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Spontaneous Magnetostriction Effects in Monoaxial Chiral Magnets $CrNb_3S_6$

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We identified the spontaneous magnetostriction effects in the chiral magnet $CrNb_3S_6$ with the monoaxial type of Dzyaloshinskii-Moriya vector. Through the powdered x-ray diffraction experiments, a prominent magnetostriction effect at the earth field was observed. The unit-cell volume remains almost constant below 170 K in the paramagnetic region, where the atomic positions of Nb and S show characteristic changes. It is considered as the so-called Invar effect. Below the magnetic ordering temperature of 127 K, the lattice constants *a* and *c* exhibit an opposite temperature dependence.

1 Introduction

The Dzyaloshinskii-Moriya (DM) interaction arises from a combination of second-order perturbation of the spin-orbit coupling (SOC) and the exchange interaction [1, 2]. The competition between the DM and exchange interactions stabilizes a long-wavelength helimagnetic order.

In a chiral space group without any rotoinversion symmetry, crystallographic chirality allows the appearance of the DM interaction. The magnetostriction in chiral magnet has been investigated in in a B20 (cubic) type of MnSi [3] (forced magnetostriction) and $Fe_{1-x}Co_xSi$ (spontaneous magnetostriction) [4]. This study investigates the magnetostriction phenomena in a typical hexagonal chiral magnet, $CrNb_3S_6$, considering lattice deformation on the atomic scale. $CrNb_3S_6$ has a stacked structure of hexagonal NbS_2 layers. The insertion of Cr^{3+} between the NbS_2 layers precludes the inversion symmetry; thus, the material crystallizes in the noncentrosymmetric hexagonal space group $P6_322$ [5]. The magnetic ordering temperature T_c at zero magnetic field is 127 K. The change in lattice parameters is related to the change in the local structural symmetry of CrS_6 octahedron with a magnetic Cr^{3+} ion.

2 Experiment

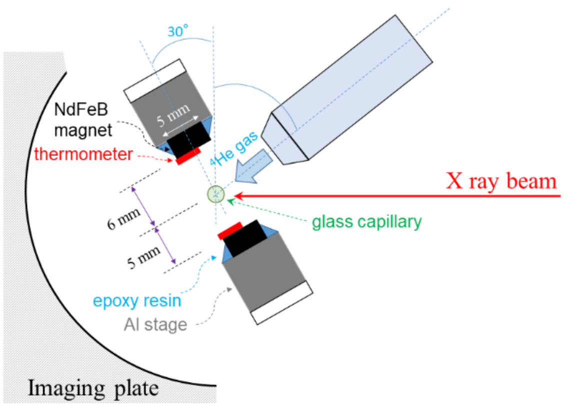

Fig. 1: Setup of XRD experiments at H = 1.2 kOe. H was obtained using two facing NdFeB magnets with opposite magnetizations [6].

We performed powder x-ray diffraction (XRD) analyses as a function of temperature T using a synchrotron radiation system with a cylindrical imaging plate of BL-8B at the Photon Factory at the Institute of Materials Structure Science of the High Energy Accelerator Research Organization. The energy of the incident x rays was 16 keV. In addition to the experiments at zero magnetic field, the magnetic field H of 1.2 kOe was applied using two permanent magnets (NeoMag, N48H) with a remanence of 13.8 kG as shown in Fig.1[6]. The remanence of a NdFeB magnet depends on its temperature. The temperature of the magnets was measured using a K-type chromel-alumel thermocouple. During the XRD measurements, the sample temperature T was increased from 92.8 to 294.7 K so that the temperature of the magnets was held at 289.2–298.2 K to maintain H. Consequently, the H value at the sample position is calculated at room temperature. The diffraction patterns for the powder sample were analyzed by Rietveld refinement. To evaluate the lattice parameters, we used 14 (*hk*3) and 11 (*hk*2) spots for the H // *ab* setup and 4 (21*l*), 4 (20*l*), and 7 (11*l*) spots for the H // *c* setup. The change in the lattice parameter will be discussed in the process of decreasing T.

