

Spin switching in $\text{Sm}_{0.7}\text{Er}_{0.3}\text{FeO}_3$ triggered by terahertz magnetic-field pulses

Zhenya Zhang¹, Minoru Kanega², Kei Maruyama¹, Takayuki Kurihara³, Makoto Nakajima⁴, Takehiro Tachizaki⁵, Masahiro Sato², Yoshihiko Kanemitsu^{1*} & Hideki Hirori^{1*}

¹*Institute for Chemical Research, Kyoto University, Uji, Kyoto 611-0011, Japan.*

²*Department of Physics, Chiba University, Chiba 263-8522, Japan.*

³*Institute for Solid State Physics, The University of Tokyo, 5-1-5 Kashiwanoha, Kashiwa, Chiba 277-8581, Japan.*

⁴*Institute of Laser Engineering, Osaka University, Suita, Osaka 565-0871, Japan.*

⁵*Department of Optics and Imaging Science and Technology, Tokai University, Hiratsuka, Kanagawa 259-1292, Japan.*

*Corresponding author: kanemitsu@scl.kyoto-u.ac.jp

*Corresponding author: hirori@scl.kyoto-u.ac.jp

Abstract

Driving spin systems to states far from equilibrium is indispensable in investigations of functional nonlinearities of antiferromagnets for spintronics. So far, it has been shown that electric-field pulses in the spectral region from the visible to the terahertz range can be used to induce ultrafast switching between different spin states. Here, we demonstrate that a multicycle terahertz magnetic-field pulse can also be used to induce non-thermal spin switching in antiferromagnets. When a strong pulse is applied to $\text{Sm}_{0.7}\text{Er}_{0.3}\text{FeO}_3$, the magnetic order parameter is first driven away from the barrier between the two potential minima of this antiferromagnet and then, in the subsequent inertial motion towards the opposite direction, it crosses the barrier. Our analysis reveals that the initial motion is driven by a dynamical modification of the magnetic potential, and this modification is enhanced through coupling between the two magnon modes.

Main

Inducing light–matter interactions with intense laser pulses is an efficient way to achieve ultrafast control of macroscopic states of materials^{1–7}, and such control is considered to be a cornerstone of future information technology. In particular, manipulation of magnetism on the picosecond time scale using laser pulses has recently attracted interest^{8–11}. For example, coherent spin switching has been demonstrated by using the inverse Faraday effect to effectively generate a magnetic field with a near-infrared light pulse⁹ and also by driving phonons with mid-infrared light to modify the exchange interaction¹¹. However, heating effects cannot be avoided with these methods because they excite either electrons or phonons. Manipulating spins by using terahertz (THz) pulses has been considered as a means of spin switching that does not involve a strong excitation of electrons or phonons because of the low photon energy (0.1–3 THz corresponds to 0.4–12.4 meV). However, because the time scale of spin switching is relatively long even for antiferromagnets^{9–11}, THz-induced spin switching via resonantly excited spin dynamics is often not possible. While it has been shown that coherent spin switching is possible by controlling spins via electronic orbital transitions induced by the electric component of a THz pulse¹⁰, nonlinear excitation of coherent spin switching using THz magnetic fields is still rather unexplored.

One possible way to advance THz-based spintronics and magnonics is to discover new phenomena related to the nonlinear manipulation of spins in antiferromagnets^{12–21}. This approach is inspired by the fact that a time-periodic external field can be used to modify a system’s energy landscape, which is a method that is recognized for its broad applicability and has attracted attention in the field of non-thermal manipulation of material properties. This method has been extensively studied for both quantum and classical (mechanical) systems, such as electrons in solids exposed to time-periodic

electric fields^{22–29} and pendulums driven by fast-oscillating forces^{30–32}, respectively. Similarly, the magnetic potential of a material can be modified by multicycle magnetic fields, which is still an unexplored but promising area. In the adiabatic limit, the potential is changed so slowly that the magnetic order will remain at the instantaneous minimum. On the other hand, if the magnetic potential is dynamically modified at a rate surpassing the adiabatic limit, the magnetic order will move away from the instantaneous minimum, and thus this method may lead to coherent spin dynamics and even spin switching. Recent studies demonstrated nonlinear interactions between the two spin-wave modes in canted antiferromagnets (also called magnon modes, since magnons are the quanta of spin waves), suggesting a powerful tool for controlling the dynamics of magnetic order^{33–36}. In this work, we exploit these interactions to significantly enhance the modification of the magnetic potential, and thereby achieve spin switching through nonlinear excitation near room temperature. In particular, we use the canted antiferromagnet $\text{Sm}_{0.7}\text{Er}_{0.3}\text{FeO}_3$ and expose it to a strong multicycle THz magnetic near-field^{37,38}. Spin switching is observed after the first half cycle of the quasi-ferromagnetic (q-FM) mode oscillation, and we interpret this phenomenon as a dynamical modification of the magnetic potential that is induced by the magnetic near-field.

Spin configuration and measurement principle

As illustrated in Fig. 1a, in $\text{Sm}_{0.7}\text{Er}_{0.3}\text{FeO}_3$, the iron sublattice magnetizations $\mathbf{m}_1$ and $\mathbf{m}_2$ are aligned antiparallel with a small canting angle due to the Dzyaloshinskii–Moriya interaction (Methods). This leads to a weak ferromagnetic vector $\mathbf{M} = (\mathbf{m}_1 + \mathbf{m}_2)/2$. The antiferromagnetic vector is defined by $\mathbf{L} = (\mathbf{m}_1 - \mathbf{m}_2)/2$. The magnetic order parameters $\mathbf{L}$ and $\mathbf{M}$ lie in the a – c plane and are orthogonal, and thus their directions can be represented by the angle θ , which is the angle between $\mathbf{L}$ and the a -axis. Note that the magnetic potential of $\text{Sm}_{0.7}\text{Er}_{0.3}\text{FeO}_3$ depends on the temperature T and exhibits a second-

order spin-reorientation transition (SRT) defined by a continuous change of the magnetic potential as the temperature increases from $T_L = 310$ K to $T_H = 330$ K, as illustrated in Fig. 1b³⁹.

For $T < T_L$, the potential has its minimum at $\theta = 90^\circ$. For $T > T_L$, the potential minimum is split into two minima at $\theta = \theta_L$ and θ_R , and as T increases, θ_L and θ_R approach 0° and 180° , respectively, because the barrier at the center increases. The three red spheres in Fig. 1b indicate the orientation of $\mathbf{L}$ under equilibrium conditions. By applying an external bias field ($B_{DC} = 2$ mT) to the sample, the potential at θ_L becomes smaller than that at θ_R , and thus $\mathbf{L}$ is oriented in the direction of the minimum at θ_L when the experiment starts. The motion of $\mathbf{L}$ and $\mathbf{M}$ can be probed by using near-infrared (NIR) pulses, because a change in θ leads to a change in the ellipticity angle of the probe light, $\Delta\eta$ (Methods, Extended Data Fig. 1).

Excitation of the q-FM mode by a single THz pump pulse

To strongly excite the spin system, we used a gold microstructure on the sample surface that allows us to generate an intense magnetic near-field $\mathbf{B}_{THz}^{NF}$ at 0.46 THz using a THz pulse, as shown in Fig. 1c^{38,40,41}. We verified that the response of the sample under this excitation condition was not due to an electric near-field^{10,42} (Supplementary Section I). The upper panel of Fig. 1d shows the c -component of the $\mathbf{B}_{THz}^{NF}$ generated at the center of the spiral calculated by a finite-difference time-domain (FDTD) simulation. The peak amplitude of $\mathbf{B}_{THz}^{NF}$ for a given excitation power is hereafter referred to as B_{amp}^{NF} .

We first investigated the sample response by using a moderate excitation power. The observed time evolution of $\Delta\eta$ is shown as the coloured curves in the lower panel of Fig. 1d for four different temperatures (Supplementary Section II). These curves exhibit a superposition of fast and slow oscillations. The Fourier spectrum of the $\Delta\eta$ curve for $T =$

315 K in Fig. 1e shows that there is a fast oscillation at 0.545 THz (blue arrow). This oscillation reflects the excitation of the quasi-antiferromagnetic (q-AFM) mode, which is confirmed by the temperature dependence of the oscillation frequency and that of the amplitude (Figs. 1f and 1g, respectively). The appearance of the q-AFM mode corresponds to an oscillation in the amplitude of $\mathbf{M}$ and a pendulum motion of $\mathbf{L}$ out of the a - c plane (Extended Data Fig. 2a). It can be simply interpreted in terms of a resonant excitation by the high-energy tail of $B_{\text{THz}}^{\text{NF}}$ (Fig. 1e)³⁸, consistent with the observed linear dependence on $B_{\text{amp}}^{\text{NF}}$ (Extended Data Fig. 3).

