there is no special value of $r_s$ within our accessible range, $r_s < 18$. We stress that this goes beyond the expected finite-size rounding of a phase transition. Our main phenomenological findings are: Radial modulation appears directly in the electron density while observation of angular modulation requires, for all cases studied here, study of the pair density. Radial localization can be substantial for an $r_s$ at which only weak incipient angular localization is visible. Increasing polarization significantly increases the amount of localization. Finally, there is a distinct tendency for small $N$ dots to display stronger modulation.

**Methods** — As a starting point, we use the Kohn-Sham orbitals obtained from a density functional calculation done in
the local density approximation. We then perform a variational Monte Carlo (VMC) calculation using a trial wave function, \( \Psi_T \), which is a linear combination of products of up- and down-spin Slater determinants of the Kohn-Sham orbitals multiplied by a Jastrow factor. The Jastrow factor effectively describes the dynamic correlation between the electrons coming from their mutual repulsion, whereas the near-degeneracy correlation is taken into account by having more than one determinant. We optimize the Jastrow parameters and determinant coefficients by minimizing the variance of the local energy. Finally, we use fixed-node diffusion Monte Carlo (DMC) to project the optimized many-body wave function onto a better approximation of the true ground state, an approximation that has the same nodes as \( \Psi_T \).

The fixed-node DMC energy is an upper bound to the true energy and depends only on the nodes of the trial wave function obtained from VMC. We have calculated the energy \( E(N, L, S) \) of a circular quantum dot for each \( N \) with angular momentum \( L \) and spin \( S \) – all good quantum numbers for our model. \( S_z \) is also a good quantum number, and all our calculations are done for \( S_z = S \); however, \( E \) is independent of \( S_z \). We investigated all possible combinations of \( L \) and \( S \) for the low-lying states, and the combination yielding the lowest DMC energy, \( E_G \), was taken as the ground state for that \( N \).

For expectation values of operators that do not commute with the Hamiltonian – the density or the pair-density, for example – we use an extrapolated estimator \( \langle \hat{F} \rangle \) (denoted \( F_{\text{QMC}} \) for an operator \( F \)) which eliminates the inaccuracy coming from the first-order error in the trial wave function. \( F_{\text{QMC}} \) is defined as \( 2F_{\text{DMC}} - F_{\text{VMC}} \) when \( F_{\text{DMC}} \geq F_{\text{VMC}} \) and as \( F_{\text{DMC}}^2 / F_{\text{VMC}} \) otherwise.

In the multi-determinant expansion of \( \Psi_T \), we keep only Slater determinants formed from the lowest energy Kohn-Sham orbitals for all of the results shown here. Our study is currently limited to \( r_s \leq 18 \) for technical reasons. The most serious is the failure of the VMC optimization as many Slater determinants need to be included for stronger interactions.

For two cases, corresponding to one moderate and one large \( r_s \), we have done preliminary calculations with higher orbitals by including all determinants involving promotion of 2 electrons across a shell gap (10 configuration state functions for \( N = 20 \)). This, then, allows for a change in the nodes of \( \Psi_T \). We find that the change in the energy, as well as the change in density and pair-density, is small, though somewhat larger for greater \( r_s \). Thus we believe that the fixed-node error in our calculations is under control.

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