Metal-insulator transition in quarter-filled Hubbard model on triangular lattice and its implication for the physics of \( \text{Na}_{0.5}\text{CoO}_2 \)

Tao Li

Department of Physics, Renmin University of China, Beijing 100872, P.R.China
(Dated: January 5, 2010)

The metal-insulator transition of the quarter-filled Hubbard model on triangular lattice is studied at the mean field level. We find a quasi-one dimensional metallic state with a collinear magnetic order competes closely with an insulating state with a non-coplanar magnetic order for both signs of the hopping integral \( t \). In the strong correlation regime \((U/|t| \gg 1)\), it is found that the metal-insulator transition of the system occurs in a two-step manner. The quasi-one dimensional metallic state with collinear magnetic order is found to be stable in an intermediate temperature region between the paramagnetic metallic phase and the non-coplanar insulating phase. Possible relevance of these results to the physics of metal-insulator transition in \( \text{Na}_{0.5}\text{CoO}_2 \) is discussed.

PACS numbers:

\( \text{Na}_\frac{2}{3}\text{CoO}_2 \) is a remarkable transition metal oxide system in which strong electronic correlation and geometric frustration interplay with each other. Unconventional superconductivity, novel magnetic and charge ordered states and state with peculiar Curie-Weiss metal behavior are observed in this system at different carrier concentrations \([3, 6, 8]\). To date, most of these observations remain not well understood.

The quarter-filled system, namely \( \text{Na}_{0.5}\text{CoO}_2 \) is especially interesting \([3, 6, 8]\). It divides the phase diagram of the \( \text{Na}_\frac{2}{3}\text{CoO}_2 \) system into two parts of qualitatively different nature. For \( x < 0.5 \), the system exhibits conventional metallic behavior in the normal state and with water intercalation a superconducting state with unknown paring symmetry in a small doping region around \( x = 0.3\) \([5]\). While for \( x > 0.5 \), the system exhibits Curie-Weiss metal behavior at high temperature and a weak static magnetic order at low temperature with ferromagnetic intra-layer spin correlation \([4, 5, 7]\). Right at \( x = 0.5 \), the observations are even more striking. The system undergoes two phase transitions at 88K and 53K \([3, 6, 8]\). At the 88K transition, the system develops a magnetic order with a ordering wave vector \( \mathbf{q} = \frac{1}{2}(\mathbf{b}_1 + \mathbf{b}_2) \) \([4, 5, 7]\). However, the longitudinal resistivity of the system seems to be totally ignorant of the transition. While at the 53K transition, the system enters the true insulating state as signaled by the rapid increase of the longitudinal resistivity below this temperature. However, both the peak position and the intensity of the magnetic Bragg peak developed at 88K seems to be ignorant of the transition at 53K. The observation of two transitions and the apparent mutual independence of the magnetic order and the charge transport have aroused many interest in the field. However, this is still not the whole story. In fact, it is found that Hall resistivity begins to drop abruptly around the 88K transition which indicates that it is not a transition in the spin sector only \([3]\).

No coherent picture yet exists on the low temperature phase transitions of the \( \text{Na}_{0.5}\text{CoO}_2 \) system. It is generally anticipated that the ordering of the Na ions outside the \( \text{CoO}_2 \) plane may plays an important role in the low temperature physics of this system \([3, 6, 11, 13]\). Charge ordering (induced by the Sodium order) in the \( \text{CoO}_2 \) plane is invoked in many works to explain the multiple phase transitions. In this paper, we suggest a possible picture for the multiple phase transitions of the \( \text{Na}_{0.5}\text{CoO}_2 \) system as an intrinsic property of a quarter-filled strongly correlated system on the triangular lattice. We backup our reasoning with a mean field study of the metal-insulator transition of the quarter-filled Hubbard model on triangular lattice. We find this seemingly oversimplified model does exhibit two successive transitions at quarter filling in its way toward the low temperature insulating state. In our picture, the magnetic transition at 88K transform the system into an quasi-one dimensional metallic state through a spin ordering induced dimensional reduction process. This quasi-one dimensional metal can then transform into the true insulating state at 53K in various possible ways. We thus predict that the intermediate phase between 53K and 88K to be a state with one dimensional transport property.

FIG. 1: (a)The magnetic structure of the proposed state for the intermediate temperature region between 53K and 88K. Note the state has no charge order and all lattice sites are still uniformly quarter-filled. (b)The insulating state with a four sublattice and non-coplanar magnetic structure predicted by the mean field theory of the quarter-filled Hubbard model on triangular lattice. The magnetic moments on the four sublattices point to the four corners of a perfect tetrahedron as shown in the inset.

