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Modulation of ultrafast laser-induced magnetization precession in $BiFeO_3$-coated $La_{0.67}Sr_{0.33}MnO_3$ thin films

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The ultrafast laser-excited magnetization dynamics of ferromagnetic (FM) $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) thin films with $BiFeO_3$ (BFO) coating layers grown by laser molecular beam epitaxy are investigated using the optical pump-probe technique. Uniform magnetization precessions are observed in the films under an applied external magnetic field by measuring the time-resolved magneto-optical Kerr effect. The magnetization precession frequencies of the LSMO thin films with the BFO coating layers are lower than those of uncoated LSMO films, which is attributed to the suppression of the anisotropy field induced by the exchange interaction at the interface between the antiferromagnetic order of BFO and the FM order of LSMO.

magnetization precession, ultrafast time-resolved magneto-optical Kerr effect, perovskite oxide, interface

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1 Introduction

Ultrafast laser pulse excitation and dynamics measurement is utilized as an effective method to investigate the fast non-equilibrium magnetization in correlated spin systems [1-7]. At both fundamental and application levels, this issue has attracted attentions for understanding and achieving fast optical detection, as well as for the control of the magnetic order [8,9]. As one of the most intriguing magnetic relaxation phenomena, the optically excited magnetization precession in magnetic media—including metals, semiconductors, and oxides—has been extensively measured and discussed [10-15], providing a probably new path to fast magnetic switching, recording, and spintronic devices. In recent years, various interesting experiments focusing on the interface effect at artificially fabricated complex oxide heterointerfaces have been performed [16-19]. Among these, the $BiFeO_3$ (BFO)- and Sr-doped $LaMnO_3$ heterostructure combines the antiferromagnetic (AFM) and ferromagnetic (FM) orders and delivers a series of novel physical properties that originate from the FM-AFM exchange interaction across the interface and have potential future applications [20-23]. Although considerable efforts have been directed toward the exploration of the transient magnetic dynamics in manganese oxides [12,24-26], few works regarding the interface-effect modulation of the optically excited magnetization behavior of perovskite oxide heterostructures with the coupling of AFM and FM orders have been reported thus far. In this study, we investigated the ultrafast laser-induced

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transient magnetic dynamics behavior of optimally doped $La_{0.67}Sr_{0.33}MnO_3$ (LSMO) thin films with BFO coating layers. The time-resolved Kerr rotations and prominent uniform magnetization precessions triggered by femtosecond laser pulse excitation were evaluated. Compared with uncoated LSMO films, the precession frequencies of the BFO-coated LSMO thin films were reduced modulated by the BFO coating. According to analysis of the frequency dependence of the magnetic field, we ascribe this modulation of the magnetization precession to the anisotropy change caused by the interface effect of the BFO coating layers. We expect this to be an effective approach for manipulating the magnetic order in future spintronic devices.

2 Experiment

The BFO/LSMO heterostructures were fabricated using a laser molecular beam epitaxy system equipped with an in-situ reflection high-energy electron diffraction system. The 10-nm-thick LSMO thin films were deposited as bottom layers on (001) $SrTiO_3$ (STO) single crystal substrates. As top layers, 3- or 20-nm-thick BFO films were coated onto the LSMO films. The crystal structures of the samples were characterized by X-ray diffraction (XRD). The magnetization curves of the LSMO thin films were measured using a commercial Physical Properties Measurement System (PPMS, Quantum Design Inc.). The femtosecond laser pulse-induced magnetization dynamics were investigated using the pump-probe method. A Ti:Sapphire amplifier laser system providing 120-fs pulses at a repetition rate of 1 kHz was used to measure the time-resolved magneto-optical Kerr effect (TR-MOKE). The 800-nm fundamental outputs were used as the pump pulses to excite the magnetization dynamics with a fluence of 1 mJ/cm^2. The 400-nm pulses generated by frequency doubling the 800-nm pulses were used for the probe with a fluence less than 0.01 mJ/cm^2. The fluence ratio between the probe and pump beams was maintained below 0.01; thus, the probe pulse had a negligible effect on the magnetization dynamics of the samples. The samples were kept at 150 K in a superconducting split-pair magneto cryostat with optical windows. The external magnetic field was applied at an angle of 45° with respect to the film plane. The pump and probe beams were also applied at an incident angle of ~45°. The polarization rotation θ of the reflected beam was measured using the balanced detection scheme.

