Terahertz light–driven coupling of antiferromagnetic spins to lattice

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Understanding spin-lattice coupling represents a key challenge in modern condensed matter physics, with crucial importance and implications for ultrafast and two-dimensional magnetism. The efficiency of angular momentum and energy transfer between spins and the lattice imposes fundamental speed limits on the ability to control spins in spintronics, magnonics, and magnetic data storage. We report on an efficient nonlinear mechanism of spin-lattice coupling driven by terahertz light pulses. A nearly single-cycle terahertz pulse resonantly interacts with a coherent magnonic state in the antiferromagnet cobalt difluoride (CoF2) and excites the Raman-active terahertz phonon. The results reveal the distinctive functionality of antiferromagnets that allows ultrafast spin-lattice coupling using light.

Their spin structure can be modeled in the simplest case by two antiparallel sublattices with the net magnetizations $\mathbf{M}_1$ and $\mathbf{M}_2$, $\mathbf{M}_1 = \mathbf{M}_2$. Alternatively, for describing the magnetic order, it is convenient to introduce the net magnetization vector $\mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2$ and the Néel (antiferromagnetic) vector $\mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2$. Typically, the frequencies of spin resonances in antiferromagnets are close to those of THz optical phonons. The objective of this study is therefore to use intense, nearly single-cycle THz pulses (18) to drive coherent spin oscillations (9) and promote interactions between otherwise noninteracting magnonic and phononic modes.

To demonstrate light-driven spin-lattice coupling, we selected a cobalt difluoride (CoF2) single-crystal plate with the tetragonal rutile crystal structure. Below the Néel temperature $T_N = 39$ K, CoF2 is a collinear antiferromagnet with a strong piezomagnetic effect (19). In a unit cell, the spins of Co$^{2+}$ ions at the cell’s center are antiparallel to those at the cell’s corners (20). These two types of ions form the two antiferromagnetic sublattices with the net magnetizations $\mathbf{M}_1$ and $\mathbf{M}_2$, respectively. The magnetizations are aligned along the crystallographic fourfold optical axis, which is the “easy axis” of magnetic anisotropy (see Fig. 1). The frequency of antiferromagnetic resonance is centered at $\Omega_m = 1.14$ THz at $T = 6$ K, whereas the nearest phonon mode of the $B_{2g}$ symmetry is at $\Omega_{ph} = 1.94$ THz (21, 22). Strong piezomagnetic properties of CoF2 imply that atomic and spin dynamics can, in principle, be coupled (4, 19), but because the energies of the phonon and the magnon are substantially different, the coupling is inefficient. In this study, we reveal that a THz photon can fill the magnon-phonon energy gap and thus induce their efficient coupling. More specifically, we show that a nearly single-cycle THz pulse centered at ~1 THz with a bandwidth in excess of $2\Omega_m - \Omega_{ph}$ is able to prepare a coherent magnonic state and subsequently interact with this coherent state by promoting an energy transfer from the coherent magnon to the $B_{2g}$ phonon.

To trace this energy transfer, we used a pump-probe technique to optically detect the coherent phonons and magnons. The atomic motion of the Raman-active $B_{2g}$ phonon mode dynamically breaks the equivalence between the crystallographic $x$ and $y$ axes and thus induces linear birefringence for the light propagating along the optical $z$ axis. Moreover, upon introducing a generalized phonon coordinate $Q_{\phi}$ corresponding to the $B_{2g}$ phonon and the $z$ component of the Néel vector $L_z$, it can be shown that if the phonon

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**Fig. 1. Geometry of the experiment and THz pumping of antiferromagnetic CoF2.** The spins of Co$^{2+}$ ions are antiferromagnetically aligned along the $z$ axis. The linearly polarized THz pump and near-infrared probe beams propagate approximately collinearly along the $z$ axis and spatially overlap on the sample. By varying the time delay $\tau_{fast}$ between the infrared probe and the THz-pump pulses, we measure pump-induced ultrafast dynamics. The angle $\alpha$ between the THz pulse magnetic field $\mathbf{B}_{THz}$ and the $y$ axis is tuned in the range of $\pm\pi/2$ using two wire-grid polarizers. The polarization of the probe pulse forms an angle $\gamma$ with the $y$ axis and is controlled by a half-wave plate.
Fig. 2. THz excitation of magnons and phonons. (A) THz pump–induced rotation of the probe’s polarization measured at different temperatures for the case of the horizontal orientation of the probe electric field ($\gamma = 90^\circ$) and the THz magnetic field ($\psi = 90^\circ$). (B) Fourier spectrum of the waveform from (A) measured at $T = 30$ K. The spectrum of the incident THz pulse is shown by the light blue area. (C) The frequencies of the magnon mode (blue open circles) and the phonon mode (green filled circles) deduced from the Fourier spectrum as a function of temperature. The solid lines are guides for the eye. The dashed line marks the Néel temperature. (D) The Fourier spectral amplitudes of the magnon mode (blue open circles) and the phonon mode (green filled circles) at $T = 6$ K as functions of the THz magnetic field strength $H_{\text{THz}} = |H_{\text{THz}}|$. The solid lines represent linear and quadratic fits. The corresponding waveforms are plotted in figs. S8 and S9. a.u., arbitrary units.

