Nonlinear coupled magnonics: Terahertz field-driven magnon upconversion

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Abstract

Tailored light excitation and nonlinear control of lattice vibrations have emerged as powerful strategies to manipulate the properties of quantum materials out of equilibrium. Generalizing the exploitation of coherent phonon-phonon interactions to nonlinear couplings among other types of collective modes would open unprecedented opportunities in the design of novel dynamic functionalities in solids. For example, the collective excitations of magnetic order – magnons – can efficiently transfer information via spin current flow\(^1\), and their coherent and nonlinear control would provide an attractive route to achieve faster signal processing for next-generation information technologies. Here, we discover that intense terahertz (THz) fields can initiate processes of magnon up-conversion via coherent magnon-magnon interactions – a phenomenon that opens the paradigm of *nonlinear coupled magnonics*. By using a suite of advanced spectroscopic tools, including a newly demonstrated two-dimensional (2D) THz polarimetry technique enabled by single-shot detection, we unveil the unidirectional nature of coupling between distinct magnon modes of a canted antiferromagnet. Calculations of spin dynamics further suggest that this coupling is a universal feature of antiferromagnets with canted magnetic moments. These results demonstrate a route to inducing desirable energy transfer pathways between coherent magnons in solids and pave the way for a new era in the development of magnonic signal processing devices.
**Main Text**

Nonlinear dynamics govern many exotic phenomena in complex systems, ranging from chaos and turbulence to neural activity. In condensed matter physics, a notable manifestation of nonlinear dynamics is *nonlinear phononics*. Intense terahertz (THz) or infrared laser pulses, derived from tailored light sources, can drive the collective vibrational modes of a lattice (phonons) into a coherent nonlinear regime, activating specific phonon-phonon interactions that would be silent otherwise\(^2,3\). These couplings have been found to play a key role in many remarkable phenomena, including transient enhancement of superconductivity\(^4\), nucleation of macroscopic polarization\(^5,6\), and manipulation of magnetic states\(^7\text{-}11\). In principle, similar coherent couplings are expected among other types of collective excitations, including magnons, plasmons, phasons, and amplitudons\(^12\text{-}14\), but their exploitation has thus far been hindered by weak interactions with light or fast decoherence. Addressing this challenge could establish an unprecedented paradigm in the nonequilibrium control of quantum materials and open the door to next-generation information technologies based on coupled coherent excitations.

Here we unveil a readily accessible coupling mechanism by studying the magnon modes of a canted antiferromagnet. We observe magnon upconversion processes mediated by nonlinear magnetic interactions, illustrating what we term *nonlinear coupled magnonics*. This concept is distinct from spin dynamics in the non-perturbative
regime\textsuperscript{15,16}, as well as previous observations of magnon harmonics, which do not involve coupling between distinct magnon modes\textsuperscript{17,18}.

The underlying physics of nonlinear coupled magnonics can be rationalized through a spring-pendulum model (Figure 1a). This simple mechanical system gives rise to rich dynamics first studied by Vitt and Gorelik in 1933\textsuperscript{19} and extensively discussed since in the context of nonlinear systems\textsuperscript{20}. In this model, a small mass \( m \) is suspended from a fixed point by a massless spring, which can both stretch and swing in a two-dimensional plane. Let \( L \) be the length of the spring at equilibrium under gravity, \( k \) be the spring constant, \( r \) be the length change of the stretched spring and \( \theta \) be the angle between the pendulum and the vertical axis. The resulting Hamiltonian of the system can be written as

\[
\mathcal{H} = \frac{1}{2} m \left( \dot{r}^2 + (L + r)^2 \dot{\theta}^2 \right) + \frac{1}{2} k \left( \frac{mg}{k} + r \right)^2 - mg(L + r) \cos \theta. \tag{1}
\]

Following the lowest order of the two degrees of freedom, \( r \) and \( \theta \), the dynamics are described by two independent equations of motion

\[
\ddot{r} + \Omega^2_B r = 0
\]

\[
\ddot{\theta} + \Omega^2_S \theta = 0, \tag{2}
\]
where \( \Omega_B = k/m \) and \( \Omega_S = g/L \) are the respective frequencies of the spring bouncing mode, in which elasticity is the restoring force, and the swinging mode, in which the system acts as a conventional pendulum. These two modes remain orthogonal to first order; yet, retaining up to the second order in the equations of motion reveals emergent nonlinear couplings:

\[
\ddot{r} + \Omega_B^2 r + \frac{1}{2} g \dot{\theta}^2 - (L + r) \dot{\theta}^2 = 0 \tag{3}
\]
\[
\ddot{\theta} + \Omega_S^2 \theta - \frac{g r \theta}{L^2} + 2 \dot{r} \dot{\theta}/(L + r) = 0.
\]

The second-order nonlinear terms in the first equation are proportional to \( \theta^2 \) and \( \dot{\theta}^2 \), indicating that a deviation in \( \theta \) alone induces a change in \( r \), while those in the second equation are proportional to \( r \theta \) and \( \dot{r} \dot{\theta} \), indicating that only deviations in the combination of \( r \) and \( \theta \) will induce further changes in \( \theta \). Thus, starting from equilibrium \( (r = \dot{r} = 0, \theta = \dot{\theta} = 0) \), the coupling is unidirectional: the pendulum swinging leads to the spring bouncing, while the bouncing motion does not induce any swinging.

