Magnetic transitions on the pseudo-ternary compounds Ho$_{1-x}$Y$_x$Rh$_2$Si$_2$

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Abstract. Magnetic measurements on the pseudo-ternary compounds Ho$_{1-x}$Y$_x$Rh$_2$Si$_2$, which substitute nonmagnetic Y for Ho, were performed. The lattice parameters $a$ and $c$ are almost constant by the change of Y composition $x$. The transition temperatures $T_{N1}$, $T_{N2}$, and $T_{N3}$ decrease with increasing $x$. The critical compositions for $T_{N1}$ and $T_{N2}$ are determined to be $x_{N1}=0.98$ and $x_{N2}=0.75$, respectively. “The successive component-separated magnetic transitions” appear for the wide $x$ region from $x=0$ to $x=0.75$. The partial ordered state, or frustration appears on these compound. The magnetic ordered state persists up to $x=0.98$ in spite of very dilute Ho-compounds; very weak magnetic interactions. The effective magnetic moments are almost constant from $x=0$ to 1; $\mu_{eff}=10.6 \pm 0.15 \mu_B$/Ho, and magnetic moment $\mu=10 \mu_B$/Ho as well. The Ho$_{1-2x}$Y$_{2x}$Rh$_2$Si$_2$ single crystal shows the quite similar magnetic behaviour to that of HoRh$_2$Si$_2$. The constant magnetic moment and constant effective magnetic moment irrespective of $x$ are confirmed from the magnetic behaviours of single crystals Ho$_{1-x}$Y$_x$Rh$_2$Si$_2$ ($x=0.05$, 0.6 and 0.8).

1. Introduction
The ternary compound HoRh$_2$Si$_2$, which crystallizes in the tetragonal ThCr$_2$Si$_2$-type structure, shows so called “successive component-separated magnetic transitions”; the $c$- and $ab$-components of magnetic moments independently order at the different temperatures, $T_{N1}=29.1$ K and $T_{N2}=12.1$ K [1]. So the partial ordered state, where only the $c$-component of magnetic moments orders and the $ab$-components disorder, appears for $T_{N1}>T_{N2}>T_{N2}$. This type of transitions often appears in frustration systems. The geometrical frustration, which comes from a competition of multiple interactions due to a geometrical ion arrangement, is responsible for most of frustrations [2, 3]. The geometrical frustration is, however, not like to occur in this compound due to the structure. The origin is very interesting. This compound has an additional transition at $T=27.3$ K [1]. The simple antiferromagnetic structure which is the propagation vector $k=(0, 0, 1)$ with magnetic moments tilted from the $c$-axis by $28^\circ$ at low temperatures has been reported from the neutron study [4]. Magnetic studies on the single crystal show complex magnetic behaviors [5, 6]; step-like metamagnetic processes appear along the symmetry directions below $T_{N2}$. For $T_{N2}<T<T_N$, a two-step metamagnetic one persists along the [001] direction whereas the processes along directions in the basal plane become a paramagnetic like one, suggesting that the $ab$-components of magnetic moments are disordered, frustrated. The $B_{100}-T$ and $B_{110}-T$ magnetic phase diagrams show the existence of peculiar phase boundary around $T_{N2}$, suggesting that a quadrupole effect play an important role for the component-separated magnetic transition. The
pseudo-ternary compounds $\text{Ho}_{1-x}\text{Gd}_{x}\text{Rh}_2\text{Si}_2$, which substitutes Gd with no quadrupole for Ho, have been studied; the quadrupole effects on the transitions were investigated. The results declare that quadrupole interactions play an important role in this compound system [7]. In order to obtain more information on an origin of the successive component-separated magnetic transitions, magnetic behaviors of pseudo-ternary compounds $\text{Ho}_{1-x}\text{Y}_x\text{Rh}_2\text{Si}_2$, which substitute non-magnetic Y for Ho, were studied. The ionic radiuses of Y and Ho ions are 1.06 Å and 1.05 Å, respectively; the ionic radiuses of Y and Ho are almost same. So it should be expected that effects of magnetic interactions on the magnetic transitions can be studied without a deformation of the lattice on $\text{Ho}_{1-x}\text{Y}_x\text{Rh}_2\text{Si}_2$.

