

Temporal and spectral fingerprint of ultrafast all-coherent spin switching

S. Schlauderer^{1,*}, C. Lange^{1,*†}, S. Baierl¹, T. Ebnet¹, C. P. Schmid¹, D. C. Valovcin², A. K. Zvezdin^{3,4},
A. V. Kimmel^{5,6}, R. V. Mikhaylovskiy^{6,†,§} and R. Huber¹

¹*Department of Physics, University of Regensburg, Regensburg 93053, Germany.*

²*Department of Physics and the Institute for Terahertz Science and Technology, University of California at Santa Barbara, Santa Barbara, California 93106, USA*

³*Prokhorov General Physics Institute and P.N. Lebedev Physical Institute of the Russian Academy of Sciences, Moscow 119991, Russia.*

⁴*Moscow Institute of Physics and Technology (State University), Dolgoprudny 141700, Russia.*

⁵*Moscow Technological University (MIREA), Moscow 119454, Russia.*

⁶*Radboud University, Institute for Molecules and Materials, Nijmegen 6525 AJ, The Netherlands.*

Future information technology demands ultimately fast, low-loss quantum control. Intense light fields have facilitated important milestones, such as inducing novel states of matter¹⁻³, accelerating electrons ballistically⁴⁻⁷, or coherently flipping the valley pseudospin⁸. These dynamics leave unique signatures, such as characteristic bandgaps or high-order harmonic radiation. The fastest and least dissipative way of switching the technologically most important quantum attribute – the spin – between two states separated by a potential barrier is to trigger an all-coherent precession. Pioneering experiments and theory with picosecond electric and magnetic fields have suggested this possibility⁹⁻¹¹, yet observing the actual dynamics has remained out of reach. Here, we show that terahertz (1 THz = 10¹² Hz) electromagnetic pulses allow coherent navigation of spins over a potential barrier and we reveal the corresponding temporal and spectral fingerprints. This goal is achieved by coupling spins in antiferromagnetic TmFeO₃ with the locally enhanced THz electric field of custom-tailored antennas. Within their duration of 1 ps, the intense THz pulses abruptly change the magnetic anisotropy and trigger a large-amplitude ballistic spin motion. A characteristic phase flip, an asymmetric splitting of the magnon resonance, and a long-lived offset of the Faraday signal are hallmarks of coherent spin switching into adjacent potential minima, in agreement with a numerical simulation. The switchable spin states can be selected by an external magnetic bias. The low dissipation and the antenna’s sub-wavelength spatial definition could facilitate scalable spin devices operating at THz rates.

30 The lowest theoretical limit of energy dissipation for manipulating one bit of information is
31 defined by the Landauer principle¹² as $Q = k_B T \ln 2$, where T is the temperature and k_B denotes the
32 Boltzmann constant. This can be seen as a result of inelastic scattering of a quasiparticle of energy Q ,
33 such as a collective spin excitation, called a magnon. At or below room temperature, Q is of the order
34 of meV, which by the uncertainty principle entails picosecond time scales for minimally dissipative
35 switching. Thus, precessional switching^{10,13,14} triggered by a single-cycle THz pulse with meV photon
36 energies and sub-picosecond duration promises ultimately fast and least-dissipative magnetic control.

37 Experimentally, ultrafast spin control has come a long way¹⁵⁻¹⁷ from the discovery of
38 subpicosecond laser-induced spin dynamics¹⁸ to all-optical non-thermal recording¹⁹. Understanding
39 strongly non-equilibrium spin dynamics triggered by THz pulses, however, is still in its infancy. In anti-
40 tiferromagnets, magnons feature resonance energies in the meV range²⁰ and can be directly addressed
41 by the magnetic field component of intense THz pulses²¹⁻²³. Since the underlying Zeeman interaction
42 is relatively weak, magnetic field amplitudes, which allow for a complete spin reversal have only been
43 reached in linear accelerators⁹, where the spin dynamics have not been accessible on the intrinsic fem-
44 tosecond scale. Also spin transfer torques mediated by THz-driven electric currents have induced
45 switching of antiferromagnetic domains, yet without ultrafast temporal resolution²⁴.

46 Conversely, electromagnons and the more universal coupling of crystal-field split electronic
47 transitions or coherent phonons with the magnetic anisotropy field have allowed the electric THz field
48 component to drive large-amplitude magnons, observed directly in the time domain^{22,25,26}. The
49 available THz peak electric field of 1 MV cm^{-1} , however, has limited the maximum spin excursion far
50 below critical values needed for a complete spin reversal. Meanwhile, the near-field enhancement in
51 custom-tailored antenna structures has been exploited to sculpt atomically strong THz waveforms,
52 sufficient to drive non-perturbative nonlinearities, such as THz-induced phase transitions²⁷ and inter-
53 band Zener tunnelling, with subdiffractive spatial definition²⁸. Such enhancement of the electric field
54 has not yet been utilized for coherent spin control.

55 Here we combine the advantages of electric-field induced anisotropy changes in an antiferro-
56 magnet with the local near-field enhancement of metal antennas. We ballistically steer spins over

57 potential barriers to achieve THz-driven switching between stable states while these dynamics are
58 observed directly on the femtosecond scale. The experiments are performed in high-quality single
59 crystals of the model antiferromagnet TmFeO_3 . The antiferromagnetically ordered Fe^{3+} spins are
60 slightly canted by the Dzyaloshinskii-Moriya interaction, resulting in a net ferromagnetic moment. As
61 the magnetic anisotropy depends on temperature²⁶, the spins undergo reorientation phase transitions at
62 $T_1 = 80$ K and $T_2 = 90$ K. The anisotropy may also be modified by THz electric dipole transitions be-
63 tween crystal field-split states of the electronic ground state of the Tm^{3+} ions, the angular momenta of
64 which are coupled with the Fe^{3+} spins by exchange and dipolar interactions²⁹. Our idea is to abruptly
65 change the magnetic anisotropy by sufficiently strong THz pulses causing the spins to switch fully
66 ballistically.

67 We fabricate custom-tailored bowtie antennas of gold (feed gap, $3.5 \mu\text{m}$) onto a $60\text{-}\mu\text{m}$ -thick
68 single crystal of TmFeO_3 (Extended Data Figure 1) to bypass the diffraction limit and maximize the
69 achievable THz amplitude. The design was guided by numerical finite-difference frequency-domain
70 simulations optimizing the near-field enhancement at a frequency of 0.65 THz (see Methods), which is
71 resonant with crystal field-split ground state transitions in Tm^{3+} . In a pump-probe scheme (Fig. 1a), an
72 intense THz transient with tuneable far-field amplitudes of up to $E_{\text{THz}} = 1.0 \text{ MV cm}^{-1}$ (see Methods)
73 excites the structure from the TmFeO_3 back side. The ensuing spin dynamics are probed via the
74 polarisation rotation, θ , imprinted on a co-propagating femtosecond near-infrared pulse (wavelength,
75 807 nm ; pulse duration, 33 fs) by the Faraday effect and magnetic linear dichroism. Our quantitative
76 simulation shows that, for the strongest electro-optically detected THz waveform, the near-field of the
77 antenna, E_{NF} , readily exceeds 9 MV cm^{-1} in the centre of the gap (Fig. 1b).

