

Ultrafast modulation of the anomalous Hall conductivity by coherent magnetization precession in Co-Pt thin films

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Ultrafast dynamics of magnetic properties in the high gigahertz (GHz) or even terahertz (THz) frequency range may be exploited in next-generation data storage and processing devices, and therefore suitable methods for detecting ultrafast dynamics of magnetization need to be developed. In this work, we investigate the impact of optically induced ultrafast precession of magnetization on the anomalous Hall conductivity in a ferromagnetic Co-Pt multilayer structure using THz-based pump-probe measurements. From the measured Faraday rotation, the time-dependent anomalous Hall conductivity σ_{xy} is extracted, and our data reveals an initial ultrafast quench due to laser-induced demagnetization and a subsequent oscillation with a frequency that increases with the strength of the applied external magnetic field $\mathbf{H}_{\text{ext}}$. For $H_{\text{ext}}=30$ kOe, the oscillation frequency reaches approximately 100 GHz. The magnetic-field dependence of the oscillation frequency is in good agreement with the prediction from the ferromagnetic resonance model, confirming that the observed oscillation is caused by coherent magnetization precession. These findings are considered to be useful for studies that aim to integrate spintronics and THz electronics.

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I. INTRODUCTION

The anomalous Hall effect (AHE) results in the emergence of a component of the electron motion that is orthogonal to the direction of the bias voltage, and this anomalous transverse current can have contributions from three mechanisms: the intrinsic mechanism [1], which arises from a nonzero Berry curvature of a Bloch band in a given solid, and two extrinsic mechanisms called skew scattering [2] and side-jump scattering [3]. With the advancement of terahertz (THz) polarimetry [4–7], it became possible to detect anomalous Hall currents with a temporal resolution better than 1 ps, and thus ultrafast manipulation of the anomalous Hall conductivity has attracted significant attention in recent years. Such ultrafast manipulation can be, for example, achieved by populating electron levels in order to modify the distribution in a band with a nonzero Berry curvature [6] or by engineering of the band structure using time-periodic electric fields (of laser pulses) to directly modify the Berry curvature [7].

In addition to the research on topological materials, there are also a few papers on ultrafast manipulation of the

anomalous Hall conductivity in magnets [8]. Since electrons with opposite spins have opposite anomalous transverse velocity components, a greater imbalance between spin-up and spin-down populations results in a stronger AHE. Consequently, the anomalous Hall conductivity in magnets is typically proportional to the out-of-plane magnetization. This physical relationship implies that the anomalous Hall conductivity can be modulated by coherent magnetization precession in a ferromagnet as illustrated in Fig. 1(a), where the modulation manifests as an oscillation of the transverse current J_c . Note that coherent magnetization dynamics in magnetic materials are being explored for next-generation data storage and processing devices based on spintronics technology [9–15]. Furthermore, the advancements in THz technology may lead to a convergence of spintronics and THz electronics. Compared to optical pulses, THz pulses can be transmitted through semiconductor materials with a small band gap and may also be less influenced by surface roughness due to significantly longer wavelengths. While the manipulation of magnetization dynamics using THz pulses has already been extensively studied (for instance, by utilizing the Zeeman torque from the magnetic-field component [16–27] or spin-orbit coupling due to the electric-field component [28,29]), research on THz-based detection has so far focused on THz emission generated through the oscillation of a magnetic dipole [30,31] or an electric dipole [32,33], where the latter type of oscillation is achieved by conversion of spin currents to charge currents

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via the inverse spin Hall effect. For the development of THz electronics, it should be advantageous to investigate whether the information encoded in coherent magnetization precession can also be directly imprinted on incident THz pulses, analogous to the polarization modulation of optical pulses via the magneto-optical effect [9,34,35]. In the THz regime, this should be possible by utilizing the AHE. In this work, we provide experimental results that clearly identify the footprint of coherent magnetization precession in the polarization of THz pulses caused by a modulation of the anomalous Hall conductivity of a ferromagnetic sample.

