Laser-induced magnetization dynamics in a van der Waals ferromagnetic Cr$_2$Ge$_2$Te$_6$ nanoflake

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ABSTRACT
Laser-induced magnetization dynamics is quantitatively investigated in a van der Waals ferromagnetic \( \text{Cr}_2\text{Ge}_2\text{Te}_6 \) nanoflake by means of time-resolved Faraday rotation. Under ferromagnetic resonance conditions, the angular dependence of spin precession dynamics gives rise to a perpendicular magnetic anisotropy with an effective field of 125 ± 8 mT. We further determine the field dependence of the effective damping coefficient, which is dominated by the inhomogeneous broadening of magnetic anisotropy in the regime of a small magnetic field while it diminishes to an intrinsic value of 0.006 ± 0.002 at high fields.

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Semiconductor spintronics promises applications in information processing of the next generation. Semiconductor spin-based quantum systems have led to numerous discoveries, such as the spin Hall effect,1–3 spin-helix states,4,5 the quantum spin Hall effect,6 the quantum anomalous Hall effect,7 and Majorana particles.8 Moreover, scalability and electrical tunability make these solid systems particularly interesting for quantum computing. Two-dimensional (2D) van der Waals (vdW) semiconductors represent a promising alternative in semiconductor spintronics.9 Compared with epitaxially grown semiconductor quantum systems, 2D vdW semiconductors are easier to fabricate by mechanical exfoliation and offer strong intrinsic quantum confinement. In addition, the thickness of 2D materials can be precisely controlled with monolayer accuracy. In such vdW structures, atomic layers can be artificially stacked with controllable orientation, thereby allowing the crystal symmetry to be tuned as desired. This unique property allows flexible engineering of the electronic band structure, either by controlling the number of layers or by stacking heterostructures of different 2D materials. The number of layers determines the symmetry along the confinement direction, which drastically affects the energy levels of spin subbands and valley subbands.10 As a result, numerous spin-related phenomena have been observed in 2D atomic crystals, such as giant magnetoresistance,11 the spin or valley Hall effect,12,13 and spin-orbit torque.14,15 Of more interest is the recent discovery of a 2D ferromagnet in which the ferromagnetism depends strongly on the number of layers,16,17 which prompted measurements of the anomalous Hall effect.18 Furthermore, the electrical control of ferromagnetism was experimentally demonstrated in different 2D systems,19–21 an example of which is our recent work that demonstrates optically addressed electrical tunability of ferromagnetism in vdW semi-insulating \( \text{Cr}_2\text{Ge}_2\text{Te}_6 \).21

Here, we study magnetization dynamics in a nanoflake \( \text{Cr}_2\text{Ge}_2\text{Te}_6 \), by means of a two-color time-resolved Faraday rotation (TRFR) with a picosecond time resolution at cryogenic temperature. Below the Curie temperature, a pump beam of femtosecond pulses intensively heats the ferromagnet to produce instantaneous demagnetization and trigger the magnetization precession. By monitoring laser-induced magnetization dynamics in an external magnetic field \( H_{\text{ext}} \), we determine the angular dependence of the precession
frequency and the transverse spin relaxation time. The precession frequency can be well fitted by the Kittel formula with an effective perpendicular anisotropic (PMA) magnetic field of $\mu_0H_0 = 125 \pm 8 \text{ mT}$. By evaluating the field dependence of the spin relaxation rate, we propose that the effective damping coefficient $\zeta_{\text{eff}}$ in the regime of a small $H_{\text{ext}}$ is dominated by inhomogeneous broadening due to PMA spatial fluctuations, and it diminishes to an intrinsic coefficient $\zeta_0 = 0.006 \pm 0.002$ at higher $H_{\text{ext}}$.

The sample investigated is a Cr$_2$Ge$_2$Te$_6$ flake exfoliated from a single crystal and encapsulated between two hexagonal boron nitride (h-BN) flakes on a fused silica substrate. To avoid degradation of the nanoflake, the whole sample fabrication is performed in a nitrogen-purged glovebox. Figure 1(a) shows a schematic description of the stacked structure, and Fig. 1(b) presents an optical micrograph showing the measurement area by the solid circle. Based on the scanning topography characterization presented in Fig. 1(c), the sample thickness is determined to be 10.5 nm. The inset of Fig. 1(c) shows the unit cell of Cr$_2$Ge$_2$Te$_6$, in which the ferromagnetism is due to the labeled Cr atoms.

