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$\text{Nd}_{0.62}\text{Pb}_{0.38}\text{MnO}_3$

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Pressure dependence of the resistivity and magnetoresistance in single-crystal $\text{Nd}_{0.62}\text{Pb}_{0.30}\text{MnO}_{3-\delta}$

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Abstract. We have measured the resistivity and magnetoresistance of single-crystal $\text{Nd}_{0.62}\text{Pb}_{0.30}\text{MnO}_{3-\delta}$ as a function of hydrostatic pressure up to 12 kbar. The pressure dependence of the resistivity at temperatures near the metal–insulator transition exceeds $d(\ln \rho)/dP \sim 3 \text{ kbar}^{-1}$, greater than that observed in the normal state of high-temperature superconductors. The extreme pressure sensitivity of the resistivity below the transition temperature provides evidence that the low-temperature phase is not a simple metal. The metal–insulator transition temperature T_{MI} decreases by 2.6 K kbar^{-1} , in accord with an expectation for greater pressure sensitivity in lower-doped materials with lower T_{MI} . The peak magnetoresistance defined as $[\rho(0 \text{ T}) - \rho(7 \text{ T})]/\rho(7 \text{ T})$ decreases with increasing pressure as the metal–insulator transition becomes less abrupt. The influence of thermal expansion upon the resistivity is expected to be large.

Recently there has been much interest [1–13] in the magnetoresistance (MR) of perovskite manganites with stoichiometry $\text{A}_{1-x}\text{B}_x\text{MnO}_{3-\delta}$ where A is a lanthanide ion, B is either an alkaline earth or lead and $x \simeq 0.2\text{--}0.5$. Both practical and fundamental physical questions remain regarding the mechanism of the unusually high MR in the manganites. The peak MR is associated with a transition from a high-temperature paramagnetic insulating phase to a low-temperature ferromagnetic metal. The temperature of peak resistivity T_{MI} varies with doping level x and the oxygen stoichiometry with a maximum T_{MI} at intermediate doping levels. Application of a magnetic field increases the transition temperature and decreases the resistivity, particularly near T_{MI} , sometimes by orders of magnitude. Hydrostatic pressure also increases T_{MI} and decreases the resistivity dramatically near T_{MI} [13–16].

Our present investigation of resistivity and MR as functions of pressure reveals significant differences between the effects of pressure and magnetic field. The resistivity is sensitively pressure dependent even at temperatures far from T_{MI} , with values of $d(\ln \rho)/dP$ near and below the transition exceeding values observed for the low-carrier-density copper oxide high- T_c superconductors [17]. Our investigations also extend previous studies on polycrystalline materials which indicated that the MR is larger for materials with lower T_{MI} . The detailed pressure dependence of the resistivity hints that the high-temperature activated regime and a re-entrant activated phase at the lowest temperatures may be manifestations of similar conduction processes.

Single crystals of $\text{Nd}_{0.62}\text{Pb}_{0.30}\text{MnO}_{3-\delta}$ were grown using a flux method following previous work [18, 19]. The crystal chosen for this study was a regular parallelepiped of dimensions $1 \text{ mm} \times 0.14 \text{ mm} \times 0.14 \text{ mm}$ with smooth reflective surfaces. X-ray

diffraction showed a single cubic phase with lattice constant 3.86 Å. Energy dispersive x-ray measurements on several parts of the sample showed a uniform stoichiometry of $\text{Nd}_{0.62}\text{Pb}_{0.30}\text{MnO}_{3-\delta}$ [20]. Current leads were attached with silver epoxy to the smaller end faces of the crystal. Voltage leads were attached 0.4 mm apart along the long axis. The resistivity was first measured at zero pressure from 10 to 300 K with and without a 7 T magnetic field applied perpendicular to the long axis of the crystal. The sample was next mounted in a self-clamping pressure cell with Flourinert 75 as a pressure medium, and a clamping pressure of approximately 6 kbar was applied at room temperature. The resistivity was again measured from 10 to 300 K with and without an applied magnetic field. The pressure inside the cell drops monotonically with increasing temperature because the thermal expansion coefficient of the cell is smaller than that of the pressure medium. A calibrated manganin coil monitored the pressure inside the cell as a function of temperature. The pressure decreased by 2–3 kbar as the temperature was lowered from 300 to 10 K. The procedure was repeated for clamping pressures of approximately 8 and 12 kbar so that resistivity $\rho(P)$ and $\text{MR}(P)$ as functions of pressure were obtained.

