

Resistivity Minimum in Highly Frustrated Itinerant Magnets

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We study the transport properties of frustrated itinerant magnets comprising localized classical moments, which interact via exchange with the conduction electrons. Strong frustration stabilizes a liquidlike spin state, which extends down to temperatures well below the effective Ruderman-Kittel-Kasuya-Yosida interaction scale. The crossover into this state is characterized by spin structure factor enhancement at wave vectors smaller than twice the Fermi wave vector magnitude. The corresponding enhancement of electron scattering generates a resistivity upturn at decreasing temperatures.

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Certain magnetic metals exhibit a resistivity minimum at low temperature. The Kondo effect explains this minimum via an effective exchange interaction J between magnetic impurities and conduction electrons [1]. Resistivity minima are also observed in compounds comprising a periodic array of localized magnetic moments such as $4f$ -electron compounds [2]. Because the Kondo effect is induced by spin-flip impurity scattering, it is expected to be strongly suppressed in systems with large local magnetic moments or with strong easy-axis spin anisotropy. Surprisingly, several compounds in this category, such as Gd_2PdSi_3 and RCuAs_2 ($R = \text{Sm, Gd, Tb, and Dy}$) [3–5] and RInCu_4 ($R = \text{Gd, Dy, Ho, Er, and Tm}$) [6,7], exhibit a pronounced resistivity minimum despite heavy suppression of the Kondo effect. These compounds are dominated by the Ruderman-Kittel-Kasuya-Yosida (RKKY) interaction, which competes against Kondo screening. It is natural to ask, therefore, if there exists a general mechanism by which a RKKY interaction can induce a resistivity minimum [8].

In this Letter, we answer the question affirmatively: frustrated itinerant magnets can exhibit a low- T liquidlike spin state with enhanced resistivity under quite general conditions. For simplicity, we focus on a 2D Kondo lattice model (KLM) with classical local moments (no Kondo effect) and a small Fermi surface (FS). For a circular FS, the bare magnetic susceptibility χ_k^0 of the conduction electrons has a flat area of maxima for $k \leq 2k_F$ (where $k \equiv |\mathbf{k}|$ and k_F is the magnitude of Fermi wave vectors). The RKKY interaction thus seeks to enhance the structure factor (SF) in the region $k \leq 2k_F$. We demonstrate that this effect leads to an increase of the electrical resistivity ρ upon decreasing the temperature over the window $T_0 \lesssim T \lesssim |\theta_{\text{CW}}|$, where the magnetic correlation length increases from one lattice space a (at $|\theta_{\text{CW}}|$) to $\xi \gg a$ (at T_0) [10]. Frustration

($|\theta_{\text{CW}}|/T_0 \gg 1$) is required just to open this window; the rest is done by the nature of the RKKY interaction. The average enhancement of the spin SF for wave vectors connecting points on the FS increases the elastic electron-spin scattering upon lowering T .

The effect of the RKKY interaction on electron transport was considered in Refs. [11] and [12]. The sign of the effect was found to be opposite (metallic) to that found in this Letter. This difference arises because we consider low filling, where the sign of $d\rho/dT$ can be shown to be insulating under quite general assumptions about the SF. In contrast, Refs. [11] and [12] considered a large FS, where the effect can have either sign depending on details of the electronic structure.

We first present an analytical derivation of the effect for the weak-coupling (WC) limit [$J\eta(\varepsilon_F) \ll 1$, where $\eta(\varepsilon_F)$ is the density of states at the Fermi level]. The resistivity is evaluated in the Born approximation and the spin SF is obtained in two ways: from a high- T expansion [13] and by using the spherical approximation [14,15]. Finally, we perform large-scale simulations of the full KLM. We use a variant of the kernel polynomial method (KPM) [16–18] to integrate Langevin dynamics (LD) and to evaluate the resistivity using the Kubo formula [19]. Our KPM-LD simulations on a triangular lattice (TL) with 256^2 sites confirm the WC results and generalize them to the intermediate and strong-coupling regimes.

