Anisotropic magnetoresistive and magnetic properties of $La_{0.5}Sr_{0.5}CoO_{3-\delta}$ film

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The magnetic and transport properties of a $La_{0.5}Sr_{0.5}CoO_{3-\delta}$ film grown on a $LaAlO_3$ substrate by pulsed-laser deposition are studied. The properties are found to be influenced by the magnetic anisotropy and inhomogeneity. Magnetoresistance anisotropy is determined by the shape anisotropy of the magnetization and the strain-induced magnetic anisotropy due to the film–substrate lattice interaction. Indications of the temperature-driven spin reorientation transition from an out-of-plane ordered state at low temperatures to an in-plane ordered state at high temperatures as a result of competition between the aforementioned sources of magnetic anisotropy are found. [DOI: 10.1063/1.1596581]

1. INTRODUCTION

Mixed-valence lanthanum cobaltites of the type $La_{1-x}Sr_xCoO_3$ have attracted much attention in recent years due to their unique magnetic and transport properties.[1,2] Study of this system is also important for understanding the nature of colossal magnetoresistance in the related oxides, mixed-valence manganites.[3,4] For technical application, epitaxial films of these compounds are mainly used. In that case the shape anisotropy (due to the demagnetizing effect) and the film-substrate lattice interaction can induce magnetization anisotropy and, therefore, magnetoresistance (MR) anisotropy (bulk samples of these compounds show no marked magnetic or MR anisotropy). This point has been studied rather intensively in manganite films (see Ref. 5 and references therein). Hardly any studies of this type can be found in the literature for cobaltites. In addition, the properties of mixed-valence cobaltites are influenced by their unavoidable magnetic inhomogeneity, which arises due to different extrinsic and intrinsic causes. The extrinsic ones are determined by various technological factors in the sample preparation. They can cause inhomogeneity in chemical composition (for example in oxygen concentration) or in crystal structure (polycrystalline or granular samples). The intrinsic sources of inhomogeneity are believed to arise for thermodynamic reasons and can lead to phase separation into two phases with different concentrations of the charge carriers and, therefore, to significant magnetic inhomogeneity.[1,2,6] In this article we present a study of a $La_{0.5}Sr_{0.5}CoO_{3-\delta}$ film which demonstrates a combined influence of the magnetic anisotropy and inhomogeneity on its transport, magnetoresistive, and magnetic properties. Indications of the temperature-driven spin reorientation transition from an out-of-plane ordered state at low temperatures to an in-plane ordered state at high temperatures as a result of competition between the aforementioned anisotropy sources are found.

2. EXPERIMENTAL

The $La_{0.5}Sr_{0.5}CoO_{3-\delta}$ film (about 220 nm thick) was grown by pulsed-laser deposition (PLD) on a (001) oriented $LaAlO_3$ substrate. The ceramic target used was prepared by a standard solid-state reaction technique. A PLD system with an Nd-YAG laser operating at 1.06 μm was used to ablate the target. The pulse energy was about 0.39 J with a repetition rate of 12 Hz and pulse duration of 10 ns. The film was deposited at a substrate temperature of (880 ± 5)°C in an oxygen atmosphere at a pressure of about 8 Pa. The film was cooled down to room temperature after deposition at an oxygen pressure about 10^5 Pa. The target and film were characterized by an x-ray diffraction (XRD) study.

The film resistance was measured as a function of temperature and magnetic field H (up to 20 kOe) using a standard four-point technique. The field was applied parallel or perpendicular to the film plane. In both cases it was perpendicular to the transport current. The magnetization M was measured in a Faraday-type magnetometer. A rotating electromagnet made it possible to measure the magnetization for different directions of H relative to the plane of the film.

