Chapter 5

Field Response in Antiferromagnetic Metals

Chapter summary: In this chapter we discuss the field response expected in an antiferromagnetic double perovskite metal. While the zero field resistivity is unremarkable in such a metal, we find that the magnetoresistance can be very large and positive. This can be a direct indicator of the metallic antiferromagnetic state. Beyond a modest field, needed for suppression of long range antiferromagnetic order, the system shows more than tenfold increase in resistivity near $T_c$ in a structurally ordered system. The ratio continues to be almost twofold even in systems with $\sim 25\%$ antisite disorder. The effect occurs because an applied field suppresses long range antiferromagnetic order leading to a state with short range antiferromagnetic correlations in the field induced ferromagnetic background. These antiferromagnetic fluctuations generate strong electronic scattering and a resistivity that can be much larger than the ordered antiferromagnetic metal. This mechanism is quite general, complementary to the colossal negative magnetoresistance process, and should operate in other local moment antiferromagnetic metals as well.

5.1 Introduction

There has been intense focus over the last two decades on magnetic materials which display large negative magnetoresistance (MR) \cite{1,3}. In these systems, typically, an applied magnetic field reduces the spin disorder leading to a suppres-
sion of the resistivity. The field may even drive an insulator-metal transition leading to ‘colossal’ magnetoresistance \[1\]. Large positive magnetoresistance 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 enormous positive magnetoresistance. The underlying principle suggests that local moment antiferromagnetic (AFM) metals \[5-12\], at strong coupling, should in general be good candidates for such unusual field response.

Like in other correlated oxides \[13\], the magnetic order in the double perovskites is expected to be sensitive to electron doping as we have discussed in detail earlier. It has been suggested \[14-16\] that the ferromagnetic metal can give way to an AFM metal on increasing electron density. The AFM order is driven by electron delocalisation and has lower spatial symmetry than the parent structure. The conduction path in the AFM background is low dimensional and easily disrupted.

There is ongoing effort \[17,18\] to obtain an AFM metal by electron doping the ferromagnetic metal. Although there is no clear evidence yet of the occurrence of an AFM metal, there is some signature of a non-ferromagnetic metal in the heavily electron doped \(\text{Sr}_2\text{FeMoO}_6\) \[18\]. The problems are twofold:

1. **Antisite defects**: in a material like \(\text{Sr}_2\text{FeMoO}_6\), 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, and suppresses magnetic order.

2. **Detection**: even if an AFM state is achieved, confirming the magnetic order is not possible without neutron scattering. The zero field resistivity is unfortunately quite similar \[19\] to that of the ferromagnetic metal.

### 5.2 Field response in ordered AFM metal

Let us first discuss the thermal and field effects in the structurally ordered double perovskite before examining the effect of antisite disorder \[20\]. As we have seen in the last chapter, upon increasing electron density the ground state changes from a ferromagnetic metal to a phase with stripe-like order. The ‘stripe’ phase involves
ferromagnetic B lines coupled antiferromagnetically in the transverse direction. We call this the A type phase. At even higher electron density there is a more traditional antiferromagnetic 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 it has a simple 3D counterpart and occurs at physically accessible electron density.

We have used field cooling (FC) as well as zero field cooling (ZFC) protocols. For zero field cooling the system is cooled to the target temperature at field \( h/t \) = 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 A type pattern has two possible orientations of the stripes, either from bottom left to top right, or from bottom right to top left. These are the two diagonals in 2D. The first corresponds to peaks in the structure factor \( D(q) \) at \( \{Q_{A1}, Q_{A2}\} \), and the second to peaks at \( \{Q_{A3}, Q_{A4}\} \). For reference, \( Q_{A1} = \{\pi/2, \pi/2\} \), \( Q_{A2} = \{3\pi/2, 3\pi/2\} \), \( Q_{A3} = \{\pi/2, 3\pi/2\} \), \( Q_{A4} = \{3\pi/2, \pi/2\} \). The ferromagnetic peak in \( D(q) \) are at \( Q_{F1} = \{0, 0\} \) and \( Q_{F2} = \{\pi, \pi\} \). The ordered configurations lead to peaks at two wavevectors since the model has both magnetic and non-magnetic sites and our wavevectors are defined on the overall B-B’ lattice.