78 To test the efficiency of the antenna, we compare the magneto-optical signal induced in TmFeO_3
79 in the transition phase ($T = 83 \text{ K}$) with and without the near-field antenna, as a function of the pump-
80 probe delay time, t . In the absence of an antenna, a THz pulse with an amplitude of $E_{\text{THz}} =$
81 1.0 MV cm^{-1} abruptly sets off coherent magnon oscillations, which decay exponentially within 40 ps
82 (Fig. 1c, black curve). The signal consists of a superposition of two frequency components centred at
83 0.09 THz and 0.82 THz (inset of Fig. 1c) – the quasi-ferromagnetic (q-fm) and the quasi-antiferro-
84 magnetic (q-afm) mode²⁶, respectively. The maximum rotation angle of the probe polarisation of

85 0.5 mrad corresponds to a magnetisation deflection by 3.5° (see Methods). In contrast, we observe a
86 qualitatively different response when the probe pulse is positioned in the centre of the antenna feed
87 gap. Here a polarisation rotation as high as 0.9 mrad is reached for a much weaker THz far-field of
88 $E_{\text{THz}} = 0.4 \text{ MV cm}^{-1}$ (Fig. 1c, blue curve). In addition, the relative spectral amplitude of the q-fm mode
89 is significantly enhanced, whereas the amplitude of the q-afm mode is suppressed. This behaviour is
90 expected since the q-fm mode is excited by the antenna-enhanced THz electric field component,
91 whereas the q-afm magnon can only be launched by Zeeman coupling to the THz magnetic field²⁶,
92 which is not enhanced in the feed gap.

93 The amplitude of the q-fm magnon is remarkably high given that the field enhancement is
94 spatially confined to the evanescent near-field region (depth, $\sim 13 \mu\text{m}$) whereas the magneto-optical
95 signal in the antenna-free case (Fig. 1c, black curve) originates from the entire thickness ($60 \mu\text{m}$) of
96 the TmFeO_3 substrate. A rough estimate (see Methods) shows that the spins in the antenna gap need to
97 undergo a rotation by as much as 24° in order to explain the observed signal strength. Hence, a further
98 increase of the incident THz field may be able to cause complete spin switching.

99 Figure 2a shows the ultrafast polarisation rotation probed in the feed gap, for various far-field
100 THz amplitudes between $E_{\text{THz}} = 0.15 \text{ MV cm}^{-1}$ and 1.0 MV cm^{-1} . For the lowest field, the spin
101 dynamics resembles the q-fm precession sampled in the unstructured crystal (Fig. 1c, black curve). For
102 increasing fields, the oscillation amplitude grows. When the incident THz field exceeds
103 $E_{\text{THz}} = 0.75 \text{ MV cm}^{-1}$, a qualitatively new behaviour emerges. The period of the first full cycle of the
104 magnetisation oscillation is distinctly stretched (see vertical dashed line in Fig. 2a) while a pronounced
105 beating feature occurs in the coherent polarisation rotation signal, seen during $25 \text{ ps} < t < 35 \text{ ps}$.
106 Simultaneously, a long-lived offset of the Faraday signal develops (Fig. 2b, red shaded area). In the
107 frequency domain (Fig. 2c), these novel dynamics are associated with an asymmetric splitting of the q-
108 fm magnon resonance superimposed on a broad spectral distribution, somewhat reminiscent of the
109 spectral fingerprint of carrier-wave Rabi oscillations³⁰. The long-lived offset (Fig. 2b) manifests itself
110 in a dc spectral component, which grows more rapidly for $E_{\text{THz}} > 0.75 \text{ MV cm}^{-1}$ (Fig. 2d and Extended
111 Data Figure 2). We will show next that the stretching of the first oscillation cycle, the beating of the
112 Faraday signal, and the spectral splitting of the magnon resonance are hallmarks of all-coherent non-

113 perturbative spin trajectories between adjacent minima of the magnetic potential energy, whereas the
114 long-lived offset directly reads out the switched spin state.

115 The dynamics can best be understood by starting out with the magnetic structure of TmFeO_3
116 (Fig. 3a). The slight canting between the magnetisations \mathbf{M}_1 and \mathbf{M}_2 of the two antiferromagnetic
117 sublattices causes a weak ferromagnetic moment $\mathbf{F} = \mathbf{M}_1 + \mathbf{M}_2$ in the x - z -plane. The antiferromagnetic
118 vector $\mathbf{G} = \mathbf{M}_1 - \mathbf{M}_2$ encloses an angle ϕ with the x -axis. In the Γ_{24} transition phase ($T_1 < T < T_2$),
119 ϕ shifts continuously between 0° and 90° as the magnetic potential $W(\phi)$ changes with the thermal
120 population of the Tm^{3+} crystal field-split states²⁶. $W(\phi)$ features four intrinsically degenerate minima.
121 To ensure that the pump-probe experiment starts with the same equilibrium spin orientation angle ϕ_0
122 for every laser shot, we apply a weak external magnetic field $B_{\text{ext}} = 100$ mT (see Methods).

123 When the intense THz near-field excites the Tm^{3+} ions, it abruptly modifies $W(\phi)$, shifting both
124 the position, ϕ_0 , and the depth of the potential minimum (Fig. 3b, inset). These non-adiabatic changes
125 give rise to a displacive and an impulsive anisotropy torque, which initiate coherent magnetisation
126 dynamics as described by the generalized sine-Gordon equation (see Methods). Figure 3b illustrates
127 two typical spin trajectories. For a peak near-field of $E_{\text{NF}} = 6$ MV cm⁻¹, the spins carry out a coherent
128 oscillation about ϕ_0 . A field of $E_{\text{NF}} = 10$ MV cm⁻¹, in contrast, allows the spins to overcome the
129 potential barrier at $t = 3.4$ ps, and relax into a new equilibrium position ϕ_1 , corresponding to a spin
130 rotation by $\sim 90^\circ$. While crossing the potential maximum the spins acquire a characteristic phase,
131 which causes a retardation by $\sim 180^\circ$ with respect to spin oscillations in the initial potential minimum,
132 seen at $t = 9.7$ ps (Fig. 3b, red solid line). Once the spins have reached their maximum positive
133 deflection they oscillate back, but do not overcome the potential barrier a second time because of
134 damping. They rather stay within the new minimum and, in a strongly anharmonic motion, accumulate
135 more phase retardation such that the red and blue trajectories in Fig. 3b oscillate in phase again,
136 around $t \approx 25$ ps.

137 To link these dynamics with the measured polarisation rotation, we calculate the expected
138 Faraday signal by projecting the ferromagnetic moment $\mathbf{F}(\phi)$ onto the wave vector of the near-infrared
139 probe pulse, \mathbf{k}_{NIR} (see Fig. 3a). By superimposing the contributions of the two spin trajectories in

140 Fig. 3b, the pronounced beating feature ($t \approx 25$ ps) can be clearly associated with the phase slip
 141 occurring during spin switching (see Extended Data Figure 3). For a quantitative analysis, we combine
 142 our calculation of the near-field induced by the experimental THz wave with a numerical solution of
 143 the local generalized sine-Gordon equation (see Methods). We then weigh the locally induced Faraday
 144 signal by the Gaussian intensity distribution of the probe beam and sum all the microscopic
 145 contributions from the probed volume. Figure 3c shows the calculated polarisation rotations, θ , for
 146 $E_{\text{THz}} = 0.4 \text{ MV cm}^{-1}$ and 1.0 MV cm^{-1} . All experimental features are quantitatively reproduced,
 147 including the quasi-monochromatic magnon oscillation, for low fields (Fig. 3c, blue curve), as well as
 148 the phase retardation of the first magnon oscillation period and the pronounced beating at $t \approx 25$ ps, at
 149 large THz fields (Fig. 3c, red curve). Moreover, the model unambiguously connects the asymmetric
 150 splitting of the q-fm resonance and the broad low-frequency components (Fig. 3d) to THz-driven all-
 151 coherent spin switching. The calculation also proves that the switched spins can be directly read out.
 152 As seen in Fig. 3e, increasing E_{THz} leads to a long-lived signal offset, caused by two distinct
 153 mechanisms: (i) THz excitation of Tm^{3+} ions slightly shifts the position of the potential minimum (Fig.
 154 3b, inset). (ii) A transfer of spins over the barrier can also change the net magneto-optical signal if \mathbf{k}_{NIR}
 155 is tilted out of the y - z -plane (Fig. 3a). In our experiment, we estimate a tilt angle of $\sim 1^\circ$. Whereas the
 156 offset caused by the shift of the magnetic potential grows slowly with E_{THz} (Fig. 3e, red circles), the
 157 slope of the long-lived Faraday signal (Fig. 3e, red spheres) increases rapidly above the switching
 158 threshold $E_{\text{THz}} > 0.75 \text{ MV cm}^{-1}$, as seen in the experiment (Fig. 2d). This steep increase is thus a direct
 159 way of reading out the switched spin population.