3 Results and discussion

XRD characterization was performed to confirm the crystal structure using standard θ-2θ scans. The XRD patterns shown in Figure 1 indicate fully epitaxial growth with distinct LSMO and BFO film peaks and no impurity phases.

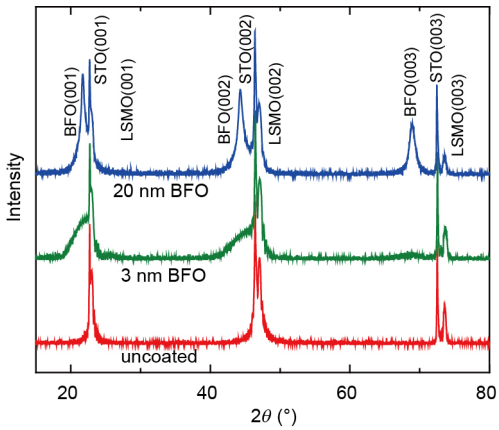

Figure 1 (Color online) XRD patterns of the uncoated LSMO thin films and the LSMO films coated with 3- and 20-nm-thick BFO layers, grown on STO substrates. The diffraction peaks are labeled for the three compounds.

We first focus on the temporal behavior observed after the photon excitation of a single sample. Figure 2(a) shows the time evolution of the Kerr rotation of the uncoated LSMO film. The TR-MOKE signal was monitored for ~500 ps under various applied external magnetic fields. In each case, a sharp decay of the magnetization is detected within the time scale of a few picoseconds after the pump pulse arrive. This is attributed to the optical "state filling" effect and the laser-induced demagnetization that occurred when the electrons were instantaneously heated by the pump pulse [10].

Subsequently, two distinct types of oscillations were launched. To investigate the oscillations, the Fourier transforms of the signals were calculated, as shown in Figure 2(b). The Fourier spectra indicate a high-frequency oscillation at ~103 GHz, which was independent of the external magnetic field. This oscillation mode was caused by the intensity change of the reflected probe beam for backward Raman scattering, which originated from the coherent acoustic phonons generated in the STO substrates by the pump-pulse irradiation [15,27,28].

The other oscillation mode occurred at a lower frequency (10-30 GHz), exhibiting a positive dependence on the external magnetic field, and is attributed to be the optically triggered precession of the magnetization, which was previously reported [8]. This precession behavior reflects the magnetic dynamics in the magnetic medium. In the equilibrium state (delay < 0 ps), the magnetization direction was along the effective field $H_{\rm eff}$, which was the sum of the applied external magnetic field $H_{\rm ex}$, the demagnetizing field $H_{\rm d}$, and the anisotropy field $H_{\rm a}$. Previous experiments involving LSMO thin films grown on STO substrates indicated an easy-plane anisotropy field due to the shape and strain effect [29]. Thus, in our case, by applying the external magnetic field, the magnetization M was canted out-of-plane along the balanced direction of the effective field $H_{\rm eff}$. When the pump pulse

