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Nanoscale magnetic-domain structure in colossal magnetoresistance islands

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Magnetic force microscopy reveals the nature of local magnetic structure in submicron islands of colossal magnetoresistance (CMR) thin films. The evolution of domains in a magnetic field reveals the importance of shape anisotropy as well as magnetostriction in determining the micromagnetics in such small CMR structures. At room temperature, a characteristic multidomain structure with perpendicular orientation, predicted by theory, is observed. The magnetization reversal of the islands in magnetic fields perpendicular and parallel to the substrate is dominated by strong domain wall pinning. Strong domain wall pinning in conjunction with geometrical confinement in these submicron CMR structures gives rise to reproducible multidomain structure.

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Doped perovskite manganites have elicited renewed interest because they exhibit a rich variety of electronic and magnetic behaviors, including colossal magnetoresistance (CMR), charge ordering, and phase segregation. Because of the large magnetoresistance effect and strong spin polarization at the Fermi level, these oxides may find important uses in magnetoresistive devices, such as magnetic random access memory and sensors. One such device is a current perpendicular magnetic tunnel junction, comprised of two layers of CMR thin films sandwiching a nonmagnetic barrier. Experimentally junction MR's of up to a factor of 10 resistance change have been reported.[1,2] The details of the magnetic response in such a device is strongly affected by the complex magnetic-domain state. Such micromagnetic effects have greatly complicated the interpretation of transport data and may contribute to the premature suppression of large magnetoresistance to temperatures well below the Curie temperature of the CMR electrodes. Thus far, imaging of the magnetic-domain state in CMR thin film materials has been limited to unpatterned thin films in zero field.[3,4] As the size of the magnetic object is decreased down to length scales comparable to the domain size and magnetic exchange length, processes that are very different from macroscopic ones may begin to dominate. At nanometer length scales, the competition between magnetostatic energy and exchange energy should suppress magnetic-domain formation, thus resulting in single magnetic-domain nanometer size particles.[5] Even before reaching this fundamental crossover point, in submicron size magnetic islands, the magnetic state is already greatly altered from macroscopic samples by the effects of strain and shape.

The structural sensitivity of the magnetic and electronic properties of this class of materials has been the focus of many experimental, as well as theoretical, studies in bulk and thin film samples. Many groups have shown that transport properties are extremely sensitive to chemical pressure (e.g., due to alkaline earth substitution of the rare-earth ion), hydrostatic pressure, as well as lattice mismatch of the film with the substrate.[6–11] Geometrical confinement (e.g., patterning films into islands) also provides an additional parameter to control the strain state of these epitaxial films. Therefore CMR nanostructures provide a rich model system for studying the influence of structure on the magnetics and transport.

In this paper, we present the nature of the local magnetic structure and its evolution with field in submicron CMR islands. By varying the thickness, diameter and thus the aspect ratio of these islands, we have found that the biaxial strain, due to the lattice mismatch with the substrate, dominates over strain relaxation effects at island edges and magnetocrystalline effects in determining the preferential orientation of the magnetization. We confirm the characteristic multidomain structure with perpendicular orientation that is predicted by Kittel. Such multidomain structure in submicron structures has only been confirmed in cobalt.[12] However, in contrast to cobalt where the magnetic structure is identical from island to island, the CMR islands exhibit a variety of reproducible magnetic structure, thus suggesting strong domain wall pinning. In addition, while the local domain structure in the CMR islands and upatterned films are similar, the magnetization reversal process in these islands differs greatly from unpatterned CMR films.

We have studied islands of $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) that are widely spaced so as to minimize magnetic interaction among islands. The islands were patterned from continuous films grown by pulsed laser deposition (PLD) on (001) oriented $LaAlO_3$ substrates. Structural characterization, discussed elsewhere, reveals excellent crystallinity.[9] We have probed the surface morphology of these films and have found that the rms roughness of the (001) films is around 1 nm for 10–90 nm thick films. We then pattern these continuous films by optical lithography into 0.3–2 μm diameter islands with height varying from 25 to 140 nm. In order to ensure that we were not degrading the CMR materials by wet and dry lithographic processing, we compared the magnetization versus temperature of continuous films with those of island arrays and found no suppression of the Curie temperature for islands thicker than 45 nm. We focus on the 70 and 90 nm high unpatterned films and islands at room temperature for most of the analysis to follow.

