Magnetic transitions on the pseudo-ternary compounds $Ho_{1-x}Y_xRh_2Si_2$

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Abstract. Magnetic measurements on the pseudo-ternary compounds $Ho_{1-x}Y_xRh_2Si_2$, 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_t, and T_{N2} 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; μ_{eff}=10.6 $\pm$0.15 μ_B/Ho, and magnetic moment μ=10 μ_B/Ho as well. The $Ho_{0.95}Y_{0.05}Rh_2Si_2$ single crystal shows the quite similar magnetic behaviour to that of $HoRh_2Si_2$. The constant magnetic moment and constant effective magnetic moment irrespective of *x* are confirmed from the magnetic behaviours of single crystals $Ho_{1-x}Y_xRh_2Si_2$ (x=0.05, 0.6 and 0.8).

1. Introduction

The ternary compound $HoRh_2Si_2$, which crystallizes in the tetragonal $ThCr_2Si_2$-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 $T_{N1}>T>T_{N2}$, 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_t=27.3 K [1]. The simple antiferromagnetic structure which is the propagation vector $\boldsymbol{k}$ = (0, 0, 1) with magnetic moments tilted from the *c*-axis by 28° 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 $T_{N2}<T<T_t$, 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 $Ho_{1-x}Gd_xRh_2Si_2$, 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 $Ho_{1-x}Y_xRh_2Si_2$, 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 $Ho_{1-x}Y_xRh_2Si_2$.

2. Experimental

Polycrystalline samples of pseudo-ternary compounds $Ho_{1-x}Y_xRh_2Si_2$ 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 *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 $Ho_{1-x}Y_xRh_2Si_2$ 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 $HoRh_2Si_2$ (x = 0) a = 4.026 Å, c = 9.899 Å, and a = 4.033 Å, c = 9.917 Å for YRh_2Si_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.

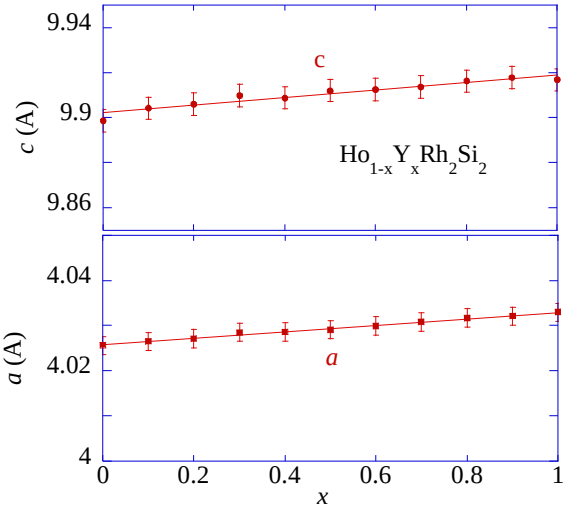

Figure. 1 Y composition x dependence of lattice parameters a and c on the polycrystalline compounds $Ho_{1-x}Y_xRh_2Si_2$.

3.2. Magnetic susceptibility

The temperature dependences of magnetic susceptibility along the main symmetry axes on the single crystals $Ho_{1-x}Y_xRh_2Si_2$ (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 $Ho_{0.95}Y_{0.05}Rh_2Si_2$ (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}=11.4 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 $HoRh_2Si_2$, although the transition temperatures become lower than those of $HoRh_2Si_2$ [1]; the successive component-separated magnetic transitions appear. The susceptibilities obey the Curie-Weiss law above 50 K. The χ^{-1}-T curves become linear and are almost parallel with each other at high temperatures. The effective magnetic moment estimated is 10.6 μ_B/Ho on the

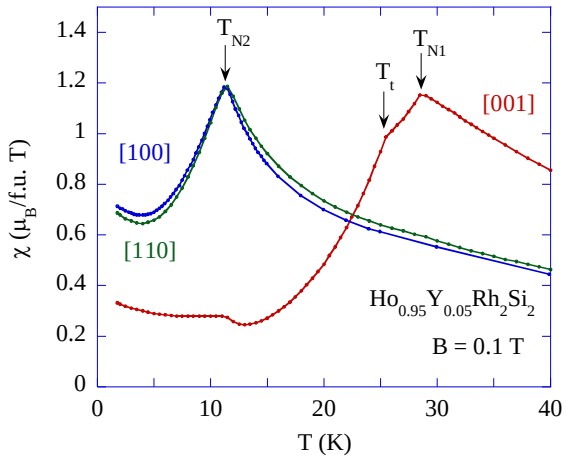

Figure. 2 Magnetic susceptibility along the main symmetry axes of tetragonal cell on the $Ho_{0.95}Y_{0.05}Rh_2Si_2$ (x=0.05) single crystal.

