Lorentz transmission electron microscope study of ferromagnetic domain walls in SrRuO₃: Statics, dynamics, and crystal structure correlation

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(Received 1 September 1998; accepted for publication 13 January 1999)

The magnetic microstructure of SrRuO₃ thin films is studied using Lorentz transmission electron microscopy (TEM). The zero-field-cooled magnetic stripe structure shows one-to-one correlation with the crystal domain structure and is used to identify the uniaxial magnetic anisotropy of these films, consistent with results on single domain films. The anisotropy is shown to be primarily magnetocrystalline in nature with the b axis as the easy axis. Lorentz TEM also yields quantitative information about the domain structure and domain wall resistivity, and allows for in situ observation of domain wall formation and dynamic response to an applied field. © 1999 American Institute of Physics. [S0021-8979(99)02908-4]

I. INTRODUCTION

SrRuO₃ is a metallic, perovskite-based oxide with a moderate carrier concentration, and a 4d itinerant ferromagnet (\(T_C = 150\) K). Early bulk studies of polycrystal and single crystal SrRuO₃ showed an orthorhombic structure, slightly distorted from cubic perovskite, with high magnetic anisotropy and a reduced magnetic moment.1–3 Because of its metallic properties and structural compatibility with technologically important oxide materials such as high \(T_c\) superconductors and ferroelectrics, SrRuO₃ was recently synthetized in thin film form, leading to new investigations of its fundamental properties.4–6 The magnetization, transport, and magnetoresistive behavior of single-crystal-like epitaxial thin films have been studied in detail; results are consistent with the bulk studies and furthermore show high uniaxial magnetic anisotropy, strong magneto-optic coupling, anomalous transport behavior about \(T_C\) and at low temperature, and strong irreversible magnetoresistive behavior.7–10

The crystalline and magnetic microstructure of these SrRuO₃ films is of considerable interest both for correlation with the macroscopic magnetization measurements and for further investigation of the magnetic properties. Lorentz transmission electron microscopy (TEM) is well suited to such microstructural analysis as well as allowing for in situ studies of magnetodynamic behavior. We have previously reported on the use of low-temperature Lorentz TEM to image the zero-field-cooled magnetic microstructure of SrRuO₃ films and to determine its relationship with crystal orientation.11,12 In this article we report in detail on the TEM magnetostatic results and extend the analysis to magnetodynamic observations of domain wall motion in an applied field. Lorentz microscopy of films with multicrystal domain (twinned) structures gives quantitative information concerning the magnetic domain structure, and allows for in situ observations of domain wall motion and annihilation. These results are correlated with previously reported magnetization measurements of highly aligned, single-crystal-like (un-twinned) films deposited on miscut substrates. The structure of the single-crystal films is also characterized. We find that the high uniaxial anisotropy of SrRuO₃ thin films is largely magnetocrystalline in origin and identify the b axis as the easy axis, measure the resistivity of domain boundaries, correlate the magnetostatics with the domain structure, and characterize the response of the domain walls to applied fields.

II. EXPERIMENT

SrRuO₃ films were synthesized by laser ablation or reactive electron beam coevaporation onto a variety of oxide substrates. The films reported on here were all deposited on SrTiO₃, because the latter can be removed by chemical etch to provide large, uniformly thin TEM samples. The films are 300–1000 Å in thickness, deposited on both standard and miscut substrates, and are removed with a chemical etch of HF:HNO₃:H₂O diluted approximately 1:1:1. They are then supported on standard carbon/formvar support films on Cu grids. The resulting TEM samples are large in extent (0.5 mm or more), uniformly thin, and robust, allowing for optimal Lorentz imaging and repeated cycling through \(T_c\) for comprehensive in situ analysis. They are, however, extensively buckled, and consequently exhibit a great deal of random diffraction contrast (due to bend contours) which must be minimized by tilting in order to clearly observe the magnetic domain walls.

