Coupled spin-charge order in frustrated itinerant triangular magnets

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We uncover four new spin-charge ordered ground states in the strong coupling limit of the Kondo lattice model on triangular geometry. Two of the states at one-third electronic filling \((n = 1/3)\) consist of decorated ferromagnetic chains coupled antiferromagnetically with the neighboring chains. The third magnetic ground state is noncollinear, consisting of antiferromagnetic chains separated by a pair of canted ferromagnetic chains. An even more unusual magnetic ground state, a variant of the 120° Yafet-Kittel phase, is discovered at \(n = 2/3\). These magnetic orders are stabilized by opening a gap in the electronic spectrum: a “band effect”. All the phases support modulations in the electronic charge density due to the presence of magnetically inequivalent sites. In particular, the charge ordering pattern found at \(n = 2/3\) is observed in various triangular lattice systems, such as, 2H-AgNiO\(_2\), 3R-AgNiO\(_2\) and Na\(_2\)CoO\(_2\).

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The influence of conduction electrons on the behavior of a system of localized magnetic moments is a well-studied topic in solid state physics. Investigations of such spin-charge coupled systems have given rise to a number of key concepts in magnetism and transport, such as, the RKKY interactions, the Kondo effect and the double-exchange (DE) mechanism. These concepts are commonly invoked in order to understand magnetism and charge transport in materials ranging from dilute magnetic semiconductors to various transition metal oxides and heavy fermion compounds. In recent years, it has been realized that the geometry of the underlying lattice plays a crucial role in determining the nature of magnetic states in such systems. In particular, geometrically frustrated lattices support unusual non-collinear and even non-coplanar spin-textures in the ground states. The electronic response is dramatically affected by these unusual spin-textures, exhibiting remarkable phenomena such as colossal magnetoresistance, anomalous and quantum anomalous Hall effects, and multiferroicity. As a result of such diversity of phenomena associated with unusual spin textures, their search in models, materials and artificial structures has become a very active field of research.

The starting point for a theoretical analysis of the interplay between spin-charge coupling and magnetic frustrations is the Kondo-lattice model (KLM) on various frustrated geometries. In the limit of weak Kondo coupling, the shape of Fermi surface can play a crucial role in determining the magnetic ground state. Moreover, a perturbative expansion of free energy to various orders in Kondo-coupling can be used to derive effective magnetic Hamiltonians. However, in strong coupling limit, the relevance of a non-interacting Fermi surface or that of a perturbative effective Hamiltonian in determining magnetic ground states is less clear. Nevertheless, there are many examples where the magnetic order in the strong coupling limit turns out to be the same as that in the weak coupling limit.

The focus of this letter is the strong coupling limit of the KLM on triangular lattice. We establish the presence of four exotic spin-charge ordered ground states at filling fractions of \(n = 1/3\) and \(n = 2/3\). Two of these phases are collinear, and consist of decorated ferromagnetic chains. The other two phases are noncollinear (NC), of which one can be visualized as AFM chains separated by a pair of canted-FM chains. The other NC phase is similar to 120° state, except that it consists of three type of spin triangles. An inequivalence between the lattice sites is induced by the peculiar spin ordering, causing an ordering of the electronic charge density. While the charge modulations are weak for phases at \(n = 1/3\), a strong charge ordering is found at \(n = 2/3\) with an ordering pattern similar to that observed in experiments on various triangular lattice systems. All the magnetic phases are insulating with a gap of the order of hopping parameter. We show that two of the four phases are further stabilized by Coulomb repulsions. The existence of such novel spin-charge orderings in a realistic model could guide the experimental search for unusual magnetic ordering phenomena.

