There has been intense focus over the last two decades on magnetic materials which display large negative magnetoresistance (MR) [1–3]. In these systems, typically, an applied magnetic field reduces the spin disorder, leading to a suppression of the resistivity. The field may even drive an insulator-metal transition leading to ‘colossal’ MR [1]. Large positive MR is rarer, and seems counterintuitive since an applied field should reduce magnetic disorder and enhance conductivity. We illustrate a situation in double perovskite [4] metals where an applied field can lead to huge positive MR. The underlying principle suggests that local moment antiferromagnetic metals [5–12], at strong coupling, should in general be good candidates for such an unusual field response.

The double perovskites (DP), \( \text{A}_2\text{B'B'O}_6 \), have been explored [4] mainly as ferromagnetic metals (FMMs) or antiferromagnetic insulators (AFMIs). The prominent example among the former is \( \text{Sr}_2\text{FeMoO}_6 \) (SFMO) [14], while a typical example of the latter is \( \text{Sr}_2\text{FeWO}_6 \). The FMMs have moderate negative MR [14–16]. As in other correlated oxides [17], the magnetic order in the ground state is expected to be sensitive to electron doping and it has been suggested [18–20] that an FMM can give way to an antiferromagnetic metal (AFMM) through increasing electron density. The AFM order is driven by kinetic energy and, typically, has lower spatial symmetry than the parent structure.

The ongoing effort [21, 22] to obtain an AFMM by electron doping the FMM has revealed some signature [22] of a non-ferromagnetic metallic state in heavily electron doped SFMO [22]. There are two problems, however: (a) Antisite defects: in a material like SFMO, substituting La for Sr to achieve a higher electron density in the Fe-Mo subsystem tends to make the Fe and Mo ionic sizes more similar (due to resultant valence change), increases the likelihood of antisite disorder (ASD) and suppresses magnetic order. (b) Detection: even if an AFMM state is achieved, confirming the magnetic order is not possible without neutron scattering. The zero field resistivity is unfortunately quite similar [23] to that of the FMM.

We discovered a remarkably simple indicator of an AFMM state. Studying a two-dimensional (2D) model of DPs, in the doping window where the ground state is AFMM, we observe that: (i) For temperatures below the zero field \( T_c \) of the AFMM, a modest magnetic field can enhance the resistivity.
more than tenfold. (ii) The MR is suppressed by structural defects (ASD), but is still almost twofold for a system with ~25% antisite disorder. (iii) The enhancement in resistivity is related to the presence of short range correlated AFM domains that survive the field suppression of the global AFM order and lead to enhanced electron scattering. The mechanism suggests a generalization to 3D, where direct simulations are difficult, and to other local moment AFMM systems where an applied field can suppress long range AFM order.

We first describe the structurally ordered DP model and then generalize it to the disordered case later.

$$H = \epsilon_B \sum_{iB} f_{ia}^+ f_{ia} + \epsilon_B' \sum_{iB} m_{ia}^+ m_{ia} - t \sum_{<ij> \rho} f_{ia}^+ f_{ij}^\dagger m_{i\rho}$$

$$+ J \sum_{iB} S_i \cdot f_{ia}^+ \sigma_{ij} f_{jB} - h \sum_i S_i - \mu N,$$

Here $f$ is the electron operator on the magnetic, B, site and $m$ is the operator on the non-magnetic, B', site. $\epsilon_B$ and $\epsilon_B'$ are on-site energies, at the B and B' sites, respectively, which alternate along each Cartesian axis. $t$ is the hopping between nearest neighbor B and B' ions on the square lattice, and we will set $t=1$ as the reference scale. $S_i$ is the core B spin on the site $R_i$. We assume $S=|S|=1$ and absorb the magnitude of the spin in the coupling $J$ at the B site. When the ‘up spin’ core levels are fully filled, as for Fe in SFMO, the conduction electron is forced to be antiparallel to the core spin. We have used $J>0$ to model this situation and considered $Jt \gg 1$ here. We have set the effective level difference $\Delta_{eff} = (\epsilon_B - J/2) - \epsilon_B = 0$. The chemical potential $\mu$ is chosen so that the electron density is in the window for the A type (stripe-like) order of core spins. $N$ is the total electron count and $h$ is the applied magnetic field in the $\hat{z}$ direction. We have ignored next-nearest neighbor hopping and orbital degeneracy in the present model. We also focus on the 2D case here, where simulation and visualization is easier, and comment on the 3D case at the end. Although there is no real $T_c$ in the infinite two dimensional system, what we indicate as $T_c$ here is roughly the scale where the correlation length crosses the system size. Since the correlation length grows exponentially at low temperature in a system with Heisenberg symmetry [24], any weak anisotropy or 3D coupling would stabilize finite $T_c$.

