Nonlinear spin control by terahertz-driven anisotropy fields

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Future information technologies, such as ultrafast data recording, quantum computation or spintronics, call for ever faster spin control by light[1-16]. Intense terahertz pulses can couple to spins on the intrinsic energy scale of magnetic excitations[5,11]. Here, we explore a novel electric dipole-mediated mechanism of nonlinear terahertz-spin coupling which is much stronger than the linear Zeeman coupling to the terahertz magnetic field[5,10]. Using the prototypical antiferromagnet thulium orthoferrite ($TmFeO_3$), we demonstrate that resonant terahertz pumping of electronic orbital transitions modifies the magnetic anisotropy for the ordered Fe^{3+} spins and triggers large-amplitude coherent spin oscillations. This mechanism is inherently nonlinear, it can be tailored by spectral shaping of the terahertz waveforms, and its efficiency outperforms the Zeeman torque by an order of magnitude. Since orbital states govern the magnetic anisotropy in all transition metal oxides, the demonstrated control scheme is expected to be applicable to many magnetic materials.

Ultrafast magnetization control has become a key incentive of modern photonics, with a broad variety of successful concepts emerging at fast pace. Examples include light-induced spin reorientation in canted antiferromagnets[3], the vectorial control of magnetization by light[6], photoinduced antiferromagnet-ferromagnet phase transitions[9], optical modification of the exchange energy[4,14], and driving spin precessions via nonlinear magneto-phononic coupling[7,16]. Despite this remarkable progress, the lion share of the photon energy in all known concepts using visible and near-infrared light is idle with respect to the light-spin interaction, and avoiding dissipation of large excess energies requires special care.

In contrast, intense electromagnetic pulses at terahertz (1 THz = 10^{12} Hz) frequencies may interface spin dynamics directly on their intrinsic energy scales[5,11]. The magnetic field component of few-cycle THz pulses has been used to coherently control magnons in the electronic ground state by direct Zeeman interaction[5,11]. Since magnetic dipole coupling is typically weak, however, THz-driven spin excitation has been confined to the linear response regime. Massive nonlinearities, such as THz-driven phase transitions[17,18] and THz lightwave electronics[19-22], in turn, have been realized by all-electric coupling to the charge degree of freedom. Apart from pioneering work on electromagnons in multiferroic $TbMnO_3$ (Ref. 13), efficient ways to exploit the THz electric field for the control of magnetic order have been missing.

Here, we introduce a conceptually new universal route to control magnetism by THz electric fields. The strength and direction of the magnetic anisotropy in practically all materials is determined by the coupling of electronic orbital states to ordered spins. Therefore, an ultrashort electric field pulse can change the orbital state of electrons abruptly, leading to a sudden modification of the magnetic anisotropy. In our proof-of-concept experiment, we exploit intense, phase-locked THz pulses to achieve such an abrupt change of the magnetic anisotropy, which in turn triggers magnon oscillations with large amplitudes that scale quadratically with the THz field strength.

Non-thermal pumping of orbital transitions in the optical range is known to induce a nonlinear spin-charge coupling on ultrashort timescales[23,24]. In the THz spectral range, this concept can be applied to any material in which selected low-energy electronic transitions change the magnetic anisotropy, for example in oxides containing both 3d and 4f ions (e.g. orthoferrites, manganites, garnets and ferroborates) and in 3d-compounds like hematite α-Fe_2O_3. However, despite the anticipated strong impact of the preparation of non-thermal orbital states on the anisotropy field, terahertz spin control exploiting orbital transitions has remained largely unexplored[25,26].

