

Frustration-induced insulating chiral spin state in itinerant triangular-lattice magnets

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We study the double-exchange model at half-filling with competing superexchange interactions on a triangular lattice, combining exact diagonalization and Monte-Carlo methods. We find that in between the expected itinerant ferromagnetic and 120° Yafet-Kittel phases a robust scalar-chiral, insulating spin state emerges. At finite temperatures the ferromagnet - scalar-chiral quantum critical point is characterized by anomalous bad-metal behavior in charge transport as observed in frustrated itinerant magnets $R_2\text{Mo}_2\text{O}_7$.

Geometric frustration is encountered in numerous magnetic condensed matter systems and gives rise to a variety of fascinating magnetic phases, such as spin-glass, spin-liquid, and spin-ice [1, 2]. Traditionally geometrically frustrated magnetism is studied in the context of wide bandgap materials with localized spins. Recently the interest broadened to frustrated magnets with metallic character such as $\text{Tl}_2\text{Mn}_2\text{O}_7$ and $\text{R}_2\text{Mo}_2\text{O}_7$ (R: rare earth ion) [3–6]. In these metallic systems, frustration in the magnetic sector has a strong impact on charge dynamics, often resulting in heavy-fermion type behavior in transport [7–10].

Besides being of geometric origin, magnetic frustrations can also be due to a direct competition between ferromagnetic and antiferromagnetic interactions. Perhaps the most elementary example is ferromagnetic (FM) double-exchange competing with antiferromagnetic (AFM) superexchange [11]. Magnetic competition of this kind has been studied extensively on hyper-cubic lattices in the context of the colossal magnetoresistance manganites, where depending on carrier concentration the competing interactions can induce new forms of magnetic order or phase-separation [12–14]. Competing FM and AFM can in principle also lead to non-collinear and even non-coplanar magnetic states, which are raising interest in the rapidly emerging and seemingly disparate fields of multiferroics and topological insulators [15, 16].

In an interesting class of materials both types of frustrations – geometric frustration *and* competing double- and super-exchange interactions – are present and give rise to a set of intriguing physical properties [17, 18]. Recent experiments on pyrochlore $\text{R}_2\text{Mo}_2\text{O}_7$, for instance, show a transition from a ferromagnetic metallic to a spin glass insulating state as R changes from Nd to Dy [8]. This transition can be controlled by external pressure. In the vicinity of the transition an unusual diffusive metallic state appears, showing a temperature independent resistivity down to very low temperatures [8]. A similar competition between FM and AFM interactions is also relevant in the triangular lattice systems such as GdI_2 and H-doped GdI_2 [19–22].

In this letter, we study in a wide temperature range the double-exchange (DE) model on a triangular lattice in the presence of frustrating antiferromagnetic superex-

change (SE) interactions. Three magnetically ordered groundstates are present in the phase diagram: an itinerant ferromagnetic state on one end, the 120° Yafet-Kittel phase on the other and an insulating non-coplanar scalar chiral (SC) state in between [23]. Apparently the SC insulating state emerges from the competition between the two metallic phases: FM and 120° . The FM and SC state are separated by a quantum critical point (QCP), whereas the transition from the SC to the 120° phase is first order in nature. At finite temperature in the vicinity of the QCP the system is characterized by a temperature-independent resistivity and a linear inverse magnetic susceptibility, down to very low temperatures. This bad metallic behavior closely resembles the experimental observations on the $\text{R}_2\text{Mo}_2\text{O}_7$ compounds.

We consider the elementary one-band DE Hamiltonian in the presence of antiferromagnetic SE interactions, introduced by De Gennes in the 1960's [11] – but now on a frustrated triangular lattice. The full Hamiltonian is

$$H = - \sum_{\langle ij \rangle} t_{ij} (c_i^\dagger c_j + H.c.) + J_S \sum_{\langle ij \rangle} \mathbf{S}_i \cdot \mathbf{S}_j, \quad (1)$$

where c_i and c_i^\dagger are annihilation and creation operators for electrons with spin parallel to the core spin \mathbf{S}_i . $\langle ij \rangle$ denotes the nearest neighbor (nn) pairs of sites on a triangular lattice. J_S denotes the strength of AF coupling between nn core spins. $t_{ij} = t[\cos(\theta_i/2)\cos(\theta_j/2) + \sin(\theta_i/2)\sin(\theta_j/2)e^{-i(\phi_i - \phi_j)}]$ denote the hopping amplitudes which depend on the polar and azimuthal angles $\{\theta_i, \phi_i, \theta_j, \phi_j\}$ of the nn core spins due to the double-exchange mechanism. All energies are measured in units of the hopping parameter t . The core spins are classical unit vectors and we focus on the case of half filling.

