Wigner Crystallization in mesoscopic 2D electron systems

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Wigner crystallization of electrons in a 2D quantum dot is reported. It proceeds in two stages: I) via radial ordering of electrons on shells and II) freezing of the inter-shell rotation. The phase boundary of the crystal is computed in the whole temperature–density plane, and the influence of quantum effects and of the particle number is analyzed.

In recent years there is growing interest in finite quantum systems at high density or/and low temperature. In particular, the behavior of a small number of electrons in quantum dots is actively investigated both experimentally and theoretically. The limiting behavior of two-dimensional (2D) finite quantum systems at zero temperature has been studied by unrestricted Hartree-Fock calculations which revealed a transition from a Fermi liquid to an ordered state called “Wigner molecule”. The same crossover at finite temperature has been recently demonstrated by fermionic path integral Monte Carlo. It has to be expected that further increase of correlations (increase of the Brueckner parameter $r_s$) will lead to a still higher ordered quantum state resembling the Wigner crystal (WC).

On the other hand, for finite classical systems, Monte Carlo simulations have shown evidence of crystallization for sufficiently large values of the coupling parameter $\Gamma$. These classical clusters consist of well separated shells, and melting proceeds in two stages: first, orientational disordering of shells takes place—neighbouring shells may rotate relative to each other while retaining their internal order. Further growth of thermal fluctuations leads to shell broadening and overlap—radial melting. The temperature of radial melting $T_r$ may be many orders of magnitude higher than the orientational melting temperature $T_m$. Large clusters with $N > 100$ have a regular triangular lattice structure and exhibit only radial melting.

Now the question arises, how does the behavior of finite electron clusters change at low temperature, i.e. in the quantum regime? In this Letter we demonstrate that, indeed, Wigner crystallization in 2D quantum electron clusters exists and that it is accompanied by two distinct—radial and orientational—ordering transitions too. However, in contrast to classical clusters, we observe a new melting scenario which is caused by quantum fluctuations and exists even at zero temperature (“cold” melting). We present a detailed analysis of the two-stage quantum melting process and provide numerical data for the phase boundaries of both crystal phases, for particle numbers in the range $N = 10 \ldots 20$.

Model and characteristic parameters. The theoretical analysis of quantum confined electrons at finite temperature requires the simultaneous account of strong correlations and quantum effects which excludes e.g. perturbation or mean field methods. We, therefore, use a path integral Monte Carlo (PIMC) approach. We consider a finite unpolarized 2D system of $N$ electrons at temperature $T$. The electrons interact via the repulsive Coulomb potential and are confined in a harmonic trap of strength $\omega_0$. The system is described by the hamiltonian

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

where $m^*$ and $\epsilon_b$ are the effective electron mass and background dielectric constant, respectively. We use the following length and energy scales: $r_0$, given by $e^2/\epsilon_b r_0 = m^*\omega_0^2 r_0^2/2$, and $E_c$ - the average Coulomb energy, $E_c = e^2/\epsilon_b r_0$. After the scaling transformations $\{r \rightarrow r/r_0, E \rightarrow E/E_c\}$ the hamiltonian takes the form

$$\hat{H} = \frac{n^2}{2} \sum_{i=1}^{N} \nabla_i^2 + \sum_{i=1}^{N} r_i^2 + \sum_{i<j}^{N} \frac{1}{|r_i - r_j|},$$

where $n$ is the dimensionless density, $n = \sqrt{2} r_0^2 / \hbar^2 = (a_B^2 r_0)_{1/2} \approx r_s^{-1/2}$, $a_B^2$ is the effective Bohr radius, and $\hbar^2 = \hbar/m^* \omega_0$ is the extension of the ground state wave function of noninteracting trapped electrons. (In the crystal phase, $r_0$ is very close to the mean interparticle distance $\bar{r}$). The dimensionless temperature is given by $T = k_B T / E_c$.

To obtain the configuration and thermodynamic properties of clusters of $N$ electrons described by the hamiltonian, we performed fermionic PIMC simulations using a standard bisection algorithm. The number of time-slices $M$ has been varied with $n$ and $T$ according to $M = \ln(T)$, where $l$ was typically in the range of $1 \ldots 10$ to achieve an accuracy better than 5% for the quantities, see below. To obtain the phase boundary of the Wigner crystal, calculations in a broad range of parameter values $\{n, T, N\}$ were performed. For each set of parameters, several independent Monte Carlo runs consisting of approximately $10^6$ steps were carried out.