Figure 1 illustrates the fundamental idea of our experiment for the case of the prototypical antiferromagnet $TmFeO_3$. This material crystallizes in a distorted perovskite structure (Fig. 1a). The four iron spins (blue arrows) per unit cell occupy two antiferromagnetically coupled sublattices, whose spin orientations are mutually canted by the Dzyaloshinskii-Moriya interaction[27]. The $^3H_6$ ground state of the paramagnetic rare-earth Tm^{3+} ions is fully split by the crystal field into a series of singlets with a characteristic energy spacing of $\sim 1-10$ meV (Ref. 28). The angular momenta of these states are coupled with the Fe^{3+} spins by exchange and dipolar interactions, which set the magnetic anisotropy. Thermal population of the singlet states within the $^3H_6$ multiplet changes the magnetic anisotropy as a function of temperature, leading to spin reorientation phase transitions[29]: In the Γ_2 phase ($T < T_1 = 80$ K), the antiferromagnetic vector **G** is aligned along the crystallographic *z*-axis, whereas it lies along the *x*-axis in the Γ_4 phase ($T > T_2 = 90$ K). For $T_1 < T < T_2$ (Γ_{24} phase), **G** rotates continuously in the (*xz*)-plane (see Fig. 1b, Eq. (6) in Methods, Supplementary Fig. 1 and Supplementary Movie 1). The spin dynamics supports two eigenmodes, the quasi-ferromagnetic (q-FM) and the quasi-antiferromagnetic (q-AFM) one. Instead of thermal activation, resonant pumping of electronic transitions between orbital states of the rare-earth ions by THz pulses may be expected to abruptly modify the magnetic anisotropy to trigger coherent magnon oscillations (Fig. 1c and Supplementary Movie 2).

We excite a 60-μm-thick window of $TmFeO_3$ by intense few-cycle THz transients generated by tilted-pulse-front optical rectification of near-infrared laser pulses[11]. The THz peak field can be tuned up to $B_{\mathrm{THz}} = 0.3$ T without changing the waveform (Fig. 2a). The spectral content lies between 0.1 and 2 THz, covering both magnon modes of Fe^{3+} spins and several transitions of the Tm^{3+} ground state multiplet[28] (Fig. 2b). Excitation of phonons can be excluded as they feature frequencies above 3 THz[28,30]. The induced ultrafast magnon dynamics is revealed by tracking the polarization rotation imprinted by the Faraday effect and magnetic linear dichroism on co-propagating near-infrared 30-fs probe pulses (Fig. 2c, Supplementary Figs. 2 and 3). Both the q-FM and the q-AFM modes are excited (see Supplementary Fig. 4), and their frequencies exhibit characteristic temperature dependences (Fig. 2d). Close to the phase transitions, we observe a dramatic softening of the q-FM mode down to a frequency of 50 GHz, in agreement with theory[28].

Next, we systematically vary B_{THz} while keeping the sample in the Γ_{24} transition phase ($T = 84.5$ K), where pumping of the rare-earth states should have maximum impact on the magnetic anisotropy. Figure 3a shows the dynamic polarization rotation as a function of the delay time t between the THz pump and the optical probe. Each transient is normalized by the corresponding THz peak field (see Supplementary Fig. 5 for a quantitative analysis of the magnetization deflection angles). The signal exhibits an oscillatory behaviour with two quasi-monochromatic components at frequencies of 0.1 THz and 0.8 THz corresponding to the q-FM and the q-AFM mode, respectively (Fig. 3b). Most remarkably, the relative strength of the q-FM mode grows with increasing THz peak fields. Figure 3c summarizes the amplitude of both modes as a function of B_{THz}. The q-AFM mode scales linearly with the THz driving field as expected for the linear Zeeman interaction. In contrast, the q-FM mode shows a distinctly nonlinear increase, which is well fit by a superposition of linear and quadratic functions of the peak field. The nonlinearity vanishes when the crystal leaves the Γ_{24} phase (Fig. 3d).

One can show (see “Model for purely magnetic interaction” in Methods and Supplementary Fig. 6), that the Zeeman torque exerted by the THz magnetic field on the Fe^{3+} spins cannot explain the nonlinear excitation of the q-FM mode. In contrast, the THz electric field can influence the magnetic system: The point group of the orthoferrites allows for an anisotropic energy term that scales quadratically with the electric field. Due to this term, the THz driving field may change the magnetic anisotropy (see “The role of the THz electric field” in Methods). Importantly, the link between the THz electric field and the spins is not restricted to a certain microscopic mechanism. In our specific experiment, the THz electric field resonantly excites electronic transitions between the singlet states of the $^3H_6$ multiplet of the Tm^{3+} ions (Ref. 28 and Supplementary Fig. 7). The concomitant non-thermal occupation change alters the magnetic anisotropy.

