Giant Effective Damping of Octupole Oscillation in an Antiferromagnetic Weyl Semimetal

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A magnetic Weyl semimetal is a recent focus of extensive research as it may exhibit large and robust transport phenomena associated with topologically protected Weyl points in momentum space. Since a magnetic texture provides a handle for the configuration of the Weyl points and its transport response, understanding of magnetic dynamics forms the basis for future control of a topological magnet. Mn$_3$Sn is an example of an antiferromagnetic Weyl semimetal that exhibits a large response comparable to the one observed in ferromagnets despite a vanishingly small magnetization. The noncollinear spin order in Mn$_3$Sn can be viewed as a ferroic order of cluster magnetic octupole and breaks the time-reversal symmetry, stabilizing Weyl points and the significantly enhanced Berry curvature near the Fermi energy. Herein, the first observation of time-resolved octupole oscillation in Mn$_3$Sn is reported. In particular, the giant effective damping of the octupole dynamics is found, and it is feasible to conduct an ultrafast switching at $<10$ ps, a hundred times faster than the case of spin-magnetization in a ferromagnet. Moreover, high domain wall velocity over 10 km s$^{-1}$ is theoretically predicted. This work paves the path toward realizing ultrafast electronic devices using the topological antiferromagnet.

1. Introduction

A Weyl semimetal is a topological system in three dimensions, which appears in either time-reversal-symmetry (TRS) or inversion-symmetry broken state.[1–4] It is characterized by gapless electronic excitations called Weyl fermions, formed as a result of a linear crossing of two non-degenerate bands at a pair of momentum points with different chiralities. As these Weyl points can be viewed as unit-size monopole of underlying Berry curvature, Weyl semimetals may exhibit various exotic phenomena such as Fermi arc, anomalous Hall and Nernst effects (AHE and ANE), chiral anomaly, and gyrotropic effects.[1–18] In particular, for TRS broken or magnetic Weyl semimetals, magnetic order determines the configuration of the Weyl points in momentum space and provides the handle for controlling the transport responses.[10,12–18] Therefore, understanding of the magnetic...
2. Spin Dynamics of an Antiferromagnet

Let us first discuss the case of collective spins in ferromagnetic materials, where each spin is coupled in parallel by exchange interaction (Supporting Information 1). A precessional motion of each spin is always in-phase as schematically shown in Figure 1c, and thus, the energy scale of the resonant frequency ($\hbar \omega_0 \approx K$) is independent of the exchange interaction and is determined by $K$ only, where $K$ is magnetic anisotropy energy mainly originating from on-site spin-orbit interaction. A typical time-scale for the magnetization switching, expressed as $(\Delta \omega)^{-1}$, can be $\approx (2\alpha K)^{-1}\hbar$. Here, $\Delta \omega$ and $\alpha$ are spectral linewidth and effective damping constant, respectively. Generally, the timescale is longer than 1 ns (e.g., $\omega = 10$ GHz and $\alpha = 0.1$).

To consider collective spins in a chiral AF metal (Supporting Information 1), the following Hamiltonian to treat the inverse triangular spin structure can be used:\[38\]

$$\mathcal{H} = J \sum_{\langle i,j \rangle} S_i \cdot S_j + D \sum_{\langle i,j \rangle} \varepsilon_{ab} z \cdot (S_i \times S_j) - \frac{K}{2} \sum_{ia} (\varepsilon_{ia} \cdot S_a)^2$$

(1)

Here, $S$, $J$, and $D$ denote spin-angular momentum, exchange interaction, and Dzyaloshinskii–Moriya interaction, respectively. $(i, j)$ and $(a, b)$ refer to the Mn sites and one of the three sublattices $(A, B,$ and $C)$ of the inverse triangular lattice structure, respectively. $K$ is introduced to describe six-fold magnetic anisotropy in Mn$_2$Sn with $k_a = (\cos \psi_a, \sin \psi_a, 0)$ and $(\psi_a, \psi_b, \psi_c) = (0, 4\pi/3, 2\pi/3)$. $\varepsilon_{ab}$ is the antisymmetric tensor which satisfies $\varepsilon_{AB} = \varepsilon_{BC} = \varepsilon_{CA} = 1$ and $z$ is the unit vector along the c-axis. Note that in-plane and out-of-plane magnetic anisotropies from the kagome plane should be determined by $K$ and $D$, respectively.

