Tunneling and enhanced magnetoresistance in $Nd_{2/3}Sr_{1/3}MnO_3$ thin films with microcracks

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We found that microcracks in thin $Nd_{2/3}Sr_{1/3}MnO_3$ films create a series of intrinsic break junctions which are ideal for investigating tunneling phenomena in this system. A comparison of films with and without cracks, which have similar ferromagnetic Curie temperature (T_c) of 140–150 K, shows that the cracked film has a lower insulator to metal transition temperature T_M (97 K vs 140 K), three orders of magnitude higher resistivity at T_M, and two times larger magnetoresistance at 1 T near T_M. At $T>T_M$ we observed that $\ln \rho\alpha T^{-1/4}$ in the uncracked film while in the cracked film a $\ln \rho\alpha T^{-1/2}$ dependence was found. This indicates that the conductivity in the first case is due to variable range hopping in three dimension, while in the second case it is dominated by thermally activated tunneling across the insulating barriers (the microcracks). © *1998 American Institute of Physics.* [S0003-6951(98)02129-9]

Rare earth manganite perovskites of the type $Ln_{1-x}A_xMnO_3$ (Ln=rare earth ion, A=Ca, Sr, Ba, or Pb) are gaining much attention due to their giant magnetoresistance (MR).[1–3] Insulator–metal transition and paramagnetic–ferromagnetic transition can be obtained in the antiferromagnetic insulator $LnMnO_3$ by partial substitution of the trivalent rare earth ions by the divalent ions, which induces mobile charge carriers.[4,5] For applications developing of junctions from these materials with high MR at high temperatures and low fields is the challenge. At temperatures much lower than the insulator–metal transition temperature (T_M), single crystals and epitaxial thin films exhibit low MR, while polycrystalline samples show high resistivity and high MR. This effect is attributed to the spin polarized tunneling across the grain boundaries.[6–8] Recently, it has been shown that tunneling through artificial grain boundary[9] or insulating barrier[10] induces a significant MR effect at low temperatures and low fields. In this letter we study the zero field transport properties and magnetoresistance in $Nd_{2/3}Sr_{1/3}MnO_3$ (NSMO) films and compare the transport properties of the films with and without microcracks.

NSMO thin films were grown on (100) $SrTiO_3$ (STO) substrates by pulsed laser deposition (PLD) at 700 °C block temperature in 200 mTorr oxygen pressure using a well sintered target. We used a KrF excimer laser ($\lambda = 248$ nm) at 5 Hz that produced ≈ 2 J/cm^2 pulse energy density on the target. After deposition the films were cooled to 450 °C in 500 Torr oxygen, annealed at this temperature for 30 min, and then cooled to room temperature. The structure of the as deposited film was examined by x-ray diffraction measurements, the surface morphology was probed by an atomic force microscope (AFM), and the electrical resistivity was measured down to 20 K. Magnetoresistance measurements at different temperatures were carried out in a magnetic field up to 1.5 T.

X-ray diffraction of the films showed only [00(2n)] peaks with c-axis parameter of 7.56 Å (cracked film) and 7.64 Å (uncracked film), both smaller than the lattice parameter of bulk NSMO ($c = 7.72$ Å). The full width at half maximum (FWHM) of the rocking curve of the (004) peak of the films was found in the range of 0.095°–0.12°, confirming the good crystallinity. The pseudocubic lattice parameter of bulk NSMO is smaller than that of the STO substrate and their lattice mismatch is about 1.2%. This leads to a tensile stress in the a-b plane and compressional strain along the c axis of the film. The c-axis contraction in the present NSMO film is similar to that observed in the case of $La_{0.7}Ca_{0.3}MnO_3$ (LCMO) films on STO.[11]

Viewing the film in an optical microscope under back side illumination reveals orthogonal microcracks in some of the NSMO films. In Fig. 1, an AFM image of such a film is

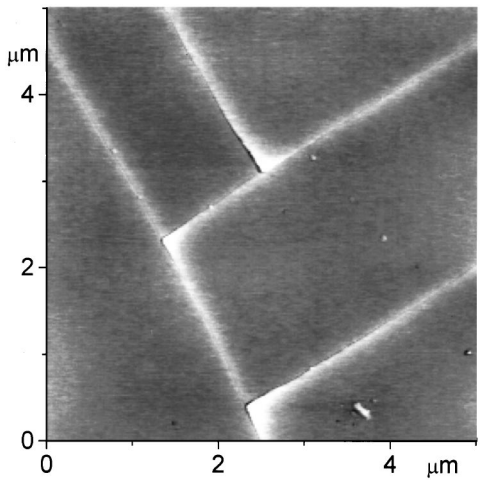

FIG. 1. An AFM image of a NSMO film with orthogonal microcracks.

