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Su, Haibin; Chia, Elbert E. M.; La-o-vorakiat, C.; Tian, Y.; Wu, T.; Panagopoulos, C.; Zhu, J.-X.

2014

La-o-vorakiat, C., Tian, Y., Wu, T., Panagopoulos, C., Zhu, J.-X., Su, H., et al. (2014).
Interface-induced magnetic coupling in multiferroic/ferromagnetic bilayer: An ultrafast
pump-probe study. Applied Physics Letters, 104(14), 141602-.

<https://hdl.handle.net/10356/104094>

<https://doi.org/10.1063/1.4870580>

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Citation: *Applied Physics Letters* **104**, 141602 (2014); doi: 10.1063/1.4870580

View online: <http://dx.doi.org/10.1063/1.4870580>

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Interface-induced magnetic coupling in multiferroic/ferromagnetic bilayer: An ultrafast pump-probe study

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(Received 5 January 2014; accepted 25 March 2014; published online 7 April 2014)

By use of optical pump-probe measurement, we study the relaxation dynamics of a multiferroic-ferromagnetic TbMnO₃/La_{0.7}Sr_{0.3}MnO₃ bilayer. The relaxation dynamics of both layers are well separated in time allowing us to investigate the magnetic coupling across the bilayer. We observe that the relaxation dynamics of the individual layers in the bilayer sample are the result of the interplay between the intrinsic magnetic order and the induced interfacial effect. Our data suggest the existence of induced ferromagnetic order in the TbMnO₃ layer and antiferromagnetic order in the La_{0.7}Sr_{0.3}MnO₃ layer. © 2014 AIP Publishing LLC. [<http://dx.doi.org/10.1063/1.4870580>]

The ability to switch ferromagnetic magnetization by an electric field has attracted a lot of attention to multiferroic materials for many prospective applications in spintronics.¹ To make practical devices, a strong coupling between ferromagnetism and ferroelectricity is a prerequisite. However, good ferromagnetic-ferroelectric-type multiferroics are rare since ferromagnetism and ferroelectricity originate from different types of symmetries that are mostly mutually exclusive.² A current strategy to make devices from more abundant antiferromagnetic-type multiferroics is to introduce a thin layer of ferromagnet to indirectly mediate the magnetic coupling across the bilayer. With this idea, the control of exchange bias by electric field was demonstrated in a bilayer of multiferroic BiFeO₃ (BFO) and ferromagnetic La_{0.7}Sr_{0.3}MnO₃ (LSMO).³

In this work, rather than BFO, we focus on a bilayer consisting of TbMnO₃ (TMO) and LSMO. TMO is among the family of rare-earth orthorhombic manganites (RMnO₃), which are spin-frustrated materials.⁴ TMO itself exhibits a large magnetoelectric effect⁵ and is therefore a subject of many studies. It has a sinusoidal spin ordering below the Néel temperature (T_N) around 40 K due to the arrangement of Mn³⁺ ions resulting from spin frustration. Below the ferroelectric transition temperature ($T_{FE} \approx T_{lock}$) around 28 K, the Mn-spin ordering becomes spiral.⁶ This spiral ordering shifts the lattice arrangement, breaks the spatial-inversion symmetry, and subsequently induces the ferroelectric polarization.² Finally, Tb ions also exhibit another long-range spin-ordering transition around 7 K (T_{Tb}) but we will not focus on this Tb-ordering transition in this work.

From SQUID magnetometry measurements,⁷ the TMO/LSMO sample exhibits signatures of phase transitions at T_N . In addition, the sample shows a strong ferromagnetic transition at Curie temperature (T_C) near 165 K due to the existence of LSMO layer. The interfacial magnetic coupling

between TMO and LSMO is observed from the exchange bias below T_N and the enhancement of magnetic coercivity below T_C . However, due to the different natures of magnetic ordering of TMO (non-collinear spiral) and LSMO (collinear), the origin on the magnetic coupling is non-trivial and is still left unexplained.

To have a better understanding on this interlayer coupling mechanism, we employed ultrafast optical spectroscopy—a powerful technique to capture the relaxation dynamics of competing phases (e.g., superconductivity, antiferromagnetism) in correlated electron materials.^{8–10} We analyzed the relaxation times of the transient reflectivity ($\Delta R/R$) across the three transition temperatures (T_{lock} , T_N , and T_C) of the sample. The relaxation dynamics of both layers are well separated in time, allowing us to investigate the role of the individual layers in the magnetic interaction across the bilayer. Our data suggest that the magnetization of LSMO induces a magnetic order in TMO layer as the temperature is lowered below T_C . Moreover, below T_N , our data point to a surprising induced antiferromagnetic state on the LSMO side of the interface.