This toy model is closely analogous to the magnon dynamics in a canted antiferromagnet, with two canted sublattice spins \( (S_1 \text{ and } S_2) \) in a magnetic unit cell. The steady-state spin Hamiltonian of such a system can be written as

\[
\mathcal{H}_0 = nJS_1 \cdot S_2 + nD \cdot (S_1 \times S_2) - \sum_{i=1,2} (K_X S_{ix}^2 + K_Z S_{iz}^2), \tag{4}
\]
where $n$ is the number of nearest neighboring spin sites, $J$ is the exchange constant that accounts for the antiferromagnetic ground state, $D$ is the antisymmetric exchange parameter that scales with the Dzyaloshinskii–Moriya interaction, which gives rise to the canting of the sublattice magnetic moments (and thereby induces a net magnetization $M$) and $K_X$, $K_Z$ are magneto-crystalline anisotropies that determine the orientation of $M$. Solving the equations of motion to first order gives rise to two magnon modes

\[
\dot{\sigma}_j = C^{\sigma}_{ji}\sigma_i \quad \dot{\gamma}_j = C^{\gamma}_{ji}\gamma_i.
\]

(5)

Here $\sigma$ and $\gamma$ are the quasi-ferromagnetic (qFM) and the quasi-antiferromagnetic (qAFM) mode coordinates; $C^{\sigma}_{ji}$ and $C^{\gamma}_{ji}$ are constants (see the Methods section). At small amplitudes, the qFM mode corresponds to a precession of the magnetization orientation, while the qAFM mode represents a periodic modulation of the magnetization amplitude. At finite amplitudes when the sublattice spins are significantly deflected from their equilibrium positions, it is necessary to consider the nonlinear couplings in the bases of both modes:

\[
\dot{\sigma}_j = C^{\sigma}_{ji}\sigma_i + D_{ijk}\sigma_i\gamma_k
\]

\[
\dot{\gamma}_j = C^{\gamma}_{ji}\gamma_i + E_{ijk}\sigma_i\sigma_k,
\]

(6)

where $D_{ijk}$ and $E_{ijk}$ are coefficients for the second-order terms. Since the dominant
nonlinear term driving of the qFM mode is proportional to $\sigma_i \gamma_k$, an excitation of the qAFM mode alone cannot launch an oscillatory response of the qFM mode. For the qAFM mode on the other hand, the nonlinear term $\sigma_i \sigma_k$ provides a coherence transfer mechanism to drive the qAFM mode. Even if the amplitude of the qAFM mode $\gamma_j$ is initially zero, the second-order interaction of the qFM mode (i.e., $\sigma_i \sigma_k$) induces an oscillation of the qAFM mode (see Fig. 1b).

To unravel this exotic physics for the first time, we performed a series of nonlinear THz experiments on the model canted antiferromagnet ErFeO$_3$ (EFO), which crystallizes in a distorted orthorhombic structure. Below the Néel temperature $T_N = 643$ K, the spins of neighboring Fe ions order antiferromagnetically, yet slightly cant due to the Dzyaloshinskii–Moriya interaction, yielding a net magnetic moment. As a result, magnon excitation follows specific selection rules: the qAFM mode can only be resonantly driven by a THz field whose magnetic field component is oriented along $M$ while the excitation of the qFM mode requires a magnetic field component perpendicular to $M$.\textsuperscript{21,22}

