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Powder Neutron Diffraction Study of $HoCoGa_5$

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We have studied successive magnetic transitions of $HoCoGa_5$ at $T_{N1} = 9.6$ K and $T_{N2} = 7.5$ K by using powder neutron diffraction. Apparent superlattice peaks were observed at temperatures below T_{N1}. With further decreases temperature, the patterns exhibit a substantial change at temperatures below T_{N2}. The observed magnetic peaks at 8 K (AntiFerromagnetic InCommensurate (AFIC) phase : $T_{N2} < T < T_{N1}$) can be represented by the propagation vector $\boldsymbol{q}_L = (1/2\ 0\ \tau)$ with $\tau = 0.35(2)$. In contrast, the magnetic structure becomes commensurate with $\boldsymbol{q}_C = (1/2\ 0\ 1/2)$ at 4 K (AntiFerromagnetic Commensurate (AFC) phase : $T < T_{N2}$). The temperature dependence of magnetic intensity shows an apparent temperature hysteresis at T_{N2}, indicates a first-order transition at T_{N2}. Analysis of the integrated intensity at 4 K reveals that the Ho moment with a size of 8.6(2) μ_B, oriented parallel to the c-axis in the AFC phase. While the successive transitions of $HoCoGa_5$ are different from those of $TbCoGa_5$, the magnetic structure in the AFC phase of $HoCoGa_5$ is the same as the AF_I^{Tb} of $TbCoGa_5$, and may indicate an additional transition at a lower temperature in $HoCoGa_5$.

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I. INTRODUCTION

As represented by heavy-fermion superconductors $CeCoIn_5$ and $PuCoGa_5$, the large family of so-called 115 compounds with the tetragonal $HoCoGa_5$-type structure has been one of the most intensively studied systems [1–5]. The interplay between magnetism and superconductivity is a key subject in the compounds. Note that a wide variety of magnetic properties is another intriguing aspect of the 115 family. In particular, itinerant members of $UTGa_5$ and $NpTGa_5$ were revealed to form various types of commensurate magnetic structures depending on the transition-metal element T [6–10]. The presence of a quadrupolar interaction and its coupling with magnetic one are suggested to a play vital role in this variety [11].

The quadrupole is suggested to be important for localized members RTX_5 (X = Ga and In) with heavy rare-earth ions R as well [12, 13]. So far, all magnetic structures of RTX_5, except for Ce, reported have a characteristic propagation vector $\boldsymbol{q} = (1/2\ 0\ 1/2)$ that breaks tetragonal symmetry [14,15]. Among them, unique successive second-order magnetic transitions in $TbCoGa_5$ at $T_{N1}^{Tb} = 36.2$ K and $T_{N2}^{Tb} = 5.4$ K are proposed as *component-separated magnetic transitions*; at temperatures below T_{N1}^{Tb}, the c-component of the Tb moment orders antiferromagnetically whereas the in-plane component remains paramagnetic and only orders at temperatures below T_{N2}^{Tb} [16,17]. This behavior is manifested in the magnetic susceptibility, in which only χ_c shows an anomaly at T_{N1}^{Tb} while the contrary behavior, *i.e.*, an anomaly only at T_{N2}^{Tb}, is found for χ_a. A recent complemental study of neutron diffraction and Nuclear Magnetic Resonance (NMR) successfully unveiled the details of magnetic transitions; magnetic moments order parallel to the c-axis at temperatures below T_{N1}^{Tb} with the propagation vector $\boldsymbol{q}_1 = (1/2\ 0\ 1/2)$ [18–20]. At temperatures below T_{N2}^{Tb}, an additional in-plane component characterized by $\boldsymbol{q}_2 = (0\ 1/2\ 1/2)$ is superposed on $\boldsymbol{q}_1$, resulting in a canting magnetic structure. These transition phenomena have similarities to those in the geometrically-

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frustrated $CsNiCl_3$ [21–25]. Hence, a novel type of frustration between magnetic and quadrupolar interactions is suggested as a possible scenario for the transitions in $TbCoGa_5$.

Recently, other members of RCoGa$_5$ (R = Dy∼Tm) have been successfully obtained as well [12, 13]. Systematic study has revealed that the magnetic anisotropy shows gradual change from Tb to Tm, where the magnetic easy-axis in paramagnetic states switches from the c- to the a-axis. As a consequence, one might expect the detailed balance between magnetic and quadrupolar interactions to be modified concomitantly. In $HoCoGa_5$, the magnetic easy-axis remains the c-axis with a smaller anisotropy [12,13]. $HoCoGa_5$ also shows successive magnetic transitions at T_{N1} = 9.6 K and T_{N2} = 7.5 K respectively [13]. Revealing the details of these transitions by using a powder neutron diffraction would be of great interest.

