Terahertz Optomagnetism: Nonlinear THz Excitation of GHz Spin Waves in Antiferromagnetic FeBO$_3$

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A nearly single cycle intense terahertz (THz) pulse with peak electric and magnetic fields of 0.5 MV/cm and 0.16 T, respectively, excites both modes of spin resonances in the weak antiferromagnet FeBO$_3$. The high frequency quasi-antiferromagnetic mode is excited resonantly and its amplitude scales linearly with the strength of the THz magnetic field, whereas the low frequency quasi-ferromagnetic mode is excited via a nonlinear mechanism that scales quadratically with the strength of the THz electric field and can be regarded as a THz inverse Cotton-Mouton effect. THz optomagnetism is shown to be more energy efficient than similar effects reported previously for the near-infrared spectral range.

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The development of high-intensity ultrashort laser pulses opened up new pathways to easily access the nonlinear optical regime, where strong light-matter coupling can lead to a plethora of fundamentally new opportunities for light frequency conversion [1–4] and controlling properties of media by light [5–7]. In magnetism these developments led to the establishment of the new research field of ultrafast magnetism [8–10] and to discussions about the potential of femtosecond optical pulses for the fastest and most energy efficient writing of magnetic data [11].

Pioneering studies of nonlinear optical phenomena in magnetics [2,12] pointed out that light can act as an effective magnetic field [13,14] that efficiently aligns spins and induces a magnetization in media [14]. Regarding symmetry, a cross product of the two orthogonal electric field components of circularly polarized light has the same symmetry as a magnetic field aligned along the wave vector of light. Also, linearly polarized light can induce magnetic anisotropy in magnetized media. Phenomenologically, these effects were described in terms of the optomagnetic inverse Faraday and Cotton-Mouton effects. Combining optomagnetism with femtosecond laser pulses yields the fastest ever and therefore unique stimulus in magnetism. Recently, it was argued that far-infrared pulses are the most energy efficient means to control magnetism [15,16]. Despite many experimental and theoretical studies in the visible, near- [17–19], and midinfrared [20], strong coherent nonlinear coupling of spins to THz light was reported exclusively for TmFeO$_3$ [21]. The mechanism of the coupling relied on resonant pumping of Tm$^{3+}$ ions and was only efficient in a narrow 10 K temperature range in the vicinity of a spin reorientation phase transition. Although the inverse Faraday and Cotton-Mouton effects are much more universal—they do not rely on absorption and must be observed in a broad temperature range—optomagnetic phenomena induced by THz light have not been reported yet and their efficiency is still an open question.

Here we report a nonlinear optomagnetic coupling of THz light to spins in a material with no strong spectral features in the THz spectral range and no phase transition. The mechanism appears to be efficient in a broad temperature range. In particular, using a single crystal of the weak ferromagnet FeBO$_3$ we demonstrate that a nearly single cycle THz pulse excites a GHz spin resonance in this antiferromagnet and the observed dependencies on the polarization of the THz pulse, temperature, and external magnetic field indicate that the mechanism can be assigned to the inverse Cotton-Mouton effect [22–24].

