

Light-induced magnetization dynamics in the chiral magnet MnSi

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We perform time-resolved magneto-optical Kerr effect measurements and record the magnetization dynamics for external magnetic fields along various symmetry axes of the chiral magnet MnSi. The different orientations of the magnetic field result in very different magnetization dynamics. For magnetic fields along the a - b plane, we observe rapid oscillations assigned to magnon modes. These oscillations are detected for various external magnetic fields and are well reproduced by our theoretical spin-wave calculations. When applying magnetic fields at 45° with respect to the a - b plane and along the c axis, changes in the net magnetization are instead observed. In the absence of external magnetic fields, the net magnetization is zero, and light-induced magnetization is observed. First-principles calculations identify the inverse Faraday effect as the mechanism leading to the observed light-induced magnetization. When sufficiently strong external magnetic fields are applied off the a - b plane, the MnSi crystal is brought into a ferromagnetic phase, where the laser pulse's demagnetizing effect is observed. This observation suggests that by changing the strength or orientation of the external magnetic field the MnSi crystal can be magnetized or demagnetized using a laser pulse.

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I. INTRODUCTION

The ultrafast manipulation of magnetic states is of technological importance in innovative magnetic storage. The progress in the development of compact ultrafast lasers and optical control of magnetic order may potentially revolutionize information processing technologies [1]. The physics that governs light-matter interactions on ultrafast timescales has been a subject of intense study over the past decades. Understanding the response of the magnetic order after the ultrafast stimulation by short laser pulses of circular or linear polarization is important not only for these applications but also from a fundamental scientific perspective. The ultrafast demagnetization of a crystal with nonzero net magnetic moment in the ground state has been observed and studied in detail [2–5], whereas the opposite effect, the generation of net magnetic moment with a laser pulse, is less observed often and is short-lived [6–8]. One method of generating ultrafast light-induced magnetism and magnetic domain switching in magnetic materials has been achieved using circularly polarized laser pulses [8–12]. Linearly polarized laser pulses do not carry any angular momentum, but they can generate magnetic fields through the inverse Cotton-Mouton effect [13], through the nonlinear coupling of electric and magnetic fields of light

[12,14], or via the impulsive stimulated Raman scattering [15]. Finally, in the absence of net magnetization the inertia-driven spin switching has been used as a mechanism to explain observed magnetization oscillations. This model assumes that the system is oscillating between two magnetization states, in a time slower than the excitation pulse duration [16]. Depending on the magnetic phase, linearly polarized light pulses can create net magnetic moment or have the opposite effect and demagnetize the crystal.

Chiral magnets such as MnSi in the helical phase are spontaneously long-range ordered without a net magnetic moment [17], but can be manipulated by light via different mechanisms [1], which include heating effects [18–24], laser-induced spin reorientation [8–12] magnon-phonon coupling [25], and anisotropy effects [26]. They show some similarities to antiferromagnets, which are thought to have great potential for application in antiferromagnetic spintronics and optospintronics [27–31], because of the potential for much higher switching speeds and the absence of dipolar fields. However, antiferromagnets are much harder to manipulate using external magnetic fields due to very large exchange energies [32–36]. The interplay of the various magnetic interactions with different energy scales often leads to the observed magnetic ordering. MnSi is a great example of this class of materials, and has played a major role in developing the theory of helical magnets in noncentrosymmetric materials exhibiting the Dzyaloshinsky-Moriya (DM) interaction

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[37–40]. Furthermore, materials that show helical spin structures are fascinating because they serve as a model system for complex magnetic ordering and possess stable magnetic vortices, also referred to as skyrmions. The recent discovery of stable skyrmions in a small area of the phase diagram has led to renewed excitement about this material [41–43]. However, more recent theoretical and experimental work has extended the temperature range where skyrmions are detected, indicating that they might be present at lower temperatures as a metastable state [44–48].

Early theoretical work proposed an explanation of the phase diagram of MnSi based on the DM interactions originating from noncentrosymmetry [49]. Currently, it is understood that the appearance of most of the magnetic phases in MnSi is a property of the hierarchy of the various energy scales of magnetic interactions appearing in B20 compounds [50,51]. In the helical ground state, the ferromagnetic exchange interaction corresponds to the strongest scale and leads to a preference for parallel spin alignment. The weaker DM interaction favors a perpendicular alignment of the spins, and the competition between them leads to the formation of a spin spiral with a large pitch of 175 Å. In the absence of external magnetic fields, the multidomain spiral helimagnetic phases are aligned along the [111] direction [52,53]. The spin spiral can be excited, leading to helimagnons according to their mapped out dispersion [54,55]. At higher temperatures the MnSi crystal is paramagnetic and only becomes helimagnetic below Curie temperature T_C of ≈ 29 K. When increasing magnetic fields at temperatures $\leq T_C$, the crystal undergoes a helical magnetic phase before becoming weakly ferromagnetic in the field-polarized phase.

Changes in the spin structure are often directly observed in the spin dynamics, such as the magnon frequency. Neutron scattering is utilized to measure the magnon energies and dispersion [56,57], whereas optical techniques are able to resolve helimagnons at $q = 0$, and complement neutron scattering which is restricted to mapping helimagnon bands at higher q [58–62]. Furthermore, the extraordinary time resolution of ultrafast lasers, coupled with the ability to track spin fluctuations via the magneto-optical Kerr effect (MOKE), offers another valuable tool in measuring the spin dynamics [63,64]. Direct measurements of magnon oscillations and their frequency provide valuable insight into the spin structure. Recent experimental work has demonstrated that spin fluctuations are initiated by a laser pulse via magnon-phonon coupling [25]. Furthermore, this mechanism is used to generate light-induced magnetization at ultrafast timescales in a spiral helimagnet without net magnetic moment in the ground state [25,65]. Materials facilitating light-induced magnetization and reversal of magnetization following an ultrafast optical excitation, also referred to as all optical magnetization switching, are crucial for ultrafast optospintronics applications [5,66–69]. Probing the magnetic response of the MnSi crystal upon optical stimulation by laser pulses along the symmetry directions of the crystal is important in understanding the light-induced spin dynamics. Thus, we perform time-resolved magneto-optical Kerr effect (TR-MOKE) experiments in the absence of external fields and with magnetic fields applied along the [100], [101], and [001] directions. When magnetic fields are applied in the [100] direction on the a - b plane, the laser

pulse generates magnons which we detect via TR-MOKE. The response is different when the magnetic fields are applied in the [101] and [001] directions, leading to laser-induced demagnetization.

