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Engineering magnetic domains in manganite thin films by laser interference

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We report on the laser interference (LI) aided conversion from maze-like to stripe-like magnetic domains in $\text{La}_{1-x}\text{Sr}_x\text{MnO}_3$ ($x \sim 0.3$) thin films grown on LaAlO_3 substrates. This conversion is attributed to the periodic, local, and rapid heating by LI which facilitates the reconfiguration of magnetic domains without damaging the film structures. By annealing the sample, the stripe-like domains can be converted back to the maze-like state. Our result represents a non-magnetic scheme for reversible magnetic domain engineering in ferromagnetic thin films. © 2012 American Institute of Physics. [doi:10.1063/1.3673870]

Controlled manipulation of magnetic domains is of paramount importance, especially for magnetic data storage, which has attracted substantial attention from the scientific community during the past decades.^{1,2} Different states of magnetic domains often exhibit non-volatile characteristics, enabling the modern age magnetic information technology. To set the magnetic states, besides the conventional way of using an external magnetic field, a number of alternative schemes have been developed, such as spin transfer torque from spin polarized electrons,³ heat-assisted magnetic recording,⁴ opto-magnetic switching using circularly polarized light,⁵ patterning with ion irradiation,^{6,7} and localized hydrogen passivation.⁸ Another recent and promising addition to this array of magnetic manipulation techniques is the laser interference (LI), which has been demonstrated to create magnetic patterns, such as single-domain nanodots and nanowires, in a number of metal-based thin films and multilayers.^{9–13} The formation or change in magnetic patterns in these works were attributed to local changes in magnetic properties due to chemical intermixing at the interface and phase separation of magnetic clusters.^{9–13} However, the magnetic modification in these cases is often associated with periodic structural modification, causing the domain formation irreversible and, hence, incompatible with data storage operations.

Despite substantial efforts on magnetic patterning in metal films by LI, there has been no such report so far on films of magnetic oxides such as manganites. Manganites are a notable subclass of magnetic materials that present fascinating physics as well as tremendous potential for technological applications.¹⁴ The magnetic and transport properties of manganites can be drastically modified by the synthesis conditions and the external parameters.^{15–17} In particular, the domain patterns of $\text{La}_{0.7}\text{Sr}_{0.3}\text{MnO}_3$ (LSMO) films exhibit rich characteristics with strong dependence on film thickness,^{18,19} substrate induced strain,²⁰ and geometric patterns.^{21–23}

In this letter, we report on nonvolatile conversion of maze-like magnetic domain patterns in LSMO to stripe-like

domains by using LI. Unlike most of the other reports on LI induced domain patterning,^{9–13} the hereby described modification on domain characteristics is not associated with any periodic structural patterning. The stripe-like phase of magnetic domain can be easily reversed back to the maze-like state by annealing, thus enabling the erasure of the magnetic modification by LI.

LSMO films with a thickness of 325 nm were grown on (100) LaAlO_3 (LAO) substrates in a pulsed laser deposition system. The details of the film deposition and the room temperature magnetic domain imaging using magnetic force microscopy (MFM) have been reported elsewhere.¹⁹ Fig. 1(a) shows the schematic of the LI patterning setup. A low intensity pulse from YG 980 Q-switched Nd:YAG laser was split into two beams by an optical prism made of fused silica,

FIG. 1. (Color online) (a) Schematics of the laser interference setup (left) and the formation of stripe-like domains after laser irradiation (right). (b) and (c) show the MFM images of the same LSMO thin film before and after laser irradiation, respectively. The scan size of both images is $3.5 \mu\text{m} \times 3.5 \mu\text{m}$. Insets show the corresponding 2-D FFT patterns of the MFM images.

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which were then allowed to interfere on the sample surface placed right at the back of the prism. The laser operates at the third (355 nm) harmonic of the fundamental wavelength (1064 nm) and provides 18 mJ/pulse. The spot radius and the full width at half maximum (FWHM) of the pulse were 4 mm and 8 ns, respectively. The surface morphology of the samples before and after the laser irradiation was complementarily checked by atomic force microscopy (AFM) and scanning electron microscopy (SEM). The magnetization characteristics were measured at room temperature using a superconducting quantum interference device (SQUID).

