

Magnetoresistance of epitaxial thin films of ferromagnetic metallic oxide SrRuO₃ with different domain structures

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We have studied the magnetoresistive behavior of epitaxial thin films of the conductive ferromagnetic oxide SrRuO₃ with different domain structures grown on both miscut (001) SrTiO₃ and exact (001) LaAlO₃ substrates. A strong anisotropic magnetoresistance (MR) has been observed in the single domain SrRuO₃ thin film on miscut (001) SrTiO₃ substrate. In contrast, the SrRuO₃ thin film on (001) LaAlO₃ substrate shows identical MR behavior in two orthogonal directions on the film due to the presence of 90° domains in the plane. For both the films, large negative magnetoresistance effects (~10%) were observed when the current and the applied magnetic field are parallel. This is attributed to a reduction in spin fluctuations near T_c and to magnetization rotation leading to a change in the angle between the current and magnetization at low temperatures.

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Recently, there has been a great deal of interest in epitaxial thin films of the conductive magnetic oxide SrRuO₃, because of its potential device applications and interesting electrical and magnetic properties. SrRuO₃ is an itinerant ferromagnet ($T_c \sim 160$ K) with a GdFeO₃-type pseudocubic perovskite structure and a bulk lattice parameter of 3.93 Å. This material exhibits a strong magnetocrystalline anisotropy in single crystal bulk¹ and single domain thin film² samples. It also exhibits unique properties when doped with Ca. Substitution of Sr with Ca progressively decreases the lattice parameter, increases the orthorhombic distortion of the lattice and suppresses the T_c until a completely paramagnetic material is obtained above 70% doping.

SrRuO₃ is also structurally and chemically similar to the LaMnO₃-based colossal magnetoresistive (CMR) materials.³ Furthermore, as in CMR materials, magnetic ordering in SrRuO₃ is known to facilitate electrical transport by decreasing resistivity which renders it attractive for magnetotransport studies.⁴ The successful growth of single crystal SrRuO₃ thin films on miscut SrTiO₃ substrates⁵ allows us to study the intrinsic anisotropic magnetotransport properties of SrRuO₃. Recently, we have reported the control of growth mechanisms⁶ and domain structure⁷ of SrRuO₃ thin films by using miscut substrates. In this letter, we present results on the magnetoresistive behavior of epitaxial SrRuO₃ thin films with two different crystallographic domain structures.

The SrRuO₃ thin films were deposited on both a 2° miscut (001) SrTiO₃ substrate and an exact (001) LaAlO₃ substrate using a 90° off-axis sputtering technique as described in Ref. 5. The thickness of the films is about 3000 Å. The film on miscut SrTiO₃ substrate grew by step flow⁶ and is single domain with (110) texture normal to the substrate. All the crystallographic planes and directions for SrRuO₃ referred to in this work are based on the orthorhombic unit cell. The in-plane epitaxial arrangement is SrRuO₃[$\bar{1}10$]/SrTiO₃[010] and SrRuO₃[001]/SrTiO₃[100].⁷ Due to the small lattice mismatch between SrRuO₃ and

SrTiO₃, this film has a coherent growth resulting in a strained lattice.

In contrast, the film on LaAlO₃ substrate has a three-dimensional island growth mechanism⁶ due to the large lattice mismatch with the substrate. Therefore, this film has a bulklike strain-free lattice. This film displays a mixture of (110) and (001) textures normal to the substrate. Both the (110) and (001) grains have two 90° domains in the plane present in equal volume fraction.

The influence of the domain structure on the magnetization of the films was studied with a superconducting quantum interference device (SQUID) magnetometer. Magnetization measurements while cooling the samples in a field of 0.05 T applied parallel to the film surface clearly showed the magnetocrystalline anisotropy in the single domain film. When the field is applied along the [$\bar{1}10$] direction, which is the easier axis for magnetization, a fully saturated magnetization is attained at low temperatures (~5 K). However, an unsaturated and lower magnetization (~65% of the value in the [$\bar{1}10$] direction) is obtained when the field is in the [001] direction. In contrast, the film on LaAlO₃ shows identical unsaturated magnetization when the field is applied along any of the two orthogonal directions on the film surface due to the presence of 90° domains in the plane of the film.