II. EXPERIMENT

The experiments presented are performed using the Oxford Spectromag, which sustains magnetic fields up to 10 T. The Spectromag is a split-coil magnet with four optical windows for optical access and can be operated in both Voigt and Faraday geometry, where the direction of light propagation is perpendicular and parallel to the magnetic field vector. The sample is held in He vapor at 3.8 K inside the Oxford Spectromag magneto-optical cryostat. A schematic of the experimental setup is shown in Fig. 1. The magnetic field was applied in three different directions with respect to the sample surface: in plane along the [100] direction [Fig. 1(b)], at 45° or along the [101] direction [Fig. 1(c)], and out of plane or along the [001] direction [Fig. 1(d)]. Two adjacent windows of the cryostat are utilized as points of entry for the pump and probe pulses overlapping on the sample for the various orientations.

The laser system used in this experiment is the Apollo-Y optical parametric amplifier (Ultrafast systems Inc.) pumped by an ytterbium fiber amplifier with 100-kHz repetition rate providing a temporal pulse width of <300 fs at 800 nm. The output of the laser amplifier is split into four beams by the multidimensional optical nonlinear spectrometer (MONSTR), where only two are used to perform measurements. One horizontally polarized beam is used to pump the sample in a pump-probe experimental configuration, whereas a smaller vertically polarized beam probes the magnetization via the magneto-optical Kerr effect. The pump beam is focused down to 300- μ m diameter and 3 mW of average power (≈ 85 μ J/cm²), whereas the probe beam is focused down to 100- μ m diameter and 100 μ W (≈ 38 μ J/cm²). The laser beam configuration is shown in Fig. 1(e). High extinction polarizers are used in the probe beam path to detect small polarization rotations that are generated by the magnetization of the sample. In the absence of perturbation by the pump beam, the reflected probe beam is rotated by 45° using a half-wave plate and sent to a Wollstone prism, which splits the beam equally into its vertical (V) and horizontal (H) components. The two equal intensity beams are aligned to a balance detector.

The pump beam generates a change in the sample's magnetization, which could be due to different mechanisms partially depending on the sample's properties. The changes in magnetization lead to a rotation of the probe polarization $\Delta\theta_K$ via the magneto-optic Kerr effect. This in turn leads to a detectable imbalance of the H and V components split by the Wollstone prism. The pump beam is modulated by an optical chopper and induced changes in the probe beam are obtained via lockin detection.

III. RESULTS AND DISCUSSION

We started the experiments with magnetic fields along the [100] direction at a temperature of 3.8 K. We perform time-resolved MOKE at different magnetic fields, cutting across various magnetic phases. The schematic of the phase diagram

