Stabilization of magnetic skyrmions by uniaxial tensile strain

S. Seki,1 Y. Okamura,2 K. Shibata,1 R. Takagi,1 N. D. Khanh,1 F. Kagawa,1 T. Arima,1,3 and Y. Tokura1,2
1RIKEN Center for Emergent Matter Science (CEMS), Wako 351-0198, Japan
2Department of Applied Physics and Quantum Phase Electronics Center (QPEC), University of Tokyo, Tokyo 113-8656, Japan
3Department of Advanced Materials Science, University of Tokyo, Kashiwa 277-8561, Japan
(Rceived 11 June 2017; published 19 December 2017)

Magnetic skyrmions with a topological particle nature have recently attracted attention as a potential information carrier for novel magnetic storage devices. For single-phase bulk crystals, skyrmions usually appear for a very narrow temperature region just below the magnetic ordering temperature \( T_c \), and the stabilization of skyrmions for a wider temperature range remains an important challenge. Here, by investigating the impact of uniaxial tensile stress for a chiral magnet \( \text{Cu}_2\text{OSeO}_3 \), we demonstrate that only less than 0.2% of uniaxial elongation can dramatically stabilize skyrmions for an entire temperature range from \( T_c \) to the lowest temperature. The stability of skyrmions essentially depends on the geometrical relationship among the directions of strain, magnetic field, and crystallographic axes, which is consistently explained in terms of the anisotropic modulation of the Dzyaloshinskii-Moriya interaction and magnetocrystalline anisotropy. Our finding may provide a good strategy for materials design to enhance the stability of skyrmions.

DOI: 10.1103/PhysRevB.96.220404

The skyrmion is a concept describing a topologically protected excitation with a particle nature [1], which appears as a nanometric vortexlike swirling spin texture in magnetic materials [Fig. 1(i)] [2–5]. Magnetic skyrmions are characterized by a unique manner of interaction with an external electric field, through the quantum Berry phase in metallic systems [6–9] or relativistic spin-orbit interactions in insulating systems [10,11]. Due to their electric controllability, particle nature, and small size, skyrmions are intensively studied as a potential information carrier for novel magnetic storage with high information density and energy efficiency [12–14].

Skyrmions are generally stabilized by a Dzyaloshinskii-Moriya (DM) interaction, i.e., an antisymmetric exchange interaction emerging under noncentrosymmetric environments [2,3,5]. In the case of single-phase compounds, the formation of skyrmions has mostly been reported for a series of magnetic materials with a chiral cubic crystal structure, such as metallic \( B20 \) alloys (\( T_c \sim 280 \text{ K} \)) [3,15,16] and Co-Zn-Mn alloys (\( T_c \sim 400 \text{ K} \)) [17] or insulating \( \text{Cu}_2\text{OSeO}_3 \) (\( T_c = 58 \text{ K} \)) [10], all of which are characterized by similar \( H \)-field-\( T \)-temperature phase diagrams, as shown in Fig. 1(f). However, the bulk crystals of these compounds host skyrmions only for a very narrow temperature range just below the magnetic ordering temperature \( T_c \), and the stabilization of skyrmions for a wider temperature range is essential for their application. For this purpose, several different approaches have been proposed. For example, the employment of thin-film samples thinner than the magnetic modulation period (<100 nm) can stabilize the skyrmion state for the out-of-plane magnetic field [18], but this approach is not applicable to the arbitrary shape of the crystal. Another potential strategy is the application of uniaxial pressure [19–21], hydrostatic pressure [22,23], or electric field [24]. However, their reported magnitude of modulation in \( T_{c2} \) (i.e., a lower critical temperature of the skyrmion state) usually remains just in the order of several degrees Kelvin, and the stabilization of skyrmions for the entire temperature range from \( T_c \) to zero temperature has yet to be achieved. Note that the rapid cooling of the sample can often create a supercooled skyrmion state [24–26], while this corresponds to a metastable state with a finite lifetime and not the thermal-equilibrium ground state.

In this Rapid Communication, we report the dramatic stabilization of the skyrmion state with a minimal uniaxial tensile strain for the prototypical chiral-lattice insulator \( \text{Cu}_2\text{OSeO}_3 \) with a cubic crystal structure. The observed strain-induced modulation of \( T_{c2} \) is one order of magnitude larger than previous reports for MnSi with a uniaxial compressive strain [20,21], and we found that only less than 0.2% of uniaxial lattice elongation is sufficient to stabilize the SkX state down to the lowest temperature. The stability of the skyrmion state essentially depends on the geometrical relationship among the directions of uniaxial strain, magnetic field, and crystallographic axes, which can be consistently explained in terms of the strain-induced modulation of the DM interaction and magnetocrystalline anisotropy.

