

# Nonlinear spin control by terahertz-driven anisotropy fields

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

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

19 In contrast, intense electromagnetic pulses at terahertz ( $1 \text{ THz} = 10^{12} \text{ Hz}$ ) frequencies may interface spin  
20 dynamics directly on their intrinsic energy scales<sup>5,11</sup>. The magnetic field component of few-cycle THz pulses  
21 has been used to coherently control magnons in the electronic ground state by direct Zeeman interaction<sup>5,11</sup>.  
22 Since magnetic dipole coupling is typically weak, however, THz-driven spin excitation has been confined to  
23 the linear response regime. Massive nonlinearities, such as THz-driven phase transitions<sup>17,18</sup> and THz  
24 lightwave electronics<sup>19-22</sup>, in turn, have been realized by all-electric coupling to the charge degree of freedom.  
25 Apart from pioneering work on electromagnons in multiferroic  $\text{TbMnO}_3$  (Ref. 13), efficient ways to exploit  
26 the THz electric field for the control of magnetic order have been missing.

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

34 Non-thermal pumping of orbital transitions in the optical range is known to induce a nonlinear spin-charge  
35 coupling on ultrashort timescales<sup>23,24</sup>. In the THz spectral range, this concept can be applied to any material in  
36 which selected low-energy electronic transitions change the magnetic anisotropy, for example in oxides  
37 containing both 3d and 4f ions (e.g. orthoferrites, manganites, garnets and ferrobates) and in 3d-compounds

38 like hematite  $\alpha\text{-Fe}_2\text{O}_3$ . However, despite the anticipated strong impact of the preparation of non-thermal  
39 orbital states on the anisotropy field, terahertz spin control exploiting orbital transitions has remained largely  
40 unexplored<sup>25,26</sup>.

41 Figure 1 illustrates the fundamental idea of our experiment for the case of the prototypical antiferromagnet  
42  $\text{TmFeO}_3$ . This material crystallizes in a distorted perovskite structure (Fig. 1a). The four iron spins (blue  
43 arrows) per unit cell occupy two antiferromagnetically coupled sublattices, whose spin orientations are  
44 mutually canted by the Dzyaloshinskii-Moriya interaction<sup>27</sup>. The  ${}^3H_6$  ground state of the paramagnetic rare-  
45 earth  $\text{Tm}^{3+}$  ions is fully split by the crystal field into a series of singlets with a characteristic energy spacing of  
46  $\sim 1 - 10$  meV (Ref. 28). The angular momenta of these states are coupled with the  $\text{Fe}^{3+}$  spins by exchange and  
47 dipolar interactions, which set the magnetic anisotropy. Thermal population of the singlet states within the  ${}^3H_6$   
48 multiplet changes the magnetic anisotropy as a function of temperature, leading to spin reorientation phase  
49 transitions<sup>29</sup>: In the  $\Gamma_2$  phase ( $T < T_1 = 80$  K), the antiferromagnetic vector  $\mathbf{G}$  is aligned along the  
50 crystallographic  $z$ -axis, whereas it lies along the  $x$ -axis in the  $\Gamma_4$  phase ( $T > T_2 = 90$  K). For  $T_1 < T < T_2$  ( $\Gamma_{24}$   
51 phase),  $\mathbf{G}$  rotates continuously in the  $(xz)$ -plane (see Fig. 1b, Eq. (6) in Methods, Supplementary Fig. 1 and  
52 Supplementary Movie 1). The spin dynamics supports two eigenmodes, the quasi-ferromagnetic (q-FM) and  
53 the quasi-antiferromagnetic (q-AFM) one. Instead of thermal activation, resonant pumping of electronic  
54 transitions between orbital states of the rare-earth ions by THz pulses may be expected to abruptly modify the  
55 magnetic anisotropy to trigger coherent magnon oscillations (Fig. 1c and Supplementary Movie 2).