II. EXPERIMENTAL DETAILS

Single crystals of $HoCoGa_5$ were grown by using the Ga self-flux method. The details of the sample preparation are published elsewhere [13]. The Obtained tiny single crystals were ground into a fine powder in a mortar. Powder neutron diffraction experiments were carried out on the Wide-Angle Neutron Diffractometer (WAND) installed at the High Flux Isotope Reactor (HFIR) at the Oak Ridge National Laboratory (ORNL). Neutrons with a wavelength of 1.48 Å were provided by the Ge (311) monochromator. A radial oscillating collimator after the sample was used in order to reduce the background. The powder sample was put in a vanadium cell and mounted on the cold finger of a standard closed-cycle refrigerator. Powder neutron diffraction patterns were collected at temperatures between 3.5 K and 20 K.

III. RESULTS

Figure 1(a) shows powder neutron diffraction patterns of $HoCoGa_5$ measured at 4 K (the AntiFerromagnetic Commensurate (AFC) phase), 8 K (the Antiferromagnetic InCommensurate (AFIC) phase) and 20 K (the paramagnetic state). All reflection peaks observed at 20 K could be indexed by using the reported structure of $HoCoGa_5$ with the space group $P4/mmm$ shown as black vertical bars. No indication of the existence of an impurity phase was found at 20 K.

Superlattice reflection peaks, in addition to the nuclear Bragg peaks, were clearly observed at 4 K and 8 K. Observed supper-lattice peaks are pronounced at low angles, suggesting their magnetic origin. The red bars in the figure indicate the expected peak position for $\boldsymbol{q}_{\mathrm{C}}$ = (1/2 0 1/2). The observed magnetic peak positions at

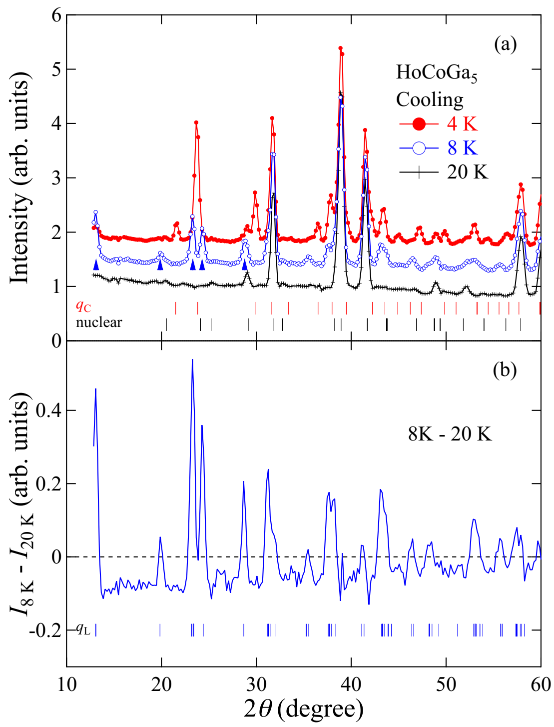

Fig. 1. (Color online) (a) Powder neutron diffraction patterns of $HoCoGa_5$ observed at 4 K (•, red), 8 K (◦, blue) and 20 K (+, black). Arrows indicates superlattice reflection peaks at 8 K (AFIC phase). Vertical bars (red and black) mark expected peak positions of the propagation vector $\boldsymbol{q}_{\mathrm{C}}$ = (1/2 0 1/2) and the nuclear reflection peaks, respectively. (b) Difference diffraction patterns at 8 K obtained by subtracting the pattern at 20 K. Vertical bars (blue) indicate expected peak positions of the magnetic propagation vector $\boldsymbol{q}_{\mathrm{L}}$ = (1/2 0 0.35).

4 K are well reproduced by the calculation; namely, the propagation vector for AFC is $\boldsymbol{q}_{\mathrm{C}}$. On the other hand, the magnetic peak positions at 8 K are distinctly different from those at 4 K. A splitting of the peak around 23° at temperatures from 4 K to 8 K suggests a lowering of symmetry at 8 K. In order to see this variation in detail, we subtracted the paramagnetic pattern at 20 K from the data at 8 K, and the obtained difference pattern $I_{8\mathrm{K}}$ - $I_{20\mathrm{K}}$, is plotted in Fig. 1(b). The negative intensity with the positive slopes can be attributed to the presence of paramagnetic scattering at 20 K. This subtraction makes it clear that large numbers of magnetic peaks were clearly observed in the spectra at 8 K. In order to explain the positions of the observed peaks, we assumed a propagation vector of $\boldsymbol{q}_{\mathrm{L}}$ = (1/2 0 τ). The observed peak positions could be reproduced by using $\boldsymbol{q}_{\mathrm{L}}$ with τ = 0.35(2), as displayed by the vertical bars.