The fact that the nonlinear excitation is achieved most efficiently in the vicinity of the magnetic phase transition temperatures, where the static magnetic anisotropy is effectively zero, indicates that the excitation of the q-FM magnon is caused by the transient anisotropy. Indeed, our model shows that anisotropy changes generated by the electric field of the THz pulse can drive large-angle excitations of the magnetic lattice, which behaves "soft" at these temperatures (see 'The role of the THz electric field' in Methods). Figure 3d depicts the temperature dependence of the modelled anisotropy torque (black curve) together with the deviation of the q-FM amplitude from a linear scaling with the THz field (red data points). Our theory traces the experimental data very well.

To put our interpretation to an ultimate test, we repeat the experiments with spectrally filtered THz pulses that selectively excite either the q-FM mode or the electronic transitions in the Tm^{3+} ions. Indeed, we find a linear dependence of the magnon amplitude on the THz field if the q-FM mode is excited only (Fig. 4a). A complementary THz spectrum (inset of Fig. 4b) that cannot couple by Zeeman interaction, resonantly prepares non-thermal orbital states of the Tm^{3+} ions, leading to a quadratic scaling (Fig. 4b). These results prove that the nonlinear spin excitation is mediated by the rare-earth ions, which exert a strong effective torque on the Fe^{3+} spin system. From our measurements, we extract that the strength of the anisotropy-mediated spin excitation is 8 times as large as the Zeeman interaction for THz pulses featuring peak magnetic fields of 0.3 T (see Methods section). With the latter, peak-to-peak magnetization deflection angles of 2.6° are reached (Supplementary Fig. 5). Neglecting saturation effects, we estimate that up-scaling the THz electric field to ~ 3 MV/cm may suffice for non-thermal switching of the magnetization direction by 90° via the nonlinear anisotropy torque (see Supplementary Fig. 8). Finally, we note that a similar nonlinear scaling of the magnon amplitude with the THz field occurs in dysprosium orthoferrite ($DyFeO_3$) (see Supplementary Fig. 9), another magnetic reference system, underlining the broad applicability of the new concept of a THz-induced anisotropy torque.

In conclusion, we demonstrated a novel interface between THz fields and the spin system of an antiferromagnet which exploits *electric-dipole* transitions coupled to the *magnetic* degrees of freedom of electrons. In this way, we realized the first nonlinear excitation of the amplitude of spin oscillations using THz pulses. The spectral sensitivity of the effect and its high efficiency compared to the Zeeman excitation open an unprecedented doorway to further raise the amplitude of THz-driven spin deflection using pulse-shaping and coherent control. Throughout the broad class of rare-earth-transition metal compounds yet-predicted field thresholds[8] for THz induced magnetic switching may be reduced by an order of magnitude. Our work exploits a new, general concept of electric field control of magnetic excitations by creating hidden states of matter which involve the spin degree of freedom. In the same spirit, one may now investigate the role of other low-energy elementary excitations, such as excitons or phonons[16], which could change the orbital wavefunctions of nearby atoms and lead to the creation of magnons by a related mechanism. Finally, the new principle of a symmetry-breaking preparation of low-energy non-thermal states may open unforeseen applications in future spin-based devices.

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Methods

Sample. A single crystal of $TmFeO_3$ (grown by a floating zone melting technique) with a thickness of ~ 60 μm and lateral dimensions of ~ 5 mm was used in the experiments. The plate was cut perpendicularly to one of the crystal's optical axes which lies in the (*yz*)-plane at an angle of 51° with respect to the *z*-axis. This orientation allows for THz excitation of both magnon modes at all temperatures. A constant magnetic field of 0.1 Tesla saturates the magnetization of the sample.

Setup. The detailed experimental setup is shown in Supplementary Fig. 10: A low-noise titanium:sapphire laser amplifier (centre wavelength, 800 nm; pulse energy, 5.5 mJ; repetition rate, 3 kHz; pulse duration, 30 fs) is used to generate intense few-cycle THz fields by tilted-pulse-front optical rectification in a cryogenically cooled $LiNbO_3$ crystal. A pair of wire-grid polarizers and different THz spectral filters (see insets to Fig. 4 for their transmission characteristics) allow for adjusting the peak fields and the spectral shape of the THz pulses. A small portion of the laser pulses (pulse duration, 30 fs; pulse energy, ~ 10 nJ) is sent through a mechanical delay line and is used as a polarization probe. The THz and the probe pulses are collinearly focused onto the $TmFeO_3$ sample, which is mounted in a helium cryostat for temperature control. The THz-induced rotation of the linear polarization of the probe pulses is analysed by polarization optics consisting of a half-wave plate, a Wollaston polarizer and a pair of balanced silicon photodiodes. A lock-in amplifier is used to record the diode signals as a function of the time delay between the THz and the probe pulses.