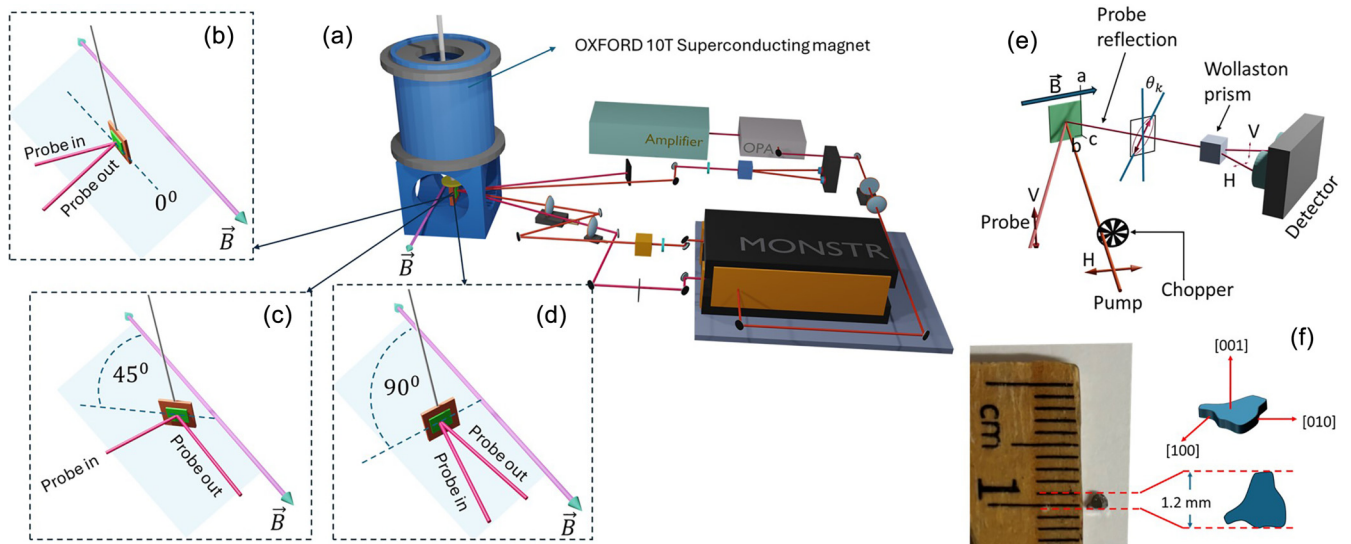


FIG. 1. (a) Schematic of the experimental setup. The laser source was provided by an amplified fiber laser with 100-kHz repetition rate and pulse duration <300 fs. The center wavelength is 800 nm with an average power of 3 mW ($\approx 85 \mu\text{J}/\text{cm}^2$) for the pump and $100 \mu\text{W}$ ($\approx 38 \mu\text{J}/\text{cm}^2$) for the probe. The laser light is split into a pump and probe beam by the multidimensional nonlinear optical spectrometer (MONSTR) instrument [70,71]. The beams are focused onto the sample surface by 40- and 50-cm focal length lenses for the pump and probe, respectively. The diameter of the pump beam on the sample is set three times larger than the probe beam. The sample is held in He vapor at 3.8 K inside the Oxford Spectromag magneto-optical cryostat. The magnetic field was applied in three different directions with respect to the sample surface: (b) in plane along the [100] direction, (c) at 45° or along the [101] direction, and (d) out of plane or along the [001] direction. The magnetic field ranges from zero up to 10 T. (e) The linearly polarized pump and probe beams are overlapped on the sample surface. The specular reflection of the probe beam is separated into the "s" and "p" components by a Wollaston prism, which are recorded by a balance detector. The magnetization of the sample leads to a rotation of the probe polarization $\Delta\theta_K$ via the magneto-optical Kerr effect, which in turn is recorded as a signal imbalance by the detector. By scanning the time delay between the pump and probe beams, the time evolution of the magnetization changes is recorded. (f) Picture of the crystal, schematic of the crystal shapes, and its orientations. The crystal orientation was determined using XRD measurements, which was oriented accordingly during the MOKE measurements. The XRD data were taken using an x-ray diffractometer Bruker D8 Focus. We collected Θ - 2Θ scans for varying angles to determine the orientation.

of MnSi is shown in Fig. 2(a), where the temperature of 3.8 K is marked by the vertical blue line and the red arrows mark the magnetic fields used for the measurements presented in Figs. 2 and 3. The time-resolved MOKE data at zero field in Fig. 2(b) show an increasing positive rotation, indicating an increase of the net magnetic moment, which peaks around ≈ 30 ps and decays over hundreds of picoseconds. This behavior has been observed in chiral magnets, including $\text{Cr}_{1/3}\text{NbS}_2$ in the ground state, and was attributed to light-induced magnetization via magnon-phonon coupling [25,65]. At a small magnetic field of 0.05 T, oscillations of the MOKE signal are observed for both polarities of the magnetic field, which is shown in Figs. 2(c) and 2(d) for +0.05 and -0.05 T, respectively. The oscillations are strongly damped with a frequency of ≈ 24 GHz corresponding to an energy of ≈ 0.1 meV.

We increase the magnetic field and perform measurements at 0.3 and 0.4 T to gain insight into the conical phase boundary. The time-resolved MOKE data are shown in Figs. 2(e) and 2(f) for 0.3 and 0.4 T, respectively. Similar oscillations are observed in the conical phase just as in the helical phase at lower magnetic fields with a frequency of ≈ 24 GHz. The frequency of the oscillations and lack of any appreciable magnetic field dependence suggest coupling to magnons [72]. Such oscillations have been previously observed in $\text{Fe}_{0.8}\text{Co}_{0.2}\text{Si}$, $\text{Fe}_{0.75}\text{Co}_{0.25}\text{Si}$, GaV_4S_8 , and MnSi skyrmion magnets using time-resolved MOKE, and have been attributed

to helimagnons and skyrmion lattice modes [61,65,73,74]. The 180° phase shift of the oscillations between 0.3- and 0.4-T data is likely due to the magnon modes belonging to two different clockwise and counterclockwise rotating $+Q$ and $-Q$ branches shown by the calculated blue curves in Fig. 4. At higher magnetic fields starting at 0.6 T, the MnSi crystal is in the field-polarized phase, and is expected to behave like a weak ferromagnet. Thus, the net magnetization could experience a torque and precession around the effective magnetic field, described by the Landau-Lifshitz-Gilbert (LLG) equation [75]. The frequency of the oscillations changes with the alteration of the magnetic field according to the Kittel formula [76,77]. This is reflected in the data shown in Fig. 3, where the time-resolved MOKE data demonstrate an increase of the oscillation frequency with increasing magnetic fields. We find close agreement with the LLG equation and Kittel formula with MnSi being in a ferromagnetic field-polarized state. In the helical and conical phases, we observe a coupling to a magnon mode which leads to oscillations of ≈ 24 GHz, remaining constant with respect to magnetic fields within our measurement certainty.

We start our discussion with the experimental observation of the helimagnons with magnetic fields aligned along the [100] direction. In order to analyze the magnetization dynamics, we use a simple phenomenological function that separates the oscillatory and nonoscillatory components seen in the data