Figure 1d,e schematically show spin-wave modes for a chiral AF metal. Mode I is an in-plane $(xy)$ optical mode. Although out-of-plane precession $(z)$ is in-plane and is governed by $D$, in-plane precession is out-of-plane and is determined by $J$. Therefore, a typical energy scale $(\hbar \omega_{II})$ of the mode I can be estimated as $\approx \sqrt{KD}$. Mode II is collective precession-like mode, that is, in-plane acoustic mode. Similarly, a typical energy scale $(\hbar \omega_{II})$ of the mode II can be $\approx \sqrt{KJ}$. As we discuss in detail in the following section, although the resonant frequencies of modes I and II are different, typical timescales for the magnetization switching are identical. $(\Delta \omega_{II})^{-1} \approx (\Delta \omega_{II})^{-1} \approx (\alpha J)^{-1}\hbar$, and thus is much shorter by the factor of $K/J$ than the ferromagnetic case. Such an $\textit{exchange-enhanced}$ ultrafast damped precession should be available in both AF and ferrimagnetic metals and would be the most significant in a fully compensated case. However, such compensation of magnetic moments has been considered to make it impossible to observe the spin-dynamics in AF metals. This is because a signal amplitude is usually proportional to the net spin-magnetization in a detection method for the spin-dynamics, e.g., the Faraday effect and the MOKE. Here, we demonstrate the time-resolved ultrafast spin precession in the AF Weyl semimetal Mn$_2$Sn by using MOKE induced by the magnetic octupole order. The spin precessions from the noncollinear texture induce the oscillations of MOKE and the Berry curvature in the momentum space.

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In this article, bulk single-crystal $D_{0_{19}}$-$Mn_{3}$Sn has been used (the Experimental Section). $Mn_{3}$Sn has the hexagonal Ni$_3$Sn-type crystal structure consisting of an ABAB stacking of the kagome lattice of Mn atoms along with the [0001] axis. Red (blue) circles in Figure 1a indicate Mn atoms in the $A$-$B$ plane of the kagome lattice. Below the Néel temperature of 430 K, an inverse triangular spin structure is stabilized by exchange and Dzyaloshinskii–Moriya interactions.$^{[10,19,40]}$ The inverse triangular spin structure possesses a uniform negative vector chirality of the in-plane Mn moments and is made of a ferro ordering of cluster magnetic octupoles. The magnetic moments cant slightly in the (0001)-plane and produce a small net spontaneous magnetization, which possesses the same symmetry as the spin-magnetization (e.g., as indicated by a mirror plane [orange line]).

3. Results

3.1. Static MOKE

In this article, bulk single-crystal $D_{0_{19}}$-$Mn_{3}$Sn has been used (the Experimental Section). $Mn_{3}$Sn has the hexagonal Ni$_3$Sn-type crystal structure consisting of an ABAB stacking of the kagome lattice of Mn atoms along with the [0001] axis. Red (blue) circles in Figure 1a indicate Mn atoms in the $A$-$B$ plane of the kagome lattice. Below the Néel temperature of 430 K, an inverse triangular spin structure is stabilized by exchange and Dzyaloshinskii–Moriya interactions.$^{[10,19,40]}$ The inverse triangular spin structure possesses a uniform negative vector chirality of the in-plane Mn moments and is made of a ferro ordering of cluster magnetic octupoles. The magnetic moments cant slightly in the (0001)-plane and produce a small net spontaneous magnetization, which possesses the same symmetry as the spin-magnetization (e.g., as indicated by a mirror plane [orange line]).

