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ABSTRACT
We examine the surface size- and shape-effects of soliton annihilation and soliton nucleation in chiral magnet CrNb$_3$S$_6$. We measure magnetization ($M$) curves of submillimeter-sized single crystals with an equal length along the $c$-axis ($L_c = 10 \mu m$) but with different cross sections in the $ab$-plane ($S_{ab} = 0.120–0.014 \text{mm}^2$). We find a ferromagnetic type of magnetizing (FMM) with a convex curve ($d^2M/dH^2 < 0$) near zero field ($H = 0$) and a major jump in $M$ near the forced ferromagnetic state, which are more conspicuous, compared with earlier samples with submillimeter $L_c$. A new finding is that the major jump in $M$ occurs at lower fields in samples with the smaller $S_{ab}$. We further perform numerical simulation of the magnetization process with the Landau–Lifshitz–Gilbert equation of the Langevin-type. Based on the numerical results, we attribute the FMM at small fields to rapid annihilation of soliton assisted by the reduction of Dzyaloshinskii-Moriya interaction near the surfaces. We also discuss possible penetration processes of chiral soliton through the ac-($bc$-)plane as well as $ab$-plane, and its relation to the major jump in $M$. Our experimental and calculated results will contribute to understanding of the effects of topological metastability in chiral magnets.

I. INTRODUCTION
Recently, an incommensurate noncollinear magnetic order called a chiral soliton lattice (CSL) has attracted much attention. $^{1,3}$ The CSL appears in a mono-axial chiral helimagnet such as hexagonal CrNb$_3$S$_6$ $^{1,3}$ and trigonal YbNi$_3$Al$_5$. $^{1,7}$ The spin texture is based on the helimagnetic spin structure at zero magnetic field, which results from the competition between exchange interaction along...
the chiral helical axis and an antisymmetric Dzyaloshinskii-Moriya (DM) exchange interaction\(^{15}\) along the chiral axis. The kink-type of spin texture, CSL, is stabilized when a DC magnetic field (\(H\)) is applied perpendicularly to the chiral helical axis,\(^{15,23}\) and it is a kind of long-ranged topological spin texture, along with the skyrmion lattice system.\(^{24-28}\) For \(H\) greater than the critical field (\(H_c\)), a forced ferromagnetic state appears. Field induced evolution from a helimagnetic state to the forced ferromagnetic state via CSL exhibits a characteristic magnetization (\(M\)) curve.

The kinks (single discommensurations), which are termed solitons, are constitutive objects of CSL and carry magnetic moments antiparallel to the magnetic field. Magnetization processes are thus accompanied by a change in soliton number.\(^{15,23}\) Topological stability hinders annihilation and nucleation of the solitons inside the sample, whereas soliton number can change via release and penetration of solitons through the surface. Near the surface, topological objects are affected by energy barriers, which causes metastability or hysteresis in the process with change in the number of topological defects\(^{29}\) (this energy barrier in chiral magnets is related to surface twist, which has been extensively discussed in Refs. 13–19). Experimentally, the existence of these solitons in CSL in CrNb\(_3\)S\(_6\) is manifested in discrete change in MR and \(M\) as well as the hysteresis in \(H\).\(^{20,21,22}\) In this paper, we study the topological effects on “soliton annihilation” and “soliton nucleation” in submillimeter-scale single crystals of CrNb\(_3\)S\(_6\) with a large area of magnetically easy plane, by conducting the \(M - H\) measurements while systematically changing the surface-size. There, as shown in Fig. 1, we assume the CSL state with ferromagnetic alignment near surfaces [Fig. 1(b)] occurs between the helimagnetic state [Fig. 1(a)] and the forced ferromagnetic state [Fig. 1(c)].

The Dzyaloshinskii’s theory and experiments\(^1\) of field induced evolution of CSL in CrNb\(_3\)S\(_6\) using the Lorentz transmission electron microscope show good agreement with each other, and thus this material can be regarded as an archetypical CSL material. It has a hexagonal layered structure consisting of NbS\(_2\) layers with an intercalation of Cr atoms.\(^{23}\) Each Cr atom has spin 3/2 and almost full magnetic moment (\(\sim 3\mu_B\)), which justifies theoretical approaches based on localized spin models and the chiral sine-Gordon model in spite of metallic conduction in this material. In zero field, CrNb\(_3\)S\(_6\) undergoes a phase transition at \(T_c = 127\) K, below which the helimagnetic state is stable. The periodicity of the helimagnetic structure at \(H = 0\) is 48 nm.\(^{4,22}\) In this state, the propagation vector is along the principal \(c\)-axis and perpendicular to the \(ab\)-plane (the NbS\(_2\) layers), which is the magnetic easy-plane. Thanks to the coupling between conduction electrons and magnetic moments, the magnetoelectrical resistance (MR) is an effective probe with which to reveal the magnetic properties of CrNb\(_3\)S\(_6\) in addition to magnetization measurements. The phase diagram at finite \(H\) has been investigated in detail from both experimental\(^14,30\) and theoretical\(^{19,31-35}\) viewpoints. Recently, the electrical magnetochiral effect has also been studied.\(^{36}\)