On the other hand, the slow but large oscillation at 0.052 THz (red arrow) shows a completely different temperature dependence; the softening at $T = T_L$ in Fig. 1f verifies that this oscillation is due to the quasi-ferromagnetic (q-FM) mode. The q-FM mode corresponds to an oscillation of θ and is therefore affected by changes in the magnetic potential. The q-FM signal is thus important for this work. The q-FM mode lies clearly out of the spectral range of $B_{\text{THz}}^{\text{NF}}$, and therefore the excitation process should have a strongly non-resonant character. Furthermore, the black $\Delta\eta$ curve in Fig. 1d obtained using a THz magnetic near-field with an inverse waveform (obtained by rotating the microstructure by 180° about the c -axis) clarifies that the initial “downward” movement of the q-FM mode oscillation (that is, the signal with $\Delta\eta < 0$) is not affected by the sign of $B_{\text{THz}}^{\text{NF}}$. This behavior is opposite to that expected for oscillations resonantly excited via the Zeeman torque^{13,16}. We also investigated the dependence of the q-FM mode amplitude on $B_{\text{amp}}^{\text{NF}}$: As shown in Fig. 1h, the q-FM mode amplitude exhibits a quadratic dependence on $B_{\text{amp}}^{\text{NF}}$.

Spin switching induced by THz double-pump pulse excitation

If the excitation is relatively weak like in Fig. 1d, $\mathbf{L}$ will always relax to the initial potential minimum in the double-well potential (Fig. 1b), and thus the spin directions

before and after excitation are the same ($\theta < 90^\circ$ in the range $T > T_L$). Spin switching results in a final orientation of $\mathbf{L}$ and $\mathbf{M}$ that is different from the initial orientation, and thus provides information about the physical meaning of the “downward” movement in Fig. 1d. To induce spin switching, we used a stronger THz magnetic near-field generated by a THz double-pump pulse (Methods, Supplementary Section III).

Figure 2b shows the sample response for different values of $B_{\text{amp}}^{\text{NF}}$ at $T = 311$ K, where the potential barrier at 90° is relatively small. In the time range $t > 20$ ps, a large offset towards positive $\Delta\eta$ values emerges as $B_{\text{amp}}^{\text{NF}}$ is increased. As shown in Fig. 2b, the offset at about 150 ps steeply grows after $B_{\text{amp}}^{\text{NF}}$ exceeds about $0.88 B_{\text{max}}^{\text{NF}}$, where $B_{\text{max}}^{\text{NF}}$ denotes the maximum peak amplitude achieved in our experiment. Here, we did not observe a gradually increasing component accompanying carrier excitation as in the case of spin switching by THz-electric-field excitation¹⁰. To characterize the dependence of the q-FM mode amplitude on $B_{\text{amp}}^{\text{NF}}$, we plot the experimental values of $\Delta\eta$ at 13 ps versus $B_{\text{amp}}^{\text{NF}}$ ² in the upper panel of Fig. 2c. The quadratic dependence on $B_{\text{amp}}^{\text{NF}}$ is evidence of a nonlinear excitation process. To evaluate the threshold behavior of the offset at $t > 20$ ps quantitatively, the experimental values of $\Delta\eta$ at 150 ps are plotted in the lower panel of Fig. 2c. The obtained result is steeper than that of the q-FM mode amplitude at 13 ps, indicating that the large positive offset in the time range $t > 20$ ps reflects the change of the orientation of $\mathbf{L}$ from θ_L (75°) to θ_R (103°); i.e., spin switching has occurred. Due to thermal fluctuations⁴³, the non-equilibrium spin distribution generated by the THz double-pump pulse relaxes to the equilibrium state before the next double-pump pulse arrives¹⁰ (Supplementary Sections IV and V).

The observed switching process clarifies that the initial “downward” movement in Figs. 1d and 2b corresponds to the motion of $\mathbf{L}$ away from the barrier at 90° . To clarify the details of the observed switching process, the sample response shown in Fig. 3a is divided

into three regions. Before pumping, $\mathbf{L}$ has an orientation according to the equilibrium condition ($\theta = \theta_L$). When the system is exposed to the strong THz magnetic field, $\mathbf{L}$ is driven from the initial minimum to the $+a$ direction (process 1 in Fig. 3a; the rotation of $\mathbf{L}$ indicated by the orange arrow leading to $\theta < \theta_L$), which results in the “downward” movement of $\Delta\eta$. After the envelope of the THz magnetic field has decayed sufficiently, $\mathbf{L}$ starts to move back to the original minimum (the condition shown in process 2, where $\mathbf{L}$ has almost reached the minimum at θ_L). Then, as a result of “inertia”⁹, it will pass the minimum at θ_L and proceed towards the barrier at $\theta = 90^\circ$ (in other words, the “kinetic energy” gained by reaching the minimum will be again converted into potential energy). If the initial driving process is strong enough, $\mathbf{L}$ can overcome the barrier and then, through dissipation, it relaxes to the other minimum at θ_R (process 3). This leads to the observed large positive offset in $\Delta\eta$.

We emphasize that process 1 describes a rather unexpected phenomenon: $\mathbf{L}$ is initially driven away from the barrier (θ becomes less than θ_L). This peculiar initial motion is in stark contrast to the results of previous investigations that used optical pulses or THz electric pulses to trigger spin switching^{9,10}; in those reports, $\mathbf{L}$ was initially driven towards the barrier. In other words, the signs of $\Delta\eta$ at the initial and final stages of the switching process were the same. This is in contrast to the data shown in Fig. 2b, where the sign of the offset ($\Delta\eta > 0$) is opposite to that of the initial deviation ($\Delta\eta < 0$).

The above interpretation relies on the relationship between the sign of $\Delta\eta$ and the rotation direction of $\mathbf{L}$ (the sign of $\Delta\theta$). While equilibrium measurements can usually be used to determine this relationship for a given sample, the large birefringence in our sample results in an uncertainty of the relation between the sign of $\Delta\eta$ and that of $\Delta\theta$ (since this relation depends on the penetration depth and the parameters in the dielectric tensor, which makes it difficult to determine the relationship unambiguously using

equilibrium measurements; see also Supplementary Section VI). On the other hand, we know that the observed positive offset of $\Delta\eta$ is not caused by a heat-induced SRT (which would lead to $\Delta\theta < 0$)^{39,44,45}: at the probe position, the time required for a temperature increase of 20 μK (due to Joule heat transferred from the spiral) is estimated to be 5 ns, while the offset already appears at about 50 ps (Supplementary Section VII). Moreover, the following section clarifies that the behaviour of the experimental data ($\Delta\eta$) can be reproduced well by the change in θ predicted by using the Landau–Lifshitz–Gilbert (LLG) equation. This indicates that, in this particular experiment, a positive sign of $\Delta\eta$ corresponds to a positive sign of $\Delta\theta$.

Model for the observed nonlinear excitation of spin motion

To explain the observed initial motion, the sine-Gordon equation based on the LLG equation is used (Methods). The numerical results of the sine-Gordon equation including fluctuations are shown by the dotted curves in Figs. 2b and 2c; the sine-Gordon equation can explain the important features observed in the experiment. The value of $B_{\text{max}}^{\text{NF}}$ calculated by the FDTD simulation based on the actual incident THz electric field at maximum power is 3.53 T, but the amplitude of the THz magnetic near-field used in the calculation to reproduce the topmost data in Fig. 2b is only 1.45 T, (maybe due to a relatively low conductivity of the gold layer). To understand the origin of the slow motion at the beginning, we consider a dynamical modification of the magnetic potential. In other words, we consider that the q-FM mode feels a new effective potential when the system is driven by the near-field (as a result of the Zeeman torque and coupling with the q-AFM mode). To obtain the effective potential during the oscillation of the THz magnetic near-field, we analytically solved the sine-Gordon equation by replacing the time-periodic energy term in the original equation with an effective energy term (Supplementary Section VIII):