Figure 2 shows the temperature dependence of the integrated intensities of representative magnetic reflections at $\mathbf{Q}_{\mathrm{I}}$ = (1/2 1 0.35(2)) and $\mathbf{Q}_{\mathrm{II}}$ = (1/2 1 1/2) for the

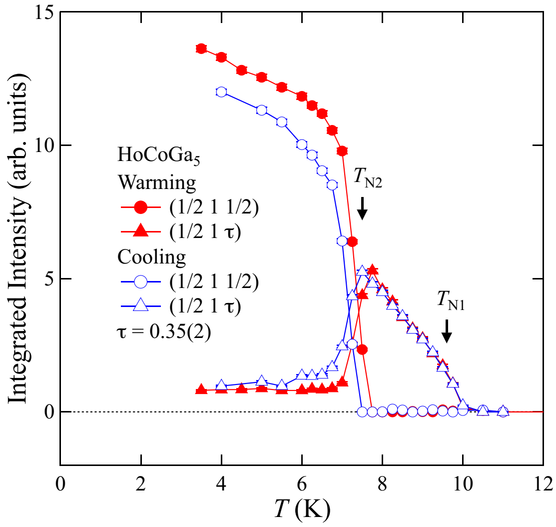

Fig. 2. (Color online) Temperature dependences of the magnetic reflection intensities for $\mathbf{Q}_{\mathrm{I}} = (1/2\ 1\ 0.35)$ (triangles) and $\mathbf{Q}_{\mathrm{II}} = (1/2\ 1\ 1/2)$ (circles). The closed symbols represent results taken on warming (red) while the open symbols correspond to those measurement on cooling (blue). The arrows indicate T_{N1} and T_{N2}.

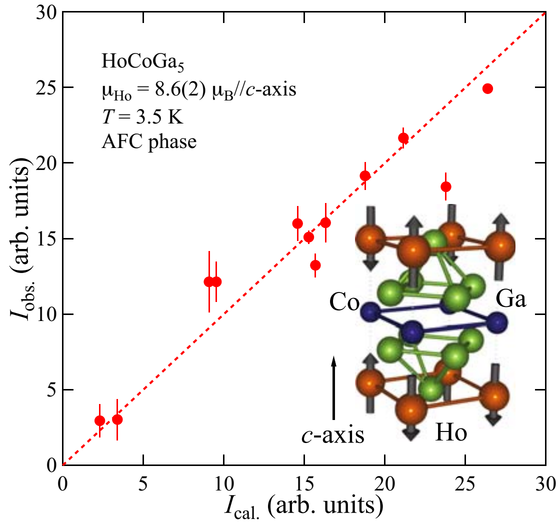

Fig. 3. (Color online) Observed magnetic reflection intensities of $HoCoGa_5$ taken at 3.5 K plotted against the calculated intensities on assuming the magnetic structure shown in the inset. The orange, blue, and green spheres in the inset denote the Ho, Co, and Ga atoms, respectively, and the gray arrows represent the magnetic moment of Ho.

AFIC and the AFC phases, respectively. Upon cooling, the reflection at $\mathbf{Q}_{\mathrm{I}}$ starts to develop at temperatures below 10 K, corresponding to T_{N1}. The peak at $\mathbf{Q}_{\mathrm{I}}$ shows a sudden drop upon further cooling to temperatures below 7.3 K and disappears at temperatures below 6 K (the residual intensity below 6 K originates from the peak tail at $\mathbf{Q}_{\mathrm{II}}$). The peak intensity at $\mathbf{Q}_{\mathrm{II}}$ exhibits sharp increase below 7.3 K. One should note that clear hysteresis exists at T_{N2} with $\delta T \sim 0.25$ K while the temperature dependence does not accompany a visible difference at T_{N1}. Furthermore, the peak at $\mathbf{Q}_{\mathrm{I}}$ coexists with the one at $\mathbf{Q}_{\mathrm{II}}$. This behavior evidently proves that the transition at T_{N2} is of first order. This is consistent with the sharp jump in the specific heat at T_{N2} [13].