In order to investigate the correlation between the magnetization and resistivity, magnetoresistance (MR) measurements were performed on these films using an Oxford Instrument MAGLAB 2000TM system. For the single domain SrRuO₃ thin film, the resistivity was measured in four possible combinations of the \mathbf{J} (current density) and \mathbf{H} (applied field) directions with respect to the in-plane [$\bar{1}10$] and [001] directions of the film (i.e., $\mathbf{J} \parallel \mathbf{H} \parallel [\bar{1}10]$; $\mathbf{J} \parallel \mathbf{H} \parallel [001]$; $\mathbf{J} \perp \mathbf{H}, \mathbf{J} \parallel [001]$; and $\mathbf{J} \perp \mathbf{H}, \mathbf{J} \parallel [\bar{1}10]$). In contrast, for the film on exact (001) LaAlO₃ substrate, the resistivity was measured along the [010] and [100] directions of the LaAlO₃ substrate in two orientations ($\mathbf{J} \parallel \mathbf{H}$ and $\mathbf{J} \perp \mathbf{H}$) because of the equivalence of the two orthogonal directions. The temperature dependence of resistivity was measured by zero-field cooling (ZFC) and field warming.

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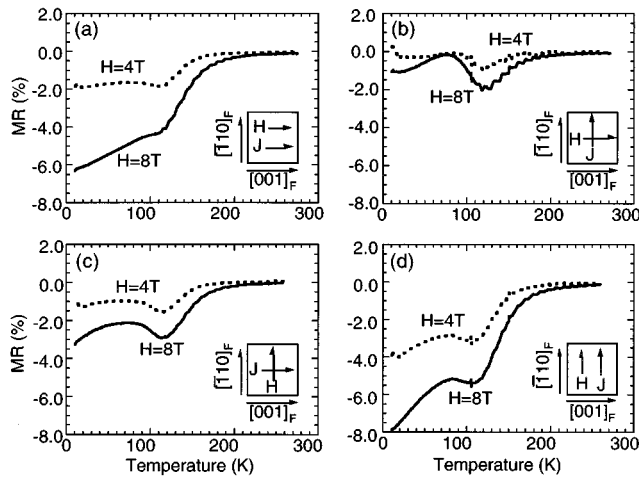


FIG. 1. MR vs temperature of a single domain SrRuO₃ film grown on miscal (001) SrTiO₃ substrate with (a) $\mathbf{J}||\mathbf{H}||[001]$, (b) $\mathbf{J}||[001]$ and $\mathbf{H}||[110]$, (c) $\mathbf{J}||[110]$ and $\mathbf{H}||[001]$, and (d) $\mathbf{J}||\mathbf{H}||[110]$. Note that the $[110]$ direction is parallel to the miscal direction.

The zero field resistivity of both the films show the characteristic change of slope at the Curie temperature (T_c) signifying a ferromagnetic phase transition. Above T_c , the resistivity increases linearly with temperature and is expected to continue without saturation.⁸ The residual resistivity ratio (ρ_{298K}/ρ_{4K}) for our samples typically varies between 2 and 4, with the films on LaAlO₃ having slightly higher values. The T_c of the film on SrTiO₃ (~ 120 K) is also lower than that for the film on LaAlO₃ substrate ($T_c \sim 155$ K). This T_c suppression in the films on SrTiO₃ substrate is not fully understood at present, but is believed to be due partly to the coherent growth induced lattice strain in these films.

From the resistivity curves at different fields, the magnetoresistance was calculated as $MR = [\rho(H) - \rho(0)]/\rho(0)$, where $\rho(H)$ and $\rho(0)$ are the resistivity at a field H and at zero field, respectively. Figure 1 compares the MR as a function of temperature for the single domain film in the four different combinations of current and field directions. The subscript “F” in $[hkl]_F$ in the inset denotes the crystallographic directions of the film. Several interesting features are observed from the figure. All the films display a large negative MR at temperatures just below T_c in all orientations of the current and field which has already been attributed to the suppression of spin fluctuations.⁹ Furthermore, the negative MR is larger when the applied field is parallel to the current, especially at low temperatures. This is due to the anisotropic magnetoresistance (AMR) effect. The AMR effect is the change in angle between the magnetization and current because of magnetization rotation observed at high fields that are smaller than the anisotropy field. Such an AMR effect has been observed in single domain SrRuO₃ thin films at a field of 6 T and is found to increase as the temperature is lowered below T_c .¹⁰

The largest MR among all four combinations of \mathbf{J} and \mathbf{H} directions is observed at low temperatures, when the current and field are parallel to the $[110]$ direction, as shown in Fig. 1(d). We believe that this is due to the fact that the $[110]$ direction is the easier axis of magnetization compared to the $[001]$ direction. The spins are more easily aligned in the

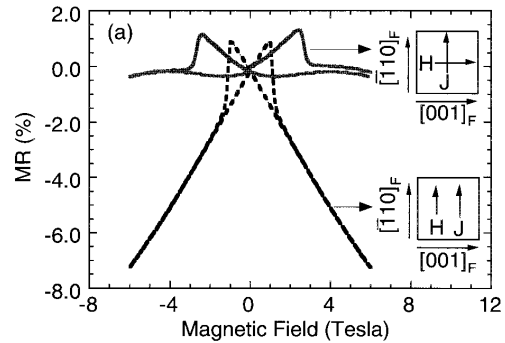


FIG. 2. MR vs field of the single domain SrRuO₃ film on miscal SrTiO₃ substrate with $\mathbf{J}||[110]$ direction.