We have used magnetic force microscopy (MFM) in order to probe the local magnetic properties of the island structures with a resolution of 50 nm. The light and dark areas in our MFM images correspond to moments pointing ''in'' and ''out'' of the plane of the substrate. Since MFM images the magnetic structure of the surface, the observed domain structure is representative of domain structure averaged over ap-

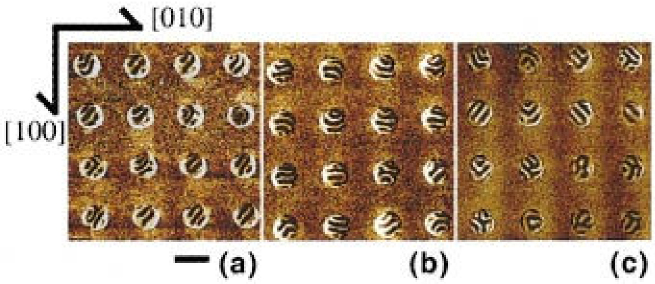

FIG. 1. (Color) Stripe domains of CMR islands in zero field after saturation at room temperature in (a) an in-plane field oriented diagonal to the array, (b) an in-plane field oriented parallel to the array, and (c) a perpendicular field.

proximately an exchange length (~50 nm) below the surface, which is a significant fraction of the film thickness.

At room temperature, the domain structure is dominated by compressive strain effects from the underlying substrate that give rise to perpendicular domains according to magnetoelastic theory. Figure 1 shows MFM images of arrays of nominally 90 nm high LSMO islands in its remnant state. In these images, the sample is placed in a magnetic field, in various directions, large enough to place the sample in a single domain state and then its magnetization is measured after the field is reduced to zero. The black line is a 1 μm bar. The magnetic images [Fig. 1(a) and (b)], measured at zero field after saturation in in-plane fields, reveal stripe domains on the order of 150 nm. These stripe domains are formed parallel to the direction of the previously applied field and are a result of the minimization of the associated Zeeman energy. The (001) CMR islands have [100] and [010] directions along the edges of the array. Magnetic fields applied parallel or diagonal to the crystal axis directions reveal no significant magnetic anisotropy in the plane of the film. MFM images of CMR islands in the remnant state after saturation in a perpendicular field reveal a variety of domain shapes. Without the associated Zeeman effects, the magnetic-domain configuration in the remnant state varies from island to island [stripes, circles, etc., in Fig. 1(c)].

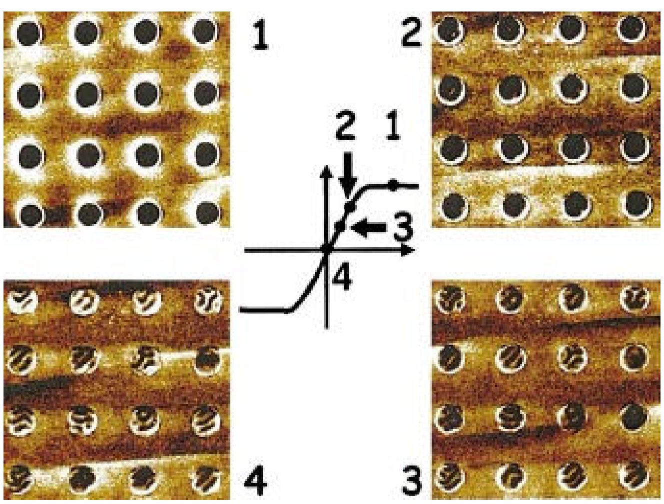

FIG. 2. (Color) Field dependence of the stripe domains of CMR islands in a perpendicular magnetic field.

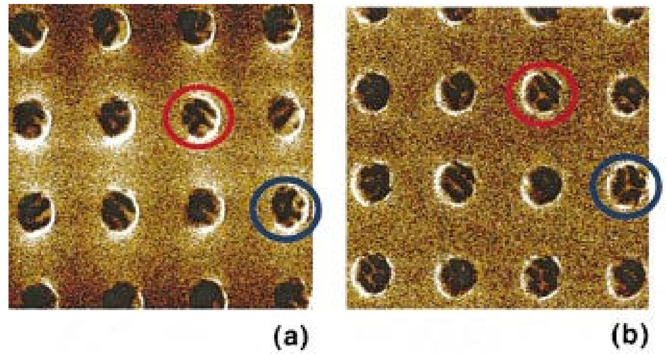

FIG. 3. (Color) Magnetic domain images of CMR islands in a perpendicular magnetic field of +/−450 Oe. The blue and red circles indicate the reversal of domains in Y-shape and T-shape domains.