Hereafter, we analyze the integrated intensity of magnetic reflection in the AFC phase at 3.5 K. The magnetic scattering intensity $I_{\mathrm{cal}}(\mathbf{Q})$ for the unpolarized powder neutron diffraction experiments is expressed as

$$I_{\mathrm{cal}}(\mathbf{Q}) \propto \mu^2 \langle \sin^2 \alpha \rangle f^2(\mathbf{Q}) |F_{\mathrm{M}}(\mathbf{Q})|^2 L(\theta) m_{hkl(\mathrm{M})},$$

where μ is the magnitude of the magnetic moment, α is the relative angle between the scattering vector and the magnetic moment, $\mathbf{Q}$ is the scattering vector, $f(\mathbf{Q})$ is the magnetic form factor, $F_{\mathrm{M}}(\mathbf{Q})$ is the magnetic structure factor, $L(\theta)$ is the Lorentz factor and $m_{hkl(\mathrm{M})}$ is the multiplicity of the magnetic reflection. As shown in Fig. 3, the observed intensity is successfully reproduced by assuming the magnetic moments to be aligned parallel to the *c*-axis (displayed in the inset). The magnitude of the moment is determined to be 8.6(2) $\mu_{\mathrm{B}}/\mathrm{Ho}^{3+}$, corresponding to 86% that of the Ho^{3+} free ion.

Concerning the AFIC phase, the present data do not allow us to obtain a conclusive solution for the long-periodic magnetic structure. One possibility may be a longitudinal sinusoidal structure along the *c*-axis [13]. Further experiments on a single-crystal sample will be indispensable for this purpose.

IV. DISCUSSION

This experiment revealed that the successive transitions in $HoCoGa_5$ were intrinsically different from those of $TbCoGa_5$. The first-order transition at T_{N2} makes a clear contrast to the second-order nature at $T_{\mathrm{N2}}^{\mathrm{Tb}}$ in $TbCoGa_5$ [16,17]. In addition, while two ordered phases in $TbCoGa_5$ can be characterized with the *common* propagation vectors $\boldsymbol{q} = \langle 1/2\ 0\ 1/2 \rangle$ [18,19], the AFIC phase of $HoCoGa_5$ has a long periodic magnetic structure with $\boldsymbol{q}_{\mathbf{L}}$. The long periodicity is a rare example among the 115 family, in which most of the compounds, form commensurate magnetic structures, with a few exceptions such as $CeRhIn_5$ [26] and the Q-phase of $CeCoIn_5$ [5]. The in-plane component of the propagation vector (1/2 0), which corresponds to the nearest-neighbor coupling, is common to the 115 compounds with heavy rare-earth ions. One should note that this in-plane component breaks the tetragonal symmetry while no evidence of structural distortion has been detected by microscopic probes so far.

Upon further cooling to temperatures below T_{N2}, the long periodic modulation $\tau \sim 0.35$ in the c-component changes to 1/2 while the characteristic in-plane nearest-neighbor interaction remains unchanged. As a result, the propagation vector of the AFC phase is described with $\boldsymbol{q}_{\mathrm{C}} = (1/2\ 0\ 1/2)$, common to RTX_5. This study revealed that the magnetic structure of AFC with moments aligned parallel to the c-axis was identical to that of the $\mathrm{AF}_{\mathrm{I}}^{\mathrm{Tb}}$ phase in $TbCoGa_5$. These facts may suggest that the in-plane magnetic component may fluctuate in the AFC phase as well, and that it may order at an additional lower-temperature transition in $HoCoGa_5$. Further experiments at lower temperatures are indispensable to deepen our understanding of this system.

V. CONCLUSIONS

In conclusion, we have studied the successive magnetic transitions in $HoCoGa_5$ by using the powder neutron diffraction technique. In the AFIC phase, clear magnetic reflections with the propagation vector $\boldsymbol{q}_{\mathrm{L}} = (1/2\ 0\ 0.35)$ were observed and then changed to $\boldsymbol{q}_{\mathrm{C}} = (1/2\ 0\ 1/2)$ in the AFC phase. The temperature dependence of the magnetic reflection intensity showed an apparent temperature hysteresis at T_{N2}, suggesting the first-order nature of this transition. Our analysis revealed that in the AFC phase, where the Ho magnetic moments with a magnitude of 8.6(2) μ_{B} oriented parallel to the c-axis. The determined magnetic structure in the AFC phase was the same as the $\mathrm{AF}_{\mathrm{I}}^{\mathrm{Tb}}$ phase and might indicate an additional transition at lower temperature in $HoCoGa_5$.

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