The films were examined in a Philips CM20FEG (field emission gun) fitted with a special lens for magnetic imaging, the twin 2 or Lorentz lens. For normal high resolution imaging the specimen is situated in the middle of the high magnetic field, approximately 2 T, of the objective imaging lens; this will significantly alter most magnetic microstructures. For Lorentz imaging this lens is switched off and the Lorentz lens, just below the objective lens, is used as the imaging lens. The advantage of the Lorentz lens, over the more traditional use of the intermediate projector lens as the magnetic imaging lens, is improved resolution and range of
magnification. In order to compensate for any residual field remaining at the specimen with the objective lens turned off and the Lorentz lens on, the current in the twin (minicondenser) lens above the specimen is reversed; calibration by observing domain wall motion with tilt in a Co film indicates a cancellation current of about −1700 mA. Exact field cancellation is difficult to reproduce with precisely the same lens settings, probably due to hysteresis in the various lenses and small variations in lens currents as imaging conditions are varied; however, when the twin lens current is not reversed, observation of magnetic microstructures indicates that there is a residual field on the order of 1 kG which is reduced an order of magnitude or greater by the current reversal.

The films are observed both in Lorentz mode (LLM) and by conventional imaging with the objective lens (OLM). The ferromagnetic transition is observed by cooling the specimens in a Gatan liquid nitrogen cold stage. Using LLM, magnetic domain walls become visible in a defocused image as alternating black and white lines due to alternating convergence and divergence of the electron beam at the wall position as it is deflected by the magnetic moment of the domains. This is described by the Lorentz force, $F = e\mathbf{v} \times \mathbf{B}$. Using OLM allows for conventional diffraction analysis; a comparison between crystalline and magnetic microstructure is thus carried out over large areas of the specimen. To study domain wall motion, the objective lens is turned on. The field of the lens is normal to the untitled specimen plane; the specimen is tilted (± 30° allowed tilt) to achieve an inplane component of field in the desired direction. In this way domain wall motion can be studied under a variety of applied field directions. The specimen is warmed and cooled repeatedly to achieve the same approximate starting domain structure.

III. RESULTS AND DISCUSSION

A. Relationship between crystal structure and magnetic microstructure

As a multicrystal domain SrRuO$_3$ film is cooled through $T_c$ in approximately zero field, a stripe magnetic microstructure of alternating black and white domain walls is observed (Fig. 1). Sets of stripes are at 45° to each other and vary in spacing. We will demonstrate, using a combination of selected area electron diffraction and Lorentz imaging characteristics, that the stripe structure shows the presence in the film of all six of the possible crystallographic domains, and the existence of a single magnetic easy axis whose orientation is close to the $b$ axis. The magnetic and crystalline domain structures are shown schematically in Fig. 2. The correlation of the uniaxial anisotropy with a specific crystal axis, shows that this anisotropy is primarily magnetocrystalline in nature and not a substrate effect.

We first describe the formation of the observed crystal domain structure; this structure occurs because the orthorhombic SrRuO$_3$, having lower symmetry than the cubic substrate, aligns in several symmetry-related orientations on the substrate: The orthorhombic structure is based on a cubic perovskite subcell ($a = 3.93$ Å), with the $c$ axis doubled and the closely spaced $a$ and $b$ axes rotated 45° with respect to the subcell ($a = 5.53$, $b = 5.57$, $c = 7.82$ Å). The relationship between the orthorhombic and subcell axes is shown in Fig. 2. During film deposition, the approximately cubic subcell aligns with the cubic axes of the substrate resulting in the six crystal domain orientations shown.

1. The orthogonal $c$-axis orientations

Since the $c$ axis is readily distinguished by electron diffraction and remains parallel to the substrate axes, a convenient way to consider the crystal domain structure is as three orthogonal±orientations of the $c$ axis, parallel to the substrate axes. The $a$ and $b$ axes of the film are then rotated with respect to the substrate axes as shown in Fig. 2. Selected area electron diffraction confirms the three orthogonal $c$-axis orientations as shown in Fig. 1, and determines the correlation with the stripe orientation as shown in the schematic. For the widely spaced walls at ±45° to the substrate axes, the $c$ axis is perpendicular to the substrate and the $a$ and $b$ axes are in the plane of the film with the walls parallel to $b$. For the narrowly spaced walls parallel to the substrate axes, the $c$ axis is in the plane of the film, perpendicular to the stripe...
direction and the $a$ and $b$ axes are out of the plane at 45°, i.e., these domains are in a [110] orientation. Electron diffraction cannot easily distinguish the $\pm c$-axis twin orientations shown in the schematic, which also correspond to reversal of the closely spaced $a$ and $b$ axes; we will use characteristics of the magnetic images to identify these twins as described below. We refer to the in-plane orientations of the $c$ axis as $c_{\perp}$, and to the normal orientation as $c_{\parallel}$. As shown in Fig. 2 there are four $c_{\parallel}$ orientations (two sets of twins at 90° to each other) and two twin-related $c_{\perp}$ regions.