Figure 1 shows the resistivity as a function of temperature in zero applied magnetic field for zero initial pressure and three different clamping pressures. The sharp narrow transitions are characteristic of a uniform high-quality crystal. Comparison of cooling and warming data show no hysteresis in temperature. The temperature T_{MI} of peak resistivity increases with application of pressure. The increased stability of the magnetic phase under pressure is consistent with the double-exchange model [21–23] in which ferromagnetic order is encouraged by an increase in electronic hopping integrals [13, 14, 24]. The resistivity drops monotonically with increasing pressure at all temperatures. The general trend to decreased resistivity is consistent with an increase in electronic overlap under pressure; the detailed behaviour of $\rho(P)$ above and below the transition merits further discussion.

As has been seen previously for a manganite of stoichiometry $\text{Nd}_{0.57}\text{Pb}_{0.39}\text{MnO}_3$ [25], the zero-pressure high-temperature conduction shown in figure 1 has a form similar to that expected for variable-range hopping, $\ln \rho \propto T^{-1/4}$ [26]. Variation in the polaron on-site energies due to spin and cation disorder can yield a hopping conductivity with an additional $T^{-1/4}$ term which could account for the measured resistivity [25]. The pressure variation in the cell as a function of temperature acts oppositely to the direct thermal expansion of the sample in determining the temperature dependence of the sample volume. The counteracting affects of direct thermal expansion and pressure variation ensure that the correction to constant volume in this context is small and the approximately $T^{-1/4}$ behaviour observed is not an artefact of variations in sample volume [27]. Pressure seems to decrease preferentially the pre-factor ρ_0 , exerting a smaller effect on the exponent T_0 in the expression $\rho = \rho_0 \exp[(T_0/T)^{1/4}]$. The pre-factor $\rho_0 \sim \sqrt{\alpha/N(E_F)}$ (α being the inverse size of the polaron [26]) is more sensitive to changes in the density of states than is the slope $T_0^{1/4} \sim (\alpha^3/N(E_F))^{1/4}$, hinting that perhaps pressure predominately affects the density of states [28]. A decrease in pre-factor is consistent with an increase in the density of states and/or an increase in the size of the polaron under pressure, both consistent with pressure destabilizing the polarons. For illustrative purposes we note that a polaron size of $\alpha^{-1} \simeq 4$ Å implies a density of states at zero pressure of $N(0) \simeq 10^{20} \text{ cm}^{-3} \text{ eV}^{-1}$.

We take this opportunity to address the effects of thermal expansion on the detailed temperature dependence of the resistivity, an issue that to our knowledge has not been accorded sufficient attention up to this time. The extreme pressure sensitivity of the resistivity in these materials strongly suggests that thermal expansion has an unusually large influence on the temperature dependence of the resistivity. Taking a rough guess at a bulk modulus of about 1000–2000 kbar, and using our results for $(d\rho/dP)$ ($P = 0$)

