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Path Integral Simulations of Crystallization of Quantum Confined Electrons

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Path integral Monte Carlo simulation results are presented which allow to characterize the equilibrium properties of a few-electron system in a 2D harmonic trap in rigorous manner. In particular, Wigner crystallization is observed and investigated in a broad range of temperature and confinement strength. An analytical expression for the phase boundary of the Wigner crystal is obtained.

In recent years, there was growing experimental [1, 2] and theoretical [3, 4] interest in *few-electron systems* confined in quantum dots. These systems promise a large variety of applications in optics and transport. An important prerequisite for this is a detailed theoretical understanding of their equilibrium properties which, until now, are only partially known. The limiting behavior at *zero temperature* has been explored in Hartree-Fock calculations [4] and exhibits a transition from a Fermi liquid (FL) to a Wigner crystal (WC) via an intermediate ordered state called "Wigner molecule" (WM). Furthermore, recently a crossover FL → WM was reported for *finite temperatures* too [3]. Up to now, it is not clear if there will be a further transition to WC-like behavior at finite temperature. In this paper, we present clear evidence for crystallization of quantum confined electrons, analytical results for the whole phase boundary and establish the relation to the crystallization in *quasi-classical systems* for which it was originally predicted by Wigner [5].

The theoretical analysis of quantum confined electrons at finite temperature is complicated since strong Coulomb correlations, degeneracy and Fermi statistics have to be accounted for simultaneously excluding e.g. perturbation or mean field approaches. The best candidate to explore this region is the path integral Monte Carlo (PIMC) method. We, therefore, have performed extensive *direct fermionic PIMC simulations* for varying numbers of electrons in a harmonic trap.

We consider a finite 2D system of N electrons with zero total spin in a thermostat of temperature T. The electrons interact via the repulsive Coulomb potential and are con-

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fined in a harmonic trap of strength ω_0. The system is described by the Hamiltonian

$$\hat{H} = \sum_{i=1}^{N} \frac{\hbar^2 \nabla_i^2}{2m_i^*} + \sum_{i=1}^{N} \frac{m_i^* r_i^2 \omega_0^2}{2} + \sum_{i<j}^{N} \frac{e^2}{\epsilon_b |\mathbf{r}_i - \mathbf{r}_j|}, \tag{1}$$

where m^* and ϵ_b are the effective electron mass and background dielectric constant, respectively. The characteristic energy scales in the system, corresponding to the three terms in Eq. (1), are the thermal energy $\langle T \rangle = k_B T$, the zero-point oscillator energy $U_0 = \hbar\omega_0$ and the average Coulomb energy $\langle V \rangle = e^2/\epsilon_b \bar{r}$; and the relevant length scales are the extension of the ground state wave function, $l_0^2 = \hbar/m^*\omega_0$, the effective Bohr radius $a_B \equiv \hbar^2 \epsilon_b / m^2$ and the mean interparticle distance $\bar{r}$ (first maximum of the pair correlation function). Our simulations showed that, throughout the WC phase (at least for $N < 40$), $\bar{r}$ is always close to r_0 given by $e^2/\epsilon_b r_0 = m^* \omega^2 r_0^2/2$. The thermodynamic state of the system is defined by the dimensionless parameters $\lambda \equiv e^2/\epsilon_b l_0 \hbar\omega_0$ and $T_\omega = k_B T/\hbar\omega_0$.

Wigner crystallization is generally known to occur when the Coulomb energy exceeds a certain threshold. Specifically, in a confined 2D system, (i) in the classical limit, WC is independent of the confinement and given by a critical value of the classical coupling parameter, $\Gamma_{cr}^{cl} \equiv \langle V \rangle / \langle T \rangle \approx 127 \ldots 137$ [6] and (ii) in the limit of strong degeneracy, WC appears when the Brueckner parameter $r_s \equiv \bar{r}/a_B$ exceeds a critical value which, in an *infinite 2D system*, was found to be $r_s^{cr} \approx 37$ [7]. Below, we present the first critical data for *finite quantum systems* and demonstrate how to connect both limits. To follow the phase boundary of the Wigner crystal, calculations in a broad range of parameter values λ, T_ω are necessary while keeping the particle number constant. Below we show results for $N = 10$ and $N = 11$. We performed path-integral Monte Carlo simulations which allow to treat quantum, spin and Coulomb interaction effects rigorously (for numerical details, see Ref. [8]).

