Optically driven collective spin excitations and magnetization dynamics in the Néel-type skyrmion host GaV₄S₈

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GaV₄S₈ is a multiferroic semiconductor hosting magnetic cycloid (Cyc) and Néel-type skyrmion lattice (SkL) phases with a broad region of thermal and magnetic stability. Here, we use time-resolved magneto-optical Kerr spectroscopy and micro-magnetic simulations to demonstrate the coherent generation of collective spin excitations in the Cyc and SkL phases driven by an optically-induced modulation of uniaxial anisotropy. Our results shed light on spin-dynamics in anisotropic materials hosting skyrmions and pave a new pathway for the optical control of their magnetic order.

The ultrafast-optical manipulation of magnetic states in quantum materials is an emerging area within condensed-matter physics, with efforts aimed at uncovering novel phases and exploring their non-equilibrium properties. Seminal examples, in this regard, include the realization of Floquet-Bloch states resulting from photon–surface-state hybridization in topological insulators and helicity-dependent control of topological-surface currents. Interest has also extended to magnetic topological defects known as skyrmions (SkS), fueled by their importance in memory technology, spintronics, and emergent electromagnetism. Recently, optical stimulus has been successfully used to write and erase individual SkS, confirm their topological robustness, and identify new metastable Sk states.

Skyrmions can be broadly classified into two varieties by their internal structure. Whirl-like Bloch-type SkS are typically found in chiral magnets and are generally stable over a relatively narrow temperature range in bulk crystals. Néel-type SkS, where spins rotate in radial planes from their cores to their peripheries, have been identified in bulk lacunar spinels, tetragonal oxides, and thin-film heterostructures. Notably, these systems all possess a polar, rather than chiral, structure and exhibit axial symmetry. Moreover, Néel-type skyrmion lattice (SkL) states in bulk crystals of these polar magnets show an enhanced thermal stability. This stems from their orientational confinement primarily due to the pattern of the Dzyaloshinskii-Moriya interaction (DMI).

The ultrafast-optical manipulation of magnetic states can generally be accomplished through mechanisms that leverage direct spin-photon coupling as well as those that exploit the thermal response of the host material. At present, however, SkL states have only been coherently excited opto-magnetically using the inverse-Faraday effect in the insulating Bloch-type–SkL-host Cu₂OSeO₃, owing to its strong linear–magneto-optical response. Another possible avenue is the optical modulation of magnetic interactions (e.g. the uniaxial anisotropy), which has proven successful in driving spin precessions in a variety of magnetic materials. Within this context, lacunar spinels, possessing large uniaxial anisotropies of the easy-axis or easy-plane varieties, represent attractive targets for the optical control of SkLS mediated by the energetic exchange between the electronic, lattice, and spin subsystems.

In this letter, we report on ultrafast time-resolved magneto-optical Kerr effect (TR-MOKE) measurements that demonstrate the generation of coherent collective excitations of the magnetic cycloid (Cyc) and SkL states in the lacunar spinel GaV₄S₈. Our results reveal GHz oscillations of the magnetization, driven by a laser-induced thermal modulation of the uniaxial anisotropy. Additionally, we observe a photo-induced enhancement of the magnetization that originates from the light-driven switching between the Cyc and ferromagnetic phases. These experiments establish an alternative route towards the optical control of the dynamic magnetic character of novel spin textures, leveraging the intimate coupling between the lattice and spin degrees of freedom.

GaV₄S₈ is a multiferroic narrow-gap semiconductor belonging to a family of lacunar spinels consisting of an FCC lattice of tetrahedral (GaS₄) and cubane (V₄S₄) clusters, the latter carrying $S = 1/2$ spins. Below $T_{JT} = 44$ K, a rhombohedral ($C_{3v}$) distortion appears, with the rhombohedral axis oriented along one of the cubic body diagonals. For $T < T_C \approx 13$ K and moderate external fields, the material is an easy-axis ferromagnet with spins oriented along the rhombohedral axis. At lower fields, a complex magnetic-phase diagram emerges consisting of Cyc and SkL ground states due to the competition between the Heisenberg-exchange inter-
action, the DMI particular to the $C_{3v}$ point-group, and the magnetocrystalline anisotropy [23, 43].