The right (left) vertical axis in Figure 2 shows the polar Kerr rotation angle measured with 660 nm continuous-wave (800 nm pulse) laser system. The difference in the Kerr rotation angle obtained by the 660 nm and the 800 nm laser systems is consistent with the previous works, where the magnetic field is applied parallel to the kagome plane (B // [21 1 0]), exhibits a clear hysteresis with a large Kerr rotation angle ($\approx$60 mdeg). This is three times larger than the previous report$^{[31]}$ and the difference may come from the optical interference effect.$^{[42]}$ In addition to the polar MOKE, the longitudinal signals, where the magnetic field is applied in [0110] and [0001] directions, were also characterized (Figure 2b). Although a clear hysteresis curve was observed in [0110], no signal was confirmed in [0001]. The observed magnetic anisotropy is consistent with the previous works, where the octupole polarization in $D_{0_{19}}$-$Mn_{3}$Sn was detected via the measurements of the AHE,$^{[10]}$ ANE,$^{[11,14]}$ and MOKE.$^{[31]}$ The right (left) vertical axis in Figure 2 shows the polar Kerr rotation angle measured with 660 nm continuous-wave (800 nm pulse) laser system. The difference in the Kerr rotation angle obtained by the 660 nm and the 800 nm laser systems is consistent with the MOKE spectroscopy reported previously.$^{[31]}$

Insets show magnetization hysteresis curves measured with the same magnetic field configuration as the MOKE measurements. From Figure 2a inset, the spontaneous magnetization of $Mn_{3}$Sn is $9 \times 10^{-3}$ $\mu_B$ per formula unit (f.u.). If we assume a conventional ferromagnet
The effective model presented in the study by Nomoto and Arita\cite{31} well reproduces the results for the domain wall dynamics and for the coherent steady precession of spins obtained by using the original spin Hamiltonian (Equation (1)).

(e.g., Fe, Co, and Ni), a possible Kerr rotation angle at zero magnetic field from the spontaneous magnetization would be as small as 0.2 mdeg with positive polarity.\cite{31} Therefore, a large negative Kerr rotation angle at zero magnetic field can be hardly explained by the spin-magnetization due to canting. The octupole has the same irreducible representation of $T_{2g}$ as the spin-magnetization, and can induce the MOKE. The results of multipole expansion of the AF structure of Mn$_3$Sn show that the octupole contributes to the expansion by more than 99.9%\cite{32}. The previous first-principles calculation finds that the Kerr rotation angle has nearly no contribution from the net magnetization.\cite{31} Moreover, a method to obtain a low-energy effective model of Mn$_3$Sn has been recently developed based on the cluster multipole theory.\cite{38} The effective model presented in the study by Nomoto and Arita\cite{31} well reproduces the results for the domain wall dynamics and for the coherent steady precession of spins obtained by using the original spin Hamiltonian (Equation (1)).

### 3.2. Time-Resolved MOKE

Our measurement of the time-resolved MOKE (TR-MOKE) was made by using an all-optical pump–probe method at room temperature (Figure 3a and the Experimental Section). This method has been conventionally used to detect spin precession for ferromagnetic metals via Kerr effect and AF insulators\cite{35-37} via the Faraday effect. For an AF metal, however, no report on time-resolved spin precession has been made to date. Pump and probe lights were configured almost perpendicular to the Mn$_3$Sn (2110)-surface. In addition, an external magnetic field normal to the surface ($// (2110)$) was applied during the measurements to direct the octupole polarization.

