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Intrinsic domain-wall resistivity in half-metallic manganite thin films

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Deciphering the intrinsic magnetic domain-wall (DW) resistivity of manganite materials by typical low-field magnetoresistance measurement is flawed due to the addition of different galvanomagnetic effects such as, colossal magnetoresistance, Lorentz force magnetoresistance, and anisotropic magnetoresistance (AMR). In this paper, by taking the advantage of rotational anisotropy and the stable rotation of the DW planes in half-metallic manganite $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ film, we deploy a remanent state resistance measurement technique to exclude all the field-dependent spurious effects from the intrinsic DW resistivity. To further refine its magnitude, we calculate the remanent state DW AMR by exploiting the three-dimensional micromagnetic simulation, which reveals a comparable but opposite contribution to the positive DW resistivity. From these results, we estimate the intrinsic DW resistance-area product in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ to be $1.9 \times 10^{-15} \Omega \cdot \text{m}^2$.

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Magnetic domain wall (DW) is a topological defect of spin configuration that has been under an intense research spotlight during the last two decades, primarily owing to its huge potential to be the building block for high-density data storage systems such as racetrack memory,¹ logic circuits,² and oscillators.³ In order to manipulate the DWs precisely and reliably in a magnetic system, it is imperative to understand its micromagnetic details and associated electrical resistivity. Due to several extrinsic effects such as anisotropic magnetoresistance (AMR) and Lorentz force magnetoresistance (LMR), it is notoriously arduous to conclude whether the DW resistivity is sourced by them or other inherent phenomena such as spin mistracking of conduction electrons or suppression of weak localization.^{4–9} Despite tremendous efforts from the research community, owing to these spurious effects, a consensus on not only the magnitude but also the sign of the DW resistivity even in extensively investigated three-dimensional (3d) transition metals like Ni, Co, and Fe has not been established yet.^{4,5,7}

Compared to the 3d transition metal ferromagnets, less attention has been paid to evaluate DW resistivity in ferromagnetic manganites, where the spin, charge, and orbital degree of freedom together with the substrate-induced strain, all play crucial roles in determining the electrical and magnetic characteristics.^{10–12} Among a number of family members of manganite, $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ is considered one of the most technologically interesting ferromagnets, especially due to its half-metallicity,¹³ environmental stability, and a Curie temperature (T_c) above room temperature. So far, only few groups have delved into the DW resistivity of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ and a wide range of DW resistance-area (DWRA) products, spanning from $10^{-11} \Omega \cdot \text{m}^2$ to $10^{-15} \Omega \cdot \text{m}^2$ have been reported.^{14–18} In another pioneering work, Mathur *et al.* reported a DWRA of $8 \times 10^{-11} \Omega \cdot \text{m}^2$ at 77 K in microstructured $\text{La}_{0.7}\text{Ca}_{0.3}\text{MnO}_3$ samples.¹⁹ However, none of these works took the AMR contribution from the detailed microscopic spin structure of the DWs into account in the evaluation of DWRA. Moreover, unlike 3d transition metal ferromagnets, manganites exhibit colossal

magnetoresistance (CMR), which also served as a source of error in the estimation of the DWRA in these experimental works. Besides, a DWRA value of $10^{-17} - 10^{-18} \Omega \cdot \text{m}^2$ has been theoretically estimated for manganites^{17,19,20} a few orders smaller than the reported experimental values. Thus, more precise evaluations are required for the correct interpretation of the four-order variation among the experimentally reported values of DWRA and the discrepancy between experiment and theory.

In most of the works in the literature, the DW resistivity is simply evaluated from the difference between the resistivity under a magnetic field (strong enough to annihilate the DW) and zero field (or a nucleation field). A fundamental shortcoming of such techniques stems from the presence of the external field during electronic transport measurement; the complexity of the problem is enhanced by the superposition of the contributions from CMR, AMR, LMR, and even the modulation of the spin structure inside the DW.^{5,21} To overcome these spurious effects and evaluate the intrinsic DW resistivity in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ thin film, we have measured the remanent state resistance with different orientations of the DW plane to the current flow direction, ranging from 0° (R_{CIW}) to 90° (R_{CPW}). Here, CIW and CPW correspond to the configurations of current in the plane of DW and current perpendicular to the plane of DW, respectively. Additionally, a combination of magnetic force microscopy (MFM) images, complemented by the detailed micromagnetic information from the 3d Object-Oriented Micromagnetic Framework (OOMMF)²² simulation and calculation of the remanent state DW AMR enabled us to further refine the magnitude of the intrinsic DW resistivity.