II. METHODS

A. Experimental setup

Ultrafast changes in the anomalous Hall conductivity can be tracked using the Faraday rotation signal of a THz pulse as illustrated in Fig. 1(b). Here, we use three wire-grid polarizers (WGP1–WGP3) to control the polarization state of the THz pulse incident on the sample and to analyze the polarization state of the THz wave after the sample [5]. Note that the polarization directions of WGP1 and WGP3 in this setup are fixed, while the angle of WGP2, α , is adjustable. The electric-field waveform of the THz pulse after WGP3 was measured by electro-optic (EO) sampling with an 800-nm gate pulse and a (110)-oriented ZnTe crystal (not shown in the figure). The time delay between the THz pulse and the gate pulse at the ZnTe crystal is hereafter denoted by t_1 . This setup enables the detection of the electric-field components projected onto the x' and y' axes, $E_{x'}$ and $E_{y'}$, respectively, which allows us to quantify the Faraday rotation. While the nonzero extinction ratio of 0.006 of WGP1 results in a small y' -axis component of the THz electric field in front of the sample, it is possible to remove the influence of this small nonideal component by the subtraction of θ (and also η) values obtained using opposite external magnetic fields. Furthermore, the THz electric field transmitted through WGP2 ($E_{x'}^\alpha, E_{y'}^\alpha$) can be expressed as shown in Eq. (1) below.

$$\begin{pmatrix} E_{x'}^\alpha \\ E_{y'}^\alpha \end{pmatrix} = R(\alpha)TR(-\alpha) \begin{pmatrix} E_{x'} \\ E_{y'} \end{pmatrix}, \quad (1)$$

where $R(\alpha)$ is the rotation matrix and T is the Jones matrix of WGP2 in the case that the wires are parallel to the y' axis:

$$T = \begin{pmatrix} t_\perp & 0 \\ 0 & t_\parallel \end{pmatrix}, \quad (2)$$

where $t_\parallel$ and $t_\perp$ are the complex transmission coefficients for THz waves polarized parallel and perpendicular to the wires, respectively ($t_\parallel/t_\perp$ is the extinction ratio, and the transmission axis is perpendicular to the wires). The

y' -axis component of the THz electric field after WGP2 passes through WGP3 and is analyzed using EO sampling. Although the nonzero extinction ratio of WGP3 also leads to a small x' -axis component after WGP3, this component cannot be detected due to the used orientation of the EO crystal. Therefore, the detected value becomes

$$F_\alpha = \begin{pmatrix} 0 & t_\perp \end{pmatrix} \begin{pmatrix} E_{x'}^\alpha \\ E_{y'}^\alpha \end{pmatrix}. \quad (3)$$

By setting $\alpha = 90^\circ$ and 45° , we can obtain F_{90° and F_{45° , respectively, and by solving Eqs. (1) and (3), $E_{x'}$ and $E_{y'}$ can be calculated.

The sample consists of a ferromagnetic Co-Pt multilayer structure on a fused silica substrate with a perpendicular magnetic anisotropy (PMA) [36]. The detailed sample structure is shown in Fig. 1(c). This layered structure exhibits a larger Faraday rotation at THz frequencies than a pure Co film of the same total thickness. The coherent magnetization precession illustrated by the yellow arrow in Fig. 1(d) is induced by ultrafast laser heating with a 1500-nm pump pulse with a duration of 80 fs and a fluence of approximately $200 \mu\text{J}/\text{cm}^2$. The time delay between peak electric field of the THz pulse and that of the pump pulse is hereafter denoted by t_2 . Furthermore, as shown by the green arrow, an external magnetic field $\mathbf{H}_{\text{ext}}$ is applied parallel to the propagation direction of the THz pulse (that is, tilted by $\phi = 30^\circ$ with respect to the sample surface normal) using a superconducting magnet. Ultrafast heating transiently suppresses the PMA in the sample and thereby creates another equilibrium direction defined by the weakened field of the PMA and $\mathbf{H}_{\text{ext}}$ [37]. Subsequently, the magnetization $\mathbf{M}$ starts to precess around the other equilibrium direction.

The details of the optical setup are as follows: First, 800-nm laser pulses with a duration of 80 fs and a pulse energy of 7 mJ were generated by an amplified Ti:sapphire system at 1 kHz. Then, the output beam was split into three beams using two beam splitters. 1% of the pulse energy was used for the gate pulse, and the remaining energy was used in equal parts (i.e., 49.5% for each beam) to generate the 1500-nm pump pulses in an optical parametric amplifier (OPA) and the THz pulses via optical rectification in a LiNbO₃ crystal using the tilted-pulse-front scheme [38]. The sample was placed inside the bore of the superconducting magnet, and the THz beam was focused onto the sample using an off-axis parabolic mirror with a focal length of 190.5 mm.

