Recent experimental studies of the colossal magnetoresistive manganites \( \text{Ln}_{1-x}A_x\text{MnO}_3 \) (\( \text{Ln} \): trivalent rare earth ions; \( A \): divalent alkaline earth ions) have led to new information which suggests the relevance of lattice effects on the conductivity and magnetism of these manganites.\(^1\)–\(^5\) Some representative experimental observation includes a strong correlation between the thickness of epitaxial films and the corresponding magnetoresistance;\(^1\) decreasing Curie temperatures \( T_C \) and increasing colossal magnetoresistance (CMR) effects with the increasing lattice distortion via the substitution of \( \text{La} \) ions by smaller ions of \( \text{Pr} \) and \( \text{Y} \);\(^2\) a significant reduction of the magnetoresistance in single crystals under a hydrostatic pressure;\(^3\) as well as a large magneto-volume effect\(^4\) and a giant oxygen isotope effect\(^5\) in \( \text{La}_{1-x}\text{Ca}_x\text{MnO}_3 \). On the physical origin for the CMR, the importance of lattice polarons induced by the Jahn–Teller coupling, and that the low-temperature \( (T \ll T_C) \) magnetoresistance may be attributed to the magnetic domain wall scattering. In contrast, the absence of the Jahn–Teller coupling and the large conductivity in \( \text{La}_{0.5}\text{Ca}_{0.5}\text{CoO}_3 \) epitaxial films yield much smaller negative magnetoresistance, which may be attributed to disorder-spin scattering. © 1997 American Institute of Physics.

The effects of lattice distortion on the physical properties of \( \text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3 \) epitaxial films are investigated. Our results suggest that larger substrate-induced lattice distortion gives rise to larger zero-field resistivity and larger negative magnetoresistance. Similar effects are also observed in samples of different thicknesses and on the same substrate material, with larger resistivity and magnetoresistance associated with thinner samples. In addition to x-ray diffraction spectroscopy, the degrees of lattice distortion in different samples are further verified by the surface topography taken with a low-temperature scanning tunneling microscope. Quantitative analyses of the transport properties suggest that the high-temperature \( (T \rightarrow T_C) \) colossal magnetoresistance (CMR) in the manganites is consistent with the conduction of lattice polarons induced by the Jahn–Teller coupling, and that the low-temperature \( (T \ll T_C) \) magnetoresistance may be attributed to the magnetic domain wall scattering. In contrast, the absence of the Jahn–Teller coupling and the large conductivity in \( \text{La}_{0.5}\text{Ca}_{0.5}\text{CoO}_3 \) epitaxial films yield much smaller negative magnetoresistance, which may be attributed to disorder-spin scattering. © 1997 American Institute of Physics.

The LCMO and LCCO epitaxial films are grown by pulsed laser deposition using stoichiometric targets. For studying the substrate-induced lattice distortion, 200 nm thick films are grown on different (001) substrates at 700 °C in 100 mTorr of oxygen, and subsequently annealed at 900 °C of 1 atm oxygen for 2 h. The Curie temperature \( T_C \) for all LCMO is 260±10 K, and that for the LCCO is \( T_C=180±5 \) K. The lattice constants \( a \), \( b \), and \( c \) (c\perp sample surface) as well as the epitaxy of the films are determined using high resolution x-ray diffraction (XRD) and x-ray rocking curves, and the results have been given elsewhere.\(^6\) The substrate-induced lattice distortion consists of lattice strain and lattice relaxation.\(^7\) The former yields intrinsic effects on the phonon modes and magnetic exchange interaction, the latter yields extrinsic effects such as dislocations and domain walls. Another batch of LCMO films are grown...
FIG. 1. The effect of lattice distortion on the magnetoresistance $\Delta R_H$ as a function of the temperature ($T$) is shown for La$_{0.7}$Ca$_{0.3}$MnO$_3$ epitaxial films on different substrates of LaAlO$_3$, YAlO$_3$, and SrTiO$_3$. Here the applied field is $H=6.0$ T. The zero-field resistivity $\rho$ vs $T$ data for these samples are illustrated in the inset.

FIG. 2. STM images of 200 nm thick LCMO epilaxial films grown on LAO with a thickness of 100 nm, and under the same conditions described above except for two different growth rates controlled by the laser fluences. Among both the LCMO and LCCO films of the same thickness, we find the largest lattice distortion in LCMO/YAO and LCCO/YAO from the x-ray data. For LCMO/LAO samples with different thicknesses and under different growth rates, the thinner sample of 100 nm is found to exhibit larger lattice distortion. The chemical properties of these samples are further characterized with x-ray photoelectron spectroscopy (XPS). The room-temperature valence band spectroscopy shows no density of states at the Fermi level for the manganites and high density of states at the Fermi level for the cobaltites, consistent with the semiconducting nature of the former and metallic nature of the latter. The effects of substrate-induced lattice strain on the optical phonon modes have been revealed by our recent infrared reflectivity studies, and the frequency shifts of the phonon modes are consistent with the semiconducting nature of the former and metallic nature of the latter.

