Excitation and control of spin waves in $FeBO_3$ by a strong-field THz pulse

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Abstract: The dynamically resolved response of the canted-antiferromagnet $FeBO_3$ excited near a magnon resonance shows fast oscillations after THz-excitation, followed by the magnons' intrinsic relaxation, enabling to probe transient magnetic relaxation dynamics over large frequency range.

Antiferromagnetic materials are preponderant in nature among the magnetically ordered materials, but extensive study started only recently [1]. The development of strong-field THz sources empowered the direct excitation of the magnon resonances [2,3] and the control of magnetic excitations. $FeBO_3$ is studied since the 1970s for two interesting properties: it is a canted antiferromagnet with a ferromagnetic moment at room temperature and is transparent at optical frequencies. The magnon frequency was determined in [4] to be located between 25 GHz and 64 GHz depending on the sample orientation, without external magnetic field and at room temperature. However, to the best of our knowledge, there is no measurement reported in the literature of dynamically resolved measurements of the relaxation following the excitation close to the magnon frequency. In this work, we report on the non-dissipative excitation and control of the magnons using a strong-field THz pump.

In the experiment layout in Fig. 1, we use 7.5 mJ of a commercial Ti:Sapphire laser operating at 1 kHz to generate strong-field THz pulses via the pulse-front tilt technique [5]. The single-cycle 30 µJ THz pulses, centered at 0.38 THz (Fig. 1(b)), are focused on a sample with two off-axis parabola to 1.4 mm x 1.9 mm spot diameter, corresponding to an electric peak field of 51 MV/m and a magnetic peak field of 0.35 T. The electric field of the THz pulses was measured by electro-optic sampling in ZnTe; the corresponding spectrum extends up to 1.5 THz. The sample is a thin $FeBO_3$ crystal (50 µm at the periphery - 200 µm in the center) placed in a DC magnetic field (B_{DC} on Fig. 1(a)) perpendicular to the magnetic field of the THz pulses (B_{THz} on Fig. 1(a)). A small fraction of the 800 nm-centered, 150 fs-long pulses from the Ti:Sapphire laser is used to probe collinearly through the sample the Faraday rotation (Polarization detection on Fig. 1(a)). The linear polarization of the probe pulses is collinear to the polarization of the pump pulses. To increase the signal to noise ratio, a chopper modulates the repetition rate of the pump beam to 500 Hz; the output of the balanced detector is sent to a locking amplifier for acquisition. The ranges used for the scans varied from 70 ps with 20 fs step size to resolve fast oscillations up to 1 ns with 1 ps step size for resolving the slow ones.

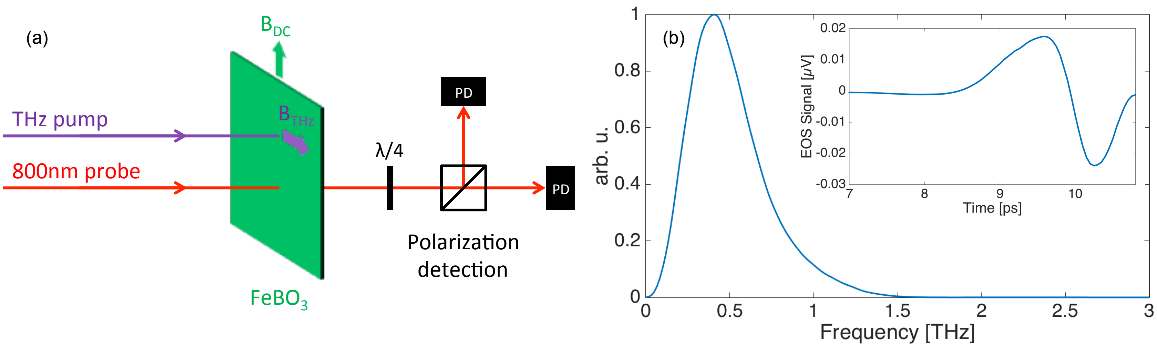

Fig. 1. (a) Experimental setup of the dynamically resolved terahertz spectroscopy of $FeBO_3$: a quarter waveplate, Wollaston prism and two balanced photodiodes are used to measure the polarization rotation of the probe induced by the strong THz pulses. PD: photodiode. (b) Spectrum of the THz excitation pulses with the electro-optical sampling trace in inset.

We first characterized the transmission of the $FeBO_3$ samples in a time-domain spectrometer for THz. To calculate the absorption and index of refraction, we assumed a thickness of 200 µm, corresponding to the middle of the sample, to account for the Gaussian spatial distribution of the THz beam. This measurement gives an index of refraction of ~3 from 0.3 THz to 2 GHz and a corresponding absorption between 50 cm-1 and 150 cm-1. The calculated Fresnel losses are then 25% on each interface, and a THz pulse twice reflected on both interfaces is delayed by 4 ps.

Figure 2 shows the dynamics of the polarization rotation, with dependency on the strength of the magnetic field of the excitation pulses (a,b) and on the DC magnetic field (c-f). We observe fast oscillations in the first 30 ps after the excitation, which seem to follow the electric field of the strong THz pulse, then slow oscillations lasting during ~500 ps, corresponding to the relaxation of the magnons. Fig. 2(a) shows the variation of the response of the magnons with decreasing THz magnetic field. A minimum magnetic field of the THz pulses of 160 mT is required to detect them in our setup.

