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# Single-pulse terahertz coherent control of spin resonance in a canted antiferromagnet

Zuanming Jin[a,b], Zoltán Mics[a], Guohong Ma[b], Zhenxiang Cheng[c], Mischa Bonn[a], and Dmitry Turchinovich[a,d]
[a]Max Planck Institute for Polymer Research, Mainz, 55128, Germany
[b]Department of Physics, Shanghai University, Shanghai, 200444, China
[c]University of Wollongong, North Wollongong, 2500, Australia
[d]Technical University of Denmark, Lyngby, 2800, Denmark

***Abstract*—We report on the coherent control of terahertz (THz) spin waves in a canted antiferromagnet, $YFeO_3$, associated with a quasi-ferromagnetic spin resonance at a frequency of 0.3 THz, using a single THz pulse. The intrinsic dielectric anisotropy of $YFeO_3$ in the THz range allows for coherent control of both amplitude and phase of the excited spin wave.**

## I. Introduction And Background

ULTRAFAST control of spins in solids has been receiving increasing attention because of its potential applications in spintronics and spin-based information processing. The direct, magnetic field - induced, ultrafast excitation and control of spin waves is most advantageous for these purposes. However, due to the lack of efficient ultrafast sources of direct magnetic excitation, the ultrafast magnetization dynamics in magnetically-ordered materials was until recently initiated and controlled indirectly, using various optically-activated mechanisms. For example, the inverse Faraday effect based on nonlinear optical process, dynamic momentum-space distribution of photoexcited spin-polarized carriers, thermal magnetic anisotropy change, and lattice distortion have been employed to excite the spin systems on femtosecond and picosecond timescales. In most of these cases, the spin excitation by ultrashort laser illumination of materials is accompanied by an undesirable thermal contribution, usually associated with electron dynamics which mediate the excitation of spin waves in the laser-induced process [1]. Ideally, the ultrafast excitation and manipulation of spin dynamics in future spintronic systems should be heat-free, i.e. not leading to any thermal load on the device.

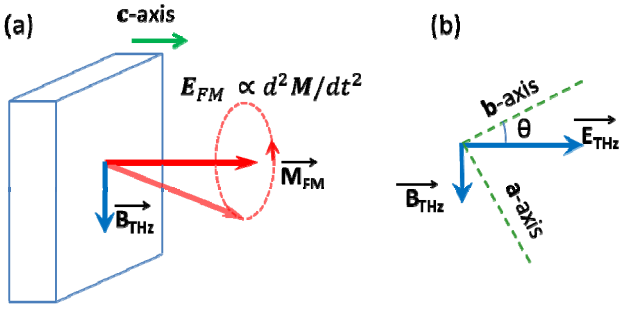


Fig.1 (a) Mechanism of THz emission by spin resonance in c-cut $YFeO_3$, and (b) orientation of the THz pulse polarization with respect to optical axes of the crystal.

The THz electromagnetic pulse can excite a spin wave in the antiferromagnetic material via an impulsive Zeeman torque, induced by the magnetic-field component of the THz pulse, as shown in Fig. 1 (a) [2-3]. The subsequent precession of the magnetization vector results in a spin wave, which in turn emits THz radiation at the spin resonance frequency, via a free-induction decay mechanism. Two optically active spin resonance modes, quasi-ferromagnetic mode (FM mode) and quasi-antiferromagnetic mode (AFM mode), can be excited in $YFeO_3$ via Zeeman torque $\mathbf{T} \propto \mathbf{M} \times \mathbf{B}_{THz}$, where $\mathbf{M}$ is the macroscopic magnetization, and $\mathbf{B}_{THz}$ is magnetic field component of a THz pulse. This radiation can be detected, and thus the spin waves can be characterized using THz spectroscopy. The excitation of either modes is governed by the experimental geometry and the cut of the crystal. In this work, only the FM resonance is excited.

