Terahertz Magnon-Polaritons in TmFeO₃

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ABSTRACT: Magnon-polaritons are shown to play a dominant role in the propagation of terahertz (THz) waves through TmFeO₃ orthoferrite, if the frequencies of the waves are in the vicinity of the quasi-antiferromagnetic spin resonance mode. Both time-domain THz transmission and emission spectroscopies reveal clear beatings between two modes with frequencies slightly above and slightly below this resonance, respectively. Rigorous modeling of the interaction between the spins of TmFeO₃ and the THz light shows that the frequencies correspond to the upper and lower magnon-polariton branches. Our findings reveal the previously ignored importance of propagation effects and polaritons in such heavily debated areas as THz magnonics and THz spectroscopy of electromagnons. It also shows that future progress in these areas calls for an interdisciplinary approach at the interface between magnetism and photonics.

KEYWORDS: polaritonics, magnonics, terahertz spectroscopy, antiferromagnetism, ultrafast dynamics

Photonics and magnonics appeal to information technologies for their potential to overcome the problems inherent to modern electronics, such as dissipation of energy due to Ohmic losses.1-4 The viability of magnonics depends on a successful increase of the frequency of magnon-based processing, which in traditional ferromagnetic materials (e.g., YIG) is restricted to the GHz regime.5-7 The use of antiferromagnetic materials could be a solution because of their very high (terahertz) frequencies of spin resonances.8,9 However, the physics of magnons at THz frequencies is far less studied due to a lack of sufficiently fast stimuli and detectors in this area. This therefore calls for an interdisciplinary approach at the intersection of magnetism and photonics. For example, the latest advances in the development of strong THz field sources have opened a doorway for coherent control over magnetism at ultrashort time scales.10

Here we report an experimental and theoretical study of antiferromagnetic magnon-polariton propagation in the orthoferrite TmFeO₃. For this, we employ THz transmission and emission time-domain spectroscopy. Our experimental findings are supported by a rigorous solution of the Maxwell equations. The rare-earth orthoferrites R-FeO₃ offer plenty of opportunities for this type of research. TmFeO₃ has been extensively used as a model system to investigate ultrafast spin dynamics.11-13 This particular orthoferrite has numerous resonances in the THz frequency range related to magnetic resonances of the iron sublattices and electronic transitions in the Tm ions.14,15

Figure 1a shows the unit-cell of the TmFeO₃ single crystal, taking into account the parameter from refs.16,17 The compound crystallizes in the Pbnm structural phase. The spins of Fe³⁺ ions are ordered antiferromagnetically. The magnetic structure can be described by four magnetic sublattices with the magnetizations M₁, M₂, M₃, and M₄, respectively. Owing to the Dzyaloshinsky-Moriya interaction, the antiferromagnetic Fe³⁺ spins are canted over a small angle from the antiparallel orientation and give rise to a net magnetization M = M₁ + M₂ + M₃ + M₄. Such a magnetic structure has two spin resonance modes, namely the quasi-ferromagnetic mode (F-mode) and the quasi-antiferromagnetic mode (AF-mode).18 For TmFeO₃, both these modes are in the THz spectral range. The F-mode resonance can be seen as a precession of the macroscopic magnetization M. The AF-mode, on the other hand, can be seen as a longitudinal oscillation of the magnetization M due to the canting of the Fe³⁺ spins induced by the Dzyaloshinsky-Moriya interaction.13,19

The single crystals of TmFeO₃ studied here were grown in a four-mirror optical floating-zone furnace using four 1.5 kW halogen lamps as the infrared radiation source. The samples had a thickness of about 1.5 mm and were oriented by using X-ray Laue photography. X-ray Laue analysis and X-ray diffraction rocking curves confirmed the high quality, precise orientation, and homogeneity of the studied crystals.