$\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ films with a thickness of 80 nm were grown on (100) LaAlO_3 substrates by pulsed-laser deposition technique. The deposition parameters, morphological, structural, and magnetic characteristics of the film are similar to our previously reported works.^{23,24} The T_c , coercivity, and the saturation magnetization have been recorded as 360 K, 25 Oe, and 270×10^3 A/m, respectively. Due to compressive

FIG. 1. (Color online) Remanent state MFM images showing the magnetic domain patterns after applying different initialization fields along the $\langle 100 \rangle$ direction. The scan size is $3 \mu\text{m} \times 3 \mu\text{m}$. In (a) the slight deviation of the stripe directions from the $\langle 010 \rangle$ direction is due to the misalignment between the sample position and field direction. The insets show the FFT images of the corresponding MFM patterns. The brightest spot of the FFT image indicates the most dominant periodic feature and its angular position with respect to the $\langle 100 \rangle$ crystal axis indicates the relative angle (α) between the current direction (along the $\langle 100 \rangle$ axis) and the normal to the DW plane.

strain, the film exhibits perpendicularly oriented magnetic stripe domain patterns, as revealed by the room-temperature MFM images of Fig. 1. A pair of the 2-mm-wide Cu pads with 2-mm edge-to-edge spacing was used for the dc resistance measurements of the 2-mm-wide $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ sample.

To determine the DW resistivity by exploiting the remanent state electronic transport characteristics, the knowledge of stability of the magnetic domain landscape after removal of the external initialization field (H) and the orientation of the DW plane is *a priori*. In order to examine such prerequisites in our $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ sample, we first applied a small field of 400 Oe to align the stripe domains along the $\langle 010 \rangle$ crystal axis and subsequently took the MFM image [Fig. 1(a)] over a $3\text{-}\mu\text{m} \times 3\text{-}\mu\text{m}$ square area region. Upon initializing the magnetic state, the external field was increased in small steps along the $\langle 100 \rangle$ direction. After every step of increase, the field was swept back to zero and kept there for 5 min, in order to facilitate the remanent state MFM imaging. As shown in Figs. 1(b)–1(d), on increasing the field strength, the remanent state stripe domains gradually rotate toward the $\langle 100 \rangle$ direction, suggesting the presence of a rotatable anisotropy, as present in typical 3d transition metal ferromagnets and their alloys.^{25–27} Furthermore, no

FIG. 2. (Color online) (a) Remanent state resistance (left y axis) after applying different initialization field along the $\langle 100 \rangle$ direction. The stripe domains were initially aligned along the $\langle 010 \rangle$ direction. As the initialization field strength increases, the normal to the DW gradually aligns with the current direction, resulting into reduction of the sample resistance. The right y axis represents the relative angle between the $\langle 100 \rangle$ direction and the normal to the DW plane, derived from the FFT images of the MFM patterns. (b) Contour plot showing the remanent state resistance change for different sample rotation angle α' and initialization field strength. Due to the incomplete rotation of the DW, the resistance is smaller when the lower initialization field is used. The curve superimposed on the contour plot represents the remanent state resistance change (right y axis) as a function of sample rotation angle using a constant initialization field of 330 Oe.

deformation of magnetic domains related to the slow relaxation procedure²⁴ was noticed in a short timescale of few minutes.

The insets in Figs. 1(a)–1(d) show the brightness-coded fast Fourier transform (FFT) images of the corresponding MFM landscapes, where the angular position of the brightest spot represents the most dominant periodic feature's relative angle (α) between the normal ($\mathbf{n}$) to the DW plane and the $\langle 100 \rangle$ crystal axis. The field dependence of thus determined α is plotted in Fig. 2(a). It is worthy to note that in spite of having a certain degree of inhomogeneity in the stripe domain pattern, the overall magnetic landscape is stable and the relative angle α increases with the initialization field above 50 to 300 Oe. The intermediate values of the relative angle α suggest that the rotation of $\mathbf{n}$ is governed by the vector sum of the external field and the rotatable anisotropy field, where the later can be roughly estimated to be around 50 Oe, above what the stripes start to rotate.