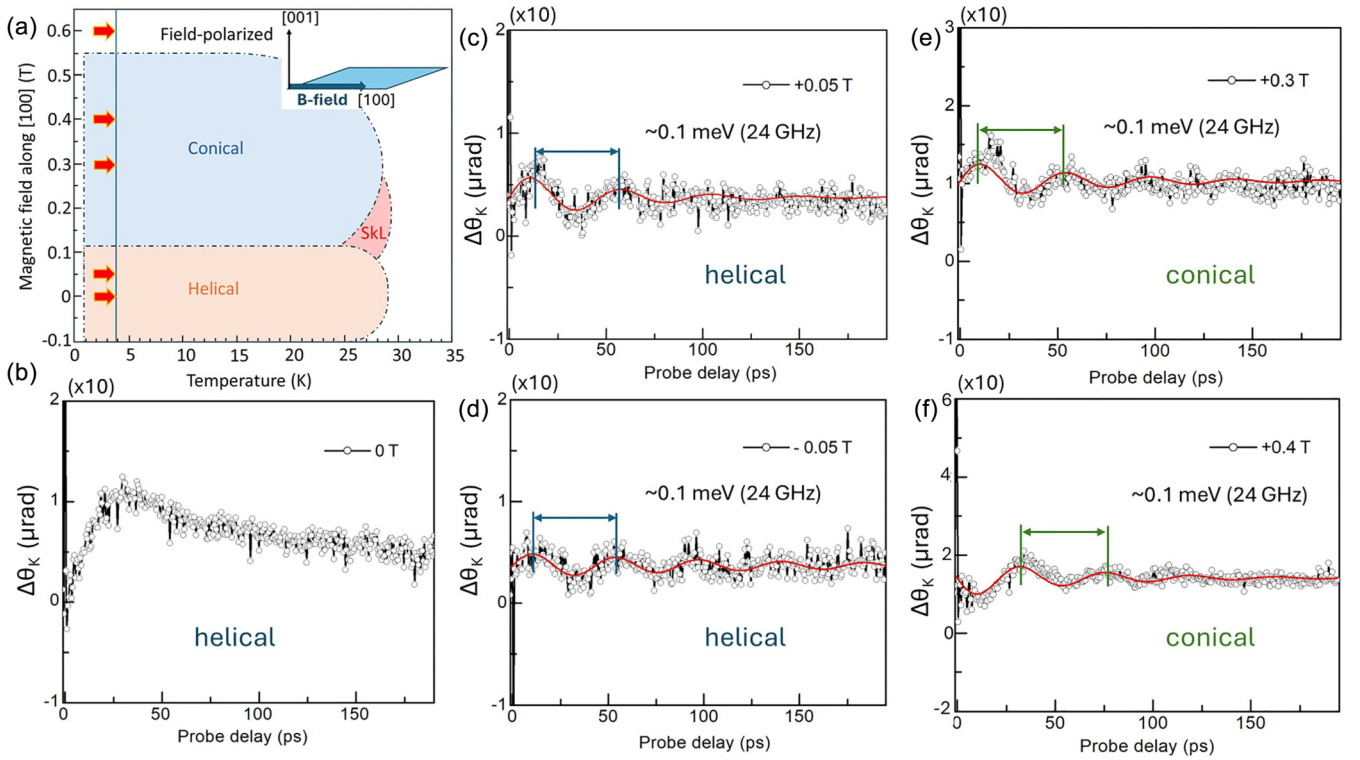


FIG. 2. (a) Phase diagram of MnSi for external magnetic field along the [100] direction following previously published work [39,41,44]. The vertical blue line marks the 3.8-K temperature where we perform our measurements. The red arrows indicate the magnetic fields at which the data were taken. The inset schematically shows the direction of the external magnetic field with respect to the sample plane. (b) Time-resolved MOKE data in the absence of magnetic fields. The change in magnetization caused by the pump pulse is recorded as a function of the delay between pump and probe pulses. (c), (d) Time-resolved MOKE data at magnetic fields of 0.05 T applied along the [100] and $\bar{1}00$ directions. At this magnetic field, the crystal is thought to be in the helical phase. An identical oscillation of ≈ 24 GHz (corresponding to ≈ 0.1 meV) is observed for both magnetic field directions. (e), (f) Time-resolved MOKE data at magnetic fields of 0.3 and 0.4 T applied along the [100] direction. At this magnetic field, the crystal is thought to be in the conical phase. An identical oscillation of ≈ 24 GHz (corresponding to ≈ 0.1 meV) is observed for both magnetic field directions (the $\bar{1}00$ direction is not shown here).

[61]. The key component of fitting our data is a decaying sinusoidal oscillation:

$$\Delta M = A + B e^{-(t/\tau)} \sin \omega t. \quad (1)$$

The ability to resolve helimagnons with femtosecond time resolution at $q = 0$ is unique to optical probes, specifically to the time-resolved MOKE. This technique complements neutron scattering, which is restricted to mapping helimagnon bands at higher q and reaches frequencies that are not achievable with conventional ferromagnetic resonance spectroscopy. This region of reciprocal space is particularly interesting in the case of helimagnets, as the periodicity introduced by the helical spin texture generates bands that are centered at $q = \pm k_h$, and therefore possess finite frequency modes at $q = 0$ even in the absence of a gap. This is in contrast to magnons in ordinary magnets, where the bands are generally centered at $q = 0$ so that the associated mode has zero frequency [78].

In order to quantitatively test the helimagnon interpretation, we take the expression for the $q = 0$ helimagnon frequency in an external magnetic field:

$$f_0 = g \mu_B H_c \sqrt{1 - \frac{1}{2} \left(\frac{H}{H_c} \right)^2} \quad (2)$$

where g is the effective electron g factor, μ_B is the Bohr magneton, and H_c is the critical field at which the spins all align with the field and the helimagnon ceases to exist as a well-defined mode. This formula expresses the magnon frequency as a function of the applied external magnetic field and predicts a weak dependency on the field [79,80].

