Domain Wall Resistivity in $SrRuO_3$

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$SrRuO_3$ is an itinerant ferromagnet with $T_c \sim 150$ K. When $SrRuO_3$ is cooled through T_c in zero applied magnetic field, a stripe domain structure appears whose orientation is uniquely determined by the large uniaxial magnetocrystalline anisotropy. We find that the ferromagnetic domain walls clearly enhance the resistivity of $SrRuO_3$ and that the enhancement has different temperature dependence for currents parallel and perpendicular to the domain walls. We discuss possible interpretations of our results.

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Ferromagnetic domain walls (DW) are interfaces between magnetic domains whose magnetization points in different directions. For decades, there have been considerable theoretical [1–6] and experimental [3,7–9] efforts to elucidate the effect of DW on electrical resistivity. Early theoretical work predicted positive domain wall resistivity (DWR) due to reflection of electrons from the DW [1,2]. More recently, a different mechanism for positive DWR has been proposed (based on spin-dependent scattering due to defects) which pointed out the similarity between the magnetic structure induced by DW and that present in giant magnetoresistance (GMR) devices [3,5]. On the other hand, other models predicted negative DWR due to the destruction of electron weak localization induced by dephasing at the domain wall [4], or predicted that DWR can have either sign, depending on the difference between the spin-dependent scattering lifetimes of the charge carriers, based on a semiclassical treatment [6]. The experimental situation is also quite confusing. Reports on observed positive DWR in materials such as Co [3], were followed by studies which indicated that the additional resistivity was actually a bulk effect [8]. Moreover, there are now strong indications that domain walls in iron (previously believed to enhance resistivity) actually decrease resistivity [8].

Contrary to most previous studies of DWR that focused on the elemental $3d$ ferromagnets, we present here a study of DWR in $SrRuO_3$ which is a metallic perovskite and a $4d$ itinerant ferromagnet ($T_c \sim 150$ K). Previously, we have reported the enhancement of resistivity due to DW in this compound for currents perpendicular to the DW at 5 K based on resistivity hysteresis loops [10]. Here,we report the conclusion of a thorough study of high-quality samples and present for the first time a detailed temperature dependence of the DWR for currents perpendicular and parallel to the domain walls while using both hysteresis loops and a comparison between zero-field resistivity and field cooled resistivity, and applying multiple tests to substantiate the intrinsic nature of the reported effect. The importance of these data which present a unique comprehensive temperature dependence of DWR goes even beyond the contribution to the understanding of the mechanisms involved in DWR. The DW in this compound are extremely narrow [11] (3 nm); therefore a justified comparison of our results can be made with models proposed for spin-dependent transport in magnetic multilayers which, as we show below, can account quantitatively for the large observed positive DWR (interface resistance of $\sim 10^{-15}\ \Omega\,\mathrm{m}^2$ at low temperatures). This may turn $SrRuO_3$ into a model system for studying the resistivity due to magnetic interfaces while avoiding material interface issues.

$SrRuO_3$ has special advantages for studying DWR for its large uniaxial magnetocrystalline anisotropy field (~ 10 T) combined with its much smaller self-field ($4\pi M \sim 0.2$ T). The large uniaxial anisotropy induces stripe magnetic structure (see inset in Fig. 1) which enables the study of DWR for currents parallel and perpendicular to the domain walls; furthermore, it is also responsible for the narrow width of the DW which contributes to the large magnitude of the DWR. The

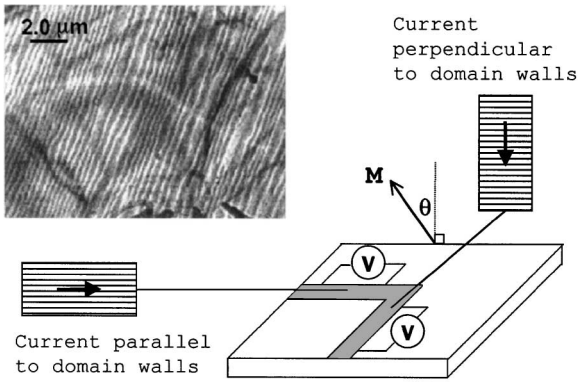

FIG. 1. The measurement configuration: films are patterned for simultaneous measurement of resistivity with parallel and perpendicular domain walls where the current is along the $[\bar{1}10]$ and [001] axes, respectively. The DW are parallel to the in-plane projection of the magnetization vector **M**. Inset: Image of DW in $SrRuO_3$ at 100 K with Lorentz mode TEM. Bright and dark lines image DW at which the electron beam diverges or converges, respectively. Background features are related to buckling of the freestanding film and are not related to magnetic variations.