The model has an FMM ground state at low electron density [18–20, 25]. This is followed by a phase with stripe-like order—with FM B lines coupled AFM in the transverse direction. We call this the ‘A type’ phase. This is followed by a more traditional AFM phase where an up spin B ion, say, is surrounded by four down spin B ions, and vice versa (the ‘G type’ phase). We focus here on the A type phase since this has a simple 3D counterpart and occurs at a physically accessible electron density.

We solve the spin-fermion problem via real space Monte Carlo using a cluster method [26]. This allows access to the system size $(40 \times 40)$. We have used field cooling (FC) as well as zero field cooling (ZFC) protocols. For ZFC the system is cooled to the target temperature at $h/t=0$ and then a field is applied. We calculate the resistivity and the magnetic structure factor peaks and also keep track of spatial configurations of spins. The optical conductivity is calculated via the Kubo formulation, computing the matrix elements of the current operator. The ‘dc conductivity’ is the low frequency average, $\sigma_{dc} = (1/\Delta \omega) \int_0^{\Delta \omega} \sigma(\omega) d\omega$. This is averaged over thermal configurations and disorder, as appropriate. Our ‘dc resistivity’ $\rho$ is the inverse of this $\sigma_{dc}$, with $\Delta \omega = 0.05t$. We have measured the resistivity in the units of $h/(\pi e^2)$, where $h$ is the Planck’s constant.

The A type pattern has two possible orientations of the stripes, either from bottom left to top right, or from bottom right to top left. The first corresponds to peaks in the magnetic structure factor $D(q)$ at $\{Q_{A1}, Q_{A2}\}$, and the second to peaks at $\{Q_{A3}, Q_{A4}\}$. For reference, $Q_{A1} = \{\pi/2, \pi/2\}$, $Q_{A2} = \{3\pi/2, 3\pi/2\}$,
Theorem 1: \(Q_A = [\pi/2, 3\pi/2], Q_A = [3\pi/2, \pi/2]\). The ferromagnetic peaks in \(D(q)\) are at \(Q_{\pi} = \{0, 0\}\) and \(Q_{\pi} = \{\pi, \pi\}\). The ordered configurations lead to peaks at two wave vectors, since the model has both magnetic and non-magnetic sites and our wave vectors are defined on the overall \(B-B'\) lattice.

Let us first examine the FC results in resistivity, figure 1. Cooling at \(h/t = 0\) leads to a sharp drop in resistivity at \(T = T_0^c = 0.032\), where \(T_0^c\) is the zero field transition temperature, and \(\rho(T) \rightarrow 0\) at \(T \rightarrow 0\). Cooling at \(h/t = 0.01\) leads to a small suppression in \(\rho_c\), but the trend in \(\rho(T)\) remains similar to \(h/t = 0\). Between \(h/t = 0.01\) and \(h/t = 0.02\), however, there is a drastic change in \(\rho(T)\), and, as we will see later, in the magnetic state. The primary effect is a sharp increase in the \(T < T_0^c\) resistivity, with the \(T \rightarrow 0\) resistivity now being almost 40% of the paramagnetic value. Even at this stage it is clear that \(\rho(T, h)\) retains significant \(h\) dependence, even for \(h/t = 0\), can be very large, as \(T \rightarrow 0\), and is \(\sim 4\) for \(T \sim T_0^c\) and \(h/t = 0.02\). Increasing the field even further leads to a reduction in \(\rho(T)\) over most of the temperature window since the field promotes a ferromagnetic state suppressing the AFM fluctuations.

We can study the impact of the magnetic field also within the ZFC scheme. Figure 2 shows the result of applying a field after the cooling of the system to different temperatures, (i) \(T_0^c + \) slightly above \(T_0^c\), (ii) \(T_0^c - \) slightly below \(T_0^c\), (iii) \(T_0^c/2\), and (iv) \(T_0^c/4\).