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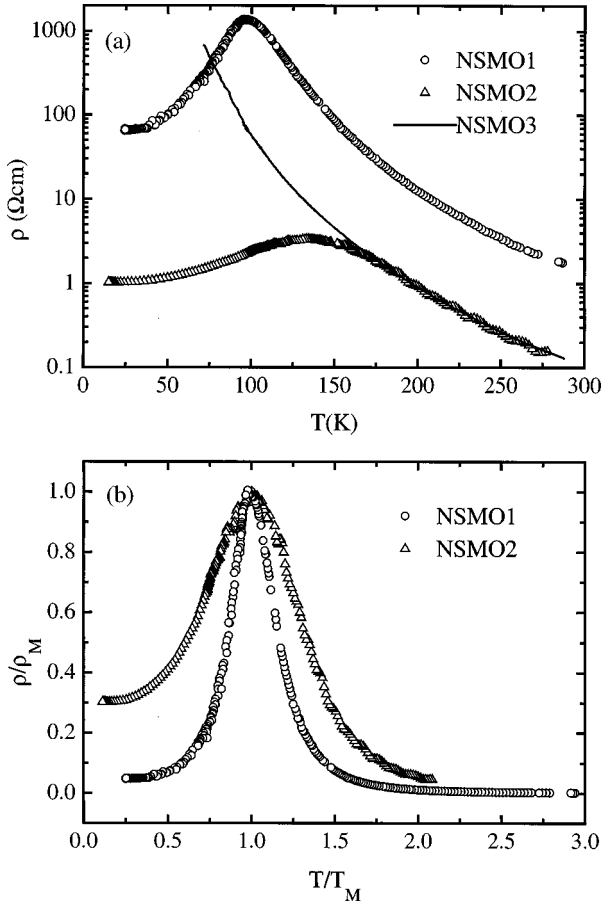

FIG. 2. (a) Resistivity as a function of temperature of cracked (NSMO1), crack free (NSMO2), and oxygen deficient (NSMO3) thin films. (b) Normalized resistivity (ρ/ρ_M) as a function of reduced temperature (T/T_M) for NSMO1 and NSMO2.