B. Extraction of the longitudinal conductivity

The longitudinal conductivity σ_{xx} was obtained from the complex transmission spectrum $T(\omega) = (E_{\text{multilayer}, +\text{Hext}} + E_{\text{multilayer}, -\text{Hext}}) / (E_{\text{ref}, +\text{Hext}} + E_{\text{ref}, -\text{Hext}})$, where $E_{\text{multilayer}, +\text{Hext}}(\omega)$ and $E_{\text{ref}, +\text{Hext}}(\omega)$ are the Fourier

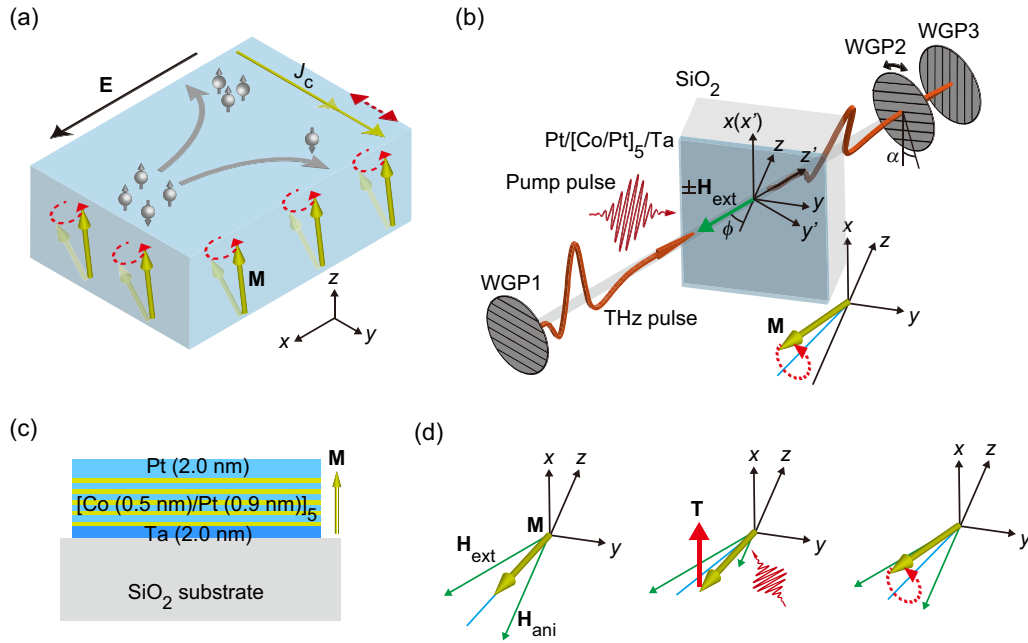


FIG. 1. (a) Schematic of the AHE in a magnet, in which the magnetization precesses as illustrated by red broken arrows. The precession leads to a modulation of the transverse current J_c (illustrated by red broken double-headed arrow). (b) Experimental setup for the measurement of the Faraday rotation of the THz pulse inside the ferromagnetic thin-film sample (light blue), which is excited by a pump pulse. The incident THz pulse is polarized along the x' axis, and its propagation direction is parallel to the z' axis. The external magnetic field $\mathbf{H}_{\text{ext}}$ is also parallel to the z' axis. The angle ϕ between $\mathbf{H}_{\text{ext}}$ and the sample surface normal (which corresponds to the z axis) was controlled by rotating the sample around the x' axis. The angle α is defined as the angle between the x' axis and the transmission direction of WGP2. (c) The ferromagnetic multilayer structure deposited on a SiO_2 substrate. (d) The orientation of the magnetization $\mathbf{M}$ at different times to clarify the mechanism that leads to magnetization precession: before excitation (left), at the moment of excitation (center), and after excitation (right). $\mathbf{T}$ refers to the torque due to the transient change of the direction of total field $\mathbf{H}_{\text{ext}} + \mathbf{H}_{\text{ani}}$.

transforms of $E_{x'}$ for the multilayer sample on the SiO_2 substrate and a bare SiO_2 substrate in the case of an external magnetic field as shown in Fig. 1(b), respectively, and $E_{\text{multilayer}, -H_{\text{ext}}}(\omega)$ and $E_{\text{ref}, -H_{\text{ext}}}(\omega)$ are the Fourier transforms in the case of an opposite external magnetic field. The multilayer sample is treated as an 11-nm-thick

single-layer film with an effective refractive index. For the static measurements (that is, when no 1500-nm pump pulse was applied), the THz pulses were normally incident on the sample ($\phi = 0^\circ$). The effective refractive index n_0 was calculated using an equation that includes multiple internal reflections,

