

THz-Driven Spin Dynamics in Orthoferrites with Kramers and Non-Kramers Rare-Earth Ions

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We study the THz-driven spin dynamics of the canted antiferromagnet ErFeO_3 across the spin reorientation temperatures 80–90 K. The amplitudes of the soft antiferromagnetic resonance mode are drastically enhanced by an order of magnitude in the middle of the spin reorientation temperature interval in ErFeO_3 . Conversely, in the phenomenologically similar magnetic system of TmFeO_3 , exhibiting the same phase transitions at very similar temperatures, this enhancement is not observed. We attribute this enhancement in ErFeO_3 to the dynamic interaction of the iron $3d$ spins with the rare-earth $4f$ -electronic transitions of the Kramers Er^{3+} ions. We confirm this interpretation by comparing measurements on TmFeO_3 , where the non-Kramers Tm^{3+} electronic states do not strongly couple to the antiferromagnetic mode. Thus, our results indicate that the character of the microscopic electronic $4f$ states may drastically alter the ultrafast magnetic response in the materials with otherwise very similar static magnetic properties and phase diagrams.

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The antiferromagnets, magnetically ordered materials with antiparallel spin alignment, have attracted interest in the last decade [1–3] owing to their unique properties distinguishing them from conventional ferromagnets. In contrast to the ferromagnets, which possess resonance frequencies in the gigahertz range, antiferromagnetic resonance frequencies are in the terahertz range, making them suitable candidates for ever-faster magnetic data storage. Moreover, the antiferromagnets are naturally more abundant than ferromagnets, are less sensitive to external magnetic fields, and generate nearly zero stray fields [4].

In general, the fastest timescale at which magnetic order can be switched is determined by the precession frequency of the ordered spins [5]. However, until recently accessing such fast spin dynamics has been a challenge due to the lack of means to excite antiferromagnets at timescales faster than their intrinsic response time. The first ultrafast stimulus for successful excitation of coherent antiferromagnetic spin precession was the femtosecond laser pulse [6–8]. More recent studies have demonstrated the possibility of switching antiferromagnetic order using electrical pulses by means of current induced spin-orbit torques [9,10]. Nonetheless, the actual writing of

information with light pulses in antiferromagnets remains a challenge due to the relatively inefficient coupling of the antiferromagnetic spin system with optical pulses [11]. Indeed, to write magnetic information, sufficiently high amplitudes of spin deflections are required so that the spins can permanently reorient themselves. Driving the spin dynamics resonantly with intense THz pulses, in contrast to the off-resonant excitation by high-energy optical photons, can overcome this challenge [12]. Only recently, using custom-made antenna's grown on Thulium orthoferrite (TmFeO_3), sufficiently high field amplitudes of THz pulses have been achieved to induce switching between stable antiferromagnetic states [13]. Besides, many recent studies have shown that THz pulses can trigger nonlinear spin dynamics. Examples include THz-assisted optically driven spin reorientation [14], demonstrations of the THz-induced anharmonic coupling between magnon modes [15–17], spin-lattice coupling [18,19], and high harmonic generation of magnon modes [20–23]. Erbium orthoferrite (ErFeO_3) has recently attracted interest due to reports of quantum phenomena such as Dicke cooperativity [24].

Here, we study the spin dynamics in the canted antiferromagnets ErFeO_3 in comparison to TmFeO_3 using intense THz pulses. The iron spin system of the orthoferrites [25] consists of two magnetic sublattices with magnetizations $\mathbf{M}_1$ and $\mathbf{M}_2$. Below the Néel temperature of $T_N \approx 600$ K, the $3d$ spins of the Fe^{3+} ions are ordered antiferromagnetically with a small canting angle induced by the Dzyaloshinskii-Moriya interaction, giving rise to a