We start from a strong coupling KLM, which reduces to a double exchange (DE) model with antiferromagnetic (AFM) exchange between the localized moments. Assuming the local moments to be classical, we directly write down the Hamiltonian on the triangular lattice as,

\[
H = - \sum_{\langle ij \rangle} t_{ij} \left( c_i^\dagger c_j + H.c. \right) + J_{AF} \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j, \tag{1}
\]

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where $c_i$ ($c_i^\dagger$) is the usual annihilation (creation) operator for electron with spin parallel to the local magnetic moment $S_i$. The angular brackets in the summations denote the nearest neighbor (nn) pairs of sites on a triangular lattice. $J_{AF}$ is the strength of AF coupling between nn localized spins. Note that $t_{ij}$ depend on the polar and azimuthal angles $\{\phi_i, \theta_i, \phi_j, \theta_j\}$ of the nn core spins, and are given by $t_{ij} = t_0 [\cos(\phi_j/2) \cos(\theta_j/2) + \sin(\phi_j/2) \sin(\theta_j/2) e^{-i(\phi_j-\phi_i)}]$. The parameters of the model are, the hopping amplitude $t_0$, the AF coupling $J_{AF}$ and the electronic filling fraction $n$. We set $t_0 = 1$ as the reference energy scale.

The model is investigated using the state of the art Monte Carlo (MC) method which combines the classical MC for spins with numerical diagonalization for fermions [31]. The solution of a fermionic problem is carried out numerically at each MC update step in order to obtain the electronic contribution to the total energy of a given classical spin configuration. We have used $6 \times 6$ and $12 \times 12$ clusters for this study, with typically $10^5$ MC steps for equilibration and an equal number of steps for averaging. The results obtained on these small clusters provide us with important clues about the nature of magnetic states. The energies of the candidate states are computed on much larger lattices in order to rule out any finite-size effects.

The important physical quantity that contains information about the nature of magnetic ordering is the spin structure factor $(S(q))$, which is defined as,

$$S(q) = \frac{1}{N^2} \sum_{ij} \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle_{av} e^{-iq\cdot(r_i-r_j)}.$$  

(2)

In the above $\langle (...) \rangle_{av}$ denotes the thermal or MC average, $N$ is the number of lattice sites, and $r_i, r_j$ are the position vectors of sites $i, j$. We begin by discussing the spin structure factor results obtained from simulations on $12 \times 12$ lattice. Fig 1 (a) and (b) display the results at low temperature, $T = 0.002t_0$, for filling fractions $n = 2/3$ and $n = 1/3$ respectively. The magnitude of $S(q)$ is indicated by the radius of the empty circles, and the $q$ values are restricted to the 1st Brillouin zone. In the low-$J_{AF}$ limit, the $S(q)$ peaks at the $\Gamma$ point indicating a ferromagnetic ground state, which is expected in the DE model. For $n = 1/3$, the peak at the $\Gamma$ point remains robust in the range $0 \leq J_{AF} < 0.08$. For $J_{AF} = 0.10$, we find two peaks in the $S(q)$ (see Fig 1(b)), one at the $M$-point and other on the $\Gamma$-$K$ axis. For $J_{AF} = 0.16$, $S(q)$ indicates the presence of another unusual magnetic phase with peaks at multiple $q$ points. We confirm by looking at the spin configurations in real space that both these phases are collinear. The $S(q)$ at $J_{AF} = 0.23$ is qualitatively different, indicating the appearance of yet another magnetic order. We will discuss the nature of these phases in detail later. At this point, we emphasize these magnetic phases have not been reported in any of the previous studies on Kondo lattice model on triangular lattice. Also, the plots at different values of $J_{AF}$ are only representative of different phases. The stability range of these phases will become clear when we discuss the phase diagrams.

For $n = 2/3$, the presence of another unusual magnetic order in the coupling range $0.04 < J_{AF} < 0.20$ is inferred from the $S(q)$. This phase is characterized by two peaks in the $S(q)$ at the $K$ and $M$ points (see Fig 1(a) plotted for $J_{AF} = 0.16$). It is also clear from the structure factor plots that all the new phases discussed above break the three-fold rotational symmetry of the triangular lattice. In order to further probe the nature of electronic states in these new magnetic phases, we compute charge structure factor defined as,

$$C(q) = \frac{1}{N^2} \sum_{ij} \langle \delta n_i \delta n_j \rangle_{av} e^{-iq\cdot(r_i-r_j)},$$  