We consider the KLM,

$$\mathcal{H} = \sum_{\mathbf{k}, \sigma} (\varepsilon_{\mathbf{k}} - \mu) c_{\mathbf{k}\sigma}^\dagger c_{\mathbf{k}\sigma} + \frac{J}{\sqrt{N}} \sum_{\mathbf{q}, \mathbf{k}, \sigma, \sigma'} c_{\mathbf{q}\sigma}^\dagger \boldsymbol{\sigma}_{\sigma\sigma'} c_{\mathbf{q}+\mathbf{k}\sigma'} \cdot \mathbf{S}_{\mathbf{k}}. \quad (1)$$

The operator $c_{\mathbf{k}\sigma}^\dagger (c_{\mathbf{k}\sigma})$ creates (annihilates) an itinerant electron with momentum \mathbf{k} and spin σ . $\varepsilon_{\mathbf{k}} = -\sum_{\delta} t_{\delta} e^{i\mathbf{k}\cdot\delta}$

is the bare electronic dispersion relation with chemical potential μ and hopping amplitudes t_δ between sites connected by δ . The second term is the exchange interaction between the conduction electrons and the local magnetic moments \mathbf{S}_k in Fourier space. We assume classical moments with magnitude $|\mathbf{S}_i| = 1$ (σ is the vector of the Pauli matrices).

The conduction electrons can be integrated out in the WC limit by expanding in the small parameter $J\eta(\varepsilon_F)$. The resulting RKKY spin Hamiltonian is

$$H_{\text{RKKY}} = -J^2 \sum_k \chi_k^0 \mathbf{S}_k \cdot \mathbf{S}_{\bar{k}} \quad (2)$$

with $\bar{k} \equiv -k$ and $\mathbf{S}_k = \sum_l e^{ik \cdot r_l} \mathbf{S}_l / \sqrt{N}$ (N is the total number of lattice sites). The effective coupling constant in momentum space is $-J^2 \chi_k^0$ with $\chi_k^0 = T \sum_{q, \omega_n} G_{q, \omega_n}^0 G_{q+k, \omega_n}^0$, where $\omega_n = (2n+1)\pi T$ are the Matsubara frequencies and $G_{k, \omega_n}^0 = \{i\omega_n - [\varepsilon_k - \mu]\}^{-1}$ is the bare Green's function. Then, the RKKY interaction favors magnetic orderings that maximize χ_k^0 .

The electrons feel an effective potential produced by the spin configuration through the exchange interaction J . If the system orders at low-enough temperature ($T \leq T_c$), the periodic array of spins only produces coherent electron scattering, which does not contribute to ρ [20]. However, the situation changes above T_c because the magnetic moments develop liquidlike correlations, which produce incoherent elastic electron-spin scattering. Within the Born approximation, the scattering cross section is proportional to the spin SF,

$$\mathcal{S}(\mathbf{k}) = \frac{1}{N} \sum_{jl} e^{ik \cdot (r_j - r_l)} \langle \mathbf{S}_j \cdot \mathbf{S}_l \rangle = \langle \mathbf{S}_k \cdot \mathbf{S}_{\bar{k}} \rangle, \quad (3)$$

where $\langle \dots \rangle$ denotes the thermodynamic average. $\mathcal{S}(\mathbf{k})$ satisfies the sum rule $\sum_k \mathcal{S}(\mathbf{k}) = N$ because $|\mathbf{S}_i| = 1$. Unlike the high- T gas regime, characterized by a nearly k -independent spin SF, short-range magnetic correlations appear in the liquid regime. The RKKY interaction is expected to enhance $\mathcal{S}(\mathbf{k})$ for wave vectors connecting points on the FS because those are the processes that more effectively reduce the electronic energy. Given that the same processes contribute to the incoherent elastic scattering in the paramagnetic state, ρ should increase upon reducing T from the high- T gas regime to the $T_0 \lesssim T \lesssim |\theta_{\text{CW}}| \sim J^2/t$ liquidlike regime.

To illustrate this point we will consider the simple case of a circular FS, relevant for most 2D lattices with a low electron (hole) filling fraction [22]. The dispersion relation near the bottom (top) of the band can be approximated by $\varepsilon_k \approx k^2/2m$. The resulting RKKY Hamiltonian is strongly frustrated: any spiral with wave vector \mathbf{k} is a ground state as long as $k \leq 2k_F$. The RKKY interaction favors these magnetic configurations because those are the only spirals that can scatter electrons between points \mathbf{q} and $\mathbf{q} + \mathbf{k}$ on the FS.