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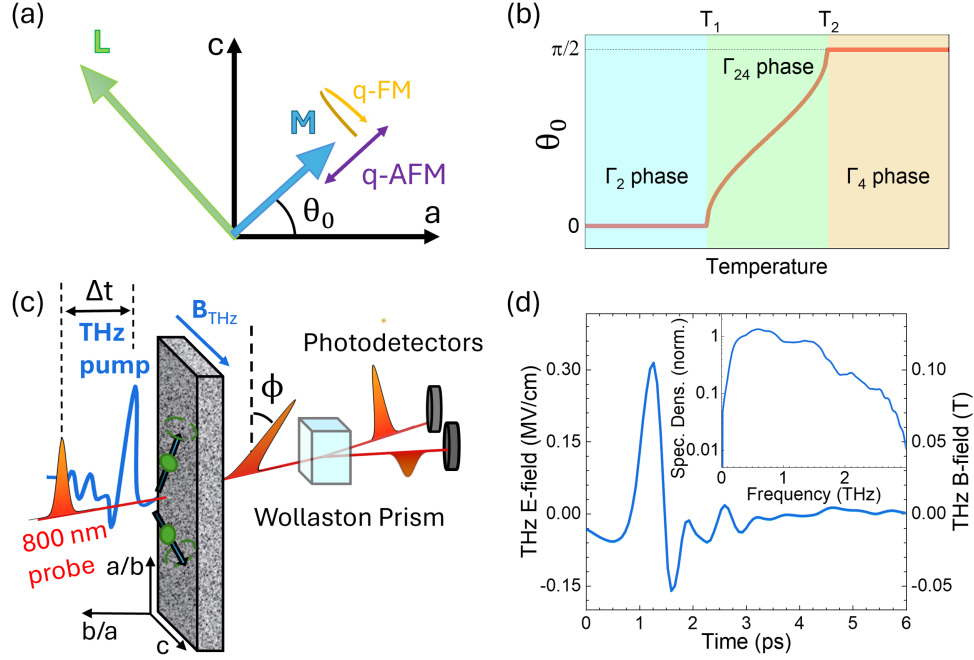


FIG. 1. (a) Illustration of the SRT in ErFeO_3 and TmFeO_3 and the geometry of the two modes of AFMR of the Fe^{3+} spins. (b) Angle of the weak ferromagnetic moment with respect to the crystallographic a axis, highlighting the SRT. (c) Schematic of the experimental setup. The crystallographic axes of the orthoferrites are indicated, and the magnetic field of the THz pump is oriented along the crystallographic c axis. (d) Experimental time-domain trace of the THz pump pulse that drives the spin dynamics. The inset shows the Fourier spectrum.

weak ferromagnetic moment [26]. Hence, it is typically convenient to describe the spin dynamics in terms of the antiferromagnetic moment $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$, and the ferromagnetic moment $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2$, which are orthogonal to each other. The macroscopic magnetic properties of ErFeO_3 and TmFeO_3 are similar and are summarized in Figs. 1(a) and 1(b). In the high temperature phase ($T > T_2 = 97$ K for ErFeO_3 , $T > T_2 = 93$ K for TmFeO_3), the weak ferromagnetic moment $\mathbf{M}$ aligns itself along the crystallographic c axis (Γ_4 phase), and in the low-temperature phase ($T < T_1 = 88$ K for ErFeO_3 , $T < T_1 = 85$ K for TmFeO_3) [27,28], $\mathbf{M}$ is oriented along the crystallographic a axis (Γ_2 phase). In between these temperatures, i.e., for $T_1 < T < T_2$, the spins undergo a spin reorientation transition (SRT) in the ac plane. The angle with respect to the c axis at temperature T is given by [11]

$$\frac{\pi}{2} - \theta_0 = \arcsin \left(\sqrt{\frac{T - T_2}{T_1 - T_2}} \right). \quad (1)$$

When the spin system is brought out of the equilibrium state, the spins will undergo precession at the two antiferromagnetic resonance (AFMR) frequencies: the quasi-ferromagnetic (q -FM) mode corresponds to the precession of the weak ferromagnetic moment about its equilibrium orientation and the quasiantiferromagnetic (q -AFM) mode

corresponds to the oscillations of the length of the weak ferromagnetic moment.

The $4f$ -electronic states of the rare-earth elements also contribute to the magnetic properties of the rare-earth orthoferrites [29]. These $4f$ electrons exhibit paramagnetism at temperatures above the rare-earth Néel temperature of $T \approx 4$ K in ErFeO_3 , and form additional antiferromagnetic sublattices below this temperature [30]. In TmFeO_3 , this ordering does not appear above 1.6 K [31]. However, the rare-earth spins interact with the iron spin system, and are thus essential for understanding the static magnetic properties in the orthoferrites, such as the SRT [32,33]. Here, we will show that this interaction strongly affects the dynamic magnetic properties of the antiferromagnetic order as well. To this end, we compare two orthoferrites with nearly identical macroscopic properties, but with two distinct characters of the rare-earth states: with Kramers symmetry in Er^{3+} and non-Kramers symmetry in Tm^{3+} . We report on the striking enhancement of the q -FM mode in the SRT of ErFeO_3 , whereas this mode in the SRT of TmFeO_3 is suppressed.