In order to understand the magnetization reversal process in the CMR islands, we have performed MFM studies under the application of a perpendicular magnetic field. Figure 2 shows four images of the local magnetic structure for a range of magnetic fields. The images are direct evidence of the domain nucleation and growth process by which magnetization reversal occurs. At large fields, the islands are saturated in the direction of the applied field. The halolike structure at the edges of the islands is believed to be an experimental artifact since it is also observed around nonmagnetic control structures. At lower fields, the bubble domains nucleate and proceed to grow as the field is reduced (No. 2 in Fig. 2). There is very little change in the domain structure as the magnetic field is further reduced (No. 3 to No. 4 in Fig. 2), thus indicating that there is strong pinning of domain walls. Moreover a snapshot of the islands at 450 Oe in and out of the plane of the substrate in Fig. 4 reveals the reproducibility of the domain structure. Figure 3 shows reversed images of a T-shape domain structure (red circle) and a Y-shape structure (blue circle) in opposite polarities of magnetic field. The reproducibility of the structure in opposite fields is further evidence for strong domain wall pinning. This domain wall pinning cannot be attributed to thickness variation of the islands, given the 1 nm surface roughness of the structures. The observed domain structure may be associated with pinning due to grain boundaries and cation disorder (including phase segregation effects) that have a characteristic length scale on the order of 100 nm.

In cobalt submicron islands, the magnetic structure is identical from island to island and the domain walls avoid

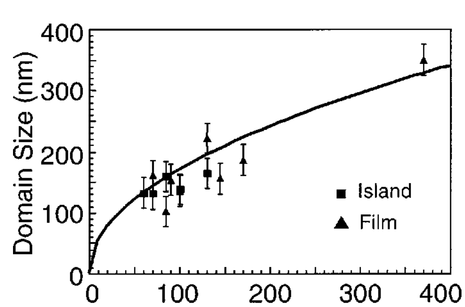

FIG. 4. Domain size versus thickness of CMR islands.

the edges in order to minimize magnetostatic energy.[12] By contrast, the CMR islands exhibit remnant magnetic-domain configurations, that vary from island to island. Studies of the evolution of magnetic-domain structure in a magnetic field in unpatterned films reveal domain sizes similar to the islands at zero field and strong pinning behavior at low fields. Patterning gives rise to edges where the cost in magnetostatic energy of having domain walls perpendicular to the edge is significant; the energy gained from pinning overcomes this magnetostatic energy to give rise to the observed reproducible domain structure (Fig. 3).

The aspect ratios of the islands were varied by varying their thickness and lateral dimensions. Domain sizes on the order of 150 nm are clearly observed in 90 nm high islands in MFM images at zero field. In 55 nm high islands, we are able to observe stripe domains, though it is hard to measure domain size due to the lateral resolution of 50 nm in our MFM. Macroscopic magnetization measurements of samples 45 nm in height reveal no suppression of T_c after patterning. MFM images reveal, however, very little magnetic structure after processing. The lack of structure in the MFM images may be an indication of resolution limited perpendicular domains or in-plane domains. The temperature dependence of the magnetization of 45 nm islands is reminiscent of the surface magnetization measured by magnetic circular dichroism and spin resolved photoemission experiments.[13] While 45 nm is substantially larger than the probe depths of 5 and 0.5 nm, respectively, for the above techniques, such suppression of magnetization at the surface combined with geometrical constraints may significantly reduce the projection of magnetic moment perpendicular to the substrate.

In our CMR islands where the lateral dimension of the islands are more than a few times the film thickness, we do not expect the strain relaxation that occurs at the edges to alter the strain state dramatically as the aspect ratio of the islands is varied.[14] In fact the compressive strain is more strongly a function of the island thickness rather than lateral dimension. Normal and grazing incidence x-ray diffraction confirm that the unit cell relaxes from a tetragonal ($a_{\text{in-plane}} = 0.3832$ nm, $a_{\text{out-of-plane}} = 0.3947$ nm) to a more cubic unit cell ($a_{\text{in-plane}} = 0.3880$ nm, $a_{\text{out-of-plane}} = 0.3897$ nm) as thickness increases from 25 to 450 nm. Therefore there is a reduction in the average strain as a function of increasing thickness which, in turn, gives rise to a reduction in magnetic anisotropy.