Below, we review the discrete change in both MR and \(M - H\) measurements in CrNb\(_3\)S\(_6\). Discrete changes accompanied by large hysteresis were first observed in the MR of the micro specimens, whose crystal sizes along the \(c\)-axis (\(L_c\)) were 10 and 25 \(\mu\)m.\(^{37}\) Afterwards, similar discreteness was also observed for exfoliated crystals with \(L_c \geq 57\) nm in the MR experiments.\(^{38}\) Interestingly, in the bulk crystals, the discrete property and hysteresis in MR was not observed.\(^{24}\) For the conventional \(M - H\) measurements requiring a larger specimen volume, initially the measurements on bulky single crystals were conducted: In 2016, some of us investigated the size dependence of \(M\) for three single crystals labeled as (A), (B), and (C), whose \(L_c\)’s were 120 \(\mu\)m,\(^{20}\) 110 \(\mu\)m\(^{39}\) and 60 \(\mu\)m,\(^{40}\) respectively. For crystals A, B, and C, the largest \(M\) jump owing to significant avalanche-like soliton nucleation (ASN) corresponds to approximately 1 \%, 1 \%, and 4 \% of the saturated magnetization \(M_s\), respectively. As \(L_c\) decreases, the stochastic feature in the discrete change in \(M\) (the \(M\) jump) is suppressed, and a more prominent \(M\) jump accompanied with hysteresis is observed in a reproducible manner.\(^{20,21}\) In the recent past, \(M\) of micro-specimens with \(L_c\) of about 10 \(\mu\)m and thickness of 0.1 \(\mu\)m perpendicular to the \(c\)-axis has been measured by using soft X-ray magnetic circular dichroism (MCD).\(^{22}\) The \(M\) hysteresis depended on the specimen geometry, suggesting a topological aspect to the soliton-release and soliton-penetration. Thus, the discrete feature accompanied by hysteresis in both MR\(^{7,38}\) and \(M - H\) measurements\(^{10,20-22}\) is experimentally found to depend on the crystal size (ex. \(L_c\)), flatness, surface quality, and other properties (e.g., crystal shape). However, for both aspects – both

\begin{figure}
\centering
\includegraphics[width=\textwidth]{fig1.png}
\caption{Overview of experimental setup of CrNb\(_3\)S\(_6\) single crystal located in magnetic field \(H\). The hexagonal layer structure of CrNb\(_3\)S\(_6\) is drawn. Green, red, purple, and yellow represents Nb, Cr, and S, respectively. The Cr atoms are intercalated between NbS\(_2\) layers. The chiral helical axis is the \(c\)-axis. The \(ab\)-plane is magnetically an easy plane. \(H\) is applied perpendicularly to the \(c\)-axis (i.e., parallel to \(ab\)-plane). In this experiment for the submillimeter scale of thin single crystals, the length along the \(c\)-axis (\(L_c\)) was reduced to 10 \(\mu\)m, and the area (\(S_{ab}\)) of the \(ab\)-plane covered 0.120–0.014 mm\(^2\). From the results of previous studies,\(^{20,21}\) \(L_c\) and \(S_{ab}\) cover 120–10 \(\mu\)m and 1.17–0.014 mm\(^2\) area, respectively. The experimental results in the present study suggest the following change in spin texture with increasing \(H\): helimagnet (a: \(H = 0\)) \(\rightarrow\) chiral soliton lattice (CSL) (b: \(0 < H < H_c\)) \(\rightarrow\) forced ferromagnet (c: \(H > H_c\)). In (b), the spins near top surface of the single crystal align ferromagnetically at small \(H\) owing to small DM interaction. \(H_c\) is the critical magnetic field for the forced ferromagnetic state. The main subject of the present study is to investigate how soliton annihilation and soliton nucleation occur in single crystals with a small DM region.}
\end{figure}
annihilation and nucleation, studies on the effect on the soliton number of systematically changing the crystal geometry are still insufficient, particularly in the submillimeter scale. Furthermore, there have been no experimental reports on MR and $M$–$H$ measurements as a function of the area, $S_{ab}$, of the $ab$-plane perpendicular to the $c$-axis.

In the present study, we investigate how the manners of both the soliton annihilation and soliton nucleation changes by decreasing $S_{ab}$ in the case when $L_c$ is maintained at 10 μm. Because the shape of the $ab$-plane is not always rectangular, we focus on $S_{ab}$ in addition to $L_c$ instead of the crystal length on the $ab$-plane. Five crystals were studied with different $S_{ab}$ from 0.120 to 0.014 mm$^2$. All the crystals with $L_c = 10$ μm were obtained from a source crystal as shown in Fig. 2, and have almost the same values for $T_c$ and $H_c$.