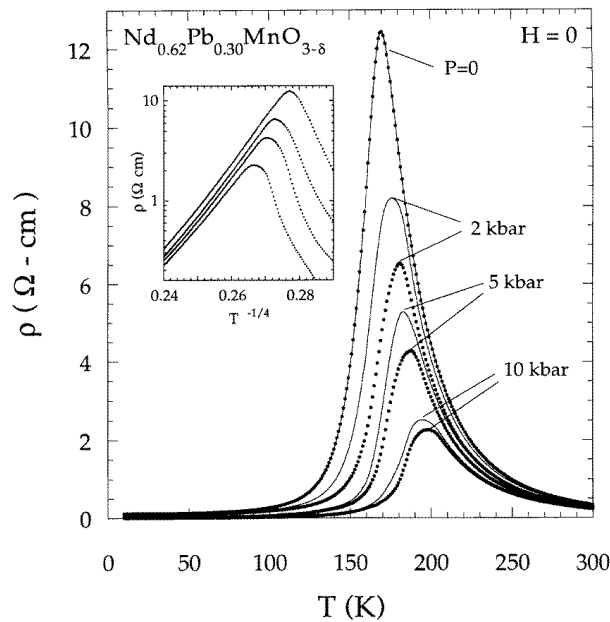


Figure 1. Resistivity of single-crystal $\text{Nd}_{0.62}\text{Pb}_{0.30}\text{MnO}_{3-\delta}$ as a function of temperature in zero applied magnetic field for clamping pressures of 0, 6, 8 and 12 kbar. The full circles are the experimental data, labelled by the pressure at 10 K. The pressure increases by 2–3 kbar as the temperature is increased to 300 K. The solid lines are corrected to constant ($T = 10$ K) pressure by interpolating the uncorrected data. The inset shows variable-range hopping behaviour at high temperatures. The constant-pressure correction is shown for illustrative purposes; the theoretically relevant constant-volume condition is actually closer to the raw variable-pressure data.

and a high-temperature linear thermal expansion coefficient of about $1.0 \times 10^{-5} \text{ K}^{-1}$ [1], the measured resistivity at 150 K and $P = 0$ is a factor of approximately 2 larger than the resistivity would have been if the $T = 50$ K volume had been maintained [29]. This upward curvature in the resistivity sharpens the low-temperature side of the metal–insulator transition. For higher temperatures the pressure sensitivity is smaller; the resistivity at 300 K is roughly about 1.05 times larger than it would have been if it had maintained the $T = 200$ K volume. Detailed modelling of the temperature-dependent resistivity should take into account the pronounced effects of thermal expansion, which contributes a substantial positive temperature coefficient to the resistivity, independent of explicitly temperature-dependent conduction processes. Correction for this effect awaits careful measurement of the bulk modulus and thermal expansion as functions of temperature.

Figure 2 shows the resistivity versus temperature in an applied magnetic field of 7 T for different clamping pressures. T_{MI} again increases with increasing pressure. The decrease in resistivity under pressure is not as dramatic as in zero field, since the resistivity peak itself is suppressed and broadened under field. Figure 3 shows T_{MI} as a function of pressure in both zero and 7 T fields, revealing a linear relationship with the same slope, 2.6 K kbar^{-1} , at zero and finite field. Since the free-energy stabilization of the low temperature phase is likely to be linear in the characteristic hopping integral, the linear relation between T_{MI} and pressure suggests that the hopping integral varies roughly linearly with pressure over the range studied [24]. This slope for our sample with $T_{MI} = 170$ K is larger

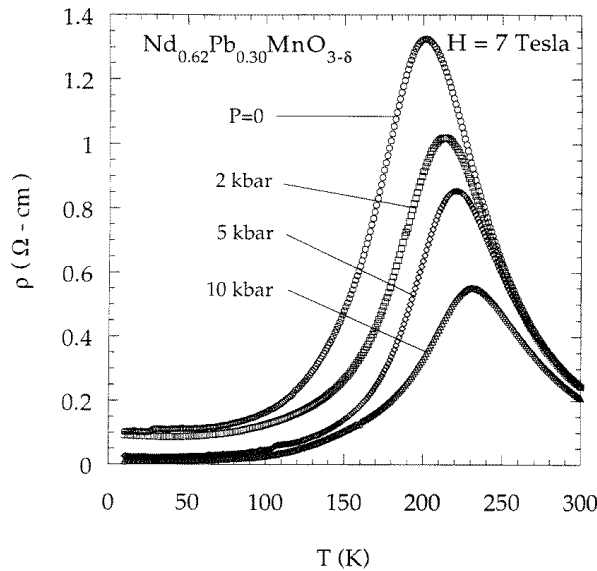


Figure 2. Resistivity of single-crystal $\text{Nd}_{0.62}\text{Pb}_{0.30}\text{MnO}_{3-\delta}$ as a function of temperature in a 7 T magnetic field for clamping pressures of 0, 6, 8 and 12 kbar. Curves are labelled with the pressure at $T = 10$ K. The pressure increases by 2–3 kbar as the temperature is raised to 300 K.