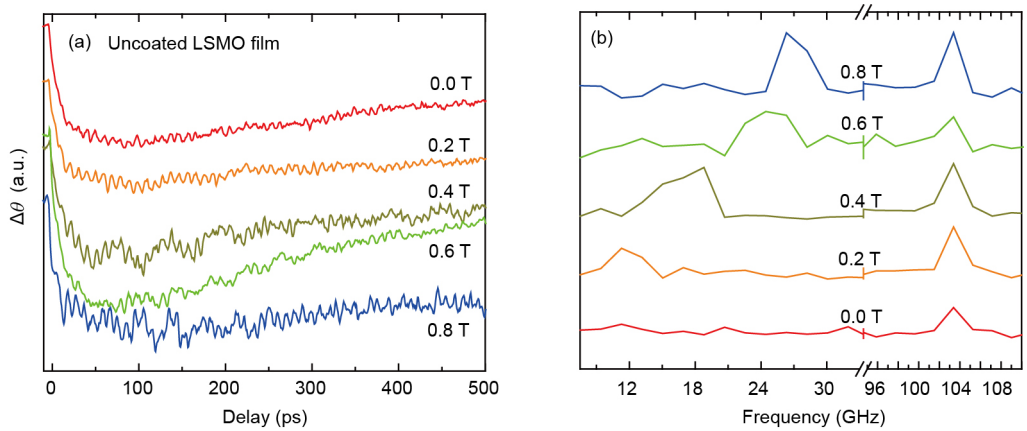

Figure 2 (Color online) (a) Transient Kerr rotation of the uncoated LSMO film under various external magnetic fields. (b) Fourier transform of the signals shown in (a), exhibiting the distinct peaks of two different oscillation modes.

instantaneously heated the LSMO layer (delay = 0 ps), the equilibrium magnetization was partly quenched by the sudden change of the demagnetization field and the anisotropy field, and subsequently rotated towards the new direction of an induced transient field H_{tr}. H_{tr} vanished after the film cooled down within a few tens of picoseconds, and the original effective field H_{eff} was restored, leaving the magnetization torque misaligned with the equilibrium direction. As a result, the magnetization started to precess around H_{eff}. In the TR-MOKE measurements, this phenomenon was monitored as an oscillation in the signal, which lasted a few hundred picoseconds. The dependence of the precession behavior on the external magnetic field is described by the Landau-Lifshitz equation. The precession frequencies increased with the external magnetic field, as shown in Figure 2(b), which is consistent with previous studies [8,25].

Using the same experimental setup, the TR-MOKE signals of LSMO films with 3- and 20-nm-thick BFO coating layers were measured, as for the uncoated film. The time evolutions of the Kerr rotation for ~500 ps are shown in Figures 3(a), (b), and (c) under applied external magnetic fields of 0.2, 0.4, and 0.6 T, respectively. A fast demagnetization response and two types of oscillations were observed, as for the uncoated LSMO film. Besides, none similar oscillation behavior depending on external magnetic field was observed in single BFO films grown on STO substrates under the same condition. Thus, it is reasonable to neglect the absorption of the laser beam by the BFO coating layers and consider that the BFO-coated LSMO films were sufficiently irradiated by the laser pulses.

We noticed the optically excited precession behavior of the different samples under the same external magnetic fields. The oscillation period of the precession appeared to be expanded for the BFO-coated LSMO films, and the sample coated with 20-nm-thick BFO exhibited a longer oscillation period than that coated with 3-nm-thick BFO in each case. The Fourier transforms show the precession frequency peaks, indicating distinct shifts of the peak position in each case for the same external magnetic field. For instance, when a 0.4 T external magnetic field was applied (Figure 3(d)), the peak positions of the precession frequencies were approximately 18.0, 16.2, and 14.3 GHz for the LSMO films with no BFO coating, the 3-nm-thick BFO coating, and the 20-nm-thick BFO coating, respectively. A similar trend was observed when the external fields were fixed at 0.2 or 0.6 T. The results are shown in Figures 3(d)-(f), clearly indicating the lower frequency of the BFO-coated LSMO films. We infer that the thicker BFO coating layers reduced the precession frequency of the LSMO films far more than the thinner coating layers did.