Within the Born approximation, the inverse relaxation time for elastic scattering is

$$\frac{1}{\tau_{k_F}} = \frac{4\pi J^2}{N} \sum_k \delta(\varepsilon_F - \varepsilon_k) \mathcal{S}(\mathbf{k} - \mathbf{k}_F) (1 - \cos \theta_{k_F, k}). \quad (4)$$

This expression is further simplified if $\mathcal{S}(\mathbf{k}) = \mathcal{S}(k)$, which is a good approximation for low carrier filling fractions in the integration domain $k < 2k_F$:

$$\frac{1}{\tau_{k_F}} = 4\pi m J^2 c \int_0^1 dx \frac{x^2}{\sqrt{1-x^2}} \mathcal{S}(2k_F x), \quad (5)$$

where c is a number that depends on the lattice, e.g., $c = \sqrt{3}/\pi^2$ for a TL. The T dependence of τ_{k_F} is then determined by the variation of $\mathcal{S}(k)$ for $k \leq 2k_F$.

We will use two independent approaches for computing the T dependence of $\mathcal{S}(k)$ in the gas and liquidlike regimes. The first approach is a straightforward high- T expansion [13]:

$$\begin{aligned} \mathcal{S}(k) = & 1 + K \tilde{\chi}_k + K^2 [\tilde{\chi}_k^2 - \langle \tilde{\chi}^2 \rangle] + K^3 [\tilde{\chi}_k^3 - \langle \tilde{\chi}^3 \rangle \\ & - 2\tilde{\chi}_k \langle \tilde{\chi}^2 \rangle + \frac{2}{5N^2} \sum_{qq'} \tilde{\chi}_q \tilde{\chi}_{q'} \tilde{\chi}_{k-q-q'}] \end{aligned} \quad (6)$$

with $K = 2J^2\beta/3$, $\tilde{\chi}_k = \chi_k^0 - \langle \chi \rangle$, and $\langle \tilde{\chi}^n \rangle = \sum_k \tilde{\chi}_k^n / N$. Figure 1(a) shows the bare magnetic susceptibility for the isotropic FS under consideration. Figure 1(b) shows the bare susceptibility for a TL with nearest-neighbor (NN) hopping t and an electron filling fraction $n = 0.09$ (the mass is $m = 1/3t$). As expected, the effect of the small C_6 lattice anisotropy (of order k_F^6) is to split the large global maxima degeneracy that would correspond to an isotropic χ_k^0 . We will see that this splitting does not alter significantly the window of stability of the liquidlike regime. Figures 1(c) and 1(d) show the momentum dependence of the SF at different temperatures obtained from Eq. (6) for the isotropic FS and the triangular KLM, respectively.

To understand the insulating sign of the temperature dependence of $1/\tau$, it suffices to analyze the second term in Eq. (6), which gives the leading order contribution to the momentum dependence of $\mathcal{S}(\mathbf{k})$. Since $\chi_k > 0$, the prefactor of the $1/T$ term in $1/\tau$ is positive as long as the average of χ_k over the interval $(0, 2k_F)$ in Eq. (5) exceeds the contribution from $\langle \chi \rangle$, which is just a constant times $\langle \chi \rangle$. Suppose that χ_k^0 does not vary dramatically in the interval $(0, 2k_F)$, where it can be estimated by some typical value $\bar{\chi}$, and falls off quickly for $k_F \ll k \ll b$, where $b \sim 1$ is the reciprocal lattice spacing. Then, the contribution of χ_k^0 to the integral in Eq. (5) is on the order of $\bar{\chi}$. On the other hand, $\langle \chi \rangle$ is an average value over the entire Brillouin zone, normalized by its area. Therefore, $\langle \chi \rangle \sim \bar{\chi} (k_F/b)^2$, and the contribution from χ_k^0 is reduced only by a small correction of order $(k_F/b)^2$ [23].

Compared with the high- T expansion, the so-called spherical approximation [14,15] is less well controlled,