2. The magnetization direction

Lorentz microscopy then shows that the stripe orientation also indicates the magnetization direction, i.e., for these zero-field-cooled conditions, the easy magnetic axis. This is done by observing the direction of beam deflection by the magnetic domains. The Lorentz lens is defocused to observe the back focal plane of the lens where the diffraction pattern is formed. The deflection of the beam by the different domains is observed as a splitting of the transmitted beam of the diffraction pattern; the direction of the spot splitting is correlated with different stripe directions by the use of a selected area aperture (in this case, the objective aperture, since the normal image and diffraction modes of the microscope are reversed). As shown in Fig. 3, it is observed that the spot splitting is always normal to the stripe direction, confirming that the magnetization of the domains is parallel to the domain walls. This is expected, since the wall energy is minimized for a parallel configuration. The magnetization is therefore in the $ac$ or $bc$ plane for $c_{\perp}$ and in the $ab$ plane for $c_{\parallel}$, indicating an easy axis along $a$ and/or $b$ (assuming the same easy axis for both orientations). The magnitude of the spot splitting from the $c_{\perp}$ regions is greater than that of the $c_{\parallel}$ domains, indicating that the magnetization is closer to or in the film plane for the former (greater Lorentz force resulting in greater beam deflection), confirming that the easy axis is approximately $a$ and/or $b$ for both types of domains.

3. Identifying $\pm c$ domains, $b$ as the easy axis

The stripe structure therefore indicates both the direction of the magnetic easy axis and the presence of three orthogonal crystal orientations of the $\text{SrRuO}_3$ structure. From details of the magnetic images, one can further determine the presence of the twin-related $\pm c$-axis crystal domains, confirming the presence of all six crystallographic domains in these films. For $c_{\perp}$ regions we observe that there are two 90° orientations of the domain walls (the more widely spaced walls) and that, for a given area, these walls always return in the same orientation during temperature cycling through $T_c$. This indicates that there is, in fact, a unique easy axis along either $a$ or $b$, and that there are twin-related $c_{\parallel}$ regions of the film where $a$ and $b$ reverse direction. It is difficult to distinguish $a$ and $b$ by absolute measurements, as they are very close in magnitude. However selected area diffraction of twinned $c_{\parallel}$ regions (those with widely spaced domain walls at 90° to each other) exhibit spot splitting which allows for measurement of the relative magnitude of the in-plane crystal axes. (This splitting effect is due to the small differences in lattice spacing and is distinct from the central spot splitting of Fig. 3 which is due to magnetic deflection.) We use this to determine that $b$ is the easy axis as follows: Spot splitting in a selected area diffraction pattern of a twinned $c_{\parallel}$ region is shown in Fig. 4. Each of a set of split spots can be assigned to one of the twin regions by shifting the selected area aperture from one twin to the other. The closer spot in reciprocal space, i.e., the larger $b$ axis in real space, is parallel to the wall direction, indicating that the $b$ axis is the magnetization direction and therefore the easy axis. Note that the direction of the spot splitting is at 45° to the $a$ and $b$ axes, as is the plane of the boundary. That is, the boundary plane is (110), which allows for atomic matching at the interface and a coherent boundary. The $a$ and $b$ axes tilt slightly across this boundary leading to the spot splitting along [110]. These tilts and the lattice mismatch with the substrate lead to severe buckling of the film which is evident in the curved black-and-white bend contours visible in the images.