Fig. 3. Phonon and magnon excitation. (A) Temperature dependencies of the Fourier amplitudes of the magnon mode (blue open circles) and the phonon mode (green filled circles). The solid lines are guides to the eye. The dashed line marks the Néel temperature. The corresponding waveforms are given in fig. S8. (B) Estimated phononic amplitude excited via the virtual state (blue open triangles; first and second terms in Eq. 3), real magnonic state (green filled circles; the third term in Eq. 3), and anharmonic magnon (red filled squares; the fourth term in Eq. 3). The dashed line marks the Néel temperature.

(23)

The typical THz pump–induced transients for the probe’s polarization rotation at different temperatures are shown in Fig. 2A. Polarization rotation with an amplitude of $150$ millidegrees (mdeg) is observed below the Néel point and reveals a strong temperature dependence. When normalized to the thickness of the crystal, the signal reaches $3$ deg/cm for a THz field on the order of $MV/cm$, which is comparable with the values reported for similar measurements on NiO (24). Figure 2B shows the Fourier transform of the time trace obtained at $T = 30$ K. It is seen that the THz-pump pulse excites two resonances centered at $1.05$ and $1.94$ THz. Figure 2C shows that the temperature dependencies of the resonance frequencies in the vicinity of the Néel point are in a perfect agreement with the behaviors expected for the magnon and the $B_{z}$ phonon in CoF$_{2}$ (25, 26). Whereas the magnon mode softens near the Néel point, the phonon frequency does not show any noticeable change in this temperature range. Figure 2D shows the dependencies of the amplitude of the magnon and the phonon resonances on the THz magnetic field strength. The magnon amplitude is a linear function of the field, which is typical for the conventional mechanism of excitation of the antiferromagnetic resonance by a magnetic field via the Zeeman torque (8, 24). By contrast, the phonon amplitude scales quadratically with the field strength similar to that in (27), thus clearly evidencing the nonlinear mechanism of the excitation. A spectrogram of the probe polarization rotation at $T = 31$ K is plotted in fig. S10.

To describe the dynamics of the Néel vector $\mathbf{L}$, one can represent it as a sum of the stationary $\mathbf{L}_{0}$ and the THz-induced $\mathbf{I}$ parts: $\mathbf{L} = \mathbf{L}_{0} + \mathbf{I}$, where $\mathbf{L}_{0} \gg \mathbf{I}$. From the Lagrange-Euler equations (28, 29), one finds that the dynamics of $\mathbf{I}$ is described by differential equations for damped harmonic oscillators (the magnon selection rules) (29). The latter gives

$$
L_{x}(t) = \frac{d}{dt}(\Omega_{m} t + \xi) e^{-t/t_{m}} \cos\psi
$$

$$
L_{y}(t) = \frac{d}{dt}(\Omega_{m} t + \xi) e^{-t/t_{m}} \sin\psi
$$

where $\tilde{H}(\Omega_{m})$ is the spectral amplitude of the THz magnetic field at $\Omega_{m}$, $t_{m}$ is the magnon damping time, and $\xi$ is the phase. Hence, any orientation of the magnetic field in the $xy$ plane excites oscillations of $\mathbf{I}$. Note that the equations of motion for $\mathbf{I}$, which can be derived either from the Lagrange-Euler or directly from the Landau-Lifshitz-Gilbert (30) equations, show that these oscillations are launched in the plane orthogonal to the THz pulse magnetic field $H_{\text{THz}}$.