We begin by studying the driven magnon response at room temperature using THz electron spin resonance (ESR) polarimetry. We focus a linearly polarized, single-cycle THz pulse with a peak magnetic field $H_{\text{THz}}$ of $\sim 0.15$ T onto a (010)-cut single-domain EFO crystal, which allows both magnon modes to be accessible. The magnetic
field component of the THz pump pulse is first aligned with respect to either the $a$ ($\perp \mathbf{M}$) or $c$ ($\parallel \mathbf{M}$) crystallographic axis to selectively drive the qFM or the qAFM mode, respectively. To measure the THz-induced free-induction decay (FID) signals, $\mathbf{H}_{\text{det}}$, of the driven modes, we use a recently developed single-shot detection method\textsuperscript{23,24} which enables the rapid acquisition of THz waveforms with high signal-to-noise ratio. A wire-grid polarizer is placed in front of the detector to select the emitted THz field either parallel or perpendicular to that of the THz excitation pulses ($\mathbf{H}_{\text{det}} \parallel \mathbf{H}_{\text{THz}}$ or $\mathbf{H}_{\text{det}} \perp \mathbf{H}_{\text{THz}}$) (See Fig. 2a and Extended Data Fig. 2). In the linear response, driving a specific magnon mode along either the $a$ or $c$ crystallographic axis can generate FID signals only parallel to the pump magnetic field ($\mathbf{H}_{\text{det}} \parallel \mathbf{H}_{\text{THz}}$) and none perpendicular to it ($\mathbf{H}_{\text{det}} \perp \mathbf{H}_{\text{THz}}$). Figure 2b shows the time-domain FID signals for both excitation and detection configurations. As expected, the two FID signals in the parallel configuration reveal the coherent excitation of each magnon mode (see Figure 2b, top panel), and the Fourier spectra in Fig. 2c matches the previously reported magnon frequencies at room temperature (qFM, 0.38 THz and qAFM, 0.67 THz)\textsuperscript{21}. However, when the THz pump field drives the qFM mode ($\mathbf{H}_{\text{THz}} \parallel a$), we clearly observe the excitation of the qAFM mode revealed by the emitted signal in the perpendicular configuration, which is not predicted in linear response (see Figure 2b, dark red trace). Conversely, no qFM response is found when the THz magnetic field component is oriented along the $a$ axis to excite the qAFM mode (see Figure 2b, light grey trace).
We achieve a comprehensive understanding of the magnon symmetry by rotating the sample and repeating the same measurements every 5°. The oscillation amplitude of each magnon mode is extracted from the Fourier spectra of the FID signals for both detection configurations and plotted as a function of the azimuthal angle, θ, i.e. the angle between the THz magnetic field orientation and the a axis in the ac plane. For the qFM mode, the parallel polarimetry pattern shows two-fold rotation symmetry (cos² θ) and reaches the largest amplitudes at θ = 0, with H_{THz} and H_{det} along the crystallographic a axis. Since the perpendicular detection configuration eliminates any FID signal that arises when the THz magnetic field directly drives the qFM mode, the responses peak at 45° off-axes, proportional to |cos θ sin θ|. For the qFM mode, both polarimetry patterns are well captured by only considering the linear Zeeman interaction with the THz magnetic field.\textsuperscript{22, 25} For the qAFM mode, the parallel polarimetry pattern also shows two-fold rotation symmetry (sin² θ), with maximum signal amplitude when H_{THz} and H_{det} are along the c axis. However, the perpendicular polarimetry pattern cannot be captured by the linear response alone. This is already apparent from the raw data in Fig. 2d: the qAFM response vanishes at 90° when the qAFM mode is directly driven by H_{THz} || c and its corresponding FID emission is blocked by the wire-grid polarizer, but the qAFM response is nonzero at 0° when H_{THz} || a drives the qFM mode. To express the full qAFM perpendicular detection pattern analytically, we perform a fit to the sum of both the linear response (|cos θ sin θ|) and the second-order magnon upconversion.
process driven by the excitation of the qFM mode \((\cos^2 \theta \cos \theta)\) and find an excellent match with the data.

To further distinguish the nature of the resonant excitation and the nonlinear magnon upconversion process, we measure the dependence of the magnon amplitudes on the peak magnetic field of the pump pulse. Since the parallel polarimetry measurement eliminates the nonlinear coupling response, both modes that are detected this way show a linear dependence on the magnetic field (see Fig. 2e). In contrast, when we drive the \(a\)-axis qFM mode with the THz pump and measure the \(c\)-axis qAFM response, we find that it scales as the square of the pump magnetic field (see Fig. 2f). This quadratic field dependence is consistent with expectations from the nonlinear equations discussed earlier.

To demonstrate that the magnon upconversion process is indeed the only excitation pathway that can rationalize our observations, we perform a 2D THz coherent spectroscopy measurement, which has proven to be an effective tool to uncover the origin of nonlinear field-induced responses.\(^{18,26-29}\) By adding a second, time-delayed THz pulse (see Fig. 3a) and recording the time-domain THz field emission with variable inter-pulse delay, \(\tau\), we extract the nonlinear THz signal generated by both pulses and perform a 2D Fourier transform to yield a frequency-frequency correlation map of the nonlinear magnonic response. The resulting 2D THz spectrum with \(H_{\text{THz}} \parallel a\) detected
in the perpendicular configuration shows a strong cross-peak with excitation and emission frequencies corresponding to the qFM and qAFM magnon modes, respectively. This peak unambiguously confirms the nonlinear upconversion process: excitation of the qFM magnon mode drives the qAFM magnon mode.

We further analyze this response through a 2D THz polarimetry measurement of this off-diagonal magnon cross-peak. Similar to our linear polarimetry measurements, we utilize our ability to rapidly measure time-dependent THz fields via our single-shot THz measurement technique to enable 2D THz polarimetry, in which we rotate the azimuthal angle \( \theta \) (see Fig. 3a) and collect 2D spectra at 5\(^\circ\) increments. The resulting polarimetry patterns of the magnon cross-peak amplitude at \((\Omega_{qFM}, \Omega_{qAFM})\) are shown in Fig. 3cd for both parallel and perpendicular detection geometries. The nonlinear upconversion signal shows two-fold symmetry in the perpendicular configuration and is strongest when \( H_{THz} \parallel a \) driving the qFM mode, whereas in the parallel configuration it is a distorted clover pattern with maximum amplitude close to 30\(^\circ\). These polarimetry patterns can be well fit with functions proportional to \( |\cos^2 \theta \sin \theta| \) and \( |\cos^2 \theta \cos \theta| \), respectively. This confirms that the upconverted THz field emission at \( \Omega_{qAFM} \) is second order in the driving field of the qFM magnon mode, i.e. proportional to the square of the THz magnetic field component along the \( a \) axis, while the emission orientation is that of the qAFM magnon, along the \( c \) axis. We note that we do not observe the corresponding downconversion peak, i.e. qAFM \( \rightarrow \) qFM, in the 2D THz spectra at any azimuthal
angle, which further confirms this coupling to be asymmetric.

