COMPLEX 2D MAGNETIC CORRELATIONS IN MANGANITES STUDIED BY SANS

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The colossal magnetoresistance (CMR) effect – the spectacular change of the resistivity in an applied magnetic field from insulating to metallic state- is an interesting unresolved problem in condensed matter physics [1]. Such CMR properties are widely studied in doped perovskite manganites, $A_{1-x}B_xMnO_3$, where A is a trivalent ion (La^{3+}, Pr^{3+}, etc) and B a divalent ion (Ca^{2+}, or Sr^{2+}). A physical problem raised in the manganese oxides is the competition between the double-exchange interaction, which favors the ferromagnetism (F) and super-exchange, which favors the antiferromagnetism (AF). From the competition between these two interactions, it was first proposed (in 1955) that a canting appeared in the magnetic structure. Recently, calculations suggested that the ground state of the CMR compounds could be an inhomogeneous electronic phase separation [2]. An elegant manner to interpret the CMR properties is a percolation of a metallic ferromagnetic phase in an insulating antiferromagnetic matrix, percolation that might be induced by a small change of the fraction or of the arrangement of the ferrodomains. This percolation scenario responsible of the CMR effect is a fascinating problem, which has attracted a lot of physicists.

Among the manganites, the $Pr_{1-x}Ca_xMnO_3$ series is one of prime interest, because Pr and Ca are about the same size and hence minimize the cationic size mismatch effect. Neutron diffraction technique is very well suited to determine their magnetic phase diagram. Figure 1 shows the different states depending on the x value. For the higher Mn^{3+} contents (typically x=0.2), the compounds are insulating ferromagnetic (FMI) at low temperature, and for larger x values (typically x=0.4) they are orbital ordered, antiferromagnetic CE like type. In between, the compositions x~0.3 show a mixing of F and AF phases as shown by neutron diffraction [3]. They also display CMR properties in close relation to the existence of the phase separation, which is now well proved by magnetoresistance, magnetization and specific heat studies [4].

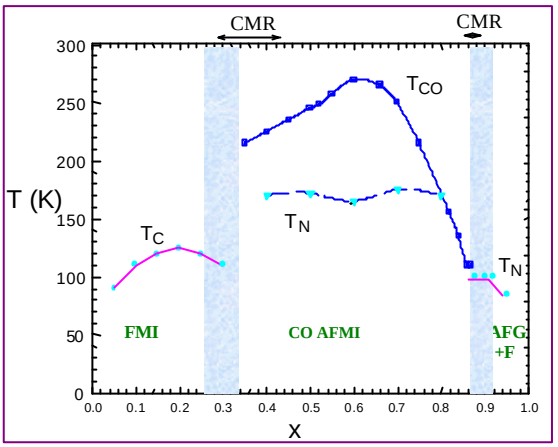

Figure 1. Magnetic phase diagram of $Pr_{1-x}Ca_xMnO_3$ compounds. The compositions close to x~0.3, which display colossal magnetoresistance, have an inhomogeneous low-temperature insulating state, where ferromagnetism (FMI) antiferromagnetism, and charge ordering (CO AFMI) coexist. Compositions close to 0.9 also have CMR properties.

As example, for the composition x=0.33, changes up to 10 orders of magnitude in the resistivity are observed at low temperature under an applied magnetic field of 7T. In revenge, the experimental situation which determines the size of the objects is not clear. Some authors have observed small nanometric ferromagnetic clusters by small angle neutron scattering (SANS), while others found much larger phase separated domains by electron microscopy or neutron diffraction. In principle, SANS is a very good technique to study the size and the shape of the domains of nanometric size and the magnetic contrast between a ferromagnet and an antiferromagnet is very large (the AF does not scatter at small angle).

Recent SANS experiments have been performed on the PAXY spectrometer at LLB to study sintered powder and single crystal samples of different compositions, x=0.20, 0.33 and 0.37. Classical magnetization and transport measurements were done to check their qualities and magnetic behaviours. x=0.2 is isolating and ferromagnetic, x=0.33 presents both ferromagnetic

and antiferromagnetic transitions and x=0.37 is mainly antiferromagnetic (see figure 1). The comparison of their SANS intensities is very interesting. Indeed, at low temperature, for the samples of composition x=0.33, the scattering signal show a q^{-2} dependence in the high q regime. This q dependence is typical of that due to infinite planar ferromagnetic sheets in an antiferromagnetic matrix [5]. The structure envisaged for the composition x=0.33, which displays CMR properties, is that of an isotropic "cabbage" as pictured in figure 2.

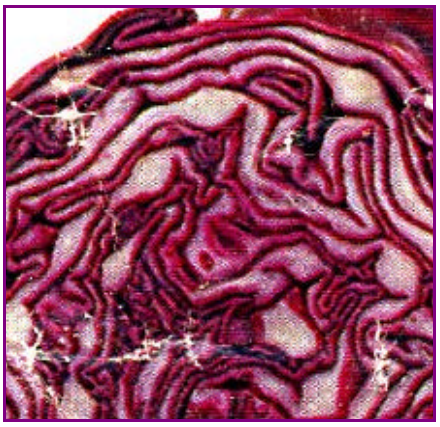

Figure 2. Cross section of a red cabbage to describe the 2D sheet structure of the F/AF phase separation in $Pr_{0.67}Ca_{0.33}MnO_3$ sample. At 30K, the phase separation between 1/3 F and 2/3 AF phase fractions occurs between thin ferromagnetic sheets of thickness of about 2.5nm.

The 2D thin sheets in this structure model could be the stripes proposed in the recent theories.

By combining magnetization and neutron diffraction data, we can determine the F and AF phase fractions in the samples. Knowing these magnetic fractions, the quantitative analysis of the SANS signal allows us to determine the ferromagnetic layers thickness of this peculiar 2D sheet structure, of about 2.5nm at 30K.

Further SANS experiments have been achieved to test the role of the magnetic field on such a nanoscopic structure. We have thus determined the magnetic field dependence of the thickness of the different magnetic domains in the phase separated x=0.33 system. The observed changes correspond to a gradual irreversible switch from AF sheets to F ones, transforming the system to complete ferromagnetic state at 6T [6]. At about 3T, the system reaches the percolation of the metallic ferromagnetic phase. The resistance decreases very rapidly, indicating the colossal magnetoresistance appearance.

Finally, SANS and classical magnetic techniques are powerful tools to study the electronic microphase separation occurring the manganese oxides.

References

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