The only unbiased method to study this model is the hybrid scheme involving exact-diagonalization (ED) of the fermion problem and Monte Carlo (MC) for classical core spins. The difficulty in handling this class of models and details on ED+MC method has been well documented in the context of manganites [12]. In order to achieve larger lattice sizes, which are essential for computing transport properties, we employ the traveling cluster approximation (TCA) [24]. This method has

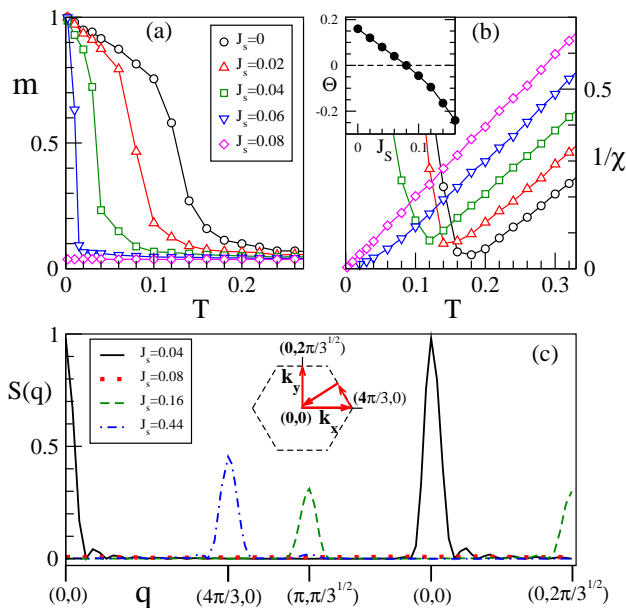


FIG. 1: (Color online) (a) Magnetization and (b) inverse magnetic susceptibility as a function of temperature for various values of superexchange coupling J_S . The inset in (b) shows the Curie-Weiss scale extracted from the $\chi^{-1}(T)$ as a function of J_S . Note that $\chi^{-1} \propto T$ down to $T = 0$ for $J_S = 0.08$. (c) Spin-structure factor $S(\mathbf{q})$ along the principal symmetry directions in the momentum space for different values of J_S . The inset shows the Brillouin zone for a triangular lattice and the path along which $S(\mathbf{q})$ is plotted.

proved very successful in studying DE models on square and cubic lattices relevant for manganites [24, 25]. Since the TCA does not rely on the lattice geometry, we extend the framework to triangular lattices. Most of the results in this work are obtained on lattices with $N = 24^2$ sites using a cluster size $N_c = 6^2$. Typically $\sim 10^4$ MC steps are used for equilibration and a similar number of steps for computing thermal averages on classical spin variables. For electronic properties, which still requires the ED of the full Hamiltonian, $\sim 10^3$ steps are used.

Fig. 1(a) shows the calculated temperature dependence of magnetization $m = \sqrt{\langle (1/N \sum_i \mathbf{S}_i)^2 \rangle_{av}}$, where $\langle \dots \rangle_{av}$ denotes thermal averaging. In the absence of J_S we find a FM groundstate with a Curie temperature $T_C = 0.15$ as inferred from the inflection point in the $m(T)$ curve. Upon increasing J_S a monotonic reduction in T_C is observed while the groundstate continues to be a saturated FM. For $J_S = 0.08$ the ferromagnetism is destroyed by the competing AFM interactions and we do not find any long-range order in the groundstate. In Fig. 1(b) we show the inverse of magnetic susceptibility computed as $\chi = T^{-1}(\langle m^2 \rangle_{av} - \langle m \rangle_{av}^2)$. The inverse susceptibility χ^{-1} clearly follows a Curie-Weiss (CW) behavior ($\chi \propto 1/(T - \Theta)$) above a characteristic temperature. The CW scale Θ , which is obtained by extrapolating the linear high-temperature behavior of χ^{-1} , is shown in the inset in Fig. 1(b). Θ reduces monotonically and changes

sign upon increasing J_S , indicating a change in the nature of effective magnetic coupling from FM to AFM. Typically a small value for ordering-temperature to Θ ratio is an indicator of the frustrations in a magnetic system. Here we encounter an unusual situation where both the ordering temperature and Θ are vanishing. A remarkable similarity of these results with those obtained on a pyrochlore lattice indicates that the essential physics is determined by the frustrated nature of the lattice [18].