To compare the present study with similar previous studies, we name the crystals used in previous studies as a continuation of the sequence A, B, and C. The source crystal is named D and the five submillimeter-sized crystals are named E–I. In total, we have investigated nine single crystals, whose $L_c$ and $S_{ab}$ varied between 120–10 μm and 1.17–0.014 mm$^2$, respectively, as shown in Fig. 3 and Table I. A decrease in $S_{ab}$ increases the ratio of the volume of the nonuniform crystal structure located in the peripheral area. Soliton number can change via release and penetration of solitons through the surface. For instance, in the previous soft X-ray MCD experiment, it has been recognized that solitons dissipate along the direction perpendicular to the $c$-axis. Thus, a change in $S_{ab}$ would influence the process of the change in soliton number. We now focus on both soliton annihilation in $H$-increasing process from a zero field and soliton nucleation in $H$-decreasing process from the forced ferromagnetic state. They become more evident in crystals with $L_c = 10$ μm, compared to those with $L_c \geq 60$ μm. As $S_{ab}$ decreases,

![Fig. 2. Pictures of seven CrNb$_3$S$_6$ single crystals. The thickness, i.e., the length along the c-axis ($L_c$), was first reduced from 75 μm to 10 μm using surface polishing. Two sub-source specimens were obtained after the process of surface polishing. The first series leading to F, G, and H were obtained from E. By machining the second part, the specimen I was cut.](image-url)
soliton annihilation becomes more successive, and avalanche-like soliton nucleation (ASN) occurs at a lower $H$ value. The hysteresis area in the $M$–$H$ curve depends on the $H$ value of ASN and the magnitude of the $M$ jump due to ASN. The crystal shape and size both influence the magnetic properties accompanying the change in the soliton number. Based on the numerical results, we discuss how both soliton release-annihilation and soliton nucleation-penetration relate to changing crystal geometry in CrNb$_3$S$_6$.

II. METHODS

Five submillimeter-sized crystals E–I were prepared from a single source CrNb$_3$S$_6$ crystal, D, with an approximate size $0.75 \times 0.50 \times 0.075$ mm$^3$, as shown in Fig. 2. The crystal D was synthesized in a manner similar to that used for the synthesis of A–C for which the procedure has been described elsewhere.$^4$ The crystal D was then fixed on a silicon substrate using resin. The surface plane is the $ab$-plane, and the thickness direction is the $c$-axis. With precise mechanical polishing, the thickness along the $c$-axis $L_c$ is slowly reduced from 75 to 10 $\mu$m (see Fig. 3). Consequently, in all the crystals labeled E–I, $L_c$ was set to 10 $\mu$m which is equal to the $c$-axis distance of the micro crystal used in the magnetoresistance measurements.$^{15}$ $S_{ab}$ was measured using a stereomicroscope. Two sub-source specimens were cut from D. Crystals F, G, and H were obtained from the first sub-source specimen E. Crystal I, which had a square shape, was prepared by cutting the second sub-source specimen which was placed, along with a wax, between a Si substrate and a cover glass. $T_c$ of CrNb$_3$S$_6$ is determined mainly by intraplane interaction on the ferromagnetic $ab$-plane.$^{9,15}$ Indeed, $T_c$ for crystals E–I was 123 K which was very similar to that of crystal D, suggesting that the intraplane interaction does not depend on either $S_{ab}$ or $L_c$. Thus, surface polishing does not influence the ferromagnetic interaction on the $ab$-plane. $H_c$ for E–I was about half of that of crystal D. It is reasonable to consider that the surface polishing creates slight strain inside the crystals, resulting in small change in the magneto-crystalline anisotropy.

The magnetization of CrNb$_3$S$_6$, $M$, was measured using a superconducting quantum interference device (SQUID) magnetometer. $H$ was applied perpendicularly to the chiral helical axis ($c$-axis), so that $H$ was parallel to the $ab$-plane. The temperature $T$ was set to 5 K, which was sufficiently lower than $T_c$. We thus reproduced conditions similar to the previous experiments conducted on specimens A, B, and C.$^{15,22}$ (details will be provided in Sec. IV). Considering the varying crystal shapes, the effect of demagnetization field was small for $H \parallel c$.

The $H$-dependence of $M$ for a three-dimensional (3D) cubic lattice was calculated using the mean-field (MF) method, and it was compared with the experimental results. In the calculation, we set the maximum soliton number to 209, corresponding to a $c$-axis length of 10 $\mu$m.$^{15}$ The details of the 3D-MF theory has been described in the recent literature on MCD experiments.$^{22}$

We calculated the magnetization dynamics by changing the external magnetic field $H$ to reproduce the $M$–$H$ curve.$^{22}$ The ratio of the variation in $H$ is 20 Oe/ns for the present calculation. The magnetization dynamics obeys the Landau–Lifshitz–Gilbert equation,

$$\frac{d\mathbf{S}}{dt} = -\gamma \mathbf{S} \times \mathbf{H}_{\text{eff}} + \alpha (\mathbf{S} \times \frac{d\mathbf{S}}{dt}),$$

where $\mathbf{S}$ is the local spin, $\gamma$ is the gyromagnetic ratio, and $\alpha (= 0.1)$ is the Gilbert damping coefficient. $\mathbf{H}_{\text{eff}}$ is the effective magnetic field obtained by $\mathbf{H}_{\text{eff}} = -\frac{\partial E}{\partial \mathbf{S}}$, where $E$ is the total energy of the spin system including the ferromagnetic exchange coupling, the DM interaction, and the external magnetic field. We use a 2-dimensional square lattice with the lattice constant $a = 1$ nm. To model the thin specimen, we consider two models for the DM interaction. The first one is the inside-surface in which the DM interaction has a spatial dependence. Because the lattice structure has an inhomogeneity around the