The presence of twin-related $c_{\parallel}$ regions, due to $\pm c$-axis orientation, is not shown by the stripe orientation because it has the same direction for such regions (see Fig. 2). The presence of this twinning can be determined by more subtle observations of variations in contrast and wall dynamics during tilting. We assume an out-of-plane magnetization for these regions approximately along the $b$ axis, as indicated by the previous results. If a $c_{\parallel}$ region is tilted about the $c$ axis, then the $b$ axis will be tilted either closer to the electron beam direction or farther away from it, depending on the $\pm$ sense of the crystal domain (see schematic of Fig. 5 where a $c_{\parallel}$ region is viewed sideways along the $c$ axis). Tilting decreases or increases the magnetic domain wall contrast, re-
spectively, because the contrast depends on the Lorentz force $F = ev \times B$, i.e., the angle between the electron beam and the magnetization. A given $c_\parallel$ region will reverse its contrast behavior (stronger versus weaker contrast) with reverse tilt, and twin-related regions will show opposite contrast changes when tilted in one direction. Such twin-related $c_\parallel$ domains are shown in Fig. 5 for ±30° tilt about the $c$ axis. Domains that decrease in contrast for one tilt direction, e.g., regions marked “X” in Fig. 5(a), show an increase in contrast for the opposite tilt, Fig. 5(b), and both twin orientations, distinguished by “X’” and “Y” in Fig. 5, are apparent in the area observed. When a field is applied (to be discussed in more detail later) parallel to the beam, the low contrast magnetic domains, having their magnetization tilted toward the beam, are the first to grow, and the domain walls of these regions of low contrast are therefore the first to annihilate [Figs. 5(c) and 5(d)]. For the same tilt, the twin-related regions of strong contrast have their magnetization almost perpendicular to the field, requiring rotation to align with the field and therefore resisting domain wall motion.

B. The emerging picture

The TEM analysis shows one-to-one correlation between the local film orientation and the magnetic stripe structure which, by itself, is sufficient to indicate uniaxial magnetic anisotropy. Furthermore, it shows that the easy magnetization axis is very close to the film $b$ axis. For $c_\parallel$ regions this implies an easy axis which is tilted out of the plane of the film at about 45°, while for $c_\perp$ regions the easy axis is in the plane of the film at 45° relative to the substrate unit axes.

These conclusions are consistent with global magnetization measurements of fully aligned films grown on miscut SrTiO$_3$ substrates which have their $c$ axis predominantly in the plane of the film (perpendicular to the miscut direction). These measurements indicated an easy axis out-of-plane at about 45° along either $a$ or $b$. The significance of the present results is not only in identifying the easy axis as the $b$ axis but, by showing the correlation between the magnetic easy axis and the crystallographic axis, which holds irrespective of the relative orientation of the film with respect to the substrate, we have substantiated the intrinsic magnetocrystalline nature of the magnetic anisotropy and excluded the possibility of a dominant substrate-induced strain effect.

C. Crystal domain morphology and crystal defects

In addition to showing us magnetization behavior, the magnetic microstructure is a more effective way to image the crystal domain morphology than conventional diffraction contrast imaging. This is because diffraction contrast between crystal domains is small, highly sensitive to small changes in orientation, and overshadowed by that from the film buckling, whereas the magnetic contrast is not sensitive to small tilt variations. The magnetic microstructure further-
more directly indicates individual domain orientation over a large field of view, allowing the degree of film alignment to be measured. This orientation distribution is not easily observed in conventional dark-field or high resolution images due to the film buckling and small field of view, respectively. However quantification of the film alignment by the Lorentz images requires caution as there are indications that some crystal domain reorientation occurs when the specimen is removed from the substrate. Figure 6(a) shows the magnetic microstructure of a film, deposited on a miscut substrate, estimated to be greater than 95% \( c_1 \) aligned from magnetization measurements. Large areas of the film are indeed \( c_1 \) aligned, as shown in Fig. 6(a) and untwinned, as confirmed by the contrast changes with tilting described previously. However, a number of \( c_{\perp} \) domains are observed, as indicated by the striped wall orientation. Overall, about 15% of the film appears misaligned, confirming some reorientation of the film upon removal from the substrate.