For all of the experiment conducted in this work, short pulses for our pump-probe setup¹¹ are provided by a Ti:sapphire regenerative amplifier system. The laser has a repetition rate of 250 kHz, central wavelength of 800 nm, and pulse duration of 70 fs at the sample position. The pump beam diameter is 50 μ m providing a pump fluence of 0.8 mJ/cm². The probe is focused to a smaller size of 25- μ m diameter to ensure a good pump-probe overlap. The probe power is attenuated 10 times weaker than the pump power. The polarizations of pump and probe are orthogonal to each other to improve the signal-to-noise ratio of the data. The sample is mounted on a continuous flow cryostat to vary the sample temperature from 10 to 250 K. Our sample is a thin film of epitaxial TMO/LSMO bilayer fabricated by a pulsed-laser deposition technique on a SrTiO₃ substrate.⁷ TMO has a thickness of 40 nm grown on top of a 3-nm-thick LSMO (8 unit cells).

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The transient $\Delta R/R$ curves of TMO/LSMO bilayer are measured as a function of temperature (Fig. 1). The dynamics are strongly dependent on the temperature but can be classified into four regions (as numbered in Fig. 1):

- (1) Initially, we observe a fast decrease (~ 300 fs) of the reflectivity signal. This fast feature is attributed to the photoexcitation of carriers from $d-d$ transition of Mn ions.¹²
- (2) Subsequently, the $\Delta R/R$ relaxes back toward the positive direction (reducing the magnitude of $\Delta R/R$), and crosses over to positive within a few picoseconds. This relaxation channel is attributed to electron-phonon thermalization.¹³ The zero crossing is a common characteristic of manganites.^{14,15}
- (3) At this point, the electron and phonon baths are almost in thermal equilibrium. However, the electron and phonon temperatures are still high compared to the temperature of the spin reservoirs. We then observe another relaxation contribution allowing the signal to once again cross over zero at low temperatures ($T < 100$ K). This intermediate timescale relaxation is commonly dedicated to the spin dynamics, and we will mainly focus on this relaxation channel in this work.
- (4) The final relaxation is due to heat transport occurring in nanosecond timescales.

To gain a fuller understanding of the data, we quantitatively analyze $\Delta R/R$ by use of exponential-model fitting function that contains five terms

$$\frac{\Delta R}{R} = -A_1 e^{-t/\tau_{ep1}} - A_2 e^{-t/\tau_{ep2}} + B_1 e^{-t/\tau_{s1}} + B_2 e^{-t/\tau_{s2}} - C e^{-t/\tau_D}, \quad (1)$$

where A 's, B 's, and C are positive numbers.^{13–16} All five exponential terms must be present to successfully fit $\Delta R/R$ across the whole temperature range by starting from the first

FIG. 1. (a) and (b) Temperature-dependent transient reflected intensity ($\Delta R/R$) of TMO/LSMO bilayer. Four general features are observed: (1) a fast decreasing spike due to photoexcitation, (2) a picosecond-scale phonon relaxation, (3) intermediate-timescale relaxation due to spin dynamics, and (4) a slow nanosecond-scale thermal-diffusion relaxation. Curve fitting results are illustrated as red dashed lines. The data are shifted for visibility.

negative peak. The first two exponential terms describe the fast negative decay (labelled as 2 in Fig. 1). The third and fourth terms fit to the intermediate relaxation dynamics (labelled as 3). And the final term is due to the slow ns-range dynamics (labelled 4). The fit results are illustrated in Fig. 1.

The first two exponential terms describe the fast negative decay due to electron-phonon thermalization; therefore, the negative signs are included. The fit results are relatively constant for $\tau_{ep1} = (0.25 \pm 0.02)$ ps and $\tau_{ep2} = (1.33 \pm 0.18)$ ps across the temperature range. The requirement of two exponential terms to describe the electron-phonon interactions suggests that hot electrons separately thermalize through two lattice channels from TMO and LSMO, which will subsequently initiate the spin dynamics of both layers.