$$T(\omega) = \frac{2n_0(n_{\text{sub}} + n_{\text{air}})}{(n_{\text{air}} + n_0)(n_{\text{sub}} + n_0)} \frac{e^{i((n_0 - n_{\text{air}})\omega d/c)}}{1 - ((n_{\text{air}} - n_0)/(n_{\text{air}} + n_0))((n_{\text{sub}} - n_0)/(n_{\text{sub}} + n_0))e^{i(2n_0\omega d/c)}}, \quad (4)$$

where n_{sub} is the refractive index of the substrate ($n_{\text{sub}} = 1.99$), n_{air} is the refractive index of air ($n_{\text{air}} \approx 1$), d is the thickness of the multilayer structure ($d = 11$ nm), and c is the light velocity in vacuum. Based on the result for $n_0(\omega)$, the effective longitudinal conductivity was calculated according to

$$\sigma_{xx} = i\varepsilon_0\omega(1 - n_0^2), \quad (5)$$

where ε_0 is the vacuum permittivity.

Similarly, the σ_{xx} after the excitation with the 1500-nm pump pulse at a certain time delay t_2 was obtained from $T_{\text{pp}} = (E_{w/, H_{\text{ext}}} + E_{w/, -H_{\text{ext}}})/(E_{w/o, H_{\text{ext}}} + E_{w/o, -H_{\text{ext}}})$, where $E_{w/, \pm H_{\text{ext}}}(\omega)$ and $E_{w/o, \pm H_{\text{ext}}}(\omega)$ are the THz spectra with and without excitation, respectively. Because the THz pulses were obliquely incident on the sample for these pump-probe measurements, the angles of incidence and refraction of the THz pulses in the substrate (ϕ and ϕ_{sub} , respectively) need to be considered. The effective refractive index n after excitation at a certain time delay t_2 was calculated by solving the equation

$$T_{pp}(\omega) = \frac{n}{n_0} \frac{(\cos \phi n_{\text{air}} + n_0)(\cos \phi_{\text{sub}} n_{\text{sub}} + n_0)}{(\cos \phi n_{\text{air}} + n)(\cos \phi_{\text{sub}} n_{\text{sub}} + n)} e^{i((n-n_0)\omega d/c)} \times \frac{1 - ((\cos \phi n_{\text{air}} - n_0)/(\cos \phi n_{\text{air}} + n_0))((\cos \phi_{\text{sub}} n_{\text{sub}} - n_0)/(\cos \phi_{\text{sub}} n_{\text{sub}} + n_0))e^{i(2n_0\omega d/c)}}{1 - ((\cos \phi n_{\text{air}} - n)/(\cos \phi n_{\text{air}} + n))((\cos \phi_{\text{sub}} n_{\text{sub}} - n)/(\cos \phi_{\text{sub}} n_{\text{sub}} + n))e^{i(2n\omega d/c)}}, \quad (6)$$

and then σ_{xx} was calculated according to Eq. (5).

C. Extraction of the anomalous Hall conductivity

The anomalous Hall conductivity σ_{xy} was obtained from the two orthogonal components of the THz spectrum, $E_{x'}(\omega)$ and $E_{y'}(\omega)$ measured by Terahertz polarimetry [5]. Based on the measurement results, the frequency dependence of the rotation angle, $\theta(\omega)$, and that of the ellipticity angle, $\eta(\omega)$, were calculated as follows:

$$\theta(\omega) = 0.5 \left[\arctan \left(\frac{\text{Re}(E_{x',H_{\text{ext}}}^*(\omega)E_{y',H_{\text{ext}}}(\omega))}{|E_{x',H_{\text{ext}}}(\omega)|^2 - |E_{y',H_{\text{ext}}}(\omega)|^2} \right) - \arctan \left(\frac{\text{Re}(E_{x',-H_{\text{ext}}}^*(\omega)E_{y',-H_{\text{ext}}}(\omega))}{|E_{x',-H_{\text{ext}}}(\omega)|^2 - |E_{y',-H_{\text{ext}}}(\omega)|^2} \right) \right], \quad (7)$$

$$\eta(\omega) = 0.5 \left[-\arcsin \left(\frac{\text{Im}(E_{x',H_{\text{ext}}}^*(\omega)E_{y',H_{\text{ext}}}(\omega))}{|E_{x',H_{\text{ext}}}(\omega)|^2 + |E_{y',H_{\text{ext}}}(\omega)|^2} \right) + \arcsin \left(\frac{\text{Im}(E_{x',-H_{\text{ext}}}^*(\omega)E_{y',-H_{\text{ext}}}(\omega))}{|E_{x',-H_{\text{ext}}}(\omega)|^2 + |E_{y',-H_{\text{ext}}}(\omega)|^2} \right) \right]. \quad (8)$$