Unlike SkLs in chiral magnets, the Néel-type SkL in GaV$_4$S$_8$ is pinned to the plane perpendicular to the rhombohedral axis [23, 44]. This is primarily due to the Lifshitz-invariants that comprise the DMI term, which energetically favor magnetic modulations with $q$-vectors perpendicular to the rhombohedral axis. Due to this and the uniaxial magnetocrystalline anisotropy, the field-stability range of the Cyc and SkL phases depend on the orientation of the magnetic field with respect to the rhombohedral axis, since different domains often coexist in these crystals. As a result, several Cyc and SkL phases can be supported simultaneously, owing to the different projections of the external field along the easy axis for the four different domains. In this work, the external magnetic field was oriented perpendicular to an as-grown (001) surface of a GaV$_4$S$_8$ crystal. This ensured that all the rhombohedral domains were magnetically equivalent, thereby hosting Cyc and SkL phases over the same field ranges [23].

We employed TR-MOKE spectroscopy to probe the magnetization dynamics of the ferromagnetic, Cyc, and SkL states. The pump and probe pulses (30 fs, 1.57 eV) were modulated at 100 kHz and 1.87 kHz and focused to 50 $\mu$m and 35 $\mu$m diameters, respectively, with an on-sample pump fluence of 0.67 $\mu$J/cm$^2$. The magnetic field and sample temperature were controlled with a superconducting-magnetic cryostat equipped with a variable-temperature insert. To detect the Kerr-rotation (KR) in the reflected probe beam, we used a polarization-sensitive bridge, the differential signal from which we measured directly at the intermodulation frequencies via a phase-sensitive-detection scheme. This allowed for rapid data acquisition, avoiding the response-time issues associated with cascaded lock-in-amplifier configurations.

Figure 1(a) shows the pump-induced change to the normalized KR angle ($\Delta \theta_k$) of the probe pulse for the ferromagnetic phase in blue and the paramagnetic phase in red. As seen in Figure 1(b), the magnitude of the demagnetization step dramatically increases below $T_C$, consistent with the onset of ferromagnetic order. We found that the demagnetization occurs over $\sim 200$ ps for all external fields and sample temperatures that coincide with the ferromagnetic phase. This timescale is consistent with other semiconducting ferromagnets and can be attributed to the slow thermalization of the spin system due to its coupling to the lattice and isolation from the electronic bath [45, 46]. This is supported by the differential-reflectivity trace shown in Figure 1(c), which contains contributions from electron-electron, electron-phonon, and phonon-phonon scattering [47, 48], all of which reach quasi-equilibrium within a few picoseconds. The change in $\Delta \theta_k$ occurs on a much longer timescale, demonstrated by its relatively small variation during the first few picoseconds. Accordingly, the thermalization of the phonon bath is a nearly instantaneous event for the spins and the time scale of the demagnetization is pri-
Figure 3. (a) Time derivative of the TR-MOKE signal at $T = 10$ K for different external magnetic fields spanning the Cyc, SkL, and ferromagnetic phases and (b) the Fourier transforms of the 20 mT (blue) and 50 mT (green) traces in (a). The dashed line in (a) marks the end of the first oscillatory period. The green lines in (a) are calculated using Eqs. (2) and (3) with $\beta = 0.03$ and $\beta = 0.1$ for the 20 mT and 50 mT curves, respectively.

We now address two fundamental questions: (1) what is the underlying mechanism driving the coherent collective spin excitations and (2) why are only certain modes excited? To answer the first, we note that the presence of coherent Cyc and SkL excitations were found to be invariant to the incident pump polarization. This is typically a fingerprint of a thermal process that does not involve a direct coupling between the pump-photon field and the spin system [39]. Thermal mechanisms of this type have been explored in the study of laser-induced spin-precessions in materials such as TmFeO$_3$ [34], Co films [32], and GaMnAs [36, 37].

Being a polar semiconductor, the electron and optical phonon subsystems in GaV$_4$S$_8$ are strongly coupled, leading to a substantial increase in the lattice temperature following the pump pulse [47]. This can, in turn, lead to a modulation of the magneto-crystalline anisotropy [30]. Though such an effect typically requires a large pump fluence, this constraint is eased in GaV$_4$S$_8$ due to the strong temperature variation of its first uniaxial anisotropy constant $(K_1)$ below $T_C$ [40]. Therefore, the laser-induced heating of the sample significantly modulates the effective field acting on the magnetic system through the anisotropy contribution, driving the magnetic excitations of the SkL and Cyc states. Owing to the relatively long time required for heat to dissipate from the photo-excited volume through diffusion [47], this can be interpreted as a step-like modulation of $K_1$ within the experimental window.