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The magnetic anisotropy of TmFeO$_3$ undergoes dramatic changes as a function of temperature and the compound is characterized by a so-called spin reorientation transition in which the net magnetization changes its orientation.\textsuperscript{20,21} In our sample, at higher temperatures ($T > 93$ K), the spins are in the $\Gamma_4$ phase, with the net magnetization $M$ along the $c$-axis and the antiferromagnetic vector $L = M_1 - M_2 + M_3 - M_4$ along the $a$-axis. At lower temperatures ($T < 85$ K), the spins are in the $\Gamma_2$ phase with the magnetization and antiferromagnetic vectors along the $a$ and $c$ axes, respectively. In the region between 85 and 93 K, there is an intermediate phase $\Gamma_{24}$ in which the antiferromagnetic and magnetization vectors rotate continuously in the $ac$ plane.\textsuperscript{22}

To reveal the strong coupling between the photons and magnons we have performed two types of experiments. The first one was transmission THz time-domain spectroscopy (THz-TDS). The corresponding experimental scheme is shown in Figure 2a. The output of a mode-locked Ti:Sapphire laser with pulse duration of 100 fs, centered wavelength of 800 nm, and repetition rate of 80 MHz (Mai Tai HP-1020, Spectra-Physics) was used to generate and detect the THz radiation. The THz waves were emitted from the sample illuminated by an intense femtosecond laser pulse. The detection of the THz radiation involved ZnTe electro-optical crystals and polarization balanced detection.

THz-TDS allows us to measure the free-induction decay (FID) signals, which result from the magnetic dipole transitions in the THz frequency range. The propagation direction of incident pulses was coaligned with the $c$-axis and the magnetic component of the pulse was pointed along the $a$-axis of the crystal ($B_{THz}\parallel a$).

Figure 3a shows typical waveforms of the THz electrical field transmitted through the TmFeO$_3$ crystal at various temperatures. Below 90 K in the $\Gamma_2$ phase, the main transmitted THz pulse is followed by a long-lived and damped harmonic oscillation component at a frequency around 0.86 THz. Upon temperature decrease the frequency of the oscillations does not
change, while the amplitude increases. Above 90 K, the oscillations disappear completely. The frequency corresponds to the quasi-antiferromagnetic (AF-mode) in this material.23 In the present geometry it can only be excited by a THz magnetic field in the \( \Gamma_2 \) phase. In the Fourier spectra in Figure 3b, this mode is seen as a narrow resonance dip at a frequency of about 0.86 THz; energy from the THz pulses is transferred to the excitation of the AF-mode, which subsequently re-emits part of this energy.

Despite the fact that there is only a single dip observed in the Fourier spectrum for all time-domain range, the changing envelope in the amplitude of the oscillations in the time-frequency domain. In Gaussian units it gives

\[
\frac{\partial}{\partial z} \left( \frac{1}{\mu(z, \omega)} \frac{\partial E_x}{\partial z} \right) + \frac{\omega^2}{c^2} \varepsilon(z) E_x = 0
\]

where the wavy line symbol is used to indicate the Fourier transform with respect to time.

The solution has the form of plane waves:

\[
\tilde{E}_x(z) = \begin{cases} 
A_1 e^{-ik_x z} + B_1 e^{ik_x z}, & z < 0 \\
A_2 e^{-ik_x z} + B_2 e^{ik_x z}, & 0 < z < d \\
A_3 e^{-ik_x z}, & d < z
\end{cases}
\]

where \( A_i \) and \( B_i \) are the amplitudes of the transmitted and reflected electromagnetic waves, \( k_0 \) is the wave vector in air, \( k_i \) is the wave vector in the material, and \( d \) is the thickness of the sample.

From Faraday’s law, the electric field \( \tilde{E}_x \) should be continuous at the interfaces. Furthermore, by integrating eq 1 along an infinitesimal interval \( \Delta z \) we come to the boundary

To understand the physical origin of the beatings we derived a model of light–matter interaction in the THz spectral range.

**MODELS**

**Terahertz Transmission.** As the THz spot on the sample is large compared to the THz wavelength, we employ a plane-wave approximation, assuming that the wave vector \( k \) of the electromagnetic waves is perpendicular to the interface. It means that the waves propagate along the \( z \)-axis, which is the \( z \)-axis of TmFeO₃. The electric \( \mathbf{E} \) and the magnetic field \( \mathbf{B} \) vectors are directed along the \( x \) - and \( y \)-axes, respectively (Figure 5a).

