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Effect of antiferromagnetic order on a propagating single-cycle THz pulse

On the quest to achieve ever faster and more energy-efficient magnetic writing, antiferromagnets are highly appealing for their outstanding properties and related functionalities. Opposite to ferromagnets which are commonly used in magnetic data storage devices but limited to the GHz frequency regime, antiferromagnets possess spin dynamics in the THz frequency range and, thus, have high potential for a faster and more energy efficient data storage. Although a theory has predicted ultrafast switching initiated by THz fields, ultrashort pulses of light, or spin currents, the actual writing of antiferromagnetic bits at the picosecond timescale and at THz repetition rates has not been demonstrated experimentally yet.

A nearly single-cycle intense THz pulse is one of the most auspicious stimuli that was employed for driving coherent spin dynamics in antiferromagnets. In antiferromagnetic CoF$_2$, the amplitudes of spin dynamics triggered by intense THz pulses were sufficiently high to push the dynamics into a nonlinear regime and opened a new channel for the development of THz control of antiferromagnetism. This single-crystal material is characterized by inherently strong piezomagnetic effects and exhibits several magnonic and phononic states in the terahertz spectral region. This makes CoF$_2$ a promising candidate to study propagation effects of THz light in antiferromagnetic insulators. Here, we investigate these effects and show that a nearly single-cycle THz pulse acquires a giant rotation and significant ellipticity upon propagation through antiferromagnetic CoF$_2$.

Cobalt fluoride (CoF$_2$) recently attracted interest as a model antiferromagnet whose properties can be changed under ultrafast pumping with polarized light in the mid-infrared and THz spectral ranges. Cobalt fluoride (CoF$_2$) possesses a tetragonal rutile crystal structure ($P4_2/mmm$ space group) as illustrated in Fig. S1(a) in the supplementary material. The lattice parameters at room temperature are $a = b = 4.695$ and $c = 6.95$ Å.
The sample used in our experiment was a ac plane single crystal plate with the b axis along the normal to the surface and thickness d ≈ 900 μm. The spins of Co2+ ions are aligned along the c axis with opposite directions at the center and corners of the unit cell below the Néel temperature TN = 39 K19,20 [see Fig. S1(a) in the supplementary material]. The simplest representation decomposes antiferromagnetic spin structure of CoF2 into two antiparallel sublattices with their net magnetizations represented by M1 and M2, respectively. Exchange coupling of these sublattices and relatively strong anisotropy lift the frequency of spin resonances in antiferromagnetic fluorides to the THz regime. Spin dynamics in antiferromagnets are described in terms of two vectors, the net magnetization \( \mathbf{M} = \mathbf{M}_1 + \mathbf{M}_2 \) and the antiferromagnetic Néel vector \( \mathbf{L} = \mathbf{M}_1 - \mathbf{M}_2 \). Conventionally, it is assumed that in antiferromagnets magnetic linear birefringence is proportional to \( \mathbf{L} \). In real materials, however, the situation can be complicated due to mechanical deformations induced by antiferromagnetic order. As a result, the temperature dependence of the linear magnetic birefringence of CoF2 in the visible spectral range, in particular, does not follow the temperature dependence of \( \mathbf{L} \).21

A mode-locked Ti:sapphire regenerative amplifier laser with 50 fs pulse duration at a center wavelength of 800 nm and a repetition rate of 1 kHz was used in the experiment. Employing optical rectification in ZnTe, we generated nearly single-cycle pulses of THz radiation which are focused tightly onto the surface of our CoF2 sample via off-axis parabolic mirrors. After passing through the sample, the THz light was directed and focused on a second ZnTe crystal where a small, time-delayed part of the infrared radiation was used to probe the waveform of the THz pulse. Hence, the THz electric field induced birefringence leading to ellipticity of the linearly polarized probe pulse at a central wavelength of 800 nm. The diameter of the THz beam is at the order of 1–2 mm and, thus, significantly larger than the one of the probe pulse which is at the order of hundreds of micrometer. The ellipticity of the probe pulse was further measured using a quarter wave plate, a Wollastone prism, and a balanced detector as explained elsewhere.22 The sample of CoF2 was placed in an optical helium cryostat, which allowed us to carry out experiments in the temperature range from 5 to 300 K.

For polarization sensitive THz spectroscopy, we employed a set of wire-grid polarizers (WGPs). Directly after the generating ZnTe crystal, we placed the first wire-grid polarizer WGP1 that allows only THz light polarized vertically, i.e., at 45° between a and c axes to pass through the CoF2 crystal. The second wire-grid polarizer WGP2 was set right after the crystal and could be oriented either along the a or the c axis, respectively. The third polarizer WGP3 was positioned right before the detecting electro-optical crystal and its orientation was again fixed at 45° between the a and c axes. We illustrate this scheme of THz polarimetry in Fig. S1(b) in the supplementary material.