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combination of the large uniaxial anisotropy with a much smaller self-field not only enables the stability of a saturated magnetization state at zero applied field but it also prevents the creation of closure domains near the sample surface when the sample is in its domain state. As we discuss below, these two properties enable unequivocal determination of DWR and avoid the need to consider bulk effects that may lead to mistaken or ambiguous identification of DWR.

Our measurements were done on high-quality single-crystal films of $SrRuO_3$ grown on slightly miscut ($\sim 2°$) substrates of $SrTiO_3$ by reactive electron beam coevaporation [12]. The orthorhombic film grows with its [001] and [$\bar{1}$10] axes in the film plane and its magnetic moment ($\sim 1.4\mu_B$ per ruthenium at saturation) along the out-of-the-plane axis of uniaxial magnetocrystalline anisotropy (see Fig. 1) whose direction varies in the (001) plane from approximately [010] (45° out of the plane) at T_c to about 30° relative to the normal to the film at low temperatures [13]. The residual resistivities of our films are as low as 4.6 $\mu\Omega$ cm (corresponding to resistivity ratio between $T = 300$ K and $T = 1.8$ K of ~ 45) and their thicknesses vary between 800 and 2000 Å. The films were patterned for simultaneous resistivity measurements along the [001] and [$\bar{1}$10] directions (see Fig. 1).

The magnetic domain structure of $SrRuO_3$ was extensively studied using transmission electron microscopy (TEM) in Lorentz mode (see inset of Fig. 1) after removing the $SrTiO_3$ substrate with a chemical etch. The details of this study are reported elsewhere [14]; here we mention the main results relevant to this report: (a) there is a *single* easy axis along which the spontaneous magnetization lies at zero applied field (no indication for flux closure domains with a different magnetic orientation); (b) the DW are parallel to the in-plane projection of the spontaneous magnetization ([$\bar{1}$10]), therefore, each time the sample is cooled through T_c the DW appear in the same direction; (c) at low temperatures the spacing of the DW is ~ 2000 Å and it does not change up to a few degrees below T_c; (d) once the DW are annihilated (at temperatures lower than a few degrees below T_c) by applying sufficiently large magnetic field, they do not renucleate when the field is set back to zero; and, only when a sufficiently high negative field is applied, new (less dense) domain walls are nucleated in the process of magnetization reversal.

The effect of the DW on the resistivity was measured using two methods. In the first method, the sample is cooled in zero field from above T_c to a temperature below T_c. Then the resistivity is measured during a hysteresis loop where the maximum field is high enough to annihilate all domain walls. Figure 2 shows a hysteresis loop taken at 5 K. We note that the initial zero-field resistivity (marked by full circles) is higher than the zero-field resistivity after the magnetization was saturated. Based on our TEM measurements, we know that the initial resistivity here is measured in the presence of DW, while

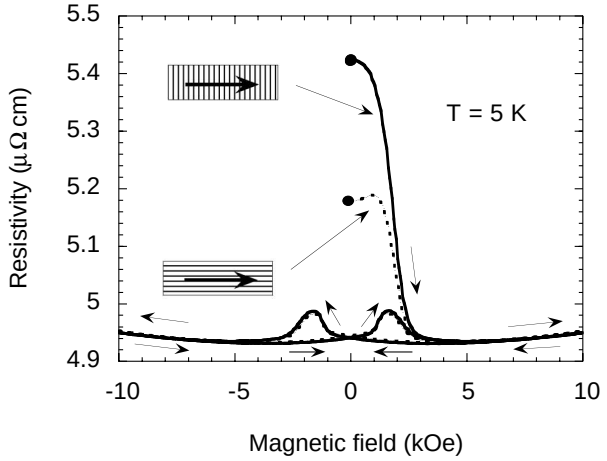

FIG. 2. Hysteresis loops of resistivity vs applied field for current parallel and perpendicular to the domain walls at $T = 5$ K. At the starting point with $H = 0$ (marked by full circle), the sample is in its domain structure. Increasing the field annihilates the DW, and when the field is set back to zero the magnetization of the sample remains saturated. We identify the difference between the initial zero-field resistivity and the subsequent zero-field resistivities as the domain wall resistivity.