56 We excite a 60- $\mu\text{m}$ -thick window of  $\text{TmFeO}_3$  by intense few-cycle THz transients generated by tilted-pulse-  
57 front optical rectification of near-infrared laser pulses<sup>11</sup>. The THz peak field can be tuned up to  $B_{\text{THz}} = 0.3$  T  
58 without changing the waveform (Fig. 2a). The spectral content lies between 0.1 and 2 THz, covering both  
59 magnon modes of  $\text{Fe}^{3+}$  spins and several transitions of the  $\text{Tm}^{3+}$  ground state multiplet<sup>28</sup> (Fig. 2b). Excitation  
60 of phonons can be excluded as they feature frequencies above 3 THz<sup>28,30</sup>. The induced ultrafast magnon  
61 dynamics is revealed by tracking the polarization rotation imprinted by the Faraday effect and magnetic linear  
62 dichroism on co-propagating near-infrared 30-fs probe pulses (Fig. 2c, Supplementary Figs. 2 and 3). Both the  
63 q-FM and the q-AFM modes are excited (see Supplementary Fig. 4), and their frequencies exhibit

64 characteristic temperature dependences (Fig. 2d). Close to the phase transitions, we observe a dramatic  
65 softening of the q-FM mode down to a frequency of 50 GHz, in agreement with theory<sup>28</sup>.

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

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

87 The fact that the nonlinear excitation is achieved most efficiently in the vicinity of the magnetic phase  
88 transition temperatures, where the static magnetic anisotropy is effectively zero, indicates that the excitation  
89 of the q-FM magnon is caused by the transient anisotropy. Indeed, our model shows that anisotropy changes

90 generated by the electric field of the THz pulse can drive large-angle excitations of the magnetic lattice, which  
91 behaves “soft” at these temperatures (see ‘The role of the THz electric field’ in Methods). Figure 3d depicts  
92 the temperature dependence of the modelled anisotropy torque (black curve) together with the deviation of the  
93 q-FM amplitude from a linear scaling with the THz field (red data points). Our theory traces the experimental  
94 data very well.

95 To put our interpretation to an ultimate test, we repeat the experiments with spectrally filtered THz pulses that  
96 selectively excite either the q-FM mode or the electronic transitions in the  $\text{Tm}^{3+}$  ions. Indeed, we find a linear  
97 dependence of the magnon amplitude on the THz field if the q-FM mode is excited only (Fig. 4a). A comple-  
98 mentary THz spectrum (inset of Fig. 4b) that cannot couple by Zeeman interaction, resonantly prepares non-  
99 thermal orbital states of the  $\text{Tm}^{3+}$  ions, leading to a quadratic scaling (Fig. 4b). These results prove that the  
100 nonlinear spin excitation is mediated by the rare-earth ions, which exert a strong effective torque on the  $\text{Fe}^{3+}$   
101 spin system. From our measurements, we extract that the strength of the anisotropy-mediated spin excitation is  
102 8 times as large as the Zeeman interaction for THz pulses featuring peak magnetic fields of 0.3 T (see  
103 Methods section). With the latter, peak-to-peak magnetization deflection angles of  $2.6^\circ$  are reached  
104 (Supplementary Fig. 5). Neglecting saturation effects, we estimate that up-scaling the THz electric field to  
105  $\sim 3$  MV/cm may suffice for non-thermal switching of the magnetization direction by  $90^\circ$  via the nonlinear  
106 anisotropy torque (see Supplementary Fig. 8). Finally, we note that a similar nonlinear scaling of the magnon  
107 amplitude with the THz field occurs in dysprosium orthoferrite ( $\text{DyFeO}_3$ ) (see Supplementary Fig. 9), another  
108 magnetic reference system, underlining the broad applicability of the new concept of a THz-induced  
109 anisotropy torque.