FIG. 3. Comparison of the thickness and growth rate dependence of the magnetoresistance $\Delta R_H(T)$ with $H=6.0$ T in LCMO/LAO films. The inset shows the corresponding zero-field resistivity $\rho(T)$ of the same samples.

FIG. 4. STM images of 100 nm thick LCMO epitaxial films grown on LAO substrates at (a) 2 J/cm$^2$ and (b) 1.6 J/cm$^2$ laser fluence. The images are 150×150 nm in size and the grey scales are 3.5 nm for (a) and 2.5 nm for (b), with rms surface roughness of 0.7 and 0.4 nm, respectively. The higher growth-rate sample (a) shows rounded "rice-paddy" terraces indicating island growth mode. The lower growth-rate sample (b) shows ragged "fish-scale" terraces suggesting a more step-flow growth mode. The terrace steps are typically 0.4 nm in height, consistent with the unit-cell lattice parameter. The less ordered surface morphology of the 100 nm films as compared with the 200 nm films (Fig. 2) is consistent with the higher lattice distortions in the former.
Assuming polaron conduction as the dominant conduction mechanism for LCMO at high temperatures, the resistivity data for all LCMO films are analyzed according to the expression

\[
\rho(T) = \alpha T \exp \left( \frac{E_b(T)}{k_B T} \right)
\]

(1)

where \(E_b\) is the polaron binding energy, \(\alpha\) a constant, and the temperature and magnetic field dependence of \(E_b\) satisfies the conditions imposed by the polaron model. That is, \(E_b \rightarrow 0\) in the limit of complete magnetic order when the increasing hopping rate of the itinerant electrons exceeds the optical phonon frequency, and \(E_b \rightarrow E_{b0} \sim \text{constant}\) in the absence of long-range magnetic order. For all LCMO films, the fitting to the high-temperature resistivity data yields \(E_{b0} \sim 0.35\) eV. This energy compares favorably to the Jahn–Teller coupling energy, suggesting that the high-temperature conduction mechanism is dominated by the lattice polaron conduction.

On the other hand, the low-temperature transport properties appear to be strongly correlated with the degree of lattice distortion. That is, samples of larger lattice distortion exhibit larger resistivity and magnetoresistance, suggesting increasing electron scattering due to a larger number of magnetic domains and grain boundaries induced by larger lattice distortion. The incompletely aligned moments of the magnetic domains due to either inhomogeneity or pinning by local defects below \(T_C\) give rise to larger scattering of conduction electrons. Therefore, an applied magnetic field has more significant effects on aligning the magnetic domains, thereby more effectively reducing the resistivity in samples with larger lattice distortion. The substrate-induced magnetic domains and grain boundaries are uniquely associated with the epitaxial films, as evidenced by the much larger low-temperature zero-field resistivity (see the inset of Fig. 1) and the significantly enhanced low-temperature magnetoresistance (Fig. 1) relative to those of the single crystals of LCMO. \(^{13}\)

To investigate the relevance of lattice polarons to the occurrence of CMR effects, the resistivity and magnetization of LCCO films on LAO and YAO substrates were studied. Despite comparable lattice relaxation and lattice strain in the manganites and in LCCO/YAO, \(^{9}\) the magnitude and temperature dependence of the resistivity in the LCMO and LCCO systems exhibit sharp contrasts, as illustrated in Figs. 1 and 5. Also shown in Fig. 5, for the LCCO/YAO sample, a faster decrease in the zero-field resistivity as well as a maximum in the magnitude of negative magnetoresistance both occur at approximately the Curie temperature \((T_C \sim 180\, \text{K})\), suggesting that magnetic ordering below \(T_C\) reduces the resistivity, \(^{9}\) and that the magnetoresistance in LCCO is of the origin of spin fluctuations rather than polaron conduction.

In summary, we have studied the effects of lattice distortion on the physical properties of \(La_{0.5}Ca_{0.5}MnO_3\) epitaxial films by varying the thickness of films on the same substrate, and by depositing the same thickness of films on various substrates with a range of lattice constants. Our studies reveal that larger substrate-induced lattice distortion gives rise to larger zero-field resistivity and larger negative magnetoresistance. The lattice distortion determined from the x-ray diffraction studies is further confirmed by the STM images of the surface topography. Our results suggest that the high-temperature \((T \rightarrow T_C)\) CMR in the manganites is consistent with the conduction of lattice polarons induced by the Jahn–Teller coupling, and that the low-temperature \((T < T_C)\) negative magnetoresistance can be attributed to the magnetic domain wall scattering. In contrast, the absence of the Jahn–Teller coupling and the large conductivity in \(La_{0.5}Ca_{0.5}CoO_3\) epitaxial films may account for the much smaller negative magnetoresistance.

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\(^{12}\) For the definition of the surface roughness, see, for example, M. Ohring, *The Material Science of Thin Films* (Academic, San Diego, CA, 1992).