A careful study of these CMR islands shows that domain sizes at zero field, after saturation, scale as the square root of thickness but not with lateral dimensions. For example, MFM images of nominally 150 nm thick islands with diameters varying from 550 nm to 2 μm reveal negligibly little change in the average domain size. Figure 4 shows that nominally 600–700 nm diameter islands have domain sizes which exhibit a square root dependence on thickness. This trend reflects the basic scaling predicted by Kittel[15] for the magnetic interactions with film thickness as well as a reduction in the magnetic anisotropy with increasing film thickness, due to reduced average strain in thicker films.[16] The scaling of domain size on the square root of thickness is also evident in unpatterned LSMO films.

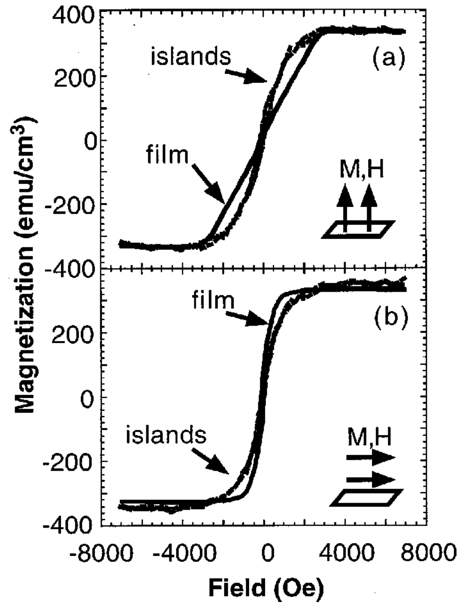

FIG. 5. Magnetization loops for an unpatterned film and submicron (90 nm high and 800 nm diameter) islands with field applied (a) perpendicular and (b) parallel to the substrate plane.

We also performed magnetization versus applied field measurements on a superconducting quantum interference device magnetometer to probe the macroscopic magnetic properties of the continuous films and island structures. Magnetization loops of unpatterned films with fields applied perpendicular and parallel to the film plane show remanence and are consistent with that of a weak stripe domain material (Fig. 5). In the weak stripe domain limit, $Q = K/2\pi M^2 < 1$ (where K is the magnetic anisotropy and $2\pi M^2$ is the maximum magnetostatic energy), relevant to our films ($Q \sim 0.2$); this condition amounts to the film thickness being greater than approximately the domain wall width [$= \pi(A/K)^{1/2} \sim 30$ nm where A is the exchange stiffness]. In magnetization loops measured parallel to the film/substrate plane, the field at which the magnetization reaches saturation (H_{sat}) provides a measure of the strain induced magnetic anisotropy in compressively strained LSMO [Fig. 5(a)]. In fact, the functional form of $H_{\text{sat}}(T)$ agrees well with strain anisotropy constants $K_\sigma(T) = 3\lambda\sigma/2$ (where λ = magnetostriction constant and σ = stress) deduced from independent experimental observation of magnetostriction[17] and moduli[18] for LSMO. In magnetization loops measured perpendicular to the film/substrate plane [Fig. 5(b)], the film saturates at a field less than the demagnetization field. This H_{sat} is a manifestation of energy gain associated with the erasure of domain walls.

Patterning has an effect on the magnetic response through shape effects and a modified strain state. When the magnetic field is applied perpendicular to the substrate plane, we observe that CMR islands do not exhibit a strict linear dependence of magnetization on applied field below H_{sat} as in the continuous film [Fig. 5(a)]. The deviation of the magnetization loops from linear behavior suggests that patterning does indeed facilitate the rotation of moments into the perpendicular direction. A detailed look at the loop reveals a small coercive field of 70 Oe that is consistent with domain wall pinning observed in the magnetic imaging. When the magnetic field is applied parallel to the substrate plane, H_{sat} of the islands is larger than that of the continuous film due to shape effects [Fig. 5(b)].

As the perpendicular magnetic-domain structure in CMR islands shows, compressive strain effects due to epitaxy on the underlying substrate are still dominant in these islands *at room temperature*. However the precise strain state of these islands is modified from that of an unpatterned film near the edges of the islands. In fact, strain is predicted to be relieved within a few island thicknesses from the edges of the islands and varies as a function of the distance from the film/substrate interface.[14] The slight reduction in the average strain from that of an unpatterned film cannot be detected in the magnetic images. However, the shape of the magnetization loops of the islands, in contrast to those of the unpatterned films, clearly show that shape anisotropy effects due to patterning are manifest although they do not overcome the biaxial strain effects that give rise to perpendicular magnetic domains.