Typical time traces of the initially vertically polarized THz electric field after propagating through the CoF2 sample at various temperatures are shown in Fig. 1. Panels (a) and (b) show the projections of the electric field \( \mathbf{E}_{THz} \) after propagating the sample on the crystallographic a and c axes. The corresponding time traces of the THz pulse propagating in free space are shown as a black solid curve and further referred to as a reference. The signatures of THz pulse reflection inside the Fabry–Pérot etalon are formed by parallel front and back surfaces of the sample crystal with the first peak around 20 ps. For the THz electric field \( \mathbf{E}_{THz} \) polarized along the a axis, the THz waveform does not experience significant changes. However, there is a slight decrease in signal amplitude with decreasing temperature. A rotation of WGP3 by 90° substantially changes the signal traces and for the THz electric field \( \mathbf{E}_{THz} \) parallel to the c axis, at temperatures below \( T_N \) the signals exhibit clear oscillations starting around 8 ps as shown in Fig. 1(b). By increasing the temperature, the amplitude of these oscillations reduces gradually. Furthermore, the signals are complicated by a clear beating. Performing a Fourier transform in a limited time range of 0–50 ps, we obtained the spectra of the signals \( S(\omega) \) and the reference \( S_{ref}(\omega) \) and calculated the normalized loss functions \( \sigma(\omega) = -\ln \left( \left( \frac{S(\omega)}{S_{ref}(\omega)} \right) \right) \),23 which are shown in Fig. S2 in the supplementary material. While the normalized loss function \( \sigma(\omega) \) for detection of the electric field \( \mathbf{E}_{THz} \parallel a \) (dotted line) is flat, in the case of \( \mathbf{E}_{THz} \parallel c \) (solid line), one can see a pronounced resonance centered at around 1 THz and present for all temperatures below \( T_N \). With increasing the temperature, this mode undergoes a red-shift and substantial broadening, i.e., experiences softening. To quantify the...
Based on the approach introduced in Ref. 28, the spectral dependence of the antiferromagnetic resonance in CoF2 and emphasize excellent agreement of the resonance frequency temperature dependence with previous reports in Refs. 21, 24, and 25.

Since spins are aligned along the c axis, an electromagnetic wave with the electric field along the a axis and the magnetic field along the c axis does not exert any torque on the spins. This confirms the fact that the antiferromagnetic resonance is seen exclusively for the case of detection of the electric field \( \mathbf{E}_{\text{THz}} \) along the crystallographic c axis. Remarkably, we also revealed a previously unreported feature evidenced by the beating-like behavior in the time domain as well as the double-peak feature in the frequency domain.

A similar feature was previously observed for THz time-domain spectroscopy of TmFeO3 and was assigned to the formation of magnon polaritons.\(^b\) In the case of CoF2, we observe similar linewidths and comparable central frequencies of the antiferromagnetic resonance. Comparing the oscillator strengths and sample thicknesses, we do not expect a large difference between the CoF\(^{2-}\) and Fe\(^{3+}\)-spins, respectively. Thus, we assign the experimentally observed double-peak feature in CoF2 to a magnon–polariton. We also emphasize the agreement of the antiferromagnetic resonance frequency dependence on temperature in our data with previously reported values from Ref. 27.

From the experimental data, we can estimate the complex refractive index, where the real part \( \text{Re}[\tilde{n}] = \varepsilon_\text{vac} \Delta \tau/\Delta \tau \) is obtained from the speed of light in vacuum \( \varepsilon_\text{vac} \), the difference of the arrival times \( \Delta \tau \) of the reference, and the THz signal and the sample thickness \( \Delta \). The difference in arrival time \( \Delta \tau \) between the signal and the reference pulse is \( \approx 5 \) ps and, thus, the real part of the refractive index \( \text{Re}[\tilde{n}] \approx 2.4 \). Based on the approach introduced in Ref. 28, the spectral dependence of \( \text{Re}[\tilde{n}(\omega)] \) and the absorption coefficient \( \alpha(\omega) \) have the following forms:

\[
\text{Re}[\tilde{n}(\omega)] = 1 + \frac{\varepsilon_\text{vac}}{2\text{Re}(\tilde{n})} \phi(\omega),
\]

\[
\alpha(\omega) = -\frac{2}{\Delta} \ln \left( \frac{S(\omega)}{S_{\text{ref}}(\omega)} \frac{[\tilde{n}(\omega) + 1]^2}{4n(\omega)} \right).
\]

Here, \( \phi(\omega) = \phi(\omega) - \phi_{\text{ref}}(\omega) \) is the phase difference of the transmitted THz pulse through the sample \( \phi(\omega) \) and the reference pulse (free-space propagation) \( \phi_{\text{ref}}(\omega) \). The corresponding spectra for \( \alpha(\omega) \) are shown in Fig. 2.