$q_{x}^{2} - q_{y}^{2}$ induces strain $\sigma_{xy}$, then the product $L_{x} \sigma_{xy}$ and the $x$ component of the magnetization $M_{x}$ transform equivalently under the symmetry operations allowed by the crystallographic 4/mmm point group of CoF$_{2}$ (the phonon selection rules) (23). This relation can also be attributed to the piezomagnetic effect (19), that is, at temperatures below the Néel temperature, coherent atomic motion can also induce magnetic circular birefringence and result in the magneto-optical Faraday effect for light propagating along the $z$ axis. The polarization rotation must be proportional to $L_{x} \sigma_{xy}$ and therefore should disappear above the Néel temperature. The magnon mode at the frequency of the antiferromagnetic resonance can also be detected optically via magnetic circular birefringence, that is, the Faraday effect, and also via magnetic linear birefringence (the magnon selection rules) (23).

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$(\text{the magnon selection rules}) (29)$. The latter gives

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L_{x}(t) = \tilde{H}(\Omega_{m}) \cos(\Omega_{m} t + \xi) e^{-t/t_{m}} \cos\psi
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where $\tilde{H}(\Omega_{m})$ is the spectral amplitude of the THz magnetic field at $\Omega_{m}$, $t_{m}$ is the magnon damping time, and $\xi$ is the phase. Hence, any orientation of the magnetic field in the $xy$ plane excites oscillations of $\mathbf{I}$. Note that the equations of motion for $\mathbf{I}$, which can be derived either from the Lagrange-Euler or directly from the Landau-Lifshitz-Gilbert (30) equations, show that these oscillations are launched in the plane orthogonal to the THz pulse magnetic field $H_{\text{THz}}$. 
In the case of atomic motion at the $B_{1g}$ phonon frequency with the generalized phononic coordinate $Q_{x_2\cdots x_2}$, one can write a conventional equation of motion for the harmonic oscillator:

\[
\frac{d^2 Q_{x_2\cdots x_2}}{dt^2} + 2\frac{dQ_{x_2\cdots x_2}}{t_{ph}} + \Omega^2_{ph} Q_{x_2\cdots x_2} = T_{x_2\cdots x_2}
\]

(2)

where the first term in the left part corresponds to acceleration and the second term accounts for damping where $t_{ph}$ is the phonon damping time. The third term corresponds to the restoring force, and $T_{x_2\cdots x_2}(t)$ is the driving torque. Obviously, the equation must be invariant under all symmetry operations of the 4/mmm point group of CoF$_2$, that is, the torque $T_{x_2\cdots x_2}(t)$ must transform equivalently to $Q_{x_2\cdots x_2}$ (see table S1). Taking into account the irreducible representations of the second order with respect to the THz electric and magnetic fields, the following four contributions to $T_{x_2\cdots x_2}(t)$ are allowed:

\[
T_{x_2\cdots x_2}(t) = C_1 [E_x(t) - E_y(t)] + C_2 [H_x(t) - H_y(t)] + C_3 [\bar{L}_x(t)H_y(t) - \bar{L}_y(t)H_x(t)] + C_4 (L_x^2(t) - L_y^2(t))
\]

(3)

where $C_1, C_2, C_3,$ and $C_4$ are phenomenological coefficients; $E_i$ and $H_i$ are the $i$-component of the THz magnetic and electric fields, correspondingly. The first and second terms correspond to electric and magnetic dipole mechanisms of two-photon excitation of the $B_{1g}$ phonon via a virtual state. A similar mechanism was described in (32, 33). In contrast to the two-photon excitation of phonons via an intermediate phononic state, as proposed in (34), the third term in Eq. 3 describes two-photon excitation of phonons via an intermediate magnonic state. The fourth term shows that the phonon can also be excited because of the anharmonicity of magnons. However, one may argue that in the case of forced oscillations of the antiferromagnetic Néel vector by a THz magnetic field, the effects of the third and the fourth terms must be similar and can hardly be distinguished.