160 Based on the microscopic understanding of the spin dynamics, we can shape the spin trajectory
 161 by tailoring the magnetic potential. As a first control parameter (see Extended Data Figure 4), we
 162 lower the temperature to $T = 82.5 \text{ K}$, where the barrier height, w , is slightly increased (Fig. 4a).
 163 Consequently, the switching dynamics are decelerated and the beating signature is delayed to $t = 45$ ps
 164 (Fig. 4e, top curve). Meanwhile, the spectrum remains qualitatively similar (Fig. 4f, top curve). The
 165 barrier height can also be raised by rotating the external magnetic bias field, \mathbf{B}_{ext} , by an angle $\alpha = 15^\circ$
 166 about the optical axis (Fig. 4b and Extended Data Figure 1), resulting in a shift of the beating feature
 167 to a delay of $t = 55$ ps (Fig. 4e). Thereby, the potential shoulder at $\phi = -115^\circ$ is lowered (Fig. 4b),

168 which enables large-amplitude oscillations throughout a slightly wider potential trough, causing a
169 weak red-shift of the spectrum (Fig. 4f). For $\alpha = 60^\circ$ (Fig. 4c), the dynamics are strongly altered
170 (Figs. 4e, f). After the spins are driven up the potential barrier at $\phi = 0^\circ$ during the first half cycle, the
171 non-switching spins oscillate back through the wide potential minimum that is extended by the
172 shoulder at $\phi = -115^\circ$. This results in a strong red-shift of the centre frequency to 50 GHz. On the
173 potential shoulder, the projection $\mathbf{F} \cdot \mathbf{k}_{\text{NIR}}$ drops below its initial value at ϕ_0 (Extended Data Figure 5),
174 leading to a transient negative offset of the Faraday signal (Fig. 4e, dashed-dotted line, and Extended
175 Data Figure 6) until the oscillations of the unswitched spins have decayed within the starting local
176 potential minimum. Still, a sufficiently large fraction of spins reach the target valley (grey sphere) for
177 beating to be observed. Finally, $\alpha = 95^\circ$ sets a new starting position and direction of acceleration (Fig.
178 4d, violet sphere and arrow), causing a reversal of the transient polarization rotation signal and offset
179 (Fig. 4e and Extended Data Figure 5). The wide potential minimum leads to a reduced centre
180 frequency reproduced by calculating the single spin dynamics (Fig. 4f, black arrows). The large barrier
181 to the neighbouring valley (grey circle) inhibits switching completely and no beating is observed.

182 The unprecedented phase slip, the asymmetric spectral splitting, and the long-lived offset in the
183 magneto-optical response occurring above a well-defined threshold peak field are the fingerprints of
184 ballistic spin switching, marking a novel regime of ultrafast all-coherent spin control throughout the
185 entire phase space. In our specific implementation of a THz-driven anisotropy torque, the absorption
186 of approximately one THz photon energy per spin suffices for switching whereas the energy
187 dissipation within the spin system remains below 1 μeV per spin (see Methods). This scheme is, thus,
188 highly scalable. Future storage devices could exploit the excellent spatial definition of antenna
189 structures (Extended Data Figure 7) to switch magnetic bits of a diameter of 10 nm with THz energies
190 of less than 1 attojoule. Owing to the absence of magnetic stray fields, these cells could be densely
191 packed, similar to vortex core structures in ferromagnetic thin films¹⁴. The readout of the spin state
192 could be combined with spintronic approaches^{20,24}. Such optimized antennas with nanoscale gaps pro-
193 viding field enhancement factors of 10^4 and more may be driven by all-electronic on-chip THz

194 sources, enabling practical implementations of novel spin memories operating at THz clock rates, and
195 ultimately low dissipation.

196

197 * These authors contributed equally to this work.

198 § Current location: Department of Physics, Lancaster University, Bailrigg, Lancaster, LA1 4YB ,UK.

199

200 **References**

- 201 1. Wang, Y.H., Steinberg, H., Jarillo-Herrero, P. & Gedik, N. Observation of Floquet-Bloch
202 States on the Surface of a Topological Insulator. *Science* **342**, 453–457 (2013).
- 203 2. Matsunaga, R. *et al.* Light-induced collective pseudospin precession resonating with Higgs
204 mode in a superconductor. *Science* **345**, 1145–1149 (2014).
- 205 3. Mitrano, M. *et al.* Possible light-induced superconductivity in K_3C_{60} at high temperature.
206 *Nature* **530**, 461–464 (2016).
- 207 4. Hohenleutner, M. *et al.* Real-time observation of interfering crystal electrons in high-harmonic
208 generation. *Nature* **523**, 572–575 (2015).
- 209 5. Garg, M. *et al.* Multi-petahertz electronic metrology. *Nature* **538**, 359–363 (2016).
- 210 6. Sivis, M. *et al.* Tailored semiconductors for high-harmonic optoelectronics.
211 *Science* **357**, 303–306 (2017).
- 212 7. Higuchi, T., Heide, C., Ullmann, K., Weber, H.B. & Hommelhoff, P. Light-field-driven
213 currents in graphene, *Nature* **550**, 224–228 (2017).
- 214 8. Langer, F. *et al.* Lightwave valleytronics in a monolayer of tungsten diselenide.
215 *Nature* **557**, 76–80 (2018).
- 216 9. Back, C. H. *et al.* Minimum Field Strength in Precessional Magnetization Reversal.
217 *Science* **285**, 864–867 (1999).
- 218 10. Tudosa, I. *et al.* The ultimate speed of magnetic switching in granular recording media. *Nature*
219 **428**, 831–833 (2004).

