A new electronic phase of $Pr(Sr_{1-y}Ca_y)_2Mn_2O_7$ deduced from high energy x-ray studies

T.A.W. Beale, P.R. Dean, P.D. Hatton and M. v Zimmermann[1]

Department of Physics, Durham University, South Road, DH1 3LE Durham, UK

[1]HASYLAB at DESY, Notkestrasse 85, 22603 Hamburg, Germany

Modern day life relies on high density information storage and retrieval. Dramatic improvements in information density have been obtained by utilising magnetic domain reversal in thin film media [1]. Spintronic memory devices such as in an ipod rely on the writing and reading of small magnetic domains. Research is now being conducted into using not the orientation of magnetic domains but of the orbital occupancy and direction [2]. The advantages of such a device would be much higher densities and potentially much faster read/write times. However controlling the orbital occupancy and direction is not easy. Previous attempts have relied on the application of epitaxial stress on thin films. Recently it was demonstrated that in a half-doped bilayer manganite $Pr(Sr_{0.1}Ca_{0.9})_2Mn_2O_7$ (PSCMO) a spontaneous reorientation of orbital stripes by 90° could be induced by temperature, with a phase transition close to room temperature [3]. The phase diagram of $Pr(Sr_{1-y}Ca_y)_2Mn_2O_7$ as a function of temperature vs. composition ($0.0 < y < 1.0$) is shown in Figure 1. This phase diagram was determined principally by resistivity and magnetization measurements [4] and the orbital stripe directions of the CO2 and CO1 phases (see Figure 2 taken from [5]) determined by electron diffraction and x-ray oscillation photographs. We have used the high-resolution diffraction capabilities of BW5 to study the phase diagram for a number of different compositions (y = 0.0, 0.4, 0.8, 0.9 and 1.0) to provide a clear structural basis for the phase diagram. Our results broadly agree with the phase diagram of Tokanuga *et al*., [4] but are very different, and far more complex, for $y = 0.8$ (see related P09 report).

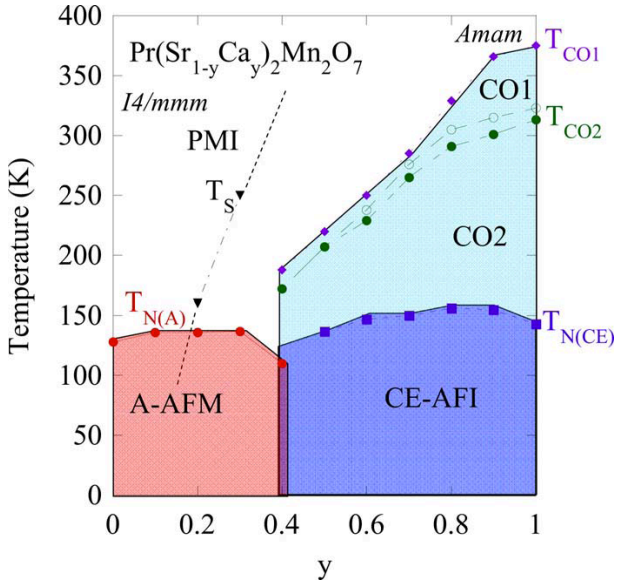

Figure 1: Electronic phase diagram of $Pr(Sr_{1-y}Ca_y)_2Mn_2O_7$ ($y = 0 – 1.0$). T_{CO1} and T_{CO2} with thermal hysteresis and T_N of A-AFM ($T_{N(A)}$) and CE-AF ($T_{N(CE)}$) were determined from resistivity and magnetisation measurements. T_s represents the structural transition from tetragonal to orthorhombic [diagram taken from ref. [4]]