A schematic illustration of the device structure employed in this study is shown in Fig. 1(a). Here, we fabricated two different types of devices: The plate-shaped \( \text{Cu}_2\text{OSeO}_3 \) single crystal with a thickness of 1 \( \mu \text{m} \) was fixed to the Si substrate by W (tungsten) bonding at one [Fig. 1(b)] or both [Fig. 1(c)] side edges at room temperature. Only in the latter case is the uniaxial tensile strain \( \sigma \) applied to \( \text{Cu}_2\text{OSeO}_3 \) due to the mismatch of thermal expansion coefficients at low temperatures, as confirmed by the simulation of thermal strain given in the Supplemental Material [Fig. S1(d)] [27]. Figure 1(d) indicates the reported temperature dependence of the lattice constant for Si [35] and \( \text{Cu}_2\text{OSeO}_3 \) [36], which suggests that the strained \( \text{Cu}_2\text{OSeO}_3 \) sample is elongated along the bridging direction by 0.12%, and this value remains almost constant below \( T_c = 58 \text{ K} \). As discussed later, the identification of the magnetic phase is possible through the analysis of the magnetic resonance modes [37–40]. For this purpose, an Au coplanar waveguide was fabricated on the substrate, which enables the measurement of microwave absorption spectra \( \Delta S_{11} \) associated with the magnetic resonance in \( \text{Cu}_2\text{OSeO}_3 \) by a vector network analyzer.
While this helical, conical, skyrmion lattice, ferromagnetic, and paramagnetic respond to ing top-view optical images of unstrained and strained samples. The modes described with dashed lines are expected to cause weaker microwave absorption than the ones with solid lines. (f) H-T phase diagram for unstrained Cu2OSeO3, obtained with the device shown in (b). In (e) and (f), H, C, SkX, FM, and PM represent helical, conical, skyrmion lattice, ferromagnetic, and paramagnetic spin states, respectively. (g)-(i) Schematic illustration of helical, conical, and skyrmion lattice spin textures, respectively.

First, we briefly discuss the H-T phase diagram for the unstrained Cu2OSeO3 sample [Fig. 1(f)], which well reproduces the ones reported in Refs. [10,39,40]. According to previous neutron diffraction experiments [41,42], a helical magnetic order is realized at $H = 0$, where the neighboring spins rotate within a plane normal to the magnetic modulation vector $q$ [Fig. 1(g)]. While this $q$ vector is weakly pinned to the (001) axes by magnetocrystalline anisotropy, the application of an external magnetic field reorients $q$ along the $H$ direction (i.e., $H \parallel q$) and stabilizes the conical magnetic structure, as shown in Fig. 1(h). A further increase of $H$ induces the transition into a collinear ferromagnetic state. The SkX (i.e., triangular lattice of the skyrmion) state [Fig. 1(i)], characterized by triple $q$ vectors normal to an external magnetic field (i.e., $H \perp q$), appears for a narrow $H$-$T$ region just below $T_c$.

Figure 1(e) indicates the theoretically predicted $H$ dependence of the magnetic resonance frequency for various magnetic phases [37], which has been experimentally verified for Cu2OSeO3 in Refs. [37,39,40]. The ferromagnetic state generally hosts a single magnetic resonance mode, whose frequency shows an $H$-linear increase. On the other hand, the helical or conical spin states host two resonance modes (i.e., the $+Q$ and $-Q$ modes, the latter of which is located at a lower frequency with a much weaker absorption intensity) and their frequency gradually decreases as a function of $H$ [37,43]. The SkX state also hosts two resonance modes, i.e., the clockwise (CW) and counterclockwise (CCW) rotational modes, where the CCW mode is characterized by a lower frequency and stronger absorption intensity [37,38]. Importantly, the resonance frequency of the CCW mode monotonically increases against $H$, which enables a clear distinction of the SkX state from the helical or conical state.

On the basis of the above knowledge, we investigated the $H$-$T$ phase diagram for a Cu2OSeO3 sample with uniaxial tensile strain $\sigma$, using the device shown in Fig. 1(c). First, we discuss the case with the in-plane $H$ applied normal to the $\sigma$ direction [Fig. 2(b)]. Figures 2(c) and 2(e) indicate the $H$ dependence of the microwave absorption spectra $\Delta S_{11}$ at 48 K. Between 23 and 43 mT, two resonance modes are identified, where the lower-lying mode shows stronger microwave absorption and its frequency increases as a function of $H$. These features are consistent with the theoretical prediction for the SkX state [Fig. 1(e)] [37,38], and the observed modes can be assigned as the CW and CCW modes of the skyrmions. Except for this $H$ region, the resonance frequency shows a negative (positive) slope against $H$ below (above) 53 mT, which represents the helical/conical (ferromagnetic) state. Note that the $-Q$ mode in the helical/conical state is too weak to be detected, in accord with theory [37]. In Figs. 2(d) and 2(f), we also plotted the corresponding data measured at 10 K. Notably, at this low temperature, the SkX state hosting the CW and CCW modes with similar characters appears between 30 and 48 mT, and a discontinuous change of resonance frequency is observed upon the transition into the ferromagnetic state. Figure 2(a) summarizes the $H$-$T$ phase diagram for the configuration shown in Fig. 2(b), with the background color representing the peak intensity of the CCW spectra. A comparison of Figs. 1(f) and 2(a) clearly demonstrates that the application of uniaxial tensile strain dramatically stabilizes the SkX state, even down to the lowest temperature.

