Component-separated magnetic transition in HoRh$_2$Si$_2$ single crystal

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Abstract. Magnetic susceptibility and specific heat measurements have been carried out on a HoRh$_2$Si$_2$ single crystal compound. Specific heat shows two $\lambda$-like anomalies at $T_N=29.1$ K and $T_{N'}=12.0$ K, and an additional sharp anomaly indicating the first order transition at $T_t=27.3$ K. The temperature dependence of susceptibility along the [001] direction shows a clear cusp at $T_N$ but a tiny kink at $T_{N'}$ and $T_t$. In contrast, those in the $ab$-plane show a clear cusp at $T_{N'}$. HoRh$_2$Si$_2$ shows a successive component-separated magnetic transition coming from a new type frustration not geometrical one.

1. Introduction

A frustration, which often comes from a geometrical ion arrangement, leads various unusual physical properties. “Successive component-separated magnetic transition”, which is unique successive transition characterized by independent ordering of the $c$- and $ab$-component of magnetic moments at different temperatures, is one of the interesting characters. The compounds CsNiCl$_3$ and DyB$_4$ are well known as compounds which show this type of transition [1, 2]. In CsNiCl$_3$, it is suggested that the transition is due to competing exchange interaction and weak Ising anisotropy in the antiferromagnetic triangular lattice [3]. In a Shastry-Sutherland lattice DyB$_4$, geometrical quadrupolar frustration in addition to geometrical spin frustration play an important role [2]. In most cases, geometrical frustration is important. Recently, N. Sanada et al. have reported that the tetragonal compound TbCoGa$_5$ shows successive component-separated magnetic transitions [4]. The magnetic, specific heat and elastic constant behavior indicate the presence of a strong frustration in the interactions in TbCoGa$_5$. The geometrical frustration is not valid for TbCoGa$_5$, because of the simple tetragonal arrangement of Tb atoms in this compound. The new class of frustration effects where quadrupolar degrees of freedom play an important role has been proposed. They suggest that this type transitions appear commonly in the rare earth compounds with a degeneracy of quadrupolar degrees of freedom below the highest magnetic transition temperatures. Further study on compounds which show a similar behavior to one of TbCoGa$_5$ should be required.

The ternary compound HoRh$_2$Si$_2$ having the tetragonal ThCr$_2$Si$_2$-type structure shows an antiferromagnetic ordering below $T_N=27$ K and an additional transition at $T_{N'}=11$ K [5, 6]. In the temperature dependence of magnetic susceptibility, two clear peaks have been observed. The neutron diffraction study have reported that the antiferromagnetic structure is a simple one; propagation vector...
$k=(0,0,1)$ with magnetic moments tilted from the c-axis by 28° at low temperatures and have given no information on the transition at $T_{N'}$. Takano et al have proposed that the transition at $T_{N'}$ is corresponding to the temperature where magnetic moments become parallel to the c-axis with increasing temperature [7]. Although the details in the transition at $T_{N'}$ have been unknown yet, multiple interactions should play a role to decline the magnetic moments. So we consider the occurrence of frustration is possible. Then HoRh$_2$Si$_2$ single crystals have been grown and magnetic study has been performed on the single crystal compounds.

2. Experimental
The single crystal HoRh$_2$Si$_2$ has been grown by the tetra-arc Czochralski method. The single phase nature has been confirmed by X-ray powder diffraction. Good quality of the single crystal has been confirmed and crystallographic orientations were determined by back Laue method. The crystal was fixed on a plastic plate so that the desired direction is perpendicular to the plate within experimental angle accuracy of one degree. Then it is used for magnetic measurements which have been carried out by use of a PPMS and SQUID magnetometer, MPMS, (Quantum Design). The specific heat has been performed by a relaxation method also using the PPMS.

![Figure 1](image1.png)  
*Figure 1*  Magnetic susceptibility along the main symmetry axes of a tetragonal cell on the HoRh$_2$Si$_2$ single crystal.