3 Results and Discussion

Figures 2(a) and 2(b) show the lattice parameters *a* and *c*, their ratio *c/a*, and the unit cell volume V of $CrNb_3S_6$ at H = 0 [6]. Ordinal thermal shrinkage appears in the paramagnetic region above 170 K. The development of magnetization at a finite H owing to the short-range order is dominant below 170 K. For 130–170 K, all of the lattice parameters change very little. It is a type of Invar effect due to competition between the thermal expansion and magnetic shrinkage. Here, *c/a* was kept nearly constant [see Fig. 2(b)]. For T < 130 K, the *a* axis expands by 0.02%, and the *c* axis becomes 0.33% shorter, resulting in a prominent change in *c/a* [see Fig. 2 (b)]. Consequently, V tends to have an almost constant value (indeed, slightly increased), even for T < 130 K. There, the increase in volume due to the expansion of the easy plane is offset by the decrease in V due to the shrinkage along the hard axis (*c* axis). Thus, this behavior below T_c, which is opposite to thermal shrinkage, is identified as spontaneous “magnetostriction” associated with the helimagnetic order.

According to the careful Rietveld analyses, the *z* coordinate of Nb at the 4*f* site, Nb(4*f*)$_z$, exhibits characteristic changes at 130, 180, and 260 K, where the change at 130 K is associated with the helimagnetic order. Under hydrostatic pressure, Nb(4*f*)$_z$ approaches 0.5 for Nb at the 2*a* site [7]. The thermal shrinkage for $T > T_c$ and hydrostatic compression have similar effects on Nb(4*f*)$_z$. Thus, the symmetry of the CrS_6 octahedron approaches that of the regular octahedron in both cases. Herein, it is noted that all of the atomic coordinates of S (S_x, S_y, and S_z) exhibit a change at T_c, similar to Nb(4*f*)$_z$. The characteristic temperatures that determine the changes in S_x, S_y, and S_z except for T_c are not uniform. Of the three atomic coordinates of S, the characteristic temperatures for S_z, such as 130, 190, and 240 K, are particularly close to those in Nb(4*f*)$_z$, suggesting interlocking motion between Nb(4*f*) and S atoms.

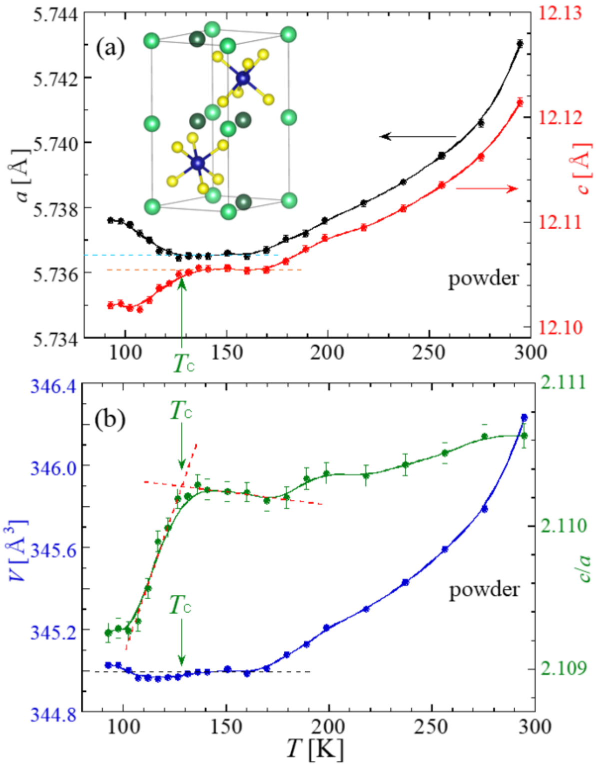

Fig. 2: Lattice parameters *a*, *c*, *V*, and *c/a* of $CrNb_3S_6$ at $H = 0$ as estimated by Rietveld analysis. The data for *a* and *c* are shown in (a), and those for *V* and *c/a* are shown in (b) [6]. Green arrows indicate the T_c value confirmed by ac magnetization at zero field.