Theoretical formalism.

Model for purely magnetic THz-spin interaction. We describe the dynamics of the quasi-ferromagnetic mode of the weak ferromagnet $TmFeO_3$ using the Lagrangian L and Rayleigh R functions of the angle θ of the normalized antiferromagnetic vector **G** with respect to the crystal axis x (see Fig. 1a), and its time derivative $\dot{\theta}$ in a form[31,32]

$$L = \frac{M_{\mathrm{Fe}}}{2\gamma^2 H_{\mathrm{E}}}\dot{\theta}^2 - \frac{M_{\mathrm{Fe}}}{\gamma H_{\mathrm{E}}} B_y \dot{\theta} - W(\theta), \qquad (1)$$

$$R = \frac{\alpha M_{\mathrm{Fe}}}{2\gamma}\dot{\theta}^2 . \tag{2}$$

Here, M_{Fe} is the magnetization of the single Fe^{3+} sublattice, H_{E} is the effective field of the *d*-*d* exchange, α is the Gilbert damping parameter and $W(\theta)$ is the free energy. For $TmFeO_3$ subjected to the magnetic field **B** of the THz pulse, one has

$$W(\theta) = K_1 \sin^2\theta + K_2 \sin^4\theta - \frac{H_{\mathrm{D}}}{H_{\mathrm{E}}} M_{\mathrm{Fe}} (B_z \cos\theta - B_x \sin\theta), \tag{3}$$

where H_{D} is the Dzyaloshinskii field, K_2 is a constant parameter and $K_1 = 2K_2 \frac{T - T_2}{T_1 - T_2}$ with $T_1 \approx 80$ K, $T_2 \approx 90$ K. Equation (3) accounts for the small spin canting angle $\varepsilon = \frac{H_{\mathrm{D}}}{H_{\mathrm{E}}}$ which is a result of Dzyaloshinskii-Moriya interaction. In Eq. (3), the term containing the magnetic field arises from Zeeman coupling. The equation of motion reads

$$\frac{d}{dt}\left(\frac{\partial L}{\partial \dot{\theta}}\right) - \frac{\partial L}{\partial \theta} + \frac{\partial R}{\partial \dot{\theta}} = 0 , \tag{4}$$

which, in the case of the functions (1) - (3), can be written as

$$\ddot{\theta} + \omega_{\mathrm{E}}\alpha\dot{\theta} + \omega_{\mathrm{E}}\omega_{\mathrm{A}} w(\theta, T) = \omega_{\mathrm{E}}\gamma\dot{B}_y - \frac{H_{\mathrm{D}}}{H_{\mathrm{E}}}\omega_{\mathrm{E}}\gamma B_z \sin\theta - \frac{H_{\mathrm{D}}}{H_{\mathrm{E}}}\omega_{\mathrm{E}}\gamma B_x \cos\theta , \tag{5}$$

where γ is the gyromagnetic ratio, $\omega_{\mathrm{E}} = \gamma H_{\mathrm{E}}$, $\omega_{\mathrm{A}} = \gamma \frac{K_2}{M_{\mathrm{Fe}}}$ and $w(\theta, T) = \sin\theta\cos\theta\left(\frac{T - T_2}{T_1 - T_2} + \sin^2\theta\right)$. Equation (5) has the form of a generalized sine-Gordon equation and is nonlinear with respect to θ. Assuming $\ddot{\theta} = \dot{\theta} = 0$ in Eq. (5) one can find the equilibrium orientation θ_0 of the antiferromagnetic vector for a given temperature as

$$\theta_0 = \begin{cases} \frac{\pi}{2}, T < T_1 \\ \arcsin\left(\frac{T - T_2}{T_1 - T_2}\right)^{\frac{1}{2}}, \quad T_1 < T < T_2 \\ 0, T > T_2. \end{cases} \tag{6}$$