no DW are present in the following zero-field resistivity measurements. Consequently, we attribute the difference between the two zero-field resistivities to DWR. In the second method the sample is cooled in zero field from above T_c to 1.8 K and the resistivity (ρ^{ZFC}) is measured as a function of temperature. Afterwards, the sample is cooled from above T_c in a field of 2 T (to prevent the formation of domain structure) down to 1.8 K where the field is set to zero and the resistivity (ρ^{FC}) is measured as a function of temperature. Since ρ^{ZFC} is measured with DW and ρ^{FC} is measured without them, we attribute the difference between the two to DWR. Figure 3 shows DWR as measured in the two methods as a function of temperature for the two different current directions. However, before discussing these plots (which are the main result of this paper), we address a crucial issue. Being aware of contentions that previous reports on positive DWR originated from anisotropic magnetoresistance (AMR) effects, we find it vital to eliminate this possibility in our case.

The AMR effect is a dependence of the resistivity in a magnetic metal on the angle between the current and the magnetic moment which results from spin-orbit coupling. If in the domain state of a magnetic metal the magnetization is along more than one axis, then saturating the magnetization of the sample must induce in some parts of the sample changes in the angle between the current and the magnetic moment, hence, an AMR effect. The existence of uniaxial anisotropy is not sufficient to ensure that the magnetization points in the same direction in all the domains. It was pointed out [8] that, when $Q = K/2\pi M_s^2 \ll 1$ (here K is the anisotropy energy and M_s is the saturated magnetization), flux closure domains in which magnetization points in different directions are created near the surface of the sample; consequently, an AMR effect contributes

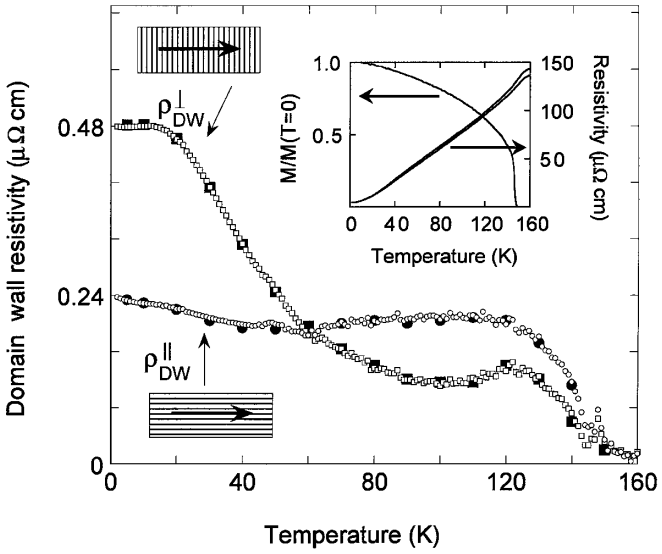

FIG. 3. Domain wall resistivity vs temperature for current parallel $[\rho^{\parallel}_{\mathrm{DW}}(T)]$ and perpendicular $[\rho^{\perp}_{\mathrm{DW}}(T)]$ to the domain walls (along the $[\bar{1}10]$ and $[001]$ axes, respectively). Close symbols are obtained by hysteresis loops (see Fig. 2) and open symbols are obtained by difference between ρ^{ZFC} and ρ^{FC}. Inset: Temperature dependence of ρ^{FC} for current along the $[\bar{1}10]$ (upper curve) and $[001]$ (lower curve) axes and of the magnetization (**M**).

to the change of resistivity when the sample is saturated. While Fe and Co are in the small Q limit, $SrRuO_3$ is in the $Q \gg 1$ limit ($Q_{SrRuO_3} > 10$). Therefore, it is not surprising that, contrary to Co and Fe, closure domains were not observed by TEM in $SrRuO_3$. This means that when we measure the resistivity with or without DW the bulk magnetization is along the same axis. This ensures not only the lack of any AMR effect but also any effect related to changes in the direction of the self-field which can give rise to changes in the regular Lorentz magnetoresistance.