2. Experimental
Polycrystalline samples of pseudo-ternary compounds $\text{Ho}_{1-x}\text{Y}_x\text{Rh}_2\text{Si}_2$ were prepared by arc-melting a stoichiometric mixture of the pure elements (Ho and Y: purity of 3N, Rh: 3N, and Si: 5N) in an argon atmosphere. Subsequently, single crystals ($x = 0.05, 0.6$ and $0.8$) were grown by the Czochralski technique using a tri-arc furnace; the polycrystalline sample was melted on a water-cooled copper hearth in the tri-arc furnace. A crystal was slowly pulled from the melt by a pulling-up rod with a seed crystal. The pulling-up rate was $5 \text{ mm/h}$ [8]. The crystal structure and the single phase nature were confirmed by X-ray powder diffraction. The quality of the single crystals was checked and crystallographic orientations were determined by the back Laue method. The magnetic susceptibility and magnetization were measured using a PPMS and/or MPMS (Quantum Design) in the Institute for Solid State Physic, the University of Tokyo.

3. Results and discussion

3.1. Lattice parameter
The lattice parameters $a$ and $c$ of the tetragonal cell on the $\text{Ho}_{1-x}\text{Y}_x\text{Rh}_2\text{Si}_2$ were determined by the least square analysis to X-ray diffraction powder patterns for polycrystalline samples. The Y composition $x$ dependences of those are shown in Fig. 1. For $\text{HoRh}_2\text{Si}_2$ ($x = 0$) $a = 4.026 \text{ Å}$, $c = 9.899 \text{ Å}$, and $a = 4.033 \text{ Å}$, $c = 9.917 \text{ Å}$ for $\text{YRh}_2\text{Si}_2$ ($x = 1$), which are agreement with the previous reports [9, 10]. The rate of change in both $a$ and $c$ from $x = 0$ to 1 is 0.18 %, and is very slight increase; no lattice deformation occurs. It is expected that the crystal field effects in this system don’t change by this substitution.

![Figure 1] Y composition $x$ dependence of lattice parameters $a$ and $c$ on the polycrystalline compounds $\text{Ho}_{1-x}\text{Y}_x\text{Rh}_2\text{Si}_2$. 
3.2. Magnetic susceptibility

The temperature dependences of magnetic susceptibility along the main symmetry axes on the single crystals Ho_{0.95}Y_{0.05}Rh_{2}Si_{2} (x = 0.05, 0.6 and 0.8) are shown in Fig. 2, Fig. 3 and Fig. 4, respectively. A magnetic anisotropy between the c-axis and directions in the basal plane is evidenced from the figure which indicates the easy magnetization direction is the [001] direction. In Fig. 2 for Ho_{0.95}Y_{0.05}Rh_{2}Si_{2} (x=0.05), the [001] magnetic susceptibility \( \chi_{[001]} \) shows a clear maximum at \( T_{N1}=28.6 \) K and a small hump at \( T_{N2}=1.4 \) K. In contrast, susceptibilities \( \chi_{[100]} \) and \( \chi_{[110]} \) for directions in the basal plane show a small hump at \( T_{N1} \) and a clear maximum at \( T_{N2} \). A small kink at \( T_{t}=25.5 \) K is also observed. This feature is similar to that of HoRh_{2}Si_{2}; although the transition temperatures become lower than those of HoRh_{2}Si_{2} [1]; the successive component-separated magnetic transitions appear. The susceptibilities obey the Curie-Weiss law above 50 K. The susceptibility on the single crystal.

Figure 2 Magnetic susceptibility along the main symmetry axes of tetragonal cell on the Ho_{0.95}Y_{0.05}Rh_{2}Si_{2} (x=0.05) single crystal.

Figure 3 Magnetic susceptibility along the main symmetry axes of tetragonal cell on the Ho_{0.4}Y_{0.6}Rh_{2}Si_{2} (x=0.6) single crystal.