Figure 3 shows the lattice parameters *a*, *c*, and *V* estimated experimentally using the single crystal at $H = 0$ and $H = 1.2$ kOe for *H//ab* [Figs. 3(a)–3(c)] and *H//c* [Figs. 3(e)–3(g)]. For *H//ab*, the increase in *a* and decrease in *c* below T_c at $H = 0$ change to an decrease in *a* and a slight increase in *c* [Figs. 3(a) and 3(b)]. The changes in the unit cell as *T* is decreased from *T*c to 90 K are shown in Fig. 3(d). The lattice constant changes notably with respect to the *H* direction against the easy plane, and it is understood with an *H*-induced magnetostriction depending on the square of magnetization. Consequently, *V* does not change greatly below *T*c between *H* =0 and *H//ab* [Fig. 3(c)], although the scenario for *H//ab* is different from that of the spontaneous magnetostriction at *H* = 0.

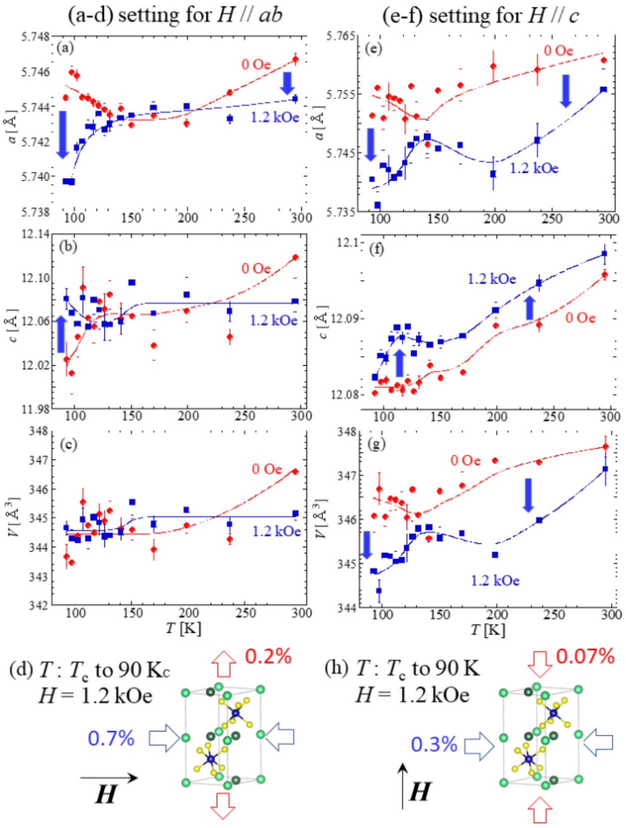

Fig. 3: *T* dependence of lattice parameters estimated in experiments using a single crystal at *H* = 1.2 kOe for (a)–(c) *H* // *ab* and (e)–(g) *H* // *c*. (d) and (h) The behavior between 90 K and *T*c. For reference, the results at *H* = 0 are also shown.

We observed spontaneous magnetostriction in $CrNb_3S_6$. The magnetostriction appears as the so-called Invar effects in two *T* regions, the paramagnetic (130–170 K) and helimagnetic (below T_c) regions. The former Invar effect originates from the changes in the symmetry of the CrS_6 octahedron, and there both lattice constants *a* and *c* do not change, resulting in no change in the unit-cell volume. The unit-cell volume remains constant also below T_c, where the shrinkage of the unit cell along the *c* axis and elongation of the *ab* plane are in competition. The Invar effect below T_c originates in the ferromagnetic alignments on the *ab* planes. This spontaneous magnetostriction was modified by a magnetic field *H*, and the manner depended on the *H* direction. Thus, in $CrNb_3S_6$ with SOC, a prominent magnetostriction effect was observed, and the actual effects are thought to depend on both the magnitude and direction of *H*.

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