$[110]$ direction which accounts for the larger change in resistivity due to the magnetic field.

The MR was also measured as a function of applied magnetic field in the four combinations of \mathbf{J} and \mathbf{H} directions. In order to erase any hysteresis effects, the samples were heated to above T_c in zero field between successive MR versus field measurements. Figure 2 shows this data at 5 K for the single domain SrRuO₃ thin film with the current along the $[110]$ direction. A strong hysteresis is observed in the MR behavior at low fields. The hysteresis effect itself is related to the magnetization hysteresis. The peak in the MR hysteresis corresponds to the coercive field and the point of overlap between the forward and backward sweeps of the field corresponds to the saturation field. As the applied field is increased beyond saturation, the magnetization of the sample does not change significantly but magnetization rotation occurs contributing to a larger negative MR when $\mathbf{J}||\mathbf{H}$ due to the AMR effect. The anisotropy of this film is evident from the MR versus field curves shown in Fig. 2. As the film is single domain, this anisotropy in MR reflects the inherent magnetocrystalline anisotropy of SrRuO₃, which is attributed to the large spin-orbit coupling of Ru.

In order to investigate the effect of the crystallographic domain structure on the magnetoresistance properties, we performed similar measurements on the film grown on (001) LaAlO₃ substrate. In contrast to the single domain film on miscal SrTiO₃ substrate, the resistivity of this film has an additional contribution from the crystallographic domain boundaries due to the incoherent three-dimensional island growth.

Figures 3(a) and 3(b) compare the magnetoresistance as a function of temperature with the field parallel and perpendicular to the current. The subscript “S” in $[hkl]_S$ in the inset denotes the crystallographic directions of the substrate. The sharp increase in MR at T_c and larger MR at low temperatures when the field and current are parallel and are similar to that of the single domain film on miscal SrTiO₃. However, for the film on LaAlO₃, the negative MR begins to decrease after having reached a maxima at low temperatures of about 30–40 K. This could be related to the temperature dependence of the magnetocrystalline anisotropy. At low temperatures, a higher field would be required to change the

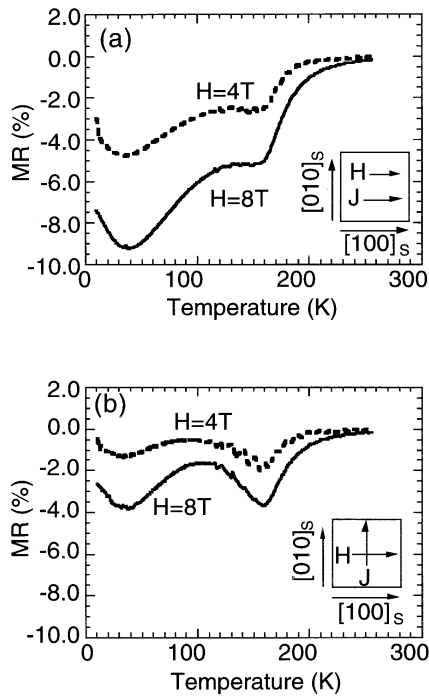


FIG. 3. MR vs temperature of a SrRuO₃ film on an exact (001) LaAlO₃ substrate with (a) $\mathbf{J} \parallel \mathbf{H}$ and (b) $\mathbf{J} \perp \mathbf{H}$.

orientation of spins in the material and thus, the negative MR decreases. A similar maxima in the MR at low temperatures has been observed by Klein *et al.*² in their films on SrTiO₃ which have a higher T_c . Such behavior is not observed in our films on SrTiO₃ probably due to its low T_c .