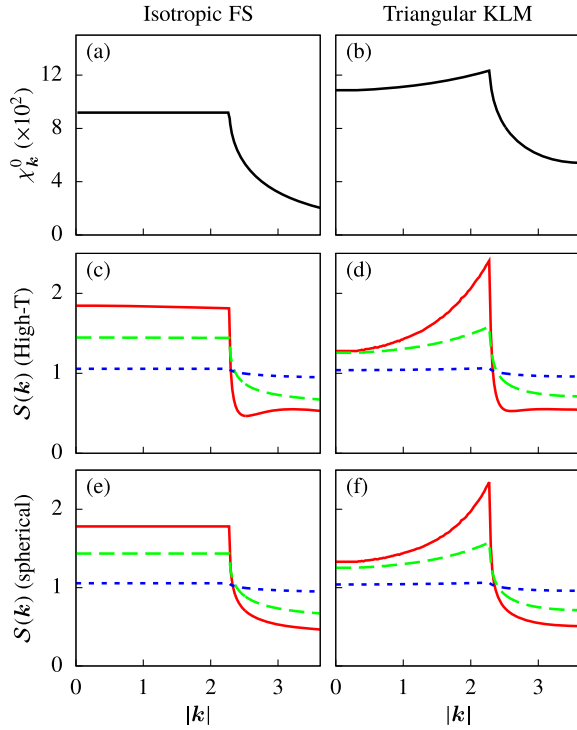


FIG. 1. Bare electronic susceptibility for (a) a 2D electron gas with isotropic dispersion $\varepsilon_k = k^2/2m$, and (b) a TL with NN hopping ($t = 1$) and filling fraction $n = 0.09$. Panels (c)–(f) show the momentum dependence of $S(\mathbf{k})$ at temperatures $T = \{0.03, 0.06, 0.45\}J^2/t$ represented by solid, dashed, and dotted curves, respectively. Panels (c) and (d) are obtained from the high- T expansion in Eq. (6), while panels (e) and (f) are obtained from the spherical approximation. Each panel is calculated using the bare magnetic susceptibility vertically above it. For panels (d) and (f) we assume $S(\mathbf{k}) = S(k)$, which is correct to within 1% relative error.

but can be applied to a wider temperature range. The hard constraints $|S_i| = 1$ are replaced with a global soft constraint $\sum_i |S_i|^2 = N$, which renders the spin Hamiltonian quadratic and can be easily integrated to give $S(\mathbf{k}) = \{3T/2[\Delta(T) - J^2\tilde{\chi}_k]\}$, where $\Delta(T)$ is determined from the self-consistency equation [23]: $\frac{1}{N}\sum_k J^2/[\Delta(T) - J^2\tilde{\chi}_k] = K$. Figures 1(e) and 1(f) show that the results for the isotropic FS and the triangular KLM agree with Figs. 1(c) and 1(d) down to $T \approx 0.03J^2/t$, at which point the high- T expansion fails.

The electrical conductivity is given by

$$\sigma = -\frac{e^2}{2} \int \frac{\sqrt{3}d^2k}{8\pi^2} \tau_k v_k^2 \frac{df(\varepsilon_k)}{d\varepsilon_k} \approx \frac{3\sqrt{3}e^2}{8\pi} tk_F^2 \tau_{k_F}. \quad (7)$$

Replacing τ_{k_F} with its expression given in Eq. (5), we obtain

$$\rho(T) = \frac{4}{\pi} \rho_0 \int_0^1 dx \frac{x^2}{\sqrt{1-x^2}} S(2k_F x), \quad (8)$$

where $\rho_0 = 8\pi J^2/(3tek_F)^2$. Figure 2(a) compares the resistivity curves $\rho(T)$ obtained from the high- T expansion and from the spherical approximation. As expected from the comparison of the magnetic SF, the resistivity curves practically coincide down to $T \approx 0.03J^2/t$. Both curves confirm our main conjecture $d\rho/dT < 0$ because the system develops stronger spin-spin correlations for wave vectors $k \leq 2k_F$. This increase should be interrupted at $T = T_0$ where precursors of magnetic Bragg peaks develop from the broad peaks of the liquid state and the Born approximation ceases to be valid.

The analytical approach that we have used for computing $\rho(T)$ is only valid in the WC regime. Away from the WC regime, the RKKY theory is no longer valid as an effective low-energy theory for the KLM and the Born approximation is no longer justified. Moreover, the two different approaches that we used for computing $S(\mathbf{k})$ fail at low T . Our calculations then need to be complemented with numerical simulations valid for any coupling strength and down to arbitrarily low T .

We perform KPM-LD simulations on a 256×256 TL with small electron filling $n = 0.09$ and $J/t = (0.2, 1.0, 1.5, 2.0)$ [25]. We integrate the dimensionless