While tunneling magnetoresistance (TMR) in CMR sandwich junctions has been observed near room temperature, it is still limited to temperatures well below the Curie temperature.[19] Our study of submicron CMR islands shows the complex multidomain state and its evolution as a function of magnetic field. With multidomain state electrodes, it is not surprising that the observed TMR is significantly less than that predicted from spin-polarized tunneling between two single domain electrodes. By increasing the thickness of the electrodes and decreasing the area of the junctions, the effects of the multidomain state can be minimized.

In summary, our results show the local magnetic structure of submicron CMR islands and their evolution as a function of applied magnetic field. At room temperature, a characteristic multidomain structure with perpendicular orientation, predicted by theory, is observed. The magnetization reversal of the islands in magnetic fields perpendicular and parallel to the substrate is dominated by strong domain wall pinning. Strong domain wall pinning in conjunction with geometrical confinement in these submicron CMR structures gives rise to reproducible multidomain structure.

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[1] X. Li, Yu Lu, G. Q. Gong, Gang Xiao, A. Gupta, P. Lecoeur, J. Z. Sun, Y. Y. Wang, and V. P. Dravid, J. Appl. Phys. **81**, 5509 (1997).

[2] Moon-Ho Jo, N. D. Mathur, J. E. Evetts, and M. G. Blamire, Appl. Phys. Lett. **77**, 3803 (2000).

[3] Qingyou Lu, Chun-Che Chen, and A. deLozanne, Science **276**, 2006 (1997).

[4] P. Lecoeur *et al.*, J. Appl. Phys. **81**, 5509 (1997); M. E. Hawley *et al.*, in *Epitaxial Oxide Thin Films II*, edited by D. K. Fork, J. S. Speck, T. Shiosoki, and R. M. Wolf, MRS Symposia Proceedings No. 401 (Materials Research Society, Pittsburgh, 1996), p. 531; C. Kwon *et al.*, J. Magn. Magn. Mater. **172**, 229 (1997).

[5] W. F. Brown, Jr., J. Appl. Phys. **39**, 993 (1968).

[6] H. Y. Hwang, T. T. M. Palstra, S.-W. Cheong, and B. Batlogg, Phys. Rev. B **52**, 15 046 (1995).

[7] M. R. Ibarra, P. A. Algarabel, C. Marquina, J. Basco, and J. Garcia, Phys. Rev. Lett. **75**, 3541 (1995).

[8] J. J. Neumeier, M. F. Hundley, J. D. Thompson, and R. H. Heffner, Phys. Rev. B **52**, R7006 (1995).

[9] Y. Suzuki, H. Y. Hwang, S-W. Cheong, and R. B. van Dover, Appl. Phys. Lett. **71**, 140 (1997).

[10] H. S. Wang and Qi Li, Appl. Phys. Lett. **73**, 2360 (1998).

[11] Y. Konishi, Z. Fang, M. Izumi, T. Manako, M. Kasai, H. Kuwahara, M. Kawasaki, K. Terakura, and Y. Tokura, J. Phys. Soc. Jpn. **68**, 3780 (1999).

[12] M. Hehn, K. Ounadjela, J.-P. Bucher, F. Rousseaux, D. Decanini, B. Bartenlian, and C. Chappert, Science **272**, 1782 (1996).

[13] J. H. Park *et al.*, Phys. Rev. Lett. **81**, 1953 (1998).

[14] I. A. Blech and A. A. Levi, J. Appl. Mech. **48**, 442 (1981).

[15] C. Kittel, Phys. Rev. **70**, 965 (1946).

[16] Yan Wu, Y. Suzuki, U. Rudiger, J. Yu, A. D. Kent, T. K. Nath, and C. B. Eom, Appl. Phys. Lett. **75**, 2295 (1999).

[17] H. Hazama *et al.*, Phys. Rev. B **62**, 15 012 (2000).

[18] R. V. Demin, L. I. Koroleva, and A. M. Balbashov, J. Magn. Magn. Mater. **177–181**, 871 (1998).

[19] See, for example, H. Q. Yin, J. S. Zhou, and J. B. Goodenough, Appl. Phys. Lett. **77**, 714 (2000).