The magnetic driving field along the c -axis has the form $B_{\text{THz}}^{\text{NF}}(t) = B_{\text{env}}^{\text{NF}}(t)\cos(\omega t)$, where $B_{\text{env}}^{\text{NF}}(t)$ describes the slow variation of the envelope, and $\cos(\omega t)$ describes the fast oscillation of the THz magnetic field. Because the intrinsic frequency of the pendulum motion of $\mathbf{L}$ in the a - c plane (θ) described by the sine-Gordon equation is very slow compared to $B_{\text{THz}}^{\text{NF}}$, we consider that θ can be separated into fast (θ_{fast}) and slow (θ_{slow}) contributions. The fast contribution θ_{fast} is a forced oscillation with a frequency equal to that of $B_{\text{THz}}^{\text{NF}}(t)$ (Supplementary Section IX). The slow pendulum motion of $\mathbf{L}$ (θ_{slow}) is given by

$$\ddot{\theta}_{\text{slow}} + c\dot{\theta}_{\text{slow}} = -\frac{\partial W_{\text{eff}}}{\partial \theta_{\text{slow}}}, \quad (1)$$

$$W_{\text{eff}}(\theta_{\text{slow}}) = W_0(\theta_{\text{slow}}) + W_{\text{ZT}}(\theta_{\text{slow}}) + W_{\text{CP}}(\theta_{\text{slow}}) \quad (2)$$

$$W_0(\theta_{\text{slow}}) = \frac{ab}{2}\sin^2\theta_{\text{slow}} + \frac{a}{4}\sin^4\theta_{\text{slow}} - \chi B_{\text{DC}}\cos\theta_{\text{slow}} \quad (3)$$

$$W_{\text{ZT}}(\theta_{\text{slow}}) = \frac{\chi^2 B_{\text{env}}^{\text{NF}^2}(t)\sin^2\theta_{\text{slow}}}{4\omega^2}, \quad (4)$$

$$W_{\text{CP}}(\theta_{\text{slow}}) = \frac{\gamma^2\omega^2(\omega_{\text{AFM}}^2 - \omega^2)B_{\text{env}}^{\text{NF}^2}(t)\sin^2\theta_{\text{slow}}}{4((\omega_{\text{AFM}}^2 - \omega^2)^2 + (\omega c)^2)}, \quad (5)$$

where $c\dot{\theta}_{\text{slow}}$ is the damping term, $\chi = \gamma^2 H_{\text{D}}$, γ is the gyromagnetic ratio, H_{D} is the antisymmetric exchange field, a corresponds to the strength of anisotropy, and b corresponds to the relative sample temperature with respect to the SRT-range (Methods). W_{eff} is the effective potential defined as the sum of the potential at equilibrium [W_0 in Eq. (3)] and the additional changes in the potential that occur when the spin system is exposed to the time-periodic THz magnetic field $B_{\text{THz}}^{\text{NF}}$ [W_{ZT} in Eq. (4) due to the Zeeman torque directly exerted by the THz magnetic near-field, and W_{CP} in Eq. (5) due to coupling between the q-AFM and q-FM modes^{33–35}]. ω_{AFM} is the frequency of the q-AFM mode. Note that the potential modification becomes larger as the frequency of the time-periodic magnetic field approaches the q-AFM resonance.

To clarify the contributions of the Zeeman-torque and the coupling terms to the slow motion at the beginning, we determined the dynamics induced by each term separately (the sine-Gordon equation was numerically solved by removing either the Zeeman-torque term or the coupling term) as shown in Fig. 3a. While both the coupling term and the Zeeman-torque term induce a slow downward motion at the beginning, the contribution from the coupling between the modes in our sample is about four times larger than that from the Zeeman torque. This can be verified in Fig. 3b, where the amplitude of the slow motion at 13 ps is shown as a function of $(B_{\text{amp}}^{\text{NF}})^2$. Furthermore, the dependence of the amplitude at 150 ps on $(B_{\text{amp}}^{\text{NF}})^2$ clarifies that, near the threshold, the combination of both terms is important.

The solution predicted by Eqs. (1)–(5) leads to a deviation from the exact solution of the sine-Gordon equation, but reproduces the overall behaviour of the downward motion (Supplementary Section VIII). Thus, the above interpretation (the dynamical modification of the magnetic potential caused by $B_{\text{THz}}^{\text{NF}}$ plays a key role in the peculiar motion of the q-FM mode) is considered reasonable. Supplementary Video 1 shows the time-dependence of the effective potential calculated using Eqs. (2)–(5). It confirms that the motion of $\mathbf{L}$ falls behind the dynamical modulation of the double-well potential. As shown in the schematics of Fig. 3c, before the THz magnetic field is applied, W_{eff} is equal to W_0 , that is, we have a double-well potential with a small barrier centered at $\theta = 90^\circ$. In process 1 of Fig. 3c, the strong periodic THz magnetic near-field modifies the potential significantly ($W_{\text{eff}} = W_0 + W_{\text{ZT}} + W_{\text{CP}}$); the potential minima shift, because the barrier at $\theta = 90^\circ$ increases. At the very beginning of this modification, the shape of W_{eff} results in a force towards the left side (as defined by Fig. 3c) since $-\partial W_0 / \partial \theta_{\text{slow}} \approx 0$ and $-\partial(W_{\text{ZT}} + W_{\text{CP}}) / \partial \theta_{\text{slow}} < 0$. Thus, $\mathbf{L}$ rotates away from the barrier at $\theta = 90^\circ$ as indicated by the arrow (process 1, which explains the observed “downward” movement

of $\Delta\eta$). Because $\mathbf{L}$ rotates away from the barrier (and because the envelope of the THz magnetic field becomes smaller with time), the force towards the right side, $-\partial W_0/\partial\theta_{\text{slow}}$, becomes more important with time. Therefore, the rotation of $\mathbf{L}$ slows down and eventually starts an acceleration towards the right side (process 2). From this moment on, the force due to W_0 and the gained “kinetic energy” of $\mathbf{L}$ dominate the motion. When $\mathbf{L}$ reaches the range $\theta_L < \theta < 90^\circ$, the force due to W_0 points to the left side (like the forces due to W_{ZT} and W_{CP}), which slows down the rotation of $\mathbf{L}$ again. $\mathbf{L}$ can approach the top of the barrier only due to the inertial effect. If $\mathbf{L}$ has gained sufficient “kinetic energy” due to the applied field, it can overcome the barrier and finally relaxes to θ_R due to damping (process 3). The term “kinetic energy” refers to $\dot{\theta}^2/2$, since the equation of motion contains the term $\ddot{\theta}$, which corresponds to the acceleration term in Newton’s equation of motion with unit mass⁹. Furthermore, as shown in Figs. 4a and 4b, $\mathbf{L}$ can be either initialized at θ_L or θ_R , depending on the direction of B_{DC} . The signs of $\Delta\eta$ of both the slow oscillation and spin switching are reversed in these two cases, but in either case $\mathbf{L}$ is initially driven away from the barrier, which supports our analysis based on the sine-Gordon equation.

Finally, on the basis of Eqs. (1)–(5), we can derive a quadratic dependence of the slow component of $\Delta\eta$ on $B_{\text{amp}}^{\text{NF}}$ (Supplementary Section X), which is consistent with the observed quadratic dependence of $\Delta\eta$ in Figs. 1h and 2c. This nonlinear excitation mechanism also explains why $\Delta\eta$ is independent of the sign of $B_{\text{THz}}^{\text{NF}}(t)$ in Fig. 1d. A brief discussion about spin switching at higher temperatures, the expected behaviour at stronger THz magnetic near-fields, and the influences of the pulse envelope and the carrier frequency on the spin dynamics are provided in Supplementary Information II, XI, and XII, respectively.

In this work, we have focused on the physics that enable non-thermal control of material properties by strong THz magnetic-field pulses. In particular, we have shown how a dynamical modification of a magnetic potential that is enhanced through coupling between q-FM and q-AFM modes can nonlinearly excite the spin system and cause spin switching. Because spin systems driven by time-periodic magnetic fields can also be described using a Floquet picture³², we anticipate that Floquet band engineering of magnon modes can be demonstrated in the future. This strategy could allow us to manipulate quantum magnetic properties in various materials⁴⁶, including atomic layers of magnetic materials^{47,48} and quantum spin systems, which may lead to new optoelectronic and coherent spintronic devices^{49,50}.

Acknowledgements

The authors thank Hiroshi Watanabe, Hideaki Obuse, and Takahisa Arima for discussions, and Takashi Oka for expressing his interest in our work. The authors also thank Takahiro Moriyama and Teruo Ono for discussions and for providing access to their electron-beam lithography system. Part of this study was supported by grants from the Japan Society for the Promotion of Science, JSPS KAKENHI Grants JP19H05465, JP19H05824, JP21H01842, JP19H05825, and JP20H02206. This work was also supported by JST SPRING Grant JPMJSP2110 and JST Grant JPMJFS2107. This work was partially supported by the Frontier Photonic Sciences Project of the National Institutes of Natural Sciences (Grant Number 01212301) and by the Canon Foundation.

Author contributions

H.H. conceived the project. Z.Z., K.M., T.T., and H.H. carried out the experiments. Z.Z., M.K. K.M., T.T., M.S., Y.K., and H.H. analysed the data and derived the expression for the effective potential. T.K. and M.N. fabricated the $\text{Sm}_{0.7}\text{Er}_{0.3}\text{FeO}_3$ crystal. Y.K. and H.H. supervised the project. All authors discussed the results and contributed to the writing of the paper.

