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Terahertz Excitation of Chiral Phonons Probed via the Faraday Effect

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***Abstract*—By combining two variably delayed THz pulses with vertical and horizontal polarizations, we create one combined THz pump pulse with circular polarization. This circular polarized THz pulse is used to excite chiral phonon motion in non-magnetic $LiNbO_3$, which in turn induces a transient magnetic moment that we probe via the ultrafast Faraday effect.**

I. Introduction

CIRCULAR motion of ions in solid materials has been posited as a means to dynamically induce or control magnetic properties, even in non-magnetic materials [1, 2]. This circular motion fundamentally requires the excitation of “chiral” phonon modes, and the ability to excite and control transient crystal chirality could have a significant impact on condensed matter physics.

The idea of dynamical multiferroicity was suggested by Juraschek and co-workers [2], where pairs of phonon modes in ferroelectric materials could be excited to create circular or elliptical motion of ions. These ion loops with right or left handedness, depending on the charge of the ions and magnitude of their motions, can induce a magnetic moment, even in originally non-magnetic materials. Therefore, exciting these ion loops in ferroelectric solids to dynamically induce magnetic ordering, will make them transiently multiferroic [2, 3].

We have recently had much success in controlling the waveform and polarization of high-field, broadband terahertz (THz) pulses [4, 5]. This waveform control is driven by the capability to perform two-dimensional (2D) THz spectroscopy [4, 6], including the ability to create circularly polarized THz pump pulses [5].

In this presentation, we report how we generated intense pulses of circularly polarized THz light to excite pairs of degenerate phonon modes. As the phonon modes are excited with proper relative phase (provided by the circularly polarized THz pulses), circular phonon motion results. The transient magnetic moment induced by these excited ions loops is then measured with the ultrafast Faraday effect.

II. Results

To generate circularly polarized light, we use our 2D THz experimental setup as shown in Figure 1a. An optical parametric amplifier (OPA) is driven by the output of a Ti:Sapphire laser system. The signal (1450 nm) and idler (1785 nm) outputs of the OPA are divided and each pumps a separate DAST organic THz generation crystal. The output THz pulses from each crystal (vertical polarization from the signal pump and horizontal from the idler pump) are brought traveling parallel to each other and as close together as possible. A three-mirror imaging system focuses both THz pulses with a tight focus to the same spot at the sample position, where the fields add together to create a single THz pulse.

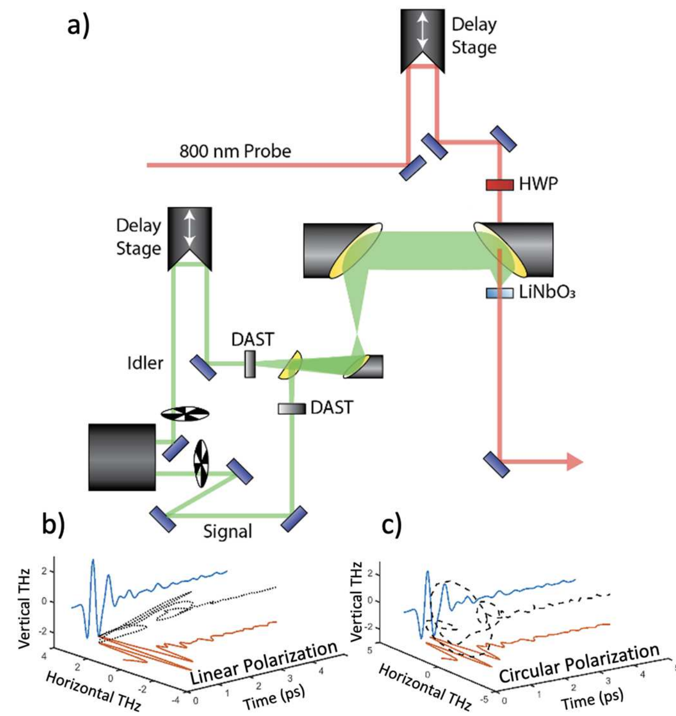

Fig. 1. a) 2D THz experimental setup. The two generated THz pulses are focused onto a $LiNbO_3$ crystal. The relative time delay of the two THz pulses can be changed by with the delay stage in the idler path to create b) linearly polarized THz pulses or c) circularly polarized THz pulses.

The relative delay of the two THz pulses is controlled by a motorized delay stage in the idler beam path. As depicted in Figure 1b, if both THz pulses arrive without any relative delay, the resulting combined THz pulse is linearly polarized. If one pulse is delayed by a quarter period with respect to the other, circularly polarized THz pulses result, as shown in Figure 1c. We therefore can easily control the ellipticity angle of the combined THz pulse. A differential chopping scheme also enables us to readily measure the sample response from a single (vertically polarized) THz pulse, the other individual (horizontally polarized) THz pulse, and the combined THz pulse with controllable polarization state, all in a succession of four laser shots; the probe response in the absence of a THz pump pulse is measured with the fourth laser shot.