- 220 11. Kim, T.H., Grünberg, P., Han, S.H. & Cho, B.K. Precessional switching of antiferromagnets
221 by electric field induced Dzyaloshinskii-Moriya torque, *Phys. Rev. B* **97**, 184427 (2018).
- 222 12. Landauer, R. Irreversibility and heat generation in the computing process. *IBM Journal of*
223 *Research and Development* **5**, 183–191 (1961).
- 224 13. Madami, M., Chiuchiù, D., Carlotti, G. & Gammaitoni, L. Fundamental energy limits in the
225 physics of nanomagnetic binary switches. *Nano Energy* **15**, 313–320 (2015).
- 226 14. Van Waeyenberge, B. *et al.* Magnetic vortex core reversal by excitation with short bursts of an
227 alternating field. *Nature* **444**, 461–464 (2006).
- 228 15. Kimel, A. V., Kirilyuk, A., Tsvetkov, A., Pisarev, R. V. & Rasing, T. Laser-induced ultrafast
229 spin reorientation in the antiferromagnet TmFeO₃. *Nature* **429**, 850–853 (2004).
- 230 16. Kimel, A.V. *et al.* Inertia-driven spin switching in antiferromagnets.
231 *Nature Phys.* **5**, 727–731 (2009).
- 232 17. Li, T. *et al.* Femtosecond switching of magnetism via strongly correlated spin–charge
233 quantum excitations. *Nature* **496**, 69–73 (2013).
- 234 18. Beaurepaire, E., Merle, J.-C., Daunois, A. & Bigot, J.-Y. Ultrafast spin dynamics in
235 ferromagnetic nickel. *Phys. Rev. Lett.* **76**, 4250–4253 (1996).
- 236 19. Stupakiewicz, A., Szerenos, K., Afanasiev, D., Kirilyuk, A. & Kimel, A. V. Ultrafast
237 nonthermal photo-magnetic recording in a transparent medium. *Nature* **542**, 71–74 (2017).
- 238 20. Jungwirth, T., Marti, X., Wadley, P. & Wunderlich, J. Antiferromagnetic spintronics. *Nature*
239 *Nanotech.* **11**, 231–241 (2016).
- 240 21. Kampfrath, T. *et al.* Coherent terahertz control of antiferromagnetic spin waves.
241 *Nature Photon.* **5**, 31–34 (2011).
- 242 22. Nova, T.F. *et al.* An effective magnetic field from optically driven phonons.
243 *Nature Phys.* **13**, 132–136 (2017).
- 244 23. Lu, J., Li, X., Hwang, H.Y., Ofori-Okai, B.K., Kurihara, T., Suemoto, T. & Nelson, K.A.
245 Coherent Two-Dimensional Terahertz Magnetic Resonance Spectroscopy of Collective Spin
246 Waves. *Phys. Rev. Lett.* **118**, 207204 (2017).

- 247 24. Olejník, K. *et al.* Terahertz electrical writing speed in an antiferromagnetic memory. *Science*
248 *Advances* **4**, eaar3566 (2018).
- 249 25. Kubacka, T. *et al.* Large-amplitude spin dynamics driven by a THz pulse in resonance with an
250 electromagnon. *Science* **343**, 1333–1336 (2014).
- 251 26. Baierl, S. *et al.* Nonlinear spin control by terahertz-driven anisotropy fields.
252 *Nature Photon.* **10**, 715–718 (2016).
- 253 27. Liu, M. *et al.* Terahertz-field-induced insulator-to-metal transition in vanadium dioxide
254 metamaterial. *Nature* **487**, 345–348 (2012).
- 255 28. Lange, C. *et al.* Extremely nonperturbative nonlinearities in GaAs driven by atomically strong
256 terahertz fields in gold metamaterials. *Phys. Rev. Lett.* **113**, 227401 (2014).
- 257 29. Staub, U. *et al.* Interplay of Fe and Tm moments through the spin-reorientation transition in
258 TmFeO₃. *Phys. Rev. B* **96**, 174408 (2017).
- 259 30. Mücke, O.D., Tritschler, T., Wegener, M., Morgner, U. & Kärtner, F.X. Signatures of Carrier-
260 Wave Rabi Flopping in GaAs. *Phys. Rev. Lett.* **87**, 057401 (2001).

261

262 **Acknowledgement.** The authors thank A. M. Balbashov for bulk crystals of orthoferrites of an
263 exceptionally high quality, J. Fabian and M. S. Sherwin for fruitful discussions and Th. Rasing for
264 continuous support. The work in Regensburg was supported by the DFG through grant no. HU 1598/2
265 and SFB 1277 (Project A05) as well as by the European Research Council through grant no. 305003
266 (QUANTUMsubCYCLE). The work in Nijmegen was supported by the European Research Council
267 ERC Grant Agreement No. 339813 (Exchange) and NWO (The Netherlands Organization for
268 Scientific Research).

269 **Author Contributions** S.S., C.L., S.B., and R.H. designed and implemented the antenna structures.
270 R.V.M. and A.V.K. identified the bulk material for the project. S.S., C.L., S.B., T.E., C.P.S. and
271 D.C.V. carried out the experiment with support from R.V.M.. The theoretical modelling was carried
272 out by C.L., S.S., S.B., A.K.Z., and R.V.M.. A.V.K. and R.H. supervised the study. All authors
273 analysed the data, discussed the results, and contributed to the writing of the manuscript.

274 **Author Information** Reprints and permissions information is available at www.nature.com/reprints.
275 The authors declare no competing financial interests. Correspondence and requests for materials
276 should be addressed to C.L. (christoph.lange@ur.de) or R.V.M. (r.mikhaylovskiy@lancaster.ac.uk).

277 **Figure 1 | Antenna-enhanced THz spin dynamics.** **a**, Schematic of the gold bowtie antenna on TmFeO₃. The
278 structure is excited from the back side by an intense THz electric field E_{THz} (red waveform) while a co-
279 propagating near-infrared pulse ($h\nu_{\text{probe}}$, light blue) probes the induced magnetisation dynamics in the centre of
280 the feed gap. **b**, Peak near-field amplitude, E_{NF} , in the antenna feed gap calculated by finite-difference
281 simulations for a real THz waveform with a peak field amplitude of $E_{\text{THz}} = 1.0 \text{ MV cm}^{-1}$ (see Extended Data Fig.
282 1c). **c**, Experimentally detected polarisation rotation signal as a function of the delay time, t , obtained for a peak
283 electric THz field of $E_{\text{THz}} = 1.0 \text{ MV cm}^{-1}$ on the unstructured substrate (black curve) and when probing the gap
284 of the bowtie antenna structure resonantly excited by a THz waveform with a peak electric far-field amplitude of
285 $E_{\text{THz}} = 0.4 \text{ MV cm}^{-1}$ (blue curve, vertically offset by 1 mrad for better visibility). Inset: Corresponding amplitude
286 spectra featuring two modes at 0.09 THz and 0.82 THz. The sample was kept at a lattice temperature of
287 $T = 83 \text{ K}$.

288

289 **Figure 2 | THz-induced nonlinear spin dynamics.** **a**, Polarisation rotation probed in the centre of the antenna
290 feed gap for various far-field amplitudes, as a function of the delay time, t . For incident THz peak fields $E_{\text{THz}} >$
291 0.75 MV cm^{-1} , the quasi-monochromatic oscillation is strongly distorted by a phase slip at delay times between
292 25 and 35 ps. The measurements are offset and scaled as indicated for clarity. Lattice temperature $T = 83 \text{ K}$. **b**,
293 Long-term evolution of the polarisation rotation for a THz peak field of $E_{\text{THz}} = 1.0 \text{ MV cm}^{-1}$. The red-shaded
294 area indicates the long-lived offset. **c**, Spectral amplitude of the time-domain data shown in **a**. The phase slip in
295 the polarisation rotation signal for highest THz fields manifests itself in a splitting of the q-fm resonance. **d**,
296 Spectral amplitude of the dc offset, $A_{0 \text{ THz}}$, as a function of the THz far-field peak amplitude, E_{THz} . $A_{0 \text{ THz}}$
297 increases monotonically with the THz field. Grey-shaded area: Spin-switching regime with increased slope of $A_{0 \text{ THz}}$.
298 Dashed lines, guides to the eye.