110 In conclusion, we demonstrated a novel interface between THz fields and the spin system of an  
111 antiferromagnet which exploits *electric-dipole* transitions coupled to the *magnetic* degrees of freedom of  
112 electrons. In this way, we realized the first nonlinear excitation of the amplitude of spin oscillations using THz  
113 pulses. The spectral sensitivity of the effect and its high efficiency compared to the Zeeman excitation open an  
114 unprecedented doorway to further raise the amplitude of THz-driven spin deflection using pulse-shaping and  
115 coherent control. Throughout the broad class of rare-earth-transition metal compounds yet-predicted field

116 thresholds<sup>8</sup> for THz induced magnetic switching may be reduced by an order of magnitude. Our work exploits  
117 a new, general concept of electric field control of magnetic excitations by creating hidden states of matter  
118 which involve the spin degree of freedom. In the same spirit, one may now investigate the role of other low-  
119 energy elementary excitations, such as excitons or phonons<sup>16</sup>, which could change the orbital wavefunctions  
120 of nearby atoms and lead to the creation of magnons by a related mechanism. Finally, the new principle of a  
121 symmetry-breaking preparation of low-energy non-thermal states may open unforeseen applications in future  
122 spin-based devices.

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182 **Methods**

183 **Sample.** A single crystal of  $\text{TmFeO}_3$  (grown by a floating zone melting technique) with a thickness of  
184  $\sim 60 \mu\text{m}$  and lateral dimensions of  $\sim 5 \text{mm}$  was used in the experiments. The plate was cut perpendicularly to  
185 one of the crystal's optical axes which lies in the  $(yz)$ -plane at an angle of  $51^\circ$  with respect to the  $z$ -axis. This  
186 orientation allows for THz excitation of both magnon modes at all temperatures. A constant magnetic field of  
187  $0.1 \text{ Tesla}$  saturates the magnetization of the sample.

188

189 **Setup.** The detailed experimental setup is shown in Supplementary Fig. 10: A low-noise titanium:sapphire  
190 laser amplifier (centre wavelength,  $800 \text{ nm}$ ; pulse energy,  $5.5 \text{ mJ}$ ; repetition rate,  $3 \text{ kHz}$ ; pulse duration,  $30 \text{ fs}$ )  
191 is used to generate intense few-cycle THz fields by tilted-pulse-front optical rectification in a cryogenically  
192 cooled  $\text{LiNbO}_3$  crystal. A pair of wire-grid polarizers and different THz spectral filters (see insets to Fig. 4 for  
193 their transmission characteristics) allow for adjusting the peak fields and the spectral shape of the THz pulses.  
194 A small portion of the laser pulses (pulse duration,  $30 \text{ fs}$ ; pulse energy,  $\sim 10 \text{ nJ}$ ) is sent through a mechanical  
195 delay line and is used as a polarization probe. The THz and the probe pulses are collinearly focused onto the  
196  $\text{TmFeO}_3$  sample, which is mounted in a helium cryostat for temperature control. The THz-induced rotation of  
197 the linear polarization of the probe pulses is analysed by polarization optics consisting of a half-wave plate, a  
198 Wollaston polarizer and a pair of balanced silicon photodiodes. A lock-in amplifier is used to record the diode  
199 signals as a function of the time delay between the THz and the probe pulses.

200

201 **Theoretical formalism.**

202 **Model for purely magnetic THz-spin interaction.** We describe the dynamics of the quasi-ferromagnetic mode  
203 of the weak ferromagnet  $\text{TmFeO}_3$  using the Lagrangian  $L$  and Rayleigh  $R$  functions of the angle  $\theta$  of the  
204 normalized antiferromagnetic vector  $\mathbf{G}$  with respect to the crystal axis  $x$  (see Fig. 1a), and its time derivative  
205  $\dot{\theta}$  in a form<sup>31,32</sup>

