Temporal and spectral fingerprint of ultrafast all-coherent spin switching

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Future information technology demands ultimately fast, low-loss quantum control. Intense light fields have facilitated important milestones, such as inducing novel states of matter[1-3], accelerating electrons ballistically[4-7], or coherently flipping the valley pseudospin[8]. 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 possibility[9-11], yet observing the actual dynamics has remained out of reach. Here, we show that terahertz (1 THz = 10^{12} 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 $TmFeO_3$ 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 principle[12] 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 way[15-17] from the discovery of

38   subpicosecond laser-induced spin dynamics[18] to all-optical non-thermal recording[19]. Understanding

39   strongly non-equilibrium spin dynamics triggered by THz pulses, however, is still in its infancy. In an-

40   tiferromagnets, magnons feature resonance energies in the meV range[20] and can be directly addressed

41   by the magnetic field component of intense THz pulses[21-23]. 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 accelerators[9], 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 resolution[24].

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 transitions[27] and inter-

53   band Zener tunnelling, with subdiffractional spatial definition[28]. 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

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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 temperature[26], 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 interactions[29]. 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 µm) onto a 60-µm-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 $E_{THz} = 1.0$ 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 ($T = 83$ 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 $E_{THz} =$

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) mode[26], respectively. The maximum rotation angle of the probe polarisation of

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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    $E_{THz} = 0.4$ 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 field[26],

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, ~13 μm) whereas the magneto-optical

95   signal in the antenna-free case (Fig. 1c, black curve) originates from the entire thickness (60 μm) 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 $E_{THz} = 0.15$ 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   $E_{THz} = 0.75$ 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 ps < t < 35 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 oscillations[30]. The long-lived offset (Fig. 2b) manifests itself

110   in a dc spectral component, which grows more rapidly for $E_{THz} > 0.75$ 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 states[26]. $W(\phi)$ 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 $B_{ext} = 100$ 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 $E_{NF} = 6$ MV cm^{-1}, the spins carry out a coherent

128   oscillation about ϕ_0. A field of $E_{NF} = 10$ 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 $\sim 90°$. While crossing the potential maximum the spins acquire a characteristic phase,

131   which causes a retardation by $\sim 180°$ 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

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Fig. 3b, the pronounced beating feature ($t \approx 25$ ps) can be clearly associated with the phase slip

occurring during spin switching (see Extended Data Figure 3). For a quantitative analysis, we combine

our calculation of the near-field induced by the experimental THz wave with a numerical solution of

the local generalized sine-Gordon equation (see Methods). We then weigh the locally induced Faraday

signal by the Gaussian intensity distribution of the probe beam and sum all the microscopic

contributions from the probed volume. Figure 3c shows the calculated polarisation rotations, θ, for

$E_{THz} = 0.4$ MV cm^{-1} and 1.0 MV cm^{-1}. All experimental features are quantitatively reproduced,

including the quasi-monochromatic magnon oscillation, for low fields (Fig. 3c, blue curve), as well as

the phase retardation of the first magnon oscillation period and the pronounced beating at $t \approx 25$ ps, at

large THz fields (Fig. 3c, red curve). Moreover, the model unambiguously connects the asymmetric

splitting of the q-fm resonance and the broad low-frequency components (Fig. 3d) to THz-driven all-

coherent spin switching. The calculation also proves that the switched spins can be directly read out.

As seen in Fig. 3e, increasing E_{THz} leads to a long-lived signal offset, caused by two distinct

mechanisms: (i) THz excitation of Tm^{3+} ions slightly shifts the position of the potential minimum (Fig.

3b, inset). (ii) A transfer of spins over the barrier can also change the net magneto-optical signal if $\mathbf{k}_{NIR}$

is tilted out of the y-z-plane (Fig. 3a). In our experiment, we estimate a tilt angle of ~1°. Whereas the

offset caused by the shift of the magnetic potential grows slowly with E_{THz} (Fig. 3e, red circles), the

slope of the long-lived Faraday signal (Fig. 3e, red spheres) increases rapidly above the switching

threshold $E_{THz} > 0.75$ MV cm^{-1}, as seen in the experiment (Fig. 2d). This steep increase is thus a direct

way of reading out the switched spin population.

Based on the microscopic understanding of the spin dynamics, we can shape the spin trajectory

by tailoring the magnetic potential. As a first control parameter (see Extended Data Figure 4), we

lower the temperature to $T = 82.5$ K, where the barrier height, w, is slightly increased (Fig. 4a).

