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Paramagnetic Magnetostriction in Monoaxial Chiral Magnets $CrNb_3S_6$

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We identified the paramagnetic magnetostriction effects in the chiral magnet $CrNb_3S_6$ with the monoaxial type of Dzyaloshinskii-Moriya vector. Through the powdered x-ray diffraction experiments, we observed a prominent magnetostriction effect at room temperature much higher than the magnetic ordering temperature of 127 K. The present study clarified that the symmetry of the CrS_6 octahedron is sensitive to magnetic field even at room temperature. The paramagnetic spin-orbit coupling should induce the distortion of CrS_6 octahedron, resulting in the changes in Cr-Nb(4*f*) distance via the change in the hybridization between Cr-a_{1g} and Nb-4d_{z^2} orbitals.

1 Introduction

In the "magnetostriction (MS) effect" due to strong magnetostructural correlation, the volume of the unit cell changes when the spin system transforms to a certain magnetically ordered state. The MS in the paramagnetic region is rarely observed, and it is related to the local moments in the paramagnetic state. On the other hand, the orbital angular momentum L presents the exotic physical phenomena via the spin-orbit coupling (SOC). The SOC also brings about magnetocrystalline anisotropy, which can enhance the MS effects.

Given these backgrounds, the MS effect has been observed in chiral magnets, where the Dzyaloshinskii-Moriya (DM) interaction can be permitted [1-4]. In 2020, in a typical monoaxial-DM type of chiral helimagnet $CrNb_3S_6$ with the magnetic ordering temperature T_c of 127 K, spontaneous MS was observed at the boundary between the paramagnetic and helimagnetic states [4]: The change in atomic position, reflecting the existence of local distortion, occurs over a wide temperature range up to room temperature. Futhermore, $CrNb_3S_6$ has a strong magnetocrystalline anisotropy on the *ab* plane. The forced MS effect was also observed at the strength of a magnetic field H of 1.2 kOe, and there a small change in the unit cell volume survives even at room temperature (RT) [4]. Thus, we wanted to investigate the paramagnetic MS in detail. In this study, to elucidate whether the paramagnetic MS occurs in the microscopic level, we performed the x-ray structural analysis experiments for $CrNb_3S_6$ as a function of H at RT [5].

2 Experiment

Single crystals of $CrNb_3S_6$ were synthesized via a chemical vapor transport method [6]. We performed XRD analyses at RT, 299.2 K, using a synchrotron radiation XRD system with a cylindrical imaging plate at BL-8B [7]. The energy of the incident x rays was 16 keV. Two facing NdFeB magnets with remanence of 13.8 kG and 14.5 kG, were located in the diffractometer [4]. The H values were varied by changing the distance between the sample and NdFeB magnets. Then, the H value was always reduced down to zero before constructing new configuration of NdFeB magnets. The diffraction spots for single crystals were observed in the vibration mode of every 1° for ±3°. On the basis of the changes in the spot pattern, the changes in the lattice constants were evaluated. The atomic positions in the unit cell did not become parameters in the analyses.

3 Results and Discussion

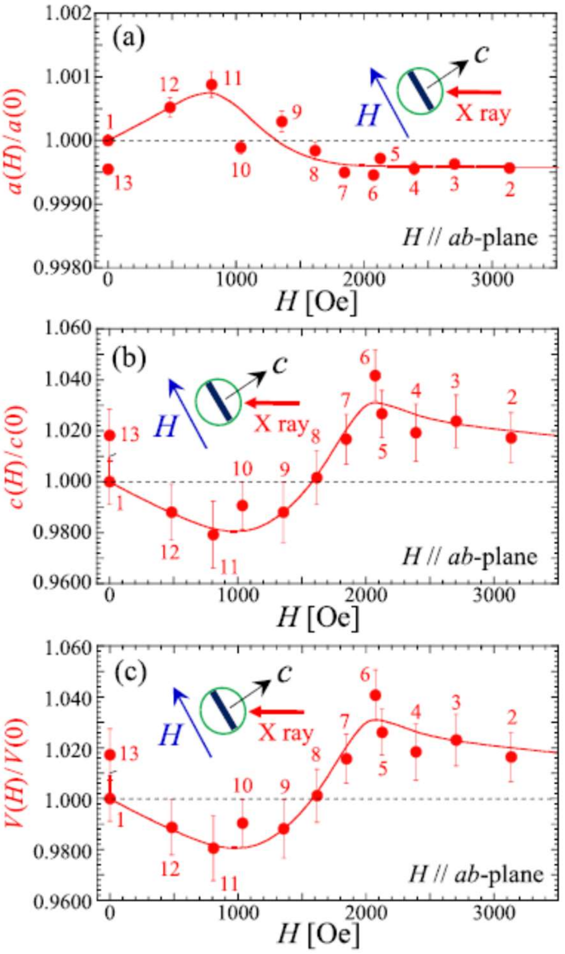

Fig. 1: H dependence of lattice constants, a (a), c (b), and V (c) of $CrNb_3S_6$ at RT for $H \perp c$ (H // ab plane) [5]. The numbers adjacent to the symbols denote the sequence of the measurement.