Finally, we study the robustness of the magnon upconversion process across a wide range of temperatures. At room temperature, EFO has a net magnetic moment along the crystallographic $c$ axis. As the crystal is cooled, the material undergoes a spin reorientation transition (SRT) between 96-87 K, which continuously rotates the magnetic moment across the $ac$ plane, such that below 87 K the magnetic moment $\mathbf{M}$ aligns along the $a$ axis.\textsuperscript{21,30} This reorientation of the spins induces a change of the selection rules for the two magnon modes for $k_{\text{THz}} \parallel b$, i.e. $H_{\text{THz}} \parallel c$ and $H_{\text{THz}} \parallel a$ drive the qFM and qAFM modes respectively. The experimentally determined excitation and emission frequencies are shown in Fig. 4b with the corresponding 2D THz spectra at selected temperatures shown in Fig. 4c. (Additional spectra can be found in Extended Data Figs. 3 and 4.) These experimental values are shown together with calculations of the qFM and qAFM mode frequencies (solid lines in Fig. 3b) and the 2D THz spectra (see Fig. 3c bottom row) derived by solving the Landau-Lifshitz-Gilbert (LLG) equation based on a single-site two-sublattice spin Hamiltonian with time-dependent effective magnetic fields (see Methods for simulation details). The excitation and emission frequencies of the magnon upconversion peak are well-matched with the calculated qFM and qAFM mode frequencies and those reported in the literature\textsuperscript{21}. The qAFM frequency is seen to be relatively constant above the SRT but hardens below the transition while the qFM frequency softens near the SRT due to the cancellation of the second-
order anisotropies along $a$ and $c$. Note that even in the region surrounding the SRT where $\Omega_{qAFM}/\Omega_{qFM} > 4$, the conversion efficiency remains significant as the process is still second-order, standing in stark contrast to other higher-order harmonic generation processes. The fact that this magnon upconversion process can be achieved with a broad range of excitation and emission frequency ratios across a wide range of temperatures illustrates the generality of the mechanism of unidirectional coherence transfer via nonlinear coupled magnonics. We note that this process is most efficient if the bandwidth of the THz pulse itself is sufficiently broad that it covers both $\Omega_{qFM}$ and $\Omega_{qAFM}/2$.

The excellent match between the experiments and the simulations also implies that the magnon upconversion is purely activated by the magnetic field of the THz pulse. Therefore, our present study highlights the unique potential of applying THz pulses to control and functionalize the properties of magnetic quantum materials out of equilibrium. For example, by using a multi-cycle THz field with the sum-frequency resonant condition satisfied (i.e., $2\Omega_{qFM} = \Omega_{qAFM}$), one can further strengthen the upconversion process to a leading-order effect. We also anticipate that this concept of nonlinear coupled magnonics will be generally applicable to many other magnetically ordered systems, including atomically thin antiferromagnets and coupled FM/AFM heterostructures, and can be utilized to further extend the frontiers of spintronics and magnonics into the ultrafast nonlinear regime.
Methods

Sample Preparation

Polycrystalline ErFeO$_3$ was synthesized by conventional solid state reaction using Er$_2$O$_3$ (99.9%) and Fe$_2$O$_3$ (99.9%) powders. According to their stoichiometric ratios, original reagents were weighed and then ground in an agate mortar with anhydrous ethanol. After grinding, the mixture was pre-sintered before being transferred back to the agate mortar and reground into powder. The powder was then pressed into round flakes using a mold and electric hydraulic press (YLJ-40T) before the second sintering and the regrinding. The polycrystalline powders were pressed into a bar by a Hydrostatic Press System (HP-M-SD-200) under an apparent pressure of 60 MPa, and then sintered again. Each sintering was performed for 1000 min at a temperature 1280 °C.

Single-crystal ErFeO$_3$ samples were prepared in an optical floating zone furnace (FZ-T-10000-H-VI-P-SH, Crystal Systems Corp.). The sintered polycrystalline rods were used both as the seed crystal and the feed rod. Single crystal growth was performed at a growth rate of 3 mm/h under an airflow of 2 L/min. A high-quality single crystal with a length of about 50 mm and a diameter of about 5 mm was obtained after about 17 hours of growth. The single crystal, shown in Extended Data Fig. 6a, was then cut to obtain the (010)-cut sample for THz measurements. Before each measurement, the sample was magnetized to reinforce the residual magnetization and ensure that a single
magnetic domain is formed. The crystallographic orientation of the single crystal was determined with Laue diffraction (Extended Data Fig. 6b).

**Single-shot time-domain THz spectroscopy**

The majority of the output of a 1 kHz Ti:Sapphire laser amplifier (12 mJ, 35 fs) was split into two equal pulses that were variably delayed and overlapped with a tilted pulse-front geometry in a MgO:LiNbO$_3$ crystal to generate a pair of time-delayed THz pulses (See Extended Data Fig. 1). These pulses were then focused onto the sample before being refocused onto a 2-mm ZnTe crystal to measure the THz waveform via electro-optic (EO) sampling. The remainder of the laser output was first expanded 9× and then reflected off an echelon mirror to generate a spatial array of 500 time-delayed and spatially shifted pulses. The pulses were focused along with the THz pulse(s) onto the EO crystal and then imaged onto a high-speed camera. The THz field-induced birefringence was detected by separating the probe array into two orthogonal polarizations with a balanced detection scheme. With this method, we can retrieve the time-domain THz signal covering 20 ps with a single THz pump laser shot while still operating at the full 1 kHz repetition rate of the laser amplifier.