Figure 3. (a) Time-domain traces of the electric field of THz pulses transmitted through TmFeO₃ single crystal for different temperatures; Fourier transforms of the time-domain traces (b) for all range and (c) for the area marked with a rectangle, the region between the THz pulse and its echo (from 14 to 45 ps).

Figure 4. (a) Time traces of the electric field of the THz radiation emitted from a TmFeO₃ single crystal at different temperatures; (b) Fourier transforms of the time-domain emission waveforms.

Figure 5. (a) Geometry of the THz transmission experiment and electromagnetic waves relevant to the problem of light-matter interaction; (b) Spectrum of the THz pulse and frequency-dependence of the real part of the TmFeO₃ magnetic permeability \( \mu(\omega) \); (c) Comparison of calculated and measured (at \( T = 40 \) K) THz waveforms transmitted through the TmFeO₃ sample for the region between the THz pulse and its echo; (d) Fourier transforms of the time traces shown in panel (c).
conditions of continuity of \( i \frac{dE_z}{\mu \, dz} \). These requirements provide the following set of equations:

\[
\begin{align*}
z = 0: & \left\{ \begin{array}{l}
A_1 + B_1 = A_2 + B_2, \\
-k_1 A_1 + k_2 B_1 = -k_2 A_1 + k_1 B_2,
\end{array} \right. \\
z = d: & \left\{ \begin{array}{l}
A_2 e^{-ik_d} + B_2 e^{ik_d} = A_1 e^{-ik_d}, \\
-k_1 A_2 e^{-ik_d} + k_2 B_2 e^{ik_d} = -k_2 A_2 e^{-ik_d},
\end{array} \right.
\end{align*}
\]

(3)

Solving this set of equations for \( A_f \), we obtain a solution for the ratio between the spectrum of light transmitted through the slab and the incident spectrum:

\[
t(\omega) = \frac{A_f}{A_i} E(\omega) = \frac{-4k_1 k_2 \mu(\omega) e^{-\alpha d}}{(k_1 - k_2 \mu(\omega))^2 e^{-\alpha d} - (k_1 + k_2 \mu(\omega))^2 e^{\alpha d}} E(\omega)
\]

(4)

where \( E(\omega) \) is the normalized spectrum amplitude of the incident THz pulse.

One expects to see manifestations of magnetic modes in transmission due to singularities of the magnetic permeability in the vicinity of resonances. For the permeability of the material, we used the standard Drude-Lorentz form (Figure 5b):

\[
\mu(\omega) = \frac{\Delta(\omega^2 - \omega_0^2 - i\omega \Delta \omega)}{\omega_0^2 - \omega^2 - i \omega \Delta \omega}
\]

(5)

with the resonant frequency \( \omega_0 \), absorption line width \( \Delta \omega \), and the effective oscillator strength \( \Delta \mu \).

Taking \( \omega_0 = 860 \text{ GHz for the resonant frequency, the sample thickness } d = 1.5 \text{ mm, tabulated values for other parameters (} \Delta \omega = 8.6 \text{ GHz, } \Delta \mu = 10^{-2}, \mu = 22-0.26i) \) and the spectrum of the incident THz pulse on the near-forward Fourier transformation to eq 5. As a result, we obtained a time-domain profile of the electric field for the radiation transmitted through the structure (Figure 5c) and its spectrum (Figure 5d).

The outcome of the model appears to be in very good agreement with the experimental data.

**Terahertz Emission.** As demonstrated in previous works, femtosecond laser excitation can effectively excite the AF-mode of magnetic resonance in \( \text{TmFeO}_3 \) due to a photoinduced change of the ratio between the exchange parameters in the canted antiferromagnet. This action of the laser pulse on spins can be described as a pulse of an effective magnetic field \( \mathbf{H}_{\text{eff}} \) along the magnetization \( \mathbf{M} \). To find the THz radiation generated by such an effective field, we solve the Maxwell equations in the frequency domain:

\[
\nabla \times \mathbf{E} = \frac{i \omega \mu(\omega)}{c} \mathbf{A} \\
\nabla \times \mathbf{H} = -\frac{i \omega \varepsilon(\omega)}{c} \mathbf{E} - 4\pi i \omega (\nabla \times \mathbf{M})
\]