Figure 3b shows typical TR–MOKE results, where a magnetic field of 2T was applied normal to the surface. First, a pump light induces a rapid increase in the Kerr rotation angle of the probe light. Because Mn$_3$Sn is metallic, a pump pulse rapidly increases the electron temperature of the system. This rapid increase in electron temperature causes a significant decrease in the size of the order parameter, namely, the cluster magnetic octupole, similar to the case in ultrafast demagnetization in ferromagnetic metals.\cite{34,38} After the excitation, a coherent spin precession starts with the aid of an effective magnetic field. As the cluster magnetic octupole is the order parameter that induces the MOKE signal as we discussed earlier, the spin precession is observed as the fluctuation of the cluster magnetic octupole. In Figure 3b, the MOKE intensity starts to recover with delay time more than 0.3 ps, exhibiting a small but clear oscillation during the recovery. To further characterize the oscillating component, a nonoscillating component was estimated as a background (black solid curve) and subtracted from the raw data (Supporting Information 2). The lower panel of Figure 3b shows the TR–MOKE signals after subtracting the background. Figure 3c also shows typical TR–MOKE results (upper panel) and the analysis for the oscillating component (lower panel) in a relatively long-time range. The orange (blue) thick curve in the lower panel of Figure 3b (3c) corresponds to the fitting to the equation, $\cos(\omega_0 t + \phi_0) \exp(-\alpha_1 I(t))$, which yields both the resonant frequency and effective damping constant at $B = 2$ T, i.e., $\omega_0 = 0.86$ THz (18 GHz) and $\alpha_1 = 0.02$ (1.0), respectively. The orange (blue) thin curves represent the envelope functions of the fit, $\pm \exp(-\alpha_1 I(t))$. Here, $t$ is the delay time. The oscillation frequencies ($\omega_0, \omega_I$) as a function of the external magnetic field are shown in Figure 3d, which will be discussed later in detail. The oscillating behavior is most significant when the magnetic field is normal to the Mn$_3$Sn surface. Because the TR–MOKE signal is proportional to the surface normal polarization of the cluster magnetic octupoles, the oscillating Kerr signal should come from a change of size in the octupole order parameter. The change of size in the octupole order parameter originates from the spin precession of each magnetic moment. Specifically for the mode I, the signal-to-noise ratio of the TR–MOKE significantly reduces with decreasing the magnetic field strength below 2 T. This is because an external magnetic field is necessary to create an effective magnetic field to drive a coherent precession (Supporting Information 3).

### 4. Discussion

Two oscillation modes I and II are found through the analyses shown in Figure 3b,c, which should come from the oscillation of the cluster magnetic octupoles (Supporting Information 2). To analyze the modes, we have derived the analytical solution

![Figure 2: Static hysteresis loops of the MOKE. a) Magnetic field dependence of the polar MOKE result. b) Magnetic field dependence of the longitudinal MOKE results. Left (right) vertical axis shows Kerr rotation angle (mdeg) as a function of the magnetic field ($B$). Insets show the corresponding magnetization hysteresis ($M$ in $10^{-3}$ μM per f.u.) obtained as a function of the magnetic field with the same configuration as the MOKE in the main panel. MOKE signal does not come from the spin-magnetization due to canting but from cluster magnetic octupole as discussed in the main text and in the study by Higo et al.\cite{31}](image-url)
to estimate the resonant frequencies from Equation (1). Note that Equation (1) includes the in-plane exchange interaction $J$ in the kagome lattice, but not the interplane exchange interaction, and thus is only valid for the modes where precessional motions in different kagome planes in-phase. The equation of motion for low energy magnetic excitation can be derived from Equation (1) as the following sine-Gordon equation (Supporting Information 4)

$$\frac{\hbar}{2\sqrt{3}(D + \sqrt{3}J)S}\frac{d^2\phi}{dt^2} + \alpha \frac{d\phi}{dt} - \frac{(\sqrt{3}D + J)S}{2\hbar} \alpha_{\text{lat}}^2 \frac{d^2\phi}{dt^2} + \frac{KS}{2\hbar} \sin 2\phi = 0$$

(2)

where $\hbar$, $\alpha$, and $\alpha_{\text{lat}}$ indicate the reduced Planck constant, damping constant, and the lattice constant of the nearest neighbor Mn atoms, respectively. Here, $S$ is the size of spin angular momentum $\approx 1.5$ for the Mn magnetic moment $\approx 3\mu_B$, $\phi$ refers to in-plane precession angle of Mn magnetic moment, and $\alpha$ is identical to the Gilbert damping constant in the Landau–Lifshitz–Gilbert equation. From Equation (2), resonant frequencies for modes I and II can be estimated as

$$\hbar \omega_1 = S\sqrt{6\sqrt{3}(\sqrt{3}D + J)D}$$

(3)

$$\hbar \omega_2 = S\sqrt{2\sqrt{3}K(D + \sqrt{3}J)}$$

(4)