Despite the indications of some reorientation, the TEM samples do indicate a high degree of alignment for SrRuO\(_3\) on miscut substrates and are expected to give a reasonable indication of crystal domain structure variations from substrate to substrate. Figures 6(b) and 6(c) compare the magnetic microstructure of two other films, deposited on (100) SrTiO\(_3\) (not miscut) with the aligned film; Fig. 6(b) shows many small \( c_{\parallel} \) domains and a few, large \( c_{\perp} \) domains, whereas Fig. 6(c) shows fewer \( c_{\parallel} \) domains and many, small \( c_{\perp} \) domains. We have also previously observed variations in domain size between SrTiO\(_3\) and LaAlO\(_3\) substrates. The crystal domain boundaries tend to follow (110) planes, as shown in Fig. 6(d), an OLM, higher magnification image of the film in Fig. 6(c). A high density of end-on dislocations (black dots) is also visible in Fig. 6(d).

**D. Analysis of the domain wall spacing**

Lorentz microscopy gives direct measurement of the domain wall spacing. We observe that the striped walls of \( c_1 \) and \( c_{\perp} \) domains differ both in spacing and in the dependence of spacing on crystal domain size. The \( c_1 \) domain walls are
more closely spaced, having an average spacing of about 2000 Å, independent of either crystal domain size or thickness for film thicknesses of about 300–1000 Å. The c$_1$ regions, on the other hand, are on the order of a micron or more in spacing and the spacing varies considerably. These observations correlate with the anisotropy and the shape dependence of the demagnetizing factor. The magnetostatic energy of c$_1$ regions, with the magnetization out-of-plane, is higher than that of c$_\perp$ regions, and the domain wall spacings are correspondingly smaller. They also show little dependence on crystal domain size since the demagnetizing factor for this out-of-plane flat-plane geometry is about $1/2\pi$ and correlates with the aspect ratio of domain size to film thickness which is always very large. When the magnetization is in-plane, as for c$_\perp$ regions, the demagnetizing factor is smaller. It also depends on the domain dimension in the direction of magnetization which now corresponds to the domain size. The striped spacing of c$_\perp$ regions therefore shows a strong dependence on the size of the domains, increasing as the domains become larger (Fig. 1).

In principle, the domain wall spacing represents a balance between the magnetostatic energy, which is reduced by domain formation, and the domain wall energy. Therefore, magnetization constants such as the domain wall energy and the anisotropy constant should be obtainable from the domain wall spacing measured by TEM combined with the saturation magnetization and exchange stiffness measured on the aligned films. An expression for single crystals with uniaxial magnetic anisotropy is given by Cullity:

$$D = (\gamma L/1.7M_t^2)^{1/2},$$

where $D$ is the domain wall spacing, $\gamma$ is the wall energy, $M_t$ is the saturation magnetization, and $L$ is the dimension parallel to the magnetization direction, i.e., approximately the film thickness for c$_1$ regions and the crystal domain dimension parallel to the walls for c$_\perp$ regions.

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\gamma$ is then related to the anisotropy constant by

$$\gamma = 2(JS^2\pi^2K/La)^{1/2},$$

where $J$ is the exchange constant, $S$ is the spin, and $K$ is the anisotropy constant. Using $M_t = 213$ emu/cm$^3$, $J = 26.33k_B$, and $S = 1$, we have the relation for the anisotropy constant,