Figure 1a shows a snapshot of the density distribution of 11 electrons in the WC phase. We observe two shells containing 8 and 3 electrons, respectively (in the case of

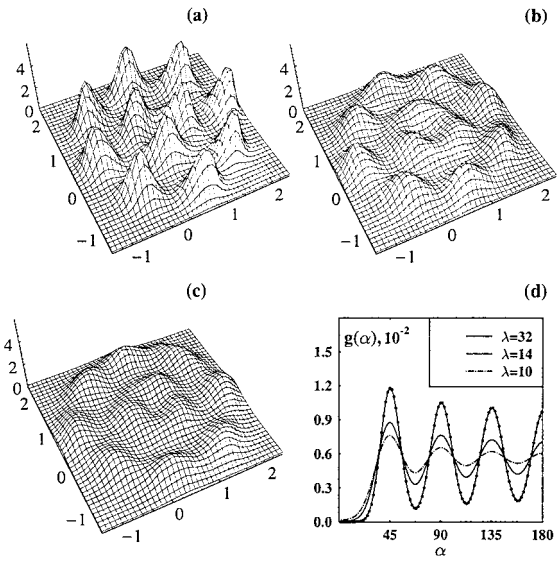

Fig. 1. Snapshots of the density distribution of $N = 11$ electrons in a harmonic trap for three values of the confinement strength λ and $T_\omega = 0.04 = \text{const}$: a) in the WC phase ($\lambda = 32$), b) near the melting point ($\lambda = 14$), c) in the WM phase ($\lambda = 10$). Coordinates are scaled in units of r_0. d) Angular pair distribution functions of electrons on the outer shell for the cases (a) to (c). α denotes the angular distance between two of the outer electrons with respect to the trap center

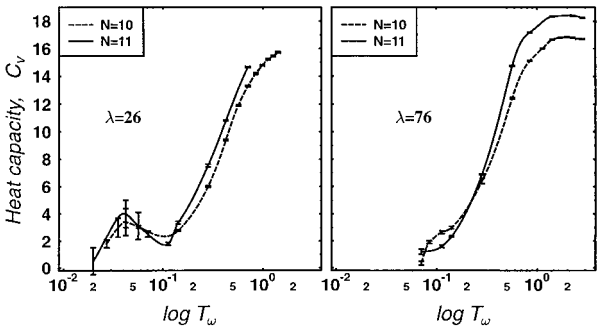

Fig. 2. Heat capacity for $N = 10$ (dashed line) and $N = 11$ (full line) electrons in the vicinity of the Wigner crystallization transition for $\lambda = 26$ (left) and $\lambda = 76$ (right). C_v in units of k_B

10 electrons, there are 2 electrons on the inner shell). Melting of the WC occurs if electrons start to hop from one shell to another which may be caused by an increase of temperature or density. Figures 1b and c show melting by compression (decrease of λ) at constant T_ω, where (b) is near the transition point and (c) in the WM phase. Evidently, from (a) to (c) the individual peaks broaden until their width becomes comparable with the inter-particle distance, and we observe increased tunneling between the two shells which eventually leads to melting of the crystal. Notice that across the phase boundary, the electron probability density (Figs. 1a to c) and the pair distribution functions (Fig. 1d) undergo continuous changes and are, therefore, not suitable to localize the phase transition. Similar, most macroscopic quantities, such as the total energy, change smoothly. A more sensitive quantity is the specific heat which is shown in Fig. 2. We have found that even more sensitive to the WC transition is the magnitude of the radial fluctuations $\langle u_r^2 \rangle = N^{-1} \sum_{i=1}^{N} \{\langle r_i^2 \rangle - \langle r_i \rangle^2\}$ (r_i is the distance of particle i from the center of the trap), which exhibits well pronounced jumps along the whole phase boundary [8] and thus is a suitable criterion for the transition.