Upon optical excitation, the magnetic moment is perturbed and deflected from its equilibrium direction. The magnetization $\mathbf{M}$ experiences a torque and precesses around the effective magnetic field $\mathbf{H}_{\text{eff}}$, following the LLG equation as given by

$$\frac{\partial \mathbf{M}}{\partial t} = \gamma \mathbf{M} \times \mathbf{H}_{\text{eff}} - \frac{\alpha}{M} \left(\mathbf{M} \times \frac{\partial \mathbf{M}}{\partial t} \right) \quad (3)$$

where α is the phenomenological damping constant, $\gamma = 2.8\pi g$ MHz/Oe is the gyromagnetic ratio of the electron, and g is the Landau splitting factor [75]. The first term describes the magnetization precession and the second term, also known as Gilbert's term, leads to damping. At low temperature and at magnetic fields above 5500 Oe, the MnSi crystal is in the field-polarized phase. In the field-polarized phase, where the MnSi crystal is thought to be a weak ferromagnet, the precession frequency as a function of the in-plane mag-

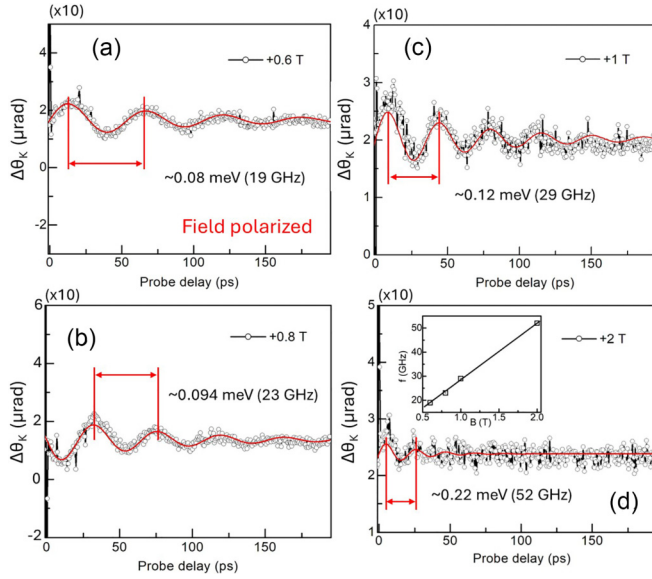


FIG. 3. Time-resolved MOKE data in the field-polarized phase at magnetic fields of 0.6 T (a), 0.8 T (b), 1 T (c), and 2 T (d) applied along the $[100]$ direction (the $[\bar{1}00]$ direction is not shown here). In the field-polarized phase, the frequency of the oscillations changes with the magnetic field following the Kittel formula, which can be approximated by a linear relationship at higher magnetic fields; the data are shown in the inset of panel (d).

netic field can be described by the Kittel equation:

$$f = \frac{\gamma}{2\pi} \sqrt{H(H + 4\pi M_s)} \quad (4)$$

where H is the magnetic field and M_s is saturation magnetization [76,77].

The theoretical model used for Fig. 4 [56,57,59,81,82] is a continuum theory which includes the exchange energy, the Dzyaloshinskii-Moriya interaction (DMI), and dipolar interactions. The ground-state configuration is determined by

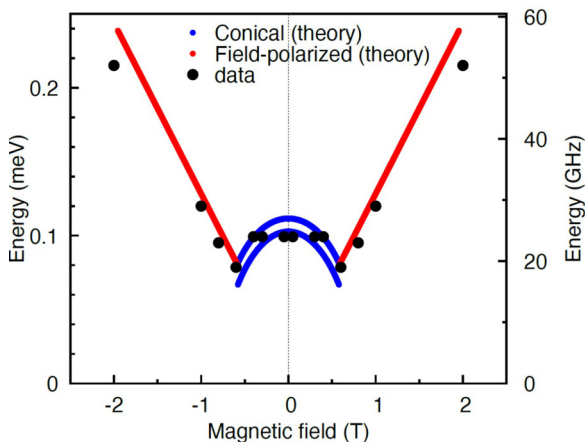


FIG. 4. Frequency of the observed oscillations as a function of the applied magnetic field along $[100]$ and $[\bar{1}00]$ directions. In the field-polarized phase, the linear relationship between the oscillation frequency and the magnitude of the external applied magnetic field is observed for both directions of the magnetic field. The solid lines correspond to the linear spin-wave model presented in Refs. [56,57,60,81].

minimizing the sum of these terms. For low field magnitudes, the helical and conical ground states are stabilized by the interplay between exchange interaction and DMI, where the pitch of the helix is given by half the ratio of their respective constants. The dynamics is determined by solving the Landau-Lifshitz equation. The calculated dispersion relations are fully determined by the interaction constants of these terms, i.e. no fitting was used for Fig. 4. Finally, we plot the oscillation frequencies as a function of the applied magnetic field for both polarities in Fig. 4. The observed oscillation frequencies in the field-polarized phase are symmetrical for both polarities of the external magnetic field. The measured experimental frequencies are shown together with the calculated dispersion, which is plotted as red lines attained from the field-polarized model presented in Ref. [81], corresponding to the Kittel equation (4) for the case of no momentum transfer. In the helical and conical phases, the observed oscillation frequency remains roughly constant and is well reproduced by the theoretical dispersion given in Ref. [81], which is displayed as blue curves in Fig. 4.

We proceed with our investigation of the magnetization dynamics by applying magnetic fields along the $[101]$ and $[001]$ directions and start our discussion with the data obtained with external magnetic fields oriented 45° with respect to the sample surface along the $[101]$ direction. The data are shown in Fig. 5, where in the absence of magnetic fields (0 T) and lower magnetic fields (0.05 T) a net increase of the net magnetization is observed, whereas for higher magnetic fields [0.3–4 T, Figs. 5(b)–5(f)] a decrease of the net magnetization is observed. This behavior is most pronounced when the external magnetic field is applied in the $[001]$ direction. In the absence of magnetic field (0 T), the laser pulse leads to an increase in net magnetization, whereas at a very small field of 0.05 T, a reduction of the magnetization is observed. This reduction in net magnetic moment persists and increases at higher magnetic fields, saturating at around 1 T. The time-resolved MOKE data are shown in Fig. 6. This observation indicates that in MnSi crystals, very small magnetic fields in the $[001]$ direction are needed to manipulate magnetization at ultrafast timescales.