3. RESULTS AND DISCUSSION

We have found a strong anisotropy in magnetic and magnetoresistive properties of the film studied. The anisotropy manifests itself as dramatic differences in those properties for magnetic fields applied parallel and perpendicular to the film plane. Consider at first the anisotropy of the magnetic properties. The magnetization curves for the fields parallel ($H_{\parallel}$) and perpendicular ($H_{\perp}$) to the film plane demonstrate a strong anisotropy (Fig. 1). At the maximum field applied (7 kOe), the magnetization seems to be rather close to saturation for the in-plane field orientation, but it is far from it for the out-of-plane one. It is reasonable to suppose that this is determined mainly by the shape anisotropy.

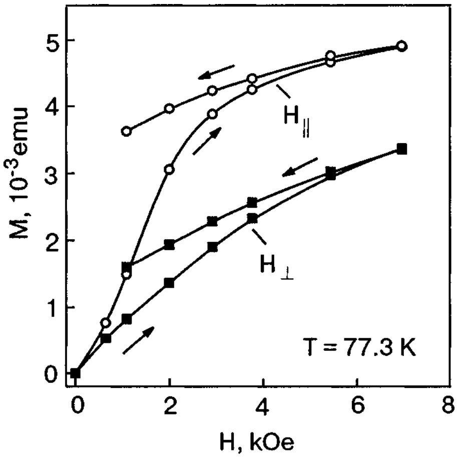

FIG. 1. Magnetization curves of the film studied for fields parallel ($H_\parallel$) and perpendicular ($H_\perp$) to the film plane.

The temperature dependence of the film magnetization for the field directions parallel ($M_\parallel$) and perpendicular ($M_\perp$) to the film plane is shown in Fig. 2. The Curie temperature T_C is found to be about 250 K. The $M_\parallel(T)$ behavior is quite common for ferromagnetic (FM) metals: it saturates with decreasing temperature. The behavior of $M_\perp(T)$ is quite different from that of $M_\parallel(T)$. At the fairly high field used, 2 kOe, the $M_\perp(T)$ curve is found to be well below the $M_\parallel(T)$ curve. Besides, in the low-temperature range the $M_\perp(T)$ curve is nonmonotonic (Fig. 2).

Figure 2 actually presents the $M(T)$ behavior only for two values of the angle θ between the field and the film plane: $\theta=0°$ and $\theta=90°$. It is helpful to consider the whole curves of the angle dependence of the magnetization, presented in Fig. 3a. Here the curves $M_{\mathrm{up}}(\theta)$ and $M_{\mathrm{down}}(\theta)$ were recorded with the field rotated in steps from 0° to 360° and back to 0°, respectively. It can be seen that the magnetization takes maximum values at $\theta\approx0°$, 180°, and 360°, that

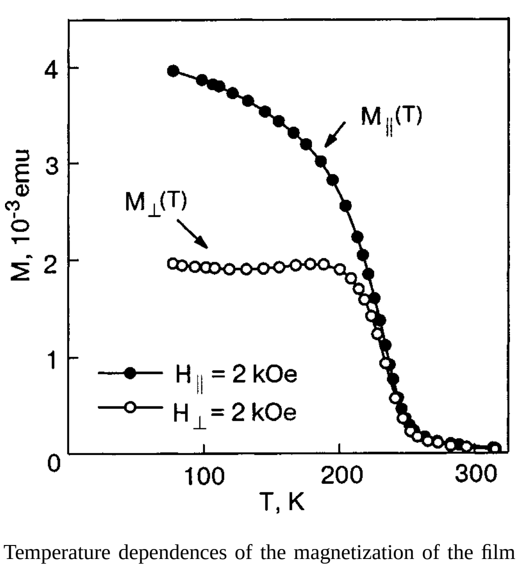

FIG. 2. Temperature dependences of the magnetization of the film studied for the magnetic field ($H=2$ kOe) applied parallel ($M_\parallel$) and perpendicular ($M_\perp$) to the film plane. The thermomagnetic prehistory: the sample was cooled down to liquid nitrogen temperature, $T\approx77.3$ K, in a field close to zero, and then the field was increased to 7 kOe and lowered to 2 kOe (see Fig. 1). After that the dependences were recorded at that field with the temperature increasing.

