

Oxygen annealing effect on $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ films deposited on LaAlO_3 , SrTiO_3 , and MgO substrates

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Extensive studies on thin films of perovskite type manganese oxides exhibiting colossal magnetoresistance (CMR) have revealed that their physical properties are quite different from their bulk congeners. The difference is largely associated with the structural strain induced by substrate as well as the oxygen defect. In order to the effects of the substrates on material properties involving CMR property, we have investigated the structural and physical properties of $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ (LCMO) films deposited on the (001) oriented SrTiO_3 , LaAlO_3 , and MgO substrates.

We have grown LCMO films on the three different substrates by pulsed laser deposition. KrF excimer laser was operated at the energy density of 2~3 J/cm² and the repetition rate of 3 Hz. During deposition, the substrates were held at 750 °C, while the oxygen partial pressure was controlled. The average thickness of film deposited for 10 min is estimated to be about 1000 Å. After deposition, some of the films were annealed under oxygen atmosphere. For comparison, we annealed the as-grown film under Ar atmosphere. We have studied physical properties of the LCMO films, with relation to effects of substrates and annealing under O₂ and/or Ar.

LCMO films grown on perovskite substrates were found to be grown epitaxially through XRD measurement. The *c*-axis lattice constants of as-grown films on SrTiO_3 and LaAlO_3 are 3.853 Å and 3.900 Å, respectively. The former is smaller than that of the LCMO target (3.877 Å) while the latter is larger. However, temperature dependent resistances for the films shown in left panel of Fig. 1 do not show any significant difference. After annealing the films at 900 °C under O₂ atmosphere, the metal-insulator transition temperatures (T_M) of the films becomes raised significantly with large MR values, which clearly demonstrates that transport properties are enhanced. After the O₂ annealing, *c*-axis lattice parameter of the film on LaAlO_3 decreases while that on SrTiO_3 increases, approaching to bulk value. The *c*-axis lattice constant in as-grown films is affected by lattice mismatch. Since lattice constant of LCMO bulk is less than that of SrTiO_3 , LCMO/ SrTiO_3 endures in-plane tensile stress whereas LCMO/ LaAlO_3 experiences compressive stress in reverse, resulting in the difference of *c*-axis lattice parameters in the as-grown films. Thus, it seems that the O₂ annealing not only increase T_M but also relieve the structural strains induced by the lattice mismatches in the as-grown films. Interestingly, after annealing LCMO/ LaAlO_3 under Ar, it is found that T_M is virtually unchanged but the *c*-axis contracts. Surprisingly the MR ratio of Ar-annealed film is increased, similar to that of the O₂-annealed film.

The O₂ annealing at high temperature drives two effects. One is the oxygen incorporation, which not only leads to the optimal doping level but also increases the Mn-O bond strength, accordingly

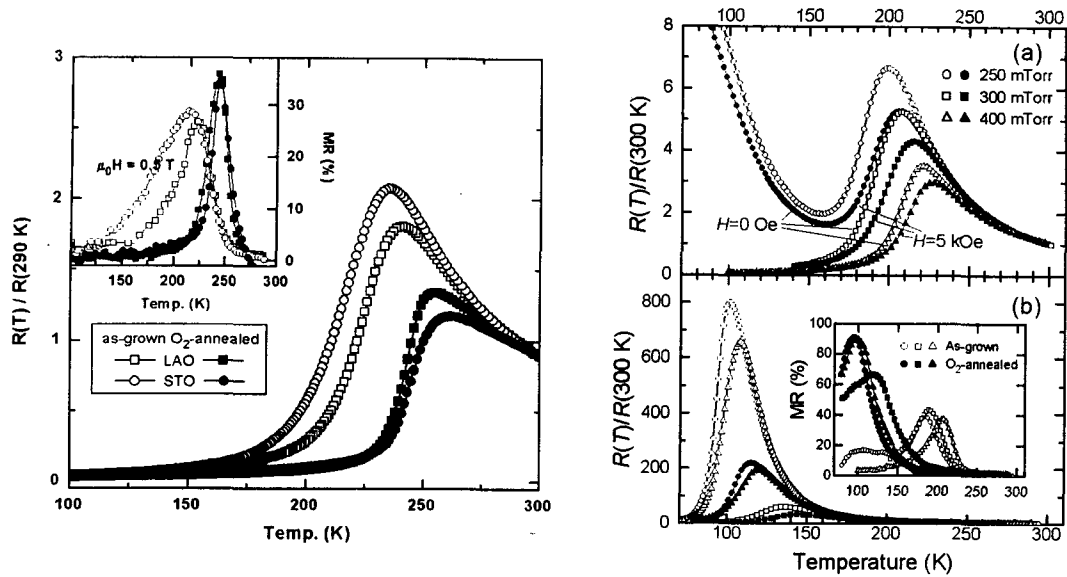


Figure 1. (left) The temperature dependent normalized resistance of as-grown (open symbol) an O_2 -annealed LCMO films (solid symbol) on $SrTiO_3$ (circles) and $LaAlO_3$ (rectangular). The inset shows MR ratios calculated as $[R(T,0Oe) - R(T,5kOe)]/R(T,0Oe)$. (Right) Normalized resistances for LCMO films deposited on MgO at different oxygen pressures. Top (a) and bottom (b) panels show the resistance curves of before and after O_2 annealing, respectively. Open symbols represent the resistance curve without applied magnetic field and Filled symbols denote that with a magnetic field of 5 kOe. Insets show MR ratios.

increasing the orbital overlap and in turn hopping electrons easily. The other is thermal effect, which would relieve the structural strain induced by lattice mismatch and also create microstructural change manifested by AFM images. The latter effect is exhibited also by Ar annealing. Thus, the MR, mainly associated with the microstructural change, is induced by the thermal effect rather than by the oxygen stoichiometry.

The as-grown films on MgO exhibit T_{MI} ranging from 200 K to 230 K, as shown in right panel of Fig. 1. Contrary to films on $SrTiO_3$ and $LaAlO_3$, T_{MI} of the film on MgO annealed under O_2 decreases by more than 100 K, featured with the colossal magnetoresistance. Furthermore, the O_2 -annealed films also show thermal hysteresis in resistivity and nonlinear I - V behavior. These results are interpreted in terms of the Mn deficiency induced by lattice mismatch between film and substrate, and also discussed in viewpoint of a percolative phase transition.

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