Angular dependence of domain wall resistivity in SrRuO$_3$ films.

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SrRuO$_3$ is a 4d itinerant ferromagnet (T$_c$ $\sim$150 K) with stripe domain structure. Using high-quality thin films of SrRuO$_3$ we study the resistivity induced by its very narrow ($\sim$ 3 nm) Bloch domain walls, $\rho_{DW}$ (DWR), at temperatures between 2 K and T$_c$ as a function of the angle, $\theta$, between the electric current and the ferromagnetic domains walls. We find that $\rho_{DW}(T, \theta) = \sin^2 \theta_{PDW}(T, 90) + B(\theta)\rho_{DW}(T, 0)$ which provides the first experimental indication that the angular dependence of spin accumulation contribution to DWR is $\sin^2 \theta$. We expect magnetic multilayers to exhibit a similar behavior.

I. INTRODUCTION

Following the discovery of the giant magnetoresistance (GMR) effect in magnetic multilayers and related phenomena considered to lay the foundation for the emerging field of spintronics there has been an intensive effort to elucidate the mechanisms involved in spin polarized transport in the presence of magnetic interfaces. One of the outcomes of this effort was the realization that naturally obtained domain structure in itinerant ferromagnets may offer an important opportunity to study basic issues relevant to spintronics while avoiding difficulties encountered when artificially grown multilayers are studied. These difficulties arise due to the intrinsic uncertainties and complications associated with the nature of magnetic interfaces in these systems.

Two major hurdles were encountered along this route when domain wall resistivity (DWR) of iron, nickel, cobalt, and FePd was studied: (a) The resistivity (or interface resistance) is quite small and hardly distinguished from the anisotropic magnetoresistance effect which is unavoidable in these systems due to the presence of closure domains (b) The estimated width of ferromagnetic domain walls (DWs) in these systems is relatively large (e.g., 10 nm in FePd, 15 in cobalt and 100 in nickel). Consequently, models applicable to magnetic multilayers with atomically sharp interfaces are not relevant and other models were used to interpret the obtained results. Therefore, limited understanding of the physics governing magnetic multilayers could be achieved from studying DWR in such systems.

These problems are absent in the itinerant ferromagnet SrRuO$_3$. This compound has relatively large magnetocrystalline anisotropy field (~10 T) that yields stripe structure with domain wall separation of 200 nm without closure domains and with narrow Bloch DWs whose estimated width is only $\delta$ ~ 3 nm. These features make SrRuO$_3$ a model system on which models suggested for magnetic multilayers can be tested.

Initial evidence that the same physical mechanisms are relevant both to DWR in SrRuO$_3$ and magnetic multilayers was given in a previous report where the magnitude of the DWR with perpendicular current was shown to be qualitatively and quantitatively consistent with resistivity induced by two sources relevant to magnetic multilayers: spin accumulation and potential step. We should note that spin accumulation was also claimed to be the source of DWR in nanowires of cobalt; however, this attribution was later challenged on grounds that the effect is expected to be suppressed in the 15 nm thick DWs.

Here we explore DWR in SrRuO$_3$ as a function of the angle, $\theta$, between the electric current and the ferromagnetic DWs. We find that DWR for any angle $\theta$ is given by a $temperature$ − $independent$ linear combination of DWR for parallel ($\rho_{DW}(T, \theta = 0)$) and perpendicular ($\rho_{DW}(T, \theta = 90)$) currents. As we demonstrate below, this fit provides the first experimental indication that the angular dependence of spin accumulation contribution to DWR is proportional to $\sin^2 \theta$, as expected based on simple theoretical considerations. This behavior is likely to be found in magnetic multilayers, as well.

