FIELD INDUCED METAL-INSULATOR TRANSITION IN (PR:CA:SR)MnO_3

J. Hemberger, M. Paraskevoupolos, J. Sichelschmidt, M. Brando, R. Wehn, F. Mayr, K. Pucher, P. Lunkenheimer, and A. Loidl
Experimentalphysik V, Elektronische Korrelation und Magnetismus, Universität Augsburg, D - 86159Augsburg, Germany

A.A. Mukhin[1] and A.M. Balbashov[2]
[1] *Moscow Power Engineering Institute, 105835 Moscow, Russia*
[2] *General Physics Institute of the Russian Acad. Sci., 117942 Moscow, Russia*

Abstract In the system $Pr_{0.65}Ca_{0.28}Sr_{0.07}MnO_3$ (PCSM) the field induced metal-insulator transition exhibits changes of the resistivity up to 11 orders in magnitude. In order to study this colossal magnetoresistance effect (CMR) we carried out measurements on a PCSM single crystal, applying a broad range of experimental methods. Resistivity, magnetization and magnetic AC-susceptibility were measured in magnetic fields up to 140 kOe and in the temperature range 1.5 K $< T <$ 800 K. With decreasing temperature and in zero magnetic field PCSM undergoes a sequence of phase transitions from a paramagnetic into a charge ordered insulating and finally antiferromagnetic phase. The conductivity in these phases is dominated by variable range hopping processes. By applying magnetic fields $H \geq 10$ kOe a ferromagnetic metallic phase is induced. This transition exhibits an extremely marked hysteresis behavior. Below $T \approx 100$ K metastable states can be generated, in which ferromagnetic metallic and insulating phases coexist.

1. INTRODUCTION

At present in the field of transition-metal oxides a lot of attention is payed to materials exhibiting the colossal magnetoresistance effect (CMR), stimulated by the observation of this effect in manganite films [1]. The system (Pr:Ca)MnO_3 shows very pronounced CMR features. In this system the metal-insulator transition (MIT) not only can be driven by a magnetic field, but the transformation of the insulating (I) into a metallic (M) and ferromagnetic (FM) phase can also be x-ray, photo- or electric field induced [2]. The system

J. Bonča et al. (eds.), Open Problems in Strongly Correlated Electron Systems, 247–252.

$Pr_{1-x}Ca_xMnO_3$ has been investigated in detail with respect to its structural, magnetic and transport properties [3, 4]. The (x, T)-phase diagram shows a orthorhombic (O) phase at high temperatures. For $x < 0.3$ a Jahn-Teller distorted O' phase develops at lower temperatures, while for concentrations $0.3 < x < 0.9$ the structural groundstate is tetragonal. For $x > 0.9$ the system stays orthorombic in the whole temperature range. Between $0.3 < x < 0.7$ an charge- and orbitally ordered state has been proposed [5]. For $x > 0.3$ the system is insulating (I) and charge order (CO) transition is followed by an antiferromagnetic (AFM) transition while for $x < 0.3$ a ferromagnetic (FM) metallic (M) phase is established [4]. The magnetic field induced transition from the AFM/CO/I state into the FM/M state is strongly of first order and shows large hysteresis effects. This MIT also can be induced by internal chemical pressure substituting Ca by Sr [6]. The reported structural and electronic properties for $Pr_{0.7}Ca_{0.3}MnO_3$ are as follows [7]: The CO transition at $T \approx 200$ K is followed by an AFM transition ($T_N \approx 150$ K). At lower temperatures the evolving of a FM component is reported, which was ascribed to a canting of the AFM order but equally well can be described assuming electronic phase separation into AFM and FM clusters, which is believed to play an important role in manganite CMR compounds [9].

To optimize the system with respect to its CMR properties, we tried to tune the Sr:Ca ratio to get as close as possible towards the border-line between the FM/M state and AFM/CO/I state in zero external magnetic field. In the following we discuss the results obtained from investigations of single crystals of the $Pr_{0.65}Ca_{0.28}Sr_{0.07}MnO_3$ (PCSM).

2. RESULTS AND DISCUSSION

The PCSM single crystals were grown by the floating-zone method, details are described elsewhere [8]. The magnetic susceptibility and the magnetization were measured using an Oxford AC-susceptometer in an Oxford cryostat for fields up to 140 kOe. The electrical resistivity was measured employing a four probe technique in the same cryostat.