Here, the resonant frequencies for the modes I and II ($\omega_1$ and $\omega_2$) correspond to the optical and collective precession-like modes shown in Figure 1d,e, respectively. The damped oscillation is often expressed by $\exp(-\alpha_{\text{lat}} t)\exp(i\omega t)$ by introducing a phenomenological effective damping constant ($\alpha_{\text{lat}}$, $\alpha$), which is expressed as

$$\hbar \omega_{1/2} = \hbar \omega_2 = 2\sqrt{3}S(D + \sqrt{3}J)\alpha$$

(5)

Note that the effective damping ($\alpha_{\text{lat}}$, $\alpha$) is not identical to the Gilbert damping constant ($\alpha$) defined in Equation (2). The Gilbert damping constant should be determined from the damping rate (Equation (5)), which is independent of the resonant frequency ($\omega_1$, $\omega_2$). Although these two damping constants are almost identical for the case of a ferromagnet, a large difference between $\alpha_{\text{lat}}$ and $\alpha$ induces large deviation of the effective damping constant from the Gilbert damping constant in Mn$_3$Sn. This is a unique property of the octupole oscillation dynamics in a chiral AF metal and should be distinguished from spin-magnetization dynamics in a ferromagnet. Recently, related discussion for the damping has been made in the magnetic domain-wall dynamics in the ferrimagnetic GeFeCo. Here, the field dependence of the resonant frequencies (Figure 3d) is understood as follows. The resonant frequency of the optical mode ($\omega_1$) is determined by interactions between Mn atoms, and thus is insensitive to an external magnetic field. However, the resonant frequency of the collective precession-like mode ($\omega_2$) is not. This is because the cluster magnetic octupole can couple with an external magnetic field via spontaneous magnetization due to canting. From the field dependence of the resonant frequencies (Figure 3d), $\omega_{2/2\pi}$ at $B = 0$ is determined to be $13.7 \pm 1.5$ GHz.
To characterize the dynamics of the cluster magnetic octupole, the six-fold magnetic anisotropy $K$ was determined from the torque measurements (Supporting Information 5). Figure 4a shows the result of out-of-plane rotation ($\gamma$-axis) in terms of the plane consisting of the kagome lattice in Mn$_3$Sn ($xy$-plane) by rotating the direction of the magnetic field (B) from $[2\bar{1}10]$ ($\phi_{B1} = 0^\circ$) to [0001] ($\phi_{B1} = 90^\circ$). The solid and open circles indicate different rotation directions. The results allow the estimation of the saturation magnetization ($M_s$), as shown in the inset of Figure 4a. The saturation magnetization should correspond to the spontaneous magnetization due to spin canting at zero field, determined by $D$ and $J$, and should be distinguished from the magnetization components due to the spin canting induced by the application of an external magnetic field. Here, the saturation magnetization is estimated to be $11.3 \times 10^{-3} \mu_B$ ($10.0 \times 10^{-3} \mu_B$) per f.u. at $B = 0$ T ($9$ T), and is comparable to the magnetization at $B = 0$ ($9 \times 10^{-3} \mu_B$ per f.u. from Figure 2a inset). In contrast, the in-plane rotation ($\zeta$-axis) measurements of the torque were made by rotating the direction of the magnetic field (B) from $[2\bar{1}10]$ ($\phi_{B2} = 0^\circ$) to [0110] ($\phi_{B2} = 90^\circ$) within the kagome-lattice plane (Figure 4b). The analysis of the result estimates an energy barrier height from the six-fold in-plane magnetic anisotropy energy ($K_{\alpha}/18$, Supporting Information 5) to be $3.1 \times 10^{-17}$ J m$^{-3}$, consistent with the value reported in the previous work ($2.2 \times 10^{-17}$ J m$^{-3}$). Here, the obtained in-plane magnetic anisotropy energy corresponds to $K = 3.1 \times 10^{-17}$ meV from Equation (I).

