

Magnetic ground state dependent magnetostriction effects on chiral magnet CrNb₃S₆Masaki MITO^{1,*} and Takayuki TAJIRI²¹ Kyushu Institute of Technology, Kitakyushu 804-8550, Japan² Fukuoka University, Fukuoka 814-0180, Japan

The mono-axial chiral magnet CrNb₃S₆ has multiple magnetic structures, such as helimagnetic, first chiral soliton lattice (CSL-1), second CSL (CSL-2), CSL-2 with irreversibility, and forced ferromagnetic phases below the magnetic ordering temperature (T_c). We performed powder x-ray diffraction analyses to investigate the effects of magnetic field and temperature on the magnetostriction. The temperature dependence of the lattice constants reveals that below T_c , the magnetostriction depends on the magnetic structure.

1 Introduction

Crystallographic chirality can be converted into a spin system by the Dzyaloshinskii-Moriya (DM) interaction deviated from the spin-orbit coupling (SOC) [1,2]. The DM interaction is allowed in a chiral space group without any rotoinversion symmetry. Further competition between the DM and exchange interactions results in the helimagnetic (HM) structure as the magnetic ground state at zero dc magnetic field (H). Magnetostriction (MS), which often appears in ferromagnets [3–5], also occurs in the magnetic materials with the DM interaction [6–10] because of a strong magnetostructural correlation.

In a prototype of the chiral magnet CrNb₃S₆ with a monoaxial DM vector, two types of MS have already been observed: (1) spontaneous MS due to the exchange interaction and SOC at zero H [6] and (2) paramagnetic (PM) MS due to the SOC and Zeeman energy at room temperature [7]. In CrNb₃S₆, the SOC originates in the hybridization between Cr and Nb [11]. These MS phenomena accompany the change in the interatomic distance between Cr and Nb. Thus, these MS effects in CrNb₃S₆ [6,7] are examples of SOC-induced MS [8–10]. However, MS in CrNb₃S₆ has not been in detail investigated over wide H and temperature T ranges.

2 Experiment

We performed powder XRD analyses at various temperatures using a synchrotron radiation XRD system with a cylindrical imaging plate at the Photon Factory at the Institute of Materials Structure Science, High Energy Accelerator Research Organization [12]. The energy of the incident x-rays was 16 keV. To produce the maximum H value of 2.2 kOe, two facing NdFeB magnets with remanence value of 13.8 and 14.5 kG were placed in the diffractometer [6]. Because the remanence of the NdFeB magnets depends on temperature, the temperature of the magnets was measured using a K-type chromel-alumel thermocouple. All XRD measurements were conducted under increasing temperature in the range 92.8–294.7 K, and the temperature of the magnets was maintained within 289.2–298.2 K.

3 Results and Discussion

Figures 1(a), 1(b), and 1(c) show the lattice constants a , c , and the unit cell volume V for CrNb₃S₆, respectively, at $H = 0, 0.71, 1.23,$ and 2.16 kOe [13]. In the PM region ($130 \text{ K} < T < 170 \text{ K}$), all of the lattice parameters change very little, suggesting a type of Invar effect due to competition between thermal expansion and magnetic

shrinkage. In the HM region ($T < 130\text{K}$), shrinkage along the a -axis and elongation along the c -axis occur simultaneously, so that V is almost constant. Thus, the decrease in volume on the easy plane is canceled by the increase in volume along the hard axis. In the present study, we consider two phenomena: (1) the PM Invar effect for $T > T_c$ and (2) MS for $T < T_c$ depending on the magnetic ground states.

First, we investigated the PM Invar effect by observing the effect of H on V [Fig. 1(c)]. The Invar effect at the unit-cell level at $T > T_c$ was also observed at $H = 0.71$ kOe, where the T range is almost the same as that at $H = 0$ Oe. The constant V characteristic of the Invar effect is not observed at $H = 1.23$ and 2.16 kOe. At $H = 0.71$ kOe, the surface is FFM and the interior is mostly HM. The HM spin alignment is assumed to stabilize the Invar effect due to competition between thermal expansion and magnetic shrinkage. Further application of H reduces the stability of HM structure, such that, for $T > T_c$, thermal expansion should exceed magnetic shrinkage at $H = 1.23$ and 2.16 kOe.

Next, the MS below T_c , depending on various magnetic ground states, was investigated by observing the T dependence of the unit cell parameters below T_c at various H values. According to the trend of the lattice constant on the ferromagnetic plane parallel to the magnetic anisotropy vector [$a(T)$ in Fig. 1(a)], the MS at $T < 103$ K can be divided into two categories; the first occurs at 0 Oe (HM) and 0.71 kOe (CSL-1), and the second occurs at 1.23 kOe (CSL-2) and 2.16 kOe [CSL-2 with irreversibility, CSL-2(NL)]. The value of a at $T < 103\text{K}$ is almost the same at $H = 0$ and 0.71 kOe. At $T = 103$ K, a discretely changes at $H = 0.71$ kOe, and for $107 \text{ K} < T < T_c$, the T dependence of a at $H = 0.71$ kOe is similar to that at $H = 2.16$ kOe. At $T < 110\text{K}$ and $H = 2.16$ kOe, the T dependence of a is opposite to that at $H = 0.71$ kOe. An increase in a due to MS at $T < T_c$ was observed at 1.23 kOe; it stabilizes the CSL-2 state at $T < 120\text{K}$ and FFM state in narrow T region of $120 \text{ K} < T < T_c$. The large increase in a during warming occurs in the CSL-2 phase. Figure 1(b) shows the T dependence of the lattice constant along the hard axis, $c(T)$; the increase at $T < T_c$ and $H = 0$ Oe changes to a discrete increase at 103 K and $H = 0.71$ kOe. Considering the behavior at 93 K, the HM and CSL phases should be placed in the same category, as suggested in the analysis of a , whereas CSL-2 and CSL-2(NL) should be categorized separately. Other features have already been observed in the results for a . The unit-cell volume V is almost constant at $T < T_c$ and

$H = 0$ Oe and just below 103 K at $H = 0.71$ and 2.16 kOe. However, at $H = 1.23$ kOe, the volume exhibits thermal expansion. Because the change in c is much larger than that in a , the T dependence of V below T_c is qualitatively the same as that of c . In addition to a , c , and V , the ratio of c and a , c/a , provides information on the uniaxial distortion of the hexagonal unit cell in the two MS phenomena mentioned above [13].