| specimen | A | B | C | D | E | F | G | H | I |
|----------|---|---|---|---|---|---|---|---|---|
| crystal series | Reference 20 | Reference 21 | Reference 21 | source | 1st | 1st | 1st | 1st | 2nd |
| $L_c$ [mm] | 0.12 | 0.11 | 0.06 | 0.075 | 0.01 | 0.01 | 0.01 | 0.01 | 0.01 |
| $S_{ab}$ [mm$^2$] | 1.17 | 0.26 | 0.16 | 0.375 | 0.120 | 0.055 | 0.020 | 0.014 | 0.039 |
| $N_{\text{soliton}}$ | 2500 | 2300 | 1300 | 1568 | 209 | 209 | 209 | 209 | 209 |
| $N_{\text{chain}}$ [$10^{12}$] | 8.1 | 1.8 | 1.1 | 2.6 | 0.84 | 0.38 | 0.14 | 0.10 | 0.27 |
| $T_c$ [K] | 133 | 130 | 130 | 130 | 123 | 123 | 123 | 123 | 123 |
| $H_c$ [kOe] | 1.90 | 2.50 | 2.50 | 2.50 | 1.21 | 1.25 | 1.25 | 1.30 | 1.40 |
| $M_0$ [$10^{-4}$ emu] | ± 0.05 | ± 0.02 | ± 0.02 | ± 0.05 | ± 0.10 | ± 0.05 |
| $\Delta M_{\text{max}}/M_0$ [%] | ± 0.4 | ± 0.1 | ± 0.4 | ± 0.4 | ± 0.4 | ± 0.4 | ± 0.3 | ± 0.3 | ± 0.3 |

$^a$Refer to Fig. 1.
$^b$Refer to Fig. 2.
surface, we set a smaller value for the DM interaction near the surface (DM$_2$) compared to that of inside the sample (DM$_1$), as shown in Fig. 11. To generate Fig. 11, we use a system size of 200 nm $\times$ 200 nm. The exchange energy along the c-axis ($J^1$) was 2.9 K and that of the in-plane direction ($J^2$) was 23.2 K. The energy of DM$_1$ was 0.3 K. The second model of the DM interaction is the anisotropic two-axes model, which has the DM interaction in the two directions. We set the small DM interaction perpendicular both to the c-axis and to the magnetic field direction, as shown in Fig. 12. To generate Fig. 12, we use a system size of 500 nm $\times$ 50 nm.

III. EXPERIMENTAL RESULTS

We show the overall $M$–$H$ curves for submillimeter sized CrNb$_3$S$_8$ crystals, D–I at $T = 5$ K in Fig. 4(a)–(d). $M$ is normalized with saturation magnetization $M_s$. The overall behavior of D is similar to the behaviors of A–C which exhibited both small ferromagnetic type of magnetizing (FMM) with a convex curve (FMM). We show the overall $M$–$H$ curves for submillimeter sized CrNb$_3$S$_8$ crystals, D–I at $T = 5$ K in Fig. 4(a)–(d). $M$ is normalized with saturation magnetization $M_s$. The overall behavior of D is similar to the behaviors of A–C which exhibited both small ferromagnetic type of magnetizing (FMM) with a convex curve $\Delta M \sim 2 \%$. There is no systematic change in $\Delta M_{max}/M_s$ as a function of $S_{ab}$. However, it is also noted that with decreasing $S_{ab}$ as $E \rightarrow F \rightarrow G \rightarrow H$, the $H$ value where a significant step occurs in $M$ ($= \Delta M_{max}$) decreases.

![FIG. 4. $M$–$H$ curve for submillimeter sized CrNb$_3$S$_8$ crystals, D–I, at $T = 5$ K. Details of the sizes of specimens D–I are described in Table I. In (a), the arrows stand for the direction of $H$ change. In (d), the solid curve expresses the MF results determined from the minimum energy conditions. The insets of (a–d) show the representative $M$–$H$ curves near $H_c$, at which hysteresis appears between the $H$-increasing and $H$-decreasing processes.](image)

![FIG. 5. Enlarged $M$–$H$ curve for sub-millimeter sized CrNb$_3$S$_8$ specimens, D–I, during the $H$-decreasing process for $0.5 \leq M/M_s \leq 1$ at $T = 5$ K. The largest discrete change in $M$ ($\Delta M_{max}$) is summarized in Table I. The $H$ value of significant ASN, which exhibits $\Delta M_{max}$, is denoted with a colored line matched to each curve.](image)
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...attribute this sample dependence of the $H$ value with $\Delta M_{\text{max}}$ solely to a difference in $S_{\text{ab}}$, not a difference of shape, based on comparison between E and F. The specimen F, which was derived from E, has a smaller $S_{\text{ab}}$ than E but the shape characteristics do not intrinsically vary within this pair. Finally, we see $L_c$ dependence in the specimens D–I together with specimens A–C. $\Delta M_{\text{max}}/M_i = 1.5$% for D (with $L_c = 75 \mu m$) is at the same level as both A (1.5% with $L_c = 120 \mu m$) and B (0.8% with $L_c = 110 \mu m$). The sample C has the smallest $L_c (= 60 \mu m)$ and the largest $\Delta M_{\text{max}}/M_i = 4.3$% amongst specimens A–D. The average of $\Delta M_{\text{max}}/M_i$ for E–H with $L_c = 10 \mu m$ is 6.6%, slightly larger than 4.3% for C. Herein, we summarize the results of Figs. 4 and 5 together with results on specimens A–D (see Table I). First, the results of specimens A–E show that (1) in general, a decrease in $L_c$ brings about an increase in $\Delta M_{\text{max}}/M_i$, along with the appearance of obvious FMM features. Next, the results of specimens E–H show that (2) the decrease in $S_{\text{ab}}$ while keeping $L_c$ constant does not bring about any systematic decrease in $\Delta M_{\text{max}}/M_i$, whereas the $H$ value with $\Delta M_{\text{max}}$ decreases with decreasing $S_{\text{ab}}$. Finally, (3) for the crystal I with rectangular edges, the small $\Delta M$ just occurs near $H_{c_i}$, and its $\Delta M_{\text{max}}/M_i$ is smaller than that for any of the specimens E–H.