$$K = 1.62 \times 10^{15}(D^4/L^2)\text{ ergs/cm}^3.$$  

For c$_\perp$ regions, with $D = 2\mu$m and $L = 35\mu$m and $11\mu$m as typical values for large and small crystal domains we obtain $K$ on the order of $1-2 \times 10^5$ ergs/cm$^3$, two orders of magnitude lower than that expected from magnetization measurements on the aligned films. The latter indicate $K \approx 10^7$ ergs/cm$^3$. However, the variation of $D$ with $L$ is reasonably consistent for c$_\perp$ domains. For c$_1$ grains, using $M_t = 151$ emu/cm$^3$, $D = 2000$ Å and $L = 1000$ Å, we obtain a slightly higher value, $K = 6.7 \times 10^5$ ergs/cm$^3$. Cullity’s description assumes $L > D$ and therefore negligible interaction between surfaces, which does not strictly hold for c$_1$ regions. A more precise expression for the magnetostatic energy by Malek and Kambersky accounts for this interaction with a prefactor summation term depending on $L/D$; for c$_\perp$ regions inclusion of this factor decreases $K$ further so that the values for both types of regions are approximately the same, also giving internal consistency. However, since the c$_1$ regions do not show an obvious dependence on $L/D$ for the film thicknesses observed here (300–1000 Å), and since the calculated values of $K$ are much lower than indicated by other measurements, it is clear that the relationship of the domain wall spacing to the magnetization parameters requires clarification beyond these standard models.

**In situ** Lorentz TEM gives valuable insight into the situation, allowing direct observation of additional factors controlling the domain wall spacing. We find that the domain wall distribution that forms during cooling does not represent magnetostatic equilibrium, even for the minimum cold stage temperature of 100 K, as will be discussed in more detail below. Also, since both the saturation magnetization and the anisotropy constant are temperature dependent, we must take into account the different temperature ranges in which the measurements are made. The magnetization constants derived from macroscopic magnetization measurements of aligned films are made at 4.2 K, whereas the TEM observations are in the range from $T_c$ to 100 K. The saturation magnetization and anisotropy constant are changing rapidly as the temperature decreases below $T_c$, the effects of which we observe by TEM, and have values quite different from those measured at low temperature. Finally, the domain wall spacing does not depend on the film thickness, as the above expressions predict, for films in the range of 300–1000 Å.

Using Lorentz microscopy, we observe directly the process of domain formation and the qualitative variation of the magnetization parameters during cooling. Figure 7 shows a series of video frames during cooling of a c$_3$ grain. As the grain is cooled through $T_c$, a very regular, faint stripe structure begins to form with a wall spacing of about 1000 Å [Fig. 7(a)]. The specimen is cooling at a rate of about 7°/min. In Fig. 7(b), one degree cooler, the stripe structure is much stronger in contrast, but still very regular with about the same spacing. As the cooling progresses, both the wall contrast and the average wall spacing increase, the latter becoming more irregular due to annihilation of pairs of black-and-white walls [Figs. 7(c) and 7(d)]. Both the magnetization and anisotropy, which have opposite influence on domain size, are increasing rapidly with decreasing temperature, the competition between them clearly favoring an increase in the domain wall spacing, i.e., the anisotropy energy contained in the wall is increasing faster than the reduction of magnetostatic energy due to the presence of domains. However once the walls have formed, their average spacing can only increase by annihilation of a pair of walls as opposed to individual wall movement, since the latter enlarges one domain at the expense of another. Furthermore, the energetics after wall formation is also influenced by the energy required to move a wall: it is observed that following wall pair annihilation, the adjacent walls tend to keep their position rather than adjusting to a more regular spacing, indicating that the walls are already immobile, relative to these magnetostatic energy changes, a few degrees below $T_c$. Wall motion and/or annihilation ceases about 15° below $T_c$ and the remaining stripe structure has a distribution of domain spacings with an average spacing of about 2000 Å for c$_1$ domains. This final spacing of the domain walls clearly does not represent magnetostatic equilibrium, but depends on magnetodynamic energetics of domain wall motion, and also becomes frozen in at temperatures fairly near $T_c$ where mag-
netization parameters are more difficult to measure than at temperatures well below $T_c$.

The fact that $D$ does not vary significantly with film thickness for $c_{\parallel}$ regions further indicates that the wall energy per area for these regions is not a constant as assumed in calculating $K$, but likely contains a magnetostatic term due to the intersection of the wall with the surface. For the range of film thicknesses studied here, the wall energy may depend significantly on the surface term and the energy per area may be therefore both variable and underestimated. We plan in the future to examine ultrathin films, 100 Å or less, as well as thicker films, $\sim$2000 Å being an upper limit for TEM transparency, to investigate further the effects of film thickness on $c_{\parallel}$ domain spacing.