(3)

where $\delta n_i = n_i - n$ is the charge density modulation w.r.t. the average charge density $n$. The $C(q)$ plots in Fig 1 (c) show that all the magnetic phases discussed exhibit charge ordering. For the phases at $n = 1/3$, the magnitude of charge disproportionation is small, and the ordering pattern is stripe-like. However, for the NC magnetic phase at $n = 2/3$, the charge ordering is strong in magnitude, and has a pattern similar to the one observed in various triangular lattice systems with active spin degree of freedom, such as, 2H-AgNiO$_2$, 3R-AgNiO$_2$ and Na$_x$CoO$_2$ [27, 30]. Typically a CO state arises either due to Coulomb repulsions at appropriate filling fractions, or due to charge-lattice couplings [32]. Therefore, it is unusual that charge ordering emerges in a model consisting of local charge-spin coupling. Indeed, this was emphasized in a recent work reporting the presence of an un-
usual spin-charge ordered state in KLM [33].

We now discuss in detail the phase diagram of the model at \( n = 1/3 \). In Fig. 2(a), we plot the ground state energy for different \( J_{AF} \) values obtained from MC simulations. Looking at the low-T spin configurations from the simulations, we infer the nature of magnetic ground states for different \( J_{AF} \). The straight lines in Fig. 2(a) correspond to the energy obtained for ideal long-range ordered spin arrangements. For small \( J_{AF} \), the MC energies fall on the straight line corresponding to a FM phase, as expected. Similarly, in the limit of large \( J_{AF} \) the MC energies match well with those of the 120° Yafet-Kittel states, respectively. The lower inset shows the snap-shot of the MC ground state with the arrows representing the spin directions and the circle sizes indicating the local charge density. The smaller circles have been filled to highlight the pattern of charge ordering.

In order to visualize these phases more clearly, we have connected all the ferromagnetically oriented spins via solid lines. This highlights the main feature of DS1 (see Fig. 2(c)), that this phase consists of diamond shaped FM chains running along one direction connected antiferromagnetically to the neighboring spins. Similarly, the DS2 phase consists of FM stripes decorated by triangular units (see Fig. 2(d)). In the strong Kondo coupling limit, the electronic hopping across a pair of sites hosting antiferromagnetically oriented spins is zero. Therefore, in the DS1 and DS2 phases, the electronic problem becomes one-dimensional. The electronic density of states (DOS) in the decorated stripe phases has large gaps at the chemical potential (see Fig. 2(b)). Opening these gaps in the DOS lowers the total energy of the system and hence these unusual phases are stabilized. Such decorated stripe paths for hopping are realized in certain organic polymers [34, 35], and are also of interest to researchers working on exactly solvable models of electrostatic correlations \[ 36, 37 \]. Interestingly, such novel structures for fermion hopping can emerge in a higher dimensional lattice via a subtle interplay between geometrical frustrations and spin-charge coupling.

The two phases discussed so far are collinear in nature and therefore allow for a description in terms of FM chains. The third new phase at \( n = 1/3 \) is NC, and consists of AF chains separated by a pair of canted FM chains. This spin arrangement also opens a gap in the electronic DOS at the chemical potential. All the phases discussed above contain inequivalent sites in terms of the orientation of neighboring spins. This causes a modulation in the local charge density, and indeed we find a charge-ordering in all the phases (see Fig. 2(c)-(e)). For opening a gap in the spectrum, that we find in all the three phases discussed above, the entire magnetic structure must be modified. This can be seen as a “band effect”. It is also interesting to note that the DS1 and DS2 spontaneously break a discrete rota-
the magnitude of the AF state. However, beyond a critical value of \( J = 2 \Delta \) and \( J = \Delta \), we obtain the Yafet-Kittel (YK) phase. We note that in the range \( 0.26 < J_{AF} < 0.36 \) the energy of another unusual spin-charge ordered state is close to that obtained from MC simulations. In fact, the same state dominates the phase diagram at \( n = 2/3 \) which we discuss next.