For THz FID measurements, one of the THz pump arms is chopped at 500 Hz (the other is blocked) and the corresponding THz FID signal averaged over 1000 shots (1 s) is recorded. For 2D THz measurements, a differential chopping scheme is used where
the two THz pulses, \(A\) and \(B\), are chopped at 500 Hz and 250 Hz, respectively. The THz signals \(H_{AB}, H_A, H_B,\) and \(H_0,\) corresponding to traces taken with the respective pulse(s) unblocked, are averaged over 5000 shots (5 s) and recorded as the inter-pump pulse delay, \(\tau,\) is scanned between 3 and 20 ps. The time-domain nonlinear signal, \(H_{NL}(t,\tau),\) is then obtained via

\[
H_{NL}(\tau, t) = H_{AB}(\tau, t) - H_A(\tau, t) - H_B(t) + H_0(t),
\]

which is Fourier transformed with respect to both \(t\) and \(\tau\) to yield the 2D THz spectrum.

Single-shot detection has been utilized in cases where only a limited number of shots is required, such as in the study of photoinduced metastable phase transitions\(^3\) and samples under extreme pulsed magnetic fields\(^4\). In our measurements, single-shot detection of the signal field permits the 2D spectrum to be collected with only one time variable (the time interval between THz pulses) swept. The single-shot THz measurement method was developed earlier to facilitate 2D THz spectroscopy\(^5\) and the approach has been applied recently\(^6,\) but its full capabilities can be exploited most effectively as demonstrated here with the use of several hundred probe pulses to record the time-dependent field at the corresponding number of time points on each shot.

\(THz\ ESR\ polarimetry\)
For both 1D and 2D THz polarimetry measurements, the sample is placed on a motorized rotation stage and the corresponding THz signal is recorded as the azimuthal angle, $\theta$, of the sample orientation relative to the incident THz polarization is rotated. A pair of wire grid polarizers, one placed before and one after the sample, is used to select the component of the transmitted/emitted THz field either parallel or perpendicular to the incident THz polarization that is detected. The data acquisition time for the complete set of 2D polarimetry measurements (144 2D spectra in all, collected with parallel and perpendicular detection configurations at 5° increments of the crystal orientation), was about 22 hours. We note that our previously reported 2D ESR spectra of magnons$^{18}$, collected without single-shot detection, required several days of data acquisition per spectrum; the present study would have required more than 2 years of data acquisition.

*Theory of nonlinear coupled magnonics*

A uniform two-sublattice Hamiltonian, eq. (4), is considered here. For convenience of description, the coordinates $\{X, Y, Z\}$ following the crystallographic axes are rotated to form two new sets of coordinates $\{x_i, y_i, z_i\}$ where the equilibrium orientation of each sublattice spin $S_i$ ($i = 1, 2$) is along $x_i$. The components of each spin can be then
expressed as

\[
\begin{bmatrix}
S_{1x} \\
S_{1y} \\
S_{1z}
\end{bmatrix} =
\begin{bmatrix}
-\cos \beta_0 & 0 & \sin \beta_0 \\
0 & 1 & 0 \\
-\sin \beta_0 & 0 & -\cos \beta_0
\end{bmatrix}
\begin{bmatrix}
S_{1X} \\
S_{1Y} \\
S_{1Z}
\end{bmatrix},
\begin{bmatrix}
S_{2x} \\
S_{2y} \\
S_{2z}
\end{bmatrix} =
\begin{bmatrix}
\cos \beta_0 & 0 & \sin \beta_0 \\
0 & 1 & 0 \\
-\sin \beta_0 & 0 & \cos \beta_0
\end{bmatrix}
\begin{bmatrix}
S_{2X} \\
S_{2Y} \\
S_{2Z}
\end{bmatrix},
\]

where \( \beta_0 \) is the canting angle between \( S_2 \) and the antiferromagnetic easy axis \( X \), which can be determined by \( \tan(2\beta_0) = \frac{nD}{nJ + K_X - K_Z} \), where \( n, J, K_X, \) and \( K_Z \) are parameters previously defined in equation (4). In these new coordinates, at equilibrium, \( S_{ix} = S \), \( S_{iy} = S_{iz} = 0 \).