Quantitative information about the domain wall resistivity is available from the $c_{\parallel}$ domain wall spacing. A zero-field-cooled, aligned film exhibits large irreversible magnetoresistance with an applied field of about 2 kG, as shown in Fig. 8. This corresponds to a resistivity of approximately $10^{-15}$ $\Omega$ m$^2$/unit area.$^{10}$ Due to the high anisotropy, the domain walls are expected to be Bloch walls. Calculation of the domain wall width using $d = (0.3kT_c\pi^2/4Ka)^{1/2}$ (see Ref. 15, p. 291), where $d$ is the wall width, $k$ is Boltzmann’s constant, and $K$ is the anisotropy constant, is assumed to be $10^7$ ergs/cm$^3$, indicates a very narrow domain wall width of about 20 Å. This is again due to the large anisotropy constant estimated at 4.2 K; the walls are expected to be wider at the TEM observation temperatures.

E. Domain wall motion in an applied field

Domain wall motion is observed by turning on the field of the objective imaging lens. This field is normal to the untitled specimen plane; an in-plane component of field is achieved by tilting the specimen. As the field is applied, pairs of black-and-white stripe domain walls move together and annihilate. An example is shown in Fig. 9 where the in-plane component of the field is parallel to the right-hand axis of the specimen.
As seen in Fig. 9, when a field is applied, the domains aligned with the field are the first to grow as evidenced by comparing the domain wall structures in (a) zero field and (b) $H=0.2\, \text{kG}$ with an in-plane direction as marked. The $c_{\perp}$ domain walls parallel to the field, such as those marked 1, 2, 3, 4, and 5, have moved and/or partially annihilated in (b). The black walls are moving up and the white walls down as aligned domains grow. It is seen that the walls become crooked, with bowed and kinked segments, as they move. The shorter white arrows show two points where black and white walls join, their segments having annihilated to the left of these points. The $c_{\perp}$ domain walls which are oriented at $90^\circ$ to the applied field, such as those marked 6, 7, 8, and 9, remain unchanged.

As the field continues to increase all of the stripe walls eventually annihilate. Above about 2 kG, when the stripe walls have completely disappeared, magnetic domain walls remain between crystallographic domains. This indicates that domain rotation has not occurred; for such a region, where neither set of domains is macroscopically aligned toward the field direction, it is likely that one set has a small magnetization component along the field which favors its growth over the other set, leading to domain wall annihilation. The final structure of domain walls at crystallographic boundaries persists even after the objective lens is increased to its maximum value of about 2 T, and then back to zero field for imaging. This is shown in Fig. 10 which compares the same region of the specimen by conventional and high resolution imaging. The high crystalline grains or domain, and the latter between twin domains or a $c_{\parallel}$ region. Most of the $c_{\parallel}$ walls visible in Fig. 9 show strong contrast and resist the field. However careful examination of the $c_{\parallel}$ walls just to the left of center shows that this area has weaker contrast in Fig. 9(a) and some movement of the walls toward each other in Fig. 9(b). The remaining $c_{\parallel}$ walls show strong contrast and resist movement until higher fields.

As the field is increased and is completely annihilated at a field estimated to be about 2 kG from the objective lens current (500 mA), consistent with the magnetoresistance measurement. Although we cannot reverse the objective lens field, we can reverse the in-plane field component by tilting the specimen in the opposite direction; this affects only the $c_{\perp}$ regions. We observe no reformation of the stripe structure, again consistent with the magnetoresistance behavior. Rather, as the specimen tilt is reversed, the magnetization of $c_{\perp}$ regions reverses by the movement of individual domain walls across the film. However the stripe structure can be reproduced, with the precise wall positions varying somewhat, by warming and cooling the film through $T_c$. In this way, the same region can be studied with the field applied in different directions (different specimen tilt direction). The stripe domains are observed to annihilate in a sequence that is consistent with the direction of tilt, or field. The most aligned domains are the first to grow and lose their striped walls, starting with an applied field of about 0.2 kG; the least aligned are last. For example, the $c_{\perp}$ domain walls on the right-hand side of Fig. 9, have moved together and partly annihilated in a field of about 0.3 kG (90 mA), while those on the left-hand side, which remain perpendicular to the field during tilting, have not changed position. For this tilt some of the $c_{\parallel}$ domains will also be favorably oriented in the field direction which will be indicated by a loss of contrast (as previously demonstrated in Fig. 5). Most of the $c_{\parallel}$ walls visible in Fig. 9 show strong contrast and resist the field. However careful examination of the $c_{\parallel}$ walls just to the left of center shows that this area has weaker contrast in Fig. 9(a) and some movement of the walls toward each other in Fig. 9(b). The remaining $c_{\parallel}$ walls show strong contrast and resist movement until higher fields.