Fig. 1(b) shows the maze-like magnetic domain patterns in the as-grown LSMO sample. The domain characteristics are influenced by the intricate interactions between the substrate and the film, which is determined by the complex competition between magnetostatic and exchange energies. The inset shows the corresponding fast Fourier transformed (FFT) pattern whose symmetric angular distribution suggests random directionality of the domains. Interestingly, the domains attain a clear directionality, resembling a stripe-like pattern, after the film is exposed to single pulse LI (the MFM image in Fig. 1(c) and the corresponding asymmetric FFT image therein). The domain size measured from the power spectral density (PSD) analysis of the FFT image reveals a substantial increase from 160 ± 8 nm for the as-grown sample to 240 ± 10 nm for the laser irradiated sample. The increased domain size is commensurate with the theoretical interference pattern periodicity, $p/2 = (\lambda/4\sin\theta) \approx 260$ nm, where λ is the wavelength of the laser and the recombination half angle $\theta \approx 20^\circ$. This suggests that the formation of the stripe-like domains is correlated to the local distribution of irradiation energy as a result of LI.

Due to the standing wave created by the LI, the absorbed energy is distributed inhomogeneously as crest and valley regions in the irradiated areas (Fig. 1(a)). Taking the transmittance of fused silica as $\sim 90\%$ and ignoring any power loss due to reflection and absorption by the substrate, we estimated the average heat absorbed by each crest region (with an approximate area of $1450 \mu\text{m}^2$) to be approximately $1.04 \mu\text{J}$. Then the corresponding temperature rise is $\Delta T = Q/V\rho c$, where Q is the total heat absorbed, V is the volume of one heated crest, $\rho = 6700 \text{ kg m}^{-3}$ is the density, and $c = 545.4 \text{ J Kg}^{-1} \text{ K}^{-1}$ is the specific heat capacity of LSMO.²⁴ Since the long-range thermalization during the nanosecond pulse is improbable, the effective thickness of the heated crest is much lower than the film thickness. Taking the thermal diffusivity of the LSMO film as $0.92 \text{ mm}^2/\text{s}$ (Ref. 24) and the pulse rise time as 1 ns, we deduced the effective thickness to be approximately 30 nm. From these parameters, we estimated a maximum local surface temperature rise of about 6500 K in the LSMO film. However, we note that this is merely a simple estimation of the surface temperature, and the actual spatio-temporal landscape can be more complex owing to the Gaussian shape of the energy distribution.²⁵

It has been reported previously that the circularly polarized light can generate a magnetic field strong enough to switch the magnetization of a $\text{Gd}_{22}\text{Fe}_{74.6}\text{Co}_{3.4}$ film near its Curie temperature (T_c).⁵ However, such a mechanism is valid only for the case where the sample temperature is close

to T_c , i.e., the magnetism still persists although very weak. As revealed by the temperature dependent magnetization data obtained from the SQUID measurements, the Curie temperature (T_c) of our LSMO samples is 370 K, and hence, the temperature rise in the crest regions as a result of LI is expected to locally destroy the ferromagnetic order. In other words, the spatiotemporal heating drives the LSMO film to the paramagnetic state for a short period of time. As the valley region receives much less energy than the crest area, during cooling, it is likely that the former will go below T_c and attain the ferromagnetism before the latter does. Thus, temporarily a periodic stripe-like structure with ferromagnet (FM)-paramagnet (PM) region is evolved, which basically follows the pattern of LI (Fig. 1(a)). A similar type of magnetic patterning has been reported previously where the periodic FM-PM regions were attained by selective N^+ ion bombardment on chemically ordered CrPt_3 films.²⁶ During the final phase of cooling to room temperature, the crest region also attains ferromagnetism, and to reduce the magnetostatic energy, its magnetization vector becomes aligned opposite to that of the valley region, creating a stripe-like domain structure in the LSMO film (Fig. 1(c)).

Unlike the other works on magnetic patterning by LI,^{9–13} the hereby described spatiotemporal heating does not leave any periodic structural fingerprint on the sample surface, which is presumably due to the lower irradiation energy used in our experiment. Nevertheless, the heating causes a significant smoothening of the sample surface, as shown in the AFM and SEM images in Fig. 2. The as-grown sample (Figs. 2(a) and 2(b)) shows a granular structure with the root mean square roughness (R_{rms}) and the maximum peak to valley roughness ($R_{\text{p-v}}$) of 3.9 nm and 19 nm, respectively. On the other hand, after the laser irradiation, the granularity of the sample diminishes (Figs. 2(c) and 2(d)), resulting in a drastically reduced R_{rms} and $R_{\text{p-v}}$ of 0.4 nm and 4 nm, respectively. Partial re-deposition of the laser ablated material is highly possible under the atmospheric pressure due to the interaction between ejected materials and the gaseous

FIG. 2. (Color online) (a) AFM and (b) SEM images of the sample before laser irradiation. (c) AFM and (d) SEM images of the laser irradiated sample. The scan size of all images is $1 \mu\text{m} \times 1 \mu\text{m}$.

molecules in air,²⁷ which may cause notable morphological modifications. Furthermore, we note that the geometric top areas of the granular islands usually receive more energies than the valley areas, so the laser ablation process effectively smoothens the surface. Besides the benefit of improved morphology, the increased smoothness reinforces the stability of the stripe-like magnetic domain as elucidated in Ref. 28, where it was demonstrated that the reduction in surface roughness assists the maze domains to attain directionality and eventually convert into the stripe-like morphology.