The interaction between the electrons in the rare-earth ion, occupying the sites with C_s symmetry, and the crystal field of the surrounding ions gives rise to an meV range energy splitting (i.e., THz frequency range) of the electronic ground state, allowing for the resonant excitation of the dipole-active transitions of the rare-earth modes [34]. Importantly, the Er^{3+} ion is a Kramers ion, containing

an odd number of electrons in the $4f$ shells, that results in splitting to the double degenerate energy levels, known as Kramers doublets [35]. In contrast, Tm^{3+} is a non-Kramers ion containing an even number of electrons in the $4f$ shell. Here, the crystal field splits the electronic levels into fully degenerate singlets.

The crystal field splitting results in the lowest excited energy levels of the Tm^{3+} ions in the orthoferrite separated by 2.28 meV (0.55 THz), 4.97 meV (1.2 THz), and 8.69 meV (2.1 THz) from the ground state [27]. The energy level diagram of ErFeO_3 is more complex due to further splitting of Kramers doublets due to the d - f exchange coupling with iron spins, resulting in a multitude of high-frequency transitions in the range of approximately 1.2–1.5 THz and low-frequency transitions within the Kramers doublets at approximately 100 GHz between the lowest two crystal field energy levels [26,34]. Thus, besides the direct coupling to the AFMR modes, the THz pulses can also reveal indirect coupling to the spins of d electrons in Fe^{3+} ions through the energy levels of the rare-earth f electrons. The efficiency of such a mechanism is seen, for instance, in changing the magnetic anisotropy experienced by Fe^{3+} spins as a result of repopulation of electronic energy levels of rare-earth ions [11].

Our experimental setup is illustrated in Fig. 1(c). We generate intense THz pulses by optical rectification in a LiNbO_3 prism, using the tilted pulse front technique

[36,37], and focus these pulses onto the sample by a set of parabolic mirrors. This allows us to achieve single cycle THz magnetic fields of the order of 0.1 Tesla [see Fig. 1(d)]. The magnetic field component of the THz pump pulse excites the spin dynamics in the orthoferrites resonantly by exerting a Zeeman torque on the $3d$ spins of the iron ions. Our ErFeO_3 and TmFeO_3 samples are bulk crystals, with a thickness of ~ 60 μm . We first studied a b -cut ErFeO_3 crystal, with the magnetic field of the THz pulse polarized along the crystallographic c axis of the sample. The spin dynamics was detected by measuring the Faraday rotation of a time-delayed 800 nm probe pulse, using a Wollaston prism and a pair of balanced photo-detectors. The sample was placed into a cryostat that allowed cooling the sample to liquid Helium temperatures. We note that the absorption of the 800 nm photons is independent of temperature in the orthoferrites [38–40] (also see Supplemental Material [41]).

We measured the spin dynamics at various temperatures across the SRT. Indicative time-domain signals for ErFeO_3 are shown in Fig. 2(a).

In the corresponding spectra [Fig. 2(b)], we observe the two modes of the antiferromagnetic resonance, depending on the temperature. In the low-temperature Γ_2 phase, the polarization of B_{THz} is orthogonal to the weak ferromagnetic moment, thus driving the q-FM mode. In contrast, B_{THz} is aligned parallel with this magnetization in the