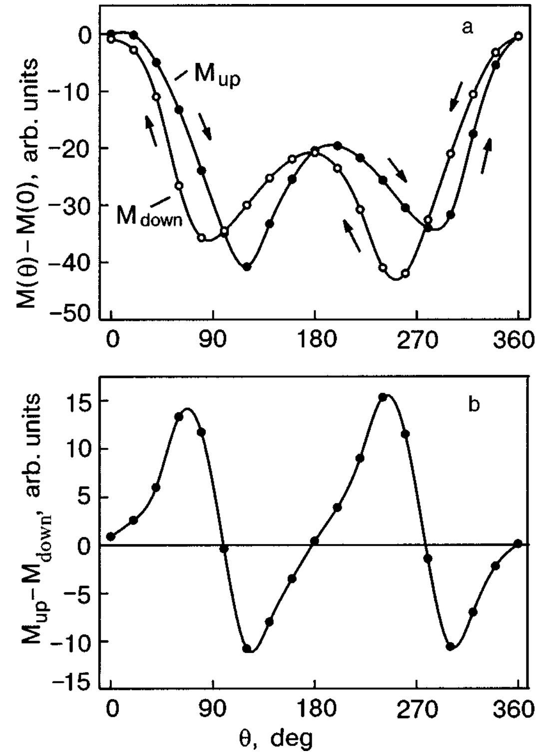

FIG. 3. Panel (a) presents the dependence of the magnetization on the angle θ between the magnetic field and the film plane (at $H=2$ kOe and $T=77.3$ K). The thermomagnetic prehistory is described in the caption to Fig. 2. The curves $M_{\mathrm{up}}(\theta)$ and $M_{\mathrm{down}}(\theta)$ were recorded with a stepwise rotation of the field from 0° to 360° and back to 0°, respectively. A considerable hysteresis effect can be seen. The angular dependence of the difference between $M_{\mathrm{up}}(\theta)$ and $M_{\mathrm{down}}(\theta)$ in panel (b) reveals this effect more clearly.

is, for the in-plane field orientations. The magnitude of the magnetization at $\theta\approx180°$ is less than those at $\theta\approx0°$ and 360°. This is determined by the shape of the magnetization loop and by the thermomagnetic prehistory of the sample. The minimum magnetization values are found, as expected, at $\theta\approx90°$ and 270°, that is, for the out-of-plane field orientations.

A considerable hysteresis effect in the $M(\theta)$ curves is seen (Fig. 3a). To present the effect more clearly, the angular dependence of the difference between the $M_{\mathrm{up}}(\theta)$ and $M_{\mathrm{down}}(\theta)$ is shown in Fig. 3b. The function $d(\theta)=M_{\mathrm{up}}(\theta)-M_{\mathrm{down}}(\theta)$ can be taken as some measure of the angular hysteresis effect. It is seen that the $d(\theta)$ dependence is close to a periodic one with a period equal to 180°. It takes zero value at the angles which are multiples of 90°, corresponding to both the in-plane and out-of-plane directions of magnetic field (Fig. 3b). The extreme values of $d(\theta)$ are situated at some intermediate angles, which are, however, closer to the out-of-plane directions than to the in-plane ones.

As indicated above, the magnetization anisotropy in the film studied should be determined mainly by the shape anisotropy. Closer inspection shows, however, that the $M_\perp(T)$ behavior cannot be attributed solely to the shape-anisotropy effect: $M_\perp(T)$ and $M_\parallel(T)$ are practically equal in a rather broad temperature range just below T_C, then (going to lower temperature) the $M_\perp(T)$ curve rather abruptly goes well below the $M_\parallel(T)$ curve and becomes nonmonotonic, with a pronounced increase in $M_\perp(T)$ at low temperatures (Fig. 2). These $M_\perp(T)$ features can be caused by the strain-induced

