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Helimagnetic and ferroquadrupole orderings in a chiral magnet $DyNi_3Ga_9$

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1 Introduction

In magnetic materials with chiral crystal structures without both an inversion center and a mirror plane, nontrivial emergent spin structures are often realized, which have been attracting wide interests. In MnSi with a cubic B20 structure, for example, a hexagonal lattice of swirling spin textures, which is called a skyrmion lattice, is realized in magnetic fields [1]. In $Cr_{1/3}NbS_2$, a monoaxial hexagonal helimagnet, a periodic lattice of twisted spin structures appears by applying a magnetic field perpendicular to the helical axis [2]. This is called the chiral soliton lattice and have recently been identified to occur also in rare-earth $Yb(Ni,Cu)_3Al_9$ [3]. In contrast to the long period of the helimagnetic structures in the 3d systems with ferromagnetic interactions, the RKKY-type antiferromagnetic interaction is dominant in the 4f systems, resulting in the short pitch of the helimagnetic structure. The degeneracy of the helicity is lifted by the weak Dzyaloshinsky-Moriya (DM) interaction.

$DyNi_3Ga_9$, isostructural to $YbNi_3Al_9$, exhibits successive phase transitions at T_1=10 K, T_2=9 K, and at T_3=6.5 K [4]. One of the interests in $DyNi_3Ga_9$ is in the relation of these transitions with the large quadrupole and magnetic moments of Dy^{3+} with J=15/2, which is contrasting to the well isolated doublet system of $Yb(Ni,Cu)_3Al_9$. Elastic constant measurement shows that the transition at T_1 is of ferroquadrupole order. At the same time, an incommensurate magnetic order is observed by neutron diffraction between T_2 and T_1. The purpose of this experiment is to clarify the details of the successive magnetic transitions.

2 Experiment

Resonant X-ray diffraction experiments have been performed at the beamline 3A. The single crystalline sample, with a mirror-polished c-plane surface, was set in the 8 Tesla vertical-field cryomagnet so that the a*-c* horizontal scattering plane was investigated with magnetic fields applied along the b-axis. We used X-ray energies around the L_3 absorption edge of Dy.

3 Results and Discussion

We scanned along the (0, 0, L), (1, 0, L), and (-1, 0, L) lines in the reciprocal space at 9 K corresponding to phase-I (T_2<T<T_1), and found Bragg peaks on the (±1, 0, L) lines at $\boldsymbol{Q}$-vectors corresponding to the incommensurate wave vector $\boldsymbol{q}$=(0, 0, 0.43), commensurate wave vectors $\boldsymbol{q}$=(0, 0, 0.5), and $\boldsymbol{q}$=(0, 0, 1.5). We call these peaks as IC, C1, and C2, respectively. These peaks were not observed on the (0, 0, L) line, indicating that the magnetic moments at the Dy-1 and Dy-2 sites on the same honeycomb layer. All peaks showed resonance enhancements at 7.798 keV.

We next performed a detailed scan at the resonance energy of 7.798 keV along (1, 0, L) with polarization analysis using a PG crystal analyzer. Fig. 1 shows the temperature dependences of the integrated intensities of the commensurate (C1 and C2) and incommensurate (IC) peaks. With decreasing temperature below T_1, the IC intensity first increases rapidly more than the C1 and C2 peaks. On further decreasing temperature, the IC intensity begins to decrease, and at the same time the C1 peak begins to develop more significantly. This result shows that the incommensurate helimagnetic structure is preferred just below T_1, where the ordered magnetic moment is still tiny and is not affected by the anisotropy energy due to the crystal field. When the magnitude of the ordered magnetic moment develops with decreasing temperature, the commensurate structure is more preferred than the incommensurate one because of the anisotropy effect. In phase I (T_2<T<T_1), the intensities of C1 and C2 peaks increase and decrease simultaneously, indicating that these two peaks correspond to the identical structure. Probably the C2 peak is the third harmonic of C1.