In order to identify non-trivial long-range ordered magnetic phases we compute the spin structure factor, $S(\mathbf{q}) = 1/N^2 \sum \langle \mathbf{S}_i \cdot \mathbf{S}_j \rangle_{av} e^{i\mathbf{q} \cdot (\mathbf{r}_i - \mathbf{r}_j)}$, where $\mathbf{r}_i, \mathbf{r}_j$ are the real-space location of spins $\mathbf{S}_i, \mathbf{S}_j$. We show the ground-state structure factor along the symmetry directions in the momentum space in Fig. 1(c). For $J_S = 0.04$, the $S(\mathbf{q})$ has a single peak at $\mathbf{q} = (0, 0)$ as expected for a FM state. In the regime $\Theta \sim 0$ ($J_S = 0.08$), the structure factor has no peak at any \mathbf{q} suggesting the absence of any long-range magnetic order in this regime. For $J_S = 0.16$, we find peaks at $\mathbf{q} = (\pi, \pi/\sqrt{3})$ and $(0, 2\pi/\sqrt{3})$. This corresponds to a four-sublattice ordered state with a non-vanishing scalar chirality $\kappa = \sum \langle \mathbf{S}_i \cdot \mathbf{S}_j \times \mathbf{S}_k \rangle_{av}$, where the sum is over indices forming a triangle taken in the counterclockwise order. The onset temperature T_{sc} for the SC state can be inferred from the temperature-dependence of κ . This state has been discussed for the Kondo-lattice model within mean-field and variational schemes, and has also been studied in the context of high- T_c superconductors and recently the topological insulators [16, 26]. Here we show for the 1st time that the SC state is in fact the groundstate for the model Hamiltonian Eq. (1) over a window of phase space. Finally as the J_S becomes large the system should approach towards the classical 120° state. This is seen in the $S(\mathbf{q})$ data for $J_S = 0.44$, where the peak is located at $\mathbf{q} = (4\pi/3, 0)$.

To investigate the repercussions of the competing magnetic interactions on electronic properties, we first focus on the evolution of the electronic density of states (DOS) with J_S . The DOS is computed as $D(E) = \langle \sum_k \delta(E - \epsilon_k) \rangle_{av}$, where we approximate the δ -function by a Lorentzian of width γ : $\pi \delta(E - \epsilon_k) \sim \gamma / [\gamma^2 + (E - \epsilon_k)^2]$. The DOS for different temperatures and values of J_S is shown in Fig. 2. For small J_S the DOS does not show any drastic changes in shape upon reducing T (see Fig. 2(a)) and an increase in bandwidth is found as expected in a DE model. Since the groundstate is a saturated FM, the low- T DOS corresponds to that of non-interacting electrons on a triangular lattice. For $J_S = 0.08$ the DOS at low temperatures is identical to that in the high temperature paramagnetic (PM) state (see Fig. 2(b)). This unusual behavior of the DOS is consistent with the facts that (i) χ^{-1} follows the Curie-Weiss behavior down to $T = 0$, and (ii) the spin-state remains disordered down to low temperatures for $J_S = 0.08$ (see Fig. 1). This observation strongly supports the possibility of finding a spin-liquid phase in this model. For $J_S = 0.16$, where the groundstate has the peculiar non-coplanar order, an energy gap appears in the DOS near E_F at low tempera-

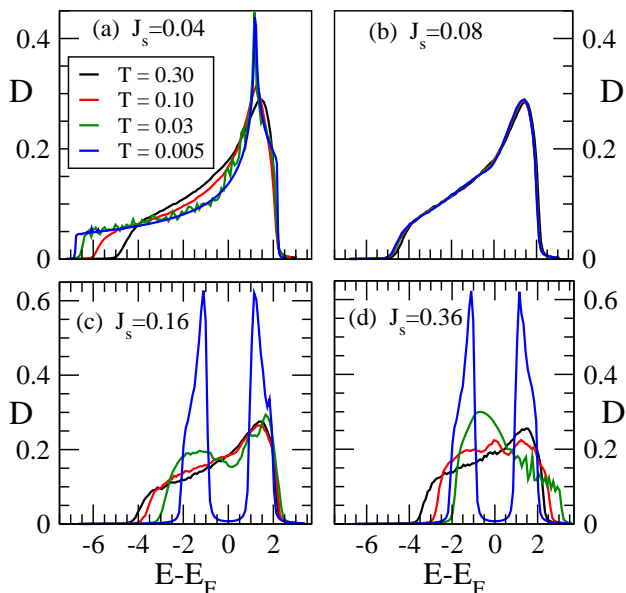


FIG. 2: (Color online) Electronic density of states for varying temperatures with (a) $J_S = 0.04$, (b) $J_S = 0.08$, (c) $J_S = 0.16$ and (d) $J_S = 0.36$. A Lorentzian broadening $\gamma = 0.04$ is used.