**Semi-reproducibility of the discrete change in $M$ during the $H$-decreasing process.** In this paragraph, reproducibility of the discrete change in $M$ is analyzed. Figure 6 presents the reproducibility of the discrete change in $M$ during the $H$-decreasing process for crystals F, H, and I. In the three runs for F, the first prominent decrease in $M$ appears at almost the same $H (\sim 1.05 \text{kOe})$ over the three runs. Characteristic branches, each of which is a continuous evolution of a metastable state, are labeled as $F_n (n = 0 – 7)$ from the high field side. The processes during $F_0$ and $F_0$ vary between the measurement runs, and all the three runs undergo the $F_1$, $F_3$, and $F_2$ branches. The crystal $H$ with the smallest $S_{\text{ab}}$ exhibits a more reproducible $M – H$ curve, and there is only one prominent discrete step ($\Delta M$) ($H_1 \rightarrow H_3$) at around 0.9 kOe. Both runs trace five characteristic branches as $H_2 \rightarrow H_4 \rightarrow H_2 \rightarrow H_3 \rightarrow H_4$. The overall behavior is reproducible, but the field value for the...
discrete change in $M$ varies between runs. Finally, the square crystal I also presents almost reproducible $M \rightarrow H$ curve tracing $I_0 \rightarrow I_1 \rightarrow I_2 \rightarrow I_3$ over four runs. The first discrete change in $M$ ($I_0 \rightarrow I_1$) appears between 1.21-1.25 kOe (1st: 1.25 kOe, 2nd: 1.21 kOe, 3rd: 1.21 kOe: 4th: 1.24 kOe) in a stochastic manner. In the lower $H$ region, a round $M \rightarrow H$ curve appears. The transition from $I_2$ to $I_3$ occurs at the same $H$ for three runs (1st, 3rd, and 4th), but it is not perfectly reproducible. Looking at the results for the three runs (2nd, 3rd, and 4th), which were conducted successively, a smooth $M$ change in branch $I_3$ as well as that in $I_3$ present high reproducibility.

Avalanche-like soliton nucleation (ASN). A successful example of quantized magnetization is analyzed. Figure 7 clearly shows that in crystal E with the largest $S_{ab}$ among A–I, each discrete change in $M$ is $\Delta M/M_s = 0.48\%$ or its multiple. The smallest step $\Delta M/M_s = 0.48\%$ corresponds to one-soliton nucleation per one chiral chain for samples with $L_c = 10$ $\mu$m. There are some discrete changes, marked by red arrows, within a continuous change in $M$. We attribute the discrete changes in $M$ to soliton nucleation. When the discrete change in $M$ is large and corresponds to nucleation of several solitons, we term it avalanche-like soliton nucleation (ASN). As $H$ decreases across $H_c$, the discrete changes in $M$ corresponding to the single-soliton nucleation occur several times before and after ASN (i.e. 1026, 1042, and 1050 Oe).

Relationship between the hysteresis area and $H_1st$. In the following two paragraphs, the hysteresis area is analyzed. Figure 8 shows the relationship between the hysteresis area and $H$ of the first primary ASN in the $H$-decreasing process ($H_{1st}$). Here, $H_{1st}$ is normalized with respect to $H_c$. For E–H, $H_{1st}/H_c$ is consistent with the $H$ value where a significant step with $\Delta M_{\text{max}}$ appears. $H_{1st}/H_c$ for I is determined to be 0.93, which is larger than the $H$ value (~0.73) with $\Delta M_{\text{max}}$. As can be seen from the data for E–I, the decrease in $H_{1st}$ is related to the increase in the hysteresis area.

$L_c$ and $S_{ab}$ dependences of the hysteresis area and $H_{1st}$. Figure 9 shows that the $L_c$ dependence of both $H_{1st}$ (a) and hysteresis area (b) for A–E, and the $S_{ab}$ dependence of $H_{1st}$ (c) and hysteresis area (d) for E–H. In the present $L_c$ range, $H_{1st}$ is almost independent of $L_c$, as seen in Fig. 9(a). Having $H_{1st}$ close to $H_c$ and a small $\Delta M_{\text{max}}/M_s$ brings about small hysteresis. Figure 9(b) shows that in B–E with various $L_c$ values, the decrease in the hysteresis area with increasing $L_c$ is related to the change in $\Delta M_{\text{max}}/M_s$ rather than to $H_{1st}$. As $S_{ab}$ decreases while $L_c = 10$ $\mu$m, $H_{1st}$ begins to decrease significantly below approximately $S_{ab} \approx 0.03$ $mm^2$ as seen in Fig. 9(c), and simultaneously the hysteresis area increases as seen in Fig. 9(d).