The stability of the SkX state largely depends on the directional relationship between $H$ and $\sigma$. On the basis of the $T$ and $H$ variation of the magnetic resonance modes, the $H$-$T$ phase diagram obtained with the in-plane $H$ parallel to $\sigma$ is indicated in Fig. 3(a). In this configuration, the SkX state is rather destabilized and completely disappears from the phase diagram. On the other hand, with the out-of-plane $H$ normal to $\sigma$, the SkX state is moderately stabilized down to 43 K [Fig. 3(b)] (see Fig. S2 in the Supplemental Material...
FIG. 2. Stabilization of the skyrmion state for Cu$_2$OSeO$_3$ with uniaxial tensile strain. (a) $H$-$T$ phase diagram for Cu$_2$OSeO$_3$ with uniaxial tensile strain $\sigma$, obtained with the device shown in Fig. 1(c). Here, in-plane $H$ is applied normal to $\sigma$, and the corresponding measurement geometry is shown in (b). The background color represents the microwave absorption intensity $|\Delta S_{11}|$ of the CCW mode in the the SkX state. (c), (d) Magnetic field dependence of microwave absorption spectra, obtained at 48 and 10 K, respectively. The background color represents the magnitude of microwave absorption, and the raw data of absorption spectra for selected magnetic field values at each temperature are shown in (e) and (f).

for the raw data of the $|\Delta S_{11}|$ spectra [27]). The above results suggest that the SkX state becomes more stable for $H \perp \sigma$, and unstable for $H \parallel \sigma$. Intuitively, these behaviors can be understood by considering the $\sigma$-induced pinning of the magnetic modulation vector $q$ [20,21]. In Figs. 3(c) and 3(d), we plotted the $H$ dependence of absorption spectra at 34 K for the $H \parallel \sigma$ and $H \perp \sigma$ setups, respectively. In the latter configuration, a discontinuous change of resonance frequency is observed at 80 mT, which corresponds to the transition between the helical and conical spin state, i.e., the reorientation of the $q$ vector along the $H$ direction. In contrast, such an anomaly is absent in the former $H \parallel \sigma$ setup [Fig. 3(e)], suggesting that the helical $q$ vector is pinned along the $\sigma \parallel [001]$ direction under zero magnetic field for Cu$_2$OSeO$_3$. To sustain such a $\sigma \parallel q$ relationship, the SkX ($H \perp q$) state can be stabilized and destabilized against the competing conical ($H \parallel q$) spin state under the $H \perp \sigma$ and $H \parallel \sigma$ configurations, respectively.

To further discuss the microscopic origin of these phenomena, we investigated the stability of the SkX state as a function of the angle ($\theta_H$) between the in-plane $H$ direction and $\sigma$ direction [Fig. 3(e)]. In Fig. 3(f), the maximum absorption intensity of the CCW mode in the SkX state at each temperature, $\theta_H = 0^\circ$ and $\theta_H = 90^\circ$ correspond to (a) in this figure and Fig. 2(a), respectively. (g), (h) $\theta_H$ dependence of free-energy terms $\langle \Delta K \rangle$ and $\langle \Delta M \rangle$, respectively, calculated for the SkX and conical spin states. Here, we assumed that $H$-induced magnetization takes half of the saturation value.
anisotropy and/or the DM interaction by the uniaxial tensile strain. In the case of the chiral cubic crystal structure, a uniaxial lattice elongation along the \( z \) direction causes the emergence of an additional free-energy term \( \Delta_K = -K_z m_z^2 \) and \( \Delta_{DM} = -d_z \langle m_i \Delta m_i \rangle \), which correspond to the contribution from magnetocrystalline anisotropy and the DM interaction, respectively [19,44]. Here, \( K_z \) and \( d_z \) represent the magnitude of each strain-induced term, and \( m_i (i = x,y,z) \) is the local magnetization component along the \( i \) direction. By considering the observed pinning of the helical \( q \) vector along the \( \sigma \) direction, the sign of \( K_z \) and \( d_z \) should be negative and positive, respectively. The strain-induced energy shift of each magnetic phase can be estimated by calculating \( \langle \Delta_K \rangle = \int \Delta_K d\mathbf{r} / \int d\mathbf{r} \) and \( \langle \Delta_{DM} \rangle = \int \Delta_{DM} d\mathbf{r} / \int d\mathbf{r} \), where the integral is taken over each magnetic unit cell. Figures 3(g) and 3(h) indicate the \( \theta_H \) dependence of \( \langle \Delta_K \rangle \) and \( \langle \Delta_{DM} \rangle \) calculated for the SkX and conical states (see the Supplemental Material for details of the calculation and a direct experimental evaluation of \( K_z \) and \( d_z \) [27]). Both terms stabilize (destabilize) the SkX state for \( \theta_H \) larger (smaller) than the critical value around 60°, well reproducing the experimental observation in Fig. 3(f).