To probe the sample response, we direct an 800-nm probe pulse through the excited region of the sample and measure the modification of the probe polarization with a transient birefringence detection scheme. This allows us to measure the transient Faraday effect to detect the creation of an ultrafast magnetic moment. However, it can also measure other ultrafast birefringence signals, like the THz Kerr effect. Fortunately, by controlling the relative polarization angles between THz and probe pulses, we can isolate the Faraday signal from the Kerr signal [7].

With circularly polarized THz pulses, we excite a z-cut sample of $LiNbO_3$. The lowest frequency E symmetry phonon modes are doubly-degenerate with perpendicular mode effect charge vectors that can be driven with THz pulses polarized along the crystallographic x and y directions. Exciting pairs of E-modes with circularly polarized THz pump pulses induces ion-loop motion, leading to a transient magnetic moment in $LiNbO_3$. Figure 2a models the motion of the pair of E modes, with the overall chiral trajectory in black. While the chiral phonon motion takes place, a transient magnetic moment is induced that can be detected with the Faraday rotation of a probe pulse. The measured Faraday signal is expected to follow the envelope of the chiral phonon motion.

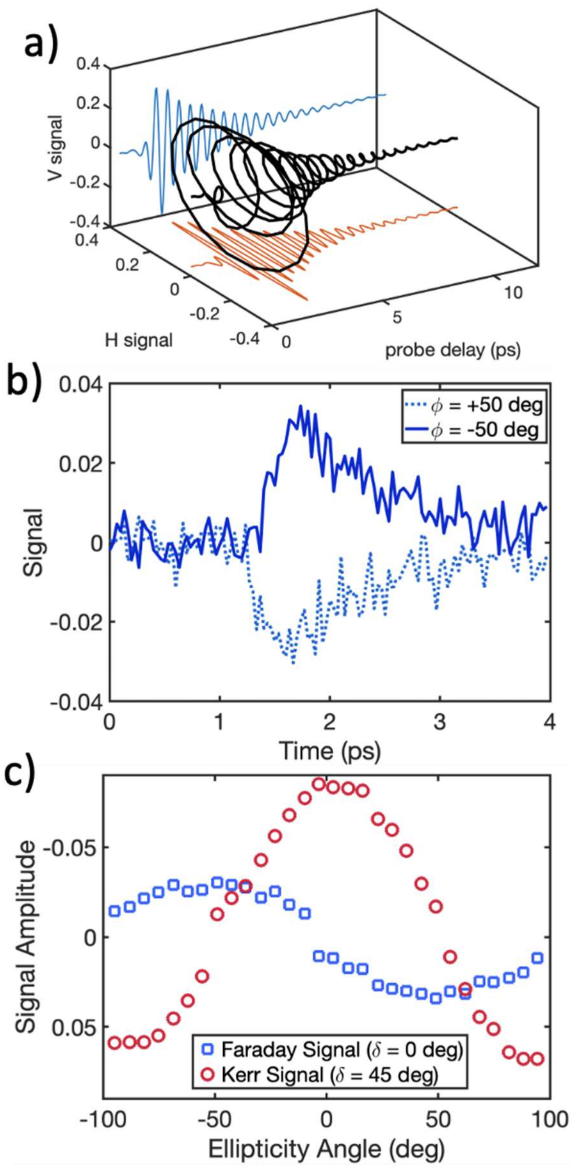

Fig. 2. a) Modeled motion of a pair of orthogonal E phonon modes in $LiNbO_3$. The black line shows the overall chiral phonon motion. b) Ultrafast Faraday effect signal due to chiral phonon motion in $LiNbO_3$, for THz pump ellipticity angles of + and - 50 degrees. c) Measuring the transient signal as a function of THz ellipticity angle, for different relative probe polarizations, allows isolation of Kerr and Faraday signals.

Figure 2b shows the time domain Faraday effect signal measured in $LiNbO_3$ for THz pump pulses of +50 and -50 degree ellipticity angles. We see a transient signal persisting for ~3 ps, which is longer than the THz pump pulse duration (<1 ps), confirming that this isn't just the THz inverse Faraday effect. The magnetic moment signal also switches sign as the THz polarization and subsequent chiral phonon motion changes handedness.

As mentioned above, our transient birefringence probe is sensitive to Faraday effect and Kerr rotation signals. However, we can isolate the Faraday signal from the Kerr signal by controlling the polarization angle of the 800-nm probe beam. Figure 2c shows how the magnitude of the transient signal changes with THz ellipticity angle for the case when the probe is sensitive to Kerr signal (red circles) and for when the signal is sensitive to the Faraday effect (blue squares), further confirming that we excited chiral phonon motion leads to transient magnetic ordering in non-magnetic $LiNbO_3$.

III. Summary

Using a 2D THz experimental setup, we can create high-field circularly polarized THz pulses. These circularly polarized THz pulses excite chiral phonon motion in $LiNbO_3$, transiently inducing a magnetic moment that persists longer than the THz excitation pulses. Isolating the Faraday signal from the Kerr signal confirms that the excited chiral phonon motion, so-called ion loops, induces magnetic ordering in non-magnetic $LiNbO_3$. The ability to use circularly polarized THz pulses to control chiral phonon motion will provide many new avenues in the study of condensed matter systems.

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