The temperature dependence $\theta_0(T)$ is shown in Fig. 1b and Supplementary Fig. 1. We numerically find the solution of Eq. (5) taking the time trace of the magnetic field $B(t)$ from the experiment (see Fig. 2a) and assume standard initial conditions $\theta(t=0)=\theta_0,\ \dot{\theta}(t=0)=0$. For calculations we take M_{Fe} = 1000 emu cm^{-3}, $H_{\mathrm{E}} = 2\times10^7$ Oe, $H_{\mathrm{D}} = 2\times10^5$ Oe and $\omega_{\mathrm{E}}\alpha \approx 0.05$ ps^{-1} (Ref. 28). The resulting time evolution of the angle $\theta(t)$ is shown in Supplementary Fig. 6 for different peak amplitudes of the driving magnetic field B_{THz} polarized along the x-axis, as in the experiment. While we confirmed the possibility of excitation of the quasi-ferromagnetic mode via the Zeeman mechanism, we did not find any deviation from the linear relation between the maximum amplitude of θ and B_{THz} below 0.3 Tesla for any temperature and orientation of the field in the (xz)-plane. Our analysis shows that the Zeeman interaction of the Fe^{3+} sublattices with the THz magnetic field cannot lead to the nonlinear excitation observed in the experiment.

The role of the terahertz electric field. To account for the observed nonlinear interaction between the Fe^{3+} spins and the THz pulses we write the free energy term quadratic with respect to the THz electric field **E** and the antiferromagnetic vector **G** as

$$W_{\mathrm{int}} = \sum_{i,k,l,m} g_{iklm} G_l G_m E_i E_k\ , \tag{7}$$

which is allowed in centrosymmetric $TmFeO_3$. Here, g_{iklm} are the components of the nonlinear magnetoelectric susceptibility tensor $\hat{\mathbf{g}}$. The symmetry point group D_{2h}^{16} of $TmFeO_3$ dictates the form of the tensor $\hat{\mathbf{g}}$ which reads (in the Voigt notation)

$$\hat{\mathbf{g}} = \begin{pmatrix} g_{11} & g_{12} & g_{13} & 0 & 0 & 0 \\ g_{12} & g_{22} & g_{23} & 0 & 0 & 0 \\ g_{13} & g_{23} & g_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & g_4 & 0 & 0 \\ 0 & 0 & 0 & 0 & g_5 & 0 \\ 0 & 0 & 0 & 0 & 0 & g_6 \end{pmatrix} . \tag{8}$$

Taking into account that the antiferromagnetic vector in $TmFeO_3$ lies in the (xz)-plane, such that $\mathbf{G} = (\cos\theta, 0, \sin\theta)$, we get

$$W_{\mathrm{int}} = \cos^2\theta\left(g_{11}E_x^2 + g_{12}E_y^2 + g_{13}E_z^2\right) + \sin^2\theta\left(g_{12}E_x^2 + g_{22}E_y^2 + g_{33}E_z^2\right) + \frac{1}{2} g_5 \sin 2\theta\, E_x E_z \tag{9}$$

which can be rearranged as

$$W_{\text{int}} = f(\mathbf{E}) + \sin^2\theta\left(\chi_x E_x^2 + \chi_y E_y^2 + \chi_z E_z^2\right) + \frac{1}{2} g_5 \sin 2\theta\, E_x E_z\,, \tag{10}$$

where $\chi_x = g_{12} - g_{11}$, $\chi_y = g_{22} - g_{12}$, $\chi_z = g_{33} - g_{13}$. The function $f(\mathbf{E}) = \left(g_{11}E_x^2 + g_{12}E_y^2 + g_{13}E_z^2\right)$ does not depend on θ and can be omitted.