## II. Results

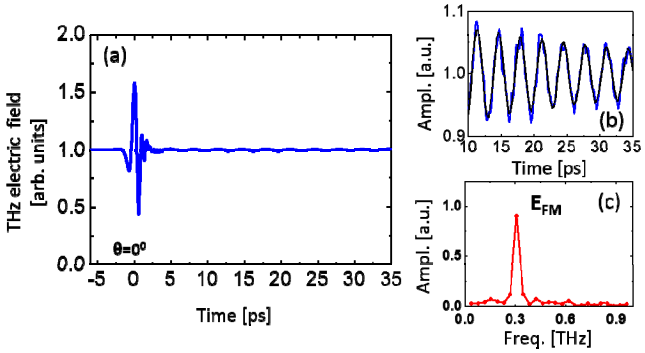


Fig. 2 (a) THz waveform transmitted through a 1.33 mm thick $YFeO_3$ crystal at azimuthal angle θ = 0°. (b) The oscillating part of the THz electric field together with the fitting result, and (c) its Fourier transform spectrum ($E_{FM}$).

As shown in Fig. 2 (a), the main peak of the THz pulse transmitted through the $YFeO_3$ crystal at azimuthal angle θ = 0° with respect to b-axis of the crystal is followed by the long lived and damped oscillation component at 0.3 THz, which is assigned to the FM mode and is highlighted in Fig. 2 (b). Fig. 2 (c) shows the Fourier transform of the oscillation components, $E_{FM}$, from Fig. 2 (b) in the time range from 10 ps to 35 ps.

First, we performed the coherent control of this spin resonance in a traditional fashion, using the sequence of two THz-pulses[2,3]. First THz pulse excites a spin wave, and the time-delayed second THz pulse excites its phase-shifted replica. Two spin waves, excited by these pulses, interfere constructively or destructively. $\Delta\varphi = \omega \cdot \Delta t$ is the phase difference between these two spin oscillations, where ω is the angular frequency of the spin resonance, and Δt is the time delay between the two pulses. As shown in Fig. 3, when the

second THz-pulse is arrived at the same time as the first one ($\Delta t$=0 ps), the combined Zeeman torque of both THz pulses enhances the spin oscillation, as compared to the excitation by only a single pulse. However, when the second pulse is timed at $\Delta t$=1.66 ps, yielding a phase difference $\Delta\varphi = \pi$, the THz signal produced by the FID of the FM spin resonance is significantly reduced. The incomplete constructive and destructive interference of the partial spin waves in the case of this two-pulse coherent control experiment is explained by slightly different amplitudes of the two THz pulses in the sequence.

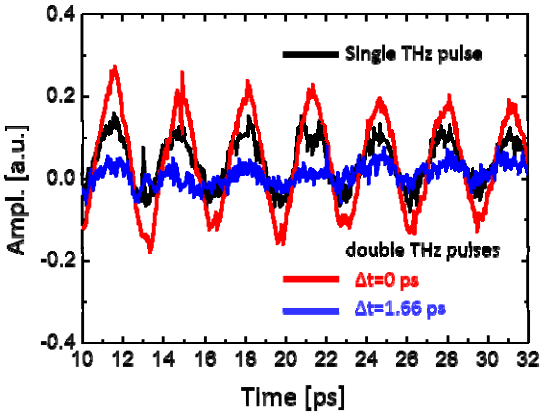


Fig. 3 The spin precession with single- and double-THz pulses excitations. Inserting the second THz control pulse at a specific time, the amplitude of FID signal enlarges ($\Delta t$=0 ps) or stops ($\Delta t$=1.66 ps).

Now we demonstrate that the same effect of coherent control of the spin resonance via excitation of two phase-shifted spin waves can be achieved with *only a single THz pulse*, using the naturally occurring THz dielectric anisotropy of $YFeO_3$ crystal. This anisotropy results from the high-frequency phonon modes in $YFeO_3$. In this fashion, using only a single incident THz pulse, we are able to control both the amplitude and the phase of the resulting spin wave [5].