To justify the above description, we used the finite-difference time-domain method to solve the Landau-Lifshitz-Gilbert equation for the SkL state using the Mumax3 code [52]. The effective field acting on the mag-
The magnetic system is given by
\[ H_{\text{eff}} = H_{\text{ext}} + H_{\text{anis}} + H_{\text{d}} + H_{\text{DMI}} + H_{\text{exch}} \]
\[ = H_{\text{ext}}\hat{e}_{\text{ext}} + \frac{2K_1}{\mu_0M_S}(\hat{e}_a \cdot \mathbf{m}) \hat{e}_a + H_{\text{d}} \]
\[ + \frac{2D}{\mu_0M_S} \left( \mathcal{L}^{(x)}_{xy} + \mathcal{L}^{(y)}_{yz} \right) + \frac{2A_{\text{ex}}}{\mu_0M_S} \nabla^2 \mathbf{m} \]

where \( A_{\text{ex}} = 0.0588 \text{ pJ/m} \) is the exchange stiffness, \( M_S = 28.8 \text{ kA/m} \) is the saturation magnetization, \( K_1 = 10 \text{ kJ/m}^3 \) is the (steady state) anisotropy constant, \( D = 0.043 \text{ mJ/m}^2 \) is the DMI constant, \( \mathcal{L}^{(i)}_{jk} = m_j \partial_i m_k - m_k \partial_i m_j \) are Lifshitz-invariants corresponding to \( C_{3v} \) symmetry, \( \hat{e}_a \) is a unit vector in the direction of the easy axis, \( \hat{e}_{\text{ext}} \) is a unit vector in the direction of the applied field, \( \mathbf{m} = \mathbf{M}/M_S \), and \( H_{\text{d}} \) is the demagnetizing field. The material parameters were estimated from literature and match the experimental periodicity of the SkL state \([23, 40]\). Here, \( \hat{x} \parallel [11\bar{1}] \), \( \hat{y} \parallel [\bar{1}0\bar{1}] \), and \( \hat{z} = \hat{e}_a \parallel [111] \). We introduced a time dependence in the effective magnetic field through a step-like decrease of \( K_1 \) by 1% of its steady state value, consistent with our estimate of the pump-induced heating of the lattice. The simulated system consisted of a \( 128 \times 64 \) grid of \( 0.8 \text{ nm}^3 \) cuboids with periodic boundary conditions along \( \hat{x} \) and \( \hat{y} \), initialized with one unit cell of a triangular Néel-type SkL with the SkL-plane normal to \( \hat{z} \). The stability of this state was established by slowly field cooling the system in the presence of an external magnetic field and the rhombohedral axis and can therefore be driven by a step-like force, in this case, representing the optical-induced modulation of the uniaxial anisotropy. This is because the modulation of \( H_{\text{eff}} \) is entirely along the \( z \)-direction (i.e. normal to the SkL plane) and can therefore only couple to the breathing mode \([54]\). As \( \alpha \) is increased, however, we see the gradual appearance of two additional peaks appearing in the \( z \)-, \( x \)-, and \( z' \)-components of the magnetization. The appearance of the new resonances is due to the the core shift characteristic of Néel-type SkLs subject to oblique external fields \([55]\). This results in a deformation of the Sk texture, reducing the six-fold rotational symmetry of the SkL to a two-fold rotation, thereby introducing a time-dependent component to the effective field in the plane of the SkL, which activates the rotational modes \([53]\). The tilting of the net magnetization and deformation of the SkL are relatively small, which accounts for the weakness of the CCW mode in our experimental results where \( \alpha = 54.7^\circ \). Further, we see that the third simulated resonance peak is relatively weak, a fact that is supported by the absence of the CW mode in our measurements. Finally, we note that the simulated resonances were blue-shifted with respect to the experimental results. This deviation is most likely due to the strong sensitivity of the mode frequencies to the values of \( D \) and \( A_{\text{ex}} \) \([53]\). Nevertheless, the order of the simulated resonances is consistent with previous ESR measurements \([39]\) as well as our experimental observations.