We have performed extensive simulations in order to obtain the phase boundary of the Wigner crystal in the whole λ–T_ω plane, the results are shown by the crosses in Fig. 3. In particular, we found that the zero temperature asymptotics is given by $\lambda_{cr}(T_\omega = 0) \approx 15 \pm 1$, for $N = 11$ (and 28 ± 1, for $N = 10$). This corresponds to a critical value of $r_s \approx 46 \pm 4$ (107 ± 5). On the other hand, for sufficiently high temperatures,

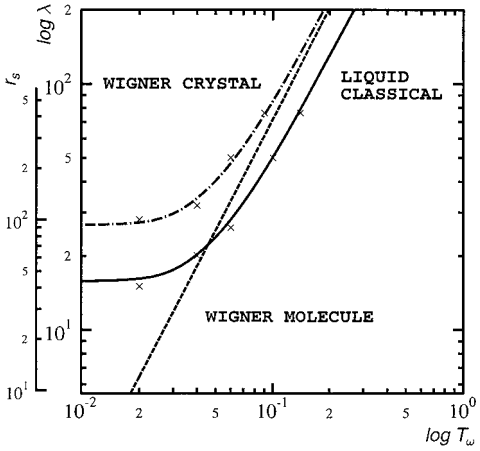

Fig. 3. Phase diagram for the Wigner crystal with $N = 11$ electrons in a harmonic trap. Crosses are the simulation points. Dashed line: phase boundary of a *macroscopic classical 2D WC*, $\Gamma_{cr} = 137$. Full line: phase boundary for the WC computed from Eq. (4) using $\gamma = 10$. For comparison, the dash-dotted line shows the analogous result for the phase boundary in the case of $N = 10$ particles

$T_\omega > 0.1$, we observe that the phase boundary is well approximated by the classical coupling parameter $\Gamma^{\rm cl}$ using critical values of $\Gamma^{\rm cl}_{\rm cr} \approx 143 \pm 9$, for $N = 10$, and 100 ± 5, for $N = 11$. Our analysis has revealed that there exists a simple analytical expression for the whole phase boundary. To derive it, we consider the mean kinetic energy of particles in a harmonic trap at temperature T,

$$\langle K \rangle (T, \omega_0) = \frac{\hbar\omega_0}{2\gamma} \coth \frac{\hbar\omega_0}{2\gamma k_{\rm B} T} , \tag{2}$$

where γ is a free parameter (the familiar result for non-interacting particles corresponds to $\gamma = 1$). This suggests to define a generalized coupling parameter (order parameter), in analogy to $\Gamma^{\rm cl}$, as

$$\tilde{\Gamma} \equiv \frac{\langle V \rangle}{\langle K \rangle} = \frac{e^2}{\epsilon_{\rm b} \bar{r}} \frac{2\gamma}{\hbar\omega_0} \tanh \frac{\hbar\omega_0}{2\gamma k_{\rm B} T} . \tag{3}$$

The phase boundary of the WC obtained from Eq. (3) is given by

$$\tilde{\lambda}_{\rm cr}(T, \omega_0; \tilde{\Gamma}_{\rm cr}, \gamma) = \frac{1}{2} \left[\frac{\tilde{\Gamma}_{\rm cr}}{\gamma} \coth \frac{\hbar\omega_0}{2\gamma k_{\rm B} T} \right]^{3/2} , \tag{4}$$

and depends on two parameters, $\tilde{\Gamma}_{\rm cr}$ and γ. It is obvious to choose $\tilde{\Gamma}_{\rm cr} = \Gamma^{\rm cl}_{\rm cr}$ to assure the correct high-temperature limit of formula (4). Further, we determine γ in order to fit the zero temperature simulation data. We found that a value $\gamma = 10$ works for both, $N = 10$ and $N = 11$ particles. Most interestingly, this choice of γ fits the *whole phase boundary* data *without any additional parameters* as can be seen from Fig. 3 (cf. the full and dash-dotted lines for $N = 11$ and $N = 10$, respectively).

The reason for this surprising agreement is that our definition of γ effectively rescales the oscillator energy, $\hbar\omega_0 \to \hbar\omega_0/\gamma$, cf. Eq. (2), which is of the order of the energy needed for inter-shell tunneling – the dominant melting mechanism at low temperatures. To generalize our result to other particle numbers and situations remains an interesting question for future investigations. We expect that Wigner crystallization should be observable in semiconductor quantum dots with sufficiently weak confinement potential (below 1 meV) at helium temperatures.

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