The state we propose for the intermediate temperature region between 53K and 88K is shown schematically in Figure 1a. In this state, the system self-organizes into a series of chains with ferromagnetic order on them. Neighboring chains are antiferromagnetically correlated with each other. Unlike other proposals, there is no charge ordering in this state and all lattice sites are still quarter-
filled uniformly and thus the ferromagnetic chains are metallic. It is interesting to note that this metallicity is of one dimensional nature as any inter-chain electron hopping must cross the spin gap. Thus the spin ordering in such a particular pattern renders the system a one dimensional metal and so we dub it as a spin ordering induced dimensional reduction process. Similar dimensional reduction process also occurs in other condensed matter system with multiple low energy degree of freedoms. The most well known example is the orbital ordering induced dimensional reduction in ruthenium oxides [15].

The magnetic structure of the state shown in Fig.1a is given by \( S_i > = \vec{m} \exp(iQ \cdot \vec{R}_i) \) with \( Q_3 = 1/2(\vec{B}_1 + \vec{B}_2) \). The states with \( Q_1 = 1/2\vec{B}_1 \) and \( Q_2 = 1/2\vec{B}_2 \) are degenerate with it by lattice symmetry. All the three states are consistent with the neutron scattering measurement and the real system will contain domains of all three kinds. Since the ferromagnetic chains are still metallic, the longitudinal resistivity of the system is not expected to change dramatically across the 88K transition. However, the transverse transport is totally different. As the system becomes quasi-one dimensional in charge transport state, we expect the Hall resistivity to drop abruptly at the 88K transition, in accordance with the experimental observation. Thus the state shown in Fig.1a has the correct characteristics to account for the phenomenology of the intermediate phase between 53K and 88K.

With further lowering of temperature, the quasi-one dimensional intermediate state will eventually become a true insulator. It is important to note that the intermediate state has an enlarged unit cell with two lattice sites. Thus the system is half-filled below the 88K transition. To arrive at the true insulating state, the system should lower its symmetry further to have a unit cell with at least four sites. Through mean field calculation on the Hubbard model on triangular lattice, we find a state with a four sublattice structure and non-coplanar spin configuration (see Fig.1b) that meet the requirement. This state has no charge order either and the magnetic structure can be written as \( S_i > = \vec{m}_1 \exp(iQ_1 \cdot \vec{R}_i) + \vec{m}_2 \exp(iQ_2 \cdot \vec{R}_i) + \vec{m}_3 \exp(iQ_3 \cdot \vec{R}_i) \). Here the three vectors \( \vec{m}_1, \vec{m}_2, \vec{m}_3 \) are of same length and are mutually orthogonal. The magnetic moments in the four sublattices points to the four corners of a perfect tetrahedron. This state has the same set of magnetic Bragg peaks as the collinear state shown in Fig.1a if the three kinds of domains of the collinear state appear with the same probability. The mean field theory presented in this paper also indicates that the transition between collinear state and the four sublattice insulating state leaves the intensity of the magnetic Bragg peak almost intact. This is in accordance with the observation at the 53K transition.

It should be noted that both kinds of order shown in Fig.1 have already been predicted in a mean field study of the quarter-filled Hubbard model on triangular lattice with a negative hopping integral [13]. In that work, both kinds of order develop as a result of the nesting property of the Fermi surface and exist for any non-zero interaction. However, the Fermi surface of the \( Na_{0.5}CoO_2 \) is almost featureless. As will be shown below, the \( Na_{0.5}CoO_2 \) system should be described as a quarter-filled system with a positive hopping integral. Since the triangular lattice has no particle-hole symmetry, any magnetic order can develop only for interaction greater than certain critical value. Indeed, we find the magnetic structures shown in Fig.1 exist only in the strong correlation regime for \( Na_{0.5}CoO_2 \).