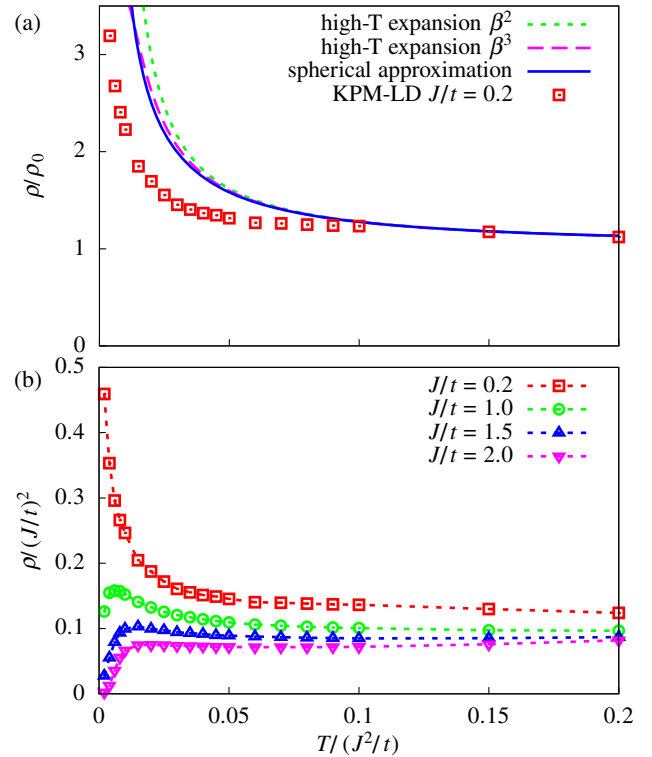


FIG. 2. (a) Temperature dependence of the resistivity for a triangular KLM with NN hopping ($t = 1$) and filling fraction $n = 0.09$. The lines correspond to calculations based on the Born approximation [see Eq. (8)] and different analytical approaches for computing the temperature dependence of $S(\mathbf{k})$. The symbols correspond to the results of KPM-LD simulations rescaled by $\rho(T = J^2/t)$. (b) Resistivity curve (in units of h/e^2) obtained from KPM-LD simulations for different coupling strengths [24].

stochastic Landau-Lifshitz dynamics with a unit damping parameter using the Heun-projected scheme [26] for a total of $(2 \times 10^3, 4 \times 10^3, 6 \times 10^3, 1 \times 10^4)$ time steps of duration $\Delta\tau = (100, 10, 5, 2)$. We estimate the effective spin forces using the gradient transformation described in Ref. [17]. To decrease the stochastic error, we use the probing method of Ref. [27] with $R = 128$ random vectors. The Chebyshev polynomial expansion order is $M = 500$. To calculate the resistivity, we expand the Kubo-Bastin formula [19,28] using the KPM [16,29] with $M = (6000, 1000, 1000, 500)$ [30]. For each temperature, we average the longitudinal conductivity over ten snapshots separated by $(100, 100, 200, 500)$ integration time steps.

Figure 2(b) shows the numerical $\rho(T)$ results for the different J/t values. Frustration decreases with J/t because higher order contributions (beyond RKKY level) split the degeneracy for $k \leq 2k_F$. For the strong-coupling limit $J \gg t$ the low-energy sector of \mathcal{H} can be mapped into a double-exchange model, which favors ferromagnetic (FM) ordering at a critical temperature T_c comparable to $|\theta_{CW}|$. Given that the temperature window with liquidlike correlations diminishes as a function of J/t , the relative low-temperature upturn of $\rho(T)$ should also decrease, as shown in Fig. 2(b).

In the intermediate-coupling regime $J/t = 1, 1.5$, and 2 , the low- T upturn of $\rho(T)$ reaches a maximum at temperature T_0 and drops rapidly for $T < T_0$. This crossover corresponds to the enhanced SF at wave vectors $k < 2k_F$. Figures 3(a) and 3(b) show the temperature dependence of $\mathcal{S}(\mathbf{k})$ for $J/t = 1$ and 2 , respectively. The roughly uniform weight of $\mathcal{S}(\mathbf{k})$ for $k < 2k_F$ starts redistributing below $T \approx 0.006J^2/t$. When $J/t \approx 1$ we observe the formation of a ring in Fourier space at $T \approx T_0$. This disordered phase is dynamically trapped at the lowest temperatures, $T \lesssim 0.002J^2/t$. As expected from the strong-coupling analysis, its radius $k_0 < 2k_F$ decreases with J/t . For larger couplings $J/t \gtrsim 2$ the FM phase clearly wins at low T . We note that, for $T > T_0$, there is strong backward scattering produced by the $k \lesssim 2k_F$ components of $\mathcal{S}(\mathbf{k})$. The resistivity drops below T_0 because the backscattering contribution ($k = 2k_F$) is reduced by the formation of a ring at $k_0 < 2k_F$ [see the integrand of Eq. (8)].