After eliminating the possibility that an intrinsic AMR effect contributes to our measured DWR, we want to exclude the possibility of a "dirt" effect, namely, that small inclusions of $SrRuO_3$ with different magnetic anisotropy are causing the effect. This is excluded not only by the low residual resistivity, and the consistent values among different samples of the DWR in both current directions, but also by a direct test of the magnetic anisotropy of the regions responsible for the observed drop in resistivity in the hysteresis loops. We identify the DWR in a finite field as the difference between the resistivity on the first branch of the loop (the initial increase of the field from zero to above the saturating field), where DW are partially annihilated, and the resistivity on the second branch of the loop (where the field is decreased from above the saturating field to zero), where no DW are present, and we look at the dependence of the finite-field DWR on the angle between the applied magnetic field and the film. If the observed effect is a dirt effect, we do not expect a preferred direction (except for geometric considerations); on the other hand, if our interpretation is correct, we expect that the change in resistivity attributed to the process of annihilating the domain walls will depend on the component of the field parallel to the known direction of the uniaxial anisotropy. Figure 4 shows finite-field DWR for perpendicular current at some of the angles at which we measured. The inset shows that the angular dependence clearly supports our scenario. Similar results were also obtained for the parallel current. Based on the above, we argue that indeed we measure DWR and not a dirt or a bulk effect.

We go back now to Fig. 3 which shows DWR as a function of temperature for currents parallel ($\rho^{\parallel}_{\mathrm{DW}}$) and perpendicular ($\rho^{\perp}_{\mathrm{DW}}$) to the DW. To the best of our knowledge, this is the most detailed temperature dependent measurement of domain wall resistivity ever reported for either current direction. The results presented in Fig. 3 were obtained by measuring the sample with the lowest residual resistivity, and, while results slightly vary among samples, we find the following characteristics in all our measured samples. At low temperatures, $\rho^{\perp}_{\mathrm{DW}}$ is always larger than $\rho^{\parallel}_{\mathrm{DW}}$ and their ratio is ~ 2 in the highest quality samples. The magnitude of the DWR of various samples in the zero temperature limit is very similar despite variations of more than a factor of 2 in the value of the residual resistivity. The specific features of the temperature dependence of $\rho^{\perp}_{\mathrm{DW}}$ and $\rho^{\parallel}_{\mathrm{DW}}$ are preserved. Up to 15 K, $\rho^{\perp}_{\mathrm{DW}}$ is flat; between 15 and 100 K there is a sharp decrease in $\rho^{\perp}_{\mathrm{DW}}$; between 100 and 120 K, $\rho^{\perp}_{\mathrm{DW}}$ slightly increases; and between 120 K and T_c, $\rho^{\perp}_{\mathrm{DW}}$ decreases to zero. $\rho^{\parallel}_{\mathrm{DW}}$ has very different behavior except for the sharp decrease above 120 K which is correlated with the sharp decrease in the spontaneous magnetization. $\rho^{\parallel}_{\mathrm{DW}}$ is quite flat up to 120 K with a shallow minimum

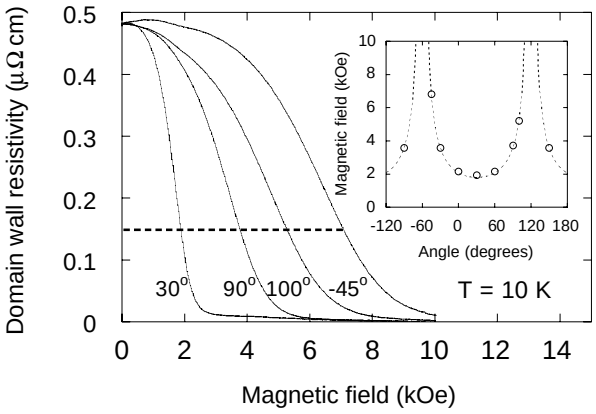

FIG. 4. Finite-field domain wall resistivity (see text) as a function of field at $T = 10$ K when the field is applied at different angles to the normal to the film. The inset shows the field needed to reduce the domain wall resistivity to 30% of its full value as a function of the angle between the applied field and the normal to the film. The dashed line is a fit with $H = H_0/|\cos(\theta - 30)|$ which is the expected behavior if only the component of the field parallel to the uniaxial anisotropy axis (whose orientation is at 30 degrees relative to normal to the film at that temperature) is relevant.