II. EXPERIMENT

SrRuO$_3$ is a pseudocubic perovskite and an itinerant ferromagnet with Curie temperature of ~160 K for bulk and ~150 K for thin films (provided the film thickness is more than 10 nm). Thin films exhibit high magnetocrystalline anisotropy field (~10 T) with uniaxial anisotropy with the easy axis roughly along the crystallographic $b$ axis in the orthorhombic notation. For this study we used high-quality thin films of SrRuO$_3$ grown on SrTiO$_3$ substrates by reactive electron beam coevaporation. To avoid twinning in the SrRuO$_3$ film and obtain a film with the same orientation of the uniaxial anisotropy throughout the sample, the cubic symmetry of the substrate surface needs to be broken. This is achieved by slightly miscutting (~2 degrees) the SrTiO$_3$ substrates which forms atomically flat terraces separated by unit cell steps. The film whose growth starts at the steps grows uniformly with the projection of the easy axes on the plane of the film perpendicular to the miscut-induced steps.

The domain structure of similarly grown films was
thoroughly studied using Lorentz microscopy on free standing films after removing the SrTiO$_3$ substrate with a chemical etch. It was found that in the domain state magnetic stripes are formed parallel to the easy axis projection on the film. Figure 1 shows DWs image taken from Ref.\textsuperscript{11}. The DWs are 200 nm apart with no detectable dependence on film thickness (in the studied range of 30 nm - 100 nm) and no detectable dependence on temperature except for few degrees below T\textsubscript{c}. The estimated thickness of the Bloch wall, $\delta$, is $\sim 3\,\text{nm}$.\textsuperscript{17} Closure domains were not observed as expected for SrRuO$_3$ whose $Q = \frac{K}{2\pi M_s} \gg 1$ (here $K$ is the anisotropy energy and $M_s$ is the saturated magnetization). The stripe structure forms spontaneously when the sample is cooled below $T_c$ in zero field. When a sufficiently high field is applied below $T_c$ the magnetization becomes uniform with no stripes. An important observation is that the uniform magnetization state remains stable when the field is set back to zero. A substantial (temperature dependent) field needs to be applied in the negative direction to start magnetization reversal (with many fewer DWs than in the initial zero-field-cooled state). These features are essential for facilitating clear identification of DWR in this system.

Using photolithography we patterned films in the form shown in Figure 1. It includes patterns at eight different angles relative to the stripes: 0, 15, 30, 45, -45, 60, 75 and 90 degrees. The distance between the two distant voltage leads is 500 $\mu\text{m}$ and the width of the current path in the patterns is 50 $\mu\text{m}$. Therefore, for $\theta = 90$ the current crosses $\sim 2500$ domain walls between the voltage leads, and for $\theta = 0$ there are $\sim 250$ parallel DWs running along the current path.

The DWR measurements were performed for each film on all eight patterns at temperatures between 2 K and 140 K. To measure the excess resistivity induced by domain walls at a specific temperature $T_{\text{measure}}$, we cooled the sample in zero magnetic field from above $T_c$ down to $T_{\text{measure}}$ and measured the resistivity there. As noted before\textsuperscript{11} when cooling in zero field a stripe domain structure forms; therefore, the zero-field-cooled resistivity is measured with the magnetic domains present. While staying at the same temperature we increased the field to obtain uniform magnetization in the film. We then decreased the field to zero and measured the resistivity once again. The uniform magnetization remains stable at zero field. Therefore, the second resistivity is measured in the absence of any DWs. Consequently, we can attribute the difference between the two values of resistivity to DWR. For each temperature where DWR was measured, the process was repeated by first warming the sample above $T_c$ and cooling in zero field to the temperature where DWR was to be measured.

FIG. 1: (a) Image of stripe domain walls in SrRuO$_3$ with transmission electron microscopy in Lorentz mode (from Ref.\textsuperscript{11}). The bright and dark lines image walls that diverge or converge the electron beam, respectively. Background features are related to buckling of the free standing film and are not related to magnetic variations. The average spacing between the walls is $\sim 200\,\text{nm}$. (b) Schematic figure of the photolithography patterns used for angular dependence study. The patterns are at eight different orientations relative to the DWs (marked by the arrow): 0, 15, 30, 45, -45, 60, 75 and 90 degrees.

III. RESULTS AND DISCUSSION

Fig. 2a shows the temperature dependence of DWR for the current flowing in eight different angles relative to the DWs. Our main objective is to understand the measured DWR changes as a function of the angle between the current and the DWs.