Structure of electron clusters. The simulations yield the spatial electron configurations in the trap, examples of which are shown in Figs. 1 and 2. One clearly sees the formation of shells. Our analysis revealed the same shell structures as reported for the corresponding classical systems. Number of shells and shell occupation depend on $N$, some examples for $N = 10 \ldots 20$ are given in Tab. 1. Clusters, in which the particle number
on the outer shells are multiples of those on the inner shells have the highest symmetry which leads to particular properties. Examples of these magic numbers are $N = 10, 12, 16, 19$, cf. Tab. I. Let us now discuss the influence of quantum effects on the clusters. In contrast to classical systems, where the electrons are point-like particles, in our case, the wave function of each electron has a finite width and may be highly anisotropic, which is typical for low temperature, as is most clearly seen in Fig. 1b. This peculiar shape results from a superposition of N-body correlations, quantum effects and the confinement potential. Varying the density and temperature, the shape changes in a very broad range, which can lead to qualitative transitions of the cluster, including cold quantum melting, as will be shown below.

Melting and phase transitions. Wigner crystallization is known to occur when the ratio of the Coulomb energy to the kinetic energy, $E_c/E_{\text{kin}}$, exceeds a certain threshold. In the quantum limit, this ratio is just the Brueckner parameter $r_s$. On the other hand, in the classical limit this ratio goes over to the classical coupling parameter $\Gamma = E_c/k_B T$. In our units ($\bar{r} \approx r_0$),

$$\Gamma = \frac{\bar{r} \approx r_0}{T} \approx \frac{1}{T}. \quad (3)$$

Let us first consider the classical case. At sufficiently low $T$ (high $\Gamma$), a classical cluster is in the orientationally and radially ordered crystal phase. Our simulations revealed that in a finite system, (in the classical part of the phase space), both orientational and radial melting are determined by characteristic values of $\Gamma$ which, however, strongly depend on the particle number $N$. We find that orientational melting occurs at temperatures which vary over many orders of magnitude, $T_r \approx 3 \times 10^{-3} \ldots 10^{-12}$ and are very sensitive to the shell configuration, as observed in Ref. [8]. Higher temperatures (lower $\Gamma$) are found for magic clusters, cf. Tab. I. The magic cluster with $N = 19$ has unusual stability against inter-shell rotation, as the ratio of particle numbers on the three shells is optimal for the formation of a triangular lattice. In contrast, nonmagic clusters have much lower orientational melting temperatures (see Tab. I), which is particularly striking for $N = 20$.

The situation is completely different at radial melting. Here, the critical temperatures for inter-shell particle jumps (radial disordering) for magic clusters have been found to be approximately 2 times smaller than those for nonmagic clusters (Tab. I).

We now investigate the phenomena of quantum orientational melting (OM). As an illustration, consider the cluster with $N = 12$ particles. Fig. 1 shows three snapshots of particle configurations at low temperature where the density increases from (a) to (c), and (b) corresponds to the critical density. One of the characteristic features of quantum OM is a high anisotropy of the electron wave function. Its main peak is along $\phi \approx \pi/4$, and we may call the orientation of the electron wave function $\phi$ the position of the electron. At quantum OM, the probability density of each inner shell electron splits in two maxima which are due to electron transitions between two closely lying energy levels.

At still higher density, Fig. 1(c), the electron wave function spreads further, splitting into three maxima, and it starts to spread in radial direction. Obviously, OM is accompanied by intensification of angular distance fluctuations, cf. Fig. 1(d). The spread of the electron wave function smears out the energy barrier for inter-shell rotation which, in turn, reduces the melting temperature. In the limit $T \to 0$ this leads to so-called “cold” orientational melting.