Competing interests

The authors declare no competing interests.

Figure Captions

FIG. 1 | Outline of the experiment and results obtained using a single THz pump pulse. **a**, Alignment of the iron spins in $\text{Sm}_{0.7}\text{Er}_{0.3}\text{FeO}_3$ slightly above room temperature (left-hand side) and the orientation of $\mathbf{M}$ and $\mathbf{L}$ in the a - c plane defined by θ (right-hand side). The shaded area indicates the a - c plane. **b**, The temperature dependence of the magnetic anisotropy potential as a function of θ . **c**, Schematic diagram of the experiment. A spiral-shaped gold microstructure was fabricated on the sample surface to generate a strong THz magnetic field $B_{\text{THz}}^{\text{NF}}$ along the c -axis. **d**, The upper panel shows the temporal evolution of $B_{\text{THz}}^{\text{NF}}$ ($-B_{\text{THz}}^{\text{NF}}$) along the c -axis. The waveform $-B_{\text{THz}}^{\text{NF}}$ is obtained by rotating the microstructure by 180° about the c -axis. The lower panel shows the temporal evolution of the ellipticity-angle change $\Delta\eta$. The curves are offset for clarity. **e**, The solid curve and the shaded area are the spectra of $\Delta\eta$ at 315 K and $B_{\text{THz}}^{\text{NF}}$, respectively. The three vertical arrows indicate the q-FM mode at 0.052 THz (solid arrow), the fast oscillation that follows $B_{\text{THz}}^{\text{NF}}$ (which has a peak at 0.460 THz, dotted arrow), and the q-AFM mode at 0.545 THz (solid arrow). **f**, The temperature dependence of the frequency of the q-AFM mode (open squares) and that of the q-FM mode (open circles). **g**, The amplitude of the q-AFM mode as a function of the temperature (open squares) and the corresponding data of the q-FM mode (open circles). The solid lines in **f** and **g** are guides to the eye. **h**, The dependence of the q-FM mode amplitude on the magnetic-field peak amplitude $B_{\text{amp}}^{\text{NF}}$ for three temperatures. The solid lines are proportional to the square of $B_{\text{amp}}^{\text{NF}}$.

FIG. 2 | Comparison with the theoretical results. **a**, The time trace of the strong multicycle THz magnetic field (solid curve) generated by irradiating the microstructure with two THz pulses (red and blue dashed curves) separated by a time interval of $\Delta t = 4.48$ ps. **b**, The experimental data of $\Delta\eta$ at 311 K for different magnetic-field amplitudes $B_{\text{amp}}^{\text{NF}}$ are shown by the solid curves. The dotted curves show the predictions for

$\Delta \cos \theta = \cos \theta - \cos \theta_L$ based on the sine-Gordon (SG) equation [Eq. (7) in the Methods section] including fluctuations. The results are offset for clarity. The two arrows at the bottom of the figure indicate the time delays of 13 ps and 150 ps. **c**, The amplitudes of the experimental and theoretical data in Fig. 2b at 13 ps as a function of $B_{\text{amp}}^{\text{NF}^2}$ (upper panel) and those at 150 ps as a function of $B_{\text{amp}}^{\text{NF}^2}$ (lower panel). The open circles and open squares represent the experimental results and the results predicted by the sine-Gordon equation, respectively. $B_{\text{max}}^{\text{NF}} = 3.53$ T is the value calculated by the FDTD simulation based on the actual incident THz electric field of the double-pump pulse at maximum power.

FIG. 3 | Three-step spin switching initiated by a dynamically modified potential. a, The experimental data at 311 K (thick curve), the exact solution of the sine-Gordon equation denoted by “SG (CP + ZT)” (thin black curve), and the theoretical results obtained by considering only coupling (CP) and only the Zeeman torque (ZT) (red and blue curves, respectively). The first two curves were already shown in Fig. 2b on a longer time scale and the grey curve is the same pulse as in Fig. 2a. The spin-switching process is divided into three time ranges, and the motion of $\mathbf{L}$ in each range is illustrated above the curve. The vertical arrow at the bottom of the figure indicates a time delay of 13 ps. **b**, The amplitude of the slow motion for each theoretical curve in panel a at 13 ps as a function of $B_{\text{amp}}^{\text{NF}^2}$ (left panel), and the theoretical amplitudes at 150 ps as a function of $B_{\text{amp}}^{\text{NF}^2}$ (right panel). **c**, The variation of the potential and the motion of $\mathbf{L}$ at four conditions: before $B_{\text{THz}}^{\text{NF}}$ is applied, during the time $B_{\text{THz}}^{\text{NF}}$ exhibits a large amplitude, soon after the peak of $B_{\text{THz}}^{\text{NF}}$, and the following spin switching.

FIG. 4 | Opposite initial states of magnetization. The initial direction of $\mathbf{L}$ with respect to the a -axis is determined by the direction of bias magnetic field $\mathbf{B}_{\text{DC}}$ as shown in the left

panel. For both single THz-pulse pumping and THz double-pulse pumping, the signals are reversed if the initial direction is changed from θ_L to θ_R .

References

1. Rini, M. et al. Control of the electronic phase of a manganite by mode-selective vibrational excitation. *Nature* **449**, 72–74 (2007). Doi: 10.1038/nature06119
2. Eichberger, M. et al. Snapshots of cooperative atomic motions in the optical suppression of charge density waves. *Nature* **468**, 799–802 (2010). Doi: 10.1038/nature09539
3. Först, M. et al. Nonlinear phononics as an ultrafast route to lattice control. *Nat. Phys.* **7**, 854–856 (2011). Doi: 10.1038/nphys2055
4. Gerber, S. et al. Femtosecond electron-phonon lock-in by photoemission and x-ray free-electron laser. *Science* **357**, 71–75 (2017). Doi: 10.1126/science.aak9946
5. Li, X. et al. Terahertz field-induced ferroelectricity in quantum paraelectric SrTiO₃. *Science* **364**, 1079–1082 (2019). Doi: 10.1126/science.aaw4913
6. Sie, E. J. et al. An ultrafast symmetry switch in a Weyl semimetal. *Nature* **565**, 61–66 (2019). Doi: 10.1038/s41586-018-0809-4
7. Mashkovich, E. A. et al. Terahertz-light driven coupling of antiferromagnetic spins to lattice. *Science* **374**, 1608–1611 (2021). Doi: 10.1126/science.abk1121
8. Kimel, A. V., Kirilyuk, A., Tsvetkov, A., Pisarev, R. V. & Rasing, T. Laser-induced ultrafast spin reorientation in the antiferromagnet TmFeO₃. *Nature* **429**, 850 (2004). Doi: 10.1038/nature02659
9. Kimel, A. V. et al. Inertia-driven spin switching in antiferromagnets. *Nat. Phys.* **5**, 727–731 (2009). Doi: 10.1038/nphys1369
10. Schlauderer, S. et al. Temporal and spectral fingerprints of ultrafast all-coherent spin switching. *Nature* **569**, 383–387 (2019). Doi: 10.1038/s41586-019-1174-7
11. Afanasiev, D. et al. Ultrafast control of magnetic interactions via light-driven phonons. *Nat. Mater.* **20**, 607–611 (2021). Doi: 10.1038/s41563-021-00922-7
12. Goto, E. The parametron, a digital computing element which utilizes parametric oscillation. *Proc. IRE* **47**, 1304–1316 (1959). Doi: 10.1109/JRPROC.1959.287195