In the case that these parameters have been obtained with a normally incident THz pulse, the anomalous Hall conductivity can be expressed as [4,39]

$$\sigma_{xy}(\omega) = (\theta(\omega) + i\eta(\omega)) \frac{1 + n_{\text{sub}} + Z_0 d \sigma_{xx}(\omega)}{Z_0 d}, \quad (9)$$

where $Z_0 = 377 \, \Omega$ is the vacuum impedance. While the values of $\theta(\omega)$ and $\eta(\omega)$ obtained in our pump-probe measurements (where we used oblique incidence) are different from those for normal incidence due to the difference between the transmittance values of the *s*- and *p*-polarized electric fields at the backsides of the multilayer structure (including the Pt and Ta layers) and the substrate, Eq. (9) remains applicable, because the differences between $\theta(\omega)$ [and also $\eta(\omega)$] for $\phi = 0^\circ$ and $\phi = 30^\circ$ are significantly small: by using the transmittance matrix from Ref. [40], $\theta(\omega)$ and $\eta(\omega)$ at $\phi = 30^\circ$ were estimated to be only 1.07 times larger than the spectra obtained using normal incidence. Then, the influence of the NA of the parabolic mirror is discussed below. The parabolic mirror has a diameter of 50.8 mm and a focal length of 190.5 mm,

which corresponds to a NA of 0.13 and a half-angle of the light cone of 7.7° . For oblique incidence, the incident angle of the THz beam within the light cone lies in the range $30^\circ \pm 7.7^\circ$. Since the calculation results of σ_{xx} and σ_{xy} derived from the measured Faraday rotation should represent an average over the entire light cone, the influence of the NA of 0.13 on σ_{xx} and σ_{xy} can be ignored. For normal incidence, the values of $\cos \phi$ and $\cos \phi_{\text{sub}}$ for the different rays within the light cone lie close to one (0.991–1 and 0.998–1, respectively), which results in a negligible influence on the derived σ_{xx} and σ_{xy} .

III. RESULTS AND DISCUSSION

A. Static conductivity spectra

The incident THz electric field is polarized along the x' axis, and due to the AHE, a nonzero y' -axis component emerges, and thus the polarization direction of the field detected after the sample is different. The black broken

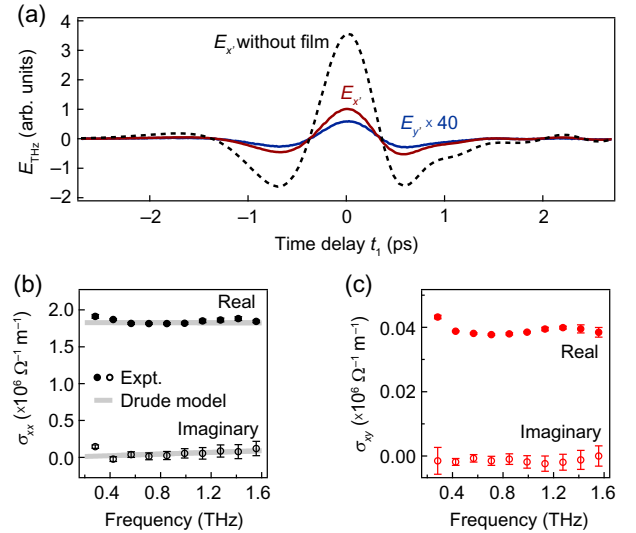


FIG. 2. (a) The x' - and y' -axis components of the electric-field waveform of the THz pulse after the sample for $H_{\text{ext}} = 14$ kOe and $\phi = 0^\circ$ in the static case (that is, no pump pulse was applied). The black broken curve represents the incident electric field. (b) The extracted spectra of the longitudinal conductivity σ_{xx} : the real and imaginary parts are shown as solid and open circles, respectively. The gray curve shows the fitting result obtained using the Drude model. (c) The extracted spectra of the anomalous Hall conductivity σ_{xy} . The error bar represents the standard deviation determined from a set of three measurements.