The differentiation between the CO1 and CO2 phases is the relative orientation of orbital stripes as shown in Figure 2. Of course high energy x-rays are not directly sensitive to such effects but are sensitive to charge disproportionation and subtle lattice distortions. The results for both $y = 0.0$ and $y = 1.0$ samples (the end members of the phase diagram shown above) were entirely consistent with the proposed phase diagram. In the $y = 0$ sample only integer Bragg peaks were observed at all

temperatures and no effects were seen of the magnetic transition at T_N. For the y = 1.0 sample splitting of the Bragg peaks was observed at all temperatures indicative of the presence of orthorhombic twins. Again no changes were apparent at T_N but the CO2 to CO1 transition was observed by a first-order transition at 340 K and a change of the lattice parameters. In addition superlattice peaks at (h±1/2, k, l) were observed at all temperatures. These are probably caused by Jahn-Teller distortions causing a doubling of the *Amam* orthorhombic unit cell. These did not disappear until 380 K suggesting that the transition from the orthorhombic CO1 orbitally ordered phase to the tetragonal paramagnetic insulating phase occurs at this temperature.

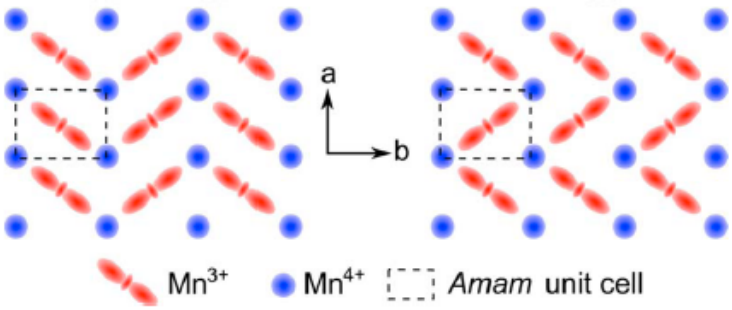

Figure 2 Schematic of the orbital stripes running along the *a*-axis in the CO1 phase and along the *b*-axis in the CO2 phase. Notice that the formation of the orbital stripes causes a doubling of the chemical *Amam* unit cell and hence superlattice reflections at half-integer positions [5].

For the y = 0.4 sample which according to the published phase diagram is on the boundary between the tetragonal and orthorhombic phases we found only Bragg peaks corresponding to a single tetragonal phase. Scans along h- and k-directions (see Figure 3) displayed half integer superlattice reflections caused by Jahn-Teller distortions. Such peaks are indicative of charge ordering within a tetragonal phase. These reduced in intensity with increasing temperature and disappeared at ~180 K, presumably at a tetragonal CE-AFI to PMI transition.

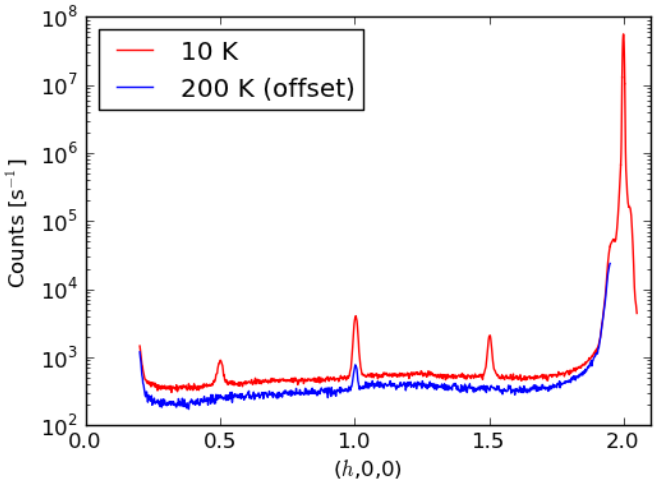

Figure 3. Recprocal lattice scans in the [100] direction at 10 K and 200 K. The Jahn-Teller reflections at (0.5,0,0) and (1.5,0,0) disappear completely, whereas a small reflection at (010) remains due to a very weak allowed Bragg reflection.

Our results suggest the existence of a new groundstate of PSCMO around y = 0.4 - 0.5 consisting of a 2 x 2 supercell tetragonal charge ordered (probably CE type) phase which contains $Mn^{3+}O_6$ Jahn-Teller distorted polyhedra. Such a phase is reminiscent of that found in $La_{2-2x}Sr_{1+2x}Mn_2O_7$ (x = 0.5) [6] and forms a natural link with other bilayer manganite phases.

References

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