The experimental results described in this section are summarized in Table II.
why in the process of decreasing $S_{ab}$ been assumed to penetrate from the perpendicular to the chiral axis was not considered and solitons have on the chiral sine-Gordon model, the manner of soliton penetration is discussed. The geometrical factor FMM SSA after FMM suggest that this field induced evolution can be regarded as a mean-field type deterministic process apart from the transition process in discrete jumps. One possible interpretation is that the major jumps $H_{ab}$ as $E \rightarrow F \rightarrow G \rightarrow H, H_{ab}$ becomes smaller. Another possibility is that an inhomogeneity along the $ab$-plane near the surface allows a larger value of $H_{ab}$ for samples with larger $S_{ab}$.

Remarks on semi-reproducibility in $H$-decreasing processes. Next, reproducibility is discussed. In crystals H and I, the $M \rightarrow H$ curve during the $H$-decreasing process is almost reproducible. This suggests that this field induced evolution can be regarded as a mean-field type deterministic process apart from the transition process in discrete jumps. One possible interpretation is that the major jumps $H_{ab}$ occur near instability of branches $H_1, I_2$ and slight differences between the runs are due to stochastic processes such as thermal tunnelling between the branches over the small residual energy barrier between them. More complex structures in $M \rightarrow H$ curve for a number of solitons $S_{ab}$ can be attributed to more inhomogeneity along a longer perimeter than in samples H and I.

Successive soliton annihilation (SSA) feature after FMM in $H$-increasing process. Here, we discuss a series of $M \rightarrow H$ data comparing the 3D-MF simulation. The following simulation suggests that the annihilation of the solitons also occurs after FMM. This successive soliton annihilation (SSA) feature after FMM is clearly observed in smaller $S_{ab}$ specimens. Figure 10 illustrates the simulated $M \rightarrow H$ curve for a number of solitons $w = 0$–209, obtained via 3D-MF theory under the periodic boundary condition. $H$ is normalized with the product of the exchange interaction along $c$-axis ($J^H$) and spin value ($S$). The simulated curve for $w = 209$ corresponds to the result for a helical magnet, while that for $w = 0$ corresponds to a forced ferromagnet. The experimental results for the $H$-increasing and $H$-decreasing processes are displayed separately in Fig. 10(a) and 10(b), respectively. We note that the initial magnetizing process during the $H$-increasing process can be well reproduced by the calculation with a fixed-$w$. We assume that after the FMM formation of surface spin at small $H$, the inside spin exhibits the CSL feature. For instance, in E and F, it is reasonable to assume that the CSL with $w = 111$ and 115 are stabilized for $H/J^H S \leq 0.0075$ and $H/J^H S \leq 0.0070$, respectively. The inside spin system tends to keep $w$ constant over a finite $H$ region. When comparing FMM for E and F (covering $S_{ab} = 0.120 - 0.055 \text{mm}^2$), it is reasonable to assume that the change in $w$ due to FMM is not dependent on $S_{ab}$. Further, it is unreasonable to believe that at each of the top and bottom surfaces, the one-quarter of the total volume corresponding to a thickness of $2.5 \mu m$ has structural inhomogeneity. Rather it should be considered that the decrease in the DM interaction near the surfaces has an influence over the deeper region via the metastable states.

### IV. DISCUSSION

Possibility of soliton penetration from ac- or bc-plane. First, the manner of soliton penetration is discussed. The $H_{1d}/H_{1c}$ values in the present study are larger than the theoretical value 0.4 which is based on the chiral sine-Gordon model, where the DM component perpendicular to the chiral axis was not considered and solitons have been assumed to penetrate from the $ab$-plane. The crystal with larger $S_{ab}$ have a longer perimeter, which will provide more chance for solitons to penetrate from the ac- or bc-plane. This scenario can explain why in the process of decreasing $S_{ab}$ as $E \rightarrow F \rightarrow G \rightarrow H, H_{ab}$ becomes smaller. Another possibility is that an inhomogeneity along the $ab$-plane near the surface allows a larger value of $H_{ab}$ for samples with larger $S_{ab}$.

Remarks on semi-reproducibility in $H$-decreasing processes. Next, reproducibility is discussed. In crystals H and I, the $M \rightarrow H$ curve during the $H$-decreasing process is almost reproducible. This suggests that this field induced evolution can be regarded as a mean-field type deterministic process apart from the transition process in discrete jumps. One possible interpretation is that the major jumps $H_{ab}$ occur near instability of branches $H_1, I_2$ and slight differences between the runs are due to stochastic processes such as thermal tunnelling between the branches over the small residual energy barrier between them. More complex structures in $M \rightarrow H$ curve for a number of solitons $S_{ab}$ can be attributed to more inhomogeneity along a longer perimeter than in samples H and I.