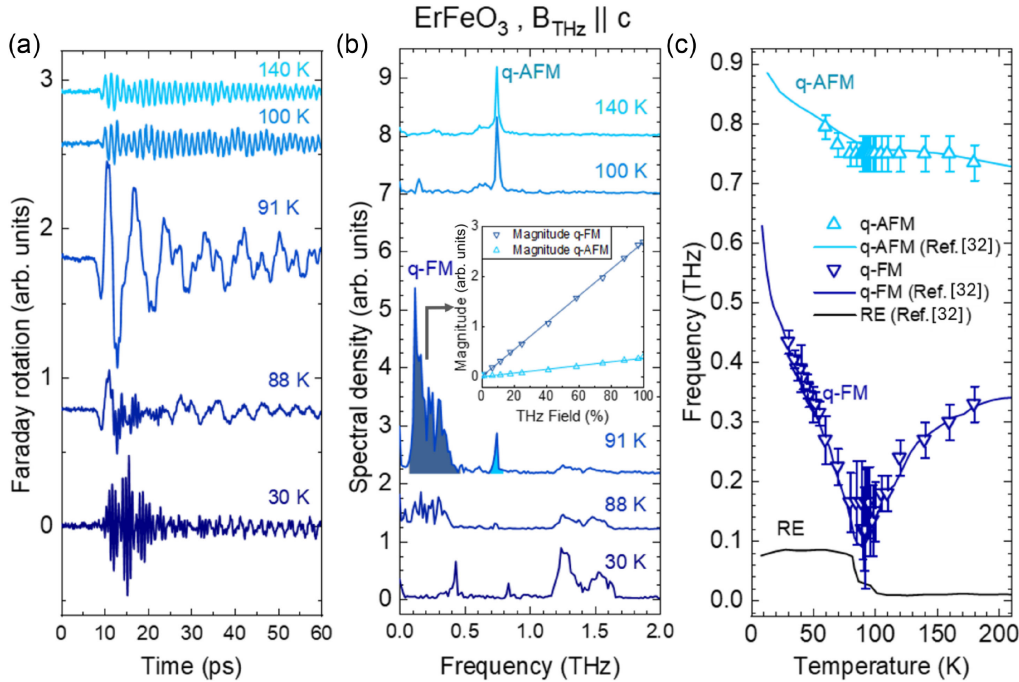


FIG. 2. THz-driven spin dynamics in ErFeO_3 for various indicative temperatures. Panel (a) shows time-domain traces of the Faraday signals and panel (b) shows the corresponding spectra. The inset in panel (b) shows the amplitudes of the q-FM and q-AFM modes as a function of the magnetic field of the THz pulse. In panel (c) the observed frequencies of the q-FM and the q-AFM mode are summarized. Error bars indicate an estimation of the half-width of the peak. The solid lines show the temperature dependence of the q-FM, q-AFM, and rare-earth (RE) modes extracted from literature [32].

high-temperature Γ_4 phase, thus exciting the q-AFM mode according to the selection rules [43].

Moreover, in the Γ_2 phase, more specifically at the low temperatures, we see a broad feature of high-frequency modes around 1.3 and 1.55 THz. We attribute this to the resonant magnetic dipole active f - f transitions of the crystal field splitting of the Er^{3+} ions induced by the THz pulse. These features are most pronounced for low temperatures and disappear upon approaching the temperature where the reorientation to the Γ_4 phase occurs. The reduction of the amplitude of the high-frequency rare-earth modes can be attributed to the thermal smearing of the population of the crystal field energy levels. As the rare-earth modes follow the same selection rules as the q-FM mode, we observe the full disappearance of the rare-earth modes upon entering the high-temperature Γ_4 phase.

Interestingly, we also see an enhancement of the q-FM mode amplitude in the middle of the SRT. Although the frequency dependence on temperature is well understood [32], the observed amplitude dependence on temperature is unexpected, as the magnetization rotates away from the orientation that is orthogonal to the THz magnetic field in the SRT. Hence, a gradual decrease in amplitude from the aforementioned selection rules would be expected, which is in contrast with our experimental observation. Here, we propose that this surprising amplitude enhancement can be attributed to the dynamic coupling between the rare-earth modes and the q-FM mode. Additionally, we observe a complex structure of multiple peaks forming around 300 GHz in the SRT, which has previously been attributed to the hybridization of the q-FM mode with the impurity modes due to the $^6\text{A}_1$ crystal field transitions of the Fe^{3+} ions [44].

The amplitude dependence raises the question of what mechanism is responsible for the increase of the q-FM amplitude and whether this mechanism is nonlinear. To determine the origin of this enhancement, we attenuated the THz pulses with a set of wire grid polarizers and checked the dependence of the amplitude of the q-FM and q-AFM mode in the middle of the SRT at $T = 91$ K on the magnetic field strength of the THz pulse. We found that the amplitudes of both modes scale linearly with the driving magnetic field component and thus conclude that the Zeeman torque drives the spin dynamics. Therefore, we exclude that this enhancement in the SRT is due to the previously reported nonlinear effects [11,15,16,18,22].