![Figure 2](image2.png)  
*Figure 2*  Inverse magnetic susceptibility along the c-axis and the directions in the basal plane

3. Results and discussion
The temperature dependence of magnetic susceptibility along the main symmetry axes for a tetragonal cell is shown in Fig.1. A magnetic anisotropy between the c-axis and directions in the basal plane is evidenced from the figure while no anisotropy is in the basal plane; the susceptibility along the [100] direction is identical to the one along [110]. The [001] magnetic susceptibility shows a clear maximum at $T_{N1}=29.5$ K and a small hump at $T_{N2}=12.4$ K. A small kink at $T_{c}=27.2$ K is also seen. In contrast, susceptibilities for directions in the basal plane, $B \parallel [001]$, show a small hump at $T_{N1}$ and a clear maximum at $T_{N2}$. Any anomaly is hardly seen around $T_c$. These results suggest that c- and ab-components of magnetic moments of the Ho$^{3+}$ ions order independently at $T_{N1}$ and $T_{N2}$, respectively. This type of transition is called “successive component-separated magnetic transitions”. Similar behaviours have been reported in TbCoGa$_5$ [4] and DyB$_4$ [2]. The existence of three phases below $T_{N1}$ is evidenced from the result. We call hereafter these the phase I ($T_{N1}>T> T_c$), the phase II ($T_c>T> T_{N2}$), the phase III ($T_{N2}>T$).
The susceptibilities along the all directions obey the Curie-Weiss law above 100 K as shown in Fig. 2. The $\chi^{-1}$-$T$ curves become linear and are almost parallel with each other at high temperatures. The effective magnetic moment estimated is 10.60 $\mu_B$ which is in good agreement with the Ho$^{3+}$ ion magnetic moment (10.6 $\mu_B$). This shows that 4$f$ electrons in HoRh$_2$Si$_2$ are well localized and Rh ion is nonmagnetic. The paramagnetic Curie temperatures along the c-axis and directions in the basal plane are $\Theta_// = 19.3$ K and $\Theta_\perp = -9.3$ K, indicating that dominant interactions along the c-axis and in the basal plane are ferromagnetic and antiferromagnetic, respectively.

The thermal variations of magnetic specific heat $C_{mag}$ and entropy $S_{mag}$ on the HoRh$_2$Si$_2$ single crystal are shown in Fig.3. The magnetic contribution of specific heat is deduced from subtracting the specific heat of a phonon contribution from total specific heat of HoRh$_2$Si$_2$. The phonon contribution is calculated from the Debye function with the Debye temperature $\Theta_D = 313$ K for HoRh$_2$Si$_2$ which is estimated from $\Theta_D = 330$ K for LaRh$_2$Si$_2$ [6]. The specific heat shows two $\lambda$-type anomalies at $T_{N1}$ and $T_{N2}$, and a sharp peak at $T_t$. This dependency is almost agreement with one reported by Sekizawa et al. [6], but they did not recognize the transition at $T_{N1}$. They determined the Neel temperature by the peak temperature of $T_t$. The existence of three phases below $T_{N1}$ is confirmed. The each phase transitions at $T_{N1}$ and $T_{N2}$ is of the second-order one, and the first-order one at $T_t$. The magnetic entropy is obtained from numerical integration of $C_{mag}/T$ vs. $T$. It is evidenced from the figure the entropy reaches Rln4 at $T_{N2}$. This indicates that the ground state is a pseudo-quartet at least. The entropy of Rln12 ($=20.6$ J mol$^{-1}$K$^{-1}$) which is released below $T_{N1}$ is smaller than Rln(2$J$+1) = 23.5 J mol$^{-1}$K$^{-1}$ for $J=8$ multiplet of Ho$^{3+}$ ion. The overall splitting of the crystalline electric field levels is slightly larger than $T_{N1}$.

The neutron diffraction study on the single crystal and powdered sample has been performed. The antiferromagnetic structure of the phase III at low temperatures below $T_{N2}$ is a simple one; the propagation vector $k=(0,0,1)$ with magnetic moments tilting from the c-axis by about 30 $^\circ$ at 4 K. The tilting angle becomes smaller with increasing temperature and almost zero at $T_{N2}$. For the phase II, ab-component of magnetic moments disappears and diffuse scatterings, indicating paramagnetic ab-component, appear. The phase I has an incommensurate structure. The detailed analysis of neutron result is now in progress which will be published soon elsewhere.

These results of HoRh$_2$Si$_2$ are quite similar to those of TbCoGa$_5$. Ho atoms in this compound are of the simple tetragonal arrangement. Then no geometrical frustration is considered. So we believe HoRh$_2$Si$_2$ is a candidate for a new type frustration. Further study such as elastic constant, magnetic and neutron measurements on this single crystal is now in progress.
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