**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. 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. The use of antiferromagnetic materials could be a solution because of their very high (terahertz) frequencies of spin resonances. 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.

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. This particular orthoferrite has numerous 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.

**Figure 1a** shows the unit-cell of the TmFeO₃ single crystal, taking into account the parameter from refs. 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 $M_1$, $M_2$, $M_3$, and $M_4$, 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 $M = M_1 + M_2 + M_3 + M_4$. Such a magnetic structure has two spin resonance modes, namely the quasi-ferromagnetic mode (F-mode) and the quasi-antiferromagnetic mode (AF-mode). 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.

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 TmFeO₃ 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. In our sample at higher temperatures (T > 93 K), the spins are in the Γ₄ 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 Γ₂ 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 Γ₂₄ in which the antiferromagnetic and magnetization vectors rotates continuously in the ac plane.\(^{22}\)

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 transients. The emitter and detector of the THz waves were photoconductive antennas (PCA) fabricated on low-temperature-grown GaAs substrates.

The second type of experiment was THz emission spectroscopy (Figure 2b). In this technique the sample was illuminated by a femtosecond laser pulse (estimated duration of \(~50\) fs, central wavelength of 680 nm, and repetition rate of 1 kHz) and served as an emitter of THz radiation. The THz radiation emitted from the sample was focused onto the 1 mm thick [110] ZnTe electro-optic crystal gated by femtosecond laser pulses at 800 nm. The terahertz-induced changes of the linear polarization of the gating pulses in the electro-optic crystal were analyzed by polarization optics consisting of a quarter-wave plate, a Wollaston polarizer and a pair of balanced silicon photodiodes. To increase the signal-to-noise ratio, we used an opto-mechanical chopper modulating the pump beam intensity combined with a lock-in amplifier.

In all these experiments the sample was placed in a coldfinger cryostat with two transparent windows and biased by in-plane magnetic fields with a strength up to 1 kG. Furthermore, the THz spectrometer and THz emission setup were purged with nitrogen in order to minimize absorption of THz radiation by atmospheric water vapor.

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 TmFeO₃ crystal at various temperatures. Below 90 K in the Γ₂ 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

![Figure 1](image1.png)  
Figure 1. (a) Crystal structure of a TmFeO₃ crystal (blue, thulium; brown, iron; red, oxygen). Fe ions are located in centrosymmetrical positions having a distorted octahedral oxygen environment; (b) Spin alignment of the four antiferromagnetic Fe³⁺ sublattices in the Γ₂ phase below 80 K.

![Figure 2](image2.png)  
Figure 2. Schematics of experimental setups. (a) Time-domain THz transmission spectroscopy (TDS). Photoconductive antennas (PCA), fabricated on low-temperature-grown GaAs substrates, were used for the generation and detection of the THz radiation; (b) THz emission spectroscopy. 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.

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and above the frequency of the AF-mode of the antiferromagnetic resonance in the $\Gamma_1$ phase of TmFeO$_3$.

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 $c$-axis of TmFeO$_3$. The electric $E$ and the magnetic field $B$ vectors are directed along the $x$- and $y$-axes, respectively (Figure 5a).

To find the THz radiation transmitted through an infinite plate of thickness $d$ with relative permittivity $\varepsilon$ and magnetic permeability $\mu = \mu(\omega)$, we use the wave equation in the frequency domain. In Gaussian units it gives

$$\frac{\partial}{\partial z} \left( \frac{1}{\varepsilon(z, \omega)} \frac{\partial E_z}{\partial z} \right) + \frac{\omega^2}{c^2} \varepsilon(z) E_z = 0$$

(1)

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

The solution has the form of plane waves:

$$E_z(z) = \begin{cases} A_1 e^{-ik_1 z} + B_1 e^{ik_1 z}, & z < 0 \\ A_2 e^{-ik_2 z} + B_2 e^{ik_2 z}, & 0 < z < d \\ A_3 e^{-ik_3 z}, & d < z \end{cases}$$

(2)

where $A_i$ and $B_i$ are the amplitudes of the transmitted and reflected electromagnetic waves, $k_0$ is the wave vector in air, $k_1$ is the wave vector in the material, and $d$ is the thickness of the sample.