Next, the intermediate relaxation time constants are observed in the range of 10–100 ps. The two timescales of both relaxation channels fall into separate ranges of 10–20 ps for τ_{s2} and 20–120 ps for τ_{s1} (Fig. 2). We ascribe the fast channel to the relaxation of TMO layers, which agree with the intermediate relaxation time of a TMO thin film (10–30 ps).¹⁶ In the same manner, the slower channel is dedicated to the relaxation through LSMO layer since the time-scale matches with the spin-lattice relaxation time of LSMO (about 100 ps) reported elsewhere.^{19,20}

Although the spin-relaxation channels are distinctive between the layer, the temperature-dependent relaxation times show a common behavior. Both τ_{s1} and τ_{s2} contain three anomalies near T_{lock} , T_N , and T_C of the sample (indicated by arrows in Fig. 2). The first two anomalies in both τ_{s1} and τ_{s2} are observed near the magnetic transition temperatures (T_{lock} and T_N) as in TMO thin film.¹⁶ The third anomaly is near magnetic transition temperature of LSMO (T_C) as it is a typical behavior of spin-lattice relaxation time of LSMO.^{14,19} The fact that the temperature-dependent signature of LSMO appears in the TMO relaxation channels, and vice versa, indicates that the spin dynamics of LSMO and TMO in our bilayer structure are interrelated.

To explain the origin of these anomalies, we generalize the model that has been used to describe the relaxation times of TMO¹⁶ and LSMO¹⁹ by incorporating three degrees of freedom: lattice (L), ferromagnetic (F), and antiferromagnetic (A) excitations. The coupled dynamics is initiated via the cooling down to the lattice temperature (T_L) after it has thermalized with the electrons. Then, the spin dynamics are activated by the heating up of ferromagnetic (T_F) and antiferromagnetic (T_A) excitations, and at the same time the spin baths are coupled together. With this three-reservoir picture in mind, the temperature dynamics can be written as the following:

$$\begin{aligned} C^A(T_A) \frac{dT_A}{dt} &= g_1(T_F - T_A) + g_2(T_L - T_A), \\ C^F(T_F) \frac{dT_F}{dt} &= g_1(T_F - T_A) + g_3(T_L - T_F), \\ C^L(T_L) \frac{dT_L}{dt} &= g_3(T_F - T_L) + g_2(T_A - T_L), \end{aligned} \quad (2)$$

where $g_1 = g_{A-F}$, $g_2 = g_{A-L}$, and $g_3 = g_{F-L}$ are phenomenological coupling constants, and C^A , C^F , and C^L are the specific heats of antiferromagnetic-spin, ferromagnetic-spin,

FIG. 2. (a) Relaxation times of the two intermediate time constants due to spin dynamics. The time constants are well separated in time and we ascribe τ_{s1} and τ_{s2} to LSMO and TMO, respectively. The anomalies at T_{lock} (28 K), T_N (40 K), and T_C (165 K) are visible as marked by the arrows. The curve fitting results of the relaxation times (Eq. (3)) are in solid lines. (b) The divergence of specific heats of TMO and LSMO near magnetic transition temperatures to describe the anomalies in τ_{s1} and τ_{s2} according to the model presented in the text. TMO specific heat data are extracted from Refs. 4 and 17; while the specific heat of LSMO is extracted from Ref. 18 and then scaled to the reported value of Ref. 19.

and lattice bath, respectively. To relate this model to our experimental relaxation times, the relaxation time from this model system can be solved analytically and written as

$$\frac{1}{\tau} = \frac{-1}{2} \left[\frac{g_1 + g_2}{C^A} + \frac{g_1 + g_3}{C^F} + \frac{g_2 + g_3}{C^L} \right] \pm \left(-4 \frac{(C^A + C^F + C^L)(g_1 g_2 + g_2 g_3 + g_3 g_1)}{C^A C^F C^L} + \left(\frac{g_1 + g_2}{C^A} + \frac{g_1 + g_3}{C^F} + \frac{g_2 + g_3}{C^L} \right)^2 \right)^{1/2} \right]. \quad (3)$$

