Phase diagram of the triangular extended Hubbard model

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The study of frustrated and strongly interacting systems in two dimensions has received an unbroken intensity of research activities in recent years. Just to name a few examples, spin-liquid states have been postulated in frustrated two-dimensional antiferromagnets \[1–4\], supersolid phases have been established for hardcore bosons on a triangular lattice \[5–7\], and the concept of deconfined quantum critical points \[8\] has sparked a tremendous interest in the search of exotic phase transitions.

Above examples involve essentially spin-like systems, where charge degrees of freedom only play a passive role. When both spin and charge degrees of freedom are considered at incommensurate filling the situation potentially becomes even more interesting. In order to study the interplay of frustration and strong interactions with spin and charge degrees of freedom at arbitrary filling \[n = (n_\uparrow + n_\downarrow)\], the extended Hubbard model on the triangular lattice is the prototypical system to study. In standard notation the model is given by the many body Hamiltonian

\[
\mathcal{H} = -t \sum_{\langle i,j \rangle, \sigma} c_{i,\sigma}^\dagger c_{j,\sigma} + \text{h.c.} + U \sum_i n_{i,\uparrow} n_{i,\downarrow} + V \sum_{\langle i,j \rangle} n_i n_j.
\]  

In addition to its fundamental importance, this model is of interest for describing the rich and complex behavior of organic conductors, such as the charge transfer salts \(\theta-(\text{BEDT-TTF})_2\chi\) \[12, 18\], where molecules are arranged on an anisotropic triangular lattice with incommensurate filling. The Hubbard model on the triangular lattice has also been considered in the context of explaining superconductivity in the layered compound \(\text{Na}_x\text{CoO}_2\) \[17, 18\] where interesting textures have been predicted recently \[18\]. Unfortunately, analytical and numerical studies of this model are far from trivial and to our knowledge it has not yet been analyzed with quantum many body simulations for incommensurate filling. In this paper we now use numerical variational Monte Carlo simulations in order to establish the phase diagram as a function of filling and interaction strength. In addition to the ordinary metallic phase, three interesting phases are found, where long-range charge order and metallic conductivity are present simultaneously as depicted in Fig. 1 which summarizes most of our findings.

In order to systematically understand the different phases let us first review the simpler case of commensurate \(2/3\) filling \((n = 2/3)\) discussed in Ref. \[15\]. Quite intuitively, for strong \(V \gtrsim U/3 \gg t\) any nearest neighbor occupation is forbidden, resulting in an insulating ordered phase with exactly two electrons on one sublattice \((200\) order), while for weaker nearest neighbor repulsion double occupancy is forbidden and instead two sublattices are half-filled in an hexagonal order \((110\) order), see Fig. 1. Neglecting hopping, these phases are also stable for lower filling \[13\]. However, using a grand-canonical point of view, the filling simply jumps as a function of chemical potential \(\mu\) in the limit of vanishing hopping. In particular, at \(\mu = 3V\) a jump occurs from the stable 100-order with \(1/3\) filling to a \(2/3\) filled 110-state. At finite hopping \(t \ll V \ll U\), however, incommensurate filling is stabilized in a finite range \(|\mu - 3V| \lesssim 3t\) due to the kinetic energy of particles (holes) on the hexagonal order \[21\]. In the following we will focus on analyzing the partially filled state in the density range \(1/2 \leq n \leq 2/3\).

For incommensurate filling, the 110 phase in Fig. 1 represents a state where one of the three triangular sub-
lattices remains empty and all the electrons occupy the other two sublattices in an hexagonal density order in order to minimize the nearest neighbor repulsion. Since the hexagonal order necessarily contains holes for \( n < 2/3 \), this phase becomes conducting. Interestingly, this coexistence of two counter-intuitive properties (order and conductivity) is directly related to the supersolid state on triangular lattices which has been established for hardcore bosons \( [4,5] \) and has analogously been postulated for spinless fermions \( [21,22] \). Simulations for hardcore bosons have shown that a separation into two types of hole-like particles is possible \( [20] \): One part creates the ordered state by keeping one sublattice empty, while the other part can move freely on the hexagonal structure (partial liquid). However, as the filling approaches \( n = 2/3 \) we observe a transition to antiferromagnetic (AF) order coexisting with conducting behavior, which must have a different mechanism as described below.

For the extended Hubbard model we find a third interesting phase in the form of a 200 order in Fig. 1. Double occupancy occurs only on one sublattice which is reduced with filling and gives an ordered state. This state has some surprising properties, since the observed conductivity implies that the other two sublattices are not completely empty either. The occupation on those two sublattices therefore increases with decreasing filling, which leads to conductive behavior. The phase transition between the 110 and 200 states is first order close to commensurate filling but may become second order for \( n \lesssim 0.57 \). For large hoppings a transition to a simple metal occurs.