FeBO$_3$ is a material with a low absorption coefficient $\sim$80 cm$^{-1}$ [25] in the visible and $\sim$160 cm$^{-1}$ in the THz spectral range. The latter was measured by means of time resolved THz spectroscopy (Fig. S1 [26]). FeBO$_3$ crystallizes in a calcite-type structure with point group $\bar{3}m$. The schematic of the crystal structure is depicted in Fig. 1(a). The spins of the Fe$^{3+}$ ions form two magnetic sublattices with magnetizations $M_1$ and $M_2$, respectively. The sublattices are
corresponding Fourier spectrum (inset). (d) Waveform of the THz electric field and the field. The crystal tilt and the orientation of the magnetic field are propagation through the tilted crystal as a function of magnetic field applied by linearly polarized light at the wavelength of 800 nm upon the nonlinear mechanism, if the optical susceptibility should be able to control spins in an antiferromagnet via a (001) plane is able to control spins in the FeBO₃ crystal. The ellipticity originates from different refraction coefficients experienced by two mutually orthogonal, linearly polarized light waves propagating in the direction of the detector. The difference is proportional to \( \mathbf{LL} \) and the effect is called the magneto-optical Cotton Mouton effect or magnetic linear birefringent (MLB). The field was applied in the (001) plane [along the \( z \) axis in Fig. 1(a)] and the probing beam propagated along the [001] crystallographic axis [\( z \) axis in Fig. 1(a)]. The observed ellipticity is even with respect to the applied magnetic field and must be assigned to the dependence of \( \chi^{(a)}_{kl} \) on \( \mathbf{LL} \). Figure 1(c) shows the polarization rotation measured in a similar geometry but for light propagating at a small angle of about 10° with respect to the \( z \) axis. The measured field dependence of the polarization rotation is odd with respect to the applied magnetic field, known as the direct magneto-optical Faraday effect that probes \( \chi^{(a)}_{kl} \). These static and earlier dynamic [41] experiments clearly show that for light at the wavelength of 800 nm the dependence of \( \chi^{(a)}_{kl} \) and \( \chi^{(s)}_{kl} \) on the magnetic order parameters results in significant magneto-optical and optomagnetic effects. In general, in the near-infrared and visible spectral ranges, it is expected that the Faraday effect, as the first term in a Taylor series, is stronger than the Cotton-Mouton effect. However, taking into account peculiarities of the electronic structure of matter, in a narrow spectral range the ratio between these two effects can be different [42]. Practically nothing is known about optomagnetic effects in the THz domain. Nonetheless, from a quantum mechanical theory of magneto-optical phenomena developed for the case of rare-earth ions [31], one can anticipate that in dielectric media with no free electrons \( \chi^{(a)}_{kl} \) and \( \chi^{(s)}_{kl} \) have rather different spectral dependencies at low frequencies of light \( \omega \): \( \lim_{\omega \to 0} \chi^{(a)}_{kl}(\omega) = 0 \) and \( \lim_{\omega \to 0} \chi^{(s)}_{kl}(\omega) \neq 0 \).
Although similar quantum mechanical calculations of the optical susceptibility for FeBO$_3$ are beyond the scope of this Letter, it is clear that, unlike previous experiments with near-infrared and visible light, in the THz spectral range the efficiency of the inverse Cotton-Mouton effect can be higher than that of the inverse Faraday effect.

To study the effect of THz light on spins in FeBO$_3$, we employed intense THz pulses generated by 100 fs optical pulses from a Ti:sapphire amplifier with an energy of about 4 mJ per pulse. The dynamics was mapped using a probe pulse at the wavelength of 800 nm. The generated THz waveform and corresponding spectrum are depicted in Fig. 1(d). Details of the THz generation technique and the experimental setup are provided in the Supplemental Material [26]. If the THz pulse triggers spin dynamics, it can be translated in the dynamics of the magnetization. FeBO$_3$ has two modes of antiferromagnetic spin resonance: a low frequency quasiferromagnetic mode (q-FM) and a high frequency quasiantiferromagnetic mode (q-AFM) [43,44]. In spherical coordinates, where $\theta$ is the angle between $\mathbf{L}$ and the $z$ axis, and $\psi$ is the angle between the $x$ axis and projection of $\mathbf{L}$ on the $xy$ plane, one finds $\mathbf{L} = 2|\mathbf{M}| \sin \theta \cos \psi \mathbf{e}_x + \sin \theta \sin \psi \mathbf{e}_y - \cos \theta \mathbf{e}_z$. It means that q-FM and q-AFM modes are seen as oscillations of $\psi$ and $\theta$, respectively.

The THz-induced spin dynamics was probed using the magneto-optical Faraday and Cotton-Mouton effects by measuring the polarization rotation of the linearly polarized probe pulse. A typical time trace of the THz induced dynamics of the polarization rotation measured at $T = 6$ K and at an external magnetic field $B_{\text{ext}}$ of 40 mT in FeBO$_3$ is shown in Fig. 2(a). The magneto-optical signal exhibits clear oscillations. After performing a Fourier transform these oscillations show up in the frequency domain as two well-defined spectral bands at very different frequencies of 20 and 480 GHz [Fig. 2(b)]. The measurements for different values of $B_{\text{ext}}$ are summarized in Fig. 2(c).

Figure 2(d) shows how the Fourier amplitudes of the modes depend on the strength of the THz electric field. The high-frequency q-AFM mode scales linearly (solid blue curve) with the THz electric field. Although there is no considerably strong spectral component of the THz spectrum near the q-FM frequency, this mode is excited rather efficiently and its amplitude (solid red curve) scales quadratically with the THz electric field. It reveals that the mechanism of the excitation is off-resonant and nonlinear.