curve in Fig. 2(a) represents the incident electric field. The x' - and y' -axis components of the electric field after sample obtained without applying the 1500-nm pump pulse are shown as the dark red and blue solid curves in Fig. 2(a), respectively, where $E_{y'}$ is magnified by a factor of 20. The longitudinal conductivity σ_{xx} extracted from the transmittance spectrum and the anomalous Hall conductivity σ_{xy} evaluated from the Faraday rotation signal at 250 K are shown in Figs. 2(b) and 2(c), respectively. (For comparison: the room-temperature values of the dc conductivity and the Hall conductivity obtained are $1.60 \times 10^6 \Omega^{-1} \text{m}^{-1}$ and $2.87 \times 10^4 \Omega^{-1} \text{m}^{-1}$, respectively.) By fitting the σ_{xx} data to the equation $\sigma_{xx} = \sigma_0 / (1 - i\omega\tau)$ from the Drude model, where σ_0 is the dc conductivity and ω is the angular frequency, the electron-electron scattering time τ is determined to be 5 fs, which is a typical value observed in metals [41]. The spectra of the real and imaginary parts of σ_{xy} are rather flat and the imaginary part is almost negligible, indicating that the AHE in our sample at 250 K is dominated by the intrinsic contribution from the Berry curvature, $\sigma_{\text{AHE}} (= \int \Omega f(\mathbf{k}) d\mathbf{k}$, where Ω is the Berry curvature and f corresponds to the Fermi-Dirac distribution).

B. Ultrafast modulation of the THz electric field by coherent magnetization precession

To clarify the impact of the 1500-nm pump pulses on the transverse current J_c , we scanned the time delay between the pump pulse and the THz pulse for $\phi = 0^\circ$ and 30° and used the detection condition $\alpha = 90^\circ$ and $t_1 = 0$ ps, which allows us to observe the pump-induced variation in the peak amplitude of $E_{y'}$, $\Delta E_{y'}$. The plots of $-\Delta E_{y'}$ in Fig. 3(a) clarify that, independent of ϕ , $E_{y'}$ first decreases very fast and then slowly recovers. The initial decrease is attributed to the suppression of the AHE by ultrafast demagnetization. Subsequently, due to energy and angular-momentum exchange between the electron and lattice systems, the spin temperature (and hence also the magnetization) gradually approaches the equilibrium condition.

Furthermore, when the angle ϕ between $\mathbf{H}_{\text{ext}}$ and the surface normal is 30° , a slow oscillation appears during the recovery, whereas no oscillation is observed when $\mathbf{H}_{\text{ext}}$ is perpendicular to the sample surface. This dependence on ϕ already suggests that the observed oscillation is caused by a precession of $\mathbf{M}$, since both the excitation and the detection of a precession of $\mathbf{M}$ are forbidden for $\phi = 0^\circ$ but allowed for $\phi \neq 0^\circ$. To verify this interpretation, we repeated the experiment for $\phi = 30^\circ$ using various different values of $\mathbf{H}_{\text{ext}}$, as shown in Fig. 3(b). The observed oscillation frequency shows a clear dependence on the field strength, and this dependence can be well described by the ferromagnetic resonance (FMR) model: the experimental data were fitted to a superposition of a double-exponential

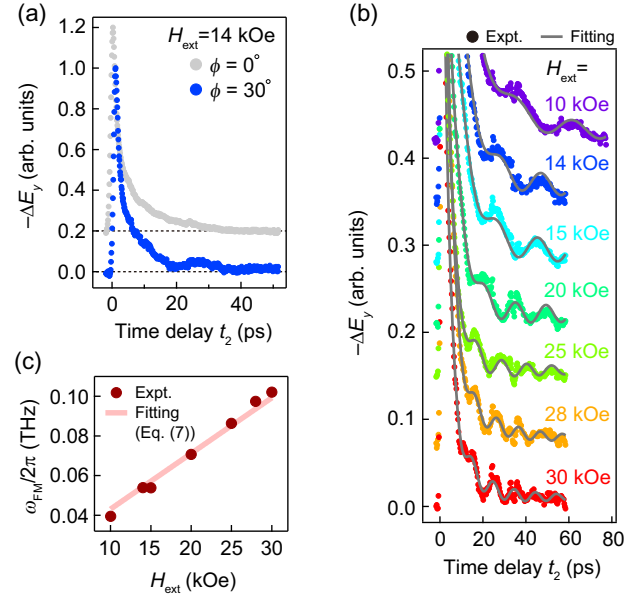


FIG. 3. (a) The change of the THz peak amplitude projected onto the y' axis, $\Delta E_{y'}$, plotted as a function of the time delay between the pump pulse and the THz pulse. Here, we used $H_{\text{ext}} = 14$ kOe. The data measured using $\phi = 0^\circ$ and 30° are shown in gray and blue, respectively. (b) The dependence of the temporal evolution of $\Delta E_{y'}$ on H_{ext} for $\phi = 30^\circ$. The gray curves are the fits to a superposition of a double-exponential function and a damped oscillation. (c) The extracted oscillation frequencies. The red curve represents the fitting result obtained using the FMR model.