We then follow the framework previously described in literature\(^{34-36}\) to treat the spin dynamics. We introduce the magnon eigenmode basis as follows:

\[
\sigma_x \equiv S_{2x} - S_{1x}, \quad \sigma_y \equiv S_{2y} + S_{1y}, \quad \sigma_z \equiv S_{2z} + S_{1z},
\]

\[
\gamma_x \equiv S_{2x} + S_{1x} - 2S, \quad \gamma_y \equiv S_{2y} - S_{1y}, \quad \gamma_z \equiv S_{2z} - S_{1z}.
\]

The free-spin precession dynamics without external magnetic field can be solved by using the equations of motion \( \frac{d\mathbf{S}}{dt} = \frac{i}{\hbar} [\mathbf{S}, \mathcal{H}_0] \). In the perturbative regime, which is the case for our experiment, we can assume \( S_{ix} \sim S \gg S_{iy}, S_{iz} \). Solving the above equations of motion to first order in \( S_{iy} \) and \( S_{iz} \) and rewriting the equations in the magnon
basis, we get

\[ \dot{\sigma}_x = 0, \]

\[ \dot{\sigma}_y = \frac{2}{\hbar}(K_X - K_Z) \cos(2\beta_0) S\sigma_z, \]

\[ \dot{\sigma}_z = -\frac{1}{\hbar} \left[n(J \cos(2\beta_0) + D \sin(2\beta_0) + J) + 2K_Z \sin^2 \beta_0 + 2K_X \cos^2 \beta_0 \right] S\sigma_y, \]

\[ \dot{\gamma}_x = 0, \]

\[ \dot{\gamma}_y = \frac{2}{\hbar} \left[n(J \cos(2\beta_0) + D \sin(2\beta_0)) + (K_X - K_Z) \cos(2\beta_0) \right] S\gamma_z, \]

\[ \dot{\gamma}_z = -\frac{1}{\hbar} \left[n(J \cos(2\beta_0) + D \sin(2\beta_0) - J) + 2K_Z \sin^2 \beta_0 + 2K_X \cos^2 \beta_0 \right] S\gamma_y. \]

These linear responses correspond to the zone-center qFM and qAFM modes with frequencies

\[ \hbar \Omega_{\sigma,qFM} = 2S \left[ n(J + K_X)(K_X - K_Z) \right]^{\frac{1}{2}}, \]

\[ \hbar \Omega_{\gamma,qAFM} = S \left[ 4nJK_X + 4K_X(K_X - K_C) + n^2 D^2 \right]^{\frac{1}{2}}, \]

which are plotted as a function of temperature in Fig. 4b with realistic parameters from the literature\textsuperscript{21,30,37}. (See Extended Data Table 1.)
The equations of motion to the next order in $S_{iy}$ and $S_{iz}$ are

\[
\dot{\sigma}_x = \frac{1}{2\hbar}\{[n(J(\cos(2\beta_0) - 1) + D \sin(2\beta_0)) + 2K_Z \cos^2 \beta_0 + 2K_X \sin^2 \beta_0]\sigma_z \gamma_y \\
+ [-n(J(\cos(2\beta_0) - 1) + D \sin(2\beta_0)) + 2K_Z \cos^2 \beta_0 + 2K_X \sin^2 \beta_0]\sigma_y \gamma_z\},
\]

\[
\dot{\sigma}_y = \frac{1}{\hbar}\{2(K_X - K_Z) \cos(2\beta_0) S\sigma_z + (K_X - K_Z) \sin(2\beta_0)(2S\sigma_x - \sigma_x \gamma_z)\},
\]

\[
\dot{\sigma}_z = \frac{1}{\hbar}\{-[n((J \cos(2\beta_0) + 1) + D \sin(2\beta_0)) + 2K_Z \sin^2 \beta_0 + 2K_X \cos^2 \beta_0]S\sigma_y \\
+ (K_X - K_Z) \sin(2\beta_0)\sigma_x \gamma_y\},
\]

\[
\dot{\gamma}_x = \frac{1}{2\hbar}\{[n(J(\cos(2\beta_0) + 1) + D \sin(2\beta_0)) + 2K_Z \cos^2 \beta_0 + 2K_X \sin^2 \beta_0]\sigma_z \gamma_y \\
+ [-n(J(\cos(2\beta_0) + 1) + D \sin(2\beta_0)) + 2K_Z \cos^2 \beta_0 + 2K_X \sin^2 \beta_0]\gamma_z \gamma_y\},
\]

\[
\dot{\gamma}_y = \frac{1}{\hbar}\{2[n(J \cos(2\beta_0) + D \sin(2\beta_0)) + (K_X - K_Z) \cos(2\beta_0)]S\gamma_z \\
- (K_X - K_Z) \sin(2\beta_0)\gamma_x^2\},
\]

\[
\dot{\gamma}_z = \frac{1}{\hbar}\{-[n(J(\cos(2\beta_0) - 1) + D \sin(2\beta_0)) + 2K_Z \sin^2 \beta_0 + 2K_X \cos^2 \beta_0]S\gamma_y \\
+ (K_X - K_Z) \sin(2\beta_0)\sigma_x \gamma_y\},
\]

where the second-order terms proportional to $\sigma_x \sigma_y$ and $\sigma_x^2$ provide the driving force for the qFM $\rightarrow$ qAFM magnon upconversion.
**LLG simulations**

Numerical calculations of the THz-induced magnon dynamics are performed by solving the LLG equations based on the following Hamiltonian\textsuperscript{18,38}

\[
\mathcal{H} = \mathcal{H}_0 + \mathcal{H}_{Zeeman}
\]

\[
= nJS_1 \cdot S_2 + nD \cdot (S_1 \times S_2) - \sum_{i=1,2} (K_a S_{ia}^2 + K_c S_{ic}^2) - \gamma [H_A(\tau, t) + H_B(t)] \cdot (S_1 + S_2).
\]