In Fig. (1) we show the $B(T)$-phasediagram of PCSM, determined from resistivity and magnetization measurements, which are illustrated in part in Fig. 2. In zero magnetic field the system undergoes a sequence of phase-transitions: At $T_{CO} \approx 210$ K a transition between two PM/I states takes place. Evidence for the inset of charge order is reported in similar systems [6]. At the same time structural changes are reflected in a clear change of the phonon-spectra [8]. At $T_N = 160$ K AFM order sets in and below $T_{irr} \approx 100$ K the system gets strongly irreversible and hysteresis phenomena with respect to field and temperature appear. In this regime a metastable FM metallic state can be induced by applying an external magnetic field. As pointed out later it can be

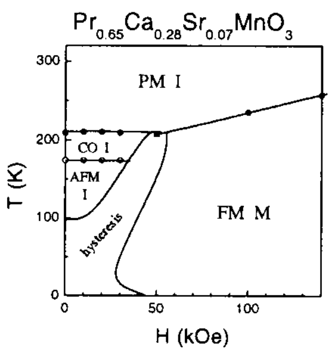

Figure 1 $B(T)$-phasediagram for $Pr_{0.65}Ca_{0.28}Sr_{0.07}MnO_3$. The 'hysteresis'-area reveals metastable behaviour.

assumed that these irreversibilities are introduced by phase separation into FM clusters within an AFM background.

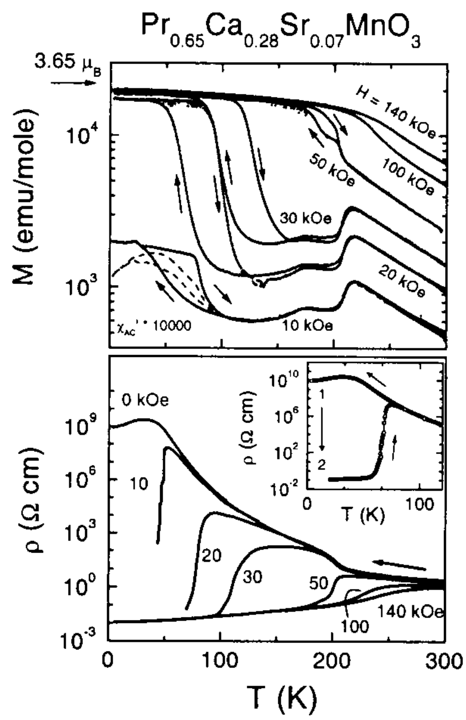

Figure 2 Resistivity and magnetization as a function of temperature for various fields up to 14 T. The dashed line (upper frame) displays the AC-susceptibility. The inset sows the thermo-remanent behaviour of the resistivity. The data was taken in zero field. Between (1) and (2) a field of 5 T was switched on for several minutes.

Fig. 2 displays the temperature dependence of the resistivity (lower frame, only cooling) and the magnetization (upper frame) for various fields up to

140 kOe and in addition the AC-susceptibility (dashed line in the upper frame). The CO transition can well be identified in the resistivity as well as the magnetization curves. For fields below 50 kOe the resistivity rises on cooling across T_c while the magnetization drops. A more detailed analysis of the DC-resistivity in zero magnetic field reveals that the underlying transport processes can be ascribed to variable range hopping (VRH) for temperatures 50 K $< T <$ 200 K and is dominated by the hopping of adiabatic small polarons for temperatures 220 K $< T <$ 650 K above T_{CO} [8].

The AFM transition only can be detected in the magnetization and susceptibility curves. Applying fields $H \geq$ 10 kOe a ferromagnetic component is induced. This phenomenon is accompanied by large hysteresis effects and a drop of the resistivity up to 11 orders of magnitude. The temperature of induced FM increases with increasing magnetic field and for $H >$ 50 kOe it is shifted above T_{CO}. It is interesting to note, that for intermediate fields ($H =$ 20 kOe, $H =$ 30 kOe) a small hysteresis of the magnetization extends up to 200 K. The saturation value of the magnetization in applied magnetic field is found to be close to the expected value of $M_S = 3.65\ \mu_B$ for $Mn^{3+}_{0.65}$ and $Mn^{4+}_{0.35}$. No additional contribution of the Pr^{3+} ions can be detected. At the same time the effective moment of $\mu_{eff} = 5.35\ \mu_B$ evaluated from the reciprocal susceptibility data up to 800 K (not shown) is clearly enhanced compared with the manganese-only expectation (μ_{eff}(Mn) $= 4.57\ \mu_B$). This fact implies that the Pr^{3+} moments order antiferromagnetically at low temperatures (see the anomaly in χ_{AC} and M(10 kOe) near $T \approx$ 25 K).