For \(T > T_0^c\), the zero field resistivity is already large and the applied field mainly suppresses the thermal fluctuations, leading to a gradual fall in the resistivity. This is weak negative MR. Below \(T_0^c\) (where there is already noticeable AFM order) and at \(T_0^c/2\), the resistivity remains almost unchanged till some value \(h_c(T) \sim 0.01\), then there is a sharp increase in resistivity, with a peak in the ratio \(\rho(T, h)/\rho(T, 0)\) around \(h/t \sim (0.02-0.03)\) and a fall thereafter. This is consistent with the trends seen in figure 1. At \(T_0^c/2\) the ratio reaches a maximum \(\sim 12\). At lower temperature, \(T_0^c/4\), the field appears to have a much weaker effect, mainly because the update mechanism that we adopt does not allow a cooperative switching of the AFM state at low \(T\) till very large fields. The field induced switching is therefore achieved most easily between \(T_0^c/2\) and \(T_0^c\). Overall, there is a window of \(T\) over which a moderate magnetic field can lead to a several-fold rise in resistivity.

A first understanding of the rise in resistivity can be obtained from the magnetic snapshots of the system at \(T = T_0^c/2\) in figure 3. We plot the correlation \(f_i = S_0 S_i\) in an equilibrium magnetic snapshot, where \(S_0\) is a reference spin (bottom left corner) and \(S_i\) is the spin at site \(R_i\). The left panel is at \(h/t = 0\) and shows a high level of \(A\) type correlation (stripe-like pattern). This is a ‘low dimensional’ electron system since the electron propagation is along the one dimensional stripes in this 2D system. The stripe pattern has a high degree of order so the scattering effects and resistivity are low. The second panel is at \(h/t = 0.008\), just below the field induced destruction of the AFM order, and the pattern is virtually indistinguishable from that in the first panel.

The third panel in figure 3 is at \(h/t = 0.02\) where the applied field has suppressed long range \(A\) type order. However, there are strong \(A\) type fluctuations that persist in the system, and they lead to a pattern of short range ordered \(A\) type patches with competing orientations, \(\{Q_{A1}, Q_{A2}\}\) and \(\{Q_{A1}, Q_{A4}\}\), in a spin polarized background. This patchwork leads to a high resistivity, higher than that in the leftmost panel, since the ferromagnetic paths are fragmented by intervening \(A\) type regions, while the \(A\) type regions are poorly conducting due to their opposite handedness. In the last panel the field, \(h/t = 0.05\), is large enough so that even the AFM fluctuations are wiped out and the spin background is a 2D ferromagnet with extremely short range inhomogeneities. The resistance here is significantly below the peak value.

Let us summarize the physical picture that emerges in the non-disordered system before analyzing the effect of antisite disorder. The ingredients of the large MR are the following: (i) An AFM metallic phase, without too much quenched disorder so that the resistivity in the magnetically ordered state is small. (ii) Field induced suppression of the AFM order at \(h = h_c(T)\), say, replacing the ordered state with AFM correlated spins in an FM background, leading to a high resistivity state. In contrast to the standard negative MR scenario, where an applied field pushes the system from a spin disordered state to a spin ordered state, here the applied field pushes the ordered (AFM) state towards spin disorder.

While our results are concrete in the case of a 2D ‘double perovskite’ model and a stripe-like ground state, the principle above is far more general and should apply to other non-ferromagnetic ordered states in two or three dimensions and to microscopic models that are very different from the double perovskites. We will discuss this issue at the end.

Defects are inevitable in any system and in particular one expects antisite disorder in the DPs. The concentration of such defects may actually increase on electron doping of a material like SFMO, due to the valence change, and we need to check if the large MR is wiped out by weak disorder. The presence of ASD affects the zero field magnetic state itself, as we have discussed elsewhere [23], and the field response has to be understood with reference to this \(h/t = 0\) state. Let us first define the augmented model to describe the presence of ASD.