299

300 **Figure 3 | Microscopic picture of ballistic spin motion.** **a**, Spin and lattice structure of TmFeO₃ in the Γ_{24}
301 phase ($T_1 < T < T_2$), showing the Fe³⁺ spins (dark blue spheres and arrows), Tm³⁺ ions (orange spheres), and the
302 ferromagnetic moment, \mathbf{F} (violet arrow). The antiferromagnetic vector \mathbf{G} (brown arrow) lies in the x-z-plane and
303 encloses a finite angle of $0 < \phi < 90^\circ$ with the x-axis. Inset: geometry of the wave vector of the probe pulse, \mathbf{k}_{NIR}
304 (light blue arrow), with respect to \mathbf{F} and the external magnetic field \mathbf{B}_{ext} (grey arrow). **b**, Numerical simulation of
305 THz-induced ballistic spin dynamics. Upon THz excitation, the magnetic potential $W(\phi)$ is abruptly modified

306 near a delay time of $t = 0$ ps (magnified in inset). Near-field THz transients with peak amplitudes of $E_{\text{NF}} = 6$
 307 MV cm^{-1} abruptly induce large-amplitude spin oscillations within the same potential valley around the initial
 308 angle ϕ_0 (blue trajectory). For a THz near-field of $E_{\text{NF}} = 10 \text{ MV cm}^{-1}$, the spins reach the adjacent local minimum
 309 (red trajectory) at ϕ_1 , where $\phi_1 \approx \phi_0 + 90^\circ$, accumulating a phase retardation relative to spins oscillating around ϕ_0
 310 (delay times $t = 9.7$ ps and 27.2 ps, respectively; red cuts through the magnetic potential). **c**, Calculated
 311 polarisation rotation in the antenna feed gap for an incident THz electric peak field amplitude of $E_{\text{THz}} = 0.4 \text{ MV}$
 312 cm^{-1} (blue curve) and $E_{\text{THz}} = 1.0 \text{ MV cm}^{-1}$ (red curve) as a function of the delay time, t , for a lattice temperature
 313 of $T = 83$ K, normalized to the experimental peak value. The experimental data are plotted as circles. **d**,
 314 Amplitude spectra of the time-domain data shown in **c**. **e**, Calculated scaling of the spectral amplitude of the
 315 long-lived offset, $A_{0 \text{ THz}}$, for no misalignment (red circles) and a misalignment angle of the near-infrared **k**-vector
 316 out of the y - z -plane of 1.25° (red spheres). In the spin-switching regime ($E_{\text{THz}} \geq 0.75 \text{ MV cm}^{-1}$, grey-shaded area)
 317 the calculations reproduce the increased slope of $A_{0 \text{ THz}}$ observed in the experiment (Fig. 2d). Dashed lines,
 318 guides to the eye.

319

320 **Figure 4 | Ballistic navigation of spins.** **a-d**, Magnetic potential $W(\phi)$ for a lattice temperature of $T = 82.5$ K
 321 and various orientations α of the static external magnetic bias, \mathbf{B}_{ext} . w , potential barrier height relevant for
 322 switching; black arrows, potential shoulder associated with the red-shift. Violet spheres and arrows: initial spin
 323 orientation and direction after excitation; grey spheres, final orientation of switched spins. **e**, Polarization
 324 rotation as a function of the delay time, t , for the potentials shown in **a-d** and a THz peak far-field amplitude of
 325 $E_{\text{THz}} = 1.0 \text{ MV cm}^{-1}$. Dashed-dotted curve: transient negative polarization rotation (see text). **f**, Amplitude spectra
 326 of the time-domain data shown in **e**. The black arrows mark the computed centre frequencies.

327 **Methods**

328

329 **Sample preparation.** We used a monocrystalline, 60- μm -thick TmFeO_3 sample obtained by floating-
330 zone melting. The sample was cut perpendicularly to one of the crystal's optical axes, which lies in the
331 y - z -plane at an angle of 51° with respect to the z -axis. The custom-tailored THz antennas with a feed
332 gap of $3.5\ \mu\text{m}$ and a resonance frequency of $0.65\ \text{THz}$ were processed on top of the crystal by
333 electron-beam lithography of a poly(α -methylstyrene-co- α -chloracrylate methylester) resist,
334 subsequent evaporation of $100\ \text{nm}$ of gold, and lift-off. The structure was kept in a helium cryostat and
335 cooled to temperatures within the Γ_{24} transition phase. For the measurements discussed in the first part
336 of the manuscript, a static bias field of $B_{\text{ext}} = 100\ \text{mT}$ from a permanent magnet was applied within the
337 y - z -plane of the crystal at an angle of 39° relative to the z -axis, defining the equilibrium spin
338 orientation ϕ_0 and ensuring the restoring of the magnetisation between subsequent laser pulses. For the
339 data shown in Fig. 4, the \mathbf{B} -field was rotated about the optical axis of the crystal, whereby an angle of
340 $\alpha = 0^\circ$ denotes the starting position within the y - z -plane as defined above.

341 **Experimental setup.** Intense single-cycle THz pulses were generated by tilted-pulse front optical
342 rectification of near-infrared pulses from a low-noise Ti:sapphire laser amplifier (centre wavelength,
343 $807\ \text{nm}$; pulse energy, $5.5\ \text{mJ}$; pulse duration, $33\ \text{fs}$; repetition rate, $3\ \text{kHz}$) in a cryogenically cooled
344 LiNbO_3 crystal (Extended Data Figure 1b). A pair of wire-grid polarisers were used to control the peak
345 field strength and the polarisation state of the THz waveforms. Extended Data Figure 1c and d show
346 the THz transient and the corresponding spectrum featuring frequency components between 0.3 and
347 $2.5\ \text{THz}$. A small portion of the near-infrared power was sent through a delay line, combined with the
348 THz pulse using a fused silica window coated with indium tin oxide, and collinearly transmitted
349 through the feed gap of the antenna structure to probe the magnetisation state. The polarisation
350 rotation was measured by subsequent optics consisting of a half-wave plate, a Wollaston prism, and
351 two balanced silicon photodiodes, read out by a lock-in amplifier.

352 **Estimate of the spin switching energy.** The Poynting theorem dictates that the absorbed electro-
353 magnetic power density $P(t)$ is given by

354
$$P(t) = j(t) E(t), \quad (1)$$

355 where $j(t)$ is the effective current density describing dissipative processes in a material and $E(t)$ is the
 356 oscillating electric field. The full energy absorbed per unit volume is therefore

357
$$W_{\text{abs}} = \int_{-\infty}^{\infty} j(t) E(t) dt. \quad (2)$$

358 By taking the Fourier transforms $j(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \tilde{j}(\omega) e^{i\omega t} d\omega$ and $E(t) = \frac{1}{2\pi} \int_{-\infty}^{\infty} \tilde{E}(\omega') e^{i\omega' t} d\omega'$,
 359 where ω is the frequency, and substituting them into Eq. (2) we obtain

360
$$W_{\text{abs}} = \frac{1}{2\pi} \int_{-\infty}^{\infty} \tilde{j}(\omega) \tilde{E}(-\omega) d\omega. \quad (3)$$

361 The current density is connected to the electric field by the effective conductivity $\sigma(\omega) = \frac{\tilde{j}(\omega)}{\tilde{E}(\omega)}$ so as

362
$$W_{\text{abs}} = \frac{1}{2\pi} \int_{-\infty}^{\infty} \sigma(\omega) \tilde{E}(\omega) \tilde{E}(-\omega) d\omega = \frac{1}{2\pi} \int_{-\infty}^{\infty} \sigma(\omega) |\tilde{E}(\omega)|^2 d\omega. \quad (4)$$

363 In the case of crystal-field split ground state transitions of TmFeO_3 in the temperature interval between
 364 80 K and 90 K, where the imaginary part of the dielectric function ε_2 is much smaller than its real part
 365 ε_1 (see Ref. 31), the effective conductivity can be approximated by $\sigma = \varepsilon_0 n_{\text{sub}} c \alpha_{\text{eff}}$. Here $n_{\text{sub}} = 4.92$
 366 is the refractive index of TmFeO_3 , and $\alpha_{\text{eff}} \approx 4000 \text{ m}^{-1}$ is the effective THz absorption coefficient
 367 obtained from data of Ref. 31, taking into account the spectral shape of our THz pulse. We obtain

368
$$W_{\text{abs}} = \frac{1}{2\pi} \varepsilon_0 n_{\text{sub}} c \alpha_{\text{eff}} \int_{-\infty}^{\infty} |\tilde{E}(\omega)|^2 d\omega, \quad (5)$$