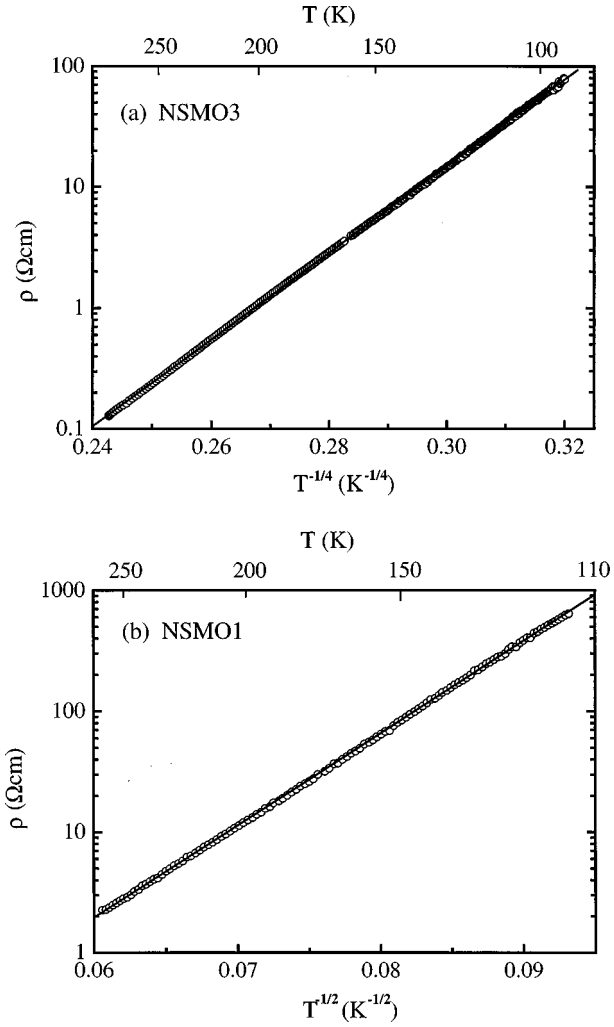

FIG. 3. (a) Plot of ρ vs $T^{-1/4}$ of the oxygen deficient film (NSMO3). (b) ln ρ versus $T^{-1/2}$ of NSMO1. The symbols represent the experimental data and the line is the theoretical fit.

shown. The orthogonal microcracks are a few micrometers apart and in the uncracked regions the root mean square (rms) surface roughness is less than 2 nm. In the uncracked films the rms roughness is larger (>4 nm). Higher smoothness can be one of the reasons for cracking.

In Fig. 2(a), we show three plots of ρ versus T, of a cracked film (NSMO1), an uncracked film (NSMO2) and an oxygen deficient film (NSMO3). NSMO1 shows a peak resistivity of 1.4 kΩ cm at its T_M of 97 K. For $T>T_M$ a negative temperature coefficient (ntc) of the resistivity is observed, while for $T<T_M$ a positive temperature coefficient of the resistivity (ptc) is found with $\rho_{97\,\mathrm{K}}/\rho_{20\,\mathrm{K}}>20$. NSMO2 shows a much higher insulator to metal transition temperature (140 K) with about three orders of magnitude lower peak resistivity (3.5 Ω cm) and $\rho_{140\,\mathrm{K}}/\rho_{20\,\mathrm{K}}$ of 3.5. However, magnetization measurements showed that saturation was reached around 140–150 K for both NSMO1 and NSMO2 indicating that ferromagnetic Curie temperature (T_c) of both types of films are virtually the same. This suggest that the higher resistivity of NSMO1 at all temperatures and the lowering of its T_M is due to the microcracks.

Generally, oxygen vacancies in these manganites cause an increase in the resistivity and a decrease of T_M.[12] It was therefore interesting to compare the temperature dependence of the resistivity of NSMO1 with that of an uncracked, oxygen deficient film with the same cation composition. Such a film (NSMO3) was prepared using the same deposition conditions as before but without the annealing step at 450 °C. As seen in Fig. 2(a) ρ (NSMO3) has ntc down to 50 K but the resistivity of NSMO1 in its ntc regime was much larger than that of NSMO3. At 100 K the ratio between the resistivities of these two samples is about two orders of magnitude. The temperature dependence of the resistivity of NSMO3 obeys Mott's variable range hopping (VRH) law $\rho=\rho\ \exp(T_0/T)^{1/4}$ over a temperature range of 200 K with $T_0\cong 5\times 10^7$ K [see Fig. 3(a)]. The behavior of NSMO2 in its ntc regime is very similar to that of NSMO3 (with similar T_0) but here the ntc regime is narrower because of the onset of the magnetic transition [see Fig. 2(a)]. In a systematic study of the well oxygenated $La_{1-x-y}Y_xCa_yMnO_3$ system[13] it was shown that T_0 increases monotonously as T_M decrease. If one plots ln T_0 vs T_M for the data of Ref. 13, our pair of values of T_0 and T_M falls on this curve within the scattering of their data confirming the reliability of our data as due to VRH in NSMO2. A completely different behavior of $\rho(T)$ was found for NSMO1, namely ln ρ showed a linear variation as a function of $T^{-1/2}$ [see Fig. 3(b)]. This kind of conductivity is generally observed in the dielectric regime of granular metal films. The metallic islands are separated by insulating barriers and the conductivity in these films is due to thermally activated tunneling across the insulating barriers