We generalize the clean model by including a configuration variable \(\eta_i\), with \(\eta_i = 1\) for B sites and \(\eta_i = 0\) for B’ sites. In the structurally ordered double perovskites the \(\eta_i\) alternate along
each axis. We consider progressively ‘disordered’ configurations, generated through an annealing process [27]. These mimic the structural domain pattern observed [28, 29] in the real double perovskites. For any specified |η| background, the electronic-magnetic model has the form:

\[ H = H_{\text{loc}} \{ \eta \} + H_{\text{kin}} \{ \eta \} + H_{\text{mag}} \{ \eta \}, \]

where \( H_{\text{loc}} \{ \eta \} = c_h \sum \eta^i f^i_{\text{loc}} f^i_{\text{loc}} + c_t \sum (1-\eta) m^i_{\text{loc}} m^i_{\text{loc}}, \) and the hopping term connects nearest neighbor sites, irrespective of whether they are B or B': \( H_{\text{kin}} \{ \eta \} = -t \sum_{i \neq j} \eta f^i_{\text{loc}} f^j_{\text{loc}} -b \sum_{i \neq j} (1-\eta) (1-\eta) m^i_{\text{loc}} m^j_{\text{loc}} -b \sum_{i \neq j} (\eta + \eta -2q\eta) (f^i_{\text{loc}} m^j_{\text{loc}} + h.c.). \)

The magnetic interactions include the Hund’s coupling on B sites, and antiferromagnetic superexchange \( J_{\text{AF}} = 0.08 t \) between two nearest neighbor B sites: \( H_{\text{mag}} \{ \eta \} = J \sum S_i S_j f^i_{\text{loc}} f^j_{\text{loc}} + J \sum_{i \neq j} \eta q S_i S_j. \) For simplicity we set the nearest neighbor hopping amplitudes \( t_1 = t_2 = t_3 = t. \)

We use the simplest indicator \( S = 1-2x \) (not to be confused with the magnitude of the core spin), to characterize these configurations, where \( x \) is the fraction of B (or B') atoms that are on the wrong sublattice. Figure 4 shows the resistivity ratio \( \rho(h)/\rho(0) \) at \( T = T_0/2 \) in panel (a), and the field dependence of structure factor peaks in panels (b)–(d). The structural motifs on which the magnetism is studied are shown in the first column of figure 5.

Down to \( S = 0.50 \) the ratio \( \rho(h)/\rho(0) \) has a behavior similar to the clean case, figure 2, but the peak ratio reduces to \( \sim 3 \) for \( S = 0.50. \) There is a corresponding field induced suppression in the principal AFM peak \( Q_{A3} \), and an enhancement of the FM peak \( Q_F. \) The complementary AF peak \( Q_{A3} \) slowly increases with \( h \), has a maximum around \( h = 0.02 \) (where the disconnected AFM domains exist) and falls at large \( h \) as the system becomes FM overall. The trend that we had observed in the clean limit is seen to survive to significant disorder. At \( S = 0.08, \) where the B–B' order is virtually destroyed, the \( h = 0 \) state, figure 5 last row, has no long range AFM order. It is already a high resistivity state and an applied field actually leads to weak negative MR. Let us now place our results in a more general context.

(a) Earlier theory: we are aware of one earlier effort [30] in calculating the MR of AFM metals (and semiconductors), assuming electrons are weakly coupled to an independently ordering local moment system. Indeed, the authors suggested that AFM semiconductors could show positive MR. Our framework focuses on field induced suppression of long range AFM order, rather than perturbative modification, and the positive MR shows up even in a ‘high density’ electron system. The electron-spin coupling is also (very) large, \( h t \gg 1, \) and cannot be handled within Born scattering.

(b) Experiments: the intense activity on correlated systems has led to the discovery of a few AFM metals, e.g., in the manganites \( \text{La}_{1-x} \text{Sr}_{x} \text{MnO}_3 \) [6], in \( \text{CaCrO}_3 \) [6], in the ruthenates \( \text{Ca}_3 \text{Ru}_2 \) [7–10], and in the heavy-fermions \( \text{CeRhIn}_5 \) [11, 12]. Of these, for the manganites and \( \text{CaCrO}_3 \) (where the resistivity is too large), we are not aware of MR results across the field driven transition. \( \text{Ca}_3 \text{Ru}_2 \text{O}_7 \) and \( \text{CeRhIn}_5 \) show large increase in the resistivity with the field induced growth of FM order. These are local moment AFM metals. On application of a magnetic field there is suppression of long range AFM order, leading possibly to a state with antiferromagnetically correlated spins in an FM background. This may be responsible for the high resistivity that is observed. Neutron diffraction in the presence of a magnetic field should be able to confirm this scenario.