function and a damped oscillation. Note that the frequency of ferromagnetic resonance, ω_{FM} , is given by [42]

$$\omega_{\text{FM}} = \gamma \sqrt{(H_{\text{ext}} + H_{\text{ani}} \sin(\pi/2 - \phi))(H_{\text{ext}} - H_{\text{ani}} \cos(\pi - 2\phi))}, \quad (10)$$

where H_{ani} is the strength of the effective field of the PMA. The modeling of the recovery process after demagnetization would require us to solve a set of coupled differential equations for the electron, spin, and lattice temperatures, if the details of the recovery were essential, but such a detailed treatment is not needed for our present aim. As shown in Fig. 3(c), the observed trend can be well described by Eq. (10). H_{ani} was determined to be 8 kOe [42], which is consistent with the literature value, and this confirms that the oscillation arises from a precession of $\mathbf{M}$.

C. Ultrafast modulation of the anomalous Hall conductivity

To investigate how the ultrafast magnetization dynamics in this sample modify the anomalous Hall conductivity σ_{xy} , we measured the Faraday rotation spectra for 1500-nm excitation at various different time delays t_2 . The

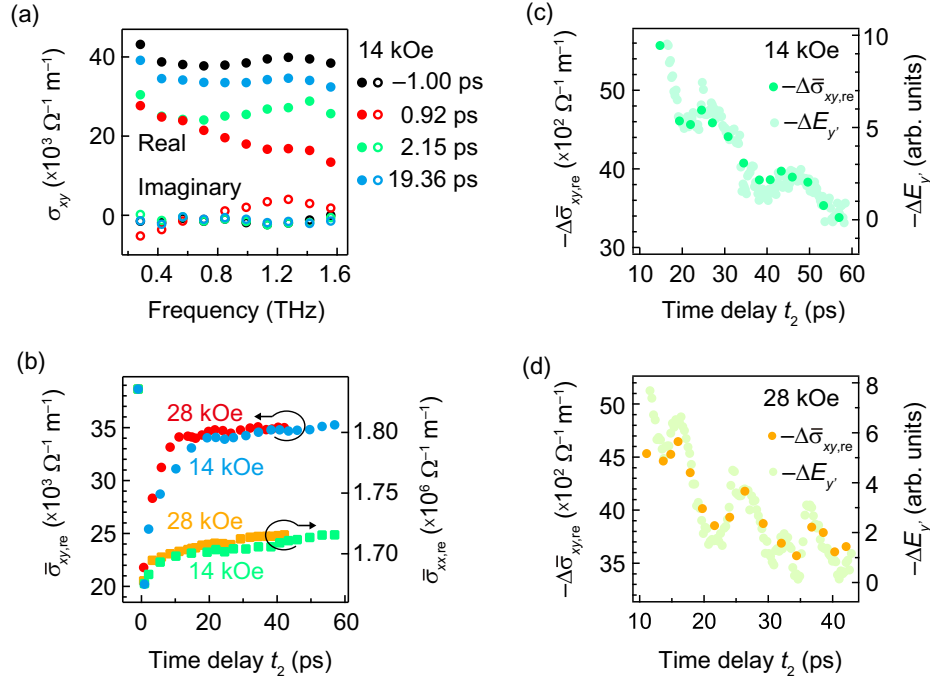


FIG. 4. (a) The spectra of the complex anomalous Hall conductivity σ_{xy} at four different values of t_2 for $\phi = 30^\circ$. The real and imaginary parts are shown as solid and open circles, respectively. (b) The temporal evolution of the averaged real part of the anomalous Hall conductivity, $\bar{\sigma}_{xy,\text{re}}$ (solid circles), and the averaged real part of the longitudinal conductivity, $\bar{\sigma}_{xx,\text{re}}$ (solid squares), for $H_{\text{ext}} = 14$ and 28 kOe, where averaging was performed over the range of 0.4 – 1.3 THz. (c), (d) Comparison between the oscillation period of $\Delta\bar{\sigma}_{xy,\text{re}}$ (green, orange) and that of $\Delta E_{y'}$ (semitransparent green, light green) for two different values of H_{ext} (14 and 28 kOe, respectively). $\Delta\bar{\sigma}_{xy,\text{re}}$ is defined as the difference between the $\bar{\sigma}_{xy,\text{re}}$ values obtained with and without the pump pulse.