The remanent state resistance of the film as a function of the initialization field is shown in the left axis of Fig. 2(a). Prior to the resistance sampling, the relative angle α was aligned at zero degrees. To minimize any perturbation due to the slow magnetic relaxation phenomenon in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$,²⁴ we capped the maximum field at 450 Oe and kept the time lag between two consecutive resistance measurement at less than a second. As the field increases, the remanent state resistance remains almost unchanged up to 50 Oe, which corroborates with the threshold field needed to rotate the stripe domains. Upon further increase of the field, the resistance reduces in a quasilinear fashion up to 200 Oe and then slowly approaches the saturation state at 350 Oe, presenting a total resistance change of $R_{\alpha=0} - R_{\alpha=90} = 110 \text{ m}\Omega$, about 0.08% of the overall sample resistance. This trend reconciles with the dependence of α on the magnetic field, evidencing the connection of the observed results to the orientation of the DW plane.

To verify that the origin of the above-described remanent state resistance change is the angular position of the DW plane with respect to the current direction, we performed a complementary set of electronic transport measurement. Instead of changing the magnetic field strength, we kept it and the current direction constant and then physically rotated the field direction in a step of approximately 10° , where $\alpha' = 0^\circ$ represents the $\langle 010 \rangle$ crystal axis. After every rotational step, the field was switched back to zero and then the corresponding sample resistance was recorded. An exemplary plot of the resistance change $\Delta R = R(\alpha') - R(0)$, for the case of 330-Oe field is shown in the line diagram of Fig. 2(b). As expected, ΔR appears as a periodic function of α' and reaches minimum approximately around $\alpha' = 90^\circ, 270^\circ$. The magnitude of maximum ΔR for this case is 80 m Ω , similar to the value observed when the DW plane was rotated by tuning the initialization field strength. As shown in the color-coded plot of Fig. 2(b), when the similar experiment is done at a field smaller than 330 Oe, ΔR is reduced. This is understandable; as a smaller field aligns the normal to the DW plane, $\mathbf{n}$ at an intermediate angle in between 0° to 90° [Fig. 2(a)], causing an angled incidence of the transport electrons on the DW plane and, in turn, a reduced ΔR . No significant change in ΔR was observed when a field value lower than 50 Oe was used, supporting the presence of rotational anisotropy and a threshold field for rotating the DW plane in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ films, as mentioned earlier.

The aforementioned results unambiguously suggest that the resistance data shown in Fig. 2 correspond to the resistivity associated with the orientation of DW. However, as mentioned earlier, the intrinsic DW resistance in a ferromagnetic manganite is often masked by several extrinsic phenomena such as CMR, LMR, and AMR. CMR causes a resistivity reduction in manganites due to the ferromagnetic alignment of t_{2g} electrons with field. *In situ* field-dependent magnetoresistance measurement reveals a 60 m Ω (about 0.045% of the total sample resistance) reduction of resistance at 330 Oe in our $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ sample, an order of magnitude comparable to the observed $R_{\alpha=0} - R_{\alpha=90}$ value. CMR effects of similar size were also present in other works in the literature^{15,16} on DW resistivity in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$, which imposes uncertainty

to exclusively determine the intrinsic DWR. In this context we emphasize the advantage of measuring the remanent state resistance to evaluate DWR, from which we can exclude not only the field-dependent CMR effects but also other galvanomagnetic contributions, such as LMR, which stems from the modulated trajectories of charges under a magnetic field.