13. Kampfrath, T. et al. Coherent terahertz control of antiferromagnetic spin waves. *Nat. Photon.* **5**, 31–34 (2011). Doi: 10.1038/nphoton.2010.259
14. Wienholdt, S., Hinzke, D. & Nowak, U. THz switching of antiferromagnets and ferrimagnets. *Phys. Rev. Lett.* **108**, 247207 (2012). Doi: 10.1103/PhysRevLett.108.247207
15. Chumak, A. V., Vasyuchka, V. I., Serga, A. A. & Hillebrands, B. Magnon spintronics. *Nat. Phys.* **11**, 453–461 (2015). Doi: 10.1038/nphys3347
16. Baierl, S. et al. Terahertz-driven nonlinear spin response of antiferromagnetic nickel oxide. *Phys. Rev. Lett.* **117**, 197201 (2016). Doi: 10.1103/PhysRevLett.117.197201
17. Baltz, V. et al. Antiferromagnetic spintronics. *Rev. Mod. Phys.* **90**, 015005 (2018). Doi: 10.1103/RevModPhys.90.015005
18. Němec, P., Fiebig, M., Kampfrath, T. & Kimel, A. V. Antiferromagnetic opto-spintronics. *Nat. Phys.* **14**, 229–241 (2018). Doi: 10.1038/s41567-018-0051-x
19. Li, J. et al. Spin current from sub-terahertz-generated antiferromagnetic magnons. *Nature* **578**, 70–74 (2020). Doi: 10.1038/s41586-020-1950-4
20. Vaidya, P. et al. Subterahertz spin pumping from an insulating antiferromagnet. *Science* **368**, 160–165 (2020). Doi: 10.1126/science.aaz4247
21. Han, J., Cheng, R., Liu, L., Ohno, H. & Fukami, S. Coherent antiferromagnetic spintronics. *Nat. Mater.* (2023). Doi: 10.1038/s41563-023-01492-6
22. Oka, T. & Aoki, H. Photovoltaic Hall effect in graphene. *Phys. Rev. B* **79**, 81406 (2009). Doi: 10.1103/PhysRevB.79.081406
23. Lindner, N. H., Refael, G. & Galitski, V. Floquet topological insulator in semiconductor quantum wells. *Nat. Phys.* **7**, 490–495 (2011). Doi: 10.1038/nphys1926
24. Wang, Y. H., Steinberg, H., Jarillo-Herrero, P. & Gedik, N. Observation of Floquet-Bloch states on the surface of a topological insulator. *Science* **342**, 453–457 (2013). Doi: 10.1126/science.1239834
25. Mentink, J. H., Balzer, K. & Eckstein, M. Ultrafast and reversible control of the exchange interaction in Mott insulators. *Nat. Commun.* **6**, 6708 (2015). Doi: 10.1038/ncomms7708

26. McIver, J. W. et al. Light-induced anomalous Hall effect in graphene. *Nat. Phys.* **16**, 38–41 (2020). Doi: 10.1038/s41567-019-0698-y
27. Shan, J. -Y. et al. Giant modulation of optical nonlinearity by Floquet engineering. *Nature* **600**, 235–239 (2021). Doi: 10.1038/s41586-021-04368-4
28. Zhou, S. et al. Pseudospin-selective Floquet band engineering in black phosphorus. *Nature* **614**, 75–80 (2023). Doi: 10.1038/s41586-022-05610-3
29. Ito, S. et al. Build-up and dephasing of Floquet–Bloch bands on subcycle timescales. *Nature* (2023). Doi: 10.1038/s41586-023-05850-x
30. Kapitza, P. L. Dynamic stability of a pendulum when its point of suspension vibrates. *Sov. Phys. JETP* **21**, 588–592 (1951).
31. Citro, R. et al. Dynamical stability of a many-body Kapitza pendulum. *Ann. Phys.* **360**, 694–710 (2015). Doi: 10.1016/j.aop.2015.03.027
32. Bukov, M., D’Alessio, L. & Polkovnikov, A. Universal high-frequency behavior of periodically driven systems: from dynamical stabilization to Floquet engineering. *Adv. Phys.* **64**, 139–226 (2015). Doi: 10.1080/00018732.2015.1055918
33. Blank, T. G. H. et al. Empowering control of antiferromagnets by THz-induced spin coherence. *Phys. Rev. Lett.* **131**, 096701 (2023). Doi: 10.1103/PhysRevLett.131.096701
34. Zhang, Z. et al. Terahertz field-driven magnon upconversion in an antiferromagnet. *Nat. Phys.* **20**, 788–793 (2024). Doi: 10.1038/s41567-023-02350-7
35. Zhang, Z. et al. Terahertz field-induced nonlinear coupling of two magnon modes in an antiferromagnet. *Nat. Phys.* **20**, 801–806 (2024). Doi: 10.1038/s41567-024-02386-3
36. Leenders, R. A., Afanasiev, D., Kimel, A. V. & Mikhaylovskiy R. V. Canted spin order as a platform for ultrafast conversion of magnons. *Nature* **630**, 335–339 (2024). Doi: 10.1038/s41586-024-07448-3
37. Mukai, Y., Hirori, H., Yamamoto, T., Kageyama, H. & Tanaka, K. Nonlinear magnetization dynamics of antiferromagnetic spin resonance induced by intense terahertz magnetic field. *New J. Phys.* **18**, 013045 (2016). Doi: 10.1088/1367-2630/18/1/013045

38. Zhang, Z. et al. Generation of third-harmonic spin oscillation from strong spin precession induced by terahertz magnetic near fields. *Nat. Commun.* **14**, 1795 (2023). Doi: 10.1038/s41467-023-37473-1
39. Fitzky, G., Nakajima, M., Koike, Y., Leitenstorfer, A. & Kurihara, T. Ultrafast control of magnetic anisotropy by resonant excitation of 4f electrons and phonons in $\text{Sm}_{0.7}\text{Er}_{0.3}\text{FeO}_3$. *Phys. Rev. Lett.* **127**, 107401 (2021). Doi: 10.1103/PhysRevLett.127.107401
40. Bayat, K., Choy, J., Baroughi, M. F., Meesala, S. & Loncar, M. Efficient, uniform, and large area microwave magnetic coupling to NV centers in diamond using double split-ring resonators. *Nano Lett.* **14**, 1208–1213 (2014). Doi: 10.1021/nl404072s
41. Polley, D., Zhou Hagström, N., von Korff Schmising, C., Eisebitt, S. & Bonetti, S. Terahertz magnetic field enhancement in an asymmetric spiral metamaterial. *J. Phys. B* **51**, 224001 (2018). Doi: 10.1088/1361-6455/aae579
42. Mashkovich, E. A. et al. Terahertz optomagnetism: Nonlinear THz excitation of GHz spin waves in antiferromagnetic FeBO_3 . *Phys. Rev. Lett.* **123**, 157202 (2019). Doi: 10.1103/PhysRevLett.123.157202
43. Weiss, M. A. et al. Discovery of ultrafast spontaneous spin switching in an antiferromagnet by femtosecond noise correlation spectroscopy. *Nat. Commun.* **14**, 7651 (2023). Doi: 10.1038/s41467-023-43318-8
44. de Jong, J. A., Kimel, A. V., Pisarev, R. V., Kirilyuk, A. & Rasing, T. Laser-induced ultrafast spin dynamics in ErFeO_3 . *Phys. Rev. B* **84**, 104421 (2011). Doi: 10.1103/PhysRevB.84.104421
45. Hortensius, J. R. et al. Coherent spin-wave transport in an antiferromagnet. *Nat. Phys.* **17**, 1001–1006 (2021). doi: 10.1038/s41567-021-01290-4
46. Kirilyuk, A., Kimel, A. V. & Rasing, Th. Ultrafast optical manipulation of magnetic order. *Rev. Mod. Phys.* **82**, 2731–2784 (2010). Doi: 10.1103/RevModPhys.82.2731
47. Kang, S. et al. Coherent many-body exciton in van der Waals antiferromagnet NiPS_3 . *Nature* **583**, 785–789 (2020). Doi: 10.1038/s41586-020-2520-5
48. Bae, Y. J. et al. Exciton-coupled coherent magnons in a 2D semiconductor. *Nature* **609**, 282–286 (2022). Doi: 10.1038/s41586-022-05024-1

49. Jungwirth, T., Marti, X., Wadley, P. & Wunderlich, J. Antiferromagnetic spintronics. *Nat. Nanotech.* **11**, 231–241 (2016). Doi: 10.1038/nnano.2016.18
50. Salikhov, R. et al. Coupling of terahertz light with nanometre-wavelength magnon modes via spin–orbit torque. *Nat. Phys.* **19**, 529–535 (2023). Doi: 10.1038/s41567-022-01908-1

Methods

General sample properties

At room temperature, the two magnetizations $\mathbf{m}_i$ ($i = 1, 2$) of the iron sublattices in $\text{Sm}_{0.7}\text{Er}_{0.3}\text{FeO}_3$ are mainly antiferromagnetically aligned along the c -axis (with a slight canting angle $\beta_0 \sim 0.6^\circ$ due to the Dzyaloshinskii–Moriya field) and exhibit a spontaneous magnetization $\mathbf{M} = (\mathbf{m}_1 + \mathbf{m}_2)/2$ along the a -axis^{39,51}. During our pump–probe experiments, the sample was in a DC magnetic field of about 2 mT to prepare an initial magnetization with an inclination towards the $+c$ -axis. In this antiferromagnet, a spin reorientation transition (SRT) occurs in the temperature range from $T_L = 310$ K to $T_H = 330$ K, where T_L and T_H are referred to as the lower and higher critical temperatures of the SRT, respectively. For temperatures $T < T_L$, $\mathbf{L}$ ($\mathbf{M}$) is parallel to the c -axis (a -axis), and for $T_H < T$, $\mathbf{L}$ ($\mathbf{M}$) is parallel to the a -axis (c -axis). The rotation of $\mathbf{L}$ ($\mathbf{M}$) from one axis to the other in the temperature range $T_L \leq T \leq T_H$ occurs in the a – c plane. This SRT process can be described by a change in the anisotropy potential with T . In the absence of the DC magnetic field, the anisotropy potential has the following normalized form:

$$W = \frac{ab}{2} \sin^2 \theta + \frac{a}{4} \sin^4 \theta, \quad (6)$$

where the variable θ is the angle between $\mathbf{L}$ and the a -axis, which is equal to the angle between $\mathbf{M}$ and the c -axis. The angular positions of the two potential minima around $\theta = \pi/2$ in the temperature range $T_L \leq T \leq T_H$ are given by $\theta_L = \arcsin [-b]^{1/2}$ and $\theta_R = \pi - \arcsin [-b]^{1/2}$, where $b = (T - T_H)/(T_H - T_L)$.