In order to simulate the model in Eq. (1) at zero temperature we have used the Variational Monte Carlo (VMC) method \( [23] \). This method gives very good results \( [24] \) even for correlated and frustrated systems by numerically sampling expectation values over a variational ansatz. A powerful correlated variational state is given by \( [25,28] \)

\[
\langle \Psi_{\text{FS}} \rangle = \mathcal{J} \langle \mathcal{F} \rangle,
\]

where \( \mathcal{F} \) is the non-interacting filled Fermi sea, to which a finite small superconductive term is added in order to regularize the wave function, i.e. to separate the highest occupied and the lowest unoccupied states by a gap. The term \( \mathcal{J} = \exp(-1/2 \sum_{ij} v_{ij} n_i n_j) \) is a density-density Jastrow factor, where the \( v_{ij} \)'s are optimized with VMC for every

\[
\frac{\sum_{ij} v_{ij} n_i n_j}{\sum_{ij} n_i n_j} \lesssim 1.
\]

We indentify a simple metallic phase, a metallic state with a 110 charge order, and a metallic state with a 200 charge order. The charge order regions are insulating at commensurate filling \( n = 2/3 \) (thick lines) and metallic otherwise. In the limiting case \( n = 1/2 \) (green line) only the 200 charge order. The charge order regions are insulating at commensurate filling and give an ordered state. This state has some surprising properties, since the observed conductivity implies that the other two sublattices are not completely empty either. The occupation on those two sublattices therefore increases with decreasing filling,

\[
\frac{\sum_{ij} v_{ij} n_i n_j}{\sum_{ij} n_i n_j} \lesssim 1.
\]
independent distance $|i-j|$ (including on-site). Backflow correlations further improve the correlated state $|\Psi_{FS}\rangle$; in this approach, each orbital that defines the unprojected state $|\Psi_{FS}\rangle$ is taken to depend upon the many-body configuration in order to incorporate virtual hopping processes \cite{29}. The non-interacting state $|\Psi_{FS}\rangle$ also includes three different chemical potentials as variational parameter, one for each sublattice. We must emphasize however, that even for a uniform variational chemical potential the charge ordered metallic states spontaneously appears in the phase diagram at arbitrary filling, which demonstrates the stability of this phenomenon. Finally, a coupling to an external field can be added to the mean-field $|\Psi_{FS}\rangle$ state in order to check if the ground state is magnetically ordered. All results presented here are obtained by fully incorporating the backflow corrections and optimizing individually \cite{30} every variational parameter in the wave function.

The static structure factor $N(q) = \langle n_q n_q \rangle$ is a good indicator for metallic behavior, where $n_q = 1/\sqrt{L} \sum_{r,\sigma} e^{iqr} n_{r,\sigma}$ is the Fourier transform of the particle density. The metallic phase is characterized by $N(q) \propto q$ for $q \to 0$, which implies a vanishing gap for particle-hole excitations. On the contrary, $N(q) \propto q^2$ for $q \to 0$, implies a finite charge gap and insulating behavior. \cite{29} We find conducting behavior everywhere except for $n = 2/3$ and $V/t \gtrsim 3$, as shown in Fig. \ref{fig:3}. Interestingly, a diverging behavior of $N(q \to 0)$ is observed in the 200 phase, which we attribute to a $q^2$ dispersion relation at effective low filling as explained below.

In order to distinguish between the different kinds of charge ordering in the model, we plot in Fig. \ref{fig:3} the electronic density per sublattice $n_{\alpha}$ with $\alpha = A, B, C$, that is the number of electrons divided by the number of sites, in each of the three sublattices. Within the non-ordered metallic phase, the electronic density is expected to be the same on each sublattice, while in the 110 region one sublattice depletes, with the electrons forming an hexagonal density order, see Fig. \ref{fig:3}. Finally, in the 200 phase one sublattice is occupied with a density much greater than one, due to the large number of double occupancies. Both the cases $n = 2/3$ and $n = 0.583$ in Fig. \ref{fig:3} show a clear distinction between the three regimes, while in the limiting case $n = 1/2$ only the 200 charge order may be observed, with a single sublattice being more and more occupied as long as the ratio $V/t$ increases.

