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Observation of step-induced magnetic domain formation in $La_{1-x}Sr_xMnO_3$ thin films by photoelectron emission microscopy

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Magnetic domain formation in thin films of $La_{1-x}Sr_xMnO_3$ (LSMO) with $x=0.2$ and 0.4 epitaxially grown on stepped $SrTiO_3$ (001) substrates has been investigated by photoelectron emission microscopy. The magnetic domains show a stripe structure elongated along the step directions, indicating uniaxial magnetic anisotropy induced by step structures. We have also found that the magnetization of the magnetic domains is slightly meandering at domain boundaries. The existence of the additional structures suggests that the magnetic domains in LSMO films are also influenced by biaxial magnetocrystalline anisotropy. The direct observation of the magnetic domain structures suggests that the competition between the two magnetic anisotropies may play an important role in magnetic properties in LSMO films. © *2006 American Institute of Physics*.
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Spin tunnel junctions employing half-metallic ferromagnets as electrodes have attracted considerable attention because of their potential applications to magnetoelectronic devices. Hole-doped manganite $La_{1-x}Sr_xMnO_3$ (LSMO) is one of the most promising materials for such applications.[1] Since the anisotropic magnetic behavior of LSMO films is very sensitive to lattice strain induced by the underlying substrates,[2–4] a great number of the experimental studies have been focused on revealing the unusual magnetic properties of LSMO films. For example, magnetic torque measurements[3–6] have revealed that LSMO films grown on $SrTiO_3$ (STO) substrates with tensile strain exhibit in-plane biaxial magnetic anisotropy with the easy axis along the [110] direction. On the other hand, the importance of step-induced uniaxial anisotropy has also been pointed out for LSMO films grown on STO substrates with a large vicinal angle of 10°.[7] Recent magnetic torque measurements suggest the coexistence of biaxial magnetocrystalline anisotropy with uniaxial step-induced anisotropy in LSMO films grown on STO substrates.[8] In the double-exchange picture, the probability of carrier hopping between parallel spins is higher than between antiparallel spins due to the energy cost of flipping the spin of a carrier. Information on the magnetic anisotropy and domain structure of LSMO films is thus crucial for understanding the carrier dynamics in LSMO-based magnetoelectronic devices.

The magnetic domain structure of LSMO films has been intensively investigated by magnetic force microscopy (MFM).[2,9] However, due to a lack of information on the in-plane domain structure, there have been only a few studies on the magnetic domain structure of LSMO films with tensile epitaxial strain, grown on STO substrates, which are widely used for magnetoelectronic devices. Direct observation of the magnetic domain structure is the key to understanding the dynamics of fully polarized conduction carriers, which dominates the characteristics of the LSMO-based devices. Photoelectron emission microscopy (PEEM) combined with x-ray magnetic circular dichroism (XMCD) is a powerful technique for studying this issue. Since XMCD provides information on the magnetization vector projected to the incident direction of the synchrotron radiation (SR) beam, it is possible to obtain all three independent components of the magnetization vector by rotating the sample. Mapping of the magnetization distribution with high spatial resolution enables us to observe the magnetic domain structures in LSMO films. In this letter, we report on direct XMCD-PEEM observation of the in-plane magnetic domain structure of LSMO films epitaxially grown on STO substrates.

LSMO films with $x=0.4$ (Curie temperature $T_C=350$ K) and $x=0.2$ ($T_C=290$ K) were grown by laser molecular beam

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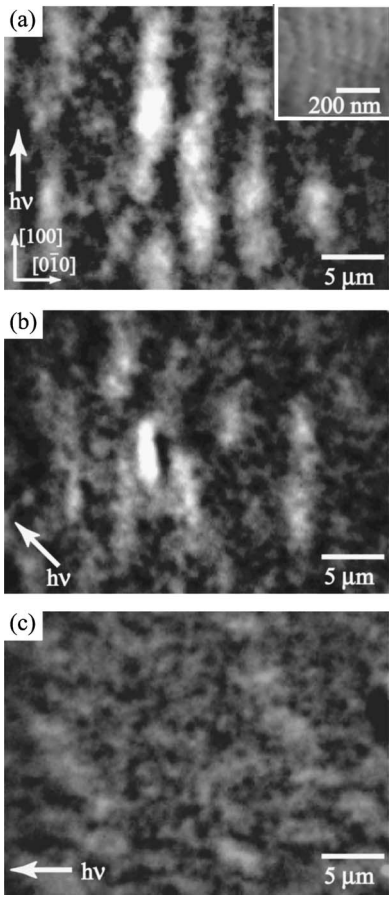