THz-pump–optical-probe measurement

The near-infrared (NIR) light source of our pump–probe measurement system shown in Extended Data Fig. 1 is an amplified Ti:sapphire laser with a repetition rate of 1 kHz, a

central wavelength of 800 nm, a pulse duration of 80 fs and a pulse energy of 7 mJ. By using a beam splitter, the laser output is first divided into two NIR beams. The weaker NIR beam with 1% of the total output power is used to probe the sample response. Furthermore, each pulse of the stronger NIR beam is divided into two pulses with equal power. A delay stage is used to control the delay time between these two strong femtosecond laser pulses. The two pulses are used to generate two intense THz pulses at different positions of the LiNbO₃ crystal based on optical rectification using the tilted-pump-pulse-front scheme^{52–54}, and then these two THz pulses are superimposed at the sample (Fig. 2a). This pair of THz pulses is referred to as the THz double pulse. The use of such a THz double-pump pulse also allows us to control the temporal profile of the THz magnetic near-field to a certain extent (Supplementary Section III). The THz pulses are focused on the triangular tail of the gold microstructure by using an off-axis parabolic mirror with a focal length of 50 mm, resulting in a spot diameter of $\approx 300 \mu\text{m}$ (full width at half-maximum). The maximum peak electric fields of the two single-cycle THz pulses before the low-pass filter (LPF) shown in Extended Data Fig. 1 were 1.0 MV/cm and 0.75 MV/cm. These THz pulses also contain frequency components that are different from the resonance frequency of the spiral-shaped microstructure, and we removed them using the LPF (cut-off frequency: 0.68 THz). This leads to a reduction in the maximum peak electric field, and the predictions for the magnetic field values presented in the main text take the effect of filtering into account. The time delay Δt between the two strong femtosecond laser pulses for THz generation was set to 4.48 ps, which allows us to avoid saturation and a reduction in the THz generation efficiency (Supplementary Section III). For the time-resolved Faraday ellipticity measurements, another delay stage is used to control the time delay between the THz pump pulse and the NIR probe pulse. The linearly polarized probe pulse is focused at the center of spiral structure by a 50 $\times$ objective lens (spot diameter $\sim 1.2 \mu\text{m}$), which enables us to measure local changes in the ellipticity

angle η defined in inset b of Extended Data Fig. 1. The polarization directions of the THz light and the probe light are parallel and almost along the b -axis. The upper right-hand side of Extended Data Fig. 1 shows the balanced detector (combined with a quarter-wave plate and a Wollaston prism) used to measure the ellipticity angle of the reflected probe pulse. The 70- μm -thick $\text{Sm}_{0.7}\text{Er}_{0.3}\text{FeO}_3$ crystal was sufficiently transparent for the used NIR probe pulses and a home-made coil heater was used to increase the sample temperature and determine the temperature dependences of the signals.

Description of the q-FM mode using the sine-Gordon equation

To explain the slow variation of $\Delta\eta$, we need to consider the behaviour of the q-FM mode, because the q-AFM mode only induces a fast variation of $\Delta\eta$. The q-FM mode and the spin-switching dynamics can be described by using a single variable, the angle θ , i.e., $\Delta\eta_{\text{FM}} \propto \Delta\cos\theta = \cos\theta - \cos\theta_L$ (Supplementary Section XIII). The spin dynamics related to the q-FM mode can be described by the sine-Gordon equation given in Eq. (7) below^{9,10,55}, while the equation of motion of the q-AFM mode is given in Eq. (9). These two equations originate from the Landau–Lifshitz–Gilbert (LLG) equation in combination with a two-sublattice model (Supplementary Section IV). The q-FM mode represents a pendulum motion of $\mathbf{L}$ in the a - c plane, and thus can be described by using the angle θ between the a -axis and $\mathbf{L}$,

$$\ddot{\theta} + c\dot{\theta} = -\chi\sin\theta[B_{\text{THz}}^{\text{NF}}(t) + B_{\text{DC}}] - a\sin\theta\cos\theta[b + \sin^2\theta] - 2\gamma B_{\text{THz}}^{\text{NF}}(t)\dot{\phi}\cos\theta\sin\theta - \gamma\dot{B}_{\text{TH}}^{\text{NF}}(t)\phi\cos\theta\sin\theta, \quad (7)$$

where we used the following definitions:

$$a = \frac{4K_2\gamma^2 H_E}{M_{\text{Fe}}}, b = \frac{T - T_H}{T_H - T_L}, c = 2\gamma H_E \alpha_{\text{LLG}}, \chi = \gamma^2 H_D. \quad (8)$$

In Eq. (7), the THz magnetic pulse $B_{\text{THz}}^{\text{NF}}(t)$ can be replaced by $B_{\text{env}}^{\text{NF}}(t)\cos(\omega t)$, that is, a multiplication of a slowly varying envelope function with a fast-oscillating component, and B_{DC} is the bias magnetic field. The first and second terms on the right-side hand are due to the Zeeman torque (of the magnetic field) and the anisotropy potential, respectively. The third term is due to coupling between the q-AFM and q-FM modes, and the fourth term is also related to coupling. Equation (8) provides the definitions of the parameters a , b , c , and χ : H_{E} is the symmetric exchange field, H_{D} is the antisymmetric exchange field, γ is the gyromagnetic ratio, M_{Fe} is the magnetization of one Fe^{3+} sublattice, K_2 is the temperature-independent anisotropic exchange field between the iron and rare-earth ions, $T_{\text{L}} = 310$ K and $T_{\text{H}} = 330$ K are the lower and higher critical temperatures of the SRT, respectively, and α_{LLG} is a damping parameter.

The q-AFM mode represents a pendulum motion of $\mathbf{L}$ out of the a - c plane, and thus can be described by the angle between $\mathbf{L}$ and the a - c plane, $\varphi\cos\theta$, where φ is defined as the angle between the a -axis and the projection of $\mathbf{L}$ on the a - b plane. The equation describing the q-AFM mode thus becomes

$$\ddot{\varphi}\cos\theta + c\dot{\varphi}\cos\theta = -\omega_{\text{AFM}}^2\varphi\cos\theta - \gamma\dot{B}_{\text{THz}}^{\text{NF}}(t)\cos\theta + 2\gamma B_{\text{THz}}^{\text{NF}}(t)\dot{\theta}\sin\theta, \quad (9)$$

where ω_{AFM} is the intrinsic frequency of the q-AFM mode. This frequency is determined by the antiferromagnetic exchange interaction, the Dzyaloshinskii–Moriya interaction, the anisotropy interactions and the bias magnetic field. The second and third terms are due to the Zeeman torque of the magnetic field and coupling to the q-FM mode, respectively.

Data availability

Source data are available for this paper. All other data that support the plots within this paper and other findings of this study are available from the corresponding author upon request.

Code availability

The simulation codes presented in this paper are available from the corresponding author upon request.

Methods-only references

51. Zhao, X. et al. Crystal growth and spin reorientation transition in $\text{Sm}_{0.4}\text{Er}_{0.6}\text{FeO}_3$ orthoferrite. *Solid State Commun.* **43**, 231–232 (2016). Doi: 10.1016/j.ssc.2016.02.002
52. Hebling, J., Almási, G., Kozma, I. Z. & Kuhl, J. Velocity matching by pulse front tilting for large area THz-pulse generation. *Opt. Express* **10**, 1161–1166 (2002). Doi: 10.1364/OE.10.001161
53. Yeh, K.- L., Hoffmann, M. C., Hebling, J. & Nelson, K. A. Generation of 10 μJ ultrashort terahertz pulses by optical rectification. *Appl. Phys. Lett.* **90**, 171121 (2007). Doi: 10.1063/1.2734374
54. Hirori, H., Doi, A., Blanchard, F. & Tanaka, K. Single-cycle terahertz pulses with amplitudes exceeding 1 MV/cm generated by optical rectification in LiNbO_3 . *Appl. Phys. Lett.* **98**, 091106 (2011). Doi: 10.1063/1.3560062
55. Zvezdin, A. K., Dynamics of domain walls in weak ferromagnets. *JETP Lett.* **29**, 553–556 (1979).