Here, we have only considered the resistivity component arising from electron-spin scattering. Electron-electron and electron-phonon scattering also contribute to ρ in real materials. These additional contributions increase with T , whereas we have argued that the electron-spin scattering produces a negative $d\rho/dT$. The combination thus yields a resistivity minimum [31]. Although we have assumed classical local spins ($S \rightarrow \infty$), our results can be extended to arbitrary S . The generalization of Eq. (6) is straightforward [32]. The main qualitative change is the Kondo effect expected for quantum spins and the antiferromagnetic exchange J . This effect becomes apparent by applying the T -matrix formalism up to order J^3 to the KLM [23], which yields

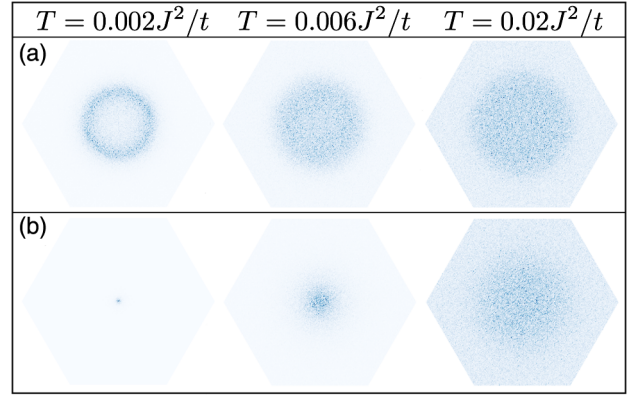


FIG. 3. Structure factor $\mathcal{S}(\mathbf{k})$ for three temperatures at intermediate couplings (a) $J/t = 1$ and (b) $J/t = 2$. At $T \approx 0.02J^2/t$, $\mathcal{S}(\mathbf{k})$ is nearly uniformly distributed in the disk $k \lesssim 2k_F$. Around $T \approx 0.006J^2/t$ the weight begins shifting toward a $k \lesssim 2k_F$ radius ring ($J/t \approx 1$) or $k = 0$ FM order ($J/t \gtrsim 2$).

$$\rho(T) \approx \rho_{\text{RKKY}}(T) \left[1 - 8J\eta(\epsilon_F) \ln\left(\frac{k_B T}{D}\right) \right], \quad (9)$$

where $\rho_{\text{RKKY}}(T)$ is given in Eq. (8). $\rho_{\text{RKKY}}(T)$ becomes T independent at $T \gg |\theta_{CW}|$, so the only T dependence arises from the Kondo effect. According to Eq. (9), the Kondo logarithmic behavior crosses over into a power law [23]

$$\rho_{\text{RKKY}}(T) \sim \frac{a}{T - T^*} + b, \quad (10)$$

upon entering the range $T_0 \lesssim T \lesssim |\theta_{CW}|$. The qualitatively different T dependence should allow us to distinguish between the two mechanisms for the resistivity upturn. Moreover, the upturn produced by the RKKY mechanism should be accompanied by a corresponding upturn in the correlation length ξ . Indeed, moderately frustrated materials, such as Gd_2PdSi_3 and RCuAs_2 ($R = \text{Sm, Gd, Tb, and Dy}$) [3–5], exhibit a nonlogarithmic resistivity upturn right above the Néel temperature. According to Refs. [33,34], the resistivity minimum of the pyrochlore oxides $\text{Pr}_2\text{Ir}_2\text{O}_7$ and $\text{Nd}_2\text{Ir}_2\text{O}_7$ is also caused by spin-spin correlations described by the spin ice model.

Furthermore, the Kondo effect is absent in transition metal oxides, where J is FM (Hund's coupling). Our results indicate that the resistivity upturn persists in the intermediate coupling regime, relevant to these materials. Indeed, a resistivity upturn has been observed in $(\text{Ga}_{1-x}\text{Mn}_x)\text{As}$ [35,36] and manganites [37] above the FM transition temperature T_c .

Our key conclusion is that the RKKY interaction enhances the elastic electron-spin scattering by increasing the magnetic SF for wave vectors connecting points on the FS. Assuming that this enhancement eventually leads to Bragg peaks (for $T < T_c$), which do not produce incoherent scattering, frustration is necessary to open a wide enough temperature window (liquidlike regime) over which the

resistivity upturn becomes noticeable. Although we have focused on 2D systems with a small FS, the conclusion applies generally to frustrated itinerant magnets, provided that χ_k^0 is larger on average for wave vectors \mathbf{k} connecting points on the FS.

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