Successive soliton annihilation (SSA) feature after FMM in $H$-increasing process. Here, we discuss a series of $M \rightarrow H$ data comparing the 3D-MF simulation. The following simulation suggests that the annihilation of the solitons also occurs after FMM. This successive soliton annihilation (SSA) feature after FMM is clearly observed in smaller $S_{ab}$ specimens. Figure 10 illustrates the simulated $M \rightarrow H$ curve for a number of solitons $w = 0$–209, obtained via 3D-MF theory under the periodic boundary condition. $H$ is normalized with the product of the exchange interaction along $c$-axis ($J^H$) and spin value ($S$). The simulated curve for $w = 209$ corresponds to the result for a helical magnet, while that for $w = 0$ corresponds to a forced ferromagnet. The experimental results for the $H$-increasing and $H$-decreasing processes are displayed separately in Fig. 10(a) and 10(b), respectively. We note that the initial magnetizing process during the $H$-increasing process can be well reproduced by the calculation with a fixed-$w$. We assume that after the FMM formation of surface spin at small $H$, the inside spin exhibits the CSL feature. For instance, in E and F, it is reasonable to assume that the CSL with $w = 111$ and 115 are stabilized for $H/J^H S \leq 0.0075$ and $H/J^H S \leq 0.0070$, respectively. The inside spin system tends to keep $w$ constant over a finite $H$ region. When comparing FMM for E and F (covering $S_{ab} = 0.120 - 0.055 \text{mm}^2$), it is reasonable to assume that the change in $w$ due to FMM is not dependent on $S_{ab}$. Further, it is unreasonable to believe that at each of the top and bottom surfaces, the one-quarter of the total volume corresponding to a thickness of $2.5 \mu m$ has structural inhomogeneity. Rather it should be considered that the decrease in the DM interaction near the surfaces has an influence over the deeper region via the metastable states.

### Table II. Geometrical effects on the change in the soliton number. Ferromagnetic type of magnetizing with convex curve during the $H$-increasing process: FMM, successive soliton annihilation during the $H$-increasing process: SSA, and avalanche-like soliton nucleation during the $H$-decreasing process: ASN.

| geometrical factor | FMM | SSA after FMM | $H$ of significant ASN | $\Delta M/S_0$ at significant ASN |
|-------------------|------|---------------|------------------------|-------------------------------|
| decrease in $L_c$ | enhanced | – | non-systematic | increase |
| decrease in $S_{ab}$ | hardly change | enhanced | decrease | non-systematic |
| rectilinear shape | hardly change | hardly change | increase | decrease |

FIG. 10 Magnetization curves of submillimeter sized CrNb$_2$S$_4$ crystals E–I, analyzed with the 3D-MF theory for a maximal soliton number of 209. The solid curve connected with black plots represents the MF results determined from the minimal energy conditions (already displayed in Fig. 4(d)), where the decrease in soliton number $w$, of as much as $\Delta w = 80$, occurs near $H_e$. However, in specimens E–I, the above response occurs near zero $H$. Afterward, the $w$-value is maintained until half of $H_e$. In H with the smallest $S_{ab}$, $w$ is less maintained. During the $H$-decreasing process, the insertion of the chiral solitons is more significantly influenced by the crystal shape as well as $S_{ab}$.
The further decrease in $S_{ab}$, however, permits SSA after FMM. According to these calculations, the $w$ value of crystal H continuously changes after FMM.

**Comparison with LLG (i) origin of FMM in H-increasing process.** In the following two paragraphs, LLG simulation yields useful information for both soliton release-annihilation and soliton nucleation-penetration. We discuss how both soliton release-annihilation after FMM and ASN from a forced-ferromagnetic state can occur through being related to the surface twisting. Figure 11 shows the $M - H$ curve for a model, in which the inner part has a finite DM$_1$ and the surface parts have DM$_2 = 0$ or 0.1 $\times$ DM$_1$. Here, we note that even if DM$_2 =$ DM$_1 \neq 0$, the calculated $M - H$ curve is not consistent with the MF results determined from the energy minimum condition, because the latter assumes an ab-plane with infinite area. As seen in Fig. 10, in and F, approximately one half of the total spins ferromagnetically aligns at a small $H$. However, it is unreasonable to assume that at each of the top and bottom surfaces, the one-quarter of the total volume has structural inhomogeneity. Thus, as a model, the surface section at each side is taken to be the region corresponding to 10% of the whole area, and the borders between DM$_1$ and DM$_2$ areas are marked with green inverse triangles. The M component along the $H$ direction ($M_z$) is normalized with the saturation magnetization. Figure 11(A)–(H) presents the snapshot of $M_z$ for some $H$ points in the case of DM$_2 = 0.1 \times$ DM$_1$. At an initial zero field, $M_z$ took a value of approximately $-0.15 \times M_s$ unintentionally. This initial $M_z$ in zero field occurs spontaneously (global rotation of spin around c-axis preserves energy). The saturation field $H_s$ is 1.4 kOe. At $H = 0.100$ kOe (only 7% of $H_s$), the magnetic moments at both surface parts align perfectly along the $H$ direction. In the process of increasing $H$ (B $\rightarrow$ C $\rightarrow$ D $\rightarrow$ E $\rightarrow$ F), solitons at the DM$_1$ region dissipate along the direction perpendicular to the c-axis. Meanwhile, in the $H$-decreasing process from greater than $H_s$ (F $\rightarrow$ G $\rightarrow$ H), solitons penetrate into the DM$_1$ region along the c-axis from the borders between the DM$_1$ and DM$_2$ areas. These manners of soliton release-annihilation and soliton nucleation-penetration are consistent with those suggested for the specimens with a small ab-plane. The present specimens are considered as the systems with the surface barriers inside the specimen.