5.2.1 Field cooling

Let us first examine the field cooling results in resistivity, Figure 5.1. Cooling at \( h/t = 0 \) leads to a sharp drop in resistivity at \( T = T^0_c \sim 0.032 \), where \( T^0_c \) is the zero field transition temperature, and \( \rho(T) \to 0 \) as \( T \to 0 \). Cooling at \( h/t = 0.01 \) leads to a small suppression in \( T_c \) 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 \( T \to 0 \) resistivity now being almost 40% of the paramagnetic value. Even at this stage it is clear that \( \rho(T,h)/\rho(T,0) \) can be very large as \( T \to 0 \) 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 antiferromagnetic fluctuations.
Figure 5.1: The resistivity $\rho(T)$ in the absence of antisite disorder for cooling in different applied fields. For temperatures below the zero field transition, $T_{c0}$, $\rho(T)$ increases on applying a field, and for $T>T_{c0}$ $\rho(T)$ decreases on applying a field. The ratio $\rho(T,h)/\rho(T,0)$ can be very large as $T\to0$ in this field cooling situation.

### 5.2.2 Zero field cooling

Now, we will study the impact of the magnetic field within the ZFC scheme. Figure 5.2 shows the result of applying a field after the cooling the system to four different temperatures, (i) slightly above $T_{c0}$, (ii) slightly below $T_{c0}$, (iii) to $T_{c0}/2$ and (iv) to $T_{c0}/4$.

For $T>T_{c0}$, the zero field resistivity is already large and the applied field mainly suppresses the antiferromagnetic thermal fluctuations, leading to a gradual fall in the resistivity. This is weak negative magnetoresistance.

Below $T_{c0}$ (where there is already noticeable antiferromagnetic order) and at $T_{c0}/2$, the resistivity remains almost unchanged till some value $h_{c}(T)\sim0.01$, 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 5.1. At $T_{c0}/2$ the ratio reaches a maximum $\sim12$.

At lower temperature $T_{c0}/4$ the field appears to have a much weaker effect, mainly because the update mechanism that we adopt does not allow a cooperative
Figure 5.2: Field dependence of resistivity at different temperatures following a zero field cooling protocol. For $T = T_c^+$ (just above $T_c$) the zero field resistivity is already large and $\rho$ decreases slightly with $h$ due to suppression of spin disorder. For $T = T_c^-$ (just below $T_c$) and at $T_c/2$ there is a sharp increase in resistivity at $h/t \sim 0.01$, with a peak around $h/t \sim 0.02 - 0.03$ and a fall thereafter. This is consistent with the trends seen in Figure 5.1. At $T = T_c/4$ this ZFC scheme does not manage to create competing magnetic structures for $h/t \sim 0.05$, possibly due to metastability of the parent AFM pattern. The field induced switching is therefore easiest achieved between $T_c/2$ and $T_c$.

switching of the antiferromagnetic state at low $T$ till very large fields. The field induced switching is therefore easiest achieved between $T_c^0/2$ and $T_c^0$. 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 $T = T_c^0/2$ in Figure 5.3. 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
Figure 5.3: Evolution of spin correlations in the clean system in response to a magnetic field. The plot shows $f_i = S_0 \cdot S_i$ in a magnetic snapshot, where $S_0$ is a reference spin (bottom left corner). $T = T^0_c/2$ and the fields are, from left to right, $h/t = 0, 0.008, 0.02, 0.05$.

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 field induced destruction of antiferromagnetic order, and the pattern is virtually indistinguishable from that in the first panel.

