MAGNETIC FILAMENTS IN RESISTIVE MANGANITES

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In oxides where magnetic cations are separated by oxygen anions, exchange interactions are mediated through the oxygen p orbitals. The resulting "super-exchange" (SE) is found to be usually antiferromagnetic. When electrons can delocalize on at least two magnetic ions, a ferromagnetic interaction called “double exchange” (DE) can take place. The manganites exhibit a rich physical behaviour where structural, transport and magnetic properties are intricately related[1]. It has also been suggested that the competition between SE and DE interactions lead to a magnetic phase separation[2]. The $Pr_{0.67}Ca_{0.33}MnO_3$ compound has a ferro-magnetic transition below which the application of a magnetic field produces a first order transition from an insulating to a conducting state[3]. In this induced metallic-like state, the magnetization relaxes with time, leading to impressive resistive transitions[4]. This can be understood in a percolation picture where ferromagnetic regions are thermally activated into an antiferromagnetic insulating state. When the last percolation path breaks, the resistivity suddenly jumps to immeasurably large values. In order to finely characterize the phase separation in $Pr_{0.67}Ca_{0.33}MnO_3$ we have carried out Polarised Small Angle Neutron Scattering (PSANS) under applied fields along with electrical transport and magnetization measurements. The results presented here were obtained on a 1x1x3 mm^3 single crystal. The PSANS measurements were carried out at the ORPHEE reactor in Saclay on the spectrometer PAPOL. The SANS intensity angular dependence gives the Fourier transform of chemical and magnetic heterogeneities with sizes ranging from 1 nm to 100 nm. As shown on the typical spectrum in the inset of Fig.1, the SANS signal is characteristic of magnetic scattering with a contribution in $sin^2\alpha$ from the direction of the applied field. The intensities recorded at 4.2K under applied fields >1.5T, follow a q^{-n} law with $1.6 < n < 1.7$. These exponents, close to 5/3, are reminiscent of fractal dimensions as found for dilute polymers in good solvent. The parallel with polymer physics is instructive here because it suggests that the phase separation is of a filamentary type. We show below that the exchange interactions in these compounds can indeed lead to such a phase separation.

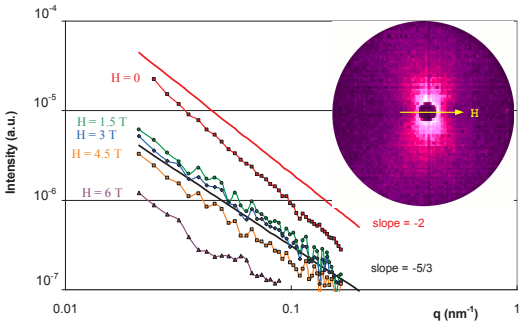

Figure 1. Magnetic SANS intensity from a $Pr_{0.67}Ca_{0.33}MnO_3$ single crystal, for a set of increasing fields from 0 to 6 T after Zero Field Cooling to 4.2K. The two straight lines are power laws with exponents –2 and –5/3. Inset: Typical PSANS spectrum showing the $\sin^2$ contribution of magnetic scattering.

In the $Pr_{0.67}Ca_{0.33}MnO_3$ compound, charge carriers are localized by a random magnetic potential. Thermal activation makes electrons hop in a random walk fashion to sites beyond their nearest neighbours. This is called “Variable Range Hopping” (VRH).The localizing potential has been attributed to random spin disorder which generates potential barriers coming from the Hund coupling energy between e_g and t_{2g} electrons on each Mn atom (of the order of 1eV). In insulating manganites, the electrons are localized on individual Mn ions. In that case, DE is not possible. Instead, we propose a picture in which, after every electronic hop, the electron gets localized in the e_g level of its new site and their spins become aligned. In the process, the total angular momentum of the electron and the Mn ion is preserved. Spatially, the interaction both follows and influences the random walk paths of hopping electrons through changes in the local magnetic potential. Because hopping happens preferentially between ions of similar spin direction, the exchange becomes stronger as spins are more closely aligned, which naturally results in a tendency to phase segregate. Indeed, once one Mn has in its vicinity another Mn with parallel spin, the hopping probability between this pair is overwhelmingly large and the ferromagnetic interaction will occur exclusively between these two ions. The remaining surrounding Mn ions interact only via SE. In order to demonstrate that this model leads to a filamentary phase segregation, we have carried out Monte Carlo

simulations treating transport and magnetism in a self-consistent manner (more details can be found in ref. 5. When hopping is turned on, magnetic filaments containing many parallel spin carriers with an enhanced mobility appear, as shown in Fig. 2.

