Ultrafast optical excitation of magnetic skyrmions

N. Ogawa¹, S. Seki¹,² & Y. Tokura¹,²

¹RIKEN Center for Emergent Matter Science (CEMS), Wako, Saitama 351-0198, Japan, ²PRESTO, Japan Science and Technology Agency, Tokyo 102-0075, Japan, ³Department of Applied Physics and Quantum Phase Electronics Center (QPEC), University of Tokyo, Tokyo 113-8656, Japan.

Magnetic skyrmions in an insulating chiral magnet Cu₂OSeO₃ were studied by all-optical spin wave spectroscopy. The spins in the conical and skyrmion phases were excited by the impulsive magnetic field from the inverse-Faraday effect, and resultant spin dynamics were detected by using time-resolved magneto-optics. Clear dispersions of the helimagnon were observed, which is accompanied by a distinct transition into the skyrmion phase, by sweeping temperature and magnetic field. In addition to the collective excitations of skyrmions, i.e., rotation and breathing modes, several spin precession modes were identified, which would be specific to optical excitation. The ultrafast, nonthermal, and local excitation of the spin systems by photons would lead to the efficient manipulation of nano-magnetic structures.
experiments\textsuperscript{17,18}. We also identified precession signals not observed in the previous reports, which would be explained by the strong impulsive excitation of the spin system.

**Results and Discussion**

The magnetic phase diagram of Cu\textsubscript{2}OSeO\textsubscript{3} and experimental setups are shown in Fig. 1 (see Methods for the detail). Figure 2 shows the representative spin precession spectra in the conical and SkX phases under the Hex // [110] (normal to the photon k-vector) [Fig. 1(b)]. In both phases, transient Faraday rotations with opposite initial phase (\(\pi\)-rad. shifted) were observed for right- and left-circular polarized pump excitations (RCP and LCP), respectively. The spin motion was negligible when excited with linearly-polarized pump pulses [Fig. 2 (a)], therefore we can safely rule out thermal effects and optically-induced changes in the magnetic anisotropy\textsuperscript{23}. We observed no pump-induced change in the probe transmittance (not shown). The spins in the Cu\textsubscript{2}OSeO\textsubscript{3} feel strong inverse-Faraday field (\(\pm H_{IF}\)) along the light k-vector of the pump pulse [Fig. 2(b) bottom inset], which tilts the magnetic moment (M) in the plane normal to the k-vector within the pump duration, triggering the spin precession generally expressed as \(\sin(\tau) e^{\frac{1}{t}}\) [Fig. 2(a) inset], where \(\tau\) is the decay constant. When the temperature was scanned from 40 to 59 K [see Fig. 1(a)] under the Hex of 165 Oe, a dispersion of the collective spin dynamics and a distinct transition to the SkX phase was observed as

![Figure 1](https://www.nature.com/scientificreports/)  
**Figure 1** | Phase diagram and experimental setups. (a) Phase diagram of a Cu\textsubscript{2}OSeO\textsubscript{3} single crystal deduced from magnetic susceptibility measurements under the in-plane magnetic field (\(\parallel [110]\)). Representative data points shown in Figs. 2 and 3 are indicated by an arrow and solid squares. (b) Schematic optical setup for detecting transient Faraday rotation of the probe light induced by the inverse-Faraday effect of the circularly-polarized pump light. (c) Absorption coefficient of the Cu\textsubscript{2}OSeO\textsubscript{3} sample with the pump and probe photon energies indicated by arrows.

![Figure 2](https://www.nature.com/scientificreports/)  
**Figure 2** | Spin dynamics in the conical and SkX phases. (a) Transient Faraday rotation of the probe light induced by the inverse-Faraday excitation in the conical (upper panel) and the SkX (lower panel) phases under the in-plane magnetic field of 165 Oe (\(\parallel [110]\)). The pump polarization was varied from right-circular (RCP), linear, to left circular (LCP). Insets show the schematics of spin precession and the magnified spectra near the time zero. (b) Temperature evolution of the collective spin dynamics excited by the RCP pump pulse (offset for clarity). The SkX phase is indicated by a shadowed box, and the beating structures in the spectra are indicated by an arrow. The response at time zero is omitted.
shown in Fig. 2(b). The large Faraday rotation at time zero is omitted from the data in the following.