(c) Generalization: We had also attempted to study the MR in the layered AFMM phase [20] expected in the 3D double perovskites. Unfortunately, the system size accessible within our cluster based MC cannot capture the thermal physics adequately. The 2D experience, however, allows us to make a phenomenological suggestion.

Let us assume that the AFM peak in \( D(q) \) is at one wave vector \( Q \), while the FM peak is at \( (0,0) \). The AFM phase has an order parameter \( m_{AF} \), say, while, for \( h > h_c(T) \), there is induced FM order of magnitude \( m_{EF}. \) Assuming that the dominant fluctuations in the relevant part of the \( (T,h) \) phase diagram are at \( q \sim Q \), we can write: \( D(q) \sim m_{AF}^2 \delta(q - Q) + A' \left(1 + (q - Q)^2 \xi^2 \right) \) when \( h < h_c(T) \), and \( D(q) \sim m_{EF}^2 \delta(q) + A' \left(1 + (q - Q)^2 \xi^2 \right) \) when \( h > h_c(T) \). Here \( m_{AF}, m_{EF} \) and \( \xi \) depend on \( (h,T) \), \( A \) depends on \( m_{AF} \) and \( \xi \), while \( A' \) depends on \( m_{EF} \) and \( \xi \). The amplitudes \( A \) and \( A' \) vanish as the corresponding \( m \) tend to saturate (since the spins get perfectly ordered). The delta functions in \( D(q) \) dictate the ‘mean field effects’ in the electronic band structure, while electron scattering is controlled by the Lorentzian part.
Consider three cases (a) $T = T_c^0$, $h = 0$, (b) $T = T_c^0$, $h = 0$, and (c) $T = T_c^0$, $h > h_c(T)$. In (a) there is no order, so $A$ is large, and $\rho = \rho_a$, say. For (b) even if $\xi$ were the same as in (a), the presence of a large order parameter would suppress $A$ and hence the scattering. We call this $\rho = \rho_b \ll \rho_a$ (assuming there is indeed a large order parameter and no significant background resistivity due to impurities). If we apply a field such that $m_{AF} \to 0$ but $m_F$ is still small, then the structure factor crudely mimics the paramagnetic case, and we should have $\rho_c = \rho_a$. If all this is true, then just beyond the field suppression of AFM order (and for $T$ just below $T_c^0$) we should get $\rho_c/\rho_b \gg 1$. Broadly, if the appearance of AFM order with reducing $T$ leads to a sharp drop in $\rho$ then the field induced resistivity ratio can be large. This is independent of dimensionality and microscopic detail.

Conclusion: We have studied the magnetoresistance in an antiferromagnetic metal motivated by the prediction of such a phase in the double perovskites. Beyond the modest field needed for suppression of long range antiferromagnetic order, the system shows an almost tenfold increase in resistivity near $T_c$. The effect originates from strong antiferromagnetic fluctuations in the field induced ferromagnetic background. The large positive magnetoresistance, though suppressed gradually, survives the presence of significant antisite disorder. We suggest how the principle behind this ‘colossal positive magnetoresistance’ could be applicable to other local moment antiferromagnetic metals as well.

Figure 5. Field response in the antisite disordered systems at $T = T_c^0/2$, for the $S$ values in figure 4. The left panels indicate the structural domains in the antisite disordered systems. The middle column shows the spin correlations at $h/t = 0$; note that rows 1–3 show significant $A$ type order, while the pattern in the 4th row has AFM domains of both orientations and so suppressed long range order. The right column shows the spin correlations at $h/t = 0.02$. In rows 1–3 the AFM pattern gets fragmented and FM regions show up. In row 4 the finite field pattern is not significantly different from the $h/t = 0$ case. Overall, the field enhancement of ‘spin disorder’ is large in the first three cases but modest at strong ASD.

\footnote{If the applied field drives a first order transition from a large $m_{AF}$ state to one with large $m_F$ the scenario above will not work.}
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