We now construct a phenomenological model of the experimental TR-MOKE traces. From Figure 3(b), we see that the magnetization dynamics resulting from a modulation of \( K_1 \) are comprised of decaying sinusoidal oscillations superimposed on a step-like offset. This reflects the transient reorientation of \( \mathbf{m} \) due to the reduced anisotropy following the optical pump. This type of response can be modeled by a damped harmonic oscillator driven by a step-like force, in this case, representing the optically-induced modulation of the uniaxial anisotropy. The incoherent de/remagnetization dynamics can be described by the sum of two exponentials convolved with a step-like function representing the response time of the spin-system to the lattice. For both the oscillatory and incoherent parts, we use the same step-like function.

\[
\frac{d^2 I_o}{dt^2} + 2\gamma_0 \frac{dI_o}{dt} + \omega_o^2 I_o = g(t) \cdot e^{-t/\tau},
\]

Figure 4. (a) The Fourier transforms of the simulated \( \dot{m}_z \), \( \dot{m}_x \), and \( \dot{m}_x' \) for various values of \( \alpha \) spanned by the external magnetic field and the rhombohedral axis and (b) the simulated \( m_z(t) \), \( m_x(t) \), and \( m_x'(t) \) for \( \alpha = 54.7^\circ \).
where $h(t) = e^{-t/\tau_1} - \beta e^{-t/\tau_2}$ and $g(t) = [\text{erf}(\alpha t) + 1]$. Here, $\tau_1$ and $\tau_2$ are the demagnetization and remagnetization time-constants, $\beta$ is a scaling parameter, $\alpha$ controls the spin-response time, and $\tau$ is the rate at which $K_1$ returns to the pre-time-zero value. Estimating $\tau_1 = 130$ ps, $\tau_2 = \tau = 2600$ ps, and $\alpha = 0.05$ from the experimental results, we obtain the curves plotted in green in Figure 3(a). The agreement between this model and the experimental results illustrates that the measured magnetization dynamics reflect the competition between incoherent and oscillatory signals.

In summary, we have demonstrated the ultrafast optical generation of coherent collective spin excitations of the Cyc and SkI phases in GaV$_4$S$_8$, driven by an optically-induced modulation of the uniaxial magnetocrystalline anisotropy. This indirect coupling between the optical pulse and the spin system is mediated by the lattice and represents a new mechanism by which magnetic excitations can be generated in skyrmion-host compounds with strong anisotropy. Furthermore, the peculiar nature of the magnetic ordering at the phase boundaries of GaV$_4$S$_8$ allows for a transient enhancement of the magnetization driven by the optically-induced heating of the lattice. This study underscores the intimate coupling between the spin and lattice subsystems in GaV$_4$S$_8$, and may provide a framework for the optical control of topological magnetic objects in semiconductors.

