Strong coupling theory of the spinless charges on triangular lattices: Possible formation of a gapless charge-ordered liquid

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We propose an existence of charge-ordered liquid state in the spinless fermion system on a triangular lattice under strong intersite Coulomb interactions, V. The classical ground state in the strong coupling limit is disordered due to geometrical frustration. The introduction of a quantum hopping term lifts the degeneracy and drives the system to a partially ordered phase, which we call a "pinball liquid." A possibly long-range-ordered Wigner-crystal solid coexists with a liquid component which is moving around them like a pinball. This liquid is dominant over a wide range of filling and even away from the regular triangle. Relevance to the organic θ-ET_2X is discussed.

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Geometrical frustration induces exotic states in the strongly correlated systems. They are explored experimentally in many materials from transition metals such as $NiGa_2S_4$,[1] Na_xCoO_2,[2] and YFe_2O_4 (Ref. 3) to organic solids, κ-$ET_2Cu_2(CN)_3$.[4] Theoretical studies on frustrated spin systems are conventionally focused on a search for "spin liquids"[5] without any long-range order. However, after the destruction of magnetic order, another nonmagnetic type of order often appears instead. A typical example is a dimer formation[6] and recently, a new gapless nematic order[7,8] is found on a frustrated square lattice as well as a supersolid on a triangular lattice.[9]

Charge degrees of freedom with frustration has also been discussed in the past,[10] and one of the latest topics is a melting of charge order (CO) into the metallic state on the triangular lattice[11] in analogy with the spin liquid. Although this seems to support an "orderless-charge-liquid" picture, there remains a possibility of another type of ordering. In this paper, we propose a new type of *partially ordered charge liquid* in such a triangular lattice system, which turns out to have very similar character with the supersolid in a hard-core boson system.[9] We find a spontaneous phase separation of charge degrees of freedom into a "statically ordered solid" and "liquid" components, which is possibly a gapless spatial ordering. The results are to be compared with an inhomogeneous metallic state found in θ-$ET_2CsZn(SCN)_4$.[12]

One of the typical cases where CO resides is the quarter-filled electronic systems under strong electronic interactions.[13] It is conventionally described by the extended Hubbard model (EHM) including the on-site and intersite Coulomb interactions, U and V, respectively,[14] besides the transfer integrals, t. Since the spin degrees of freedom plays only a secondary role when $t<V\ll U$, i.e., $J\sim t^2/U\ll t$, treating the spinless fermions at half filling, $\rho=0.5$, is enough to understand the CO phenomena realized in the temperature range of $J<T<T_{CO}$, where T_{CO} denotes the onset temperature of CO. We introduce a Hamiltonian of such a t-V model as

$$H_{t\text{-}V}=\sum_{\langle i,j\rangle}(-t_{ij}c_i^\dagger c_j+\text{H.c.}+V_{ij}n_in_j). \tag{1}$$

Here, c_j denotes the annihilation operator of fermions and $n_j(=c_j^\dagger c_j)$ is its number operator. The index $\langle ij\rangle$ are the nearest-neighbor (NN) pair sites. We deal with the triangular lattice with anisotropy in one of three directions. The geometry is reflected in t,t' and V,V' as shown in Fig. 1.

The classical limit, $t=t'=0$, of this model has a macroscopically degenerate and disordered ground state at $V=V'$, which is classified into two groups.[15] One group has the staggered alignment of charges in one direction but with still disordered interstacking patterns, as shown in Fig. 1. The other states are basically made up of three sublattices as shown in Fig. 2(a). As long as the two sublattices (A and B) are filled and empty (denoted as 1 and 0), the third sublattice (C) can have an arbitrary configuration. The binding energy of its hexagon unit in Fig. 2(b) is always $-3V$.[16] There are many irregular contingent freedoms included in this group such as the one in Fig. 2(c). Since the binding energy is unchanged over the filling, $1/3\leqslant\rho\leqslant 2/3$, the system is gapless at $\rho=0.5$.[16] This is in sharp contrast to the striped states with the excitation gap of $2V$.