369 which can be rewritten in the time domain (compare Eqs. (2) and (3)) as

370
$$W_{\text{abs}} = \varepsilon_0 n_{\text{sub}} c \alpha_{\text{eff}} \int_{-\infty}^{\infty} E^2(t) dt. \quad (6)$$

371 The absorbed energy density in the rare-earth system for a near-field THz transient with a peak electric
 372 field of 7.8 MV cm^{-1} , which exceeds the threshold for spin switching, is $W_{\text{abs}} = 20 \text{ J cm}^{-3}$. TmFeO_3
 373 crystallises in a distorted perovskite structure with a unit cell volume of $V_{\text{uc}} = 2.22 \times 10^{-28} \text{ m}^3$ (lattice
 374 constants, $a = 525 \text{ pm}$, $b = 557 \text{ pm}$, and $c = 758 \text{ pm}$) (see Ref. 32), which contains 4 Fe^{3+} spins. Thus,
 375 an upper bound for the absorbed energy in the rare-earth system per spin is given by $W_{\text{spin}} = W_{\text{abs}} \times$
 376 $\frac{V_{\text{uc}}}{4} = 7.15 \text{ meV}$, which is on the order of the energy of one THz photon. The dissipation by the spin

377 system is even smaller: The energy required to overcome the potential barrier, separating two
378 neighbouring potential minima (see Fig. 3b), normalized by the number of spins in the switched
379 volume is less than 1 μeV . This value can, thus, be regarded as an upper limit for the maximal energy
380 dissipated by one spin upon switching.

381 **Estimate of the magnetisation deflection in the near-field volume.** In the case of unstructured bulk
382 TmFeO_3 , the total polarisation rotation, θ , results from approximately equal contributions across the
383 entire sample thickness of 60 μm . In order to calibrate the relation between θ and the spin angle ϕ , we
384 enforce a full switching of the magnetisation (change of ϕ by 180°) by reversing the external static
385 magnetic bias field. This scenario rotates the probe polarisation by 24 mrad. Thus, we conclude that a
386 polarisation rotation of $\theta = 0.5$ mrad, as induced by a THz amplitude of 1.0 MV cm^{-1} in the antenna-
387 free sample, corresponds to a transient spin excursion of $\Delta\phi = 3.5^\circ$. Taking into account the quadratic
388 dependence of $\Delta\phi$ on the electric field amplitude²⁶, we link the polarisation rotation to the THz peak
389 electric field by $\theta = \xi \times L \times \bar{E}_{\text{peak}}^2$, where L is the crystal length, $\xi = 472 \text{ mrad cm (MV)}^{-2}$ is the
390 coupling constant, and $\bar{E}_{\text{peak}} = 0.42 \text{ MV cm}^{-1}$ is the peak electric THz amplitude averaged over the
391 length of the unstructured TmFeO_3 sample. In the antenna-covered structure, the magneto-optical
392 signal can be divided into two contributions: the antenna near-field region extending down to a depth
393 of 13 μm below the antenna (Extended Data Figure 8, red-shaded area), where electric fields strongly
394 exceeding the far-field amplitude are encountered, and a bulk part (Extended Data Figure 8, blue-
395 shaded area), where the electric field assumes an average value of 0.3 MV cm^{-1} . Accordingly, the
396 polarisation rotation by the bulk part is $\theta_b = \xi \times 47 \mu\text{m} \times (0.3 \text{ MV cm}^{-1})^2 = 0.2$ mrad, such that
397 0.7 mrad of the total magneto-optical signal result from the near-field volume. This contribution
398 corresponds to an average spin deflection angle of $\Delta\phi = 24^\circ$.

399 **Numerical calculation of antenna response.** The THz response of the entire structure, including the
400 near-field of the custom-tailored antenna as well as the substrate, was obtained by solving Maxwell's
401 equations using a finite-difference frequency-domain (FDFD) approach. The refractive index of
402 TmFeO_3 is set to $n_{\text{sub}} = 4.92$, while the gold structure is implemented as a perfect metal. The THz near-
403 field waveforms were subsequently calculated based on the measured far-field THz waveform,

404 employing the results of the FDFD calculations as a complex-valued transfer function. These near-
405 field waveforms enabled us to retrieve the local dynamics of the spin deflection angle, ϕ , by time-
406 domain numerical integration as detailed below. The overall polarisation rotation was obtained by
407 integrating the local contributions along the entire probe volume, weighed by the intensity profile of
408 the probe beam. We used a diameter of 6 μm (FWHM) in the direction parallel to the capacitor plates,
409 and 2 μm (FWHM) in the orthogonal direction in order to account for diffraction effects near the
410 capacitive plates. While calibrating near-fields in excess of $\sim 10 \text{ MV cm}^{-1}$ is challenging^{27,28}, the total
411 polarisation rotation is robust against variations of the maximum near-fields occurring only in the
412 close vicinity of the capacitive plates, as confirmed by calculations. A grid resolution of $(100 \text{ nm})^3$
413 was chosen for proper convergence.

414 **Calculation of spin dynamics.** We adapted the previously derived formalism for THz-induced spin
415 dynamics based on the generalized sine-Gordon equations for our high-field setting²⁶. The vectorial
416 spin orientation can be mapped onto the angle ϕ between the antiferromagnetic vector \mathbf{G} and the x -axis
417 (Fig. 3a). The magnetic potential $W(\phi)$ of TmFeO_3 is given by²⁶

$$418 \quad W(\phi) = K_1 \sin^2 \phi + K_2 \sin^4 \phi - \frac{H_D}{H_E} M_{Fe} (B_{\text{ext}} \cos \alpha \cos \phi - B_{\text{ext}} \sin \alpha \sin \phi - B_{\text{THz}} \sin \phi),$$

419 (7)

420 where $H_D = 2 \times 10^5 \text{ Oe}$ is the Dzyaloshinskii field, $H_E = 2 \times 10^7 \text{ Oe}$ is the effective field of the d - d
421 exchange, and $M_{Fe} = 1000 \text{ e.m.u. cm}^{-3}$ is the magnetisation of a single Fe^{3+} sublattice³³. The parameter
422 $K_1 = 2K_2 \frac{T-T_2}{T_2-T_1}$ for $T_1 < T < T_2$, where K_2 is a constant, sets the potential curvature by the frequency of
423 the quasi-ferromagnetic mode $\omega_{\text{q-fm}}^2 = \frac{1}{2} \omega_E \omega_A \sin^2 \phi_0$ in the linear regime of spin dynamics. Here,
424 $\omega_E = \gamma H_E$, $\omega_A = \gamma \frac{K_2}{M_{Fe}}$, γ is the gyromagnetic ratio, T is the spin lattice temperature, and $T_1 = 80 \text{ K}$
425 and $T_2 = 90 \text{ K}$ are the lower and upper temperature bounds of the Γ_{24} transition phase, respectively.
426 The thermal excitations of the crystal-field-split ground states determine the equilibrium angle of the
427 spin vector, $\phi_0 = \arcsin \left(\frac{T-T_2}{T_2-T_1} \right)^{\frac{1}{2}}$ (see Ref. 26). For our numerical simulations, we calibrated the
428 effective magnetic potential $W(\phi)$ by the experiment with bulk TmFeO_3 , and we included an external

429 magnetic field along the z -axis ($\alpha = 0$) of $B_{\text{ext}} = 150$ mT compatible with the experimentally
 430 determined value. As we are operating in the high-field regime, where the THz-induced nonlinear
 431 anisotropy torque dominates²⁶, we neglect the magnetic THz spin interaction with the THz magnetic
 432 field, B_{THz} , which is oriented along the crystallographic x -axis.