We now present the results of the mean field study of the metal-insulator transition in the quarter-filled Hubbard model on triangular lattice. The Hamiltonian of the model reads

\[
H = -t \sum_{<i,j>,\sigma} (c_{i\sigma}^\dagger c_{j\sigma} + h.c.) + U \sum_i n_{i,\uparrow} n_{i,\downarrow}.
\] (1)

It is generally believed that the \( Na_{x}CoO_2 \) system resides in the strongly correlated regime with \( U \gg 1 \). Since the triangular lattice has no particle-hole symmetry, the sign of the hopping integral \( t \) is important. For \( Na_{x}CoO_2 \) system, it is negative as inferred from ARPES measurement [12]. In \( Na_{0.5}CoO_2 \), each \( Co \) site hosts 1.5 electrons on average is thus a \( 4/5 \)-filled system with a negative hopping integral. After a particle-hole transformation, it is equivalent to a quarter-filled system with a positive hopping integral. For purpose of comparison, we also consider the case of quarter-filled system with a negative hopping integral which is studied already in Ref 12.

From the weak coupling point of view, a quarter-filled system on triangular lattice with a positive hopping integral has no speciality at all as compared to a general incommensurate filling. The Fermi surface has a almost perfect rounded shape. Thus, it is hard to guess what would happen in the strong correlation regime from weak coupling analysis. To the contrary, a quarter-filled system on triangular lattice with a negative hopping integral is special in the sense that the chemical potential rests just on the Van Hove singularity of the density of state (\( \mu = -2|t| \) at zero temperature). The Fermi surface is nested with nesting vectors \( Q_1, Q_2, \) and \( Q_3 \). As the result of the nesting property of the Fermi surface, the system develops magnetic order at an arbitrarily small \( U \). In Ref. 12, the collinear state shown in Fig. 1a and the non-coplanar state shown in Fig.1b are found to be the most favorable choice for the magnetic structure. Among the two, the non-coplanar state is found to be slightly more favorable.

To have an idea on what would happen in the strong correlation regime for the quarter-filled Hubbard system with a positive hopping integral, we have conducted an unrestricted mean field search at zero temperature on a
12 × 12 lattice. The search is done in the mean field space with the local density \( n_1 \) and the three components of local spin density \( \mathbf{S}_i \) as variational parameters. We have used both the conjugate gradient method and the simulated annealing method to perform the search and used more than 100 random initial configurations for the local density and local spin density. We find the search always converge to the two configurations shown in Fig.1 (up to global spin rotations and point group operations) for large enough value of \( \frac{U}{t} \). Thus, although the quarter-filled system with positive hopping integral has no nesting property, it has the same magnetic ordering pattern in the strong correlation regime as the negative hopping system.

The mean field equations for the collinear state is given by

\[
1 = \frac{2U}{N} \sum_{\mathbf{k}} \left[ \frac{(\xi_{\mathbf{k}} - E_{\mathbf{k}}^+)}{(\xi_{\mathbf{k}} - E_{\mathbf{k}}^+)^2 + (Um)^2} f(E_{\mathbf{k}}^+) + \frac{(\xi_{\mathbf{k}} - E_{\mathbf{k}}^-)}{(\xi_{\mathbf{k}} - E_{\mathbf{k}}^-)^2 + (Um)^2} f(E_{\mathbf{k}}^-) \right]
\]

in which the sum is limited in the magnetic Brillouin zone and \( \xi_{\mathbf{k}} = -2t(\cos k_x + \cos k_y + \cos(k_x + k_y)) - \mu \) is the dispersion on the triangular lattice. Here we have used the convention for the momentum that \( \mathbf{k} = k_x \mathbf{b}_1 + k_y \mathbf{b}_2 \). In this convention, the first Brillouin zone of the triangular lattice is given by \( \{k_x, k_y\} \in [-\pi, \pi] \oplus [-\pi, \pi] \) and the magnetic Brillouin zone of the collinear state is given by \( [0, \pi] \oplus [-\pi, \pi] \). \( E_{\mathbf{k}}^\pm = \frac{\xi_{\mathbf{k}} + \xi_{\mathbf{k} + \mathbf{Q}_n} \pm \sqrt{(|\xi_{\mathbf{k}} - \xi_{\mathbf{k} + \mathbf{Q}_n}|^2 + (Um)^2)}}{2} \) is quasiparticle energy in the magnetic ordered state.