In Fig. 2(c), we extracted the frequencies of the observed q-FM mode and q-AFM modes (triangles) and compared these with AFMR frequencies reported in literature. We observe an excellent match of the frequencies with previous reports [32]. Moreover, the literature data show us that the low-frequency rare-earth modes in ErFeO_3 exhibit an avoided crossing with the q-FM mode, implying that there is a strong interaction between these modes. We note that the gap is about 10 GHz, which corresponds to a coupling energy of 0.04 meV, compared to the ~ 0.1 meV energy

scale of the f mode and the ~ 1 meV scale of the d modes. However, we do not directly observe this low-frequency mode in our signals, probably because it does not modulate the magneto-optical response, which is primarily determined by the Fe^{3+} states.

To check if this enhancement is also present in other orthoferrites, we performed the same set of measurements on a -cut TmFeO_3 , which has very similar magnetic properties to ErFeO_3 . Remarkably, as shown and discussed in the End Matter, we do not observe any similar enhancement in TmFeO_3 .

To understand the observed enhancement in ErFeO_3 , we compared the amplitudes of the q-FM mode in ErFeO_3 and TmFeO_3 (Fig. 3) and tracked the integrated spectral weight of the q-FM mode in ErFeO_3 as a function of the temperature [Fig. 3(b)]. The spectral weight is normalized with respect to the spectral density of the THz pulse at the resonant frequencies to account for the uneven THz driving strength at different frequencies (i.e., at different temperatures).

The comparison of the amplitudes in the two different orthoferrites confirms opposite behaviors. In TmFeO_3 the amplitude of the q-FM mode decreases rapidly across the spin reorientation, whereas in ErFeO_3 this mode is enhanced. This is in stark contrast with the expected amplitude of the q-FM mode from following the projection of $\mathbf{M}$ on the axis orthogonal to the THz magnetic field $\sim \cos \theta_0$. The angle θ_0 depends on the temperature, as given by Eq. (1) [see also Fig. 1(b)].

On the other hand, the amplitude of the q-FM mode in ErFeO_3 matches the symmetry of the $\sin \theta_0 \cos \theta_0$. Taking in mind the presence of strong interaction between the q-FM mode and the rare-earth mode as seen from the avoided crossing reported in Ref. [32], we explain this by introducing a coupling between the spins of the rare-earth $4f$ -electronic transition, and iron d -electron spin q-FM resonance, based on the theoretical formalism derived in Ref. [29]. As the rare-earth magnetic moments are deflected, this deflection transfers resonantly to the iron spin system due to this d - f interaction. Using the Lagrangian formalism, we calculate the additional torque that the deflected rare-earth moments exert on the iron spins (see Supplemental Material [41]) and show that

$$\Delta\tau_{df} = \frac{dL_{df}}{d\delta\theta} = \frac{-dF_{df}}{d\delta\theta} \propto \sin(\theta_0) \cos(\theta_0), \quad (2)$$

where L_{df} and F_{df} represent the Lagrangian and the free energy terms, describing the d - f interaction respectively, $\delta\theta$ is the deflection of the rare-earth spins, and θ_0 is the equilibrium orientation of the weak ferromagnetic moment of the iron subsystem with respect to the crystallographic a axis [see Fig. 1(a)], which is given by Eq. (1).

This calculation shows that the torque is exactly proportional to $\sin \theta_0 \cos \theta_0$, excellently matching our experimental observation. We conceptually illustrate the mechanism