$CrNb_3S_6$ has the ferromagnetic layer (*ab* plane) with the easy-plane anisotropy, which stacks along the chiral axis (*c* axis). The comparison between MS for $H \perp c$ and that for *H* // *c* is a matter of the greatest concern. Figures 1(a) and 1(b) show the *H* dependence of normalized lattice constants *a* and *c* with those at *H* = 0 for *H* ⊥ *c* at RT. The lattice expands toward the *a*-axis at small fields and the expansion becomes 0.09% at 0.8 kOe. At the same time, the *c* axis shrinks, which in turn decreases the unit cell volume *V* as shown in Fig. 1(c). With further increasing *H*, *a* starts to decrease. It becomes smaller than that at *H* = 0 and the shrinkage reaches 0.04%. Conversely, *c* tends to increase around 1 kOe and it exhibits the 4% expansion at 2 kOe. The 2% expansion remains even above 2.4 kOe, because the value of *c*(*H*)/*c*(0) keeps almost to 1.020. The magnitude of the aforementioned forced MS is much larger than that of the spontaneous magnetostriction below T_c [4]. Thus, the relative change in *c* is 50 times larger than that in *a*. We assume that the structural change along the *c*-axis originates from the change in *c* along the hybridization between z^2 orbital of Nb(4 *f*) and delocalized a_{1g} orbital of Cr. The change in the unit cell volume *V* is closely correlated with the change in *c*. Indeed, their baselines in both *c* and *V* exhibit negative slope with respect to *H* in the considered *H* region. Next, we investigate whether the aforementioned lattice change occurs in another *H* orientation.

Figures 2(a)–2(c) show the *H* dependence of lattice parameters *a*, *c*, and *V* for *H* // *c*. In the figures, *a* and *c* do not exhibit meaningful changes with respect to the change in *H*. Furthermore, *V* maintains a constant value. The comparison among two *H* orientations reveals that the MS occurs when *H* is applied in a direction parallel to the magnetic easy plane. It is natural to identify any important factors for producing the easy-plane type of magnetic anisotropy. The present MS at RT is observed sufficiently higher than T_c. The order of the easy-plane anisotropy (~1K) as well as the DM interaction (~1K) is much smaller than the thermal energy at RT [8]. Hence, as a possible scenario, it may be worth considering that the paramagnetic MS originates directly from the change in the orbital angular momentum *L*.

In this study, we observed magnetostriction in $CrNb_3S_6$ at room temperature. This intrinsically differs from forced MS due to the SOC near T_c. The distortion of the CrS_6 octahedron accompanies a change in the hybridization between Cr-a_{1g} and Nb-4d_{z^2} orbitals, resulting in the change in the Cr-Nb(4*f*) distance. The resultant movement of Nb(4*f*) along the *z* direction leads to a change in the unit cell volume. We consider that the present paramagnetic MS would originate from significant SOC in $CrNb_3S_6$

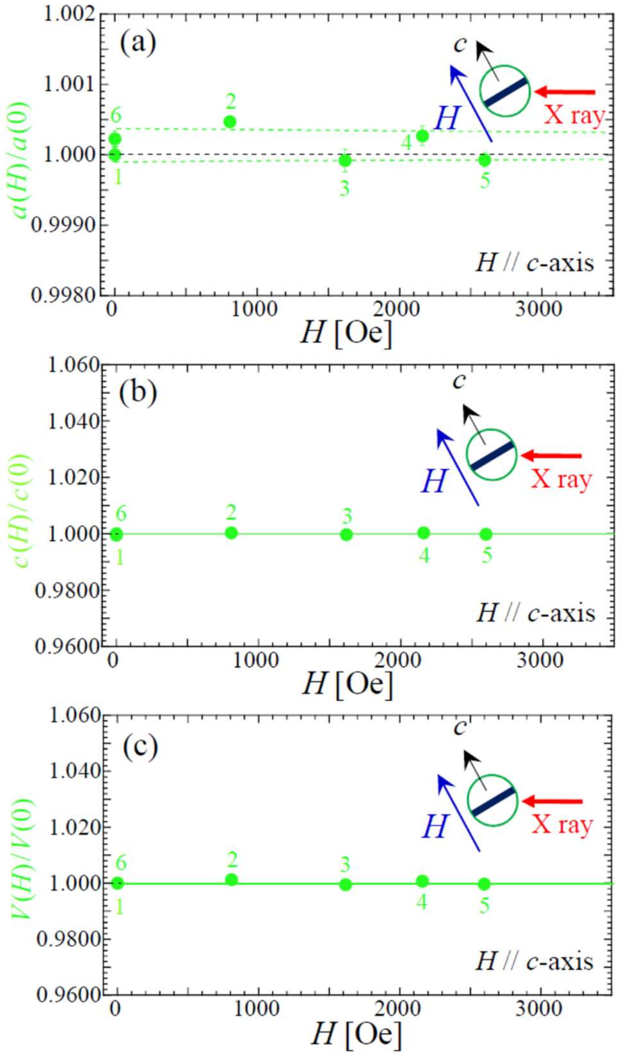

Fig. 2: *H* dependence of lattice constants, *a* (a), *c* (b), and *V* (c) of $CrNb_3S_6$ at RT for *H* // *c* [5]. The numbers adjacent to the symbols denote the sequence of the measurement.

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