In principle, six spin-wave modes should be confirmed in Mn$_3$Sn as the unit cell consists of six Mn atoms. Previous neutron scattering studies$^{[47,48]}$ have revealed the three modes in the low energy region ($\hbar \omega < 20$ meV) and the other three modes in the high energy region ($\hbar \omega \approx 100$ meV) at $q = 0$, where $q$ is the momentum vector. The low and high energy modes are the ones where precessional motions in different kagome-planes are in-phase and out-of-phase, respectively. Here, both the modes I and II in this article correspond to the low energy modes. From TR-MOKE results (Figure 3b), the energy of the mode I is $3.6$ meV (0.86 THz). However, this is only 26% of the value obtained in the neutron scattering ($\approx 14$ meV).$^{[47]}$ This strongly suggests that the size of the magnetic moment (see $S$ in Equation (3)) is reduced. This might be because laser heated the sample close to its Néel temperature ($\approx 430$ K),$^{[39]}$ which is not so far from room temperature, during the TR-MOKE measurements. In fact, it has been reported that the AHE and MOKE are significantly suppressed by heating Mn$_3$Sn.$^{[49]}$ When $S \approx 0.4$ ($= 1.5 \times 26\%$) is used in Equation (3) and (4), $J$ and $D$ are estimated to be $\approx 10$ and $\approx 0.7$ meV, respectively, which is in good agreement with the previous studies.$^{[47,48,50]}$ Gilbert damping constant of Mn$_3$Sn, which is estimated for the first time, is found to be $\alpha = 0.0005$ from Equation (5). The Gilbert damping constant of Mn$_3$Sn is as small as the theoretical prediction for Mn–Ge$^{[51]}$ and Mn–Ga$^{[52]}$ alloys (0.0005–0.001).

As discussed, the dynamics characteristic of the cluster magnetic octupoles is an ultrafast damped oscillation due to the exchange interaction (Equation (5)). Interestingly, the exchange interaction dramatically increases the effective damping ($\alpha(t_1 = 1.0)$. Thus, the typical switching time can be ultrafast ($1/\omega_{a1} \approx 1/\omega_{a1}t_1 \approx 9$ ps) although the resonant frequency for the collective precession-like motion is relatively slow ($\omega_{a1} = 13$ GHz). As discussed, $S \approx 0.4$ was used for the aforementioned analysis. If the sample temperature can be held much lower than the Néel temperature ($S = 1.5$), the switching time would be reduced to $\approx 2$ ps. From the sine-Gordon equation (Equation (2)), the Néel type-domain wall velocity in Mn$_3$Sn can be estimated as fast as $3$ km s$^{-1}$ ($12$ km s$^{-1}$) for $S = 0.4$ (1.5) (Supporting Information 6), which is greater than the recently reported value for the ferrimagnetic systems in the vicinity of the angular momentum compensation temperature.$^{[53–55]}$

For device applications, not only dynamic but also static properties of the cluster magnetic octupoles would be important. As discussed, energy barrier height ($K_{\alpha}/18$) for the six-fold magnetic anisotropy energy of Mn$_3$Sn is $3.1 \times 10^{-17}$ J m$^{-3}$, which corresponds to the in-plane magnetic anisotropy field of $3.8$ T ($=K_{\alpha}/M_s$). If we employ $10$ nm thick Mn$_3$Sn, the areal magnetic anisotropy energy should correspond to $3 \mu$J m$^{-2}$. To obtain a thermal stability factor of $60$ at $300$ K, which is defined as magnetic anisotropy energy divided by thermal fluctuation energy ($k_B T$), relatively large magnetic cell as $320$ nm in diameter ($8 \times 10^5$ nm$^3$) is needed. However, such small magnetic anisotropy energy ensures that the octupole polarization can