In contrast to the CMR and LMR effects, a portion of AMR contribution cannot be excluded from the experimental data even for our remanent state resistance measurement scheme, as the magnetization vector is inherently inhomogeneous inside the DW. Therefore, we theoretically assess the magnitude of the remanent state AMR effect superimposed on the measured $R_{\alpha=0} - R_{\alpha=90}$ data. To get the information about the magnetization landscape, we performed 3d OOMMF simulation²² over a sample size of $1 \mu\text{m} \times 1 \mu\text{m} \times 80 \text{ nm}$ using a cubic cell of $5 \times 5 \times 5 \text{ nm}^3$. Taking experimentally evaluated values of the key parameters, such as Curie temperature $T_c = 360 \text{ K}$, lattice constant $a = 0.384 \text{ nm}$, saturation magnetization $M_s = 270 \times 10^3 \text{ A/m}$, and effective magnetic anisotropy energy (obtained from the difference between areas enclosed by $M-H$ loops along the $\langle 100 \rangle$ and $\langle 001 \rangle$ direction²⁸) $K = -1.5 \times 10^4 \text{ J/m}^3$, we estimate the constants needed for micromagnetic simulations such as exchange integral $J = \frac{3K_B T_c}{2zS(S+1)} = 33.12 \times 10^{-23} \text{ J}$ and exchange stiffness constant $A_{ex} = \frac{Js^2}{a} = 1.94 \times 10^{-12} \text{ J/m}$. Here, we assume that the spin $s = 3/2$ and the magnetic interaction is restricted to the nearest-neighbour Mn sites, i.e., the coordination number $z = 6$. These parameters result in a mazelike domain pattern, as the color-coded magnetization state of the xy plane (at a depth $z = 40 \text{ nm}$) in Fig. 3(a) shows. The power spectral density (PSD) analysis of the corresponding FFT image shown in the inset of Fig. 3(c) estimates the period of the most dominant feature, i.e., the average domain size (D) to be 90 nm, very close to the D experimentally evaluated (84 nm) from the analysis of the MFM images.

To elucidate the internal structure of the DW, in Fig. 3(b), we plot a cross-sectional image of the enclosed square region of Fig. 3(a). The micromagnetic landscape at the cross-section of the DW shown in Fig. 3(b) clearly suggests that the DW structure is quite complex, and it converts from a Bloch type at the core to the Néel type at the closure region near the surface, a similar scenario as the typical ferromagnets with weak perpendicular anisotropy exhibits.²⁹ For simulating the AMR contribution due to the DW, we took a sample portion of the image containing half of two oppositely polarized domains and a straight DW in between them [as indicated by the enclosed square area in Fig. 3(a)]. The results obtained for such a sample cell was combined using a series-parallel network to estimate the remanent state AMR effect for the true sample size.

The electric field due to the AMR contribution can be calculated from the following expression:³⁰

$$\mathbf{E} = \rho_{\perp} \mathbf{j} + \mathbf{m}(\mathbf{j} \cdot \mathbf{m})(\rho_{\parallel} - \rho_{\perp}) + \rho_{H0} \mathbf{m} \times \mathbf{j}, \quad (1)$$

where $\mathbf{m}$ is the unit vector for magnetization, $\rho_{\parallel}$ and $\rho_{\perp}$ are the resistivities when the current and magnetization vector are aligned parallel and perpendicular to each other, respectively, and ρ_{H0} is the extraordinary Hall resistivity. Since the

resistance is measured at zero external field and the current density $\mathbf{j}$ is always along $\langle 100 \rangle$, from Eq. (1) we can deduce the expression $R'_{\text{CPW}} = \rho_{\perp}(1 - \cos^2\theta \cos^2\varphi) + \rho_{\parallel}\cos^2\theta \cos^2\varphi$ and $R'_{\text{CIW}} = \rho_{\perp}(1 - \cos^2\theta \sin^2\varphi) + \rho_{\parallel}\cos^2\theta \sin^2\varphi$, where R'_{CPW} and R'_{CIW} correspond to the resistance of each cell for CPW and CIW case, respectively; we consider only the AMR contribution, while neglecting the DW resistance. Here θ and φ are the azimuthal and polar angles, respectively. To evaluate the DW resistivity quantitatively, we assume that the resistance in the CIW case can be calculated by the AMR contribution of each cell, because the electrons do not need to pass through the DW in the CIW case. Unlike typical transition ferromagnetic metals such as permalloy, $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ is known to exhibit negative AMR ($\rho_{\perp} > \rho_{\parallel}$) with a typical magnitude of 1%.^{31,32} From the experimentally obtained value of $R_{\alpha=90}$ and with the assumption of a reasonable 1% AMR, we can estimate $\rho_{\perp} = 1.076 \times 10^{-5} \Omega \cdot \text{m}$, $\rho_{\parallel} = 1.065 \times 10^{-5} \Omega \cdot \text{m}$, and the AMR difference $R'_{\text{CPW}} - R'_{\text{CIW}}$ for our experimental sample size to be $-145 \text{ m}\Omega$. This suggests that the AMR magnitude in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ can be comparable to the DW resistance and therefore should not be neglected. In addition, the sign of the AMR contribution due to the inhomogeneity of magnetization inside the DW is opposite to that of experimentally obtained $R_{\text{CPW}} - R_{\text{CIW}}$, which