The sample is mounted in a helium-free cryostat, and all measurements presented in this Letter are done at a sample temperature of 10 K, which is well below the Curie temperature of Cr$_2$Ge$_2$Te$_6$.\textsuperscript{22} We first measure the magnetization hysteresis loops by using magneto-optic Kerr rotation in Faraday geometry. More technical details are described in our previous work.\textsuperscript{21} We define $H_{\text{c}}(0^\circ)$ as the angle between $H_{\text{ext}}$ and the sample normal (i.e., $\theta_1 = 0^\circ$ in this configuration). As shown in Fig. 1(d), the out-of-plane coercivity of the Cr$_2$Ge$_2$Te$_6$ investigated is determined as $H_{\text{c}}(0^\circ) = 8.0 \pm 0.1 \text{ mT}$. This coercivity is similar to the values previously reported for similar samples from 3.5 to 20 nm thick.\textsuperscript{23}

The sample normal is rotated through the angle $\theta_1$ with respect to $H_{\text{ext}}$, which is always perpendicular to the optical axis. Hereinafter, we record magnetization by means of Faraday rotation, which presents the magnetization component along the optical axis. The probe beam is focused on the sample in a cryostat by using an achromatic lens with a numerical aperture of 0.42, and the transmitted beam is recollimated by another identical lens. The spatial resolution of this setup is estimated to be $d_0 = 2 \mu\text{m}$ in Voigt geometry (i.e., $\theta_1 = 90^\circ$), and we expect that the beam focus is elliptically lengthened to be $d_0/\sin(\theta_1)$ along the tilted axis.

Figure 1(d) shows more magnetization hysteresis loops, which are obtained by changing $\theta_1$ to 30°, 40°, 50°, and 60°. The respective magnetic coercivity $H_{\text{c}}(\theta_1)$ is 9.5 ± 0.1 mT, 10.4 ± 0.1 mT, 12.4 ± 0.1 mT, and 16.0 ± 0.1 mT. Experimentally, the relation $H_{\text{c}}(0^\circ) = H_{\text{c}}(\theta_1)\cos\theta_1$ is well followed, and this demonstrates that the field component in the out-of-plane direction dominates the magnetization reversal. These results are consistent with the previous findings that only the c axis is the easy axis in ferromagnetic Cr$_2$Ge$_2$Te$_6$ with perpendicular magnetic anisotropy.\textsuperscript{22,23}

To study magnetization dynamics in the Cr$_2$Ge$_2$Te$_6$ ferromagnet, we have developed TRFR of a two-color pump–probe technique, which is schematically described in Fig. 2(a). The pump beam consists of a pulse train with a 78 MHz repetition rate generated by a

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**FIG. 1.** (a) Schematic of a Cr$_2$Ge$_2$Te$_6$ flake encapsulated by two h-BN layers on a fused silica substrate and (b) optical micrograph of the Cr$_2$Ge$_2$Te$_6$ sample (scale bar: 15 $\mu$m). (c) Height profile measured by atomic force microscopy along the sample area. The inset is a schematic drawing of the unit cell for Cr$_2$Ge$_2$Te$_6$. (d) Magnetization hysteresis loops by varying $\theta_1$ at 10 K, where the scale bar on the lower left indicates the measurement error.

**FIG. 2.** (a) Schematic description of a two-color TRFR technique. (b) Normalized magnetization dynamics due to laser heating in Cr$_2$Ge$_2$Te$_6$ for different $H_{\text{ext}}$ at $\theta_1 = 50^\circ$. Solid lines are fits and dashed lines are baselines in the absence of pumping. Inset: Magnetization dynamics (markers) and cumulative Gauss fit (line) in the first 10 ps (left axis). Dashed line (right axis) is the differential curve of the fit, which gives a time resolution of 1.2 ± 0.01 ps.
mode-locked Ti: sapphire laser, with a pulse width of 150 fs at a center wavelength of 770 nm. The probe beam with a sub-picosecond pulse width is produced by exciting a photonic crystal fiber by another sequence of synchronized ultrafast pulses to generate supercontinuum pulses from which the probe beam is selected at 815 nm. Both the pump and probe beams are focused on the sample surface by the same achromatic lens. The energy density of a single pump (probe) pulse is chosen to be constant at 123 μJ/cm² (2 μJ/cm²). The time delay Δt between the pump and probe pulses is controlled by using a motorized translation stage to vary the optical path of the pump beam with respect to the probe beam. The intensities of the pump beam and the probe beam are modulated at 10 kHz and 83 Hz, respectively. The Faraday rotation of the probe beam is detected by a balanced bridge of the probe beam are modulated at 10 kHz and 83 Hz, respectively. The time resolution of the following pump–probe measurements is estimated to be 1.2 ± 0.01 ps [see the inset of Fig. 2(b)], and the spatial resolution is δl/sin(θ/2) as described above.