Below T_2=8 K in this experiment, the IC peak completely disappear, but the C1 and C2 peaks survive weakly. The main ordered structure in phase-II below T2 is the canted antiferromagnetic order described by $\boldsymbol{q}$=(0,0,0) with a ferromagnetic moment. Also by applying a magnetic field, these peaks of IC, C1, and C2 at zero field

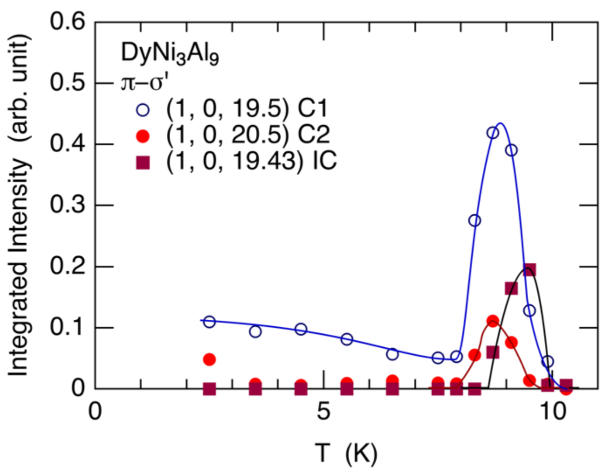

Fig. 1: Temperature dependences of the integrated intensities of the three Bragg peaks at the resonance energy of 7.798 keV.

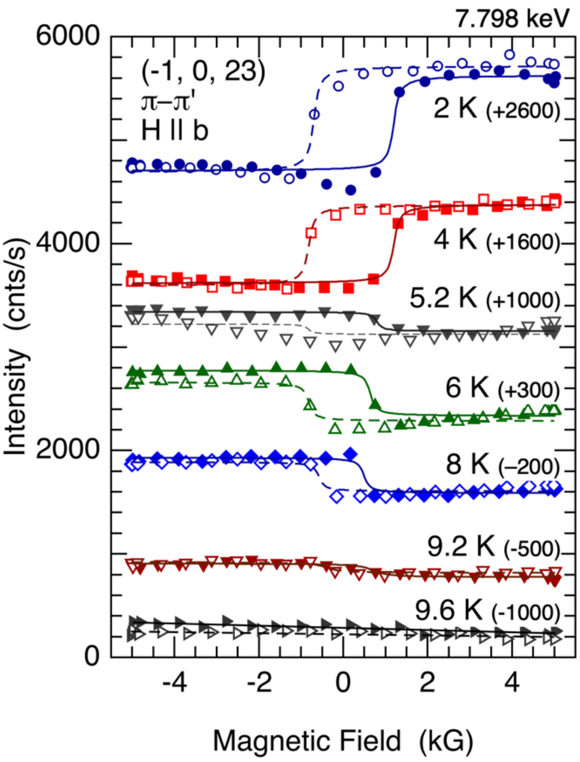

Fig. 2: Magnetic field dependences of the Bragg intensity at (-1, 0, 23) fundamental lattice spot for H ll b.

soon disappear at around 2 kG and the $\boldsymbol{q}$=(0,0,0) structure is realized.

Fig. 2 shows the field dependences of the (-1, 0, 23) fundamental Bragg intensity for H ll b with polarization analysis. The magnetic scattering for the π - π' channel reflects the magnetic component perpendicular to the scattering plane (ll b). The change in intensity for +H and -H shows that the magnetic and Thomson scatterings interfere and the magnetic structure changes when the field direction is reversed. Interestingly, the behavior of the intensity changes below and above 5 K. This temperature corresponds to T_3 in Ref. [3], where the macroscopic magnetization vanishes.

Although the detailed analysis of the change in magnetic structure is still under progress, this anomalous behavior shows that some kind of spin flop takes place by changing the temperature. Since this kind of spin flop usually takes place by applying a magnetic field because of the competition between magnetic anisotropy and the Zeeman energy, it is speculated that the similar spin flop takes place through a temperature sweep.

4 Conclusion

We have studied the successive magnetic phase transitions in $DyNi3Ga_9$ by resonant X-ray diffraction at low temperatures and in magnetic fields. It was clarified that the helimagnetic order first appears just below the Neel temperature T_1 with ferroquadrupole order. Although this statement sounds controversial, we consider that this is one of the interesting points in $DyNi3Ga_9$ with large quadrupole moments. The crossover and the transition from incommensurate to commensurate structure shows that the magnetic exchange interaction, Dzyaloshinsky-Moriya interaction, and the anisotropy energies are in the same order of magnitudes and competing.

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