To allocate the melting temperatures and densities accurately, we examine the relative mean angular distance fluctuations of particles from different shells

$$u_\phi \equiv \sqrt{\langle \delta \phi^2 \rangle} = \frac{2}{m_{s_1} m_{s_2}} \sum_i \sum_j \left[ \frac{\langle \phi_j - \phi_i \rangle^2}{\langle \phi_j - \phi_i \rangle^2} \right] - 1, \quad (4)$$

where $\phi_i$ and $\phi_j$ are the angular positions of particles on shells $s_1$ and $s_2$, respectively, and $m_{s_1}$, $m_{s_2}$ are the total number of particles on the shells. We further consider the magnitude of the relative distance fluctuations

$$u_r \equiv \sqrt{\langle \delta r^2 \rangle} = \frac{2}{N(N-1)} \sum_{i<j} \sqrt{\frac{r_{ij}^2}{(r_{ij})^2}} - 1, \quad (5)$$

We measured the $s_1$ dependence of the fluctuations $u_\phi$ and $u_r$ along a radial line (Fig. 1l). The region of phase transition, which corresponds to the classical region of the phase space, where Wigner crystallization...
In the vicinity of the orientational (OM) and radial melting (RM) for $N = 12$ and $N = 19$ versus density. Upper three figures show snapshots of the magic cluster with $N = 19$ in the three phases (left $n = 0.025$, middle $n = 0.06$, right $n = 0.14$). $T = 5.0 \times 10^{-4}$. Shown error bars are typical for all curves.

Fig. 2 shows the $n$-dependence of the fluctuations $u_\phi$ and $u_r$, at a fixed temperature $T = 5 \cdot 10^{-4}$, for the magic clusters $N = 12$ and $N = 19$. The first jump of the fluctuations at comparatively low critical densities $n_\phi^*$ corresponds to quantum OM. At densities $n \geq n_\phi^*$, the angle between particles from neighbouring shells can take arbitrary values. There is a significant difference in the magnitude of the melting densities for the clusters under consideration. As in the classical case, the highest stability (highest value $n_\phi^*$) is found for the magic clusters, with the maximum value found for $N = 19$. In particular, we obtained the densities of “cold” orientational melting by extrapolation of our data to zero temperature. The corresponding values for the density and Brueckner parameter $r_(s)^(\phi)$ are listed in Tab. 1.

Let us now proceed in Fig. 2 to higher densities. We observe a clear second jump of the fluctuations $u_\phi$ and $u_r$, which is related to total melting of the WC. It is instructive to compare the state of the cluster before and after this jump, see middle and right snapshots in Fig. 2, respectively. Evidently, when the density is increased, the peaks of the wave functions broaden in radial (and angular) direction until their width becomes comparable with the inter-shell spacing. As a result, the probability of inter-shell transfer of electrons grows rapidly, causing a sudden increase of the radial fluctuations and the onset of radial melting. Our simulations revealed that the jumps of $u_\phi$ and $u_r$ are clearly visible along the whole WC phase boundary. In the limit of zero temperature, we observe “cold” radial melting, the corresponding values for the density and $r_(s)^(r)$ are presented in Tab. 1.

Phase boundary of the mesoscopic Wigner crystal. The obtained values of the melting temperatures and densities are now used to draw the phase boundaries of the radially and fully ordered states of mesoscopic clusters. The results are summarized in Fig. 3 and Tabs. 3, 4 for various particle numbers. Consider first the line of radial melting “RM”. At low densities, $n < 0.03$, crystallization occurs at critical values of $\Gamma = 1/T^*_\phi$, see the data in Tab. 3. We found that, for magic (non-magic) clusters, $\Gamma$ is above (below) the 2D bulk value $\Gamma^\infty = 137$, e.g. 6. This shows that non-magic clusters are more stable against radial disordering. The same tendency is observed for “cold” melting, where the critical values of $r_s$ are systematically lower for non-magic clusters. Interestingly, for all comparatively small clusters with $10 \leq N \leq 20$, the critical $r_s$, (Tab. 3), exceeds the known value of an infinite 2D system $r_s^\infty \approx 37$ 12. This systematic deviation from the critical data of a macroscopic system points to the existence of a different scenario of radial melting typical for finite systems. For example, for clusters with $N = 10$ (19), when approaching the phase boundary, we observe increasingly frequent transitions between the shell configurations $\{2,8\} \leftrightarrow \{3,7\}$ $\{1,6,12\} \leftrightarrow \{1,7,11\}$). The explanation is the lowering of the potential barrier for inter-shell transitions of individual electrons between these two energetically rather close configurations.

A particularly interesting feature which is missing in infinite systems is the existence of the second transition characterized by freezing of the inter-shell rotation, cf. areas bounded by the lines “OM” in Fig. 3. This highly ordered state is confined to the region of low density and low temperature. Its phase boundary is highly sensitive to the angular symmetry of the cluster. The most symmetric clusters are the magic ones which are essentially more stable against orientational melting which is seen in their relatively low values of $\Gamma_\phi$ and $r_(s)^(\phi)$, cf. Tabs. 3 and 4. The critical values for the non-magic clusters are several orders of magnitude larger, confining their orientationally ordered state to much lower densities and temperatures. Finally, the last two columns of Tab. 4 contain the maximum temperatures at which the orientationally and radially ordered crystal phases may exist.