We continue our discussion with the observations of light-induced magnetization and demagnetization effects. Below T_c and for applied fields > 0.1 T an applied magnetic field unpins the helical order and aligns its wave vector parallel to the field. This leads to a conical phase with a net magnetic moment. At these magnetic fields, the light pulse randomizes the spin structure, having a demagnetizing effect. In the absence of external magnetic fields or at low fields below 0.1 T, the spins are thought to be purely helical without significant net magnetic moment. Thus, the laser pulse can lead to a conical tilt of the spin structure [64]. Although linearly polarized laser pulses do not carry any angular momentum, they can generate magnetic fields through the inverse Faraday effect (IFE) [13], through the nonlinear coupling of electric and magnetic fields of light [12,14], or via the impulsive stimulated Raman scattering using short laser pulses [15]. In order to get an insight on the light-induced magnetization dynamics effects we perform first-principles calculations of the IFE in MnSi. IFE is a nonlinear optomagnetic effect in which magnetization $\delta \mathbf{O} \propto \mathbf{E} \times \mathbf{E}^*$ is induced as a second-order response to the

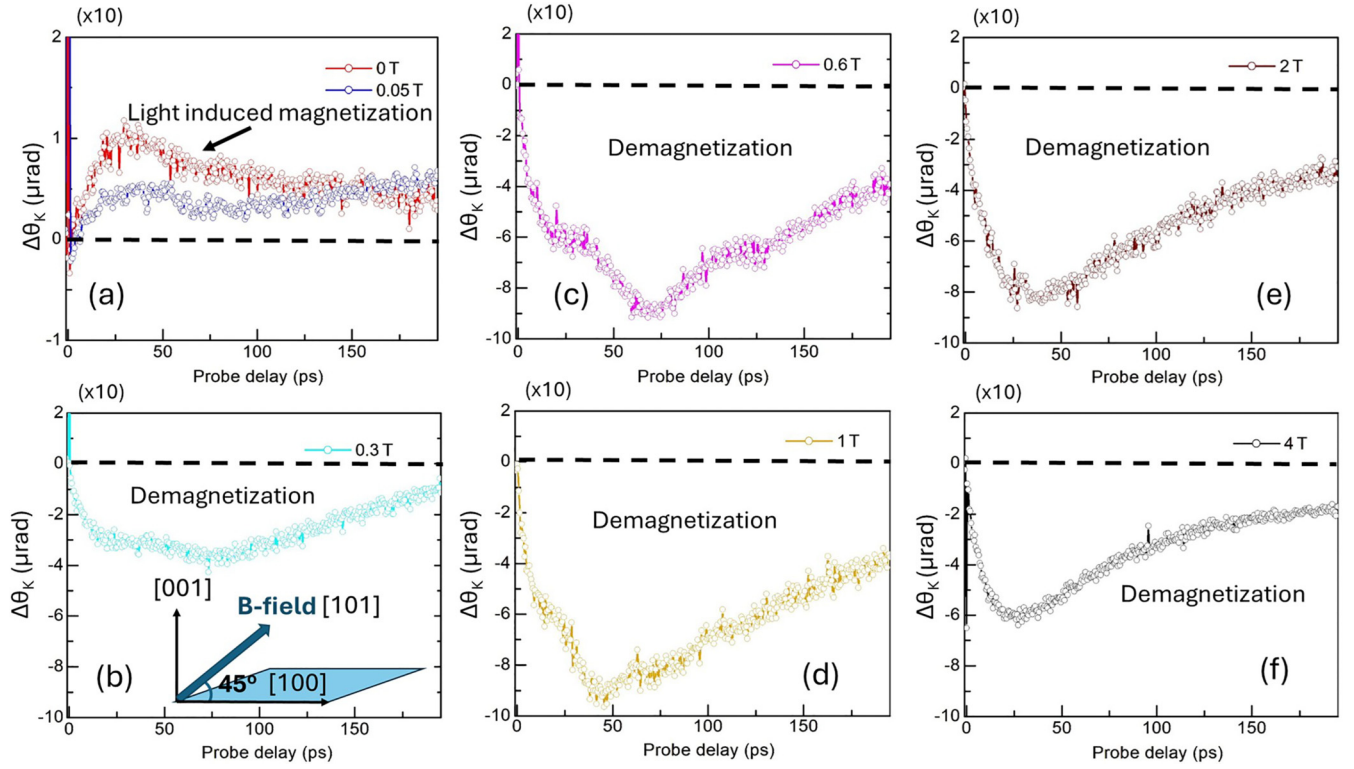


FIG. 5. Time-resolved MOKE data with external magnetic fields applied along the [101] direction. The inset in panel (b) shows the direction of the applied magnetic field with respect to the sample surface. (a) At 0 and 0.05 T in the helical phase, a positive signal is observed, which we attribute to a net increase of the magnetic moment caused by the pump pulse (light-induced magnetization). (b) At 0.3 T in the conical phase, a negative signal is observed, which we attribute to a net decrease of the magnetic moment caused by the pump pulse (demagnetization). (c)–(f) In the field-polarized phase 0.6, 1, 2, and 4 T, the same net decrease of the magnetic moment caused by the pump pulse is observed.

electric field $\mathbf{E}$ of a laser pulse [83–86]. We explore the angular dependence of IFE by calculating the effect for various directions of the ferromagnetic ground-state magnetization of MnSi.