Figure 4 Magnetic susceptibility along the main symmetry axes of tetragonal cell on the Ho_{0.2}Y_{0.8}Rh_{2}Si_{2} (x=0.8) single crystal.

Figure 5 Temperature dependence of magnetic susceptibility on the polycrystalline compounds Ho_{1-x}Y_{x}Rh_{2}Si_{2} (x = 0.2, 0.4, 0.6 and 0.8)
compound which is in good agreement with the Ho$^{3+}$ free ion magnetic moment ($g(J+1)^{1/2}=10.6 \mu_B$). This shows that 4$f$ electrons in this compound are well localized and Rh ion is nonmagnetic. The paramagnetic Curie temperatures along the c-axis and [100], [110] directions in the basal plane are $\Theta_{001}=18.5$ K, $\Theta_{000}=-5.4$ K and $\Theta_{110}=-7.6$ K, respectively, indicating that the dominant interaction along the c-axis and in the basal plane is ferromagnetic and antiferromagnetic, respectively. With increasing $x$, the transitions shift to lower temperatures; for $x=0.6$, $T_{N1}=13.6$ K and $T_{N2}=2.5$ K, and for $x=0.8$, $T_{N1}=6.4$ K and $T_{N2}<2$ K. The transition of $T_t$ is hardly observed in the figures 3 and 4.

Figure 5 shows temperature dependences of magnetic susceptibility on the polycrystalline compounds Ho$_{1-x}$Y$_x$Rh$_2$Si$_2$ ($x=0.2$, 0.4, 0.6 and 0.8). It can be confirmed that the larger $x$ make the lower transition temperatures. The magnetic susceptibilities of all the compounds obey the Curie-Weiss law for high temperatures. The effective magnetic moments per Ho atom estimated are almost constant irrespective of change in $x$: $\mu_{eff}=10.6 \pm 0.15 \mu_B$/Ho which is good agreement with the Ho$^{3+}$ theoretical moment. This means that 4$f$ electrons in this compound system are well localized and Rh ion is nonmagnetic in this compound system.

**Figure. 6** Magnetization curves at 1.8 K along the main symmetry axes on the Ho$_{0.95}$Y$_{0.05}$Rh$_2$Si$_2$ ($x=0.05$) single crystal.

**Figure. 7** Magnetization curves at 14 K along the main symmetry axes on the Ho$_{0.95}$Y$_{0.05}$Rh$_2$Si$_2$ ($x=0.05$) single crystal.

**Figure. 8** Magnetization curves at 1.8 K along the main symmetry axes on the Ho$_{0.4}$Y$_{0.6}$Rh$_2$Si$_2$ ($x=0.6$) single crystal.