The situation changes under the anisotropy of $V\neq V'$. At $V>V'$, the vertical striped CO in Fig. 1(a) becomes a unique ground state. As for $V<V'$, we have the staggered alignment along the anisotropic direction still with interstacking disorder under a semimacroscopic (2^{N_x}-fold) degeneracy. The classical binding energy E_C of the anisotropic triangle at the fixed value of $V+V'\equiv V_{\text{sum}}$ is given in Fig. 3(a). The energy

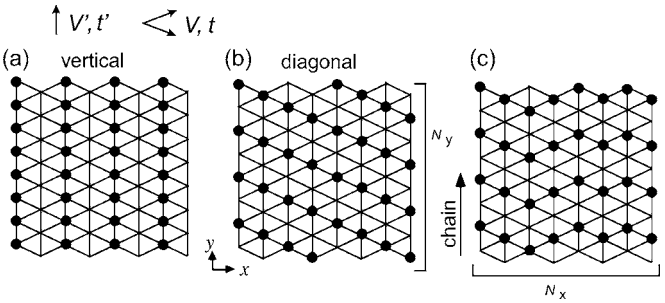

FIG. 1. Schematic description of the typical CO on an anisotropic triangular lattice. The staggered alignment of charges in one particular direction (CO chain) with either (0101)- or (1010)-type of configuration is present. Representative regular stacking of CO chains are (a) vertical and (b) diagonal-striped states. One example of the random alignment is given in (c).

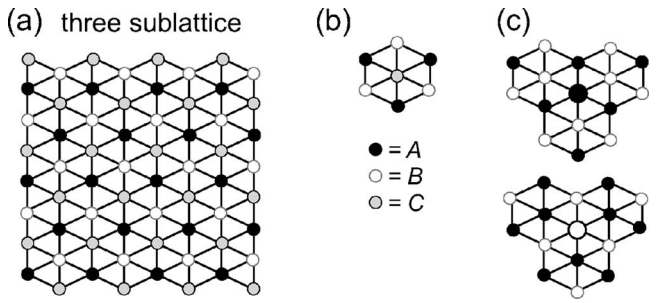

FIG. 2. (a) Three-sublattice structure: when two sublattices (A and B) are filled (1) and empty (0), respectively, the third one (C) could individually choose either 0 or 1. (b) Hexagonal unit of (a): The center site (C) is free, surrounded by the staggard edges. (c) Representative contingents: When the three nearest C sites happened to have the same configuration, the center site (A or B in the big circle) can switch its number without the energy loss.

line follows $E_C \propto VN/2$ and $-VN/2$ at $V<V'$ and $V>V'$, respectively, and becomes exactly equal to the three-sublattice one at $V=V'$.

In the highly frustrated disordered region an introduction of small t_{ij} is expected to lift the degeneracy. Let us start from the regular triangle at $V=V' \gg t=t'$. The first term of Eq. (1) is considered as perturbations. In the striped states, the first-order correction is absent and the energy changes to $\sim(V-4t^2/V)N/2$. Since this effect is small, the stripes are quite robust.

In contrast, the three sublattice states are modified at the first-order level. For demonstration, we first deal with the simplest $\rho=1/3$ case. The charges align equally spaced like a Wigner crystal, e.g., in the A sublattice. Next we add a single charge to one of the B or C sites, and then it can hop by t to its neighboring B or C sites as in Fig. 4(a) without the loss of binding energy. Resultantly, the charge can move around avoiding the A site just like a pinball, so we call this "a pinball liquid." The same situation holds at $\rho=2/3$ where we