around 60 K. In the zero temperature limit we find that $\rho_{\mathrm{DW}}^{\perp} \sim 0.48\ \mu\Omega\,\mathrm{cm}$ and $\rho_{\mathrm{DW}}^{\parallel} \sim 0.24\ \mu\Omega\,\mathrm{cm}$. The width of the domain wall is $\sim$3 nm while the spacing between the domain walls is $\sim$200 nm. Therefore, in terms of bulk resistivity, the resistivity for the perpendicular current within the domain wall is $\sim 35\ \mu\Omega\,\mathrm{cm}$ and the interface resistance is $\sim 10^{-15}\ \Omega\,\mathrm{m}^2$. This value is more than 3 orders of magnitude larger than the interface resistance of $6 \pm 2 \times 10^{-19}\ \Omega\,\mathrm{m}^2$ reported for cobalt [8]. We believe that the huge difference in the magnitude of the DWR is related to the width of the DW in cobalt estimated as 5 times larger than in $SrRuO_3$.

Models proposed for DWR [1–6] have concentrated on the limit where the width of the DW is much larger than the Fermi wavelength. This limit, however, is not applicable here, and therefore we cannot compare our results to these models. Instead, we compare our data to results obtained for magnetic multilayers. The configuration in which $\rho_{\mathrm{DW}}^{\perp}$ is measured is similar to the so-called CPP (current perpendicular to plane) geometry in GMR structures (only that in our case we study solely the effect of the magnetic interface without having to consider issues such as surface roughness and matching between different materials). Therefore, mechanisms considered for the GMR structures may also be responsible for the observed DWR. Such a mechanism is the potential step scattering, previously studied for layered magnetic structures by Barnas and Fert [15]. The two found that an interface between a magnetic and a nonmagnetic metal, or between different magnetic domains within a magnetic metal, acts similar to a potential step whose height is related to the exchange splitting. While there is no closed form equation for the interface resistance, the numerical solution of Barnas and Fert indicates that for commonly used materials in magnetic multilayers (e.g., Co or Cu) the interface resistance is on the order of $10^{-15}\ \Omega\,\mathrm{m}^2$. Since DW in our case are so narrow, we believe that it is possible to treat them as potential steps. The exchange splitting and the Fermi energy in $SrRuO_3$ are 0.65 and 2 eV, respectively [16]. Therefore, we can expect an interface resistance on the order of $10^{-15}\ \Omega\,\mathrm{m}^2$, as observed.

Another potential source for DWR also considered for GMR structures is spin accumulation [17]. When a polarized current crosses an interface, there is spin accumulation near the interface that induces a potential barrier which results in excess resistivity [see Eq. (25) in Ref. [17]]. The spin accumulation (and its related resistivity) is strongly affected by the spin diffusion length l_{sf}. This length is strongly affected by magnetic scattering; therefore, the decrease in $\rho_{\mathrm{DW}}^{\perp}(T)$ above $T = 15$ K may be related to the sharp decrease in the spin-accumulation resistivity induced by the shortening of l_{sf} due to magnetic scattering.

Contrary to $\rho_{\mathrm{DW}}^{\perp}(T)$ which exhibits complex temperature dependence, $\rho_{\mathrm{DW}}^{\parallel}(T)$ is almost flat up to 120 K despite big changes particularly in the resistivity but also in the magnetization of $SrRuO_3$. We have no model for this behavior, although it is interesting to note that we would have obtained temperature-independent resistivity if we excluded the volume of the DW including a distance from the DW proportional to the charge carrier mean free path.

In conclusion, special properties of domain walls in $SrRuO_3$ enable clear observation of large *positive* DW resistivity. Our main result here is the detailed temperature dependence of $\rho_{\mathrm{DW}}^{\perp}$ and $\rho_{\mathrm{DW}}^{\parallel}$ which requires further theoretical consideration. This, we hope, will yield deeper understanding not only of DWR but also of transport mechanisms in magnetic multilayers. Further experiments in which the mean free path will be changed by electron irradiation, and the magnetization will be changed by doping, are planned for quantitative identification of the different contributions to DWR.

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