tures (see Fig. 2(c)). This is consistent with the previous discussions on this non-trivial state [16, 26]. The effect of this energy gap persists at higher temperatures in the form of a pseudogap feature at E_F . For larger J_S an unusual flip in the DOS shape w.r.t. the high temperature DOS occurs near $T = 0.03$ (see Fig. 2(d)). This can happen in a triangular lattice upon introducing an overall phase in the hopping [27]. For DE systems such a phase in the effective hoppings t_{ij} can be spontaneously generated at low temperatures. We find that this flip in the shape of DOS is correlated with the rise in $S(\mathbf{q})$ at $\mathbf{q} = (4\pi/3, 0)$ and hence indicates a 120° state.

We compute the dc conductivity ($\sigma(\omega)|_{\omega \rightarrow 0}$) using the Kubo-Greenwood formula and the exact eigenspectrum [28]. In a disorder-free metal the entire contribution to optical conductivity is in the coherent part at $\omega = 0$, which can not be accessed on a finite lattice due to the discrete eigenvalue spectrum. The lowest accessible frequency ω_0 decreases with increasing lattice size ($\omega_0 \propto N^{-1}$). In the presence of thermal or quenched disorder a broadening of $\sigma(\omega)$ to finite frequencies occurs and $\sigma(\omega_0)$ can be used as the dc conductivity to a good approximation. In the clean limit, the conductivity sum rule can be used to estimate the coherent part of the conductivity [29]. In this work we use $\sigma(\omega_0)|_{\omega_0=0.03}$ as an approximation to dc conductivity except for low-temperature metallic states, where we make use of the conductivity sum rule.

A typical double-exchange behavior in resistivity is found in cases where the groundstate is FM (see Fig. 3(a)). For $J_S = 0.08$, which corresponds to a disordered magnetic groundstate as seen in $S(\mathbf{q})$, we find an unusually flat resistivity. Further increase in J_S leads to an-

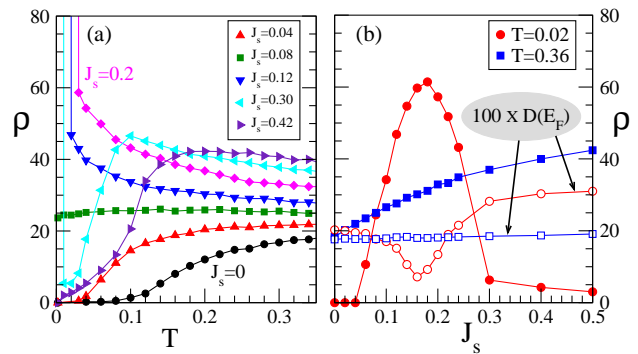


FIG. 3: (Color online) (a) Resistivity as a function of temperature for various values of J_S . (b) J_S -dependence of resistivity ρ (filled symbols) and scaled DOS (open symbols) at Fermi level E_F for low and high temperatures.

ulating behavior ($d\rho/dT < 0$) at low temperatures with a diverging resistivity at $T \sim 0.01$. For $J_S = 0.30$ one finds a sharp reduction in ρ at a characteristic temperature associated with the rise in $S(\mathbf{q})$ at $\mathbf{q} = (4\pi/3, 0)$. $d\rho/dT$ remains positive in the low temperature regime until the system enters the scalar chiral insulating state near $T = 0.005$. Eventually, beyond $J_S \sim 0.40$ the groundstate becomes the 120° -state. The evolution of $\rho(T)$ with J_S is therefore guided by the presence of an unusual insulating state for intermediate J_S . For small and large J_S the groundstates are FM-metal and 120° -metal respectively, however, for the intermediate values of J_S the SC insulating state is energetically favored. This represents an unusual situation where from the competition between two metallic states an insulating state emerges.