It is notable that the spin precessions cannot be expressed with a single sinusoidal wave with an exponential decay [a characteristic modulation, called beating below, is exemplified by an arrow in Fig. 2 (b)]. This is naturally expected because the conical spins exhibit two fundamental excitation modes in our experimental geometry, called $\pm Q$ helimagnons$^{24}$, which have been detected in Cu$_2$OSeO$_3$ with the microwave resonance spectroscopy$^{17,18}$. We also found that the spin precession in the SkX phase cannot be expressed with a single frequency. Therefore, we fitted all the spectra with two sinusoidal functions, as shown in Fig. 3(a), which allowed us to avoid arbitrary determination of the phase boundaries. (In the lower panel of Fig. 3(a), the spectrum near the phase boundary with a clear shoulder structure is shown.) The fitting works reasonably well in both conical and SkX phases, yielding temperature- and field-dependent oscillation frequencies, amplitudes, and damping constants. The oscillation frequencies are plotted as a function of temperature in Fig. 3(b), showing a discontinuous transition from the conical phase into the SkX phase. By comparing to the previous reports$^{17,18}$, we can assign the observed spin dynamics to the $\pm Q$ helimagnons in the conical phase and rotation modes of the skyrmions [Fig. 3(b) insets], as will be discussed in detail below. By rendering the oscillation frequencies (of the $-Q$ mode and the CCW rotation) in the phase diagram, we can nicely visualize the SkX phase [Fig. 3(c)].

When the external magnetic field $H_{\text{ex}}$ is applied along the sample normal [|| [110], Fig. 4], only one collective spin mode is expected in the SkX phase (and no mode in the other phases)$^9$. In this setup, the Faraday rotation of the probe light is smaller than that of the former setup, consistent with the previous report$^{18}$. We found that the observed spin dynamics can be fitted with a single sinusoidal function [Fig. 4(a)]. The deduced oscillation frequencies fell into those of the skyrmion breathing motion, which also nicely reproduce the phase diagram deduced from the magnetic susceptibility measurements [Fig. 4(b)].

The oscillation frequencies of the helimagnons, and those of the rotation and the breathing modes in the SkX phases nicely match to the values found in the previous reports$^{17,18}$. Judging from this fact and also from the phase maps in Figs. 3(c) and 4(b), it is concluded that we have successfully detected skyrmions from their optically-excitied dynamics. We note that the CW rotation mode has been inferred only from the nonreciprocal directional dichroism$^{17}$, due probably to the small spectral weight as expected from the numerical calculations$^9$. In contrast, we clearly identified two spin precessions in the SkX phase as shown in Fig. 3. There, the high frequency mode ($\sim 1.5$ GHz) is not due to the mixing of the conical phase, judging from the discontinuous transition from the conical phase to the SkX phase seen in Fig. 3(b).

To scrutinize the observed spin modes, we plot the magnetic-field dependence of the spin precessions in Fig. 5 for the different magnetic-field orientations. For the $H_{\text{ex}}$ [|| [110] [Fig. 5(a)], the precession frequency decreases with increasing the $H_{\text{ex}}$ in the conical spin phase$^{25}$, whereas increases with increasing $H_{\text{ex}}$ in the SkX phase. For the $H_{\text{ex}}$ [|| [110] [Fig. 5(c)], the frequency decreases in the SkX phase. These observations are consistent with the expected behaviors of the conical spin phase, rotation modes and the breathing mode of the skyrmions, respectively$^{17}$. Therefore, in addition to the reasoning from its absolute frequency, the higher-lying mode in the SKX phase can be assigned to the CW rotation. The reentrant behavior of the conical phase by increasing the $H_{\text{ex}}$ after experiencing a pocket of the SkX phase at 56.5 K [Fig. 5(a)], also supports the identification of the SkX phase, since this pocket is well isolated from other spin phases. It is also expected that the damping increases in the SKX phase$^{25}$, which is roughly captured in our data in Fig. 5(b). The long-
The intermediate phase has larger spin fluctuations\(^1\), expected to host isolated skyrmions or fragmented skyrmion domains, and to be susceptible to the magnetic history of the sample. The spin dynamics invoked in this region may indicate the existence of these fragmented domains\(^1\), or other induced spin excitation by the strong inverse-Faraday field.

In the metallic helimagnet Fe\(_{0.8}\)Co\(_{0.2}\)Si, which is also known to host skyrmions, the spin dynamics has been analyzed with time-dependent Gilbert dampings\(^2\), since no beating features in the precession signals were detected by the optical spectroscopy. In the case of Cu\(_2\)OSeO\(_3\) here, the beating and shoulder structures in the spin signals revealed the distinct multiple precession dynamics.

**Conclusions**

To summarize, we have successfully detected the collective excitation modes of skyrmions in the insulating chiral magnet Cu\(_2\)OSeO\(_3\) by optical pump-probe techniques. The rotation and breathing modes are identified in the skyrmion phase, and several additional spin precession modes have also been detected. We discussed the difference between the inverse-Faraday and microwave excitations, which may explain the observed multi-mode spin excitations.