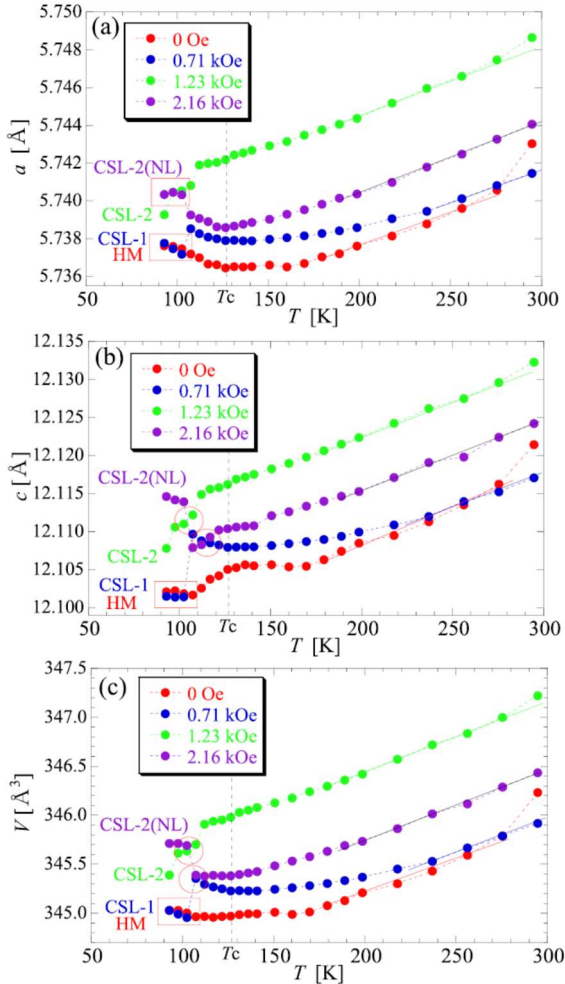


Fig. 1: Lattice parameters a (a), c (b), and V (c) at $H = 0$, 0.71, 1.23, and 2.16 kOe [13]. The position of T_c is represented with a broken line. The qualitative behavior above 200 K, characterized with a straight line, in all of a , c , and V is independent of H . The T region categorized as a group is presented with red rectangle or circle.

Acknowledgement

We thank many collaborators, Dr. Y. Kousaka, Prof. J. Akimitsu, Prof. J. Kishine, and Prof. K. Inoue, in the present study. This work was also supported by the Centre for Chiral Science at Hiroshima University (the MEXT program for promoting the enhancement of research universities, Japan).

References

[1] I. E. Dzyaloshinskii, J. Phys. Chem. Solids **4**, 241 (1958).
 [2] T. Moriya, Phys. Rev. **120**, 91 (1960).
 [3] A. Katsuki and K. Terao, J. Phys. Soc. Jpn. **26**, 1109 (1969).

[4] M. Shimizu, J. Magn. Magn. Mater. **20**, 47 (1980).
 [5] S. Khmelevskiy and P. Mohn, Phys. Rev. B **69**, 140404(R) (2004).
 [6] T. Tajiri, M. Mito, Y. Kousaka, J. Akimitsu, J. Kishine, and K. Inoue, Phys. Rev. B **102**, 014446 (2020).
 [7] M. Mito, T. Tajiri, Y. Kousaka, Y. Togawa, J. Akimitsu, J. Kishine, and K. Inoue, Phys. Rev. B **105**, 104412 (2022).
 [8] M. Matsunaga, Y. Ishikawa, and T. Nakajima, J. Phys. Soc. Jpn. **51**, 1153 (1982).
 [9] S. Wang, Y. Hu, J. Tang, W. Wei, J. Cong, Y. Sun, H. Du, and M. Tian, New J. Phys. **21**, 123052 (2019).
 [10] K. Shimizu, H. Maruyama, H. Yamazaki, and H. Watanabe, J. Phys. Soc. Jpn. **59**, 305 (1990).
 [11] M. Mito, H. Ohsumi, T. Shishidou, F. Kuroda, M. Weinert, K. Tsuruta, Y. Kotani, T. Nakamura, Y. Togawa, J. Kishine, Y. Kousaka, J. Akimitsu, and K. Inoue, Phys. Rev. B **99**, 174439 (2019).
 [12] A. Fujiwara, K. Ishii, T. Watanuki, H. Suematsu, H. Nakao, K. Ohwada, Y. Fujii, Y. Murakami, T. Mori, H. Kawada, T. Kikegawa, O. Shimomura, T. Matsubara, H. Hanabusa, S. Daicho, S. Kitamura and C. Katayama, J. Appl. Cryst. **33**, 1241 (2000).
 [13] M. Mito, T. Tajiri, Y. Kousaka, J. Akimitsu, J. Kishine, and K. Inoue, Phys. Rev. B **107**, 54427 (2023).

* mitoh@mns.kyutech.ac.jp