Figure 3(a) shows how the amplitude of the q-FM mode depends on the azimuthal angle $\alpha$ of the linearly polarized THz pump pulse. The azimuthal angle $\alpha = 0^\circ$ corresponds to the THz polarization in the $xy$ plane, perpendicular to the applied magnetic field. The Fourier amplitude of the q-AFM mode has a maximum when the THz magnetic field is...
The experiment shows that a THz pulse triggers harmonic oscillations of spins on a much longer time scale than the pulse duration. Such dynamics in antiferromagnets is described by differential equations for damped harmonic oscillators corresponding to the q-FM and q-AFM modes, respectively [26]. These equations are derived from the Lagrangian, being a solution of the Euler-Lagrange equations for $\psi$ and $\theta$ [47]. In the case of normal incidence of the THz pulse and the external magnetic field along the $x$ axis, the torque that triggers the q-AFM mode is $T_{\text{AFM}} \sim H(t) \cos \alpha$ [30]. For the torque that is generated due to the inverse Cotton-Mouton effect and that triggers the q-FM mode one finds $T_{\text{FM}} \sim E^2(t) \sin 2\alpha$ [26]. Here $E(t)$ and $H(t)$ are the electric and magnetic fields of the THz pulse, respectively. A comparison between the outcomes of the model and the experimental results confirms that the q-AFM mode is excited resonantly due to the Zeeman coupling of the THz magnetic field and the sublattice magnetizations, while the q-FM mode is excited due to the mechanism of the inverse Cotton-Mouton effect.

The sum and the difference of the time traces measured at the different polarities of the field $B_{\text{ext}} = \pm 40$ mT are shown in Fig. 3(c). Figure 3(d) shows that the dominating part of the THz-induced dynamics is even with respect to the applied magnetic field. For FeBO$_3$ it is known [23] that the q-FM mode is the easiest to detect via the direct magneto-optical Cotton-Mouton effect, i.e., due to the symmetric part of the optical susceptibility. Figures 3(c) and 3(d) show that
the oscillations at the frequency of the q-FM mode are present only in the sum, but not in the difference plot. The signal that appears in the difference is well above the noise level, but its Fourier spectrum [Fig. 3(d)] does not allow ascribing it to spin dynamics. Hence the signal must have a different origin [48]. Similar to Ref. [23], the observed independence of q-FM oscillations on the polarity of the applied magnetic field allows us to confirm that the spin dynamics is excited and detected via the inverse and direct Cotton-Mouton effects, respectively. The detection of magnetization dynamics is discussed in the Supplemental Material [26].

Changing the value of the applied magnetic field one changes the frequency of the q-FM mode. According to Ref. [23], the amplitude of the q-FM mode excited in FeBO$_3$ due to the inverse Cotton-Mouton effect must be inversely proportional to the frequency of the mode itself. This expectation is confirmed experimentally, as shown in Fig. 3(c). The inverse Cotton-Mouton effect in the THz spectral range reported here results in a spin oscillation of the magneto-optical signal with an amplitude of about 10 mdeg for a pump fluence of $10^{-2}$ mJ. (b) Frequencies of the q-AFM (open blue circles) and q-FM (closed red circles) modes of the waveform from panel (a).

Finally, in order to demonstrate that the observed mechanism of THz-induced spin excitation is very different from the one recently reported for TmFeO$_3$ [21], we studied the efficiency of the excitation as a function of temperature, as shown in Fig. 4(a). Although the measurements at temperatures above 160 K are hampered by a reduction of the net Néel vector and consequently of the magneto-optical Cotton-Mouton effect, even these data show that the q-FM mode excitation mechanism remains efficient in a much broader temperature range as compared to the mechanism reported in Ref. [21]. Temperature dependencies of the frequencies of the excited q-FM and q-AFM modes [see Fig. 4(b)] are in a good agreement with previously reported data [49] and this agreement is the best evidence of negligibly small THz-induced heating in our experiment.

In summary, we show that intense THz pulses efficiently trigger spin oscillations corresponding to the two modes of the antiferromagnetic resonance in FeBO$_3$. While the q-AFM mode is resonantly excited by the magnetic field of the incident THz pulse, the q-FM mode is excited off-resonantly and its amplitude scales quadratically with the strength of the THz electric field. The mechanism of the excitation has all the features of the inverse opto-magnetic Cotton-Mouton effect and is at least as efficient as the similar effect in the near-infrared spectral range. Although until now the effect has only been reported for FeBO$_3$, symmetry analysis shows that the corresponding term in the energy of light-matter interaction is not restricted to a specific crystal point group. Similar to the direct Cotton-Mouton effect, the inverse effect must be allowed in materials of other symmetries. The mechanism is also efficient in a broad temperature range and we expect similar nonlinear effects to be found in other magnetically ordered media. This work opens up a previously ignored channel to control spins in magnetically ordered materials in an energy efficient way.

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