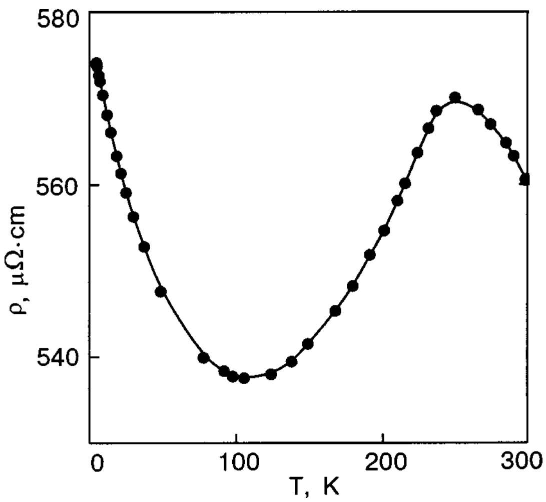

FIG. 4. Temperature dependence of the film resistivity.

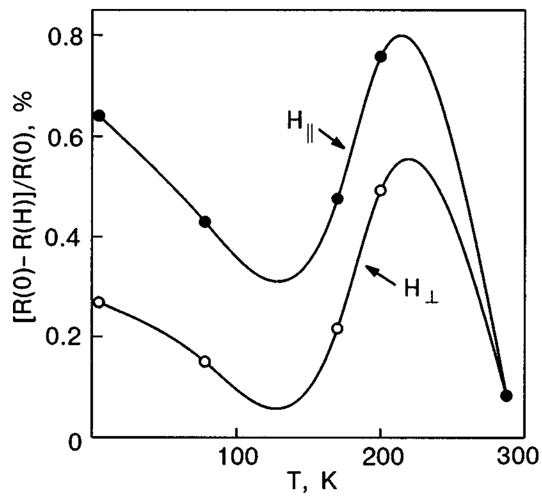

FIG. 5. Temperature dependence of the magnetoresistance at $H=20$ kOe for fields parallel ($H_{\parallel}$) and perpendicular ($H_{\perp}$) to the film plane. In both cases the fields were perpendicular to the transport current. The solid lines present a *B*-spline fitting.

magnetic anisotropy due to lattice mismatch between the film and the substrate. This guess is supported by our XRD study, which has revealed that the film has an out-of-plane tensile strain. For materials with positive magnetostriction this must favor an out-of-plane easy magnetization. Additional corroborations of this suggestion have been found in the MR properties of the film, described below.

Now turn to the transport properties of the film. The temperature dependence of the resistivity, $\rho(T)$, is found to be nonmonotonic with a maximum at $T\approx 250$ K and a minimum at $T\approx 107$ K (Fig. 4). $La_{0.5}Sr_{0.5}CoO_{3-\delta}$ samples with fairly perfect crystalline structure and δ close to zero are known[2] to be metallic ($d\rho/dT>0$) in the whole range below and above T_C. The $\rho(T)$ behavior in Fig. 4 reflects an inhomogeneous structure of the film and some oxygen deficiency. Due to the last factor, the hole concentration is less than the nominal one (at $\delta=0$). This is responsible for a resistance peak at $T=250$ K, which is common[2] for low-doped $La_{1-x}Sr_xCoO_3$ with $0.2\leq x\leq 0.3$. The low-temperature resistance minimum is typical for systems of FM regions (grains or clusters) with rather weak interconnections. For example, it has been frequently seen in polycrystalline manganites.[7–9] The inhomogeneous structure can be determined by technological factors of sample preparation (causing a polycrystalline structure with rather high tunneling barriers between the grains) or by phase separation into hole-rich and hole-poor phases.[1,2] The conductivity of inhomogeneous systems of this type is determined by the intragrain conductivity and the tunneling of charge carriers through the boundaries between the grains. A competition between these two contributions can lead to a resistance minimum.[8,9] For an extended discussion of the most obvious reasons for the appearance of the resistance minimum in polycrystalline cobaltites see Ref. 6.

