Coupled spin-charge order in frustrated itinerant triangular magnets

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We uncover four spin-charge ordered ground states in the strong coupling limit of the Kondo lattice model on triangular geometry. The results are obtained using Monte Carlo simulations, with a classical treatment of localized moments. 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 2\( H \)-AgNiO\(_2\), 3\( R \)-AgNiO\(_2\), and Na\(_4\)CoO\(_2\).

The focus of this Rapid Communication 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 (FM) chains. The other two phases are noncollinear (NC), of which one can be visualized as antiferromagnetic (AF) chains separated by a pair of canted-FM chains. The other NC phase is similar to the 120° state, except that it consists of three types 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 [27–33]. We show that two of the four phases are further stabilized by Coulomb repulsions. The existence of such spin-charge orderings in a realistic model could guide the experimental search for unusual magnetic ordering phenomena.

We start from the KLM consisting of localized moments coupled to itinerant fermions. Assuming the moments to be classical, the model in the strong coupling limit reduces to a DE model with additional AF exchange [15,34–36]. The resulting Hamiltonian on the triangular lattice is

\[
H = -\sum_{(ij)} (t_{ij} c_i^\dagger c_j + \text{H.c.}) + J_{AF} \sum_{ij} \textbf{S}_i \cdot \textbf{S}_j,
\]

where \( c_i \) (\( c_i^\dagger \)) is the usual annihilation (creation) operator for electron with spin parallel to the local magnetic moment \( \textbf{S}_i \). 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 \( \{\theta_i, \phi_i, \theta_j, \phi_j\} \) of the NN core spins, and are given by \( t_{ij} = t_0 [\cos(\theta_i/2) \cos(\theta_j/2) + \sin(\theta_i/2) \sin(\theta_j/2) \cos(\theta - \phi)] \) [34]. Previous studies show that the classical approximation is a good starting point, unless the localized moments are spin \( \frac{1}{2} \) [37,38]. 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 [39]. 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 \( 6^2 \) and \( 12^2 \) clusters with periodic boundary conditions and typically \( 10^4 \) MC steps for equilibration and averaging. While the MC on larger lattices is not feasible due to computational costs, the energies of candidate states have been compared on \( 120^2 \) sites [40].

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

\[
S(q) = \frac{1}{N^2} \sum_{ij} (S_i \cdot S_j)_{av} e^{-i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)}.
\]

In the above, \( \langle \cdots \rangle_{av} \) denotes the thermal or MC average, \( N \) is the number of lattice sites, and \( \mathbf{r}_i, \mathbf{r}_j \) are the position vectors of sites \( i, j \). We begin by discussing the spin structure factor results obtained from simulations on a \( 12 \times 12 \) lattice. Figures 1(a) and 1(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 open circles, and the \( q \) values are restricted to the first 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 the 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. 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 different phases discussed above break the threefold rotational symmetry of the triangular lattice. In order to further probe the nature of electronic states in these magnetic phases, we compute the charge structure factor, defined as

\[
C(q) = \frac{1}{N^2} \sum_{ij} \langle \delta n_i \delta n_j \rangle_{av} e^{-i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)},
\]

where \( \delta n_i = n_i - n \) is the charge density modulation with regard to the average charge density \( n \). The \( C(q) \) plots in Fig. 1(c) show that all the magnetic phases discussed above exhibit charge ordering. For the phases at \( n = 1/3 \), the magnitude of charge disproportionation is small, and the ordering pattern is stripelike. 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 an active spin degree of freedom, such as \( \text{HfAgNiO}_2 \), \( \text{YbAgNiO}_2 \), and \( \text{Na}_3\text{CoO}_2 \) [27–33]. Typically, a CO state arises either due to Coulomb repulsions at appropriate filling fractions, or due to charge-lattice couplings [41]. 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 unusual spin-charge ordered state in KLM [42].

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 \( 120^\circ \) Yafet-Kittel (YK) phase. We require four additional magnetic phases in order to fit the MC data in the intermediate \( J_{AF} \) regime. The spin arrangement for three of these magnetic phases are shown in Figs. 2(c)–2(e). We label two of these phases as decorated stripes (DS1 and DS2), and the third as canted AF (C-AF). The fourth phase will be discussed later, as it turns out to be the dominant phase at \( n = 2/3 \).

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
and therefore allow for a description in terms of FM chains. Interestingly, such structures for fermion hopping can emerge as a "band effect." It is exactly solvable models of electronic correlations [45,46], and are also of interest to researchers working on these unusual phases are stabilized. Such decorated stripe phases have large gaps at the chemical potential [see Fig. 2(b)]. The opening of these gaps in the DOS lowers the total energy of the system and hence causes a modulation in the local charge density, and indeed we find a charge ordering in all the phases [see Figs. 2(c)–2(e)]. For opening a gap in the spectrum, which we find in all the phases discussed so far, 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 rotational symmetry. Since there cannot be any magnetic order at finite temperatures in a rotationally invariant continuous spin model [47], it may give rise to an interesting nematic magnetic order at finite temperatures. These magnetic phases exist in the wide parameter range $0.08 < J_{AF} < 0.26$. For larger values of $J_{AF}$ the magnetic order gradually changes towards the 120° 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. 3, 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 patterns. 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 phase. A MC snapshot of this magnetic phase at low temperature is shown in the inset in Fig. 3. 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 phase, there are three different types of triangles, as shown in the real-space plot in Fig. 3. The first type is the usual 120° orientation, the second type is formed with two antiparallel 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 the inset in Fig. 3) that closely resembles the charge modulations observed in various triangular lattice materials [27–30]. In particular, a state with six magnetically inequivalent Co ions has recently been observed in NMR experiments on Na$_2$CoO$_2$ [31, 32]. The electronic DOS in this magnetic phase supports two gaps (see the inset in Fig. 3), 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, $H_1 = V \sum_{\langle ij \rangle} n_i n_j$. An unrestricted Hartree-Fock analysis is performed by keeping the magnetic order fixed [40]. 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 spin-charge ordered phases at filling fractions of $n = 1/3$ and $n = 2/3$ in the strong coupling KL$_M$ on a triangular lattice. Two of these phases are collinear and stripelike in their magnetic arrangement, and the other two are noncollinear. The 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 an active spin degree of freedom, such as 2H-AgNiO$_2$, 3R-AgNiO$_2$, and Na$_2$CoO$_2$ [27–33]. 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. The stability of these states relies on the nature of the electronic spectrum for itinerant fermions, which develops a gap at the chemical potential. Consequently, all the phases reported in this study are electrically insulating with a gap of the order of the bare hopping amplitude. These insulators can neither be called Slater type nor Mott type since the opening of the gap can neither 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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