As noted before, we analyze our results by applying models used for magnetic multilayers for which two important contributions to resistivity were considered theoretically: spin accumulation\textsuperscript{18} and potential step\textsuperscript{19}. For parameters appropriate for SrRuO$_3$ the two effects are expected to yield interface resistance for perpendicular current on the order of $10^{-15}\,\Omega\,\text{m}^2$, as experimentally observed\textsuperscript{11}.

Spin accumulation is generated when spin-polarized
current crosses an interface between two domains with opposite magnetization. Valet and Fert showed that such a current yields spin accumulation near the interface that induces a potential barrier which results in interface resistance $r$ given by $r = 2\beta^2 \rho_F l_{sf}$ (see Eq. 25 in Ref. 18) where $\beta$ is the spin asymmetry coefficient (zero for unpolarized current and 1 for fully polarized current), $\rho_F$ is the average resistivity (for spin-up and spin-down currents) and $l_{sf}$ is the spin diffusion length which is the characteristic length over which the polarization of crossing current equilibrates with the equilibrium polarization. The spin-accumulation resistivity, $\rho_{SA}$, proportional to the interface resistance, $r$, with the number of domain walls per unit length being the proportionality factor. $\rho_{SA}$ is expected to decrease with temperature due to the expected fast decrease in $l_{sf}$ due to increase in magnetic scattering (e.g., due to magnons) which becomes more probable with increasing temperature.

Since spin accumulation is associated with the net current crossing the interface, no spin accumulation contribution to DWR is expected for current parallel to the domain walls. Here we explore in what way spin accumulation contribution, that we note by $\rho_{SA}$, changes as a function of angle.

Two factors relevant to spin accumulation resistivity change as a function of angle (see inset to Figure 3a): the net current flowing perpendicular to the interface is multiplied by $\sin \theta$ and the number of domain walls per unit length crossed by a current is multiplied by another factor of $\sin \theta$. The bottom line is that spin accumulation contribution to DWR for any angle $\theta$ is expected to be the spin accumulation resistivity for perpendicular current multiplied by $\sin^2 \theta$. The problem, however, is to determine the spin accumulation part in the DWR with perpendicular current.

As we noted before, no spin accumulation contribution is expected for parallel current. Therefore, assuming that the sources responsible for DWR for parallel current (presumably, related to potential steps) are present for other angles as well and assuming that (similarly to spin-accumulation) the contribution at other angles is a temperature-independent function of the angle alone, $A(\theta)$, we can expect that DWR for any temperature and angle will be given by the following equation:

$$\rho_{DW}(T, \theta) = \frac{\sin^2 \theta (\rho_{DW}(T, 90) - A(90) \rho_{DW}(T, 0)) + A(\theta) \rho_{DW}(T, 0)}{\rho_{DW}(T, 0)}$$

The term $(\rho_{DW}(T, 90) - A(90) \rho_{DW}(T, 0))$ is the spin accumulation part in the DWR for perpendicular current and $B(\theta) = A(\theta) - A(90) \sin^2 \theta$.

To test this model we check whether Equation 1 can reproduce, using measured $\rho_{DW}(T, 90)$ and $\rho_{DW}(T, 0)$, the measured DWR at all other angles by using a single fitting function, $B(\theta)$. The success of this fit is visible in Figure 2b.

As a sensitivity check on our assumption of the $\sin^2 \theta$ dependence of $\rho_{SA}$, we looked for the best fit (allowing $B(\theta)$ to vary) when $\sin^2 \theta$ is replaced by $\sin \theta$ or $\sin^3 \theta$. The different fits are shown in Figure 3a for $\theta = 45$. Figure 3b shows an alternative comparison of the three fits which is independent of any fitting parameter. It shows $(\rho_{DW}(T, 45) - f(\theta) \rho_{DW}(T, 90))/\rho_{DW}(T, 0)$ as a function of temperature with $f(\theta)$ being $\sin \theta$, $\sin^2 \theta$, or $\sin^3 \theta$. A fit consistent with the assumption that the sources of DWR for parallel current (that do not include spin accumulation) are present at other angles as well and their relative effect at other angles is temperature independent should yield a temperature independent curve. Figure 3b demonstrates that the best fit is indeed obtained with $f(\theta) = \sin^2 \theta$.