Importantly, the interaction term in Eq. (10) can be present in any crystal with D_{2h}^{16} point group regardless of the exact microscopic origin (electronic excitations, phonons, excitons, etc.) of coupling between the antiferromagnetic vector and the electric field. In our experiment the THz magnetic field is linearly polarized along the *x*-axis and therefore $E_x = 0$. Thus, the interaction energy (10) reduces to

$$W_{\text{int}} = (\chi_y E_y^2 + \chi_z E_z^2)\sin^2\theta \tag{11}$$

and can be seen as a modulation of the anisotropy energy $W_{\text{A}}(\theta) = K_1 \sin^2\theta + K_2 \sin^4\theta$. The THz electric field changes the anisotropy parameter K_1 in Eq. (3) by $\Delta K_1 = \chi_y E_y^2 + \chi_z E_z^2$.

The coupling described by Eq. (11) generates a torque acting on spins which results in an additional term on the right-hand side of the generalized sine-Gordon equation (7) as

$$\ddot{\theta} + \omega_{\text{E}}\alpha\dot{\theta} + \omega_{\text{E}}\omega_{\text{A}} w(\theta, T) = -\omega_{\text{E}}\omega_{\text{A}} \sin\theta\cos\theta\, a(t), \tag{12}$$

with

$$a(t) = \chi_y E_y^2(t) + \chi_z E_z^2(t) \tag{13}$$

being proportional to the THz intensity time trace. In Eq. (12) we do not include the additive Zeeman torque considered above to isolate the effect of the THz-induced transient anisotropy. If the duration of the THz pulse and the relaxation time of the excited Tm^{3+} states is much shorter than the period of the quasi-ferromagnetic mode one can show that the generated torque acts as an instantaneous excitation described by the initial conditions

$$\theta(t=0) = \theta_0, \quad \dot{\theta}(t=0) = -\omega_{\text{E}}\omega_{\text{A}} \sin\theta_0 \cos\theta_0 \int_0^{\tau_{\text{THz}}} a(t)dt\,, \tag{14}$$

where τ_{THz} is the duration of the THz pulse. The transient anisotropy torque depends on the temperature as $\sin\theta_0(T)\cos\theta_0(T)$ which is shown in Fig. 3d. It is non-zero only in the temperature interval corresponding to the intermediate magnetic phase Γ_{24}. Obviously, the torque is quadratic with respect to the THz field **E** (Eq. (13) and (14)). The Zeeman torque and the transient anisotropy torque are additive, but scale differently with the THz peak field.

Finally we note that our phenomenological theory correctly reproduces the spin dynamics on timescale longer than the duration of the THz-driven perturbation of the anisotropy fields. To describe the strongly non-equilibrium state during the interaction of the medium with the intense THz pulse, one may need to account for the THz carrier wave within a time-dependent many-body theory.

Estimation of the strength of the Zeeman torque. Applying a THz low-pass filter with a nominal cut-off frequency of 0.3 THz allows us to selectively excite the q-FM mode (see inset to Fig. 4a). In this measurement, the q-FM amplitude amounts to A_z = 8% of the one induced by the unfiltered THz transients (Fig. 4a). Taking into account the filter transmission of T_F = ~ 75% at the q-FM resonance frequency, we conclude that the anisotropy induced torque is $\frac{1-A_z/T_F}{A_z/T_F} \approx 8$ times as large as the Zeeman interaction.

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Author contributions

S.B., A.V.K., R.H. and R.V.M. conceived the study, carried out the experiments and analysed the data. A.K.Z. and R.V.M. developed the theoretical model. S.B., M.H., A.V.K., R.H., R.V.M. wrote the manuscript. All authors discussed the results.

Additional Information

Supplementary information is available in the online version of the paper. Reprints and permissions information is available online at www.nature.com/reprints. Correspondence should be addressed to R.H. (rupert.huber@physik.uni-regensburg.de) or R.V.M. (R.Mikhaylovskiy@science.ru.nl).

Competing financial interests

The authors declare no competing financial interests.