Even though an high density of electrons on one sublattice is the expected behavior for a small hopping $t$ and $3V > U$, the 200 ordered metal has rather unusual properties. First of all it is far from obvious why this ordered state is conducting. In the 110 order the conductivity can be explained by mobile holes moving on a hexagonal substructure \cite{20,22}. In the 200 order on the other hand, holes only appear on one sublattice which is not connected, so this argument fails. Moreover, we have checked that all the electrons that are in a doubly occupancy in each of the three sublattices. Within the non-ordered phase, otherwise, if $\phi > 3D^2$ holds, all the double occupancies participate to the charge order, otherwise, if $\phi < 3D^2$, a fraction of the double occupancies is mobile outside the 200 pattern, with the limiting case $\phi = D^2$ corresponding to an uniform distribution of double occupancies in the lattice. According to the result shown in Fig. \ref{fig:3} for $V/t = 10$, the relation $\phi = 3D^2$ is verified in all the doping range and the system separates into charge ordered double occupancies and free electrons that are responsible for the conduction mechanism. Therefore, conductivity appears to require a small density of electrons on the two sublattices which
are empty for $n = 2/3$, i.e. the density on the two almost empty sublattices must increase with decreasing $n$. In Fig. 5 we show $\delta n$, that represents the electronic filling on the hexagonal substructure, which is available for a conducting band in the 200 regime. In the case $V/t = 11$ it is clear that $\delta n$ increases at increasing doping, while in the case $V/t = 10$ there is a small decrease in $\delta n$ when doping becomes large, i.e. $n \lesssim 0.57$. This is just a consequence of the small and almost constant number of double occupancies that occurs at $V/t = 10$ in the range $1/2 \leq n \lesssim 0.57$, see Fig. 4. Indeed, in this density range the transition between the 110 and the 200 phases becomes second order with a smooth increase of the number of double occupancies as a function of $V/t$. The effective filling of electrons on the hexagonal substructure $\delta n$ is rather low in the range $0.57 \lesssim n \leq 2/3$. Accordingly, the electrons follow a $q^2$ dispersion relation at the bottom of the band, which explains the divergence of $N(q \rightarrow 0)$ in this phase, as discussed above in Fig. 2.

Finally, we also tested for magnetic order and found that an antiferromagnetic state has lower variational energy for fillings $0.62 \lesssim n \leq 2/3$ as indicated in Fig. 1. While antiferromagnetic order is expected for commensurate insulating fillings, it should immediately be destroyed by moving holes on the hexagonal substructure. However, for very small doping close to filling 2/3 the energy gain from hopping of order $t(2/3 - n)$ is not sufficient to overcome the energy gain from long-range antiferromagnetic order of order $nt^2/U$. Nonetheless, second order hopping processes of holes via the depleted sites are still possible without changing the spin orientation, so that a finite conductivity is observed in co-existence with antiferromagnetic order in this special case. This phase is stabilized for larger second order hopping amplitude $t^2/V$ in agreement with our finding in Fig. 1.

In conclusion, we have analyzed the extended Hubbard model on the triangular lattice as a function of interaction strength and filling. The phase diagram in Fig. 1 shows three metallic phases at incommensurate filling. A simple metallic phase is confirmed for large hopping. With increasing interaction strength an ordered metal with a 110-type order is observed, due to the appearance of holes on a stable hexagonal order, which is analogous to the underlying mechanism for supersolidity [20]. For filling close to 2/3 we observe a phase transition to an antiferromagnetically ordered metal. A 200-ordered phase with one double occupied sublattice is found for still larger nearest neighbor repulsion, which surprisingly also shows conductive behavior. The observed occupancy of the sublattices B and C and the electronic properties are consistent with a band on the hexagonal substructure with very low filling. This is surprising, since the strong nearest neighbor repulsion naively presents a large energy barrier for electrons on the hexagonal substructure next to the double occupied sites. The detailed mechanisms of the conductive behavior both in the 200 phase and in the 110 antiferromagnetic phase remain a topic of future research.

Experimentally, charge ordering phenomena in charge transfer salts have been researched with a large variety of methods, e.g. NMR, X-ray and Infrared/Raman spectroscopy [31]. Coexistence of metallic behavior and charge ordering has only been observed in few cases for $\beta$-(BEDT-TTF)$_2$X and $\beta''$-(BEDT-TTF)(TCNQ) charge transfer salts and only for short range charge order [32]. The scenario we have proposed in this paper predicts a coexistence of metallic behavior and long-range order, which is not due to a partial instability of the Fermi surface. Instead we can identify a separation into two functional classes of particles (or holes): part of them contribute to a stable order on one sublattice, while another part forms a partially filled band on the remaining hexagonal substructure.

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