Figure 4 shows MR versus magnetic field curves for the SrRuO₃ film on (001) LaAlO₃ substrate at 5 K. The MR is shown in two orthogonal directions, with $\mathbf{J} \parallel \mathbf{H}$ and $\mathbf{J} \perp \mathbf{H}$. The MR is significantly larger when the current is parallel to the field, as observed in the single domain film. The hysteresis in MR is observed in both orientations of the field and the current. However, compared to the single domain film, the hysteresis on this film is smaller. It is believed that the film

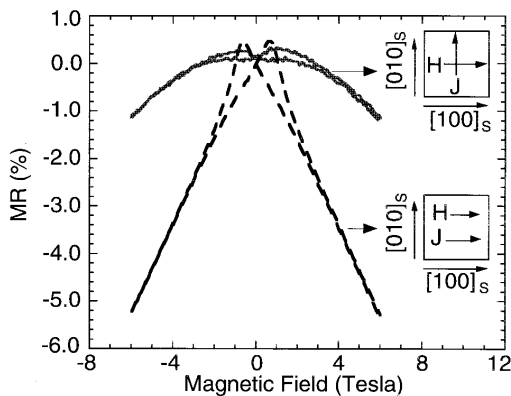


FIG. 4. MR vs field of a SrRuO₃ film on an exact (001) LaAlO₃ substrate with $\mathbf{J} \parallel \mathbf{H}$ and $\mathbf{J} \perp \mathbf{H}$.

on LaAlO₃ does not have any magnetic domain walls, because the grain size of these films [as observed from the scanning tunneling microscopy (STM) images]⁶ is of the same order as the spacing between magnetic domain walls (~ 2000 Å) observed by Lorentz microscopy imaging.¹¹ Therefore, the crystallographic domain boundaries in this film act as magnetic boundaries as well and the change in relative orientation of the magnetization of adjacent crystallographic domains contributes to the hysteresis. Finally, the sample was rotated by 90° and the MR versus field measurement was repeated in both orientations ($\mathbf{J} \parallel \mathbf{H}$ and $\mathbf{J} \perp \mathbf{H}$). The MR behavior obtained was identical to that shown in Fig. 4. This is due to the equivalence of the two orthogonal directions within the plane.

In summary, we have observed a strong anisotropic magnetoresistance in single domain SrRuO₃ thin film grown on 2° miscut (001) SrTiO₃ substrate. For the SrRuO₃ film grown on (001) LaAlO₃ substrate, the presence of 90° domains in the plane results in identical magnetotransport properties in two orthogonal directions within the plane. For both the films, large negative magnetoresistance effects ($\sim 10\%$) were observed when the current is parallel to the applied magnetic field, due to a reduction in spin fluctuations near T_c and to the AMR effect at low temperatures. Such studies on the influence of crystallographic domain structure on the anisotropic magnetotransport properties will help develop a better understanding of the intrinsic magnetocrystalline anisotropy and the anomalous electrical transport behavior of SrRuO₃.

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- ¹A. Kanbayashi, J. Phys. Soc. Jpn. **41**, 1879 (1976); G. Cao, S. McCall, M. Shepard, and J. E. Crow (unpublished).
- ²L. Klein, J. S. Dodge, C. H. Ahn, J. W. Reiner, L. Mieville, T. H. Geballe, M. R. Beasley, and A. Kapitulnik, J. Phys. Chem. Solids **8**, 10111 (1996).
- ³S. Jin, T. H. Tiefel, M. McCormack, R. A. Fastnacht, R. Ramesh, and L. H. Chen, Science **246**, 413 (1994).
- ⁴L. Klein, J. S. Dodge, C. H. Ahn, G. J. Snyder, T. H. Geballe, M. R. Beasley, and A. Kapitulnik, Phys. Rev. Lett. **77**, 2774 (1996).
- ⁵C. B. Eom, R. J. Cava, R. M. Fleming, J. M. Phillips, R. B. van Dover, J. H. Marshall, J. W. P. Hsu, J. J. Krajewski, and W. F. Peck, Science **258**, 1766 (1992).
- ⁶R. A. Rao, Q. Gan, and C. B. Eom, Appl. Phys. Lett. **71**, 1171 (1997).
- ⁷Q. Gan, R. A. Rao, and C. B. Eom, Appl. Phys. Lett. **70**, 1962 (1997).
- ⁸P. B. Allen, H. Berger, O. Chauvet, L. Forro, T. Jarlborg, A. Junod, B. Revaz, and G. Santi, Phys. Rev. B **53**, 4393 (1996).
- ⁹S. C. Gausepohl, M. Lee, K. Char, R. A. Rao, and C. B. Eom, Phys. Rev. B **52**, 3459 (1995).
- ¹⁰L. Klein (private communication).
- ¹¹A. F. Marshall, presented at the Materials Research Society—1997 Spring Meeting: Epitaxial Oxide Thin Film Symposium (1997) (unpublished).