Figure 1

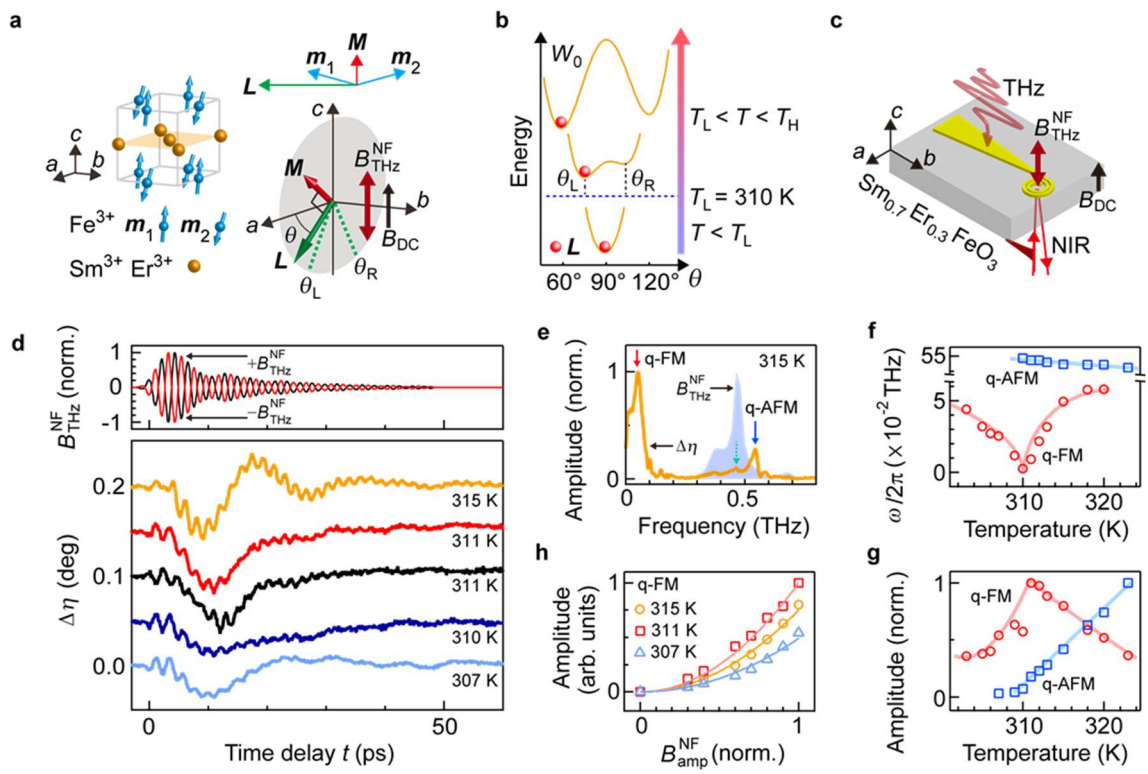


Figure 2

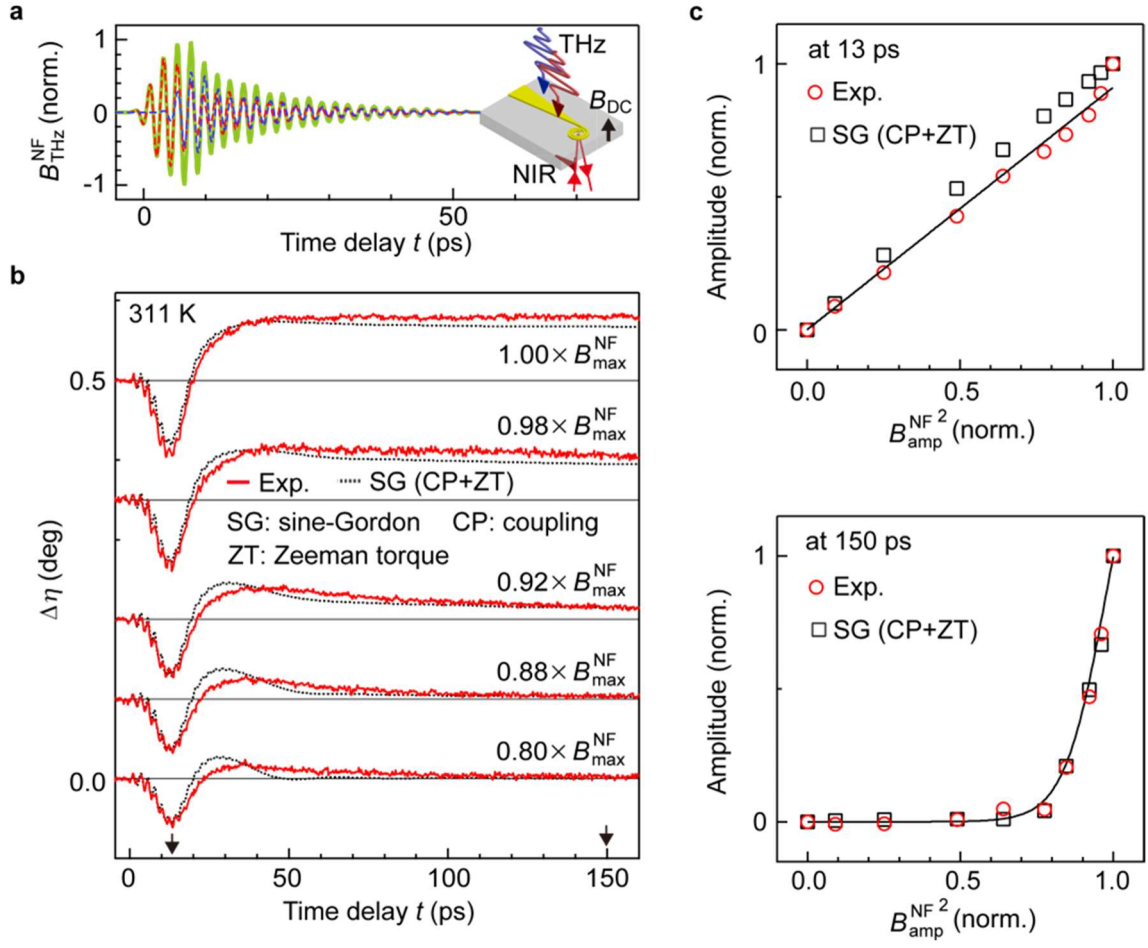


Figure 3

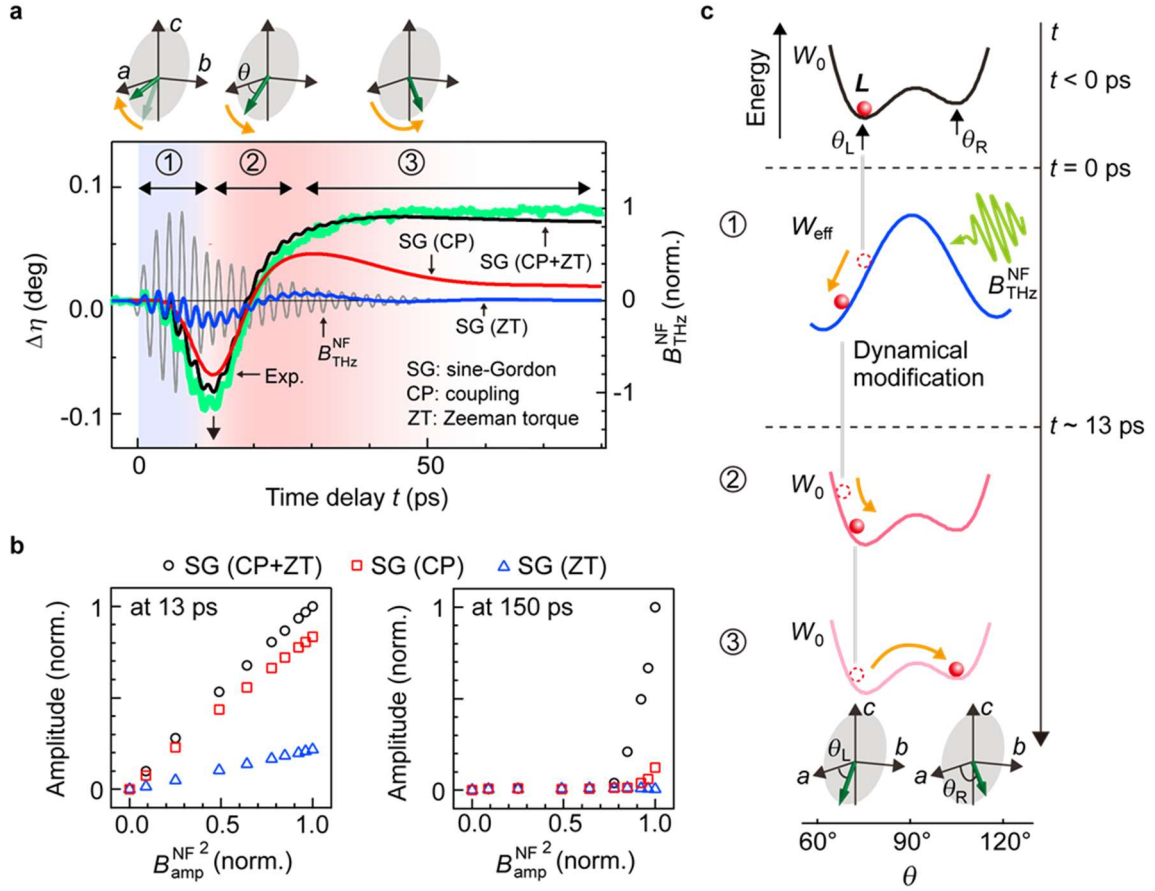
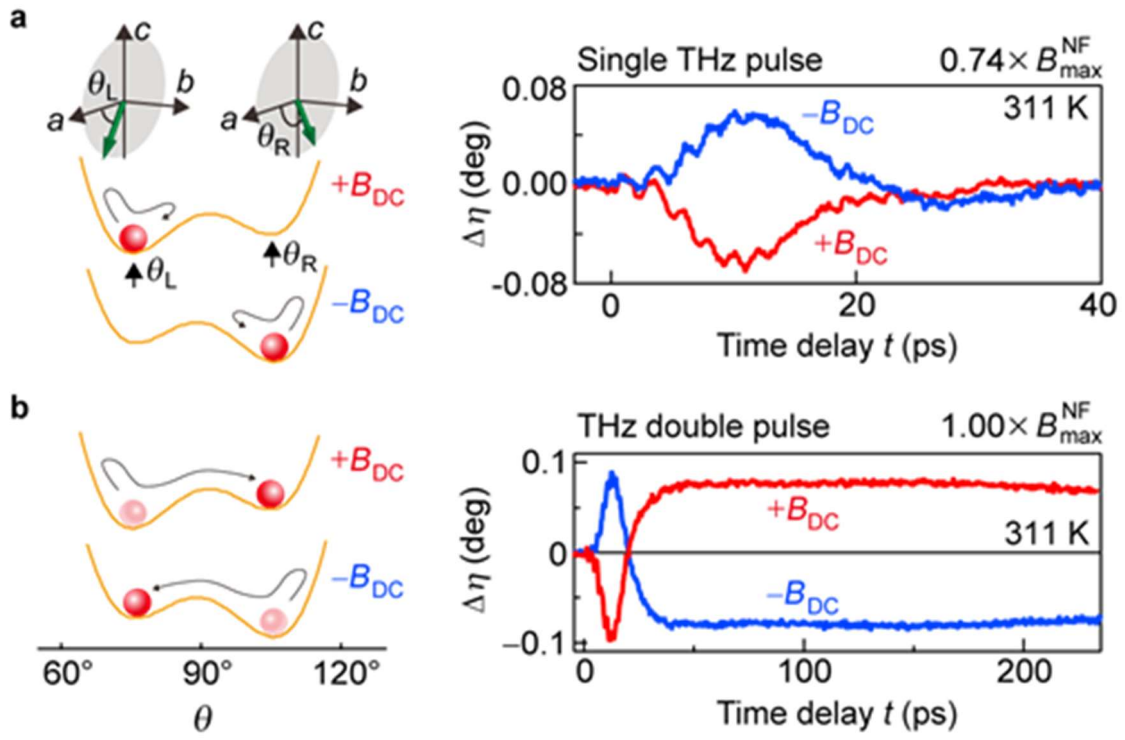