Our SQUID measurements confirm that the LI process affects the magnetic properties of the LSMO sample. Fig. 3(a) shows the M-H loop along $\langle 100 \rangle$ (perpendicular to the stripe direction) and $\langle 010 \rangle$ (parallel to the stripe direction) edges of the sample before and after exposure to the laser irradiation. It is clear that the laser irradiation does not affect the saturation magnetization, indicating minimal damage on the LSMO thin film. But the shape of the hysteresis loops changes, showing a larger magnetization at small field, which indicates that the magnetization vector is easier to be aligned by the external field in the LI-treated sample. Furthermore, the laser irradiation process reduces the coercivity of the LSMO film from ~ 30 to ~ 5 Oe. This change of magnetic properties may be related to the modification of the surface morphology, i.e., the smoother surface of the LI-treated sample causes little pinning of the domain walls (DWs), making the LSMO film appear “softer” magnetically. Furthermore, the laser irradiation may locally modify the oxygen content in the films, which can affect the magnetic properties of manganites^{29,30} and warrants future investigations.

Furthermore, the laser treatment effectively breaks the magnetic symmetry in the LSMO thin films. Fig. 3(b) reveals that for the untreated sample with maze-like domains, the M-H loops along $\langle 100 \rangle$ and $\langle 010 \rangle$ directions are almost identical, which is supported by the lack of directionality in the

FIG. 3. (Color online) (a) M-H characteristics for the as-grown and the laser irradiated samples. (b) Close-up views of the hysteresis loops.

FIG. 4. (Color online) (a) AFM, (b) SEM, and (c) MFM images of the laser irradiated sample after annealing at 750°C for 5 min. The stripe-like domains are reversed back to the maze-like state. The scan size for (a) and (b) is $1\ \mu\text{m} \times 1\ \mu\text{m}$ and for (c) is $2.7\ \mu\text{m} \times 2.7\ \mu\text{m}$.

domain pattern (Fig. 1(b)). On the other hand, after the LI treatment and the formation of stripe domain due to LI, the absolute magnetization along $\langle 100 \rangle$ is found to be slightly higher than that along $\langle 010 \rangle$. Such anisotropy could be a manifestation of the magnetization direction inside DWs separating two perpendicularly oriented and oppositely polarized domains. This is reasonable if the DWs in our samples is the Bloch type,³¹ where the magnetization at the center of the DWs is directed in plane and transverse to the stripe direction. This contributes an extra magnetization component to the $\langle 100 \rangle$ direction, giving rise to the observed in-plane magnetic anisotropy. For the maze-like case, such DWs are isotropically distributed, thus exerting no effect on the M-H results.

Interestingly, by annealing the LI irradiated samples at 750°C for 5 min, the stripe-like domains can be reversed back to the maze-like state, as shown in Fig. 4. This suggests that the LI induced stripe-like domains are metastable and lie closely to the maze-like state in the energy landscape. However, these maze-like domains in the annealed sample show a size distribution not as uniform as the as-grown samples. The PSD analysis of the FFT images (inset of Fig. 4(c)) indicates a nominal domain width of 225 nm, similar to that of the previous stripe-like domains. Furthermore, the annealing process brings back the granularity of the samples, as shown in Figs. 4(a) and 4(b). The roughness parameters, R_{rms} and $R_{\text{p-v}}$, increase to 2.8 nm and 10 nm, respectively. This gives further support to the correlation between the stability of the stripe-like domains and the surface smoothness, and the underlying mechanism warrants further investigation.

In summary, we show that by using single-pulse laser interference, the maze-like domains in manganite films can be modified to stripe-like domains within nanoseconds without leaving any permanent periodical structural pattern on the sample. The stripe-like domains are metastable and can be reversed back to the maze-like state by uniformly heating the film at high temperatures. Such a reversible approach of engineering the magnetic patterns is promising for non-volatile magnetic data storage technology, and further investigations are ongoing to shed more light on the underlying mechanisms.

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