Here the additional Zeeman term is introduced to describe the interaction between the magnetic field of the THz pulses and sublattice spins. The magnetic fields used here \((H_A \text{ and } H_B)\) are the second derivatives of a Gaussian function, which ensure that the temporal integration of the pulses reduces to zero. \(\gamma = \frac{g \mu_B}{\hbar}\) is the gyromagnetic ratio and \(g = 2\) is the g-factor. An equation of motion can be derived for each sublattice spin \(S_i\) \((i = 1, 2)\)

\[
\frac{dS_i}{dt} = \frac{\gamma}{1 + \alpha^2} [S_i \times H_i^{eff} + \frac{\alpha}{|S_i|} S_i \times (S_i \times H_i^{eff})],
\]

where \(\alpha\) is a phenomenological Gilbert damping constant that accounts for energy dissipation, \(H_i^{eff}\) is the effective magnetic field for each lattice \(i\), which can be calculated as \(H_i^{eff} = -\frac{1}{\gamma} \frac{\partial \mathcal{H}_i}{\partial S_i}\).

To account for the temperature dependence of the qFM and qAFM modes, we use
parameters defined in Extended Data Table 1. To simulate the 2D spectra, we solve for the time evolution of each sublattice spin $S_i$ as a function of both $\tau$ and $t$ at each temperature and then extract the nonlinear response of the magnetization $M = S_1 + S_2$ generated by both magnetic fields. Performing 2D Fourier transforms yields the simulated 2D THz spectra (Fig. 4c and Extended Data Fig. 4). The detection configurations are chosen to be the same as those used in the experiments. The simulated polarimetry patterns are shown in Extended Data Fig. 5.

**Data availability** All data that support the findings of this study are available from the corresponding authors on reasonable request.

**Code availability** All LLG simulation codes are available from the corresponding authors on reasonable request.

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Energy Frontier Research Center, under Grant No. DE-SC0019140. P.N. acknowledges support as a Moore Inventor Fellow through Grant No. GBMF8048 and gratefully acknowledges support from the Gordon and Betty Moore Foundation.

**Author contributions** Z.Z. conceived the study; Z.Z. and F.Y.G. designed and performed the experiments and analyzed the data, supported by Z.-J.L. and Y.-C.C.; X.M. grew, cut, and characterized the high quality single crystals used in the experiments under the guidance of W.R. and S.C.; Z.Z., Y.-C.C, and F.Y.G. performed simulations with the LLG equations, supported by J.B.C. and E.R.S.; Z.Z., F.Y.G., J.B.C., P.N., A.v.H., E.B., and K.A.N. interpreted the data and conceived the concept of nonlinear coupled magnonics; Z.Z., F.Y.G., E.B., A.v.H. and K.A.N. lead the manuscript preparation with input from all the authors; K.A.N. and E.B. supervised the project.

**Competing interests** The authors declare no competing interests.

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Fig. 1: Analogy between nonlinear dynamics in a spring pendulum and a canted antiferromagnet. a, In the spring-pendulum model, there are two normal modes: a swinging mode, which corresponds to the swinging motion of a rigid pendulum, and a bouncing mode, which corresponds to the oscillating motion of a spring. Due to the asymmetric coupling, driving the swinging mode starting from equilibrium leads to a motion of the bouncing mode, but the reverse is not true. b, A canted antiferromagnet with two sublattice spins possesses two magnetic-dipole allowed zone-center magnon modes: the qFM mode can be interpreted as a precession of the net magnetization and the qAFM mode can be understood as an amplitude oscillation of the magnetization. Analogous to the coupled dynamics in the spring-pendulum model, exciting the qFM mode also induces a response of the qAFM mode, while the coupling is unidirectional so that driving the qAFM mode alone does not couple to the qFM mode.
Fig. 2: THz ESR polarimetry and field dependence measurements at room temperature. a, Schematic representation of the experimental setup for the time-domain FID and polarimetry measurements. A wire grid polarizer (WGP) is used to select for either the parallel (blue) or perpendicular (red) polarized THz field emission. b, FID signals corresponding to excitation of either the $a$ axis qFM mode or the $c$ axis qAFM mode for both parallel and perpendicular signal detection configurations. c, Fourier transforms of the FID signals in b. The power spectra of the qFM (orange) and qAFM (purple) modes driven by direct THz magnetic excitations are shown at the top. The bottom power spectrum shows the qAFM mode response driven by the excitation of the qFM mode (red). d, (top) Parallel polarimetry patterns of the qFM (left) and qAFM (right) mode amplitudes. The experimental data (circles) of the qFM (qAFM) mode are fitted to functions of the form $\cos^2 \theta$ ($\sin^2 \theta$). (bottom) Perpendicular polarimetry patterns of the qFM (left) and qAFM (right) mode amplitudes. The responses are plotted on the same scale with their relative amplitudes labeled. The experimental data of the qFM mode are fitted to functions of the form $\cos \theta \sin \theta$, but the qAFM mode responses are fitted to a sum of the form $|\cos \theta \sin \theta|$ (light purple shaded area) and $|\cos^2 \theta \cos \theta|$ (light red shaded area). e, Pump magnetic field dependence of qFM and qAFM amplitudes with $H_{\text{det}} \parallel H_{\text{THz}}$. f, Pump magnetic field dependence of qAFM amplitudes with $H_{\text{det}} \perp H_{\text{THz}}$. The data are fitted with a quadratic function (dashed line).
**Fig. 3:** 2D THz ESR spectroscopy at room temperature.  