![Figure 4. Magnetic anisotropy. a) Results of the torque measurements for the out-of-plane magnetic field rotation, namely, upon changing the direction of the magnetic field B from [2\bar{1}10] ($\phi_{B1} = 0^\circ$) to [0001] ($\phi_{B1} = 90^\circ$). The inset shows the saturation magnetization ($M_s$) as a function of an external magnetic field B. b) Results of the torque measurements for the in-plane magnetic field rotation, namely, upon changing the direction of the magnetic field B from [2\bar{1}10] ($\phi_{B2} = 0^\circ$) to [0110] ($\phi_{B2} = 90^\circ$). Solid and open circles represent the torque obtained with increasing and decreasing rotation angles of $\phi_{B1}$ and $\phi_{B2}$.](Image 48x743 to 145x765)
be efficiently controlled by electric current- or voltage-driven torque.\textsuperscript{[56]}

5. Conclusion

To conclude, our work demonstrates the giant effective damping of the octupole dynamics in an AF Weyl semimetal. The exchange interaction significantly increases the damping rate for the collective precession-like mode, which is directly related to a typical switching time in device operation.\textsuperscript{[57]} The introduction of the key concept of cluster magnetic octupole, instead of spin-magnetization in ferromagnets, has provided the basis for carrying out the experiments by using the conventional schemes developed for ferromagnetic spintronics. Thus, our observation will certainly foster the development of the topological spintronics.

6. Experimental Section

Sample Preparation: Polycrystalline samples were prepared by melting the mixtures of Mn and Sn in an Al\textsubscript{2}O\textsubscript{3} crucible sealed in an evacuated quartz ampoule in a box furnace at 1050 °C for 6 h. In preparation for single-crystal growth, the obtained polycrystalline materials were crushed into powders, compacted into pellets, and inserted into an Al\textsubscript{2}O\textsubscript{3} crucible that was subsequently sealed in an evacuated SiO\textsubscript{2} ampoule. Single-crystal growth was performed using a single-zone Bridgman furnace with a maximum temperature of 1080 °C and growth speed of 1.5 mm h\textsuperscript{-1}. These were exactly the same methods for fabricating the single crystals as those used for the previous study on the Kerr effect\textsuperscript{[31]} and the chiral anomaly due to Weyl fermions.\textsuperscript{[13]} Analysis using inductively coupled plasma spectrometry showed that the composition of the single crystal was Mn\textsubscript{7.07}Sn\textsubscript{93}. The bulk-Mn\textsubscript{3}Sn sample was cut and polished so that the sample had optically smooth surfaces along the (2\text{1} 1 0) plane. Then, the sample was annealed at 600 °C under vacuum (\textapprox 2 \times 10\textsuperscript{-6} Pa) for 1 h. Without breaking vacuum, 4 nm AlO\textsubscript{3} was subsequently prepared onto the Mn\textsubscript{3}Sn surface by electron beam deposition method to prevent degradation during MOKE measurements.

TR–MOKE Measurements: Time-resolved polar MOKE signals were measured in a conventional all-optical pump–probe setup using a Ti–sapphire laser with a regenerated amplifier.\textsuperscript{[83]} The incident light was almost perpendicular to the Mn\textsubscript{3}Sn surface, and the polar MOKE signal was proportional to the cluster magnetic octupole component normal to the Mn\textsubscript{3}Sn surface. The laser wavelength, pulse width, and repetition rate were 800 nm, 120 fs, and 1 kHz, respectively. A penetration depth of the incident light was \textapprox 20 nm. The pump beam was modulated with a frequency of 360 Hz using an optical chopper, and the signal was detected using a lock-in amplifier. Spot size and averaged laser power were \textapprox 1 mW and 40.66 mm for pump, and 0.05 mW and 40.14 mm for probe beams, respectively. The delay time dependence of the Kerr rotation signal was recorded. The Kerr rotation signal was detected by the differential method. For the case of the TR–MOKE measurements, nonmagnetic backgrounds (e.g., a quadratic background confirmed in the static MOKE results (Figure 2a in the main text) attributed to an artifact from measurement setup) were cancelled by averaging the data measured for the reversed magnetic field. All measurements were performed at room temperature.

Supporting Information

Supporting Information is available from the Wiley Online Library or from the author.

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Conflict of Interest

The authors declare no conflict of interest.

Data Availability Statement

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

Keywords

antiferromagnetic spintronics, damping, Mn\textsubscript{3}Sn, spin dynamics, Weyl semimetal

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