The third panel in Figure 5.3 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 $\{Q_{A3}, Q_{A4}\}$, in a spin polarised background. This patchwork leads to a high resistivity, higher than that in 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 antiferromagnetic 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 summarise the physical picture that emerges in the non disordered system before analysing the effect of antisite disorder. The ingredients of the large magnetoresistance are the following:

1. An antiferromagnetic metallic phase, without too much quenched disorder so that the resistivity in the magnetically ordered state is small.

2. Field induced suppression of the antiferromagnetic order at $h = h_c(T)$,
Figure 5.4: Field response in the presence of antisite disorder. The temperature is $T = T_0^c/2$, where $T_0^c$ is the $T_c$ at $h/t = 0$ in the non disordered system. The results are obtained via ZFC. (a) Field dependence of resistivity, normalised to $h/t = 0$. (b) Magnetic structure factor at the major antiferromagnetic peak $Q_{A1}$. The value is same at $Q_{A2}$ also. (c) Growth in the ferromagnetic structure factor with $h$. (d) Growth in the complementary antiferromagnetic peak $Q_{A3}$, result same for $Q_{A4}$.

Let us highlight the contrast to the standard negative magnetoresistance 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 (antiferromagnetic) state towards spin disorder, before the high field polarised state occurs. Our concrete results are in the case of a 2D double perovskite model and a stripe-like ground state, but 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.
5.3 Response in antisite disordered AFM metals

Defects are inevitable in any system and in particular one expects antisite disorder in the double perovskites. The concentration of such defects may actually increase on electron doping a material like Sr$_2$FeMoO$_6$, due to the valence change, and we need to check if the large magnetoresistance is wiped out by weak disorder. The presence of antisite disorder affects the zero field magnetic state itself, as we have discussed elsewhere $[19]$, and the field response has to be understood with reference to this $h = 0$ state.

Figure 5.4 shows the resistivity ratio $\rho(h)/\rho(0)$ at $T = T_c/2$, in panel (a), and the field dependence of structure factor peaks in panels (b)-(d). Figure 5.5 first column shows the structural motifs on which the magnetism is studied.

Down to $S = 0.50$ the ratio $\rho(h)/\rho(0)$ has a pattern similar to the clean case, Figure 5.2, but the peak ratio reduces to $\sim 3$ for $S = 0.50$. There is a corresponding suppression in the principal antiferromagnetic peak $Q_{A1}$ and an enhancement of the ferromagnetic peak $Q_F$. The complementary antiferromagnetic peak $Q_{A3}$ slowly increases with $h$, has a maximum around $h/t = 0.02$ (where the disconnected antiferromagnetic domains exist) and falls at large $h$ as the system becomes ferromagnetic 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 $h/t = 0$ state, Figure 5.5 last row, has no long range antiferromagnetic order. It is already a high resistivity state and an applied field actually leads to weak negative magnetoresistance.

Since a real double perovskite has a three dimensional structure, we have also studied a one band three dimensional model. In contrast to the two dimensional case, where the effective magnetic lattice is bipartite, the three dimensional lattice has a geometrically frustrated face centered cubic structure. This promotes various non-collinear phases, studied in detail elsewhere $[16]$. We had tried to study the impact of a magnetic field on these phases. These results, unfortunately, are still preliminary and more work is needed to improve their quality.
Figure 5.5: Field response in the antisite disordered systems at $T = T_{c}^{0}/2$, for the $S$ values in Figure 5.4. The left panels indicate the structural domains. 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. 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 antisite disorder.

5.4 Discussion

Let us place our results in the general context of antiferromagnetic metals.

5.4.1 Earlier theory

We are aware of one earlier effort [21] in calculating the magnetoresistance of antiferromagnetic metals (and semiconductors), assuming electrons weakly coupled to an independently ordering local moment system. Indeed, the authors suggested that antiferromagnetic semiconductors could show positive magnetore-
sistance. Our framework focuses on field induced suppression of long range antiferromagnetic order, rather than perturbative modification, and the positive magnetoresistance shows up even in a high density electron system. The electron-spin coupling is also (very) large, $J/t \gg 1$, and cannot be handled within Born scattering.