206

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

207 
$$R = \frac{\alpha M_{\text{Fe}}}{2\gamma} \dot{\theta}^2. \quad (2)$$

208 Here,  $M_{\text{Fe}}$  is the magnetization of the single  $\text{Fe}^{3+}$  sublattice,  $H_{\text{E}}$  is the effective field of the  $d$ - $d$  exchange,  $\alpha$  is  
 209 the Gilbert damping parameter and  $W(\theta)$  is the free energy. For  $\text{TmFeO}_3$  subjected to the magnetic field  $\mathbf{B}$  of  
 210 the THz pulse, one has

211 
$$W(\theta) = K_1 \sin^2 \theta + K_2 \sin^4 \theta - \frac{H_{\text{D}}}{H_{\text{E}}} M_{\text{Fe}} (B_z \cos \theta - B_x \sin \theta), \quad (3)$$

212 where  $H_{\text{D}}$  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,

213  $T_2 \approx 90$  K. Equation (3) accounts for the small spin canting angle  $\varepsilon = \frac{H_{\text{D}}}{H_{\text{E}}}$  which is a result of

214 Dzyaloshinskii-Moriya interaction. In Eq. (3), the term containing the magnetic field arises from Zeeman  
 215 coupling. The equation of motion reads

216 
$$\frac{d}{dt} \left( \frac{\partial L}{\partial \dot{\theta}} \right) - \frac{\partial L}{\partial \theta} + \frac{\partial R}{\partial \dot{\theta}} = 0, \quad (4)$$

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

218 
$$\ddot{\theta} + \omega_{\text{E}} \alpha \dot{\theta} + \omega_{\text{E}} \omega_{\text{A}} w(\theta, T) = \omega_{\text{E}} \gamma \dot{B}_y - \frac{H_{\text{D}}}{H_{\text{E}}} \omega_{\text{E}} \gamma B_z \sin \theta - \frac{H_{\text{D}}}{H_{\text{E}}} \omega_{\text{E}} \gamma B_x \cos \theta, \quad (5)$$

219 where  $\gamma$  is the gyromagnetic ratio,  $\omega_{\text{E}} = \gamma H_{\text{E}}$ ,  $\omega_{\text{A}} = \gamma \frac{K_2}{M_{\text{Fe}}}$  and  $w(\theta, T) = \sin \theta \cos \theta \left( \frac{T - T_2}{T_1 - T_2} + \sin^2 \theta \right)$ .

220 Equation (5) has the form of a generalized sine-Gordon equation and is nonlinear with respect to  $\theta$ . Assuming

221  $\ddot{\theta} = \dot{\theta} = 0$  in Eq. (5) one can find the equilibrium orientation  $\theta_0$  of the antiferromagnetic vector for a given  
 222 temperature as

223 
$$\theta_0 = \begin{cases} \frac{\pi}{2}, & T < T_1 \\ \arcsin \left( \frac{T - T_2}{T_1 - T_2} \right)^{\frac{1}{2}}, & T_1 < T < T_2 \\ 0, & T > T_2. \end{cases} \quad (6)$$

224 The temperature dependence  $\theta_0(T)$  is shown in Fig. 1b and Supplementary Fig. 1. We numerically find the  
 225 solution of Eq. (5) taking the time trace of the magnetic field  $B(t)$  from the experiment (see Fig. 2a) and  
 226 assume standard initial conditions  $\theta(t=0)=\theta_0, \dot{\theta}(t=0)=0$ . For calculations we take  $M_{\text{Fe}} = 1000 \text{ emu cm}^{-3}$ ,  
 227  $H_{\text{E}} = 2 \times 10^7 \text{ Oe}$ ,  $H_{\text{D}} = 2 \times 10^5 \text{ Oe}$  and  $\omega_{\text{E}}\alpha \approx 0.05 \text{ ps}^{-1}$  (Ref. 28). The resulting time evolution of the angle  $\theta(t)$  is  
 228 shown in Supplementary Fig. 6 for different peak amplitudes of the driving magnetic field  $B_{\text{THz}}$  polarized  
 229 along the  $x$ -axis, as in the experiment. While we confirmed the possibility of excitation of the quasi-  
 230 ferromagnetic mode via the Zeeman mechanism, we did not find any deviation from the linear relation  
 231 between the maximum amplitude of  $\theta$  and  $B_{\text{THz}}$  below 0.3 Tesla for any temperature and orientation of the  
 232 field in the  $(xz)$ -plane. Our analysis shows that the Zeeman interaction of the  $\text{Fe}^{3+}$  sublattices with the THz  
 233 magnetic field cannot lead to the nonlinear excitation observed in the experiment.