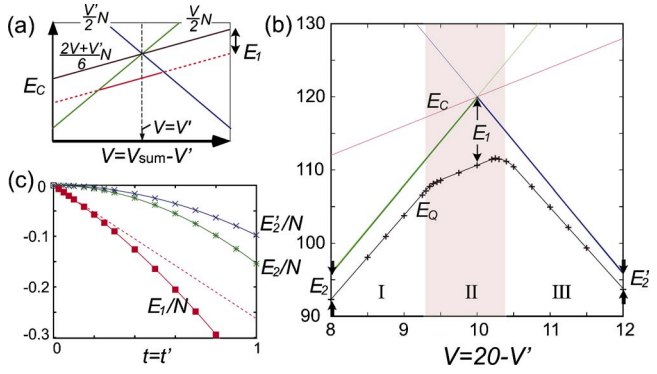

FIG. 3. (Color online) (a) Classical energy E_C at fixed $V+V'=V_{\mathrm{sum}}$. The vertical and disordered stripes and three-sublattice states have $VN/2, V'N/2$, and $(2V+V')N/6$, respectively. (b) Ground-state energy E_Q of Eq. (1) at $N=24$ with $V_{\mathrm{sum}}=20$ and $t=t'=1$, classified into (I) disordered (horizontal) stripe, (II) pinball liquid, and (III) vertical stripe, where E_Q behaves almost linear to respective E_C's. (c) Energy correction, $E_{1/N}$(II) and $E_{2/N}$(I,III). Solid lines are the results of fitting. A t-linear term in a broken line is present only at (I).

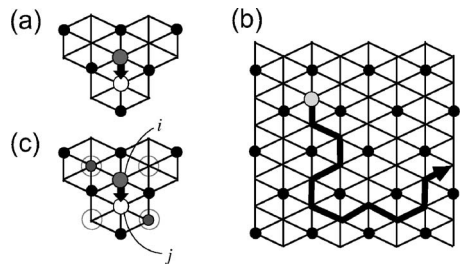

FIG. 4. (a) Representative bases of the three-sublattice states at $\rho\sim1/3$ that mix in the first order of t. (b) Schematic illustration of a pinball model. (c) Representative bases away from $\rho=1/3$, which are allowed to mix by t. The ball can move from the ith to the jth site when the extra two neighboring sites of the ith site with big circles together have the same number of balls with those of the jth site.

regard the "pins" as a hole-rich site after the particle-hole transformation. In the dilute limit, the extra charges thus form a honeycomb band.[17] Such simple understanding is, however, nontrivial when the charge density becomes close to half filling, $\rho\sim0.5$, where about one-fourth of the B and C sites are filled. The "balls" can at least move from one site to another following the rule shown in Fig. 4(c). Still, there are several unsettled issues. The first problem is that a significant number of states [including the contingents in Fig. 2(c)] will prevail, which might mix by t and destroy the pins. Also, the instability of the Fermi liquid might replace the pinball liquid with another ordering. Another thing is that the free pinball picture no longer holds since there are interactions between balls themselves. Finally, the validity of the above discussion is not clear when the anisotropy is introduced.

To clarify these points we proceed with the numerical calculations in Eq. (1) at $\rho=0.5$. We execute the exact diagonalization at $T=0$ on the $N=4\times6=24$ cluster. Then both the striped type of twofold states and the three-sublattice states are compatible. Size dependence is confirmed to be small.[18] Figure 5 shows the structural factor of the charge-charge correlation at $V=V'=10$ and $t=t'=1$ under the periodic boundary condition. It is defined as $C_k=\frac{1}{N}\Sigma_{lm}\langle(n_l-1/2)(n_m-1/2)\rangle e^{i(l-m)k}$. The three-sublattice type of peak structures at $\vec{k}=(\pi,\pm2\pi/3),(0,\pm4\pi/3)$ are observed, which grow with increasing V, while those of the twofold stripes are suppressed.

The pinball liquid is sustained even under the anisotropy

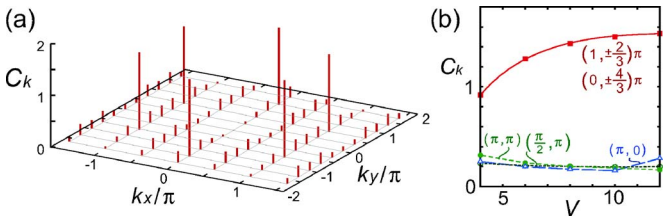

FIG. 5. (Color online) (a) Charge structural factor C_k at $V=V'=10$, $t=t'=1$. (b) $V(=V')$ dependence of the peak at $\vec{k}=(0,\pm4\pi/3),(\pi,2\pi/3)$ representing (II). Those of the stripes at (I) (π,π), $(\frac{\pi}{2},\pi)$, and (III) $(\pi,0)$ have only small amplitudes comparable to those in other $\vec{k}$ points.