(6)

(7)

where \( \mathbf{M} = \chi(\omega) \mathbf{H}_{\text{eff}} \) and \( \chi(\omega) \) is the magnetic susceptibility. Dividing eq 6 by \( \mu(\omega) \), taking the curl of the cross product \( \nabla \times \mathbf{E} \) and using eq 7 we obtain:

\[
\nabla \times \left( \frac{1}{\mu(\omega)} \nabla \times \mathbf{E} \right) + \frac{\omega^2 \mathbf{E}}{c^2} = -\frac{4\pi i \omega^2 \chi(\omega)}{c \mu(\omega)} (\nabla \times \mathbf{M})
\]

(8)

This simplifies to:

\[
\mu(\omega) \frac{\partial}{\partial z} \left( \frac{1}{\mu(\omega)} \frac{\partial \mathbf{E}_z}{\partial z} \right) + k^2 \mathbf{E}_z = -\frac{4\pi i \omega^2}{c \mu(\omega)} \frac{\partial \chi(\omega)}{\partial z} \mathbf{H}_{\text{eff}}
\]

(9)

where \( k = \frac{\omega}{c} \sqrt{\varepsilon(\omega)} \) is the wave vector.

The effective magnetic field \( \mathbf{H}_{\text{eff}} \) follows the intensity envelope of the optical Gaussian pump pulse. The propagation speed of this pulse is \( v_p \), the penetration depth into the material is \( l \).

Taking the partial solution in the frequency domain, this can be represented as:

\[
\mathbf{H}_{\text{eff}}(z) = f(\omega) e^{-i \omega z / v_p} e^{-i z / l}
\]

(10)

where \( f(\omega) = A_0 \frac{\tau}{\sqrt{2 \pi} \sigma^2} e^{-\omega^2 \sigma^2 / 4} \) is the spectrum of the envelope of the optical intensity, \( \tau_p \) is the pulse duration, \( A_0 \) is the spectral amplitude of the effective magnetic field.

The solution of eq 9 has the form:

\[
\mathbf{E}_z(z) = \begin{cases} 
B_z e^{i k_d z}, & z < 0 \\
A_z e^{-i k_d z} + B_z e^{i k_d z} + U(z), & 0 < z < d \\
A_z e^{-i k_d z}, & d < z
\end{cases}
\]

(11)

where \( U(z) \) is a partial solution, \( k_1 \) is the wave vector in air, \( k_1 \) is the wave vector in the material, and \( d \) is the thickness of the sample.

Taking the partial solution in the form \( U(z) = G(\omega) e^{-i \omega z / v_p} e^{-i z / l} \), it is found from eq 9 that:

\[
U(z) = -\frac{4\pi i \omega^2 \chi(\omega)}{c (\omega^2 - k_1^2)} \mathbf{H}_{\text{eff}}(z)
\]

(12)

where \( \alpha = \frac{\omega}{v_p} + \frac{1}{l} \) is the effective optical penetration decay.

By integrating Maxwell’s equations over an infinitesimal length crossing the interfaces and taking the limit as the thicknesses of the transition regions go to zero, one gets the boundary conditions. These boundary-matching conditions imply continuity of the electric field, while its derivative exhibits a finite discontinuity:

\[
\left\{ \begin{array}{l}
\frac{1}{\mu(\omega)} \frac{\partial \mathbf{E}_z}{\partial z} \\
\mu(\omega) \frac{\partial \mathbf{E}_z}{\partial z} \\
\end{array} \right\} = \pm \frac{4\pi i \omega^2 \chi(\omega)}{c \mu(\omega)} \mathbf{H}_{\text{eff}}
\]