In summary, we have presented a detailed PIMC-analysis of Wigner crystallization of finite electron systems confined in a 2D harmonic trap including the critical data in the whole density-temperature plane. We have shown that there exist two phases characterized by radial and radial plus angular ordering, respectively, which is essentially different from macroscopic systems.
The phase boundaries have been found to be very sensitive to the electron number and to the shell symmetry. In contrast to classical clusters, we observed “cold” orientational and radial melting which is governed by the spread of the electron wave functions in angular and radial direction. The predictions of our model calculations are expected to be relevant in particular for electrons in external fields. Furthermore, they lead us to expect Wigner crystallization also in small 2D islands of electrons [holes] in semiconductor heterostructures: for example, in GaAs/AlGaAs systems, crystallization is predicted for carrier densities below approximately $10^8\text{cm}^{-2} \left[ (10^9 \ldots 10^{10})\text{cm}^{-2} \right]$ and for temperatures below $1.6K \ldots 5.5K$, for confinement potentials of $3meV \ldots 10meV$.

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TABLE I: Classical Wigner crystal: critical temperature $T^*$ and coupling parameter $\Gamma^*$ corresponding to the orientational (o) and radial (r) melting for clusters of size $N$ in the classical limit ($n \to 0$). Second column contains the shell occupancy, starting from the inner shell. Non-magic clusters are typed italic.

| $N$ | $T^*_o$ | $\Gamma^*_o$ | $T^*_r$ | $\Gamma^*_r$ |
|-----|---------|--------------|---------|-------------|
| 10  | 2.8     | $1.5 \times 10^{-5}$ | $6.7 \times 10^4$ | $5.9 \times 10^{-3}$ | 169 |
| 11  | 3.8     | $2.6 \times 10^{-7}$ | $3.85 \times 10^6$ | $1.25 \times 10^{-2}$ | 80 |
| 12  | 3.9     | $8.0 \times 10^{-4}$ | 1250    | $6.0 \times 10^{-3}$ | 166 |
| 16  | 1.5, 10 | $1.7 \times 10^{-3}$ | 590     | $6.2 \times 10^{-3}$ | 161 |
| 19  | 1.6, 12 | $3.0 \times 10^{-3}$ | 330     | $6.5 \times 10^{-3}$ | 154 |
| 20  | 1.7, 12 | $2.9 \times 10^{-12}$ | $3.4 \times 10^{11}$ | $1.2 \times 10^{-2}$ | 83 |

TABLE II: Quantum Wigner crystal: critical density and Brueckner parameter $r_s$ for orientational (o) and radial (r) melting at zero temperature. $\tilde{T}^*_o(r)$ is the highest possible melting temperature, $\tilde{T} \equiv n^* T$. The critical parameters $n_o^*$, $r_s^{(o)}$, $T^*_o$, $T^*_r$ for $N = 11$ and 20 are estimates [4].

| $N$ | $n_o^*$ | $r_s^{(o)}$ | $n_s^*$ | $r_s^{(r)}$ | $T^*_o$ | $T^*_r$ |
|-----|---------|-------------|---------|-------------|---------|---------|
| 10  | $4.5 \times 10^{-6}$ | $4.9 \times 10^{-3}$ | 0.120   | 69          | $5.0 \times 10^{-8}$ | $2.2 \times 10^{-4}$ |
| 11  | $5 \times 10^{-5}$  | $4 \times 10^{-6}$  | 0.148   | 45          | $7 \times 10^{-11}$ | $9.4 \times 10^{-4}$ |
| 12  | $1.4 \times 10^{-2}$ | 5102         | 0.123   | 66          | $5.0 \times 10^{-6}$ | $2.4 \times 10^{-4}$ |
| 19  | $5.0 \times 10^{-2}$ | 400          | 0.125   | 64          | $6.0 \times 10^{-5}$ | $2.5 \times 10^{-4}$ |
| 20  | $1.7 \times 10^{-6}$ | $3 \times 10^{11}$ | 0.140   | 51          | $3 \times 10^{-18}$ | $7.5 \times 10^{-4}$ |