The MR in the film studied is found to be anisotropic. The absolute values of negative MR in fields parallel to the film plane are considerably above those in perpendicular fields (Fig. 5). The temperature behavior of the ratio between the in-plane and out-of-plane MRs is shown in Fig. 6. It is seen from Figs. 5 and 6 that this MR anisotropy takes place only in the FM state and disappears for $T>T_C$. Since the conductivity of mixed-valence cobaltites increases with enhancement of the magnetic (spin) order, this behavior just reflects the circumstance that the magnetization increases more easily in a magnetic field parallel to the film plane, as has indeed been found in this study (Figs. 1, 2, and 3).

In polycrystalline samples (beside an intrinsic MR, which depends on magnetic order inside the grains) a significant contribution to the MR comes from grain boundaries, and this contribution increases with decreasing temperature. Discussion of the possible mechanisms for this extrinsic type of MR can be found in Refs. 10–13. The film studied does indeed show a continuous increase in MR with decreasing temperature (for temperatures well below T_C) (Fig. 5). This behavior is expected for polycrystalline FM samples with poor enough intergrain conductivity.[10,11] In contrast, for cobaltite and manganite samples with fairly good crystal perfection and even for polycrystalline samples of these materials but with a good intergrain connectivity, the MR goes nearly to zero with decreasing temperature.[10,14] It should be mentioned that grain boundaries in FM oxides are regions of perturbation of the structural and magnetic orders, and,

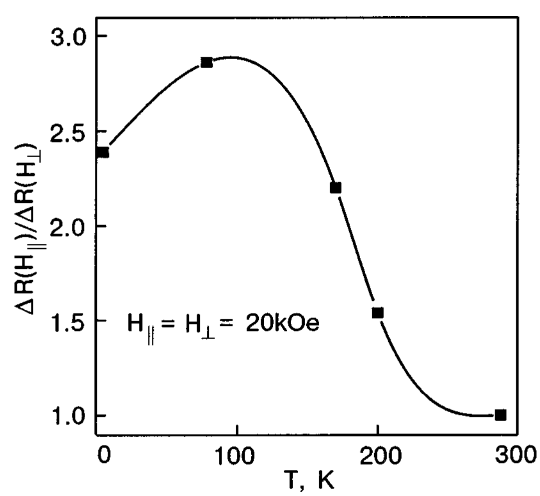

FIG. 6. Temperature dependence of the ratio of magnetoresistances for fields parallel ($H_{\parallel}$) and perpendicular ($H_{\perp}$) to the film plane. The fields were equal to 20 kOe.

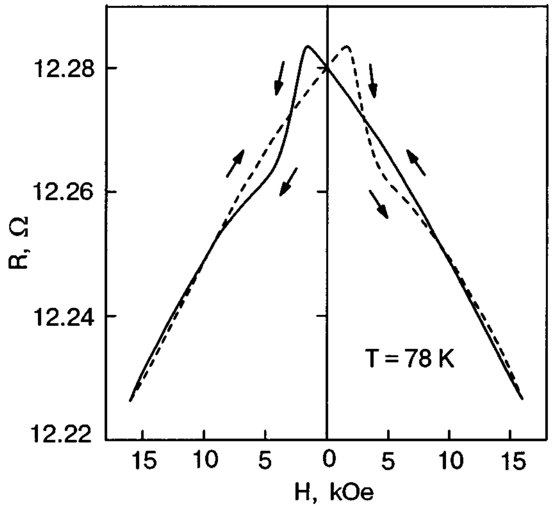

FIG. 7. Magnetoresistive hysteresis at $T = 78$ K for fields parallel to the film plane and perpendicular to the transport current.