spectra of the extracted real and imaginary parts of σ_{xy} (denoted by $\sigma_{xy,\text{re}}$ and $\sigma_{xy,\text{im}}$, respectively) are presented in Fig. 4(a) for four values of t_2 . For $t_2 = 0.92$ ps (that is, immediately after the pump pulse), both $\sigma_{xy,\text{re}}$ and $\sigma_{xy,\text{im}}$ exhibit pronounced changes compared to the equilibrium values observed at $t_2 = -1$ ps, while the spectra for $t_2 = 19.36$ ps are close to the equilibrium values. We find that the spectral changes (relative to the equilibrium spectra) for $t_2 = 2.15$ and 19.36 ps are nearly independent of the frequency, whereas the spectra for $t_2 = 0.92$ ps show a clear frequency dependence with larger variations at higher frequencies. This result seems to be the relatively large FWHM of the THz electric-field envelope (1.4 ps): when the peak electric field of the THz pulse temporally overlaps with that of the pump pulse (with a duration of 80 fs) at a certain value of t_2 , only the trailing part of the THz pulse ($t_1 > t_2$) is modified by the pump pulse [43], which means that a strong reduction of the high-frequency components in the Fourier transform is only observed as long as a significant part of the THz peak electric field is not suppressed. Figure 4(b) shows the temporal evolution of the real part of the anomalous Hall conductivity averaged over the frequency range 0.4 – 1.3 THz, $\bar{\sigma}_{xy,\text{re}}$, for $H_{\text{ext}} = 14$ and 28 kOe. Figures 4(c) and 4(d), which plot $\Delta\bar{\sigma}_{xy,\text{re}}$ (that is, the difference between the $\bar{\sigma}_{xy,\text{re}}$ values obtained with and without the pump pulse) as a function

of t_2 , clarify that a clear oscillation with a frequency that matches that observed in Fig. 3(b) is observed for both $H_{\text{ext}} = 14$ and 28 kOe, which confirms that the coherent magnetization precession in this sample induces a modulation of the anomalous Hall conductivity at high GHz frequencies.

It has been shown that $\sigma_{xy,\text{re}}$ can be expressed in terms of a product of the out-of-plane magnetization $M_\perp$ and a frequency-dependent coefficient $\beta(\omega)$, where $\beta(\omega)$ is associated with the product of the electron distribution and the Berry curvature [44]. By comparing the variation of the $\bar{\sigma}_{xy,\text{re}}$ data in Fig. 4(b) with the corresponding data of $\bar{\sigma}_{xx,\text{re}}$, it should be possible to distinguish between the regions where either the change in β or $M_\perp$ governs the observed variation of $\bar{\sigma}_{xy,\text{re}}$, because variations of $\bar{\sigma}_{xx,\text{re}}$ are usually governed by a change in the electron distribution. As shown in Fig. 4(b), the decrease of $\bar{\sigma}_{xx,\text{re}}$ around $t_2 = 0$ ps corresponds to the process of ultrafast excitation of the electron distribution, and the increase in the latter region corresponds to the subsequent electron-phonon thermalization. After approximately 15 ps, the inclination of the gradual increase of $\bar{\sigma}_{xy,\text{re}}$ is similar to that of $\bar{\sigma}_{xx,\text{re}}$. On the other hand, in the region $t_2 < 15$ ps, the recovery behavior of $\bar{\sigma}_{xy,\text{re}}$ is significantly different from that of $\bar{\sigma}_{xx,\text{re}}$, indicating that the change in $M_\perp$ is dominant during this early stage. Note that the relatively large Faraday rotation

of the multilayer structure used in this work compared to the pure Co film is caused by a smaller electrical conductivity, and there is maybe also a contribution from a larger coefficient β .

IV. CONCLUSIONS

We investigated the ultrafast precession of magnetization in a ferromagnetic thin film after optical excitation. We have demonstrated that coherent ferromagnetic resonance induces a modulation of the anomalous Hall conductivity that can be tracked by THz polarimetry, and this detection scheme is considered to be useful for the field concerned with the convergence of spintronics and THz electronics. The presented method may, for example, also be used to explore the coherent manipulation of the Berry curvature in the topological antiferromagnet MnBi_2Te_4 or the magnetic Weyl semimetal $\text{Co}_3\text{Sn}_2\text{S}_2$ where the band topology is determined by the spin orientation. Such investigations are considered to be essential for the controlled excitation of dynamical axion quasiparticles in condensed-matter systems [45].

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DATA AVAILABILITY

The data that support the findings of this article are openly available [46].

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