These requirements provide the following system of equations:

\[
\begin{align*}
\begin{cases}
A_1 + B_1 = A_2 + B_2, \\
-k_1 A_1 + k_2 B_1 = -k_2 A_1 + k_1 B_2,
\end{cases}
\end{align*}
\]

\[
\begin{align*}
\begin{cases}
A_2 e^{-i k_d} + B_2 e^{i k_d} = A_1 e^{-i k_d}, \\
-k_1 A_2 e^{-i k_d} + k_2 B_2 e^{i k_d} = -k_2 A_2 e^{-i k_d},
\end{cases}
\end{align*}
\]

(13)

Solving these equations for \( A_f \), we obtain the solution for the spectrum of the THz radiation emitted from the slab.
\[ A_i = \left[ e^{-i\xi \omega}\left(\gamma(1+i\xi)\left(-2e^{i(k\mu \omega)} + e^{2i(k\mu \omega)} + 1\right) + \gamma^2(1 + e^{2i(k\mu \omega)})\right) - i\gamma(-1 + e^{i(k\mu \omega)})\right] \left(1 + \gamma^2\right)^{\frac{1}{2}} \]

where \( \gamma = \frac{\gamma(\mu, \omega)}{k_0(\omega)} \) and \( \xi(\omega) = -\frac{\alpha(\omega)}{k_0(\omega)} \) are introduced to simplify the expression.

Figure 6 shows such a spectrum (Figure 6b) and the corresponding waveform (Figure 6c). For the calculation we used \( \nu_r = \frac{\nu}{n} \), \( n = 2.3 \) at \( \lambda = 680 \text{ nm} \), \( I = 50 \mu\text{m}^2 \), and \( \tau_p = 50 \). It is seen that the model is in good agreement with the outcome of the experiment.

\section*{DISCUSSION}

We would like to note that, although our models do not contain fitting parameters, they correctly predict the bandwidth of the THz spectrum, position of the beat frequencies, and the positions of the Fabry-Perot peaks.

The dispersion of photons (\( k(\omega) \) dependence from eq 9) in the absence of magnetic resonances (\( \mu(\omega) = \text{const} \)) is a linear function (Figure 7, black line). In the presence of a spin resonance (\( \mu(\omega) \neq \text{const} \)) without spatial dispersion, the \( \omega(k) \) curve has an avoided crossing at the resonance frequency (Figure 7, dash line). However, taking into account dissipation of the magnetic permeability, one finds that a coupled photon-magnon (polariton) is formed (Figure 7, blue solid line). In the vicinity of the resonance frequency for each wave vector, one would get not one, but two solutions corresponding to two magnon-polaritons (red circles) at close frequencies.

Figure 7. Magnon-polariton dispersion in the vicinity of a spin resonance (blue dashed line for an ideal system, infinite discontinuity; blue solid line for the real TmFeO3). The linear dispersion of photons in the absence of the spin resonance is shown for comparison (black line). In the vicinity of the resonance frequency for each wave vector, one would get not one, but two solutions corresponding to two magnon-polaritons (red circles) at close frequencies.

As the magnon-polaritons propagate in space it is instructive to analyze their beating as a function of the crystal thickness. In Figure 8 we show the beating spectra for the region between the THz pulse and its echo in the time-domain range for different crystal thicknesses; (b) emission spectra for different crystal thicknesses.

Figure 8. (a) Calculated transmission beating spectra for the region between the THz pulse and its echo in the time-domain range for different crystal thicknesses; (b) emission spectra for different crystal thicknesses.

beating interferes with the Fabry-Perot echoes of the broadband THz pulse for thicknesses less than 1 mm, which further hinders the observation of beatings in this type of measurement. This analysis is in line with the absence of a clear beating in previous experiments [e.g., refs 13, 22, 24], in which much thinner crystals with thicknesses less than 200 \( \mu\text{m} \) were studied.

\section*{CONCLUSIONS}

Time-domain spectroscopy of THz magnetic resonances in orthoferrites shows that the observed spectra cannot be described without accounting for propagation of electromagnetic waves. Instead of a single peak in the spectra, two peaks with frequencies slightly below and slightly above the frequency of the magnetic resonance are observed. Our findings have implications for several other areas of physics including THz magnonics\(^{27}\) and spectroscopy of electromagnons.\(^{28}\) Therefore, further progress in these areas calls for an interdisciplinary approach at the interface between magnetism and photonics.
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