evidences that our observed results originate from the intrinsic resistivity of the DW in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$. From this consideration, taking a total DW resistance $= 255 \text{ m}\Omega$, the average DW density $n_{\text{avg}} = 1.1 \times 10^4/\text{mm}$, and the area of a single DW $= 2 \text{ mm} \times 80 \text{ nm}$, we can estimate an intrinsic average DWRA_{CPW} value as $1.86 \times 10^{-15} \Omega \cdot \text{m}^2$. We note that due to the irregularities of domain direction, locally some of the DWs' resistance may fluctuate due to inhomogeneous current distribution. However, the power spectral density of the fast Fourier transformed MFM image provides a fairly accurate measure of n_{avg} . Here, we assumed evenly spaced domains and used n_{avg} in all the calculations.

Now we turn to a discussion about the magnitude of the DWRA product in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ films. Among the reports found in the literature, the claim of Wolfman *et al.* ($10^{-11} \Omega \cdot \text{m}^2$) and Arnal *et al.* ($10^{-13} \Omega \cdot \text{m}^2$) are on the higher side.^{14,15} However, these estimations are flawed due to the negligence of the CMR effect. Moreover, no direct evidence for the presence of the DW in these works was provided; hence, the accountability of the DW resistivity for the reported large low-field MR remains highly questionable. A more reasonable result was reported by Y. Wu *et al.*¹⁷ (DWRA $= 10^{-15} \Omega \cdot \text{m}^2$), in which such artifacts were removed. The reported value is approximately half of our estimation because they neglected the AMR contribution, which is of the same order of magnitude as the DWRA.

An intrinsic DWRA value of $1.9 \times 10^{-15} \Omega \cdot \text{m}^2$ is 2 to 3 orders larger than that for other conventional ferromagnets such as Co ($10^{-17} \Omega \cdot \text{m}^2$),³³ as well as the theoretical expectation for manganites also.^{17,19} However, for these theoretical calculations, it was assumed that the DW is purely a Bloch type, having a width (δ) of 30 or 100 nm, which we believe is the reason for the much lower estimation. As mentioned above, the evaluation of the DW width is not so simple because the spin structure also varies along the z axis. If we assume the 50% M_z (magnetization along $\langle 001 \rangle$) criterion as the boundary for the DW, from Fig. 3(c) we find that the DW width increases from 5 nm at the center to 25 nm at the surface. The theoretical expectation³⁴ for intrinsic DW resistivity in manganite is to be proportional to $(\frac{1}{\delta})^3$ and hence two orders of resistivity difference between the wall core and the closure region is possible, which may explain the discrimination between the theoretical expectation and the value found in our experiment.

In summary, we exploit the rotational anisotropy of stripe domains and remanent state resistance measurements to accurately determine the intrinsic DWRA in half-metallic manganite thin films. The technique is advantageous particularly in excluding the spurious galvanomagnetic effects such as CMR, AMR, and LMR from the DW resistivity estimation. We also show that the sign of the DW AMR in $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ is negative, whereas that for the intrinsic DW resistance is positive.

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FIG. 3. (Color online) (a) Simulated micromagnetic landscape for $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ showing a mazelike pattern at remanent state. Red (dark gray) and blue (medium gray) pixels correspond to $+M_z$ and $-M_z$ respectively. (b) The cross-sectional image of the area enclosed by the dotted rectangle in Fig. 3(a) shows that the DW converts from a Bloch type at the core region to the Néel type at the closure area. (c) M_z as a function of distance from the wall core, suggesting a gradual widening of the DW region from center to the surface of the film. The inset shows the PSD graph of Fig. 3(a), whose peak position corresponds to the inverse of the average D in the simulated image.

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