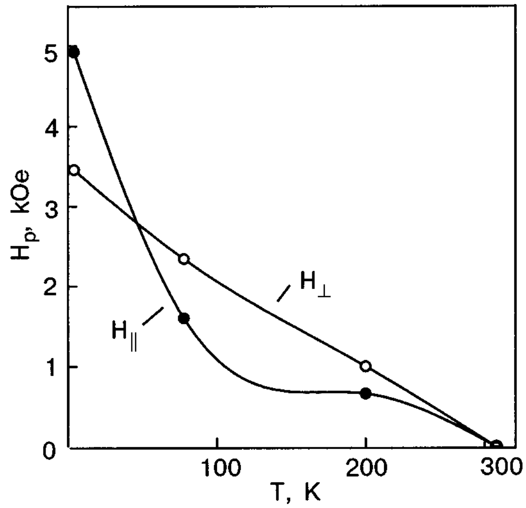

FIG. 8. Temperature dependence of characteristic field, H_p, at which resistance peaks in the magnetoresistive hysteresis curves (Fig. 7), for fields parallel ($H_\parallel$) and perpendicular ($H_\perp$) to the film plane. The field H_p corresponds to the coercive force (H_c) in the magnetization loops.

therefore, induce a magnetic inhomomogeneity as well. These boundaries (and, maybe, other sources of inhomogeneity, e.g., phase separation) can cause the significant angular hysteresis effect found in this study (Fig. 3), since they hinder the motion of FM domains upon rotation of the magnetic field. It is noteworthy, however, that the hysteresis effect is minimal at the angles corresponding to both the in-plane and out-of-plane directions of the magnetic field. In summary, the behavior of the resistivity, MR, and magnetization of the film corresponds to that of a system of weakly connected grains.

The data presented in Fig. 5 correspond to negative MR for fairly high fields. In general, the MR curves are hysteretic and have specific features in the low-field range (Fig. 7). Symmetric hysteresis curves, like that in Fig. 7, were obtained for the film after some number of repeated sweeps between the chosen maximum (positive and negative) field magnitudes. For the first sweeps, the hysteresis curves were some-what asymmetric. Actually, their behavior correlates with that of magnetization loops.[13] In particular, the field $H = H_p$ at which the resistance peaks (Fig. 7) corresponds to the value of the coercive force H_c. The value of H_p decreases with increasing temperature and goes to zero on approaching T_C. The magnitude of positive MR in the low-field range, $\Delta R(H_p) = [R(H_p) - R(0)]/R(0)$, is some measure of the remanent magnetization.

We found that H_p and $\Delta R(H_p)$ depend on the field direction and in this way reflect the magnetization anisotropy. The temperature dependences of H_p for the in-plane and out-of-plane directions of magnetic field are shown in Fig. 8. It is seen that at $T \simeq 4.2$ K the value of H_p in the out-of-plane field is less than that in the in-plane field, but at $T \approx 78$ K and higher temperatures the opposite relation is true. For high enough temperature ($T > T_C$) the H_p values go to zero for both field directions. The $\Delta R(H_p)$ values are found to be higher for the out-of-plane field direction as compared with the in-plane one at $T \simeq 4.2$ K. At $T \approx 78$ K and $T = 200$ K, the opposite relation holds true. All this implies that at low temperatures the out-of-plane magnetization is favored, whereas for higher temperatures the in-plane magnetization becomes dominant. The pronounced increase in $M_\perp(T)$ at low temperatures (Fig. 2) and decrease in the ratio between the in-plane and out-of-plane MRs below $T \approx 80$ K (Fig. 6) also support this suggestion. All these are indications of a temperature-driven spin reorientation transition which can be determined by competition between the shape anisotropy and the strain-induced anisotropy. This transition has been studied rather intensively (theoretically and experimentally) for films of common FM metals[15,16] but has never been mentioned for cobaltite films. It should be noted, however, that theoretical models like those of Refs. 15 and 16 are applicable only for ultrathin magnetic films (up to 10 monolayers), whereas the film studied is much thicker and rather disordered. Consequently, the spin reorientation transition in the film studied can have a different nature than those proposed for ultrathin films.

In conclusion, we have revealed and investigated magnetic and magnetoresistance anisotropy in a $La_{0.5}Sr_{0.5}CoO_{3-\delta}$ film. Among other things, we found indications of a temperature-driven spin reorientation transition in the film studied: at low temperature, the magnetization vector tends to be perpendicular to the film plane, but with increasing temperature the magnetization vector goes entirely to the in-plane direction.

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