433 The equation of motion accounting for a THz-induced change of the magnetic potential energy reads

$$\ddot{\phi} - C^2 \nabla^2 \phi = -\gamma_D \dot{\phi} + \omega_E \omega_A \cos(\phi) \sin(\phi) \times (\eta + \sin^2(\phi)) +$$

$$434 \quad \kappa \cos(\phi) \sin(\phi) \varepsilon_0 n_{\text{sub}} c \alpha_{\text{eff}} E_{\text{THz}}^2 - \frac{H_D}{H_E} \gamma w_E B_{\text{ext}} \sin \phi. \quad (8)$$

435 Here, γ_D is the damping. The excitation by the crystal field transitions is modelled by both an
 436 impulsive and a displacive mechanism, accounting for an increase of the angular velocity, $\dot{\phi}$, and a
 437 shift of the equilibrium spin angle, ϕ_0 , respectively, in conceptual analogy to Ref. 34. The impulsive
 438 excitation is implemented by the term proportional to the constant κ , coupling the spin dynamics to
 439 the instantaneous THz power density $\varepsilon_0 n_{\text{sub}} c \alpha_{\text{eff}} E_{\text{THz}}^2$. To account for the displacive term, we
 440 implement a strong THz-induced excitation of the crystal field transitions, leading to an increase of the
 441 population density $\Delta\rho(t)$ of the excited states of the Tm^{3+} ions. In our model, this is described by the
 442 excitation parameter $\eta = \frac{(\rho(T) + \Delta\rho(t)) - \rho_2}{\rho_2 - \rho_1}$, where $\rho(T)$, ρ_1 , and ρ_2 are the equilibrium population
 443 densities of the crystal-field split states at the temperature T , T_1 , and T_2 , respectively. The THz-induced
 444 change of the population density leads to an abrupt change of the magnetic potential, $W(\phi)$, of the iron
 445 spins, resulting in a displacive anisotropy torque. Quantitatively, we calculate
 446 $\Delta\rho(t) = \Gamma \int_{-\infty}^t \frac{\varepsilon_0 n_{\text{sub}} c \alpha_{\text{eff}}}{\hbar \omega_{\text{CFT}}} E_{\text{THz}}^2(t') dt'$, where Γ is a coupling parameter, \hbar is Planck's constant, and
 447 ω_{CFT} is the resonance frequency of the electric dipole active Tm^{3+} ground state transition³⁵. The term
 448 $C^2 \nabla^2 \phi$ accounts for the interaction between different magnetic domains of the sample, where C is the
 449 spin wave velocity that sets the maximal speed of a domain boundary. In the orthoferrites, $C = 2 \times 10^6$
 450 cm s^{-1} (see Ref. 36, 37). One can see that, on the ~ 1 ps timescale of our experiment, the regions of the
 451 sample exposed to the THz fields of different strengths can be assumed to be practically non-
 452 interacting as the magnetic excitations travel a distance of 10 nm during this time. This distance is also

453 much smaller than the characteristic spatial scale of the THz near-field of $>1 \mu\text{m}$. We therefore
454 neglected the term $C^2 \nabla^2 \phi$ in our numerical simulations.

455 The local dynamics of the spin deflection angle, ϕ , are calculated by solving equation (8) separately
456 for each near-field cell using the corresponding THz near-field transient (see Supplementary Video 1).
457 As confirmed by polarimetry, the THz-induced change of the magnetisation leads to a rotation of the
458 near-infrared probe polarisation. A switch-off analysis shows that the Faraday rotation is almost
459 exclusively caused by the ferromagnetic component of the magnetisation, while the dynamics of the
460 antiferromagnetic response plays a minor role. Thus, the microscopic Faraday rotation is obtained by
461 projecting the ferromagnetic vector, $\mathbf{F}(\phi)$, of each cell onto the wave vector of the near-infrared probe
462 beam, \mathbf{k}_{NIR} . Integration of these contributions along the optical axis allows us to quantitatively
463 reproduce the experimentally detected polarisation rotation, θ , (see Fig. 3c). In the non-perturbative re-
464 gime, the actual spin trajectory depends sensitively on the exact location within the near-field region
465 of the antenna. Yet the total magneto-optical response integrated over the entire near-field volume is
466 fairly robust against minor field fluctuations. For our measurement with a far-field THz peak
467 amplitude of $E_{\text{THz}} = 0.4 \text{ MV cm}^{-1}$, we obtain the best agreement (Fig. 3c, blue curve) using the
468 experimentally determined spin dephasing rate $\gamma_{\text{D}} = 45 \text{ GHz}$, as well as the following values: $\omega_{\text{q-fm}} /$
469 $2\pi = 88.7 \text{ GHz}$, $\kappa = 3.58 \times 10^8 \text{ m}^2 \text{ W s}^{-2}$, and $\Gamma = 2.09 \times 10^{-10} \text{ m}^3 \text{ s}$. For a THz peak amplitude of $E_{\text{THz}} =$
470 1.0 MV cm^{-1} (Fig. 3c, red curve), we slightly adjust some of the parameters to $\omega_{\text{q-fm}} / 2\pi = 90.0 \text{ GHz}$,
471 $\kappa = 1.02 \times 10^8 \text{ m}^2 \text{ W s}^{-2}$, and $\Gamma = 1.01 \times 10^{-10} \text{ m}^3 \text{ s}$. Magnon-magnon scattering can effectively be
472 accounted for by introducing a momentum dependent damping in the spin system. Extended Data
473 Figure 9 shows the results of a switch-off analysis considering three scenarios including the full
474 calculation (solid lines), only the displacive (dashed lines), and only the impulsive contribution
475 (dashed-dotted lines). Whereas for a field amplitude of $E_{\text{THz}} = 0.4 \text{ MV cm}^{-1}$, the sum of displacive and
476 impulsive contributions approximates the full calculation, the strong-field dynamics at $E_{\text{THz}} = 1.0 \text{ MV}$
477 cm^{-1} are only rendered correctly by the full calculation. In all cases, a purely displacive effect yields an
478 exclusively positive magneto-optical signal and a non-zero signal offset, while the impulsive
479 component is responsible for the strong oscillatory component.

480

481 **Methods References**

482 31. Zhang, K. *et al.* Resolving the spin reorientation and crystal-field transitions in TmFeO₃ with
483 terahertz transient. *Scientific Reports* **6**, 23648 (2016).

484 32. Leake, J. A., Shirane, G. & Remeika, J. P. The magnetic structure of thulium orthoferrite,
485 TmFeO₃. *Solid State Communications* **6**, 15–17 (1968).

486 33. Srinivasan, G. & Slavin, A.N. High Frequency Processes in Magnetic Materials (*World*
487 *Scientific*, 1995).

488 34. Garrett, G.A., Albrecht, T.F., Whitaker, J.F. & Merlin, R. Coherent THz Phonons Driven by
489 Light Pulses and the Sb Problem: What is the Mechanism? *Phys. Rev. Lett.* **77**, 3661 (1996).

490 35. Kozlov, V. V. *et al.* Observation of Magnetic Dipole and Electric Dipole Electron Transitions
491 in the Ground Multiplet of the Rare-Earth Ion in TmFeO₃. *Phys. Lett. A* **52**, 264 (1990).

492 36. Zvezdin, A. K. Dynamics of domain walls in weak ferromagnets. *Pisma Zh. Exp. Teor. Fiz.*
493 **29**, No. 10, 605–610 (1979).

494 37. Chetkin, M.V. & de La Campa, A. Maximum velocity of a domain wall in a weak
495 ferromagnet. *JETP Letters* **27**, 157 (1978).

496

497 **Data Availability.** The data supporting the findings of this study are available from the corresponding
498 authors upon request.