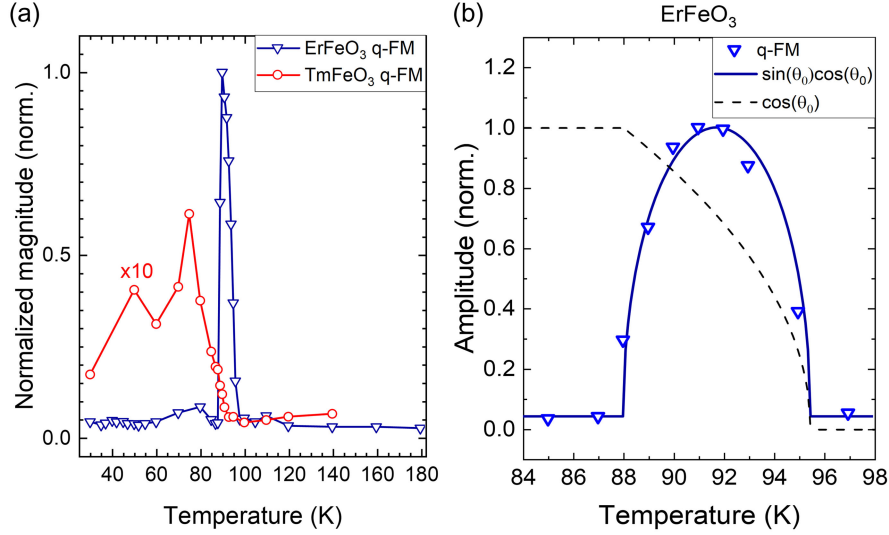


FIG. 3. (a) Comparison of the amplitudes of the q-FM mode in ErFeO₃ (blue) and TmFeO₃ (red). (b) Integrated spectral weight of the q-FM mode in ErFeO₃ across the SRT [as marked with the shaded areas in Fig. 2(b)]. The solid line shows a function $\sim \sin(\theta_0)\cos(\theta_0)$ (dark blue line, q-FM) and the expected amplitude of the q-FM mode from the projection of $\mathbf{M}$ on the crystallographic a axis is plotted as $\cos(\theta_0)$ (dashed black line).

for the enhancement in Fig. 4. The frequencies of the rare-earth modes closely match with the frequency of the q-FM mode in the SRT interval in ErFeO₃. In contrast, the transitions between the energy levels in TmFeO₃ occur at much higher frequencies and thus does not allow for a resonant transfer of energy between the rare-earth and iron spin systems. Thus, as our experiment indicates, the coupling of the rare-earth modes to the iron spin dynamics will only happen efficiently in ErFeO₃ and is not observed in TmFeO₃.

One may also speculate that the discrepancies in the observed amplitudes of the q-FM modes in ErFeO₃ and TmFeO₃ may have originated from variations of the magneto-optical constants. However, comparisons of the

static magneto-optical responses in our experiment and comparisons with literature data [45] do not indicate significant deviations of the magneto-optical constants in this temperature region of interest.

To conclude, we have demonstrated the strong enhancement in the amplitude of the q-FM mode in ErFeO₃ in the middle of the SRT. Similar experiments in TmFeO₃, which has nearly identical macroscopic magnetic properties as ErFeO₃, did not reveal such enhancement. We proposed the mechanism for this unexpected observation and attributed this to the dynamical coupling between the rare-earth $4f$ -electron spins and the iron $3d$ -electron spins and found that the amplitude of the AFMR modes scale linearly with the THz magnetic field. Therefore, accounting for microscopic interactions underlying macroscopic magnetic properties can be critical for understanding ultrafast spin dynamics in different materials, even if they exhibit the same static magnetic properties. Our proposed mechanism scales linearly with the magnetic field of the THz pulse, however enhancing the strength of THz pulses towards fields of more than 1 MV/cm (0.3 Tesla) may facilitate the search for nonlinear effects in spin dynamics coupled to low-energy electronic transitions in transition metal oxides.

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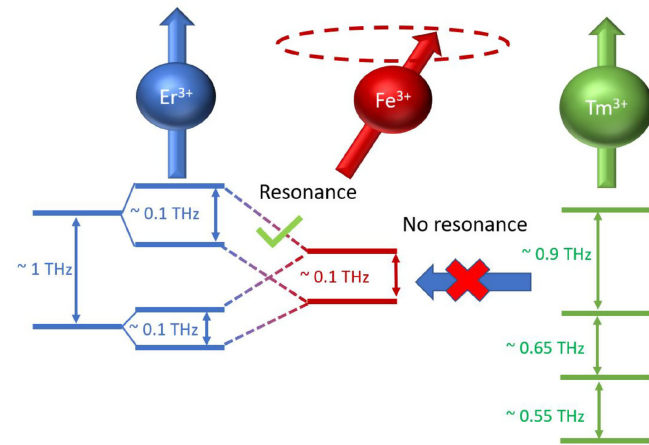


FIG. 4. Schematic of the energy levels of the Er³⁺ and Tm³⁺ ions, explaining the resonant enhancement of the q-FM mode in ErFeO₃ and the lack thereof in TmFeO₃.