Consequently, the switching dynamics are decelerated and the beating signature is delayed to $t = 45$ ps

(Fig. 4e, top curve). Meanwhile, the spectrum remains qualitatively similar (Fig. 4f, top curve). The

barrier height can also be raised by rotating the external magnetic bias field, $\mathbf{B}_{ext}$, by an angle $\alpha = 15°$

about the optical axis (Fig. 4b and Extended Data Figure 1), resulting in a shift of the beating feature

to a delay of $t = 55$ ps (Fig. 4e). Thereby, the potential shoulder at $\phi = -115°$ 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 $\alpha = 60°$ (Fig. 4c), the dynamics are strongly altered

170   (Figs. 4e, f). After the spins are driven up the potential barrier at $\phi = 0°$ 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 $\phi = -115°$. This results in a strong red-shift of the centre frequency to 50 GHz. On the

173   potential shoulder, the projection $\mathbf{F} \cdot \mathbf{k}_{NIR}$ 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, $\alpha = 95°$ 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 films[14]. 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.

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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.

Figure 1 | Antenna-enhanced THz spin dynamics. a, Schematic of the gold bowtie antenna on $TmFeO_3$. The structure is excited from the back side by an intense THz electric field E_{THz} (red waveform) while a co-propagating near-infrared pulse ($h\nu_{probe}$, light blue) probes the induced magnetisation dynamics in the centre of the feed gap. **b,** Peak near-field amplitude, E_{NF}, in the antenna feed gap calculated by finite-difference simulations for a real THz waveform with a peak field amplitude of $E_{THz} = 1.0$ MV cm^{-1} (see Extended Data Fig. 1c). **c,** Experimentally detected polarisation rotation signal as a function of the delay time, t, obtained for a peak electric THz field of $E_{THz} = 1.0$ MV cm^{-1} on the unstructured substrate (black curve) and when probing the gap of the bowtie antenna structure resonantly exited by a THz waveform with a peak electric far-field amplitude of $E_{THz} = 0.4$ MV cm^{-1} (blue curve, vertically offset by 1 mrad for better visibility). Inset: Corresponding amplitude spectra featuring two modes at 0.09 THz and 0.82 THz. The sample was kept at a lattice temperature of $T = 83$ K.

Figure 2 | THz-induced nonlinear spin dynamics. a, Polarisation rotation probed in the centre of the antenna feed gap for various far-field amplitudes, as a function of the delay time, t. For incident THz peak fields $E_{THz} >$ 0.75 MV cm^{-1}, the quasi-monochromatic oscillation is strongly distorted by a phase slip at delay times between 25 and 35 ps. The measurements are offset and scaled as indicated for clarity. Lattice temperature $T = 83$ K. **b,** Long-term evolution of the polarisation rotation for a THz peak field of $E_{THz} = 1.0$ MV cm^{-1}. The red-shaded area indicates the long-lived offset. **c,** Spectral amplitude of the time-domain data shown in **a**. The phase slip in the polarisation rotation signal for highest THz fields manifests itself in a splitting of the q-fm resonance. **d,** Spectral amplitude of the dc offset, $A_{0\ THz}$, as a function of the THz far-field peak amplitude, E_{THz}. $A_{0\ THz}$ increases monotonically with the THz field. Grey-shaded area: Spin-switching regime with increased slope of A_0 $_{THz}$. Dashed lines, guides to the eye.

Figure 3 | Microscopic picture of ballistic spin motion. a, Spin and lattice structure of $TmFeO_3$ in the Γ_{24} phase ($T_1 < T < T_2$), showing the Fe^{3+} spins (dark blue spheres and arrows), Tm^{3+} ions (orange spheres), and the ferromagnetic moment, **F** (violet arrow). The antiferromagnetic vector **G** (brown arrow) lies in the x-z-plane and encloses a finite angle of $0 < \phi < 90°$ with the x-axis. Inset: geometry of the wave vector of the probe pulse, $\mathbf{k}_{NIR}$ (light blue arrow), with respect to **F** and the external magnetic field $\mathbf{B}_{ext}$ (grey arrow). **b,** Numerical simulation of THz-induced ballistic spin dynamics. Upon THz excitation, the magnetic potential $W(\phi)$ is abruptly modified

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

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

Methods

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

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

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

354
$$P(t) = j(t) E(t), \tag{1}$$

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

358 By taking the Fourier transforms $j(t) = \frac{1}{2\pi}\int_{-\infty}^{\infty} \tilde{\jmath}(\omega)e^{i\omega t}\, d\omega$ 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
$$W_{\text{abs}} = \frac{1}{2\pi}\int_{-\infty}^{\infty} \tilde{\jmath}(\omega)\tilde{E}(-\omega)\, d\omega. \tag{3}$$