**Methods**

Single crystals of Cu\(_2\)OSeO\(_3\) were grown by the chemical vapor transport method, and were polished down to 300 \(\mu\)m in thickness to expose (110) planes. The conical spin and SkX phases were checked by the magnetic susceptibility measurements [Fig. 1(a)], in which some demagnetization effects make small deviations from sample to sample\(^3\). We studied the spin dynamics by the time-resolved magneto-optics with pulsed laser sources (~120 fs, 1 kHz) at near-normal incidence [Fig. 1(b)]. The pump pulse (2 mW on ~100 \(\mu\)m spot) excite the spin system at 0.95 eV (within the optical gap) by the inverse-Faraday effect, and the induced spin precession was

---

**Figure 4** | Spin mode under the out-of-plane magnetic field. (a) Transient Faraday rotation of the probe light with the external magnetic field applied along the [110] axis (nearly parallel to the photon \(k\) vector). The red line shows the fit with a single spin precession with exponential decay. (b) Contour map of the spin precession frequencies plotted together with the phase boundaries deduced from the magnetic susceptibility (open circles).

---

**Figure 5** | Spin dynamics as a function of external magnetic field. Spin precession frequencies and decay constants as a function of the external magnetic field, for (a),(b) \(H_{ex} \parallel [110]\) and (c) \(H_{ex} \parallel [110]\).
detected by the Faraday rotation of the linearly-polarized probe at 2.2 eV (Figs. 1(b) and (c)). Thus the probe pulse detects the change in the out-of-plane ($[[110]]$) component of the magnetization. Note that the pump pulse at 0.83 eV induced qualitatively the same spin dynamics with slightly reduced efficiency (not shown).