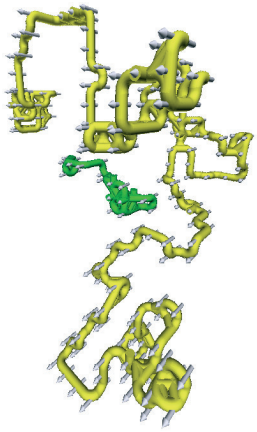

Figure 2. Magnetic filaments as obtained by Monte-Carlo simulations. The long yellow filament has a spin aligned with the external field whereas the small green one has its spin at 25° (the spins are all parallel within the filaments).

In the zero field-cooled (ZFC) state at H=0, the PSANS intensity can be well fitted with an exponent close to -2 (Fig. 1), i.e. a Debye function. This is consistent with previously published SANS data recorded at zero field and interpreted as an average coherence[6] or a "red cabbage structure"[7]. Debye functions being reminiscent of polymer melts, this indicates, in our picture, that the magnetic filaments are entangled and do not self-avoid. Here, “SE-screening” does not work since electrons tunnel over distances longer than the screening length. Hence, gaussian, entangled, randomly magnetized filaments are generated by a ZFC procedure as shown in Fig.3a. When a field is applied, those filaments with their magnetization parallel to the field grow while others shrink. Within the filaments, mobility is large and carriers proceed by nearest-neighbour-hops, mediating ferromagnetic "hopping exchange". Super-exchange interactions screen the filaments to make them self-avoiding (see Fig.3b) and the measured power laws are -5/3. This exponent remains unchanged ; only the global SANS intensity decreases with field (fig.1). The variation in intensity results from a combination of a reduction in magnetic contrast as the background is forced to become more ferromagnetic and an increase in density due to the growth of the filaments.

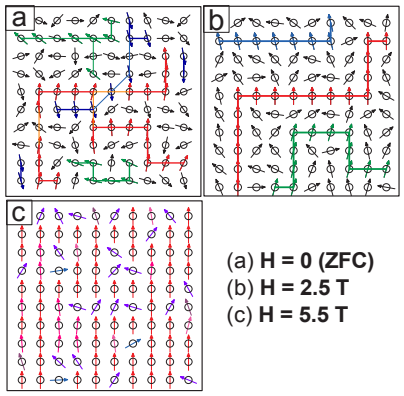

Figure 3. Schematics of the evolution of the filamentary phase with the applied field. From a disordered assembly of small filaments after the ZFC procedure, the applied field makes filaments with spins parallel to H grow, and shrinks the other ones. As the field continues to increase, the filaments percolate at 3.9T and then fade into the background when the difference in magnetization is too low to keep the carriers inside the filaments.

Complementary magnetization measurements allow us to conclude that the volume fraction of the filamentary phase increases monotonically as the field is raised. This naturally leads to a picture of magnetic filaments existing across the entire range of fields and becoming fainter as the background magnetization increases. At higher fields, the carriers leak out of the filaments into the entire volume (fig. 3c) which is almost fully magnetized, and produces a homogeneous ferromagnetic phase. The percolation at 3.9 T has no signature since nothing dramatic happens for the magnetic configuration.

In conclusion, we propose here that a ferromagnetic interaction due to electron hopping is responsible for the phase separation in resistive manganites. The random walk motion of the charge carriers leads to the appearance of magnetic filaments which were evidenced by neutron scattering in $Pr_{0.66}Ca_{0.33}MnO_3$ single crystals and supported by Monte-Carlo simulations.

References

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