First, we apply this equation to describe the relaxation time τ_{s2} (TMO channel). We extract the numbers of C_A and C_L from literature^{4,17} (Fig. 2(b)) but treat the induced C_F as a

constant fitting parameter along with the g 's. The electronic contribution is neglected. By performing a curve fitting to τ_{s2} , we obtain an excellent fitting (Fig. 2, blue line) that show two anomalies as a result from the peaks of C_A near T_N and T_{lock} of TMO (Fig. 2(b)). The fit yields $C_F = (0.045 \pm 0.003)$ J/(mol K) and $g_{A-F} = (9.2 \pm 0.6) \times 10^8$ W/(mol K). Interestingly, the other two coupling constants are negligible, $g_{A-L} = g_{F-L} \approx 0$, indicating that our model is reduced to two-temperature coupling between ferromagnetic and antiferromagnetic reservoirs with the relaxation time $1/\tau = g_{A-F}(1/C^A + 1/C^F)$, consistent with that previously proposed for the pump-probe study of TMO thin film.¹⁶ To achieve the best fit, we have to shift the temperature axis of TMO specific heat (T_A) up by 7.5 K; the temperature shift can be explained from the analysis of temperature dynamics where the lattice temperature is slightly higher than the antiferromagnetic-spin temperature during the timescale of τ_{s2} (≈ 40 ps).²¹ The fitted value of g_{A-F} is an order of magnitude smaller than the reported g_{A-F} of 2×10^{10} W/(mol K) of TMO thin film due to the assumption that $C_A = C_F = 1$ J/(mol K). In addition, the reported g_{A-F} was based on the 30 K data point only.¹⁶

To assign physical meaning to spin reservoirs, the antiferromagnetic reservoir (A) is ascribed to the *intrinsic* antiferromagnetic order in TMO layer; while the ferromagnetic reservoir (F) is proposed to be the *induced* magnetization on TMO due to the proximity effect to the LSMO layer. This interpretation is supported by a significant coercivity enhancement of TMO/LSMO at low temperature.⁷ This enhancement of coercivity was shown to be correlated to the induced local magnetic ordering at the interface on the antiferromagnetic side of exchange bias materials according to the previous theoretical model²² and x-ray magnetic circular dichroism (XMCD) studies.^{23,24} With this, we can attribute the third anomaly near T_C to the divergent behavior of C_F near Curie temperature resulting in a peak of the relaxation time described in Eq. (3).

Next, we shift our attention to the other relaxation time of LSMO (τ_{s1}). In this case, we are considering the coupling between the *intrinsic* ferromagnetic order and the *induced* antiferromagnetic order inside LSMO, which is supported by clear anomalies of τ_{s1} near antiferromagnetic transition temperatures T_N and T_{lock} (Fig. 2, red). By applying the calculated relaxation time from Eq. (3), we could explain the anomaly near T_C in a similar fashion. Since the Curie temperature of LSMO is strongly dependent on the sample condition and the specific heat measurement of LSMO beneath the heterostructure is non-trivial, we estimate the specific heat of LSMO film (Fig. 2(b)) from single crystal data by scaling both Curie temperature and the magnitude based on the available information.^{18,19} As before, we fit the relaxation time to $1/\tau$ (Fig. 2, red line). In this case of LSMO, we found that the lattice becomes increasingly important for the relaxation dynamics giving by the fit results, $g_{F-L}/g_{A-F} = 7.5 \pm 1.3$, while $g_{A-L} \approx 0$; therefore, the ferromagnetic-spin-lattice process dominates the relaxation of LSMO at the temperature near T_C . The observation agrees with the reported spin-lattice mechanism for the relaxation dynamics of LSMO.^{19,20}

Finally, we discuss the nature of the antiferromagnetic order that coexists in LSMO relaxation channel. Since the

formation of antiferromagnetic order inside a ferromagnet is quite surprising, we explain its origin as due to an interfacial effect in following analysis of the relaxation amplitudes B_1 and B_2 .

Since the optical reflectivity is related to the dielectric function, the amplitudes B_1 and B_2 represent the optical spectral weight. It follows from the optical sum rule that the spectral weight is related to the kinetic energy due to the virtual charge fluctuations and is in turn given by the super-exchange energy for a bond along a given polarization direction via the Hell-Feynman theorem.²⁵ Therefore, B_1 and B_2 are measures of the local exchange energy between the neighboring spins.¹⁶ In the case of the photon polarization being in the plane parallel to the interface, the local exchange energy is given by $\mathcal{K}_E = J_{\parallel} \langle \vec{S}_i \cdot \vec{S}_j \rangle$, where $J_{\parallel}$ is an effective exchange interaction while the symbol $\langle \dots \rangle$ denotes the quantum mechanical average. We note that for a generic inhomogeneous system, the quantity $\mathcal{K}_E$ should be understood as a spatial average. As temperature decreases, $\mathcal{K}_E$ increases resulting in a corresponding increase of B_1 and B_2 (Fig. 3).