5.4.2 Experimental results

The intense activity on oxides has led to the discovery of a few antiferromagnetic metals, e.g., in the manganite (La$_{0.46}$Sr$_{0.54}$MnO$_3$) $^5$, in CaCrO$_3$ $^6$, in the ruthenates (Ca$_3$Ru$_2$O$_7$) $^7$-$^10$, and in the heavy-fermions (CeRhIn$_5$) $^{11,12}$. Of these for the manganites and CaCrO$_3$ (where the resistivity is too large), we are not aware of magnetoresistance results across the field driven transition. Ca$_3$Ru$_2$O$_7$ and CeRhIn$_5$ show large increase in the resistivity with the field induced growth of ferromagnetic order, as shown in the Figure 5.6. These are local moment antiferromagnetic metals. On application of a magnetic field there is suppression of long range antiferromagnetic order, leading possibly to a state with antiferromagnetically correlated spins in a ferromagnetic 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 conform this scenario.

5.4.3 Qualitative analysis

The information about the spin configurations at any $(T,h)$ is encoded in the structure factor, $D(q)$. Let us put down a form for $D(q)$ and suggest how it affects the resistivity. To simplify notation we will assume that the antiferromagnetic peak is at one wavevector $Q$, while the ferromagnetic peak is at $\{0,0\}$. The antiferromagnetic phase has an order parameter $m_{AF}$, say, while, for $h > h_c(T)$, there is induced ferromagnetic order of magnitude $m_F$. Assuming that the dominant fluctuations in the relevant part of the $(T,h)$ phase diagram are at $q \sim Q$, we can write: $D(q) \sim m_{AF}^2\delta(q - Q) + A/(1 + (q - Q)^2\xi^2)$ when $h < h_c(T)$, and $D(q) \sim m_{F}^2\delta(q) + A'/(1 + (q - Q)^2\xi^2)$ when $h > h_c(T)$. $m_{AF}, m_F$ and $\xi$ depend on $(h, T)$, $A$ depends on $m_{AF}$ and $\xi$ and $A'$ on $m_F$ and $\xi$. The amplitudes $A$ and $A'$ vanish as the corresponding $m$ tend to saturate (since the spins get perfectly
Figure 5.6: Left column: Top row shows $\rho_c$ as a function of magnetic field for Ca$_3$Ru$_2$O$_7$ [7]. It has antiferromagnetic metallic phase between 48K to 55K, while the bottom row shows the low temperature in-plane field-induced change in resistivity $\Delta \rho = \rho(H) - \rho(0)$ $(H \parallel c)$ for antiferromagnetic metal CeRhIn$_5$ with $T_N = 3.8$K [11]. Right column: Plots magnetoresistance in the antiferromagnetic metallic phase for ruthenate (top panel) and heavy-fermions (bottom panel). Respective antiferromagnetic transition curve is also shown.

ordered). The delta functions in $D(q)$ dictate the bandstructure while electron scattering is controlled by the Lorentzian part.

Consider three cases (a) $T = T_c^+, h = 0$, (b) $T = T_c^-, h = 0$, and (c) $T = T_c^-, h > h_c(T)$. 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 $\rho = \rho_b << \rho_a$ (assuming there is indeed a large order parameter and no significant background resistivity due to impurities). If we apply a field such that $m_{AF} \rightarrow 0$ but $m_F$ is still small, then the structure factor crudely mimics the paramagnetic case, and we should have $\rho_c \approx \rho_a$. If all this is true, then just beyond field suppression of
antiferromagnetic order (and for $T$ just below $T_c$) we should get $\rho_c/\rho_b \gg 1$. Broadly, if the appearance of antiferromagnetic 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. A caution: as $T \to 0$, the applied field would drive a first order transition from a large $m_{AF}$ state to one with large $m_F$ and weak scattering. The scenario above will not work, as our Figure 5.2 illustrated.

5.5 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 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. The principle that we uncover behind this “colossal positive magnetoresistance” should be applicable to other local moment based antiferromagnetic metals as well.
Bibliography