In the non-coplanar insulating state, the magnetic Brillouin zone reduces further into the region given by \( [0, \pi] \oplus [0, \pi] \). The mean field equation for the non-coplanar state is given by

\[
m = \frac{1}{N} \sum_{\mathbf{k}, \mathbf{n}} W^n_{\mathbf{k}} f(E^n_{\mathbf{k}}),
\]

in which \( W^n_{\mathbf{k}} = \frac{1}{2\sqrt{3}} \sum_{\alpha, \beta} \phi^n_{\alpha}(\mathbf{k}) M_{\alpha, \beta} \phi^n_{\beta}(\mathbf{k}) \), \( \alpha, \beta, n = 1 \cdots 8 \) and the sum is limited in the magnetic Brillouin zone. \( \phi^n_{\alpha}(\mathbf{k}) \) is the \( \alpha \)-th component of the eigenvector for the mean field Hamiltonian with eigenvalue \( E^n_{\mathbf{k}} \). The mean field Hamiltonian matrix is of the following form

\[
H_{\text{MF}} = \begin{pmatrix}
\xi_{\mathbf{k}} I & M_1 & M_2 & M_3 \\
M_1 & \xi_{\mathbf{k} + \mathbf{Q}_1} I & M_2 & M_3 \\
M_2 & M_1 & \xi_{\mathbf{k} + \mathbf{Q}_2} I & M_4 \\
M_3 & M_4 & M_2 & \xi_{\mathbf{k} + \mathbf{Q}_3} I
\end{pmatrix},
\]

here \( I \) is a 2 × 2 identity matrix, \( M_1 = -\frac{U_m}{3} \begin{pmatrix} 1 & \sqrt{2} \\ \sqrt{2} & -1 \end{pmatrix} \),

\[
M_2 = -\frac{U_m}{3} \begin{pmatrix} 1 & -1 + i\sqrt{3} \\ -1 - i\sqrt{3} & \sqrt{2} \end{pmatrix} \].

The matrix \( M \) is given by

\[
M = -\frac{3}{U_m} \begin{pmatrix} 0 & M_1 & M_2 & M_3 \\
M_1 & 0 & M_2 & M_3 \\
M_2 & M_3 & 0 & M_1 \\
M_3 & M_2 & M_1 & 0
\end{pmatrix}.
\]

FIG. 2: The order parameters of magnetic ordered states at zero temperature as functions of the coupling strength \( U/|t| \) for both signs of the hopping integral. (a) \( |t| > 0 \), (b) \( |t| < 0 \).

Fig.2 shows the order parameter \( m \) as a function of \( U/|t| \) at zero temperature for both signs of the hopping integral. For negative \( t \), the magnetic order exist only for all non-zero \( U \) as a result of the nesting property of the system. While for positive \( t \), the magnetic order exists for all \( U/|t| > U_c/|t| = 7.2 \). For both sign of \( t \), the non-coplanar state is slightly more stable than the collinear state at zero temperature.

Fig.3 shows the mean field phase diagram at finite temperature for both signs of the hopping integral. At finite temperature, the gapless Fermion excitation in the collinear state will contribute more entropy than the gapped Fermion excitation in the non-coplanar insulating state. We find for sufficiently large \( U/|t| \), the collinear state will become more stable than the non-coplanar state above a critical temperature at which a first order transition between the two occurs. In the phase diagram, the first order transition line between the collinear state and the non-coplanar state end at a critical end point on the magnetic-paramagnetic transition line. For positive \( t \), the critical end point locates at \( U/|t| = 7.9 \), while for negative \( t \), it locates at a smaller value of \( U/|t| = 5.5 \). Thus for both sign of the hopping integral, the collinear state is stable only in the strong correlation regime.

It is interesting to note that the transition between the collinear state and non-coplanar state, though is of first order in nature, has almost no observable effect on the peak position and intensity of the magnetic Bragg peak. As we have explained above, the non-coplanar state has the same sets of magnetic Bragg peak as the collinear state, if the three domains of the latter as mixed with equal probability. In Fig.4, we plot the change of the magnitude of the order parameter at the collinear-non-coplanar transition as a function of \( U/|t| \). We find the change to be always positive but its absolute value is always less than 1.5% of the ordered moment for all value of \( U/|t| \) and for both signs of \( t \). Thus we conclude that the neutron scattering experiments are not sensitive to the collinear-non-coplanar transition.

However, as the system opens a full gap abruptly on the quasiparticle spectrum at the collinear-non-coplanar transition, the longitudinal resistivity is expected to increase dramatically. Below the transition point, the system will enter the true insulating state. Thus in this system, the metal-insulator transition occurs in a two-step
manner. The quarter-filled system first breaks the lattice symmetry by a two-fold enlargement of the unit cell and enters a half-filled quasi-one dimensional metallic state at the paramagnetic-magnetic transition and then reduces its symmetry further by again a two-fold enlargement of the unit cell at the collinear-non-coplanar transition and enters a integer-filled insulating state. It is interesting to note that such a multi-step transition behavior is not limited to the quarter-filled system. At a more general filling fraction $\frac{1}{2q}$, the system can first enter a $\frac{1}{p}(\frac{1}{q})$-filled intermediate metallic phase by a $q(p)$-times enlargement of unit cell and then arrive at the integer-filled true insulating state by a second transition with a $p(q)$-times enlargement of the unit cell.