than the value $dT_{MI}/dP = 2.0$ K kbar $^{-1}$ reported previously on a single-crystal sample of $\text{Nd}_{0.5}(\text{Sr}_{0.36}\text{Pb}_{0.14})\text{MnO}_{3-\delta}$ with $T_{MI} = 204$ K at $P = 0$ and $H = 0$ [13]. This pair of results confirms and extends measurements on polycrystalline samples [14, 15] which indicated that dT_{MI}/dP is larger for samples with lower T_{MI} .

The MR defined as $[R(0 \text{ T}) - R(7 \text{ T})]/R(7 \text{ T})$ for different clamping pressures is shown in figure 4. The MR peak is suppressed and moved to higher temperatures under pressure. This behaviour is similar to the variations in MR and T_{MI} with doping level; in general, higher values of the MR are associated with lower values of T_{MI} , since at lower temperatures the metal–insulator transition is necessarily more abrupt. The MR above T_{MI} at different pressures is not a universal function of temperature and field; the provisional factorized form of ρ in [13] is not upheld by the present work. The slight positive MR under pressure at the lowest temperatures may be related to the usual positive MR observed for normal metals.

A compilation of published results [13] has shown a clear inverse trend between T_{MI} and the maximum MR. Some insight into this relationship can be gained by looking at simple phenomenological forms for the low- and high-temperature resistivities. Taking the phenomenological expressions $\rho \sim a + \rho_1 T^3$ for the low-temperature phase and $\rho \sim \rho_0 \exp[(T_0/T)^{1/4}]$ for the high-temperature phase [30], and assuming that the magnetic field is large enough to induce a complete transition from the insulating to the metallic phase at the temperature of maximum MR, one obtains a maximum MR at T_{MI} of $\text{MR} \sim \{\rho_0 \exp[(T_0/T_{MI})^{1/4}] - a - \rho_1 T_{MI}^3\}/(a + \rho_1 T_{MI}^3)$. High-temperature activated conduction creates the divergence as $T_{MI} \rightarrow 0$. The MR at high temperatures disappears near a temperature at which the metallic-like resistivity exceeds the activated resistivity.

Within either a lattice polaron [12, 13, 25, 31] or an Anderson localization [32] model, the decrease in average electronic hopping rate upon entry to the high-temperature spin-disordered state induces electron localization. Within double exchange the low-temperature

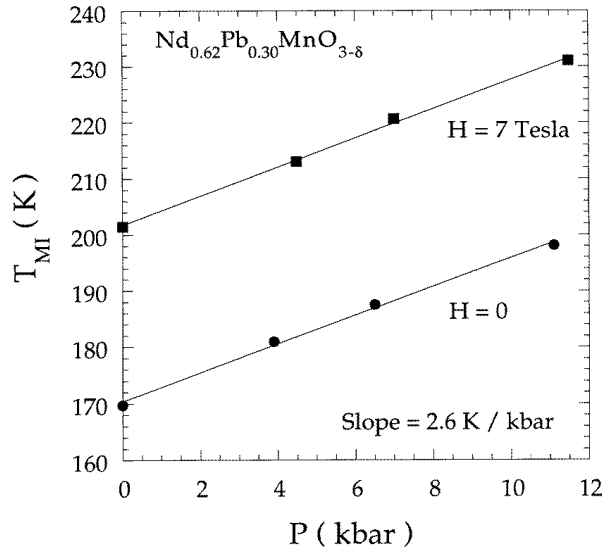


Figure 3. Temperature T_{MI} of peak resistivity versus pressure for zero applied field and $H = 7$ T. Application of the magnetic field shifts the curve upwards but does not significantly affect the slope.

