Quantum Phase Transitions to Charge Order and Wigner Crystal Under Interplay of Lattice Commensurability and Long-Range Coulomb Interaction

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Relationship among Wigner crystal, charge order and Mott insulator is studied by the path-integral renormalization group method for two-dimensional lattices with long-range Coulomb interaction. In contrast to Hartree-Fock results, the solid stability drastically increases with lattice commensurability. The transition to liquid occurs at the electron gas parameter \( r_s \sim 2 \) for the filling \( n = 1/2 \) showing large reduction from \( r_s \sim 35 \) in the continuum limit. Correct account of quantum fluctuations are crucial to understand charge-order stability generally observed only at simple fractional fillings and nature of quantum liquids away from them.

Coulomb interaction between electrons drives various types of electron crystallization ranging from Wigner lattice, charge order including stripes to Mott insulator. The Wigner transition in the continuum space was studied on two-dimensional (2D) electron systems by quantum Monte Carlo calculations and the transition point was estimated to be \( r_s \sim 35 \) at zero temperature \( \square \), where \( r_s = r_0/a_B \) with the Wigner-Seitz radius \( r_0 = 1/\sqrt{\pi \varepsilon} \), the Bohr radius \( a_B = 4\pi e^2/m^*\varepsilon^2 \) and the electron density \( n_e \). This 2D Wigner transition in quantum region was experimentally realized in GaAs heterostructure with extremely high mobility and the transition at \( r_s \sim 35 \) was in good agreement with the theoretical prediction \( \square \).

The most dramatic effect of atomic periodic potential in crystal not contained in the electron gas is apparently the band formation in electronic spectra due to Bloch theorem. In this letter, we stress another crucial concept, lattice commensurability, generates different dramatic effects when the Coulomb interaction is present. In correlated electron systems in solids as in transition metal compounds \( \square \) and organic systems \( \square \), charge ordering phenomena including stripe-type are common when the electron filling \( n \) satisfies simple fractional number such as \( 1/2 \) and \( 1/3 \). These phenomena have attracted much interest in relation to the mechanisms of high-Tc superconductivity in the cuprates and colossal magnetoresistance in the manganites. However, in those systems, as we discuss later, the effective value of \( r_s \) is usually estimated to be rather small in the range \( r_s < 10 \), though we have some uncertainty in the evaluation of the effective mass \( m^* \) and the dielectric constant \( \varepsilon \). At such low values of \( r_s \), we normally expect the quantum melting of electrons in terms of the above electron gas picture. When the electron filling deviates from a simple fractional number, such charge orders indeed usually melts to metals or the charge periodicity is pinned at the simple fillings with added carriers localized by disorder. It implies that a mechanism of stabilizing the solid only at simple commensurate filling works very efficiently.

Another known result is that the Mott insulator at integer fillings can be stabilized even at lower effective \( r_s \). The Mott insulator can be viewed as the extreme limit of strong commensurability. A common feature of the charge order and the Mott insulator is that they are charge incompressible state resulted from the short-ranged part of the Coulomb interaction.

In this letter, we show that the mechanism for such common stabilization of the solid phase only at simple fractional fillings is not clear in the Hartree-Fock (HF) calculations, whereas the stabilization and melting can indeed be understood only when the quantum fluctuation effects are seriously considered.

To understand the basic mechanism, we employ a Hamiltonian given as

\[
\hat{H} = \hat{H}_0 + \hat{H}_I, \\
\hat{H}_0 = - \sum_{<ij>,\sigma} t_{ij}(c_{i\sigma}^\dagger c_{j\sigma} + h.c.), \\
\hat{H}_I = U \sum_i n_i^\uparrow n_i^\downarrow + \frac{1}{2} \sum_{i\neq j} V_{ij} n_i n_j, \\
\]

where the notations follow the standard one in the Hubbard type models. We take the long-ranged Coulomb term as \( V_{ij} = V | r_i - r_j | \), where we ignore possible screening effects arising from electrons in other bands. The jellium model is employed by assuming a uniform positive charge as a background while the effect of ionic periodic potential is represented by the lattice with single-band electrons located near the Fermi level. The onsite interaction \( U \) measured from \( V \) may depend on the detailed structure of atomic orbitals. Here we take \( U/t = 4 \) throughout this paper for simplicity. This ratio may be more or less the lower bound for the real situations. In any case our results in this letter for \( n \leq 1/2 \) do not sensitively depend on this ratio. We study spinless fermion models in addition to spin-1/2 electrons. For spinless fermions, the \( U \) term does not exist. This corresponds to the fully polarized ferromagnetic case. We impose the periodic boundary condition in the numerical studies. The Coulomb interactions between images across the periodic boundaries are taken by the Ewald summation.