499

500 **Supplementary Information**

501 **Supplementary Video 1 | Visualisation of calculated local spin dynamics in the antenna near-**
502 **field.** Top panel, measured (grey curve) and calculated (red curve) polarisation rotation signal for
503 $E_{\text{THz}} = 1.0 \text{ MV cm}^{-1}$ (Fig. 3c, red curve). Lower set of panels, *y-z*-, *x-y*- and *x-z*-projections of the
504 calculated spin dynamics in the antenna near-field as a function of the delay time, *t*.

505 **Extended Data Figure 1 | Experimental setup.** **a**, Microscope image of the gold bowtie antenna with a
506 resonance frequency of 0.65 THz and a feed gap of 3.5 μm , structured onto the TmFeO_3 sample. **b**, Ti:sapphire
507 amplifier, centre wavelength, 807 nm; pulse energy, 5.5 mJ; pulse duration, 33 fs; repetition rate, 3 kHz. The
508 grating (G), imprints a pulse front tilt onto the near-infrared beam. Two cylindrical lenses image and focus the
509 laser light into a cryogenically cooled lithium niobate crystal (LiNbO_3). WG, pair of wire grid polarisers
510 controlling the intensity and the polarisation state of the generated THz pulses. ITO, indium tin oxide coated
511 calcium fluoride window. The THz-induced polarisation changes are decoded with the help of a half-wave plate
512 ($\lambda/2$), a Wollaston polariser (WP) and a pair of photodiodes and subsequently detected with a lock-in amplifier.
513 DL, mechanical delay line. E_{NIR} , near-infrared probe pulse polarisation. E_{THz} , THz polarisation. The inset depicts
514 the orientation of the static magnetic field, \mathbf{B}_{ext} , as a function of the angle α relative to the orientation $\mathbf{B}_{\text{ext},0}$ used
515 for the measurements in the first part of the manuscript. **c**, Electro-optically detected THz field, E_{THz} , generated
516 by tilted-pulse front optical rectification. **d**, Corresponding spectral amplitude of the THz transient shown in **c**.
517 The blue arrows indicate the frequencies of the Tm^{3+} ground state transitions relevant for our experiment.

518

519 **Extended Data Figure 2 | Scaling of the residual offset for large delay times.** Polarisation rotation signal at a
520 delay time of $t = 950$ ps as a function of the THz electric peak field, E_{THz} . The data are extracted from time-
521 resolved measurements in the feed gap of an antenna structurally similar to the one discussed in the main text
522 with a feed gap of 3.5 μm and a broad resonance around 0.65 THz, optimized to the Tm^{3+} ground state
523 transitions. Lattice temperature $T = 81$ K. In the spin switching regime $E_{\text{THz}} > 0.65$ MV cm^{-1} the slope of the
524 polarisation rotation signal is significantly increased. Error bars, standard deviation of θ for the integration time
525 of 1 s. Dashed lines, guides to the eye.

526

527 **Extended Data Figure 3 | Qualitative simulation of the beating signature.** **a**, Polarisation rotation calculated
528 by superimposing the responses shown in Fig. 3b, that is, spins oscillating in the equilibrium potential minimum
529 at ϕ_0 (relative weight, 0.8) and spins driven into the neighbouring local minimum at ϕ_1 (relative weight, 0.2).
530 **b**, Amplitude spectra of the time-domain data shown in **a**.

531

532

533 **Extended Data Figure 4 | Temperature dependence of spin dynamics. a,** Transient polarisation rotation
534 probed in the centre of the feed gap of the antenna discussed in Fig. 4, for a THz far-field amplitude
535 $E_{\text{THz}} = 1.0 \text{ MV cm}^{-1}$ and different lattice temperatures, T , between 82.0 K and 84.0 K. **b,** Corresponding
536 amplitude spectra of the data shown in **a**.

537

538 **Extended Data Figure 5 | Faraday signal for spin dynamics in different magnetic potentials. a,** Magnetic
539 potential (red curve) for a lattice temperature, $T = 82.5 \text{ K}$, and an angle of \mathbf{B}_{ext} , $\alpha = 60^\circ$, as shown in Fig. 4c.
540 Violet (grey) sphere, initial (switched) spin state. Inset: projection (grey dotted horizontal lines) of the
541 magnetization $\mathbf{F}(\phi)$ (arrows) onto the near-infrared wave vector, $\mathbf{k}_{\text{NIR},z}$ (light blue arrow), for different angles ϕ .
542 For $\phi < \phi_0$, the projection drops below its initial value and becomes negative for $\phi < -90^\circ$, causing a negative
543 transient Faraday signal (Fig. 4e). For $\phi_0 < \phi < \phi_1$, $\mathbf{k}_{\text{NIR}} \cdot \mathbf{F}(\phi) > \mathbf{k}_{\text{NIR}} \cdot \mathbf{F}(\phi_0)$, resulting in the positive initial half-
544 cycle of the Faraday rotation signal (Fig. 4e). **b,** Magnetic potential for $\alpha = 95^\circ$ (dark red curve) as shown in
545 Fig. 4d. For $\phi < \phi_0$, the initial spin deflection leads to $\mathbf{k}_{\text{NIR}} \cdot \mathbf{F}(\phi) < \mathbf{k}_{\text{NIR}} \cdot \mathbf{F}(\phi_0)$, causing a negative onset of the first
546 oscillation period (Fig. 4e, bottom curve).

547

548 **Extended Data Figure 6 | Field dependence of spin dynamics for $\alpha = 60^\circ$. a,** Polarisation rotation signal as a
549 function of the delay time, t , for different THz fields, E_{THz} , between 0.42 and 1.0 MV cm^{-1} , probed in the centre
550 of the feed gap of the antenna discussed in Fig. 4. The transient negative Faraday signal (dashed-dotted curves)
551 builds up for $E_{\text{THz}} \geq 0.87 \text{ MV cm}^{-1}$. **b,** Corresponding amplitude spectra of the data shown in **a**.

552

553 **Extended Data Figure 7 | Electric field enhancement in the near-field of a THz nanoantenna.** Enhancement
554 factor $E_{\text{NF}}/E_{\text{THz}}$ of the near-field peak amplitude E_{NF} compared to the THz electric far-field E_{THz} calculated by
555 finite-difference simulations for a real THz waveform in the near-field of an antenna structure with a feed gap of
556 10 nm. Assuming a switching threshold of $\sim 10 \text{ MV cm}^{-1}$ a far-field amplitude of only 1 kV cm^{-1} is sufficient to
557 drive coherent spin switching by 90° in the centre of the antenna structure.

558

559 **Extended Data Figure 8 | Calculated electric near-field characteristics of antenna.** Near-field amplitude E_{NF}
560 as a function of depth z in the center of the antenna feed gap, for a THz far-field amplitude of $E_{\text{THz}} = 0.4 \text{ MV cm}^{-1}$
561 cm^{-1} (red curve). The electric field distribution expected in the unstructured substrate, for $E_{\text{THz}} = 1.0 \text{ MV cm}^{-1}$ is
562 shown for comparison (black line). The near-field region of the antenna, where the electric field exceeds the
563 value of the bulk structure, is indicated by the red-shaded area.

564

565 **Extended Data Figure 9 | Simulated magneto-optical response for different driving forces.** Calculated
566 polarisation rotation signals expected from the antenna structures for a THz far-field amplitude of 0.4 MV cm^{-1}
567 (blue curves) and 1.0 MV cm^{-1} (red curves). Calculations including only the displacive (dashed lines) or
568 impulsive (dashed-dotted lines) anisotropy torque do not fit the experimental data. For the switch-off analysis,
569 the parameters Γ for the displacive and κ for the impulsive torque of the full calculation (solid lines) are used.
570 The curves are offset and normalized to the experimental peak value.