We performed a preliminary analysis of the precession behavior by solving the Landau-Lifshitz equation including the anisotropy field. We neglected the relaxation term for simplicity. Thus, the equation can be written as:

$$\frac{\mathrm{d}M}{\mathrm{d}t} = -\gamma M \times H_{\mathrm{eff}},$$

where H_{eff} is the effective field, and γ is the gyromagnetic ratio. As illustrated in Figure 4, the magnetization made an angle α with the film plane in the equilibrium condition, and θ is the angle between the applied magnetic field direction and the film plane. For thin films, the equation has the following solution:

$$\omega = \gamma\sqrt{\left(H_{\mathrm{ex}}\cos(\theta-\alpha)+H_{\mathrm{a}}\sin 2\alpha\right)\left(H_{\mathrm{ex}}\cos(\theta-\alpha)+H_{\mathrm{a}}\sin^2\alpha\right)}\,.$$

According to this solution, increasing the external field enhances the precession frequency, which agrees with our results for a single sample. We ascribe the reduction of the precession frequency for the BFO-coated LSMO films to the changes in the anisotropy field H_a. For FM LSMO thin films, the AFM order in the BFO coating layer should have a significant influence on the FM order through the exchange interaction at the interface; thus, the anisotropy field in the LSMO

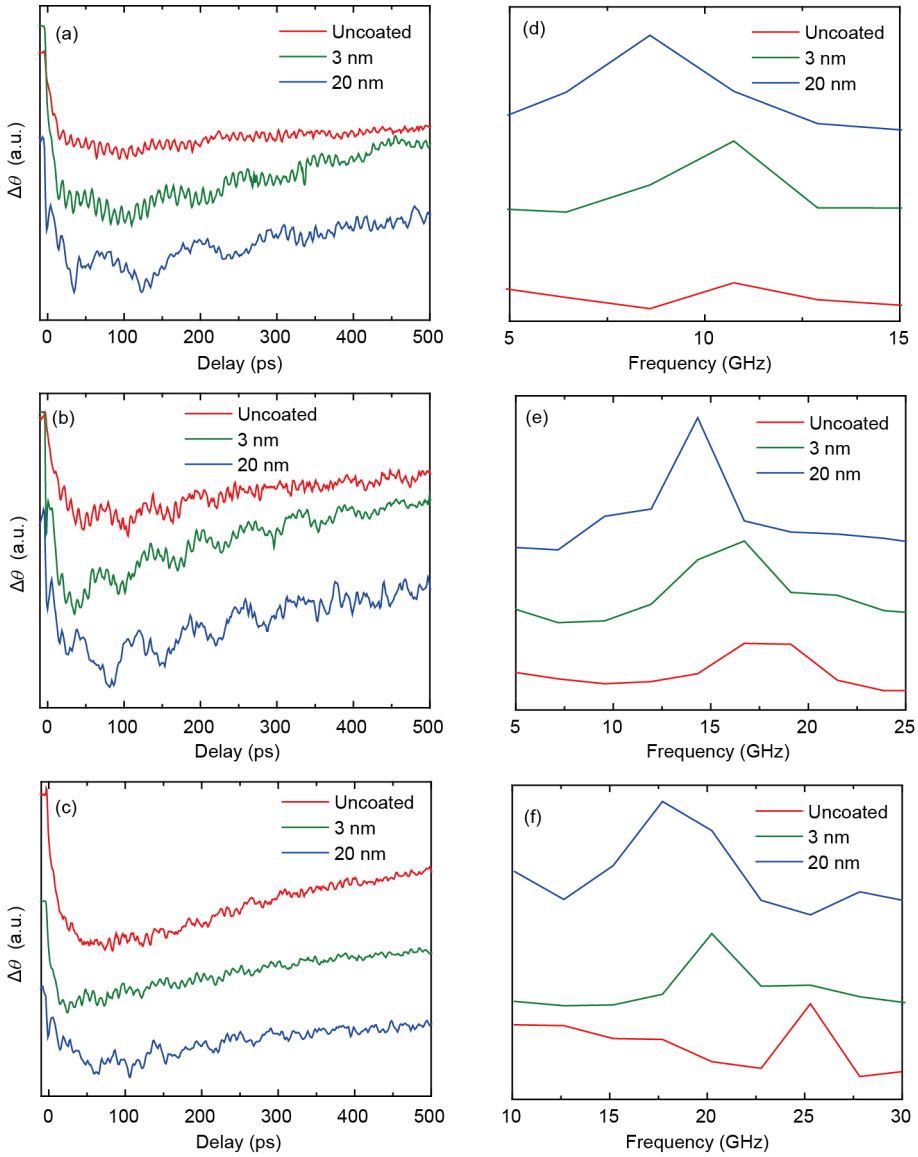

Figure 3 (Color online) Transient Kerr rotation signals of the uncoated LSMO film and the LSMO films coated with 3- and 20-nm-thick BFO layers under external magnetic fields of 0.2 T (a), 0.4 T (b), and 0.6 T (c). The Fourier transforms of the signals shown in (a), (b), and (c) for external magnetic fields of 0.2 T (d), 0.4 T (e), and 0.6 T (f), respectively.

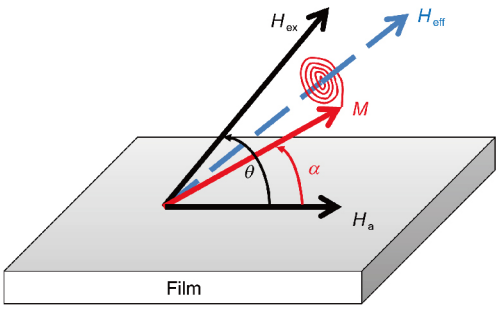

Figure 4 (Color online) Schematic of the magnetization precession.

can be changed effectively. To verify this speculation from a macroscopic perspective, the magnetization curve was measured for all the samples. Figure 5 shows the dependence of the magnetization—measured in the plane direction—on the external magnetic field. The sharper increase and larger area integral of the magnetization curve indicate that the magnetization occurred more easily in the