Ultrafast optical manipulation of interfacial magnetoelectric coupling

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Abstract: We demonstrate a new paradigm for all-optical detection and control of interfacial magnetoelectric coupling on ultrafast timescales, achieved by using time-resolved second harmonic generation (SHG) in a ferroelectric/ferromagnet oxide heterostructure.

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Magnetoelectric multiferroics have attracted much attention due to their potential applications in areas including sensing and data storage [1]. However, the goal of developing bulk multiferroics with strong magnetoelectric (ME) coupling has been elusive, prompting researchers to explore artificial multiferroic composites [2], in which the ME coupling can be engineered through the interface geometry and constituent materials [3]. The most well known approach uses strain to indirectly enhance the ME coupling in ferroelectric (FE)/ferromagnet (FM) heterostructures [1,2], by using a magnetic field to modify the spin alignment in the FM layer, changing the lattice constant through magnetostriction. This in turn modifies the strain on the FE layer and, through the piezoelectric effect, the FE polarization P, [4] significantly enhancing the ME coupling [1].

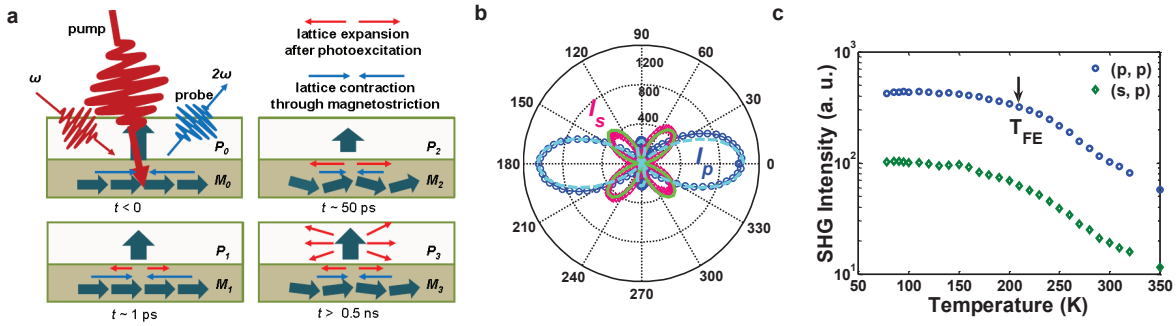

Fig. 1. (a) Idea of optically-induced interfacial ME coupling. The piezoelectric response of BSTO results from interfacial strain relaxation after photoexcitation of ferromagnetic LCMO. (b) Polar plot of the measured p and s polarized SHG intensity at 10 K. The cyan dashed and green solid lines are numerical fits to I_p and I_s. (c) Static temperature-dependent SHG for (p,p) and (s,p) polarization combinations. In LCMO, T_c ~240 K, while in BSTO, T_{FE} ~215 K [6], as indicated by the arrow.

Despite these impressive advances, an important aspect of multiferroics has received little attention: namely, their dynamic properties [5], which will impact many of their potential applications. Here, we use femtosecond laser pulses to optically induce interfacial ME coupling in a $Ba_{0.1}Sr_{0.9}TiO_3$ (BSTO)/$La_{0.7}Ca_{0.3}MnO_3$ (LCMO) heterostructure and selectively probe the piezoelectric response using SHG (proportional to P). In our experiments, the pump pulses photoexcite non-equilibrium quasiparticles in FM LCMO, which rapidly interact with phonons before undergoing spin-lattice (s-l) relaxation on a timescale of tens of picoseconds (ps). This relaxes the lattice contraction resulting from spin-spin interactions in LCMO, inducing a change in the lateral strain imposed on FE BSTO through magnetostriction. This in turn leads to a transverse ME effect that occurs much faster than photoinduced heat diffusion from absorbing LCMO to transparent BSTO. The concept underlying this optically-induced ME effect is sketched in Fig. 1(a).

The sample used here is a 50 nm thick BSTO film grown on a 50 nm thick LCMO film, using an MgO substrate [6]. Our optical experiments are based on an amplified Ti:sapphire laser system, in which pump pulses at 1.59 eV are used to excite quasiparticles in LCMO and 1.59 eV probe pulses are used for SHG from compressively strained BSTO at 3.18 eV (Fig 1(b)). We note that the static SHG intensity decreases upon heating BSTO (Fig. 1(c)).

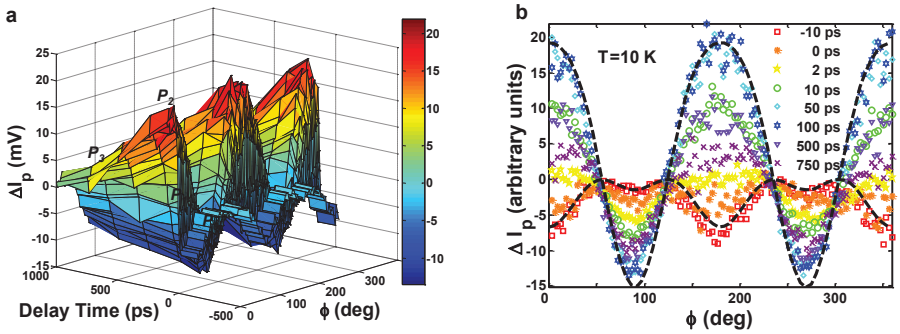

Fig. 2. (a) Time-resolved *p*-polarized SHG signal as a function of the incident light polarization. P_0-P_3 are piezoelectric responses to the demagnetization (arising from M_0-M_3 in Fig. 1(a)). (b) Polarization-dependent changes in I_p measured at 10 K for various time delays.

After photoexciting LCMO, we measured the polarization-dependent time-resolved SHG (TRSHG) signal at low temperatures (Fig. 2); we note that standard optical pump-probe experiments revealed no polarization-dependent time-resolved change in the reflectivity at either 1.59 or 3.18 eV, ensuring that photoinduced changes in the TRSHG signal are not due to changes in the optical constants. At early times ($\sim$ 1 ps), photoexcited carriers in LCMO relax through electron-phonon coupling, increasing the lattice temperature and initiating lattice expansion in LCMO, which in turn relaxes the interfacial compressive strain in BSTO. This modifies P, causing a small change in the TRSHG signal within $\sim$ 7 ps, given by the timescale for the strain relaxation to propagate through the BSTO film.

A much larger polarization-dependent change in the TRSHG signal occurs on a relatively slow ($\sim$50-100 ps) timescale.This is only observed below the critical temperature for FM order (T_c) in LCMO, indicating that this signal has a non-thermal and magnetic origin. In fact, the observed timescale matches well with that for s-l relaxation in LCMO, in which demagnetization reduces the lattice contraction originating from spin-spin interactions. This further relaxes the interfacial compressive strain in BSTO, initiating a stronger piezoelectric response that modulates P and thus the SHG signal. Finally, at long timescales, the remaining changes in P are due to thermal diffusion from LCMO to BSTO (which reduces the SHG intensity through heating BSTO). We note that identical experiments were performed on separate 50 nm films of BSTO (non-absorbing with a large static SHG signal) and LCMO (absorbing without a significant static SHG signal), and no TRSHG signal was observed. This further verifies that the observed TRSHG signal is due to optically-induced ME coupling at the BSTO/LCMO interface.

In conclusion, we demonstrate ultrafast all-optical interfacial ME coupling in a multiferroic heterostructure by using TRSHG to optically perturb FM order in LCMO and selectively probe the resulting changes in the FE properties of BSTO. This presents intriguing possibilities for future high speed optically controlled magnetoelectric devices.

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