The THz-range birefringence of our sample along a- and b-crystal axes $\Delta n = n_a - n_b$ is found to be about $\Delta n = 0.2$ over the whole spectral range of our experiment. Owing to the crystal birefringence, the single incident THz pulse, if launched in the polarization state at non-zero angle to either of the crystal axes, excites *two* phase-shifted spin waves associated with the two orthogonal components of THz field experiencing anisotropic propagation. The amplitudes of these two spin waves are dictated by the field strengths of the two orthogonal components of the THz field, which can be controlled by the polarization of the incident THz pulse relative to the crystal axes. The angular dependence of the amplitude of the THz field detected in horizontal poilarization is

$$\left|\mathbf{E}^{horiz}\right| = \left|\mathbf{E}^{in}_{THz}\right| e^{\left(-\frac{1}{2}\alpha_b d\right)} \left[\cos^2(\theta)\, e^{i\Delta\Gamma} e^{-\Delta\alpha d} + \sin^2(\theta)\right]$$

where $\Delta\alpha = \alpha_a - \alpha_b$ and $\Delta\Gamma = \Gamma_a - \Gamma_b = \frac{\omega d}{c}\Delta n$ are the absorption coefficient difference and the phase retardation between two orthogonal polarization components of THz pulses, and hence between the excited spin waves ($d$ is the crystal thickness). This equation is thus a coherent control function, using the azimuthal angle $\theta$ (see Fig. 1(b)) as control parameter [5].

In Fig. 4, the symbols show the dependency of the measured amplitude ($E_{FM}$) of the FID signal of the FM-mode spin wave on the azimuthal angle θ, and the solid line shows the analytical coherent control function. Excellent agreement between the model and the measured data is observed. In the same figure we show the separate contributions from the refractive index and the absorption coefficient anisotropies to the coherent control function. As the dashed and dotted lines, respectively, we show this function including only refractive index anisotropy (assuming $\alpha_a$=$\alpha_b$=0), and only absorption anisotropy (assuming $n_a$=$n_b$) of the $YFeO_3$ crystal. Both index and absorption contributions play a significant role in the observed coherent control effect. Hence, the coherent control of the resulting spin resonance is readily achieved by control of the relative amplitudes of the partial THz waves, by rotating the crystal with respect to the polarization of the incident single-THz pulse.

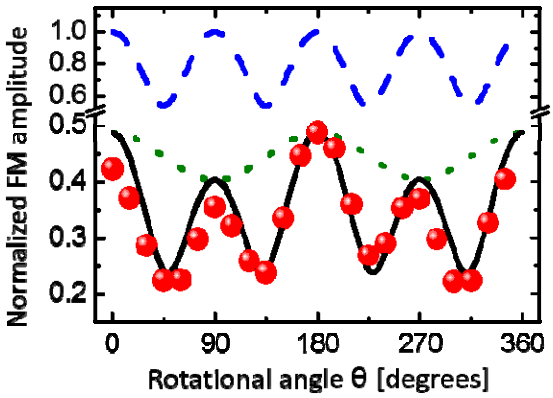


Fig. 4 Amplitude of $E_{FM}$ as a function of azimuthal angle θ (symbols). Model including full dielectric anisotropy at spin resonance (solid line), only index anisotropy (dashed line), and only absorption anisotropy (dotted line) are also shown.

In summary, we have demonstrated the coherent control of a quasi-ferromagnetic resonance at 0.30 THz in a canted antiferromagnet $YFeO_3$ using only a single THz pulse. The coherent control effect is mediated by the dielectric anisotropy of the $YFeO_3$ crystal. Taking into account both refractive index and absorption coefficient anisotropy of the crystal, a coherent control function is developed, using the THz pulse polarization with respect to the crystal axes as a control parameter. In our 1.33 mm – thick $YFeO_3$ crystal the THz spin wave amplitude could be modulated by a factor of two, and the phase could be shifted by about -3π/2 simply by choosing the polarization of an incident THz pulse with respect to the crystal axes. The suggested method can thus further expand the range of capabilities in ultrafast THz manipulation of magnetization by adding one more degree of freedom, *polarization*, to existing coherent control schemes.

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