FIG. 1. Magnetic images of ferromagnetic $La_{0.6}Sr_{0.4}MnO_3$ thin films taken at room temperature (T=300 K) with a photon energy of 641 eV, corresponding to the Mn L_3 edge. The incident directions of the synchrotron radiation beam are along the (a) [100], (b) [110], and (c) [010] directions, where the atomic step direction is aligned with the [100] direction. The inset shows an AFM image of the measured LSMO films.

epitaxy.[10] We describe the orientation of the epitaxial LSMO films in terms of the pseudocubic lattice. LSMO films with a thickness of 40 nm were deposited on atomically-flat TiO_2-terminated Nb-doped STO (001) substrates with regular straight steps. STO crystals with an approximately 0.2° miscut with respect to the crystallographic (001) surface toward the [010] direction were used in order to obtain straight surface steps along the [100] direction. The films were grown in a step-flow mode at a substrate temperature of 950 °C at an oxygen pressure of 1×10^{-4} Torr.[10,11] The coherent growth of the LSMO films was confirmed by four-circle x-ray diffraction[10,11] The surface morphology of the measured films was analyzed by atomic force microscopy (AFM), and atomically flat step-and-terrace structures were clearly observed, as shown in the inset of Fig. 1(a). The magnetization was measured by a superconducting quantum-interference device (SQUID) magnetometer.

The XMCD-PEEM experiments were carried out using Elmitec PEEMSPECTOR at the soft-x-ray undulator beamline BL25SU at SPring-8. The polar angle between the incident direction of the SR beam and the sample surface was set at 30°. The spatial resolution of XMCD-PEEM images is estimated to be better than 100 nm. The PEEM images were recorded with the photon energy at the Mn L_3 absorption edge (641 eV).[12] Magnetic images were obtained from the difference between two images taken with right- and left-circularly polarized lights.

Figures 1(a)–1(c) show magnetic domain images of a LSMO (x=0.4) film taken at azimuthal angles of 0°, 45°, and 90° between the incident SR beam direction and the [100] direction, respectively. The bright and dark parts in the images correspond to magnetic domains that have their magnetization vectors parallel and antiparallel to the incident SR beam, respectively. The contrast of the XMCD-PEEM images is normalized by the x-ray absorption images, namely, summation of images taken with right- and left-circularly polarized lights. The magnetic images shown in Fig. 1 are drawn on the same gray scale for comparison. The elongated magnetic domains along the [100] direction can be clearly observed in Fig. 1(a) where the incident SR beam is parallel to the [100] direction. The average length and width of the magnetic domains are estimated to be approximately 30 and 3 μm, respectively. The existence of the "stripe" domains indicates that magnetic domains with the magnetization components parallel or antiparallel to the [100] direction are predominantly formed in the LSMO films.

The presence of stripe magnetic domains that are magnetized either parallel or antiparallel to the [100] direction is clearly indicative of the uniaxial magnetic anisotropy of the LSMO thin films. The uniaxial magnetic anisotropy may originate from breaking of rotational symmetry at the surface and/or interface, since the [100] and [010] directions are crystallographically equivalent in the case of LSMO films grown on STO substrates. The anisotropic behavior of LSMO films may be induced by the step structure on the surface of the LSMO films and/or at the interface between the LSMO films and STO substrates, since the step directions of the substrates and the LSMO films are both aligned along the [100] direction in the present case. These results suggest that the structural symmetry dominating magnetic anisotropy can be broken by a periodic array of atomic steps generated by cutting the substrate crystal at a small angle off a low-index plane and consequently the step structure controls the direction of the magnetization easy axis in LSMO films.[8] Similar uniaxial magnetic anisotropy has been reported to exist in 3d transition metal or alloy films grown on vicinal substrates.[13,14] For LSMO films, Wang *et al.* have observed uniaxial magnetic anisotropy in a $La_{0.67}Sr_{0.33}MnO_3$ film grown on a STO substrate with a large vicinal angle of 10°.[7] Although the step density of the LSMO films in the present study is smaller by about two orders of magnitude, we have clearly found magnetic domain formation with step-induced uniaxial anisotropy, suggesting the importance of symmetry breaking by the step structure in LSMO films.