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

$$237 \quad W_{\text{int}} = \sum_{i,k,l,m} g_{iklm} G_i G_m E_i E_k, \quad (7)$$

238 which is allowed in centrosymmetric  $\text{TmFeO}_3$ . Here,  $g_{iklm}$  are the components of the nonlinear  
 239 magnetoelectric susceptibility tensor  $\hat{\mathbf{g}}$ . The symmetry point group  $D_{2h}^{16}$  of  $\text{TmFeO}_3$  dictates the form of the  
 240 tensor  $\hat{\mathbf{g}}$  which reads (in the Voigt notation)

$$241 \quad \hat{\mathbf{g}} = \begin{pmatrix} g_{11} & g_{12} & g_{13} & 0 & 0 & 0 \\ g_{12} & g_{22} & g_{23} & 0 & 0 & 0 \\ g_{13} & g_{23} & g_{33} & 0 & 0 & 0 \\ 0 & 0 & 0 & g_4 & 0 & 0 \\ 0 & 0 & 0 & 0 & g_5 & 0 \\ 0 & 0 & 0 & 0 & 0 & g_6 \end{pmatrix}. \quad (8)$$

242 Taking into account that the antiferromagnetic vector in  $\text{TmFeO}_3$  lies in the  $(xz)$ -plane, such that  
 243  $\mathbf{G} = (\cos\theta, 0, \sin\theta)$ , we get

$$244 \quad W_{\text{int}} = \cos^2 \theta (g_{11} E_x^2 + g_{12} E_y^2 + g_{13} E_z^2) + \sin^2 \theta (g_{12} E_x^2 + g_{22} E_y^2 + g_{33} E_z^2) + \frac{1}{2} g_5 \sin 2\theta E_x E_z \quad (9)$$

245 which can be rearranged as

$$246 \quad W_{\text{int}} = f(\mathbf{E}) + \sin^2 \theta (\chi_x E_x^2 + \chi_y E_y^2 + \chi_z E_z^2) + \frac{1}{2} g_5 \sin 2\theta E_x E_z, \quad (10)$$

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

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

$$253 \quad W_{\text{int}} = (\chi_y E_y^2 + \chi_z E_z^2) \sin^2 \theta \quad (11)$$

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

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

$$258 \quad \ddot{\theta} + \omega_E \alpha \dot{\theta} + \omega_E \omega_A w(\theta, T) = -\omega_E \omega_A \sin \theta \cos \theta a(t), \quad (12)$$

259 with

$$260 \quad a(t) = \chi_y E_y^2(t) + \chi_z E_z^2(t) \quad (13)$$

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

$$266 \quad \theta(t=0) = \theta_0, \quad \dot{\theta}(t=0) = -\omega_E \omega_A \sin \theta_0 \cos \theta_0 \int_0^{\tau_{\text{THz}}} a(t) dt, \quad (14)$$

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

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

276 **Estimation of the strength of the Zeeman torque.** Applying a THz low-pass filter with a nominal cut-off  
277 frequency of 0.3 THz allows us to selectively excite the q-FM mode (see inset to Fig. 4a). In this  
278 measurement, the q-FM amplitude amounts to  $A_z = 8\%$  of the one induced by the unfiltered THz transients  
279 (Fig. 4a). Taking into account the filter transmission of  $T_F = \sim 75\%$  at the q-FM resonance frequency, we  
280 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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297 **Author contributions**

298 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.  
299 and R.V.M. developed the theoretical model. S.B., M.H., A.V.K., R.H., R.V.M. wrote the manuscript. All  
300 authors discussed the results.