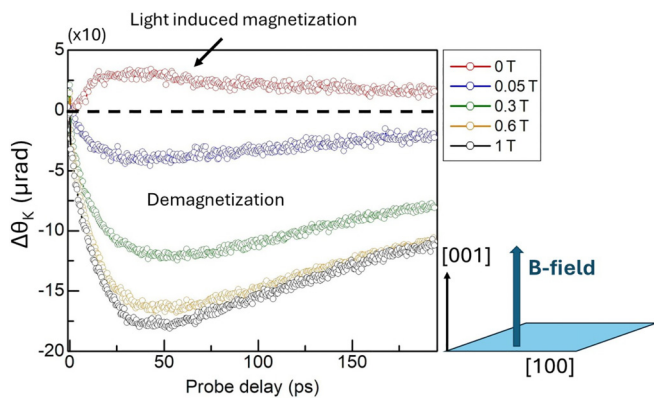


FIG. 6. Time-resolved MOKE data with external magnetic fields applied along the [001] direction. The inset on the right hand side shows the direction of the applied magnetic field with respect to the sample surface. At 0 T, a positive signal is observed in the helical phase, which we attribute to a net increase of the magnetic moment caused by the pump pulse (light-induced magnetization). At 0.05, 0.3, 0.6, and 1 T, a negative signal is observed, which we attribute to a net decrease of the magnetic moment caused by the pump pulse (demagnetization).

We calculate the first-principles electronic structures of MnSi by means of the full-potential linearized augmented plane-wave FLEUR code [87], within the local density approximation [88], and by using the VWN [89] exchange correlation functional. The parameters of our first-principles calculations, i.e. lattice constants, muffin-tin radii, plane-wave cutoffs, etc., are taken from Ref. [90]. The Mn 3s and 2p states are considered as local orbitals. An external magnetic field in the form of a simple Zeeman term equal to -0.295 eV is applied to the Mn atoms in order to constrain the spin moments to the observed value of $0.4 \mu_B/\text{Mn}$ [91]. Next, we construct maximally localized Wannier functions (MLWFs) by employing the WANNIER90 code [92] and its interface with the FLEUR code [93]. Similarly to Ref. [90], we choose an $8 \times 8 \times 8$ interpolation k mesh, and d and p orbitals for the initial projections of Mn and Si atoms, respectively, and disentangle 64 MLWFs out of 100 Bloch states within a frozen window of 3.0 eV above the Fermi energy, after excluding the 32 local orbital states and 8 Mn 4s states. Lastly, at a postprocessing step, we calculate the light-induced spin δS and orbital δL magnetic moments within the framework of the Keldysh formalism [94,95] at an $128 \times 128 \times 128$ interpolation k mesh. In all calculations a light energy $\hbar\omega = 1.55$ eV is used which corresponds to a wavelength of 800 nm, similarly to our experimental setup. Additionally, light is considered linearly polarized along the [100], [010], and [110] in-plane directions, with an intensity $I = 10 \text{ GW}/\text{cm}^2$. In our simulations disorder effects are taken into account by considering