**Figure. 9** Magnetization curves at 2 K along the main symmetry axes on the Ho$_{0.2}$Y$_{0.8}$Rh$_2$Si$_2$ ($x=0.8$) single crystal.
3.3. Magnetization process
In order to elucidate more detail magnetic behaviour by a change of x, magnetization processes of some single crystal compounds (x=0.05, 0.6 and 0.8) were studied. Magnetization curves at 1.8 K is shown on the Ho_{1-x}Y_{x}Rh_{2}Si_{2} single crystal (x=0.05) in Fig.6. Sharp step-like metamagnetic transitions appear along the all symmetry directions. This behaviour is similar to that of HoRh_{2}Si_{2} [5], indicating that magnetic moments tilt from the main symmetry axes. The [001] magnetization process, easy axis magnetization one is a two-step metamagnetic one; the magnetization increases rapidly at critical fields \( B_{c1}=1.9 \) T and \( B_{c2}=4.6 \) T. The magnetization curve is almost linear with a slight slope except around transition fields. The magnetization just above \( B_{c1} \) and \( B_{c2} \) is \( M_{1}=4.0 \) \( \mu_{B} \)/Ho and \( M_{2}=8.0 \) \( \mu_{B} \)/Ho, respectively. The \( M_{2} \) is two times as large as \( M_{1} \). If a Ho atom in this compound has the theoretical Ho\(^{3+}\) free ion moment of \( gJ=10 \) \( \mu_{B} \), the tilt angle of magnetic moments from the c-axis can be deduced from \( M_{2} \) to be 37°. This tilt angle should become 0° and the magnetization should saturate 10 \( \mu_{B} \)/Ho by higher magnetic fields as the case of HoRh_{2}Si_{2} [6]. A clear step-like metamagnetic transition also appears in the basal plane processes. The critical fields are \( B_{100}=2.6 \) T and \( B_{110}=1.9 \) T in the [100] and [110] processes, respectively. The magnetization just above the transition \( B_{100} \) and \( B_{110} \) is \( M_{100}=5.7 \) \( \mu_{B} \)/Ho and \( M_{110}=3.5 \) \( \mu_{B} \)/Ho, respectively. The higher temperature behaviours shown in Fig. 7 are also quite similar to that of HoRh_{2}Si_{2}; the two-step metamagnetic process along the [001] direction persists up to \( T_{t} \) whereas the basal plan processes become a paramagnetic one above \( T_{N2} \). This indicates the partial ordered state, where the magnetic moments of c-component order while ab-components disorder, appears for \( T_{N2}<T<T_{t} \). Figures 8 and 9 show the magnetization curves at 1.8 K and 2 K on the Ho_{1-x}Y_{x}Rh_{2}Si_{2} (x=0.6 and 0.8) single crystals, respectively. A two-step metamagnetic transition appears in the [001] process, and paramagnetic like processes appear in the basal plane on the both compounds. These behaviours are similar to that in Fig. 8. They are corresponding to those in the partial ordered state. The critical fields are \( B_{c1}=0.8 \) T and \( B_{c2}=1.9 \) T for \( x=0.6 \), and \( B_{c1}=0.3 \) T and \( B_{c2}=0.8 \) T for \( x=0.8 \). The larger x, weaker magnetic interactions, make lower critical fields, which is reasonable because it should be expected that the antiferromagnetic interactions become smaller with decreasing magnetic Ho. The saturation magnetization (at \( B=7 \) T) is 7.7 \( \mu_{B} \)/Ho and 7.8 \( \mu_{B} \)/Ho for \( x=0.6 \) and \( x=0.8 \), respectively, which is almost same to the moment of \( x=0 \) and \( x=0.05 \) around 7 T. It should be speculated the saturation magnetic moment reaches 10 \( \mu_{B} \) as same to that of HoRh_{2}Si_{2}. This means that the Ho magnetic moment keeps the theoretical Ho\(^{3+}\) magnetic moment (\( gJ=10 \) \( \mu_{B} \)) irrespective of x, in the diluted Ho composition compounds as well.

3.4. Composition dependence of transition temperature
The Y composition x dependences of magnetic transition temperatures \( T_{N1} \), \( T_{t} \), and \( T_{N2} \) on the Ho_{1-x}Y_{x}Rh_{2}Si_{2} compounds are shown in Fig. 10. The transitions were determined from measurements of \( \chi' \) \( T \) above mentioned. The magnetic ordered state is divided to 3 states by the transition lines, which are

![Figure 10](image_url)