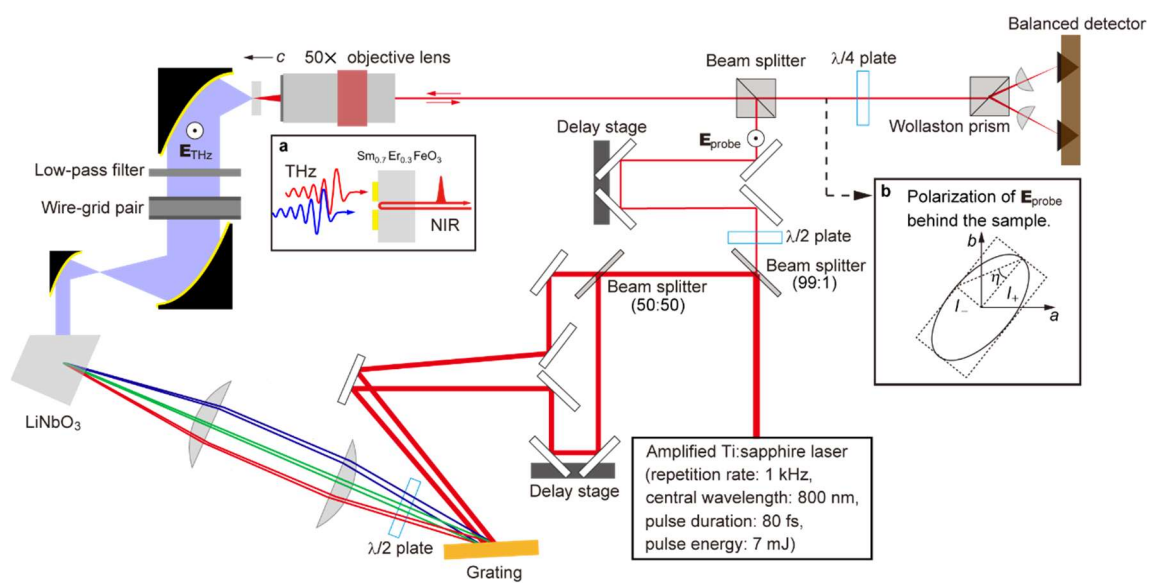


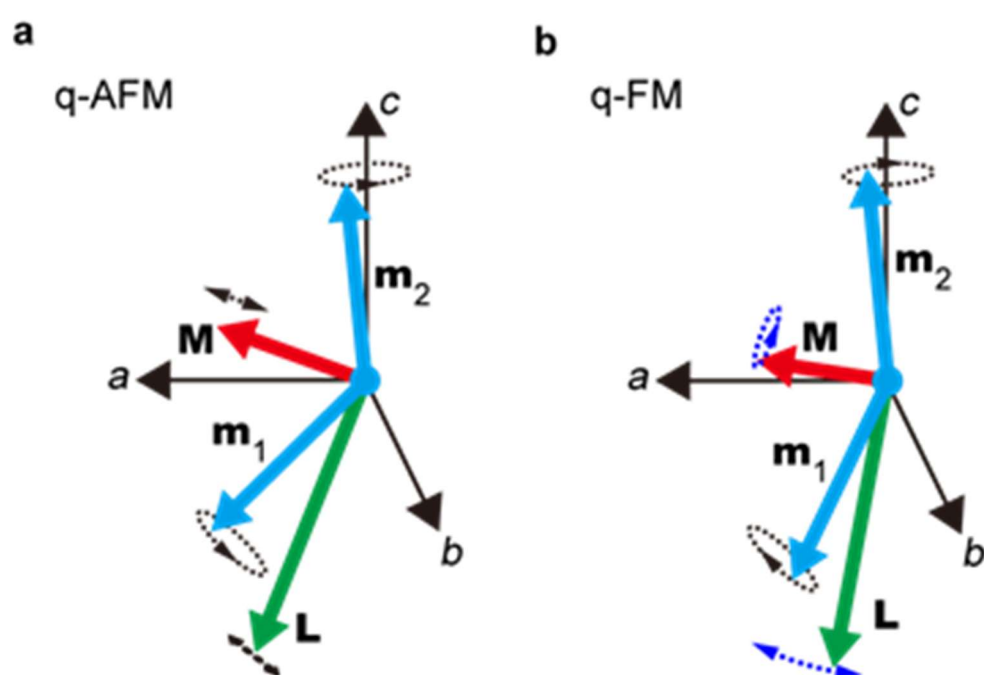
Figure 4



Extended Data Figure 1



Extended Data Figure 2



Extended Data Figure 3