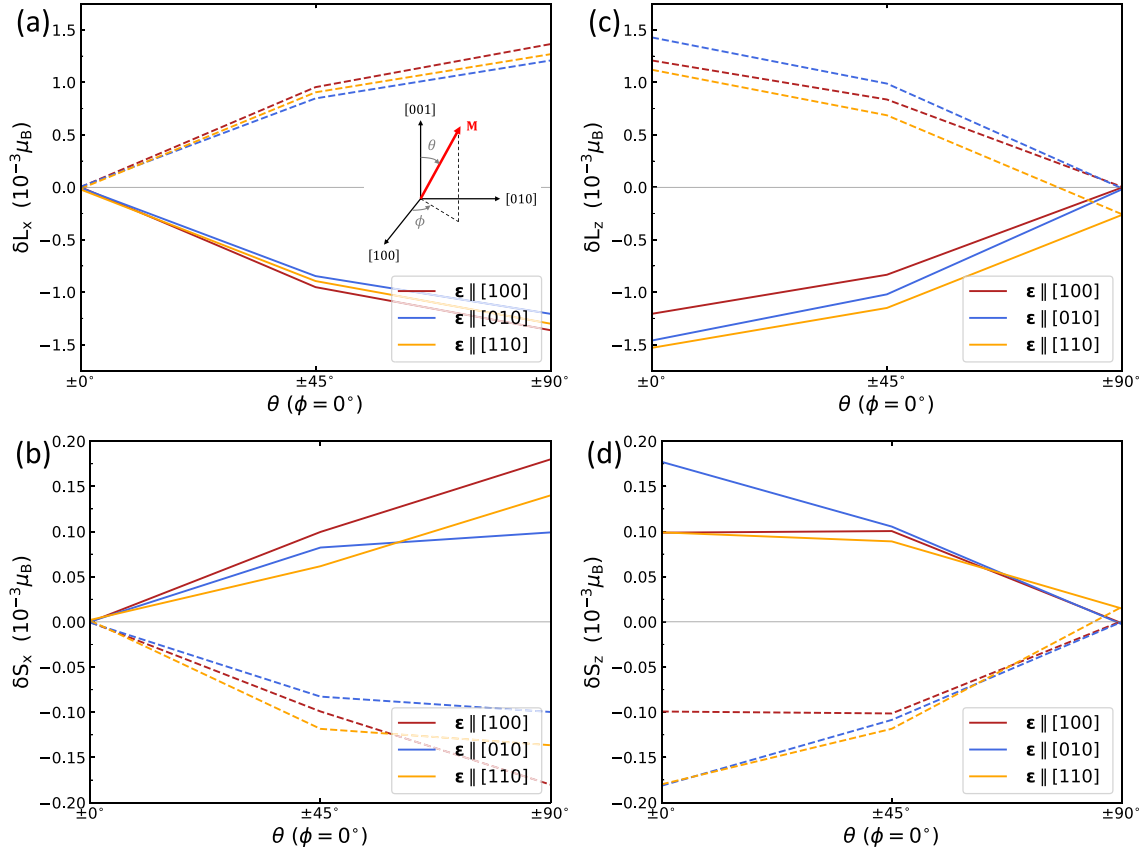


FIG. 7. Angular dependence of the inverse Faraday effect on the polar angle θ (the azimuthal angle ϕ is set to 0°) of the ferromagnetic magnetization of MnSi with respect to the [001] direction. (a), (b) The orbital δL_x and spin δS_x light-induced magnetic moments. The inset shows schematically the angles θ and ϕ . (c), (d) The orbital δL_z and spin δS_z light-induced magnetic moments. Similarly to the experimental setup, the x axis is along the [100] direction, while the z axis is along the [001] direction. Light is considered to be linearly polarized along the [100] direction (red line), the [010] direction (blue line), and the [110] direction (yellow line). The results for negative polar angle θ are depicted with dashed lines and describe the cases of opposite magnetization. The light frequency is $\hbar\omega = 1.55$ eV, the lifetime broadening $\Gamma = 25$ meV, and the calculations are performed at the true Fermi energy level.

a lifetime broadening $\Gamma = 25$ meV that corresponds to room temperature.

Since the external magnetic field is applied along the [100], [010], and [001] directions in our experiment, we study the angular dependence of IFE for MnSi with magnetization along these directions. Therefore, we explore different ferromagnetic configurations within the [101] plane by varying the polar angle θ of the magnetization from 0° to 90° , while keeping the azimuthal angle $\phi = 0^\circ$. The results of our calculations of IFE in MnSi are presented in Fig. 7. In general we observe a typical behavior of IFE in ferromagnets with δL and δS being induced along the direction of ferromagnetic magnetization, with the effect being of the same order as in previous works and odd in the magnetization [85,86]. In fact, by taking into account the small values of the ground-state spin and orbital moments, $0.4\mu_B/\text{Mn}$ and $0.01\mu_B/\text{Mn}$, respectively, IFE is remarkably more pronounced in MnSi than in other ferromagnets with larger magnetic moments. Moreover, the orbital nature of the effect is evident with δL being almost one order of magnitude larger than δS . We note that the y component of δL and δS is vanishing. We observe a similar behavior of IFE when we study in-plane magnetized configurations within

the [110] plane (not shown) by varying the azimuthal angle ϕ from 0° to 90° , while the polar angle is fixed at $\theta = 90^\circ$. In the latter scenario it is the x and y components of δL and δS that are sizable, reaching similar magnitudes as in the case with magnetization within the [101] plane, marking IFE in MnSi as a magnetic effect that depends strongly on the magnetization. It is important to highlight that IFE strongly depends on the degree of disorder as it exhibits a nonlinear behavior with respect to the lifetime broadening Γ and reaches colossal values in the clean limit [86,96]. For example, in MnSi with magnetization along [001] and for linear polarization along [110], we obtain $\delta L_z = -3.8 \times 10^{-3}\mu_B$ for $\Gamma = 10$ meV and $\delta L_z = -3.3 \times 10^{-4}\mu_B$ for $\Gamma = 100$ meV, indicating that by tuning the sample disorder, IFE may possibly vary even up to two orders of magnitude.

In our experimental geometry we are always sensitive to the [001] direction of the magnetization, therefore we are probing the IFE-induced dynamics described by δL_z and δS_z in Figs. 7(c) and 7(d), respectively. Our prediction of sizable light-induced moments by IFE along the z axis when the magnetization points along z [$\theta = 0^\circ$ in Figs. 7(c) and 7(d)] is in agreement with the experimental observations in Fig. 6 where

the external magnetic field is applied along [001]. When the external field is applied along [101], a smaller portion of the effect is measured in Fig. 5, in comparison to Fig. 6, again in agreement with our prediction when the magnetization points along the [101] direction [$\theta = 45^\circ$ in Figs. 7(c) and 7(d)]. On the other hand, when the external magnetic field is applied along [100], no magnetization dynamics are induced by IFE in the z direction and the observed oscillations in the signal are assigned to magnons, as already discussed earlier.

IV. CONCLUSIONS

We study laser-induced magnetization dynamics in the chiral magnet MnSi for different orientations of the external magnetic field with respect to the crystal axes. We obtain different responses for different orientations of the external magnetic field. When magnetic fields are applied parallel to the a - b plane, we observe damped oscillations of the magnetization as a function of time. At lower magnetic fields corresponding to the helical and conical phases, the oscillations have a very weak dependence on the magnetic fields. In the forced ferromagnetic phase, the oscillation frequencies change linearly with the magnetic field following the Kittel equation. These oscillations are attributed to magnon modes, which are impulsively excited by the first laser pulse and detected as polarization rotation of the second probe laser. The experimental data are well described by our theoretical model based on a spin-wave continuum theory which includes exchange energy, the Dzyaloshinskii-Moriya interactions, and dipolar interactions.

When external magnetic fields are applied off the a - b plane at 45° and along the c axis, the oscillations due to magnons are no longer observed. We instead observe an increase in the net magnetization at zero or low magnetic fields and demagnetization effects at higher magnetic fields. We employ first-principles calculations of the inverse Faraday effect and calculate the effect of the laser pulses on the magnetic order. These calculations confirm sizable moments δL_z and δS_z in the [001] direction generated by the laser light via the IFE. The first-principles calculations reproduce the experimental observation well, where the light-induced magnetization is most pronounced in the [001] direction and less pronounced at 45° with respect to the a - b plane. At zero or low magnetic fields in the helical phase, the crystal possesses no appreciable net magnetization, and the laser pump pulse generates net magnetization. At higher magnetic fields in the conical and forced ferromagnetic phases the pump pulse demagnetizes the crystal by disarranging the magnetic order. The ability to manipulate the magnetization by light and small magnetic fields is an important first step for many spintronic applications.

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