FIG. 3: The finite temperature phase diagram of the model for both signs of the hopping integral. (a) $t > 0$, (b) $t < 0$.

FIG. 4: The jump of the order parameter at the collinear-non-coplanar transition point as a function of $U/|t|$ for both signs of the hopping integral.

In the intermediate state between 53K and 88K, the collinear spin ordering induces dimensional reduction in the charge transport. In fact, all metal-insulator transition can be viewed as a dimensional reduction process in the charge sector: an insulator can be viewed as a system in which the charge transport is quenched in all dimension. Such a dimensional reduction process in the charge sector can occur either directly, or like our example, in a multi-step manner in spatial dimension larger than 2. In the intermediate state, the charge transport is quenched in some dimension but remain active in other dimension. Such dimensional reduction in the bulk of the system can occur in condensed matter system with multiple low energy degree of freedoms. One famous example is the orbital ordering induced dimensional reduction in ruthenium oxides in which the electron hopping is blocked except in one direction as a result of the symmetry property of the electron orbital wave function. Another example is the stripe-like structure in some cuprates superconductors in which the charge motion becomes effectively one dimensional as a result of microscopic phase separation.

Finally, we discuss the relevance of our results to the physics of the $Na_{0.5}CoO_2$ system. The quasi-one dimensional metallic state with collinear magnetic order has the proper characteristics to account for the experimental observations between 53K and 88K. Thus it is not unreasonable to assign this state to the intermediate temperature region. However, as our model is purely two-dimensional, additional three dimensional coupling should be invoked to explain the finite temperature existence of the magnetic order in the experiment. Here we assume that such three dimensional coupling will not alter the essential physics discussed in this paper. Furthermore, as our treatment of the model is limited to the mean field level, it is important to know how fluctuation effect will change the results. We leave this important issue to future study. As to the insulating state below 53K, we note that in real system, the perfect hexagonal symmetry may be broken by perturbation such as the sodium order potential, or spontaneously through the Peries instability process of the quasi-one dimensional intermediate state, the real system have many different ways to become a true insulator. It is important to make sure that the non-coplanar state studied in this paper is stable with respect to these perturbations before assigns it as the true ground state of the $Na_{0.5}CoO_2$ system.

T. Li is supported by NSFC Grant No. 10774187, National Basic Research Program of China No. 2007CB925001 and Beijing Talent Program.

[1] K. Takada et al., Nature, 422 53(2003).
[2] I. Terasaki, Y. Sasago, and K. Uchinokura, Phys. Rev. B 56, R12685 (1997).
[3] Maw Lin Foo et al., Phys. Rev. Lett. 92, 247001(2004).
[4] R. Ray, A. Ghoshray, K. Ghoshray, and S. Nakamura, Phys. Rev. B 59, 9454 (1999).
[5] Y. Wang, N. S. Rogado, R. J. Cava, and N. P. Ong, Nature 423, 425 (2003).
[6] G. Gasparovic, R. A. Ott, J.-H. Cho, F. C. Chou, Y. Chu, J. W. Lynn, and Y. S. Lee, Phys. Rev. Lett. 96, 046403(2006).
[7] N. L. Wang et al., Phys. Rev. Lett. 93,147403(2004).
[8] F. L. Ning et al., Phys. Rev. Lett. 100, 086405 (2008).
[9] $\mathbf{b}_1 = \frac{2\pi}{\sqrt{3}}(0, 1)$ and $\mathbf{b}_2 = \frac{2\pi}{\sqrt{3}}(1, -1/2)$ are the two reciprocal lattice vectors of the triangular lattice.
[10] G. Baskaran, Phys. Rev. Lett 91, 097003 (2003).
[11] N. Zhou and Z. Wang, Phys. Rev. Lett. 98,226402(2006).
[12] M.Z.Hasan et al., Phys. Rev. Lett. 92,246402(2004).
[13] I. Martin and C. D. Batista, Phys. Rev. Lett. 101, 156402 (2008).
[14] J. M. Tranquada et al., Nature (London) 375, 561 (1995).
[15] S. Lee et al., Nature Materials 5, 471 (2006).