The temperature dependencies of the detected signal at the phonon and magnon frequencies are shown in Fig. 3A. The substantially low (30–30 times) phononic oscillations above the Néel point is a result of the dominating role of the magneto-optical Faraday effect in the detection of the phonon. Because the phonon-induced Faraday rotation of the probe pulse is proportional to the $L_x \tau_{sp}$ product, the detection efficiency becomes zero above the Néel temperature. Intriguingly, the detected phononic amplitude stays nearly constant in the range from 6 to 25 K and even has a maximum at 30 K, where the equality $\Omega_{ph} = 2\Omega_m$ holds. By contrast, the magnon frequency $\Omega_m$ smoothly and monotonically decreases from 1.14 THz at $T = 6$ K to zero at the Néel temperature, and this frequency change is accompanied by a broadening of the magnon spectral line $\Delta \Omega_m$ (see fig. S4). Such changes affect the efficiencies of phononic excitations in different ways because of the mechanisms that correspond to different terms in Eq. 3. To elucidate this, we modeled temperature dependencies of the mechanisms that correspond to the first, third, and fourth terms in Eq. 3. In particular, our model explored how temperature affects the strength of the oscillations induced at the frequency of the phonon (the phonon selection rules) (23). In the modeling, we also took into account that the phonon and magnon are detected optically and that the sensitivity of the detection is practically proportional to the Néel vector. Comparing the simulated dependencies with that observed experimentally clearly emphasizes the dominating role of the magnon-mediated mechanism in the excitation of the phonon (see Fig. 3B).

To demonstrate that the nonlinear excitation of phonons is mediated by a coherent magnonic state, we performed double-pump experiments using two identical THz pulses separated by a time delay $\tau_{exc}$ with a peak magnetic field of 100 mT. For different $\tau_{exc}$, we measured the probe’s polarization rotation as a function of time delay between the first THz pump and the probe pulse $\tau_{det}$. Using the same method of data acquisition as explained in (35), we measured the dynamics triggered by both THz-pump pulses $\alpha_{exc}(\tau)$ by the first pump only, that is, when the second THz pump is blocked $\alpha_{exc}(\tau)$ and by the second THz pump only (the first THz pump is blocked) $\alpha_{det}(\tau)$. Here, $\tau$ is the vector $\tau = (\tau_{exc}; \tau_{det})$. We deduce the intrinsically nonlinear response of the medium by subsequently calculating the difference between the signals in the time-domain:

\[
\alpha_{NL}(\tau) = \alpha_{det}(\tau) - \alpha_{exc}(\tau) - \alpha_{det}(\tau)
\]

(4)

where $\alpha_{NL}(\tau)$ is nonzero only if the first pump changes the spin susceptibility to the THz magnetic field. A Fourier transform of the extracted signals $\alpha_{NL}(\tau_{exc}; \tau_{det})$ allows us to plot the spectrum as a 2D graph $\omega_{NL}(\omega_{exc}; \omega_{det})$ (see Fig. 4A). Here, $\omega_{exc}$ and $\omega_{det}$ are the Fourier frequencies of the corresponding time delays $\tau_{exc}$ and $\tau_{det}$ respectively. The spectrum $\omega_{NL}(\omega_{exc}; \omega_{det})$ clearly reveals one maximum at $\omega_{det} = \omega_{ph}$ and $\omega_{exc} = \Omega_m$, revealing that the excitation of the $B_{1g}$ phonon via a nonlinear THz light–driven mechanism is only possible when the magnon is excited. This means that the macroscopic coherent magnonic state with the frequency $\Omega_m$, resonantly generated by the broadband THz pulse, has an essentially different susceptibility to the THz magnetic field compared with that associated with its unperturbed state. More particularly, the interaction of the second THz photon at the frequency $\Omega_{ph} - \Omega_m$ with the coherent magnonic state is able to excite the $B_{1g}$ phonon at the frequency $\Omega_{ph}$ (see Fig. 4B).

The ultrafast coherent transfer of spin energy to the lattice driven by THz light pulses
opens up possibilities for the fields of nonlinear phononics, ultrafast magnetism, THz magnonics, and antiferromagnetic spintronics. The dynamic coupling of the lattice to the spins is crucial for ultrafast control of magnetism and phase engineering through transient changes of magnetic and structural states. Exactly the same light-driven spin-lattice coupling is allowed in other antiferromagnetic fluorides that have the same point group (MnF$_2$, FeF$_2$, and others), but the effect must be more general and can be extended to other materials. Because a coherent magnonic state in any ferro-, ferri-, and even antiferromagnetic material can induce dynamic magnetization, the excitation of this coherent state with a magnetic field at the frequency matching the gap between the magnon and the phonon might even lead to nontrivial ultrafast phenomena associated with the physics of the Einstein–de Haas effect. This mechanism is especially appealing for ultrafast coherent control of materials with complex charge and spin ordering, for example, in multiferroics and 2D magnets.

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SUPPLEMENTARY MATERIALS
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