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End Matter

Absence of the q-FM amplitude enhancement in TmFeO_3 —We investigated further if the enhancement of the q-FM amplitude in the SRT in ErFeO_3 is also present in other orthoferrites. To this end, we performed the same set of measurements on *a*-cut TmFeO_3 , which

has similar magnetic properties to ErFeO_3 . The indicative results are shown in Fig. 5.

Again, we observe the q-FM mode in the Γ_2 phase and the q-AFM mode in the Γ_4 phase. We note that the rare-earth modes that we observed at low temperatures in

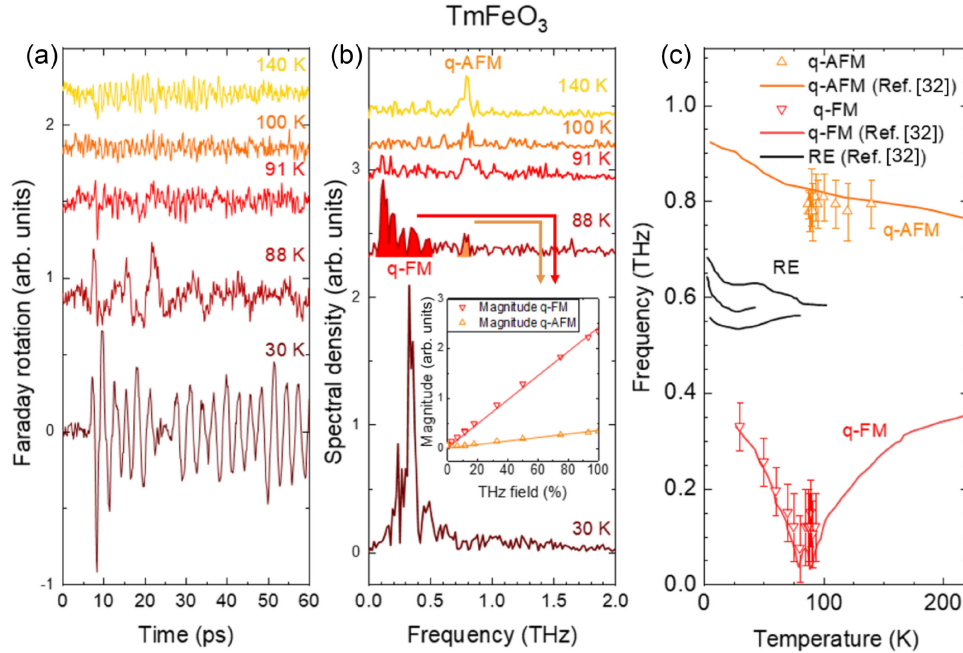


FIG. 5. THz induced spin dynamics of TmFeO_3 for a variety of temperatures. Panel (a) shows the time-domain signals and panel (b) shows their spectra. The inset in panel (b) shows the integrated spectral weight for the q-FM and the q-AFM mode as a function of the field amplitude of the THz pulse. The triangles in panel (c) indicate the extracted frequencies from the spectra in panel (b). Error bars indicate an estimate of the half-width of the peaks. The solid lines represent the frequencies of the q-FM, q-AFM, and rare-earth (RE) modes extracted from literature [32].

ErFeO_3 are not visible here. Moreover, in contrast to the measurements on ErFeO_3 , we see that the q-FM mode is further suppressed in the SRT region. We repeated our investigation to see if the excitation of the antiferromagnetic resonance showed any sign of nonlinearity, and found, similarly to ErFeO_3 that the amplitudes scale linearly with the fluence of the THz pulse. We note that this linear response is in contrast with the previous report on THz-driven dynamics in TmFeO_3 [11]. We find that this discrepancy is the result of a different THz excitation polarization, and that our sample orientation is not sensitive to the nonlinear mechanism previously reported. We

confirmed that the nonlinear effect can be reproduced by applying the THz magnetic field along the b axis of TmFeO_3 . This indicates that different mechanisms are responsible for the spin dynamics in the SRT, and the linear effect we report here is distinct to the nonlinearity previously observed in TmFeO_3 . The drastic difference in the response in the SRT region for ErFeO_3 and TmFeO_3 implies that the rare-earth ion character is responsible for the enhancement of the dynamics in ErFeO_3 . In Fig. 5(c), we again tracked the frequencies of the AFMR modes in TmFeO_3 and observe that in contrast to ErFeO_3 , these modes do not show any crossing over the rare-earth modes.