Since our purpose in this letter is to understand a generic and universal feature of the interplay between quantum fluctuations and commensurability, we restrict our study to the square lattice with the anisotropy of lattice constants \( a_x \) and \( a_y \) as well as \( t_x \) and \( t_y \) taken as simple as possible to avoid a possible complexity which may intervene the clarification of the essence. The transfer is limited to the nearest-neighbor pairs in \( x \) and \( y \).
directions denoted by $t_x$ and $t_y$ as in Fig. 1. We choose the anisotropy $d = a_y/a_x$ to make the charge ordering on a right triangular lattice possible. By this choice, the charge order stability is optimized and can be estimated on the same ground as the continuum limit. To see a systematic dependence on $r_s$, we control $t_x$ and $t_y$ by a single parameter of the effective mass $m^*$ as $t_2 = \hbar^2/2m^*a_x^2$ with $z = x$ or $y$, which is identified in the two-body correlation function for fixed electron number $N_e$ as in Fig. 4. The transition point is estimated as $r_s \rightarrow 0$, with $r_s$ fixed, the above Hamiltonian is reduced to that of conventional electron gas.

We first show results of the standard HF calculation for spinless fermions. For the fillings $n < 1/2$, the first-order transitions are clearly visible from the jump of $\langle d\hat{H}/dr_s \rangle$ e.g. in Fig. 3 at $r_{sc} = 3.45 \pm 0.05$ for $n = 1/32$ on 32 by 32 lattice. Note that the jump of $\langle d\hat{H}/dr_s \rangle$ indicates the level crossing in the ground state as a function of the control parameter $r_s$ and gives an evidence for the first-order transition. The right ordinate is the scale for the second Hamiltonian $\hat{H}_R$ defined by $\Xi = \partial (\hat{H}_R)/\partial r_s = (d/\pi n)\partial \hat{H}/\partial r_s$. From Fig. 3, the electron crystallization is indeed identified in the two-body correlation function defined by $C(r_i) = \frac{1}{N} \sum_{r_j} \langle n_{r_j} n_{r_i} \rangle$. For fixed electron number $N_e$, the transition point is extrapolated to the continuum limit $n \rightarrow 0$. These results are further extrapolated to the thermodynamic limit $N_e \rightarrow \infty$ as in Fig. 4. This indicates that the system size dependence is small and the result at $N_e = 8$ is already a good estimate of the thermodynamic limit. At $n = 1/2$, the transition is of continuous type with the absence of a jump in $\langle d\hat{H}/dr_s \rangle$ while the peak value of the Fourier transform of two-body charge correlation diverges as a Bragg peak with increasing system size for $r_s \geq r_{sc}$. Then the transition point is estimated as $r_{sc} = 0.9 \pm 0.1$ after finite-size scaling.
To understand effects of spin degrees of freedom, spin-1/2 electrons are also analyzed at \( n = 1/4 \) and 1/2. The transitions appear to be of first order at \( r_{sc} = 2.05 \pm 0.05 \) and continuous at \( r_{sc} = 1.35 \pm 0.15 \), respectively. The orders of transitions are the same as the spinless case and the difference of \( r_{sc} \) between these two cases shrinks with decreasing filling. We discuss this tendency later. We conclude that the unrestricted HF approximation predicts the crystallization of 2D electrons ranging from \( r_{sc} \sim 1 \) at \( n = 1/2 \) to \( r_{sc} \sim 3.69 \) in the continuum limit. We note that the filling dependence is rather small.

To understand how quantum and many-body fluctuations modifies the HF results, we have calculated the ground state of this system by applying recently developed PIRG method [3]. This algorithm allows us to start from and improve the above unrestricted HF solutions and reach the correct ground state by taking account quantum fluctuations in a controlled fashion. By following the path integral formalism, the convergence to the ground state is obtained after repeated operations of \( \exp[-\tau \hat{H}] \) to Slater determinants with small \( \tau \) and by the break-up of the kinetic and Coulomb energy terms. The long-ranged Coulomb term is rewritten by using the Stratonovich-Hubbard transformation which allows this term to operate to the Slater determinants, though it increases the number of Slater determinants in the linear combination of the wave function [3]. With increasing the dimension of truncated Hilbert space in a nonorthogonal basis optimized by this path integral operation, an extrapolation method using the variance \( \Delta E = (\langle \hat{H}^2 \rangle - \langle \hat{H} \rangle^2)/\langle \hat{H} \rangle^2 \), is employed to reach the true ground state. For the extrapolation, we took the number of nonorthogonal Slater determinants up to 256. We show results when the extrapolation linearly converges well to those of the full Hilbert space.