1. Nagaoa, N. & Tokura, Y. Topological properties and dynamics of magnetic skyrmions. Nat. Nanotechnol. 8, 899–911 (2013).
2. Schulz, T. et al. Emergent electrodynamics of skyrmions in a chiral magnet. Nat. Phys. 8, 301–304 (2012).
3. Mühlbauer, S. et al. Skyrmion Lattice in a Chiral Magnet. Science 323, 915–919 (2009).
4. Yu, X. Z. et al. Real-space observation of a two-dimensional skyrmion crystal. Nature 465, 901–904 (2010).
5. Jonietz, F. et al. Spin Transfer Torques in MnSi at Ultralow Current Densities. Science 330, 1648–1651 (2010).
6. Yu, X. Z. et al. Skyrmion flow near room temperature in an ultralow current density. Nature Commun. 3, 988 (2012).
7. White, J. S. et al. Electric field control of the skyrmion lattice in Cu$_2$OSeO$_3$. J. Phys.: Condens. Matter 24, 432201 (2012).
8. Mochizuki, M. et al. Thermally driven ratchet motion of a skyrmion microcrystal and topological magnon Hall effect. Nature Mater. 13, 241–246 (2014).
9. Mochizuki, M. Spin–Wave Modes and Their Intense Excitation Effects in Skyrmion Crystals. Phys. Rev. Lett. 108, 017601 (2012).
10. Iwasaki, J., Beekman, A. J. & Nagaoa, N. Theory of magnon–skyrmion scattering in chiral magnets. Phys. Rev. B 89, 064412 (2014).
11. Petrova, O. & Tchernyshyov, O. Spin waves in a skyrmion crystal. Phys. Rev. B 84, 214433 (2011).
12. Miide, P. et al. Unwinding of a Skyrmion Lattice by Magnetic Monopoles. Science 340, 1076–1080 (2013).
13. Seki, S., Yu, X. Z., Ishiwata, S. & Tokura, Y. Observation of Skyrmions in a Multiferroic Material. Science 336, 198–201 (2012).
14. Adams, T. et al. Long-Wavelength Helimagnetic Order and Skyrmion Lattice Phase in Cu$_2$OSeO$_3$. Phys. Rev. Lett. 108, 237204 (2012).
15. Mochizuki, M. & Seki, S. Magneto-electric resonances and predicted microwave diode effect of the skyrmion crystal in a multiferroic chiral-lattice magnet. Phys. Rev. B 87, 134403 (2013).
16. Seki, S., Ishiwata, S. & Tokura, Y. Magneto-electric nature of skyrmions in a chiral magnetic insulator Cu$_2$OSeO$_3$. Phys. Rev. B 86, 060403(R) (2012).
17. Okamura, Y. et al. Microwave magnetoelectric effect via skyrmion resonance modes in a heli-magnetic multiferroic. Nature Commun. 4, 2391 (2013).
18. Onose, Y., Okamura, Y., Seki, S., Ishiwata, S. & Tokura, Y. Observation of Magnetic Excitations of Skyrmion Crystal in a Helimagnetic Insulator Cu$_2$OSeO$_3$. Phys. Rev. Lett. 109, 037603 (2012).
19. Finazzi, M. et al. Laser-Induced Magnetic Nanostructures with Tuneable Topological Properties. Phys. Rev. Lett. 110, 177205 (2013).
20. Ogasawara, T., Iwata, N., Murakami, Y., Okamoto, H. & Tokura, Y. Submicron-scale spatial feature of ultrafast photoinduced magnetization reversal in TbFeCo thin film. Appl. Phys. Lett. 94, 162507 (2009).
21. Kirilyuk, A., Kimel, A. V. & Rasing, Th. Ultrafast optical manipulation of magnetic order. Rev. Mod. Phys. 82, 2731–2784 (2010) and references therein.
22. Satoh, T. et al. Directional control of spin-wave emission by spatially shaped light. Nature Photon. 6, 662–666 (2012).
23. Hansteen, F., Kimel, A., Kirilyuk, A. & Rasing, Th. Femtosecond Photomagnetic Switching of Spins in Ferrimagnetic Garnet Films. Phys. Rev. Lett. 95, 047402 (2005).
24. Katakba, M. Spin Waves in Systems with Long Period Helical Spin Density Waves Due to the Antisymmetric and Symmetric Exchange Interactions. J. Phys. Soc. Jpn. 56, 3635–3647 (1987).
25. Zang, J., Mostovoy, M., Han, J. H. & Nagaoa, N. Dynamics of Skyrmion Crystals in Metallic Thin Films. Phys. Rev. Lett. 107, 136804 (2011).
26. Schloemann, E., Green, J. J. & Milan, U. Recent Developments in Ferrimagnetic Resonance at High Power Levels. J. Appl. Phys. 31, 3865–3955 (1960).
27. Sokoloski, M. M. & Tanaka, T. Excitation spectra of magnetic bubble lattices. J. Appl. Phys. 45, 3091–3101 (1974).
28. Schlickeiser, F. et al. Temperature dependence of the frequencies and effective damping parameters of ferrimagnetic resonance. Phys. Rev. B 86, 214416 (2012).
29. Janson, O. et al. The quantum nature of skyrmions and half-skyrmions in Cu$_2$OSeO$_3$. Nature Commun. 5, 5376 (2014).
30. Ozerv, M. et al. Establishing the Fundamental Magnetic Interactions in the Chiral Skyrmionic Mott Insulator Cu$_2$OSeO$_3$, by Terahertz Electron Spin Resonance. Phys. Rev. Lett. 113, 157205 (2014).
31. Reid, A. H. M., Kimel, A. V., Kirilyuk, A., Gregg, J. F. & Rasing, Th. Optical Excitation of a Forbidden Magnetic Resonance Mode in a Doped Lutetium-Iron-Garnet Film via the Inverse Faraday Effect. Phys. Rev. Lett. 105, 107402 (2010).
32. Koralek, J. et al. Observation of Coherent Helimagnons and Gilbert Damping in an Itinerant Magnet. Phys. Rev. Lett. 109, 247204 (2012).

Acknowledgments
The authors thank G. Tatara, W. Koshihiae, F. Kagawa, Y. Nii, and N. Nagaoa for stimulating discussions. This research is granted by the Japan Society for the Promotion of Science (JSPS) through the “Funding Program for World-Leading Innovative R&D on Science and Technology (FIRST Program),” initiated by the Council for Science and Technology Policy (CSTP), and by the JSPS Grant-in-Aid for Scientific Research(S) No. 24224009 and for Challenging Exploratory Research No. 26610109. N.O. was supported by RIKEN Incentive Research Projects.

Author contributions
N.O., S.S. and Y.T. conceived the experiments. S.S. fabricated the sample, and N.O. carried out the optical experiments. N.O., S.S. and Y.T. wrote the manuscript. All authors contributed considerably.

Additional information
Competing financial interests: The authors declare no competing financial interests.

How to cite this article: Oogawa, N., Seki, S. & Tokura, Y. Ultrafast optical excitation of magnetic skyrmions. Sci. Rep. 5, 9552; DOI:10.1038/srep09552 (2015).

This work is licensed under a Creative Commons Attribution 4.0 International License. The images or other third party material in this article are included in the article’s Creative Commons license, unless indicated otherwise in the credit line; if the material is not included under the Creative Commons license, users will need to obtain permission from the license holder in order to reproduce the material. To view a copy of this license, visit http://creativecommons.org/licenses/by/4.0/