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[25] T. Kurumaji, T. Nakajima, V. Ukleev, A. Feoktystov, T.-h. Arima, K. Kakurai, and Y. Tokura, Phys. Rev. Lett. 119, 237201 (2017).
[26] S. D. Pollard, J. A. Garlow, J. Yu, Z. Wang, Y. Zhu, and H. Yang, Nat. Commun. 8, 14761 (2017).
[27] J. Zhao, A. V. Bragas, D. J. Lockwood, and R. Merlin, Phys. Rev. Lett. 93, 107203 (2004).
[28] R. Iida, T. Satoh, T. Shimura, K. Kuroda, B. A. Ivanov, Y. Tokunaga, and Y. Tokura, Phys. Rev. B 84, 064402 (2011).
[29] C. Tzschaschel, K. Otani, R. Iida, T. Shimura, H. Ueda, S. Günther, M. Fiebig, and T. Satoh, Phys. Rev. B 95, 174407 (2017), arXiv:arXiv:1702.05666.
[30] A. Kirilyuk, A. V. Kimel, and T. Rasing, Rev. Mod. Phys. 82, 2731 (2010).
[31] H. Shibata, M. Okano, and S. Watanabe, Phys. Rev. B 97, 014438 (2018).
[32] N. Ogawa, S. Seki, and Y. Tokura, Sci. Rep. 5, 9552 (2015).
[33] R. B. Versteeg, I. Vergara, S. D. Schäfer, D. Bischoff, A. Aqeel, T. T. M. Palstra, M. Grüninger, and P. H. M. van Loosdrecht, Phys. Rev. B 94, 094409 (2016).
[34] A. V. Kimel, A. Kirilyuk, A. Tsvetkov, R. V. Pisarev, and T. Rasing, Nature 429, 850 (2004).
[35] J.-Y. Bigot, M. Vomir, L. Andrade, and E. Beaurepaire, Chem. Phys. 318, 137 (2005).
[36] J. Wang, I. Cotoros, K. M. Dani, X. Liu, J. K. Furdyna, and D. S. Chemla, Phys. Rev. Lett. 98, 217401 (2007).
[37] Y. Hashimoto, S. Kobayashi, and H. Munekata, Phys. Rev. Lett. 100, 067202 (2008).
[38] H. Shibata, M. Okano, and S. Watanabe, Phys. Rev. B 97, 014438 (2018).
[39] D. Ehlers, I. Stasinopoulos, V. Tsurkan, H.-A. K. von Nidda, D. Grundler, and A. Loidl, Phys. Rev. B 94, 014406 (2016).
[40] D. Ehlers, I. Stasinopoulos, I. Kézsmárki, T. Fehér, V. Tsurkan, H.-A. K. von Nidda, D. Grundler, and A. Loidl, J. Phys. Condens. Matter 29, 065803 (2017).
[41] Z. Wang, E. Ruff, M. Schmidt, V. Tsurkan, I. Kézsmárki, P. Lunkenheimer, and A. Loidl, Phys. Rev. Lett. 115, 207601 (2015).
[42] J. Hlinka, F. Borodavka, I. Rafalovskyi, Z. Docelorova, J. Pokorny, I. Gregora, V. Tsurkan, H. Nakamura, F. Mayr, C. A. Kuntscher, A. Loidl, S. Bordacs, D. Szaller, H.-J. Lee, J. H. Lee, and I. Kézsmárki, Phys. Rev. B 94, 060104 (2016).
[43] A. Bogdanov and A. Hubert, J. Magn. Magn. Mater. 138, 255 (1994).
[44] J. S. White, A. Butykai, R. Cubitt, D. Honecker, C. D. Dewhurst, L. F. Kiss, V. Tsurkan, and S. Bordacs, Phys. Rev. B 97, 020401 (2018).
[45] E. Kojima, R. Shimano, Y. Hashimoto, S. Katsumoto, Y. Iye, and M. Kuwata-Gonokami, Phys. Rev. B 68, 193203 (2003).
[46] J. Hlinka, F. Borodavka, I. Rafalovskyi, Z. Docekalova, J. Pokorny, I. Gregora, V. Tsurkan, H. Nakamura, F. Mayr, C. A. Kuntscher, A. Loidl, S. Bordacs, D. Szaller, H.-J. Lee, J. H. Lee, and I. Kézsmárki, Phys. Rev. B 94, 065803 (2017).
[47] A. Othonos, J. Appl. Phys. 83, 1789 (1998).
[48] P. Y. Yu and M. Cardona, Fundamentals of Semiconductors, 4th ed. (Springer-Verlag, New York, 2010) pp. 292-295.
[49] C. Kittel, Phys. Rev. 110, 836 (1958).
[50] A. V. Kimel, R. V. Pisarev, J. Hohlfeld, and T. Rasing, Phys. Rev. Lett. 89, 287401 (2002).
[51] E. Ruff, S. Widmann, P. Lunkenheimer, V. Tsurkan, S. Bordacs, I. Kézsmárki, and A. Loidl, Sci. Adv. 1, e1500916 (2015).
[52] A. Vansteenkiste, J. Leliaert, M. Dvornik, M. Helsen, F. Garcia-Sanchez, and B. Van Waeyenberge, AIP Adv. 4, 107133 (2014).
[53] V. L. Zhang, C. G. Hou, K. Di, H. S. Lim, S. C. Ng, S. D. Pollard, H. Yang, and M. H. Kuok, AIP Adv. 7, 055212 (2017).
[54] M. Mochizuki, Phys. Rev. Lett. 108, 017601 (2012).
[55] A. O. Leonov and I. Kézsmárki, Phys. Rev. B 96, 214413 (2017).