measurement direction, indicating a larger anisotropy field H_a in that direction. The results shown in Figure 5 indicate that among the film samples, the uncoated LSMO films were most easily magnetized, followed by the samples with the 3-nm-thick BFO coating and then the samples with the 20-nm-thick BFO coating. This demonstrates that the anisotropy field was weakened as the thickness of the BFO coating layers on the LSMO films increased on the macroscale. Therefore, the suppressed anisotropy field for the BFO-coated LSMO films is a possible reason for the observed reduction of the magnetization precession frequencies. Obviously, this suppression originated

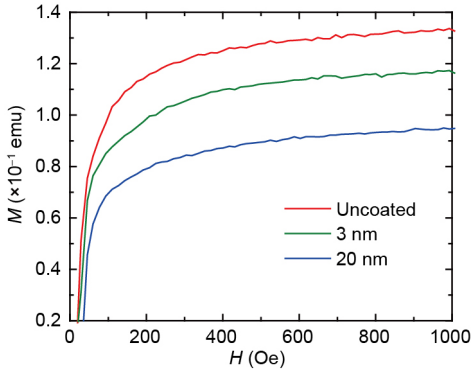

Figure 5 (Color online) Magnetization curve measured in the plane direction.

from the interaction across the interface of the heterostructures. As previously mentioned, the interface effect in BFO/LSMO heterostructures has been extensively investigated in previous studies focusing on intriguing phenomena such as the exchange bias, magnetoelectric coupling, and multiferroic field control [20-23], etc.

4 Conclusion

In conclusion, we studied the coherent uniform magnetization precession behavior of LSMO films epitaxially grown on STO substrates with BFO coating layers via TR-MOKE measurements. The BFO coating layer reduced the magnetization precession frequency, and a thicker BFO coating layer caused a larger reduction in the precession frequency. This allowed the modulation of the magnetization order in the artificially designed perovskite oxide structures. We attribute the reduction of the precession frequency to the suppression of the anisotropy field, which was induced by the exchange interaction between the AFM order of BFO and the FM order of LSMO. The intrinsic mechanism underlying this interesting phenomenon should be further studied and might need to be combined with many other means. Our findings may provide an effective approach for controlling the spin behavior in magnetic oxide films through structural design and shed light on the potential application in spintronic devices in the future.

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