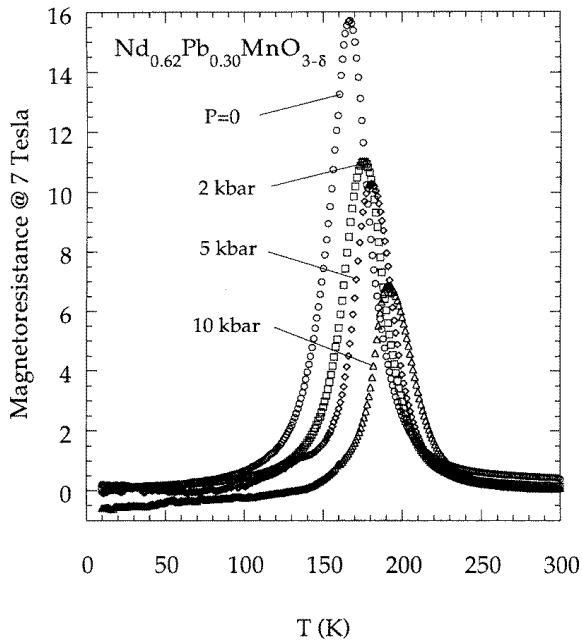


Figure 4. Temperature dependence of the MR $[R(7\text{ T}) - R(0\text{ T})]/R(7\text{ T})$ of single-crystal $\text{Nd}_{0.62}\text{Pb}_{0.30}\text{MnO}_{3-\delta}$ at four different clamping pressures. Curves are labelled by the pressure at $T = 10$ K.

spin-ordered state is stabilized by the decrease in electronic kinetic energy upon relaxation of the energy constraint against electrons occupying opposite-spin ionic sites. The application of field clearly favours the low-temperature spin-ordered state. The application of moderate pressure favours the low-temperature spin-ordered state by increasing the importance of electronic hopping in the total energy of the system. Pressure also directly destabilizes the high-temperature localized phase by increasing the electronic hopping rate between sites. Both the magnetic field and the external pressure increase T_{MI} and are expected to cause a large change in the resistance at temperatures near the transition.

Far from the transition temperature the pressure continues to depress the resistivity strongly whereas the magnetic field has little effect. The magnetic field affects the bare electronic hopping rate through changes in the magnetization which are only significant near T_{MI} . Pressure, in contrast, increases the bare hopping rate at all temperatures. The bare hopping rate determines the activation energy, with the activation energy for conduction a sensitive function of average electron on-site residency time through the magnetic transition.

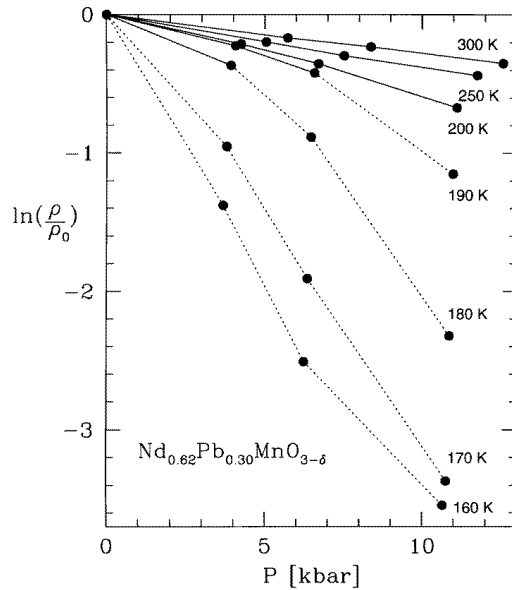


Figure 5. The logarithmic resistivity versus applied pressure for temperatures near and above the magnetic transition. The solid lines refer to resistivities in the high temperature insulating phase. The broken lines refer to samples in the low-temperature metallic phase. Note the nearly linear behaviour, except across the metal–insulator transition. ρ_0 is the zero-pressure resistivity.