The inset of Fig. 2 shows the resistivity measured in zero external field on cooling and heating. Between the points (1) and (2) the FM/M state was induced by applying a magnetic field of $H =$ 50 kOe. After switching off the field the system remains metallic up to approximately 70 K. One should mention that in spite of the persistence of the conductivity at low temperatures the detected macroscopic magnetization returns to zero after switching off the external field in an analogous experiment (not shown). This can be explained by domain effects according to the soft magnetic character of the ferromagnetism in this system. The metastable conducting state below this temperature decays with time, as it is illustrated in Fig. 2. Fig. 2 shows the magnetic AC-susceptibility (upper frame) and resistivity (lower frame) versus time after switching off the magnetic field $H =$ 50 kOe at $T =$ 66 K. With increasing time χ_{AC} decays smooth and continuously. At the same time in the resistivity sharp steplike jumps appear. Such behavior can not be explained in the framework of a homogeneous canted AFM state but has to be considered as clear evidence for a inhomogeneous, phase separated state with FM. The susceptibility of the FM state is higher than that of the AFM state. The decay of $\chi_{ac}(t)|_{H=0}$ documents the vanishing fraction of ferromagnetic sample volume and can be described as stretched exponential behavior $\chi(t) = \chi_{FM}\left(1 - \exp\left(-\left(t/\tau\right)^{\beta}\right)\right) + \chi_{AFM}$

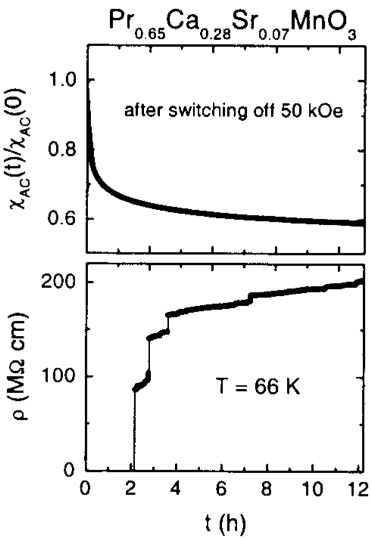

Figure 3 Time dependence of the normalized AC-susceptibility and the resistivity at $T = 66$ K after switching off a magnetic field of $H = 50$ kOe.

with $\beta \approx 0.3$. The steps in the resistivity denote the breakdown of conducting percolation paths. Phase separation scenarios in manganites are documented in literature experimentally and theoretically [8]. In the present case it stays unclear, if the groundstate of the system is intrinsically phase separated or if the observed phenomena are only due to induced metastable inhomogeneities with respect to the nearly degenerate AFM/CO/I and FM/M states of the system.

3. SUMMARY

In the system $Pr_{0.65}Ca_{0.28}Sr_{0.07}MnO_3$, which was optimized with respect to the CMR properties, investigations of the DC-resistivity, the magnetic AC-susceptibility and the magnetization were carried out in fields up to 140 kOe. This system is very close to the boundary between a FM metallic and a AFM charge-ordered insulating phase. In this system metal insulator-transitions of more than 11 orders of magnitude in resistivity can be induced by applying an external magnetic field of $H \approx 10$ kOe. At low temperatures ($T < 100$ K) the system becomes strongly irreversible and metastable transport and magnetization phenomena appear. Those nonergodic states are related to the coexistence of nearly degenerate FM/M and AFM/CO/I phases. In this regime the nature of the transport properties appears to be determined by percolation processes.

Acknowledgments

This work was supported in part by the Sonderforschungsbereich 484 of the Deutsche Forschungsgemeinschaft.

References

[1] R. von Helmolt *et al.*, Phys. Rev. Lett. **71**, 2331 (1993); S. Jin *et al.*, Science **264**, 413 (1994).

[2] Y. Tomioka *et al.*, J. Phys. Soc. Jpn. **64**, 3626 (1995); V. Kiryukhin *et al.*, Nature **386**, 813 (1997); K. Miyano *et al.*, Phys. Rev. Lett. **78**, 4257 (1997); A. Asamitsu *et al.*, Nature **388**, 50 (1997)

[3] Z. Jirak *et al.*, J. Magn. Magn. Mat. **53**, 153 (1985).

[4] Y. Tomioka *et al.*, Phys. Rev. B **53**, R1689 (1996).

[5] E.O. Wollan and W.C. Koehler, Phys. Rev. **100**, 545 (1955).

[6] H. Yoshizawa *et al.*, Phys. Rev. B **55**, 2729 (1997).

[7] H. Yoshizawa *et al.*, Phys. Rev. B **52**, R13145 (1995); D.E. Cox *et al.*, Phys. Rev. B **57**, 3305 (1998).

[8] J. Sichelschmidt *et al.*, to be published

[9] A. Moreo *et al.*, Science **283**, 2034 (1999); Uehara *et al.*, Nature **399**, 560 (1999); I.F. Voloshin *et al.*, 71, 106 (2000)