Figure legends

Figure 1 | Principle of spin control by a THz induced anisotropy torque. a, Spin and lattice structure of $TmFeO_3$ shown in the Γ_{24} phase. Green/blue spheres: Tm^{3+}/Fe^{3+} ions. Oxygen atoms are not shown for clarity. The iron spins (blue arrows) form two antiferromagnetically coupled sublattices $\mathbf{M}_1$ and $\mathbf{M}_2$, which are mutually canted by the Dzyaloshinskii-Moriya interaction, giving rise to a weak ferromagnetic moment $\mathbf{F} = \mathbf{M}_1 + \mathbf{M}_2$. In the Γ_{24} phase (i.e. for 80 K < T < 90 K), the antiferromagnetic vector $\mathbf{G} = \mathbf{M}_1 - \mathbf{M}_2$ encloses a finite angle $0° < \theta_0 < 90°$ with the x-axis. **b**, Spin reorientation phase transitions: In the Γ_2 phase (i.e. T < 80 K) the antiferromagnetic vector **G** is aligned along the crystallographic z-axis, whereas it lies along the x-axis above $T = 90$ K (Γ_4 phase). In the Γ_{24} phase, **G** rotates continuously in the (xz)-plane (see also panel **a**). **c,** The crystal field splits the ground state $^3H_6$ of the rare-earth Tm^{3+} ions into several energy levels with an energy spacing of ~1 – 10 meV (schematic level scheme). The corresponding orbital wavefunctions set the magnetic anisotropy for the iron spins in thermal equilibrium (upper panel). Lower panel: Ultrafast transitions between these energy levels resonantly induced by THz pulses should exert an abrupt torque on the spins and act as an efficient trigger for coherent spin dynamics. The small canting angle is not shown for clarity.

Figure 2 | Overview of the experiment. a, THz transients used to excite magnon/Tm^{3+} resonances in $TmFeO_3$. **b**, Amplitude spectrum of the waveform shown in panel **a**. Arrows mark the frequencies of the magnon/Tm^{3+} resonances. **c**, Schematic of the experiment: THz-pump (red) and near infrared probe pulses (NIR, blue) are collinearly focused onto the $TmFeO_3$ sample with variable time delay t. Using a $\lambda/2$ plate, a Wollaston prism, and two balanced photodiodes, THz-induced magnetic dynamics in $TmFeO_3$ are measured by polarization rotation of the probe pulses. **d,** Resonance frequencies of the q-FM (red circles) and q-AFM (blue triangles) modes in dependence on the sample temperature T. Black curves are guides to the eye.

Figure 3 | Nonlinear THz-magnon interaction. a, Normalized magnon traces for various THz excitation strengths B_{THz}: Whereas quasi-monochromatic oscillations are found for the lowest THz field, a low-frequency oscillation is superimposed onto the dynamics for higher pump fields. **b**, Amplitude spectra of the time domain data shown in panel **a** allow for the identification of the q-FM and q-AFM modes at 100 and 830 GHz, respectively. **c,** Scaling of the amplitudes from panel **b**: The q-AFM mode (blue triangles) scales linearly with the THz field strength, whereas the q-FM mode (red circles) shows a quadratic dependence on the latter. Error bars denote the standard deviation interval for the THz amplitude, arising from uncertainties in the THz spot size and the repeatability of the polarizer angle. **d,** Deviation of the experimental field-scaling of the q-FM mode with the THz field strength from a linear behaviour (red data points) and THz-induced anisotropy torque exerted on the spins (black curve) as computed by our model for various temperatures at the magnetic phase transitions. The nonlinear behaviour vanishes outside the Γ_{24} phase. Error bars take account of the uncertainty in the extracted nonlinearity owing to noise in the measurements.

Figure 4 | Control of THz-induced nonlinear torque by spectral shaping: a, Low pass filtering the pristine THz spectrum (grey shaded curve in inset) results in the black shaded spectrum (inset) featuring a dominant maximum at the q-FM resonance frequency. Main graph: Linear scaling of the q-FM amplitudes obtained by Zeeman excitation with the constrained THz spectrum (red circles). The q-AFM mode (blue triangles) is suppressed since the THz amplitude at the corresponding frequencies is strongly reduced by the low pass filter. **b**, The Zeeman type excitation of the q-FM mode is switched off by bandpass filtering (centre frequency 1.2 THz) of the THz pulse (black/grey: filtered/pristine spectrum). Main graph: Tm^{3+} excitation triggers q-FM oscillations, whose amplitudes scale quadratically with the THz fields (red circles). Additionally, q-AFM magnons (amplitude: blue triangles) are excited by the Zeeman interaction. All curves are normalized to the maximum amplitude of the q-FM mode shown in Fig. 3c. Dotted/solid lines are linear/quadratic fits. Error bars are retrieved as in Fig. 3c.