The interpretation of the data in terms of uniaxial magnetic domain formation is further supported by the azimuthal angle dependence of XMCD-PEEM images, as shown in Fig. 1. Since the contrast in XMCD-PEEM images reflects the components of the magnetization vector projected along the incident SR beam direction, the modulation of the XMCD-PEEM images as a function of the azimuthal angle corresponds to magnetic torque measurements at zero magnetic field. The images in Fig. 1 show that the contrast of the stripe magnetic domain, which is the strongest at 0° [Fig. 1(a)], becomes weaker with increasing azimuthal angle. The stripe magnetic domains completely disappear when the direction of the SR beam is perpendicular to the step direction [Fig. 1(c)], as expected for step-induced uniaxial magnetic

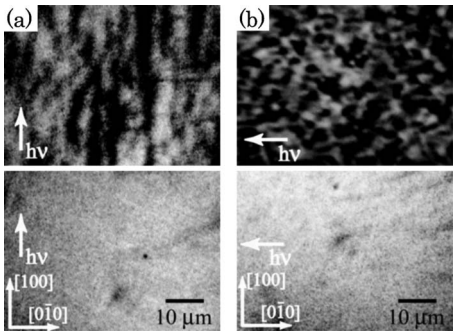

FIG. 2. Magnetic images of ferromagnetic (upper panel, $T=160$ K) and paramagnetic (lower panel, $T=300$ K) $La_{0.8}Sr_{0.2}MnO_3$ thin films. The incident direction of the SR beam is (a) parallel and (b) perpendicular to the step structure. The photon energy is set at the Mn L_3 edge.

anisotropy. On the other hand, alternative "bubblelike" images appear in Fig. 1(c) suggesting the existence of additional magnetization components perpendicular to the step direction. In order to clarify whether the anomalous images observed in the LSMO films are magnetic in origin, we have performed magnetic imaging of LSMO films with $x=0.2$ across the ferromagnetic transition. The magnetic images of ferromagnetic LSMO ($x=0.2$) films with $T_C=290$ K were similar to the images of LSMO ($x=0.4$) films, as shown in Fig. 2. The magnetic domain structures completely disappeared above T_C, indicating that the observed magnetic images are intrinsic. The possibility of explaining the bubblelike features by the presence of out-of-plane magnetization can be eliminated, since the out-of-plane component is hardly detectable in MFM measurements of LSMO films grown on STO substrates.[2]

The domain contrast seems to be blurred at the domain walls, as shown in Fig. 1(a). The blurred parts may correspond to the bubblelike images in Fig. 1(c). The same behavior in magnetic images is also observed in ferromagnetic LSMO ($x=0.2$) films. In general, ferromagnets with uniaxial magnetic anisotropy form stripe magnetic domains in which the magnetization distribution is alternatively parallel or antiparallel to the easy axis. The magnetic images from typical ferromagnets with uniaxial magnetic anisotropy show clear straight stripe domain patterns with alternating bright (parallel) and dark (antiparallel) contrasts.[15] In contrast, the present PEEM images of LSMO films exhibit unusual blurred contrasts indicative of meandering magnetization at the domain boundary. Such magnetic domain formation strongly suggests the presence of additional magnetic anisotropy. It should be noted that the blurring in the magnetic images is not due to insufficient spatial resolution, since the spatial resolution of magnetic imaging by PEEM is much higher (<100 nm).

Finally, we discuss the origin of the imperfect stripe domain formation observed in the ferromagnetic LSMO films. The observation of additional magnetization components perpendicular to the step direction strongly suggests that the magnetization direction in the LSMO films is not completely aligned parallel or antiparallel to the step direction. A reasonable explanation is the influence of magnetocrystalline anisotropy in LSMO films. Magnetic torque measurements have elucidated the existence of biaxial magnetocrystalline anisotropy with the easy axis along the [110] direction in LSMO films on STO substrates.[4] Therefore, it is reasonable to conclude that the observed meandering magnetic stripe domains originate from the competition between the step-induced uniaxial magnetic anisotropy and the biaxial magnetocrystalline anisotropy. In fact, the coexistence of two anisotropies with different symmetries is suggested by recent magnetic torque measurements on LSMO films with regular steps, where magnetization showed twofold symmetry with respect to the step direction at room temperature in spite of the fourfold symmetry with the easy axis along the [110] direction at low temperature.[8] These results suggest that LSMO films can show complicated magnetic domain structures as a result of competition between two magnetic anisotropies with different symmetries. In order to understand the unusual magnetic behavior of LSMO films, systematic XMCD-PEEM studies as a function of film thickness, step density, and/or temperature are needed.

In summary, we have performed an XMCD-PEEM study on LSMO ($x=0.2$ and 0.4) epitaxial thin films on stepped STO substrates in order to investigate the complicated magnetic domain structure of LSMO films. The overall magnetic domain structure can be explained by step-induced uniaxial magnetic anisotropy with the easy axis along the step direction. The azimuthal angular dependence of the magnetic images reveals the presence of an additional magnetic anisotropy in LSMO thin films, suggesting the existence of competition between step-induced uniaxial magnetic anisotropy and biaxial magnetocrystalline anisotropy in strained LSMO films.

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