Note that the domain walls observed in Fig. 10 are no longer $180^\circ$ walls but are approximately $60^\circ$ and $90^\circ$ walls, the former occurring between two orthogonal $c$ axis $c_{\parallel}$ domains or a $c_{\parallel}$ and $c_{\perp}$ domain, and the latter between twin-related $\pm c_{\parallel}$ $c_{\parallel}$ grains or $c_{\perp}$ domains. The tip of the small $c_{\parallel}$ twin just to the left of center in Fig. 10(c) does not show domain boundary contrast because the magnetic de-
flection of the beam is parallel to the boundary in this region rather than perpendicular to it.

There are several features of the domain walls that are unusual and indicate directions for future analysis. One is the central spot observed in the $c_{ij}$ Lorentz diffraction pattern, suggesting a stable state of perpendicular magnetization occurring in some fraction of the domains or domain wall structure. Another is the response of the domain walls as the applied field is increased: the walls first move together in a fairly continuous way with increasing field, but some then reach a state where they remain “stuck” on each other, i.e., close together (estimated as 900 Å or less), but failing to annihilate until noticeably higher fields are reached. A possible explanation for such behavior is the concept of 360° walls, i.e., a state of a pair of walls where the magnetization rotation across both walls varies continuously by 360°. This is in contrast to rotation of $+180°$ across one wall and $-180°$ across the adjacent one. Annihilation of the former state would be expected to require more energy than the latter. A third feature of the domain wall anihilation sometimes observed is a fading of wall segments just prior to anihilation, also indicating an intermediate or transient state in the anihilation process. All of the above suggest complex domain wall structures, perhaps metastable states, not predicted by basic models. Finally when a magnetized film is warmed through $T_c$, the domain walls are observed to return just below $T_c$ and (in a apparent reversal of the cooling process) to become more finely spaced with the increasing temperature until $T_c$ is reached and they disappear. Although it is not clear if this latter effect, which is again due to magnetization parameters changing with temperature, will reveal any new physics of the domain walls, there is the potential of using the initial few domains as a starting point for studying reversal effects.

IV. CONCLUSIONS

Lorentz TEM is a powerful tool for analyzing the magnetic microstructure, crystal structure, and in situ magneto-dynamic behavior of these SrRuO$_3$ films. Furthermore, their highly organized crystal structure and uniaxial magnetic anisotropy make these films ideal for such studies. Using this technique we have: (1) independently confirmed the uniaxial anisotropy found for aligned films and found it to be largely magnetocrystalline in nature; (2) identified the easy axis as the orthorhombic $b$ axis; (3) compared the crystal domain structure of films on conventional and miscut substrates; (4) analyzed the domain wall spacing and observed details of its formation relative to the ferromagnetic energy state; (5) measured the domain wall resistivity, and (6) characterized the
magnetodynamic behavior of wall anihilation. The results also indicate future directions for using Lorentz microscopy to study magnetodynamics and details of domain wall structures in these films.

ACKNOWLEDGMENT

This work was supported by the NSF-MRSEC program through the Center for Materials Research at Stanford University.

13 That the center of the pattern lies between the split spots in Fig. 4(a) was confirmed by overlapping the patterns from the two areas. A peculiar and consistent feature of $c_i$ regions is the occurrence of a central spot between the split spots. This suggests regions of magnetization parallel to the beam (or possibly unmagnetized regions), and is believed related to the domain wall structure. Attempts to further identify the source of this spot by observing changes with tilting and applied field have been ambiguous and will be pursued in more detail in future experiments.
14 C. B. Eom (private communication).