EVOLUTION OF THE FERROMAGNETIC AND NON-FERROMAGNETIC PHASES WITH TEMPERATURE IN PHASE SEPARATED $La_{1-x}Sr_xCoO_3$ BY HIGH FIELD ^{139}La NMR

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Intrinsic phase separation in doped transition metal oxides is important in determining the unusual properties in these materials. An important example is colossal magneto-resistance in the manganites. Previous ^{59}Co NMR [1,2] and Small Angle Neutron Scattering (SANS) has demonstrated, for $La_{1-x}Sr_xCoO_3$, that ferromagnetic (FM) and non-ferromagnetic (NFM) phases co-exist for a wide range of doping levels. Co ions are present in different charge (Co^{3+} and Co^{4+}) and spin states (0,1,2) leading to a rich variety of properties as a function of temperature T and applied magnetic field H, including large negative magneto-resistance for $x < 0.2$ and a very large anomalous Hall effect. Using ^{139}La NMR it has been possible to determine the evolution of all three phases, ferromagnetic (FM), spin glass (SG), and paramagnetic (PM) over a wide range of temperature and x.

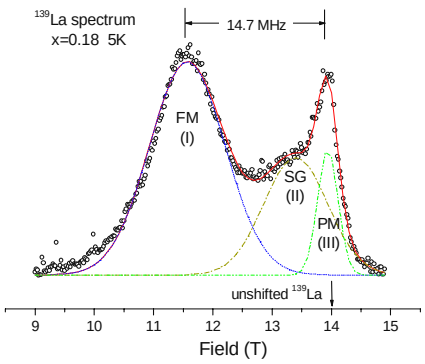

Fig. 1. Typical ^{139}La spectrum showing Gaussian fits for $x = 0.18$, $T = 5$ K.

Fig. 1 shows a typical ^{139}La spectrum and the Gaussian fits corresponding to the three phases. The spectra were taken by scanning H while keeping spectrometer frequency and sensitivity constant so the relative area of each Gaussian is proportional to the relative concentration of the corresponding phase. The low frequency line comes from the FM component, with the large negative shift due to the negative hyperfine coupling, the broad intermediate signal is from the SG, and the narrow unshifted line comes from paramagnetic regions. The large width of the FM and SG lines are due to a wide distribution of hyperfine couplings. Data were taken from 2.2 to 280 K and adjusted using the Boltzmann factor. The results are shown in Fig. 2.

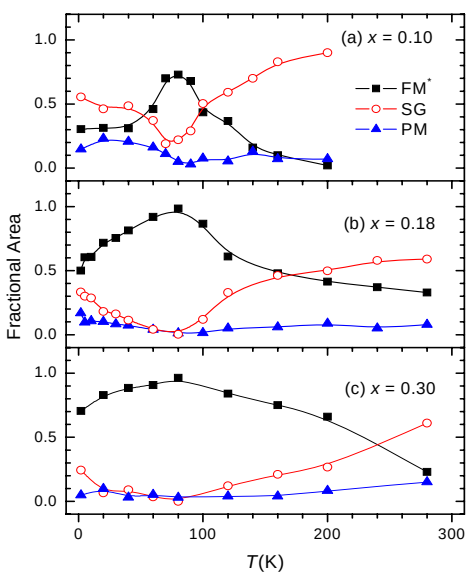

Fig. 2. ^{39}La Gaussian areas (proportional to the relative concentration) for peaks I (FM or possibly spin polarons at high temperature), II (SG) and III (PM) versus T for (*a*) $x = 0.10$, (*b*) $x = 0.18$ and (*c*) $x = 0.30$.

The proposed model is that metallic FM clusters imbedded in a SG or cluster glass matrix, with some residual low spin, hole poor, regions. A percolation limit is reached at the MI transition, $x = 0.18$. SANS data, through correlation length measurements, yields a maximum FM cluster size of order 3 nm essentially independent of x. The correlation lengths estimated from NMR T_1 measurements are consistent with this and are also independent of x [Ref. 2]. Note that the FM fraction grows at the expense of the spin glass component with increasing] T up to 80 K, and then decreases. The concentration trends are also reflected in resistivity data. The maximum in the FM component is likely to be due to Co^{3+} low-spin to intermediate-spin conversion that leads to growth in the FM cluster size. Above 90 K there appears to be a conversion from FM to SG, which is not currently well understood. Also, the "FM" component persists appreciably above the temperature at which the SANS data shows the FM correlation length goes to zero, so it may well be that at the highest temperatures the NMR shifts are due to magnetic polarons, which have been previously postulated to occur in these systems.

References

[1] Kuhns, P.L., *et al.*, Phys. Rev. Lett., 91, 127202 (2003).
[2] Hoch, M.J.R., *et al.*, Phys. Rev. B, 69, 014425 (2004).