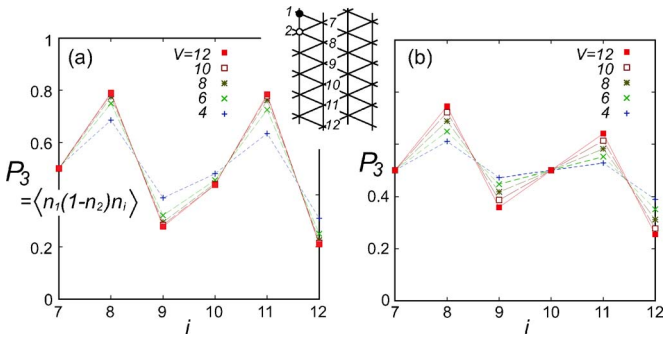

FIG. 6. (Color online) Three-body charge-correlation function $P_3(j)$ along the second chain ($i=7-12$) at $t=t'=1$ for several choices of $V=V'$. Here, i denotes the location of the site indicated in the inset. Those of the (a) spinless fermion and (b) hard-core boson systems are shown.

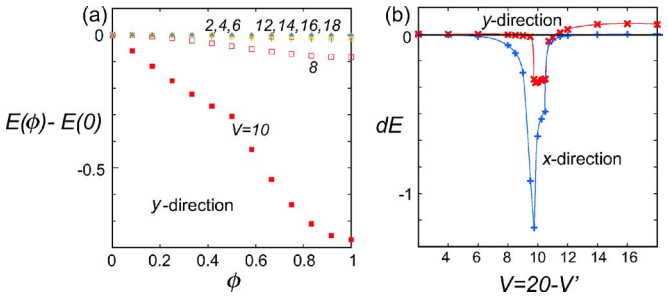

FIG. 7. (Color online) (a) Variation of the ground-state energy $E(\phi)-E(0)$ as a function of $\phi=0-\pi$, under the twisted boundary condition in the y direction for several choices of V with $V+V'=20$ fixed. (b) The amplitude of variation $E(\pi)-E(0)$ as a function of V along $V_{\mathrm{sum}}=20$, when the boundary is twisted in the x and y directions.

of $V\neq V'$ or $t\neq t'$. The binding energy per "ball" is modified from $-3V$ to $-2V-V'$. If only the first-order correction of energy by t, E_1, is finite the three-sublattice state should overcome the others as in Fig. 3(a) and form a new phase at $V\sim V'$. Figure 3(b) shows the ground-state energy of Eq. (1) together with E_C at fixed $V+V'=20$. The diagram is separated into three regions; the energy line behaves parallel to different classical states: (I) disordered stripe, (II) three sublattice, and (III) vertical stripe. The pinball liquid realized in (II) extends at $V\neq V'$ over a width of order t. The estimated energy gain per site, $(E_Q-E_C)/N$, is given in Fig. 3(c). We find larger gain, $E_1\propto t$, in (II) than the second-order gain ($E_2\propto t^2$) of the (I) and (III) regions. In this way, the existence of a finite pinball-liquid phase is guaranteed at $V\sim V'$.

To see the details, we calculate the three-body correlation function, $P_3(j)\equiv\langle n_1(1-n_2)n_j\rangle$, which reveals the population of the jth site when the first and the second site are present and absent, respectively. The ones along the y direction for several choices of $V=V'$ are shown in Fig. 6(a). A clear threefold structure of A-B-C type with different density, $P_3(A)+P_3(B)\sim 2P_3(C)$, suggests the presence of particle-hole symmetry. Here, $P_3(A)$ denotes the correlation when the jth site belongs to the A sublattice. This characteristic three-body correlation originates from the geometrical frustration and essentially differs from the two-body ones in the conventional CO states. The amplitude of the "pins" are squeezed to ~ 0.8, but grows with V to form a firm structure at least over considerable distance. Whether or not this correlation has a long-range order is out of our scheme. Just for reference we remind one of a supersolid long-range order of the hard-core boson.[9] This boson system has very similar $P_3(j)$ with that of fermions as shown in Fig. 6(b). Hence the long-range order of the pinball liquid is speculated as well.