In an equilibrium state, as denoted in Fig. 2(a), the angle θ_M by which the magnetization deviates from the easy axis is determined by using $H_k \cos \theta_M \sin \theta_M = H_{ext} \sin \theta_M - \theta_M) ^2$, where $H_k$ is defined as $H_k = 2 K_u / M_s - 4 \pi M_u$ with $K_u$ being defined as the uniaxial magnetic anisotropy energy along the c axis and $M_s$ being the saturation magnetization. The out-of-plane effective field is $H_{eff} = H_k \cos \theta_M$, so the static magnetization energy remains along the total effective field $H_{tot} = H_{eff} + H_{ext}$. Based on this relation, at a certain $θ_M$, $θ_M$ asymptotically reaches $θ_M$ by increasing $H_{ext}$, and in the case of a fixed $H_{ext}$, $θ_M$ increases with $H_{ext}$.

An intensive optical pulse can remarkably perturb the magnetization of a ferromagnet. Figure 2(b) plots the laser-induced magnetization dynamics up to 1500 ps for $θ_M = 50°$ at three external fields: $H_{ext} = 211, 418, and 680$ mT. The inset presents the initial demagnetization dynamics in the first few picoseconds. This ultrafast process can be qualitatively understood by considering a drastic increase in the spin temperature leading to a reduction of both the magnetization M and the effective magnetic field $H_{eff}$. As shown in Fig. 2(a), the total magnetic field $H_{tot} = H_{eff} + H_{ext}$ is modified to tilt the magnetization M away from its equilibrium direction. After spin cooling of a few picoseconds, M recovers its saturation magnetization value while the electron temperature cools through phonon emission. Meanwhile, $H_{eff}$ recovers dynamically to the value at thermal equilibrium, so M recovers according to $H_{tot} = H_{eff} + H_{ext}$ with spin precession.

The change $ΔM(Δt)$ of magnetization dynamics probed by the Faraday rotation can be phenomenally described as $ΔM(Δt) ≈ θ_M(Δt) = A_m e^{-\lambda M t \cos(2πf_0 t + \phi_m) + A_p e^{-\lambda P t} + A_e e^{-\lambda E t}}$. The first term describes the spin-precession dynamics in which $A_m$, $\tau_p$, $f_0$, and $\phi_m$ denote the magnetization amplitude, the transverse spin relaxation rate, the precession frequency, and the initial phase, respectively. The second and third terms describe the magnetization recovery process, where $A_p$ ($\lambda_p$) and $A_e$ ($\lambda_e$) are, respectively, the demagnetization magnitude and the characteristic time constant of a short (long) process. By fitting the experimental data, we obtain the precession frequency as $f_0 = 7.34 ± 0.01$ GHz, 12.89 ± 0.01 GHz, and 20.32 ± 0.03 GHz, and $A_m = 0.095 ± 0.016$, 0.073 ± 0.087, and 0.055 ± 0.015 for $H_{ext} = 211, 418, and 680$ mT, respectively. The precession frequency clearly increases, while the oscillation magnitude decreases. Qualitatively, this coincides with the spin detection scenario described in Fig. 2(a), i.e., a large $H_{ext}$ contributes to a larger $H_{tot}$ giving rise to a larger precession frequency and simultaneously a larger $θ_M$, which in turn decreases the spin projection on the optical axis. In addition, we obtain the magnetization recovery-time constants $τ_p ≈ 400 ± 150$ ps and $τ_e ≈ 8 ± 2$ ns, which are similar for different $H_{ext}$. Two relaxation processes are quite likely related to energy relaxation by optical phonons and acoustic phonons in semiconductor structures at low temperatures. For each curve, a cumulative heating effect is indicated at $Δt < 0$ by a clear offset from the dashed baseline.

To more systematically investigate the magnetization dynamics of this sample, TRFR measurements of $H_{ext}$ dependence at various $θ_M$ are performed and fitted by using the method described above. The frequency $f$ and the transverse spin relaxation rate $τ_p$ as a function of the external magnetic field are shown in Figs. 3(a) and 3(b), respectively. To analyze the data more quantitatively, we use the expression $f = (H_0/H_s) ^2 / 2\pi$ with a PMA based on the Kittel formula. Here, $H_0 = H_{ext} \cos(θ_M - θ_M) + H_k \cos^2 θ_M$, $H_s = H_{ext} \cos(θ_M - θ_M) + H_k \cos(2θ_M)$, and $γ = γ_B / h$, in which γ, $μ_B$, and h are the spectroscopic splitting factor, the Bohr magneton, and the reduced Planck constant, respectively. In addition to using $H_{ext} \cos θ_M \sin θ_M = H_{ext} \sin θ_M - θ_M)$, we fit all curves of $f$ vs $H_{ext}$ by applying the same set of parameters $H_k$ and $γ$ [see the solid lines in Fig. 3(a)]. We obtain $μ_B H_k = 125 ± 8$ mT and a g factor of 2.04 ± 0.03. Based on the definition $H_k = 2 K_u / M_s - 4 \pi M_u$ and the previous measurement of $M_u = 192$ emu/cm³, we determine the magnetic anisotropy energy density $K_u = 3.4 \times 10^5$ erg/cm³, which is similar to the value obtained by ferromagnetic resonance for bulk Cr₃Ge₂Te₆. In addition, Fig. 3(c)
plots the corresponding $\theta_M$ as a function of the external magnetic field, which is consistent with the scenario described in Fig. 2(a), i.e., $\theta_M$ increases to $\theta_{H1}$ upon increasing $H_{ext}$.