**Comparison with LLG (ii) role of DM component perpendicular to c-axis.** The simulation of Fig. 11 does not exhibit the soliton penetration from the a- or b-axis. From the TEM experiment, it is known that the direction of propagation vector in the helimagnetic structure of CrNb$_3$S$_6$ varies near a hole defect. We can assume that a structural inhomogeneity near the crystal surface can create a dislocation, leading to a change in the direction of propagation vector in the helimagnetic structure. We therefore perform the LLG simulation for a new model, in which the DM vector has a component $S_{ab}$ perpendicular to both $H$ and c-axis in addition to that parallel to the c-axis (DM$_1$). The magnitude of DM$_1$ is assumed to be a half of DM$_1$ so as to enhance the effect of DM$_1'$. Figure 12 shows the $M - H$ curve for the above model. Figure 12(A)–(F) presents snapshots of $M_z$ for some $H$ points in the $H$-decreasing process. Figure 12(B) shows that initial soliton penetration starts from the surfaces perpendicular to the c-axis. However, in (C) with $M_z = 0.8$, the soliton motion along the direction perpendicular to the c-axis is also observed. This tendency of soliton nucleation-penetration from all surfaces is enhanced in the process of (C) $\rightarrow$ (F). The LLG results suggest the importance of the role of DM$_1'$ in the process of the soliton nucleation-penetration in CrNb$_3$S$_6$.

**Remark on shape dependence of hysteresis area.** We also discuss the effect of crystal shape. In Fig. 8, the specimen that has the smallest hysteresis area is not the large specimen E, but the square specimen I. Thus, specimen I with the most regular shape exhibits smooth transfer from the forced ferromagnetic state to the CSL state, and has the smallest irreversibility between soliton annihilation and soliton nucleation. This reveals that the perimeter length is also the important geometrical factor in determining $H_{1st}$, suggesting the geometrical feature characteristic of topological magnets.

**Similarities and differences between chiral-soliton penetration and superconducting-vortex penetration.** Finally, we summarize chiral-soliton penetration comparing it with superconducting-vortex penetration. The penetration of chiral solitons is caused by a disappearance of the energy barrier due to competition between outward- and inward-forces near surfaces, in a similarly way.

FIG. 11. LLG simulation for the surface-inner model. $M_z$ has the magnetization along the $H$ direction perpendicular to the c-axis. The inner region has DM$_1$, while the region of surface perpendicular to the c-axis has DM$_2 = 0$ or 0.1$\times$DM$_1$. (A)–(H) presents the snapshot of $M_z$ for some $H$ points in the case of DM$_2 = 0.1 \times$ DM$_1$. The x and y-axes defining the plane perpendicular to the $H$ direction (c-axis) are drawn with broken lines only in (A). The borders between DM$_1$ and DM$_2$ areas are marked with green inverse triangles. In (A), there are 23 solitons. Blank arrows in (E) and (H) stand for the direction of soliton release and that of soliton penetration, respectively.
to that of superconducting vortices.\textsuperscript{12} In the former case, the outward force stems from exchange interaction and Zeeman energy, while the inward force does from DM interaction. In the latter case, the outward force is due to attraction between vortex inside and the image vortex outside. The inward force comes from interaction between vortex and supercurrent (Meissner current) near the surface.\textsuperscript{12} In superconductors, the density of vortices varies continuously as a function of $H$ because of inhomogeneity of $H$ near surfaces. However, the density of chiral solitons is discontinuous along with the disappearance of the energy barrier. The chiral soliton, being a plane-type object is not influenced with pinning centers less than the superconducting vortex, which is a line-type object.

V. CONCLUSION

We measured the magnetization curve of five submillimeter-sized single crystals of CrNb$_3$S$_6$ with a $c$-axis length of 10 $\mu$m, where the area of $ab$-plane was in the range 0.120–0.014 mm$^2$. These results were compared with those for four larger crystals. In the submillimeter-sized crystals, multiple solitons can escape near zero $H$, while over a finite $H$ range below the critical field $H_c$ for the forced ferromagnetic state, the soliton number is almost unchanged. The magnetic behavior was quite different from the MF theory determined under minimum energy conditions. The difference between the experimental results and the aforementioned MF theory was more clearly observed during the $H$-decreasing process from $H_c$, resulting in the appearance of significant hysteresis in the experimental results.

The magnetic properties of the submillimeter-sized crystals were characterized as follows: (1) With a small magnetic field, ferromagnetic type of magnetizing (FMM) with a convex curve occurs in the region with a small DM vector, (2) The crystals with a smaller surface exhibit successive soliton annihilation after FMM. (3) The crystals with a smaller surface exhibit a large jump in $M$ due to avalanche-like soliton nucleation-penetration at lower magnetic field, because the decrease in perimeter length with decreasing $S_{ab}$ disturbs the motion of soliton penetration along the direction perpendicular to the $c$-axis. (4) In the single domain level of $L_c$, for the full field scanning from 0 $\rightarrow$ above $H_c$ $\rightarrow$ 0, prominent magnetic hysteresis appears, and its magnitude is related to the magnetic field of the first discrete change in $M$ due to significant avalanche-like soliton nucleation-penetration. (5) The crystal shape (i.e., perimeter length) is also a physical factor which affects the energy barrier for soliton penetration. Thus, in the submillimeter-sized crystals with the same $L_c$, the shape and size both are key factors to determine the magnetic properties accompanying the change in the soliton number.

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