It is natural to anticipate the gapless situation at $t\neq 0$ from its classical limit. Since the direct estimation of a gap in small clusters is unreliable, we instead focus on the coherence length of each characteristic state. We twist the boundary condition as $e^{i\phi}$, where $\phi=0$ and π correspond to the periodic and antiperiodic boundary conditions, respectively. The ground-state energy $E(\phi)$ should have considerable ϕ dependence only when the coherence lasts longer than the system length. Figure 7(a) shows the variation of $E(\phi)-E(0)$ as a function of ϕ when twisting the boundary in the y direction with $V+V'$ fixed. In the regions (I) and (III), $E(\phi)$ remains almost constant, reflecting the insulating character. In contrast, the ones in (II) show large ϕ dependence, indicating that pinball wave function has a delocalized character at least over dozens of sites. Figure 7(b) shows the amplitude of the energy variation, $E(\pi)-E(0)$, when twisting ϕ in the x and y directions. One actually finds a significant difference that separates (II) from the other two states.

In summary, we proposed the existence of a pinball-liquid state on a triangular lattice in the spinless fermion systems with NN Coulomb interactions over a wide range of filling, $1/3<\rho<2/3$. It is an ordered liquid formed out of disorder characterized by a three-body correlation. Thus, although the state is metallic, it is driven by the local correlation of electrons at $V\gg t$ and is not related to the details of the Fermi surface, namely, the geometrical structure of t. This nontrivial fact gives the reason why the statistically different systems can share the analogous situation.[9] It is also insensitive to the anisotropy of V. Further, it is expected to be sustained when the degrees of freedom other than charges such as spins and orbitals are introduced, e.g., EHM at quarter filling.

Recently there have been other theoretical studies on the anisotropic triangular lattice that argue the existence of the threefold states.[19,20] Unfortunately, their treatment was based on the mean-field wave functions, which somewhat takes account of the instability of the Fermi liquid towards the two-body correlations. Therefore, they could not capture the essential highly correlated features of the pinball liquid. The present theory gives the explicit and clear-cut answers to the nature of this system alluded to preliminarily in such previous studies.[11,19,20]

Related experimental studies are explored in θ-ET_2X. This family is well described by the 1/4-filled EHM on a triangular lattice[21] with $V/V'\sim 0.85-0.9$.[22] The conventional insulating striped CO was considered as relevant for a long time, but recently a coexistence of the short-range diffuse spot of twofold and threefold periodicity is observed,[23] which now requires theoretical support. From our strong coupling viewpoint, both twofold striped and pinball-liquid states are compatible (though not coexisting) without having

special assumptions such as long-range interactions or the particular shape of the Fermi surface. If the systems were in the critical region of these two competing characteristic states, some additional effects such as impurity, temperature (entropy effect), or electron-phonon interaction might induce a coexistence or domain structure that cope with these anomalous experimental findings.[12] Theoretical development in the search for further unusual aspects particular to the fermion system remains a future problem.

In conclusion, we found a charge-ordered liquid under geometrical frustration, which is possibly relevant at a temperature range of $J<T\ll V$. In the strong-coupling limit, the frustration among electronic interactions V drives the system to disorder, which consists of macroscopically degenerate classical states. Then, the introduction of small but finite t $(\ll V)$ lifts the degeneracy and transforms the system to a strongly correlated quantum liquid with a three-sublattice correlation A-B-C whose electronic density is $n_A+n_B\sim 2n_C$. About half of the charges become a Wigner-crystal-solid while the rest remain a liquid. An originally unique charge degrees of freedom spontaneously separates into the solid and liquid-like part and coexists in the same system. Such correlation-induced order out of disorder distinctively differs from the usual charge ordering or charge-density waves formed from the instability of the Fermi liquid. This state remains valid regardless of electronic filling, the shape of the Fermi surface, and anisotropy of V. Based on the simple fundamental model, it will surely become a testbed for the study of an extremely correlated metal.

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