301 **Additional Information**

302 Supplementary information is available in the online version of the paper. Reprints and permissions  
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305 **Competing financial interests**

306 The authors declare no competing financial interests.

## 307 **Figure legends**

308 **Figure 1 | Principle of spin control by a THz induced anisotropy torque. a**, Spin and lattice structure of TmFeO<sub>3</sub>  
309 shown in the  $\Gamma_{24}$  phase. Green/blue spheres: Tm<sup>3+</sup>/Fe<sup>3+</sup> ions. Oxygen atoms are not shown for clarity. The iron spins (blue  
310 arrows) form two antiferromagnetically coupled sublattices  $\mathbf{M}_1$  and  $\mathbf{M}_2$ , which are mutually canted by the  
311 Dzyaloshinskii-Moriya interaction, giving rise to a weak ferromagnetic moment  $\mathbf{F} = \mathbf{M}_1 + \mathbf{M}_2$ . In the  $\Gamma_{24}$  phase (i.e. for  
312  $80 \text{ K} < T < 90 \text{ K}$ ), the antiferromagnetic vector  $\mathbf{G} = \mathbf{M}_1 - \mathbf{M}_2$  encloses a finite angle  $0^\circ < \theta_0 < 90^\circ$  with the  $x$ -axis. **b**,  
313 Spin reorientation phase transitions: In the  $\Gamma_2$  phase (i.e.  $T < 80 \text{ K}$ ) the antiferromagnetic vector  $\mathbf{G}$  is aligned along the  
314 crystallographic  $z$ -axis, whereas it lies along the  $x$ -axis above  $T = 90 \text{ K}$  ( $\Gamma_4$  phase). In the  $\Gamma_{24}$  phase,  $\mathbf{G}$  rotates  
315 continuously in the  $(xz)$ -plane (see also panel **a**). **c**, The crystal field splits the ground state  $^3H_6$  of the rare-earth Tm<sup>3+</sup> ions  
316 into several energy levels with an energy spacing of  $\sim 1 - 10 \text{ meV}$  (schematic level scheme). The corresponding orbital  
317 wavefunctions set the magnetic anisotropy for the iron spins in thermal equilibrium (upper panel). Lower panel: Ultrafast  
318 transitions between these energy levels resonantly induced by THz pulses should exert an abrupt torque on the spins and  
319 act as an efficient trigger for coherent spin dynamics. The small canting angle is not shown for clarity.

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

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

338 **Figure 4 | Control of THz-induced nonlinear torque by spectral shaping: a**, Low pass filtering the pristine THz  
339 spectrum (grey shaded curve in inset) results in the black shaded spectrum (inset) featuring a dominant maximum at the  
340 q-FM resonance frequency. Main graph: Linear scaling of the q-FM amplitudes obtained by Zeeman excitation with the  
341 constrained THz spectrum (red circles). The q-AFM mode (blue triangles) is suppressed since the THz amplitude at the  
342 corresponding frequencies is strongly reduced by the low pass filter. **b**, The Zeeman type excitation of the q-FM mode is

343 switched off by bandpass filtering (centre frequency 1.2 THz) of the THz pulse (black/grey: filtered/pristine spectrum).  
344 Main graph:  $Tm^{3+}$  excitation triggers q-FM oscillations, whose amplitudes scale quadratically with the THz fields (red  
345 circles). Additionally, q-AFM magnons (amplitude: blue triangles) are excited by the Zeeman interaction. All curves are  
346 normalized to the maximum amplitude of the q-FM mode shown in Fig. 3c. Dotted/solid lines are linear/quadratic fits.  
347 Error bars are retrieved as in Fig. 3c.