As a key parameter in the Landau–Lifshitz–Gilbert equation, the damping coefficient is regarded to more conveniently describe the magnetization relaxation. By combining the Landau–Lifshitz–Gilbert equation and the Kittel formula, the effective damping coefficient $x_{eff}$ is given by the relation $x_{eff} = 2/(\tau_0 H_{1} + H_{2})$. Figure 4 shows $x_{eff}$ as a function of frequency $f$, which is obtained by using $\tau_f$ from Fig. 3(b) and $\theta_M$ from Fig. 3(c) to determine $H_1$ and $H_2$.

Despite the intrinsic damping constant $x_0$ being regarded independent of the external field, it clearly decreases with increasing precession frequency and approaches a constant value that is independent of $H_{ext}$ and $\theta_M$. For this point, we analytically decompose the effective damping coefficient into an intrinsic damping $x_0$ and an extrinsic damping $x_{eff}$ (i.e., $x_{eff} = x_0 + x_{ex}$), in which $x_{eff}$ is due to inhomogeneous broadening and depends on $f$. Here, we propose the spatial fluctuations of the perpendicular magnetic anisotropy as the main extrinsic source, and we write $x_{eff} = x_0 + (2\pi n)^{0.5}[\gamma(H_1 + H_2)]^{-1}|\Delta H|/H_{1}$ by using the expression derived in Ref. 32, where $\Delta H$ is the deviation of the PMA magnitude from $H_{1}$ and $|\Delta H|/H_{1}$ is numerically calculated by applying the Kittel formula. As shown by the solid lines in Fig. 4, $x_{eff}$ is well fit with $x_0 = 0.006 \pm 0.002$ and $\mu_0 \Delta H = 8.0 \pm 2.0$ mT. The PMA deviation $\Delta H$ in this sample can be produced during crystal growth or in the preparation of layered $\text{Cr}_2\text{Ge}_2\text{Te}_6$, e.g., by introducing defects or forming terraces by exfoliation, and these local effects are present on a length scale of nanometers which is too far beyond the optical resolution to be distinguished. This inhomogeneous broadening is suppressed in the regime of a large $H_{ext}$, so $x_{eff}$ approaches the intrinsic Gilbert damping coefficient for different $\theta_M$.3,4

Note that the Gilbert damping constant $x_0 = 0.006 \pm 0.002$ for this semi-insulating $\text{Cr}_2\text{Ge}_2\text{Te}_6$ is lower than that measured for many ferromagnetic materials with PMA.3,5,6 This result may be qualitatively understood by considering two facts that are mostly absent in metallic ferromagnets. First, the deviation of the $g$ factor of 2.04 $\pm$ 0.03 from the free-electron value $g = 2$ indicates a tiny orbital contribution to the magnetization. This points to a weak spin–orbit interaction, which induces spin–photon scattering and thereby strongly suppresses the transverse magnetization relaxation.7 In addition, since electrons are mostly localized in a semi-insulating system, spin relaxation due to electron–electron scatterings is highly suppressed in this layered $\text{Cr}_2\text{Ge}_2\text{Te}_6$.

In summary, we report herein the laser-induced magnetization dynamics of a 10.5 nm vdW ferromagnet. By using Kerr and Faraday rotations, we record the magnetization hysteresis loops at various $\theta_M$ and observe the PMA of the $\text{Cr}_2\text{Ge}_2\text{Te}_6$ nanoflake. The magnetization dynamics measured by the in-house-developed TRFR technique reveals an effective PMA field of $\mu_0 H_0 = 125 \pm 8$ mT and an intrinsic damping coefficient of $x_0 = 0.006 \pm 0.002$. Experimentally, the effective damping coefficient is dominated by extrinsic inhomogeneous broadening in the regime of a small external magnetic field and it approaches an intrinsic damping coefficient at high external magnetic fields. We attribute the extrinsic damping to the inhomogeneous broadening of the PMA deviation, with $\mu_0 \Delta H = 8.0 \pm 2.0$ mT. Both the low intrinsic damping coefficient and the PMA of the nanoflake make semi-insulating vdW ferromagnets candidates for low-dissipation magnetic devices, such as spin-transfer–torque magnetic random-access memory. Future studies of particular interest are to investigate the magnetization dynamics in a vdW magnet of a few atomic layers down to a monolayer.

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DATA AVAILABILITY

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

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