We took lattice sizes up to 144 sites (12 \( \times \) 12 lattice) with electrons up to 72 at quarter filling \( n = 1/2 \) for both spinless and spin-1/2 electron systems (Then for 144 sites of spin-1/2 system, we took 36 up and down spin electrons each). For fillings 1/8 and 1/18, we show only on the spinless system for 64 sites and 144 sites, respectively with 8 electrons. We restricted to the spinless case for these low fillings since the difference of \( r_{sc} \) from the system with spins is estimated to be small. This is inferred from the comparisons of the PIRG with the HF results at \( n = 1/2 \) together with the HF results at lower filling with spins. In fact, the spin effects appear mainly through the exchange process. The exchange is scaled by \( t^2 \) where \( t \) is the effective transfer between neighboring electrons. At lowering fillings, it forms lower-energy hierarchy as compared with that of the charge order, \( V \). Therefore, though spin is important in determining the magnetic order, this small energy scale hardly changes the solid-liquid transition point for \( n \leq 1/2 \). From comparisons for different sizes, the system-size dependence of the transition point appears to be small at the low fillings because the transition becomes first order. Therefore, though our systems at the low fillings are rather small, the results are expected to be close to the thermodynamic results.

![FIG. 5: PIRG result for \( dH/dr_s \) vs. \( r_s \) for 8 spinless electrons on 12 by 12 lattices.](image)

![FIG. 6: PIRG result for two-body correlation function for 8 spinless electrons on 12 by 12 lattice.](image)

We first show spinless cases. Fig. 3 shows a typical example showing the first-order transition in the PIRG calculation at \( n = 1/18 \) for 8 fermions on 12 by 12 lattice. The behavior of jump is similar to the HF results with a clear change of the two-body correlation function as in Fig. 4. However, this first-order crystallization takes place at substantially higher \( r_s \) than the Hartree-Fock result. The transition at \( n = 1/8 \) is also first order while that is of continuous type again at \( n = 1/2 \). The transition point is estimated to be \( r_{sc} = 1.75 \pm 0.25, 13.5 \pm 0.5 \) and 24.5 \( \pm 0.5 \) for \( n = 1/2, 1/8 \) and 1/18, respectively. We have also studied spin-1/2 electrons at \( n = 1/2 \) and obtained a continuous transition at \( r_{sc} = 2.0 \pm 1.0 \), which is comparable to the spinless case.
In Fig. 7, the phase boundary of the solid and liquid phases, \( r_{sc} \), are shown and the HF and PIRG results are compared for \( n = 1/l \) with integer \( l \). At simple fractional filling such as \( n = 1/2 \), \( r_{sc} \) by the HF approximation is relatively good. However, it deviates from the PIRG results rapidly with increasing \( l \). The ratio of these two estimates increases from \( \sim 1.5 - 2 \) at \( n = 1/2 \) to \( \sim 10 \) in the continuum limit. In terms of the electron density, this difference means from 3-4 to 100, since the density is scaled by \( r_s^2 \). From the PIRG results, we can understand why charge orders are commonly observed in organic and transition metal compounds at simple fractional fillings as 1/2 and 1/3 while it is pinned or melts away from such fillings because \( r_{sc} \) increases dramatically with increasing denominator of the irreducible fraction while the parameters of many compounds may lie in this range of variation. For example, when we assume a layered perovskite structure with the lattice constant \( a = 4.1 \m A \) and the effective ratio \( \epsilon/m^* \) being the twice of the bare value, \( n = 1/2, 1/3 \) and \( 2/3 \) are located in the solid region while \( n = 1/4, 3/4 \) and fractions with higher denominators are located in the liquid region as we see in Fig. 7. We note that the filling control at fixed lattice constants follows a contour line of \( \epsilon/m^* \) in Fig. 7. As we also see in Fig. 7, the HF approximation fails in accounting this general experimental trend. The general trend can be understood only by taking account of quantum fluctuations sufficiently.

Even metallic states near the charge order may inherently contain such charge-order and stripe proximities e.g. formation of quantum and dynamical defect structure, in the region where the static solid could be stabilized on the mean-field level. Competitions with hierarchical structure formation by various levels of fractional numbers of fillings become intrinsic properties leading to self-organized structure. This may be particularly conspicuous near the simple commensurate order such as the Mott insulator, because the mass renormalization increases effective \( r_s \) with more continuous character of the transition. Reanalyses of mechanisms of high-Tc superconductivity in the cuprates and the colossal magnetoresistance in the manganites from the present viewpoint would be desired in future, since they appear in the proximity of insulator stabilized by the commensurability.

In summary, we have studied the liquid-solid transition of two-dimensional electrons with long-range Coulomb interaction. We have shown how the Mott insulator at \( n = 1 \), charge orders at \( 0 < n < 1 \) and the Wigner lattice at \( n \rightarrow 0 \) are connected. The charge order transition does not sensitively depend on the spin degrees of freedom for \( n < 1/2 \). Although the HF transition points are rather insensitive to the electron filling and lattice commensurability, the PIRG result with quantum fluctuations taken in a correct way shows rapid increase of \( r_s \) at the transition with decreasing filling. The present PIRG results explain why charge orders or stripes are commonly observed only at simple fractional fillings such as 1/2 and 1/3 but usually are not away from them. We stress that such interplay between quantum fluctuations and the commensurability may play crucial role also in metals (quantum liquids). It would be interesting if the interplay can be experimentally studied by microfabrication of periodic potential to high-mobility 2D electron systems.

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