**a,** Schematic illustration depicting the excitation scheme in the 2D THz ESR spectroscopy setup.  

**b,** Room temperature 2D THz spectra collected in the perpendicular detection geometry with $\mathbf{H}_{\text{THz}} \parallel c$ showing the strong off-diagonal magnon upconversion peak.  

**c,** and **d,** Parallel (blue) and perpendicular (red) polarimetry patterns showing the amplitude of the 2D THz upconversion peak collected upon rotation of the azimuthal angle, $\theta$, between $\mathbf{H}_{\text{THz}}$ and the $a$ axis. One complete polarimetry scan for each detection configuration, i.e. 72 2D THz spectra, takes about 11 hours. The asymmetric shape in **c** is likely due to laser drift or a slight misalignment of the rotation stage, e.g. being placed off-center. Fits to functions of the form of $|\cos^2 \theta \sin \theta|$ and $|\cos^2 \theta \cos \theta|$ are shown in **c** and **d** respectively.
Fig. 4: Temperature dependence of magnon upconversion. **a**, Schematic representation of the canted spin configurations for temperatures above and below the spin re-orientation transition (87-96K) in EFO. **b**, Excitation (orange diamonds) and emission frequencies (purple diamonds) of magnon-magnon peak obtained from temperature-dependent perpendicular polarized 2D THz measurements are shown along with the frequencies of qAFM (purple solid) and qFM (orange solid) magnon modes derived from simulations of the spin dynamics using the LLG equation. **c**, Raw experimental and simulated 2D THz spectra for selected temperatures above and below the SRT. Note that $\mathbf{H}_{\text{THz}} \parallel a$ (c) above (below) the SRT drives the qFM mode. All spectra are in the perpendicular detection configuration ($\mathbf{H}_{\text{det}} \perp \mathbf{H}_{\text{THz}}$).
Extended Data

Extended Data Fig. 1: Single-shot time-domain THz magnetic resonance setup.

a, Schematic of the experimental setup. BS–beam splitter, LN Crystal–MgO:LiNbO₃, HWP–half wave-plate, PBS–pellicle beam splitter, QWP–quarter wave-plate, CL–cylindrical lens, BD–calcite beam displacer.

b, Waveform of the free-space single-cycle THz pump pulse B used in the experiments. The THz pump pulse A has a nearly identical shape and field strength. The THz field was attenuated and measured by EO sampling in a 300-um GaP crystal.

c, Spectrum of the single-cycle THz field, showing a bandwidth of ~1 THz around the central frequency of 0.4 THz.
Extended Data Fig. 2: Raw one-dimensional THz ESR polarimetry data. a and c, time-domain signals for parallel and perpendicular detection configurations. b and d, corresponding Fourier spectra. Weak peaks alongside the main qAFM mode resonance are Fourier transform artifacts.
Extended Data Fig. 3: Extended experimental temperature-dependent 2D THz spectra. Experimental 2D THz spectra for the remaining temperatures in Fig. 4b that are not otherwise shown. All spectra are in the perpendicular detection configuration ($H_{\text{det}} \perp H_{\text{THz}}$).
Extended Data Fig. 4: Extended calculated temperature-dependent 2D THz spectra. Calculated 2D THz spectra for the remaining temperatures in Fig. 4b that are not otherwise shown. All spectra are in the perpendicular detection configuration (H_{det} \perp H_{THz}).
Extended Data Fig. 5: Simulated polarimetry patterns corresponding to magnon upconversion. Derived from simulations of nonlinear magnon dynamics following irradiation with a pair of THz pulses using the LLG equation. (left) Parallel detection configuration. (right) Perpendicular detection configuration.

Extended Data Fig. 6: ErFeO$_3$ single crystal characterization. a, A photo of the single crystal grown by optical floating zone method. b, Laue diffraction of the ErFeO$_3$ single crystal used in the experiments.
## Extended Data Table 1: Parameter values used in the LLG simulation.

Realistic values from the literature are used here\textsuperscript{21,30,37}. The temperature-dependent magnetic anisotropy $K_c$ is obtained by fitting the experimentally observed magnon frequencies to a modified Curie–Weiss law.\textsuperscript{37}

| Parameter                                           | Value                               |
|-----------------------------------------------------|-------------------------------------|
| Total spin $S$                                       | $\frac{5}{2}$                       |
| Nearest neighboring number $n$                       | 6                                   |
| Exchange constant $J$                                | 4.6 (meV)                           |
| Antisymmetric exchange constant $D$                  | 0.1066 (meV)                        |
| Temperature-independent magnetic anisotropy $K_a$    | 0.00905 (meV)                       |
| Temperature-dependent magnetic anisotropy $K_c$      | $0.00265 \frac{0.9847}{T+62.38}$    |