The fitting function is $B(\theta) = A(\theta) - A(90) \sin^2 \theta$. To identify the spin accumulation part in the DWR of the perpendicular current we need to determine $A(90)$. To identify $A(90)$ we consider that the closer we are to $T_c$ the less is the relative contribution of spin accumulation to DWR; hence, the angular dependence of DWR is dominated by the angular dependence of $A(\theta)$. Therefore, we look for $A(90)$ for which $A(\theta)$ is the most similar to the angular dependence of DWR obtained at high temperature. Figure 4a presents $A(\theta)$ for two different samples and Figure 4b shows the angular dependence of the DWR at different temperatures. The main feature is the non monotonic or even oscillatory (note correlation between the two samples) behavior of $A(\theta)$ which is reflected in the angular dependence of the DWR particularly near $T_c$ where $\rho_{SA}$ diminishes. It is likely that the source that displays this behavior is related to interface resistance associated with potential steps; however, the specific cause of this behavior is unclear at the moment. It is important to note that the exact form of $A(\theta)$ depends on the thickness of the film so we hope that more study focused on the thickness dependence of DWR will yield more understanding.

Having found $A(90)$, we can determine the spin accumulation interface resistance as a function of temperature. Figure 5 shows the spin accumulation interface resistance for two different samples and the inset shows their extracted $l_{sf}$ using Valet-Fert equation. It is important to note that this derivation of $l_{sf}$ is expected to be reliable only in the low-temperature limit where Valet-Fert equation is valid. From the low temperature limit we find $l_{sf}$ on the order of 40-50 nm. This value is consistent with our findings that DWR for perpendicular current scales with the density of DWs which implies that the scattering at neighboring DWs is independent, suggesting that spin diffusion length is smaller than the separation between the DWs (200 nm).

IV. SUMMARY AND CONCLUSION

We present here for the first time data on the angular dependence of resistivity induced by stripe domain
structure in a system that can adequately serve as a model system for magnetic multilayers. We find a simple fitting equation whose success provides the first experimental indication that the angular dependence of the spin-accumulation resistivity \( \rho_{DA}^{SA} \) is \( \sin^2 \theta \).

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FIG. 2: (a): DWR as a function of temperature with current flowing at different angles relative to the DWs: $\theta$=0, 15, 30, 45, 60, 75 and 90 degrees. The data points for $\theta$=45 are on top of the data points for $\theta$=45 and are not shown in the figure. (b) DWR as a function of temperature with $\theta$=15, 45, and 75. The symbols are the actual data points for the different angles whereas the line is the one-parameter fit based on Equation 1.
FIG. 3: (a) $\rho_{DW}(T,45)$ with three different fits assuming different angular dependence of the spin accumulation contribution to DWR: $\sin \theta$ (dotted), $\sin^2 \theta$ (full) and $\sin^3 \theta$ (dashed). Inset: Schematic illustration of a current path with current $J$ at an angle $\theta$ relative to the DWs. The current flowing perpendicular to the wall is $J \sin \theta$ and the distance the current flows between the walls $d$ is $(\text{domain width})/\sin \theta$. (b) $(\rho_{DW}(T,45) - f(\theta)\rho_{DW}(T,90))/\rho_{DW}(T,00)$ as a function of $\theta$ with $f(\theta) = \sin \theta$ (circles), $f(\theta) = \sin^2 \theta$ (squares) and $f(\theta) = \sin^3 \theta$ (triangles). Following Equation 1, the quality of the fit is manifested in temperature independence of $(\rho_{DW}(T,45) - f(\theta)\rho_{DW}(T,90))/\rho_{DW}(T,00)$.
FIG. 4: (a) $A(\theta)$ as a function of angle for two different samples (b) $\rho_{DW}(T, \theta)$ as a function of angle at different temperatures. The symbols are the actual data points whereas the line is the one parameter fit based on Equation 1.
FIG. 5: Spin accumulation contribution to the interface resistance as a function of temperature for two different samples. The inset shows the extracted spin diffusion length based on Valet-Fert relation.