In Fig. 2(b) we show resistivity as a function of J_S for low and high temperatures. This data can be compared with the pressure dependence of resistivity reported in [8], since application of external pressure alter the J_S/t ratio and tunes the system across the FM to PM transition. We find the resistivity at high temperature increases monotonically, whereas at low temperatures it shows a non-monotonic behavior. These features qualitatively agree with the experimental observation. We also plot the DOS at E_F as a function of J_S for low and high temperatures. Naively at low temperatures one expects the resistivity to be inversely related to the DOS at E_F , since only states near the Fermi level contribute. We indeed find this behavior at low temperatures: the peak in ρ corresponds to a dip in $D(E_F)$.

In Fig. 4(a) we plot the peak values in $S(\mathbf{q})$ at various \mathbf{q} at $T = 0.005$ as a function of J_S . Peak in $S(\mathbf{q})$ at $\mathbf{q} = (0, 0)$ is a measure for ferromagnetism, that at $\mathbf{q} = (4\pi/3, 0)$ corresponds to 120° state and simultaneous peaks at $\mathbf{q} = (\pi, \pi/\sqrt{3})$ and $(0, 2\pi/\sqrt{3})$ indicates a SC state. Upon increasing J_S , $S(0, 0)$ vanishes before a rise in $S(\pi, \pi/\sqrt{3})$ and $S(0, 2\pi/\sqrt{3})$, indicating that the associated transition at $J_S \sim 0.07$ is quantum critical in nature. On the other hand the transition between chiral and 120° state is 1st order. The dashed curve shows the

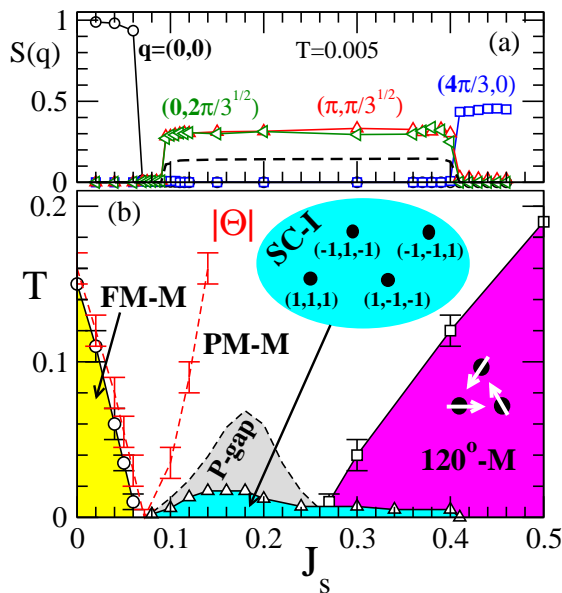


FIG. 4: (Color online) (a) Low-temperature spin structure factor $S(\mathbf{q})$ as a function of superexchange coupling J_S for various \mathbf{q} . The dashed line is the scalar chirality κ . (b) The phase diagram of the model in the temperature- J_S plane. The spin directions within a unit cell are schematically shown for the 120° state, and are indicated as un-normalized (S_x, S_y, S_z) on each site for the non-coplanar SC state.

scalar chirality κ as a function of J_S . κ is finite only

for the SC state, therefore it can also be considered an order-parameter for the SC state. The results are summarized in a phase diagram in Fig. 4(b). The ferromagnetic transition temperature (T_C) is inferred from the $m(T)$, the onset of pseudogap behavior is determined by directly looking at the DOS at various temperatures, and the transition to the 120° state is determined from the T -dependence of the $S(\mathbf{q})$ at $\mathbf{q} = (4\pi/3, 0)$. The absolute value $|\Theta|$ of the Curie-Weiss temperature is plotted as an estimate of the effective magnetic interaction in the system. The FM scale closely follows the value of Θ , indicating the unfrustrated nature of the system in this regime. In the intermediate- J_S regime the groundstate is SC-I and a pseudogap behavior appears just above T_{sc} . This simple looking model provides an elegant example where a competition between two metallic phases leads to an intermediate insulating phase.

To conclude, a highly non-trivial scalar-chiral insulating state emerges when double- and super-exchange interactions are competing on a triangular lattice. The transition from ferromagnetic metal to a scalar-chiral insulator is quantum critical in nature with both T_C and T_{sc} vanishing at a critical value of J_S . The anomalous transport and magnetic properties that we find theoretically in the vicinity of the quantum critical point are in line with experimental observations on $R_2Mo_2O_7$ [8].

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