Finally, using the time-traces for the two projections of the THz electric field transmitted through the sample \( \mathbf{E}_{\text{THz}} \parallel a \) and \( \mathbf{E}_{\text{THz}} \parallel c \), we reconstructed a three-dimensional waveform of the corresponding pulses. The THz electric field \( \mathbf{E}_{\text{THz}} \), sampled for free space propagation is linearly polarized (see the blue line in Fig. 3), while the THz electric field after interaction with the CoF2 sample exhibits considerable changes of polarization. The most pronounced deviation from the linear polarization appears as an elliptical feature between 6 and 8 ps (10 K, see Fig. 3) and occurs during the temporal overlap of THz and probe pulses.

From this simple three-dimensional illustration, we identified the region of the first 10 ps as hosting the most significant changes. To express as ellipticity and rotation, we employed a direct (i.e., non-iterative) and ellipse specific in terms of occlusion and noise sensitivity least squares fitting method proposed for generic applications in Ref. 29. Starting from the general expression of a conic as which an ellipse can be modeled and referring to Ref. 29 for detailed steps, we arrive at a generalized eigenvalue problem. Solving the same, we compute fitting parameters for an ellipse, namely, the major \( a \) and minor \( b \) axes as well as the rotation \( \theta \) with respect to the center coordinates. An exemplary illustration for such a fit of the a and c axis projection is indicated by the red ellipse in Fig. 3.

We can easily deduce (see Fig. 3) the relative change in ellipticity \( \varepsilon_{\text{rel}} \) and rotation \( \theta_{\text{rel}} \) of the THz pulse by \( \varepsilon_{\text{rel}} = \varepsilon - \varepsilon_{\text{ref}} \) and \( \theta_{\text{rel}} = \theta - \theta_{\text{ref}} \) with \( \varepsilon_{\text{ref}} = \arctan(a_{\text{ref}}/b_{\text{ref}}) = 1.5 \) and \( \theta_{\text{ref}} = 44 \). Having ellipticity and rotation extracted, we plot them as functions of temperature dependence of the magnon frequency.

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**Fig. 2.** Absorption coefficient for THz transmission along the crystallographic a axis (dotted lines) and c axis (solid lines) as a function of temperature. Inset: Temperature dependence of the magnon frequency.

**Fig. 3.** Three-dimensional representation of the THz pulse shape after propagating through CoF2 at \( T = 10 \) K (red). Elliptical fitting to the two-dimensional projection along the crystallographic a and c axes is indicated in the exposed plot (red). The THz pulse propagating in free space is linearly polarized and shown as reference (blue).
temperature in Fig. 4. Our data are in qualitative agreement with the temperature dependence of birefringence reported in the visible spectral range (632 nm) in Ref. 21. We believe that the ellipticity and the rotation are due to linear birefringence in the antiferromagnetic CoF_2 crystal. Maximum amplitudes of ordinary and extraordinary refractive indices were determined in Ref. 21 as n_o - n_e \approx 2 \times 10^{-4} (see the normalized data in Fig. 4). Despite a huge difference in the photon energy practically by a factor of 10^3, the THz rotation and ellipticity and the birefringence in the visible spectral range closely follow a similar law. It was shown that the latter is determined by the interplay of different contributions, including the pure magnetic linear birefringence and the magnetostriction, and, thus, is anomalous and does not follow the conventional dependence of L^2.

In conclusion, by performing THz-TDS, we showed that (i) antiferromagnetic CoF_2 and, in particular, the antiferromagnetic mode impose significant rotation and ellipticity on the polarization state of a propagating nearly single-cycle THz pulse; (ii) rotation and ellipticity can be well quantified by employing elliptical fits; (iii) both the THz ellipticity and polarization rotation are due to magnetic linear birefringence and pronounced in the spectral range of the antiferromagnetic resonance. Their temperature dependencies follow a law that is qualitatively similar to the linear birefringence (magnetic and natural) measured in the visible spectral range; and (iv) our findings clearly show the importance of accounting for propagation effect in THz spintronics and magnonics.

See the supplementary material for an extended discussion on the spin structure of CoF_2, the experimental setup, and the calculation of Fourier transformation and loss function.

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**AUTHOR DECLARATIONS**

Conflict of Interest

The authors have no conflicts to disclose.

**Author Contributions**

**Thomas WJ. Metzger:** Data curation (lead); Formal analysis (lead); Investigation (lead); Methodology (lead); Software (lead); Visualization (lead); Writing – original draft (lead); Writing – review & editing (lead).

**Kirill A. Grishunin:** Data curation (supporting); Formal analysis (supporting); Investigation (supporting); Methodology (supporting); Writing – review & editing (supporting).

**Dmytro Afanasiev:** Supervision (supporting); Visualization (equal); Writing – review & editing (supporting).

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**Alexey V. Kimel:** Conceptualization (lead); Funding acquisition (lead); Methodology (lead); Project administration (lead); Supervision (lead); Validation (lead); Writing – original draft (equal); Writing – review & editing (equal).

**DATA AVAILABILITY**

The data that support the findings of this study are available from the corresponding author upon reasonable request.

**REFERENCES**