We first examine B_2 , the amplitude of TMO relaxation (Fig. 3(a)). We see that B_2 starts to increase as temperature decreases below 90 K—this trend matches the rise in coercivity of TMO/LSMO (squares, taken from Ref. 7). Since coercivity is a measure of the induced local magnetic orders

FIG. 3. (a) The amplitude B_2 (circles) from the TMO channel shows the gradual rising trend starting from below T_C at 165 K as the ferromagnetic order is induced in TMO layer. The interpretation is supported by the enhancement of coercivity data of TMO/LSMO bilayer (squares) overlaid on top of the data points.⁷ (b) The amplitude B_1 of the LSMO channel increases gradually from T_C and becomes relatively constant until 50 K (the green dashed line as a guide to the eye). However, it shows discontinuities at low temperatures especially below T_{lock} .

as mentioned earlier, the temperature dependence of B_2 thus supports our assumption that the relaxation channel is related to the induced ferromagnetic magnetization in the TMO layer, via proximity effect with LSMO (Fig. 4(a)).

We next examine the relaxation amplitude B_1 due to the LSMO relaxation channel (Fig. 3(b)). B_1 exhibits a slight dip at T_C , then increases gradually with decreasing temperature (green dashed line in Fig. 3(b)). Below 40 K, B_1 increases sharply near T_N (40 K) and T_{lock} (28 K). This suggests a sudden change in the local exchange energy in the LSMO layer as the temperature crosses below T_N and T_{lock} . We attribute this sharp increase in B_1 , and the anomalies in the relaxation time τ_{s1} , to the interfacial antiferromagnetic order in LSMO (Fig. 4(b)). As previous studies have shown, the interfacial Mn spins from the LSMO side are prone to be antiferromagnetically aligned as an A-type (planar) antiferromagnetic ordering by the interface effect, while the rest of Mn planes are still ferromagnetically aligned.^{26–29} This interfacial antiferromagnetic state has been previously observed from the XMCD study of BFO/LSMO bilayer,²⁶ where the orbital reconstruction causes the interfacial Mn on the LSMO side to antiferromagnetically couple to the second Mn layer via super-exchange mechanism. A similar result has been reported from the first-principles calculation of an interface between a ferromagnetic $\text{La}_{1-x}\text{A}_x\text{MnO}_3$ ($\text{A} = \text{Ca}, \text{Sr}, \text{Ba}$) and multiferroic BaTiO_3 ²⁷—the electrical polarization of BaTiO_3 causes an antiferromagnetic coupling of the first two layers of Mn. Our data are consistent with these scenarios. A future XMCD study is one possible approach to confirm our proposed spin configurations on both TMO and LSMO sides.

In conclusion, our pump-probe data suggest magnetic interlayer coupling between multiferroic manganite TMO and ferromagnetic LSMO in a heterostructure bilayer. From the analysis of relaxation times, we speculated an induced ferromagnetism in TMO by its proximity to LSMO and also an induced interfacial antiferromagnetic state at the LSMO side of the bilayer. The two induced states are important during the relaxation dynamics since they couple to the intrinsic (anti-) ferromagnetic magnetic orders of (TMO) LSMO. Our current work points to the possibility of using TMO/LSMO bilayer for the next generation of spintronic devices, by taking advantage of multiferroic properties of TMO, half

FIG. 4. A possible spin configuration of Mn ions inferred from our pump-probe data at the temperature (a) between T_N and T_C and (b) below T_{lock} . Below T_C , a ferromagnetic order (FM) is induced into the TMO layer due to the proximity effect (blue), while below T_{lock} , an interface effect induces an interfacial antiferromagnetic (AFM) order inside LSMO (red). The dashed arrows on the right side of the figure illustrate the induced ferromagnetism.

metallicity, and colossal magnetoresistance of LSMO, and the magnetic coupling between them to control the magnetism with electric field.

We acknowledge the support from Singapore NRF CRP (NRF-CRP4-2008-04) and MOE AcRF Tier 1 (RG 13/12) and the LANL LDRD Program. Part of this work was supported by the Center for Integrated Nanotechnologies, a U.S. DOE user facility.

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