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

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

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

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

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

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 $W_{\text{abs}} = 20$ J cm^{-3}. $TmFeO_3$

373 crystallises in a distorted perovskite structure with a unit cell volume of $V_{\text{uc}} = 2.22 \times 10^{-28}$ m^3 (lattice

374 constants, $a = 525$ pm, $b = 557$ pm, and $c = 758$ 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 $\frac{V_{\text{uc}}}{4} = 7.15$ meV, which is on the order of the energy of one THz photon. The dissipation by the spin

15

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 $\Delta\phi = 3.5°$. Taking into account the quadratic

388   dependence of $\Delta\phi$ on the electric field amplitude[26], 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, $\xi = 472$ mrad cm (MV)$^{-2}$ is the

390   coupling constant, and $\bar{E}_{\text{peak}} = 0.42$ 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 $\theta_{\text{b}} = \xi \times 47$ μm $\times$ (0.3 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 $\Delta\phi = 24°$.

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 ~10 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 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 setting[26]. The vectorial

416   spin orientation can be mapped onto the angle ϕ between the antiferromagnetic vector **G** and the *x*-axis

417   (Fig. 3a). The magnetic potential $W(\phi)$ of TmFeO$_3$ is given by[26]

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

419   (7)

420   where $H_D = 2 \times 10^5$ Oe is the Dzyaloshinskii field, $H_E = 2 \times 10^7$ Oe is the effective field of the *d-d*

421   exchange, and $M_{Fe} = 1000$ e.m.u. cm^{-3} is the magnetisation of a single Fe^{3+} sublattice[33]. 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 $T_1 = 80$ K

425   and $T_2 = 90$ 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 $B_{ext} = 150\,mT$ compatible with the experimentally

430   determined value. As we are operating in the high-field regime, where the THz-induced nonlinear

431   anisotropy torque dominates[26], 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

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

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

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 $\varepsilon_0 n_{sub} c \alpha_{eff} E_{THz}^2$. 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     $\Delta\rho(t) = \Gamma \int_{-\infty}^{t} \frac{\varepsilon_0\ n_{sub}\ c\ \alpha_{eff}}{\hbar\omega_{CFT}} E_{THz}^2(t')\ dt'$, 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 transition[35]. 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 μm. 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, $\mathbf{k}_{\mathrm{NIR}}$. 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 $E_{\mathrm{THz}} = 0.4\,\mathrm{MV\,cm^{-1}}$, we obtain the best agreement (Fig. 3c, blue curve) using the

468 experimentally determined spin dephasing rate $\gamma_{\mathrm{D}} = 45$ GHz, as well as the following values: $\omega_{\mathrm{q-fm}}/$

469 $2\pi = 88.7$ GHz, $\kappa = 3.58 \times 10^8\,\mathrm{m^2\,Ws^{-2}}$, and $\Gamma = 2.09 \times 10^{-10}\,\mathrm{m^3 s}$. For a THz peak amplitude of $E_{\mathrm{THz}} =$

470 $1.0\,\mathrm{MV\,cm^{-1}}$ (Fig. 3c, red curve), we slightly adjust some of the parameters to $\omega_{\mathrm{q-fm}}/2\pi = 90.0$ GHz,

471 $\kappa = 1.02 \times 10^8\,\mathrm{m^2\,Ws^{-2}}$, and $\Gamma = 1.01 \times 10^{-10}\,\mathrm{m^3 s}$. 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 $E_{\mathrm{THz}} = 0.4\,\mathrm{MV\,cm^{-1}}$, the sum of displacive and

476 impulsive contributions approximates the full calculation, the strong-field dynamics at $E_{\mathrm{THz}} = 1.0$ 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.

Methods References

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

Supplementary Information

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

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

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

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

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

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

Extended Data Figure 6 | Field dependence of spin dynamics for $\alpha = 60°$. a, Polarisation rotation signal as a 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 of the feed gap of the antenna discussed in Fig. 4. The transient negative Faraday signal (dashed-dotted curves) builds up for $E_{THz} \geq 0.87$ MV cm^{-1}. **b,** Corresponding amplitude spectra of the data shown in **a**.

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

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 $E_{THz} = 0.4$ MV

561 cm^{-1} (red curve). The electric field distribution expected in the unstructured substrate, for $E_{THz} = 1.0$ 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.