**Figure 10** Y composition x dependences of magnetic transition temperatures \( T_{N1} \), \( T_{t} \), and \( T_{N2} \) on the Ho_{1-x}Y_{x}Rh_{2}Si_{2} compounds. Closed and open symbols are transition temperatures determined from single crystal and polycrystalline samples, respectively. The lines are drawn by fitting to quadratic functions.
referred to as phase I, II, and III in the order of increasing temperature as shown in the figure. The phase I has a simple antiferromagnetic structure with the propagation vector \( k = (0, 0, 1) \) and magnetic moments tilted from the \( c \)-axis [4]. The phase II and III are the partial ordered states as reported previously [5]. The transition temperatures \( T_{N1} \), \( T_{N} \), and \( T_{N2} \) decrease with increasing \( x \). The tendency is consistent with the fact that the magnetic interactions become weaker with decreasing Ho composition (increasing \( x \); nonmagnetic) in this compound system. The lines in the figure are drawn by fitting to quadratic functions. The dependencies well obey quadratic functions. The curves of \( T_{N1} \) and \( T_{N2} \) have upward curvatures while one of \( T_i \) has a downward curvature. This suggests that the main origin which induces the transitions of \( T_{N1} \) and \( T_{N2} \) is different from that of \( T_i \). The critical composition, which is a composition where a transition temperature becomes \( T = 0 \) K, for \( T_{N1} \) and \( T_{N2} \) is determined to be \( x_{N1} = 0.98 \) and \( x_{N2} = 0.75 \), respectively by extrapolating the quadratic functions to \( T = 0 \). It should be worth noting that the ordered state persists in spite of very dilute Ho compounds: only a few percent Ho-compound orders. The successive component-separated magnetic transitions appear for wide \( x \) range up to \( x_{N2} = 0.75 \). The \( x \)-dependency of a magnetic transition should have a downward curvature if it follows the de Gennes factor: \( (g-1)^2 J(J+1) \). The \( x \)-dependency of \( T_i \) is not scaled by de Gennes factor though it has an upward curvature. The RKKY interaction may take an important part for the transition of \( T_i \).

3.5. Summary

Magnetic studies on the pseudo-ternary compounds Ho\(_{1-x}\)Y\(_x\)Rh\(_2\)Si\(_2\), which substitute non-magnetic Y for Ho, were performed using either single crystal \((x = 0.05, 0.6, 0.8)\) or polycrystalline \((x = 0 - 1\) every 0.1\) compounds. The lattice parameters \( a \) and \( c \) are almost constant by the change of Y composition \( x \). The effects of magnetic interactions on the magnetic transitions can be studied without a change in the crystal field effects on this compound system. The single crystal compound Ho\(_{0.95}\)Y\(_{0.05}\)Rh\(_2\)Si\(_2\) \((x = 0.05)\) exhibit quite similar magnetic features to those of HoRh\(_2\)Si\(_2\). It shows “successive component-separated magnetic transitions” and step-like metamagnetic transitions along all the symmetry directions at low temperatures. The metamagnetic transitions in the basal plane disappear for \( T > T_{N2} \) for the phase II: the partial ordered state. The similar behaviours appear on the single crystals Ho\(_{1-x}\)Y\(_x\)Rh\(_2\)Si\(_2\) \((x = 0.6 \) and \( 0.8)\) at low temperatures as well. So these compounds is in the phase II at low temperatures. The magnetic moment per Ho atom of the compounds \((x = 0.95, 0.6, 0.4)\) is same to that of HoRh\(_2\)Si\(_2\): a Ho magnetic moment and effective magnetic moment keep constant in the diluted Ho compounds. The partial ordered state, or frustration appears on these compound. “The successive component-separated magnetic transitions” appear in this system as well. The Y composition \( x \) dependence of magnetic transitions were determined. The transition temperatures \( T_{N1} \), \( T_{N} \), and \( T_{N2} \) decrease with increasing \( x \). The critical compositions, where the transition temperatures of \( T_{N1} \) and \( T_{N2} \) become zero, are determined to be \( x_{N1} = 0.98 \) and \( x_{N2} = 0.75 \), respectively. It should be noted that the ordered state persists in spite of very dilute Ho region: only a few percent Ho-compound orders magnetically. The effective magnetic moments are almost constant from \( x = 0 \) to 1: \( \mu_{\text{eff}} = 10.6 \pm 0.15 \mu_\text{B}/\text{Ho} \) which is good agreement with the Ho\(^{3+}\) theoretical moment. This means that 4f electrons in this compound system are well localized and Rh ion is nonmagnetic. The successive component-separated magnetic transitions appear for wide \( x \) range up to \( x_{N2} = 0.75 \). The \( x \)-dependency of \( T_i \) indicates the RKKY interaction may take a part of the cause for the transition of \( T_i \). In spite of decreasing magnetic interactions, the partial ordered state is stable. The magnetic interaction is not main cause for the appearance of the partial ordered state, the frustration. Other effects should be investigated.

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