In figure 5 a plot of the logarithm of the resistivity versus pressure near and above the zero-pressure magnetic transition temperature reveals a roughly exponential dependence of resistivity on pressure, as would be expected for hopping conduction if pressure has a roughly linear relation to the spatial extent and/or activation energy of localized electronic states away from the transition, a supposition which is consistent with a linear increase in T_{MI} with increasing pressure. The solid lines in figure 5 denote the behaviour in the insulating phase while the broke lines denote the behaviour in the metallic phase. The crossing of the metal–insulator transition as a function of pressure for temperatures of 180 and 190 K induces the deviation from exponential pressure dependence visible in these curves.

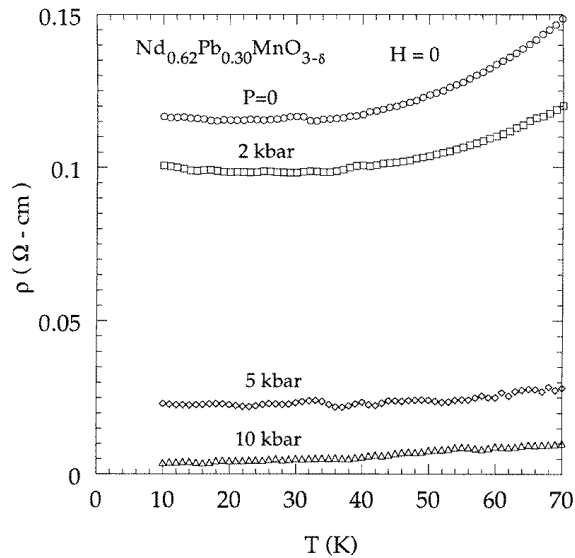


Figure 6. Low-temperature resistivity for the four different clamping pressures. The upturn in the resistivity at lowest temperatures and pressures is suppressed by the application of greater than 5 kbar pressure. Curves are labelled by the pressure at $T = 10$ K.

The resistivity at lower temperatures retains a strong pressure sensitivity (up to $d(\ln \rho)/dP \simeq 3 \text{ kbar}^{-1}$), as shown in the resistivity curves in figure 6. The pressure derivative $d(\ln \rho)/dP$ at low temperatures and moderate pressures is as large as that observed near T_{MI} . This sensitivity away from the transition may seem surprising since at low temperatures the system is securely fastened in a magnetically ordered state which is ostensibly metallic. A clue towards this pressure sensitivity is perhaps apparent from the upturn in the resistivity at the lowest temperatures and pressures in the sample studied. Measurements on polycrystalline samples with lower doping levels show a similar but more pronounced upturn at low temperatures[33]. Underdoped compounds can show a re-emergence of hopping conduction below the resistive transition which suggests that the increase in bare electron hopping rate at temperatures below the transition is sometimes insufficient to unbind the polarons. The continuous evolution of low-temperature activated behaviour as a function of doping suggests that the upturn observed in our more optimally doped samples has the same origin, a pressure-dependent incipient activation barrier in the nominally metallic low-temperature phase. As long as the hopping barrier is small or moderate compared with kT , the conduction can present a metallic-like temperature dependence, particularly since thermal expansion biases $\rho(T)$ upwards. A measure of covert hopping or ‘bad metal’ [34] behaviour is consistent with a large absolute value of the resistivity in the metallic-like phase. Figure 6 suggests that the low-temperature pressure dependence of ρ may be weaker at the lower pressures. A detailed correction for thermal expansion is impossible at this time, but the data suggest that lower values of $d\rho/dP$ (at both low and high temperatures) are associated with an activated constant-volume resistivity and very large values of $d\rho/dP$ are associated both with the vicinity of the transition and with regions of positive temperature coefficient of the resistivity. This common pressure dependence of both activated regimes suggests a common conduction mechanism.

Measurements of the pressure dependence of the resistivity in the high-MR doped

manganites signals the importance of thermal expansion and suggests a common kinship for the high- and low-temperature activated regimes. As observed in the high- T_c superconductors, large values of $d(\ln \rho)/dP$ are associated with behaviour characteristic of bad metals.

Acknowledgments

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