From Faraday’s law, the electric field $\vec{E}_z$ should be continuous at the interfaces. Furthermore, by integrating eq 1 along an infinitesimal interval $\Delta z$ we come to the boundary...
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\[ \nabla \times \mathbf{E} = \frac{i \omega E}{c} \mathbf{H} \]

\[ \nabla \times \mathbf{H} = -\frac{i \omega \mathbf{E}}{c} - 4\pi i \alpha \nabla \times \mathbf{M} \]

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

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

This simplifies to

\[ \mu(\omega) \frac{\partial}{\partial z} \left( \frac{1}{\mu(\omega)} \frac{\partial \mathbf{E}_z}{\partial z} \right) + k^2 \mathbf{E}_z = -\frac{4\pi \omega^2}{\mu(\omega)} \frac{\partial(\chi(\omega) \mathbf{H}_{\text{eff}})}{\partial z} \tag{9} \]

where \( k = \frac{\omega}{\sqrt{\varepsilon \mu(\omega)}} \) 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 \( v_\text{p} \), the penetration depth into the material is \( l \). In the frequency domain, this can be represented as

\[ H_{\text{eff}} = f(\omega) e^{-i\omega z/v_\text{p}} e^{-z/l} \tag{10} \]

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

The solution of eq 9 has the form

\[ \mathbf{E}_z(z) = \begin{cases} B_0 e^{ik_0 z}, & z < 0 \\ A_0 e^{-ik_0 z} + B_0 e^{ik_0 z} + U(z), & 0 < z < d \\ A_0 e^{-ik_0 z}, & d < z \end{cases} \quad \tag{11} \]

where \( U(z) \) is a partial solution, \( k_0 \) 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 \( U(z) = G(\omega) e^{-i\omega z/c} e^{-z/l} \), it is found from eq 9 that

\[ U(z) = -\frac{4\pi i \alpha \chi(\omega)}{c(a^2 - k^2)} \mathbf{H}_{\text{eff}}(z) \quad \tag{12} \]

where \( \alpha = \frac{i \omega}{c} + \frac{1}{l} \) 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:

\[ \begin{cases} 1 \frac{\partial \mathbf{E}_z}{\partial z} = \frac{4\pi \omega^2 \chi(\omega)}{\mu(\omega)} \mathbf{H}_{\text{eff}} \end{cases} \]

These requirements provide the following system of equations:

\[ z = 0: \begin{cases} B_1 = A_2 + B_2 + U(0), \\ ik_0 B_1 = ik_1 A_2 - ik_1 B_2 - \frac{\alpha}{\mu(\omega)} U(0), \end{cases} \]

\[ z = d: \begin{cases} ik_0 A_2 - ik_1 A_2 + \frac{\alpha}{\mu(\omega)} U(d) = -ik_0 A_2 e^{-ik_0 d} \end{cases} \quad \tag{13} \]

Solving these equations for \( A_2 \), we obtain the solution for the spectrum of the THz radiation emitted from the slab.
Figure 6. (a) Geometry of THz generation in a slab of orthoferrite and the electromagnetic waves relevant to the problem of light–matter interaction; (b) Calculated and measured Fourier amplitudes $E_{FT}$ of the electric field for radiation generated in the TmFeO$_3$ sample slab by a laser pulse at 40 K; (c) Time traces corresponding to the spectra shown in (b).

used $v_g = c/n$, $n = 2.3$ at $\lambda = 680$ nm; $l = 50 \mu m$, and $\tau_p = 50$.

It is seen that the model is in good agreement with the outcome of the experiment.

**DISCUSSION**

We would like to note that, although our models do not contain fitting parameters, they correctly predict the linewidth 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) = $ const) is a linear function (Figure 7, black line). In the presence of a spin resonance ($\mu(\omega) \neq $ 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 magneton-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.

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 $\mu m$ were studied.

**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. Therefore, further progress in these areas calls for an interdisciplinary approach at the interface between magnetism and photonics.
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Notes
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