In Fig. 4 we show the low-T MC energy for different values of \( J_{AF} \) at \( n = 2/3 \). Following the analysis at \( n = 1/3 \), we compare the MC energies with those obtained for ideal ordered spin structures. We require only three phases to perfectly describe the MC energy data across the full \( J_{AF} \) range. Two of these phases are the expected limiting phases: a ferromagnet at small values of \( J_{AF} \) and a 120° YK phase at large \( J_{AF} \). The entire intermediate range belongs to another exotic spin-charge ordered phases. A MC snap-shot of this magnetic phase at low temperature is shown in inset in Fig. 4. The spin structure remains planar, as in the 120° phase. In fact, for a specific choice of global orientation, all spins are pointing towards the neighboring sites, which is also similar to the 120° phase. The important difference is that in this new phase, there are three different type of triangles, as shown in the real-space plot in Fig. 4. First type is the usual 120° orientation, the second type is formed with two anti parallel spins with the third one pointing at 60°. The third type of triangle can be obtained from the second type by flipping the spins. Similar to other magnetic phases discussed so far, this magnetic arrangement also generates inequivalent sites in terms of the hopping amplitudes. This inequivalence is reflected via a charge ordering pattern (see inset in Fig. 4) that closely resembles the charge modulations observed in various triangular lattice materials [27, 30]. The electronic DOS in this magnetic phase supports two gaps (see inset in Fig. 4), corresponding to filling fractions of \( n = 2/3 \), and \( n = 1/3 \), thereby justifying the existence of the NC-CO phase at both filling fractions.

In order to test the stability of these unusual spin-charge ordered phases in the presence of electron-electron interactions, we add to the Hamiltonian Eq. (1) a nn repulsive interaction, \( H_1 = V \sum_{\langle i,j \rangle} n_i n_j \). An unrestricted Hartree-Fock analysis is performed by keeping the magnetic order fixed. The \( C(q) \) is then computed for the self-consistent solutions for local charge densities. We plot in Fig. 4 the magnitude of the \( C(q) \) at characteristic values of \( q \) as a function of \( V \) for each of the four ordered phases. Two of the phases at \( n = 1/3 \), DS2 and C-AF, are not affected by the nn repulsive interaction (see Fig. 4 (a)). However, beyond a critical value of \( V \), these phases are destabilized in favor of the expected charge ordered phase consisting of a high density site surrounded by low density sites. On the contrary, the structure factors for DS1 and NC-CO states increase with increasing \( V \), indicating that both these states are further stabilized by a nn repulsive interaction.

To conclude, we have reported four new spin-charge ordered phases at filling fractions of \( n = 1/3 \) and \( n = 2/3 \) in the strong coupling KLM on a triangular lattice. Two of these phases are collinear and stripe-like in their magnetic arrangement, and the other two are noncollinear. Presence of magnetically inequivalent sites leads to charge ordering in all the phases. The charge ordering pattern for the noncollinear phase at \( n = 2/3 \) is identical to that observed in various triangular lattice materials with active spin degree of freedom, such as, 2H-AgNiO\(_3\), 3R-AgNiO\(_2\) and Na\(_2\)CoO\(_2\) [27, 30]. The inclusion of a nn Coulomb interaction enhances the charge ordering further, indicating that mutually supportive mechanisms could be involved in stabilizing such ordering in real materials. All the states reported in this work break the discrete rotational symmetry of the triangular lattice via their charge and spin arrangements. This may lead to interesting states with charge-spin nematicity at finite temperatures. The stability of these novel states relies on the nature of the electronic spectrum for itinerant fermions, which develops a gap at the chemical potential. Consequently, all the new phases reported in this study are electrically insulating with gap of the order of bare hopping amplitude. These insulators can neither be called Slater-type nor Mott-type since the opening of gap cannot be understood from the Fermi surface nesting arguments nor from the infinite coupling limit. Therefore, such exotic spin-charge ordered insulators are prototype examples of cooperative many-body effects which are not easy to understand within effective single particle theories.

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