In Fig. 4(e), the \( H-T \) phase diagrams for the strained and unstrained \( \text{Cu}_2\text{OSeO}_3 \), measured with various directional combinations of \( H \) and \( \sigma \) under different crystallographic orientations, are summarized. In the case of the unstrained \( \text{Cu}_2\text{OSeO}_3 \), the SkX state appears between 56 and 58 K for any \( H \) direction, reflecting the cubic symmetry of the original crystal structure [Fig. 4(c)] [41,45]. By applying \( \sigma \parallel H \), the SkX state is always suppressed [Fig. 4(d)]. In contrast, the application of \( \sigma \perp H \) generally enhances the stability of the SkX state. Here, the SkX state is stabilized down to the lowest temperature for the in-plane \( H \) normal to \( \sigma \) [Fig. 4(a)], while the lower critical temperature \( T_{c2} \) for the SkX state is of the order of 40 K for the out-of-plane \( H \) normal to \( \sigma \) [Fig. 4(b)]. In general, a magnetic dipole-dipole interaction favors orienting the magnetization along the in-plane direction. Under the out-of-plane \( H \), the conical spin state has a larger in-plane spin component than the SkX state, which may explain the relatively limited degree of SkX stabilization in this experimental geometry. Notably, for the configuration with \( \sigma \parallel [110] \) and \( H \parallel [110] \), such \( \sigma \)-induced stabilization of the SkX state does not occur. Since the \( q \) vector favors orienting along the \( (100) \) axes in the unstrained \( \text{Cu}_2\text{OSeO}_3 \) [41,42], the application of \( \sigma \parallel [110] \) probably selects the \( q \parallel (100) \) closest to the \( \sigma \) direction (i.e., \( q \parallel [100] \) and \( q \parallel [010] \)) rather than realizing the \( q \perp \sigma \) configuration. In this situation, stable \( q \) directions do not exist within the \( H \perp q \) plane for \( H \parallel [110] \), therefore, the stability of the SkX state is not enhanced. This result suggests that the appropriate choice for crystallographic orientation is also important for the \( \sigma \)-induced control of SkX stability.

In this Rapid Communication, we experimentally demonstrated that uniaxial elongation only by 0.2% in the isotropic chiral magnet can stabilize the skyrmion state for the entire temperature range from \( T_c \) to the lowest temperature. Note that some metallic \( B20 \) alloys such as \( \text{MnSi} \) [20,21] and \( \text{FeGe} \) [44] are reported to host opposite signs of \( K_z \) or \( d_z \) from the current case with insulating \( \text{Cu}_2\text{OSeO}_3 \), and further investigation of the relationship between the electronic band structure and the magnitude/sign of the strain effect, in particular, for other chiral-lattice magnets with higher \( T_c \), would be an interesting issue [46]. Our present findings also provide a strategy for materials design toward the enhancement of SkX stability at ambient pressure, since a similar magnitude of lattice symmetry reduction is often possible through chemical substitution approaches such as tuning of the tolerance factor [47] or the introduction of a Jahn-Teller active ion [48]. Considering the present fact that the skyrmion lattice is stabilized within an anisotropic two-dimensional plane including the \( \sigma \) axis, the noncentrosymmetric magnets without an isotropic two-dimensional plane, such as the ones with orthorhombic or monoclinic crystal lattice symmetries, may also be promising candidates to realize skyrmions with enhanced stability.

The authors thank N. Nagaosa, Y. Iwasa, M. Kawasaki, Y. Otani, K. Kondou, Y. Nii, M. Nakamura, T. Nakajima, N. Kanazawa, T. Kurumaji, and D. Morikawa for enlightening discussions and experimental help. This work was partly supported by the Mitsubishi Foundation, Grants-In-Aid for Scientific Research (Grants No. 17H05186 and No. 16K13842) from JSPS, and PRESTO from JST.