Fig. 2(c) and (e) shows the measured dependency of the magnon frequency with the static magnetic field and Fig. 2(d) and (f) the calculated Fourier transforms: the magnon frequency shifts from 40 GHz (*i.e.* period of 25 ps) for 82 mT to 64 GHz (*i.e.* period of 16 ps) for 185 mT. The resonance frequency of the ferromagnetic resonance is located on the edge of the spectrum of the excitation pulse.

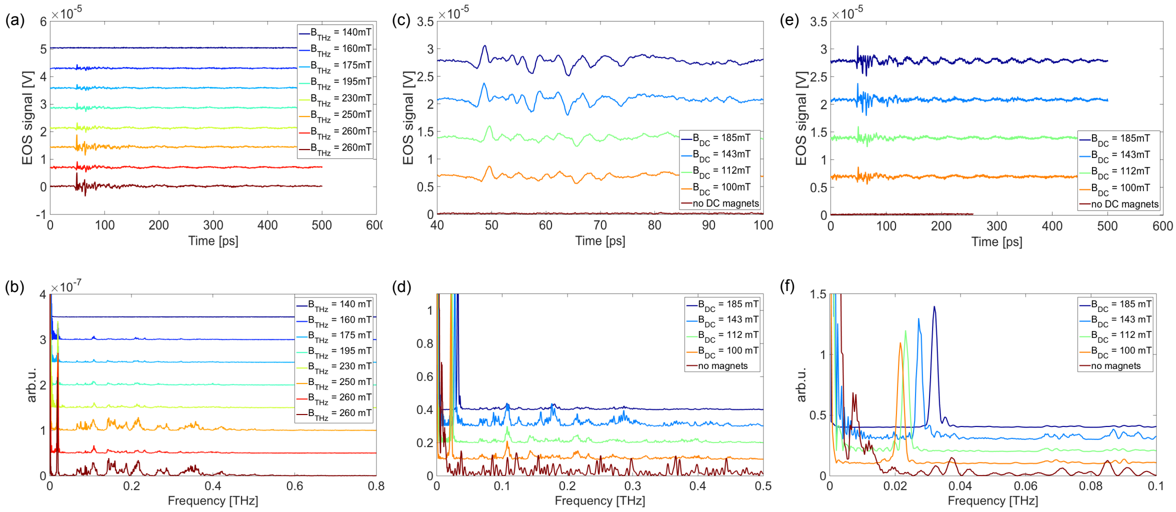

Fig. 2. Oscillations measured after excitation by a strong THz pulse in the FeBO3 sample (upper row) and their Fourier transform (lower row). The input THz magnetic field (a,b) and the magnitude of the DC magnetic field (d-f) were varied. The scans shown here were performed with 500 ps range and 100 fs resolution. (a), (b): the external DC magnetic field was fixed to 82 mT. (c-f) the magnetic field of the THz pulses was setup to 260 mT. (e) shows the relaxation of the magnons over the full range of the scans of (c) (zoomed in on the fast oscillations). (d) and (f) corresponding Fourier transforms of the signals centered on the fast oscillations overlapping in frequency with the excitation spectrum and on the magnon response, respectively. EOS: electro-optical sampling.

In this work, we report the dynamically resolved magnetic response of the canted antiferromagnet FeBO3, excited with a few-cycle THz pulse. At early times after the excitation, we observe fast oscillations close to the frequency of the exciting pulse, followed by slow oscillations at the frequency of the intrinsic ferromagnetic resonance of the crystal. Our studies open new avenues to probe the transient magnetic relaxation dynamics in magnetic materials over a wide range of frequencies.

7. References

[1] A. V. Kimel, A. Kirilyuk, A. Tsvetkov, R. V. Pisarev, and T. Rasing, "Laser-induced ultrafast spin reorientation in the antiferromagnet $TmFeO_3$," Nature **429**, 850 (2004).
[2] T. Kampfrath, A. Sell, G. Klatt, A. Pashkin, S. Mährlein, T. Dekorsy, M. Wolf, M. Fiebig, A. Leitensdorfer, and R. Huber, "Coherent terahertz control of antiferromagnetic spin waves," Nat. Phot. **5**, 31 (2011).
[3] S. Baierl, M. Hohenleutner, T. Kampfrath, A. K. Zvezdin, A. V. Kimel, R. Huper, and R. V. Mikhaylovskiy, "Nonlinear spin control by terahertz-driven anisotropy fields," Nat. Phot. **10**, 715 (2016).
[4] W. Jantz, J. R. Sandercock, and W. Wettling, "Investigation of FeBO3 by Brillouin scattering from thermal magnons", AIP Conference Proceedings 1976 29:1, 268-269
[5] X. Wu, S. Carbajo, K. Ravi, F. Ahr, G. Cirmi, Y. Zhou, O. D. Mücke, and F. X. Kärtner, "Terahertz generation in lithium niobate driven by Ti:sapphire laser pulses and its limitations," Opt. Lett. **39**, 5403 (2014)