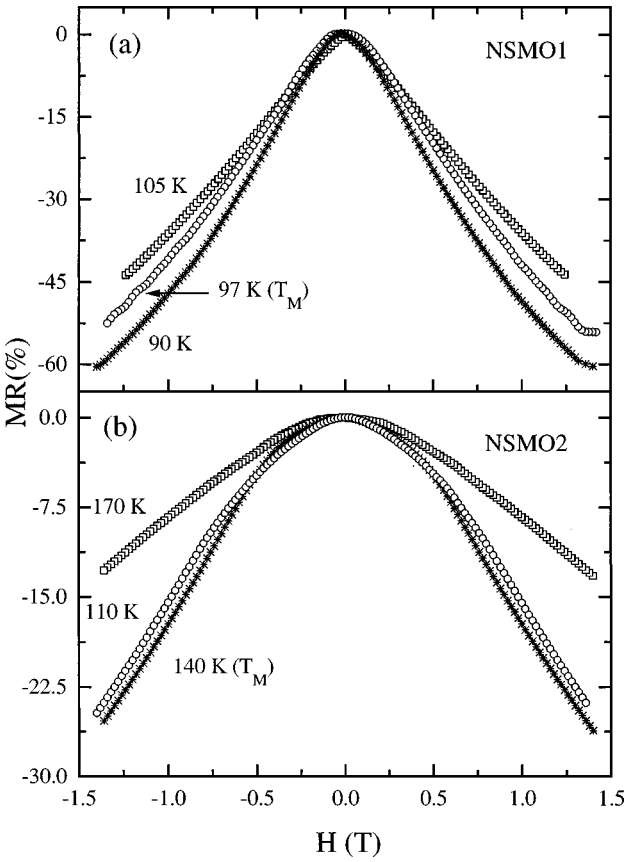

FIG. 4. Magnetoresistance as a function of magnetic field in NSMO1 (a) and NSMO2 (b).

which yields the $\ln \rho \alpha T^{-1/2}$ behavior.[14] Our observation of this behavior in NSMO1 suggests that microcracks in the films behave as the insulating barriers and govern the transport properties of the film.

Magnetoresistance measurements at different temperatures were carried out on NSMO1 and NSMO2 up to a field of 1.5 T and the results are shown in Figs. 4(a) and 4(b), respectively. In NSMO1 the MR is more than 65% at 1.5 T and more than 50% at 1 T with a sharp decrease in resistance with increasing field near T_M. In contrast, NSMO2 shows only a 25% change in resistance near T_M at 1.5 T. Earlier studies show that in the case of epitaxial manganite films, T_M and T_c are almost the same and the MR is high in the vicinity of the transition temperature and low at $T \ll T_c$. Even though the cracked film NSMO1 has a T_c of 140–150 K, we observed a maximum MR at 90 K (near T_M). On the other hand NSMO2 showed a maximum MR at 140 K (at $T_c \approx T_M$).

In polycrystalline manganite films and magnetic tunnel junctions, high MR at low temperatures has been observed as compared to that of epitaxial films and single crystals.[6–8] This effect has been explained on the basis of spin polarized tunneling across the grain boundaries. An enhanced MR at room temperature due to the spin polarized tunneling in $Fe/Al_2O_3/Fe$ junctions has been reported by Miyazaki and Tezuka.[15] Further a tunneling type resistivity and enhanced magnetoresistance in Co–Al–O granular films has been observed.[16] The microcracks in the NSMO films acts like tunnel barriers and therefore these films exhibit a tunneling type resistivity and the enhanced magnetoresistance may be attributed to the spin polarized tunneling.

The mechanism of microcracks formation in the NSMO thin films is still an open question. Rare earth manganites are known to show a sudden change in volume near their insulator–metal transition temperature.[17] Also, the NSMO films on STO are under tensile stress.[18] Probably because of these two factors some of the films develop microcracks mostly during the low temperature cycling. Further experimental study is required to understand the mechanism of microcrack formation.

In conclusion, we found that transport in thin NSMO films with microcracks occurs via a thermally activated tunneling process where the microcracks act as the insulating tunnel barriers. We also observed enhanced magnetoresistance near T_M, which demonstrates the high sensitivity of the MR to the local defects in this tunneling junction system.

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