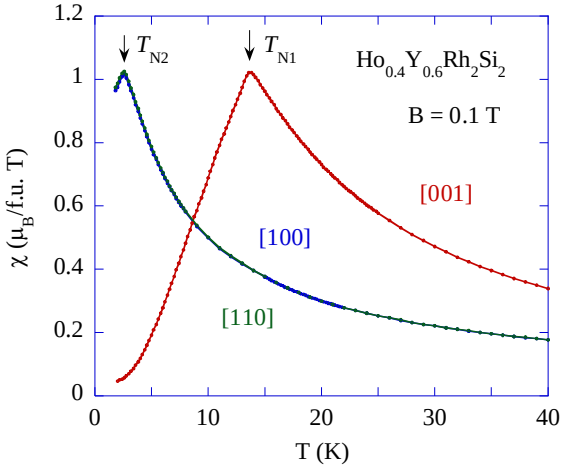

Figure. 3 Magnetic susceptibility along the main symmetry axes of tetragonal cell on the $Ho_{0.4}Y_{0.6}Rh_2Si_2$ (x=0.6) single crystal.

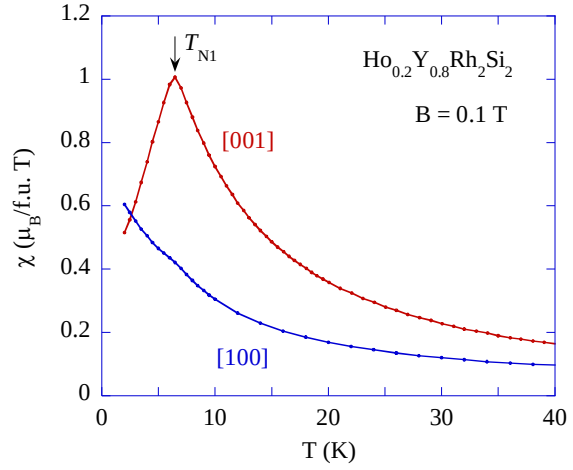

Figure. 4 Magnetic susceptibility along the main symmetry axes of tetragonal cell on the $Ho_{0.2}Y_{0.8}Rh_2Si_2$ (x=0.8) single crystal.

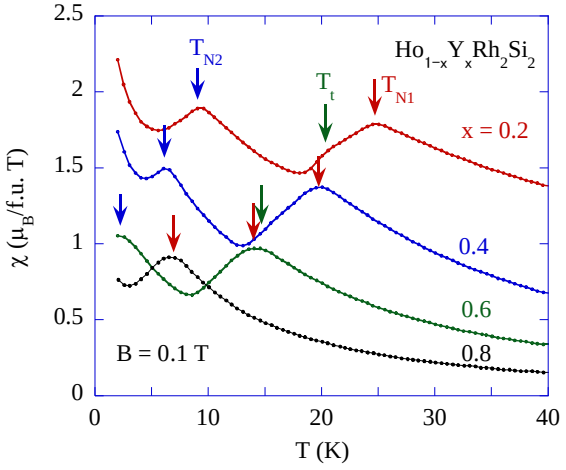

Figure. 5 Temperature dependence of magnetic susceptibility on the polycrystalline compounds $Ho_{1-x}Y_xRh_2Si_2$ (x =0.2, 0.4, 0.6 and 0.8)

compound which is in good agreement with the Ho^{3+} free ion magnetic moment ($g[J(J+1)]^{1/2}$=10.6 μ_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 Θ_{001}=18.5 K, Θ_{100}=-5.4 K and Θ_{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 $Ho_{1-x}Y_xRh_2Si_2$ (*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*: μ_{eff}=10.6 ±0.15 μ_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.

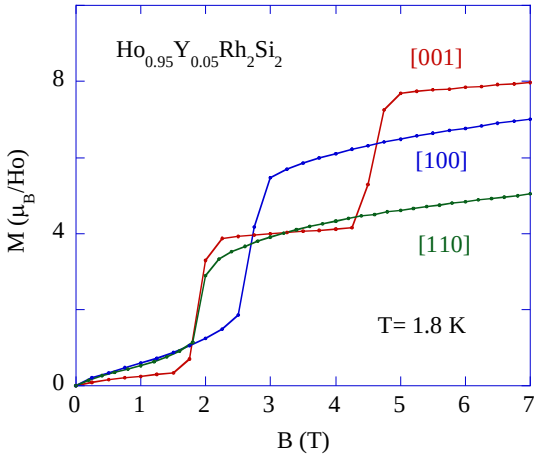

Figure. 6 Magnetization curves at 1.8 K along the main symmetry axes on the $Ho_{0.95}Y_{0.05}Rh_2Si_2$ (*x*=0.05) single crystal.

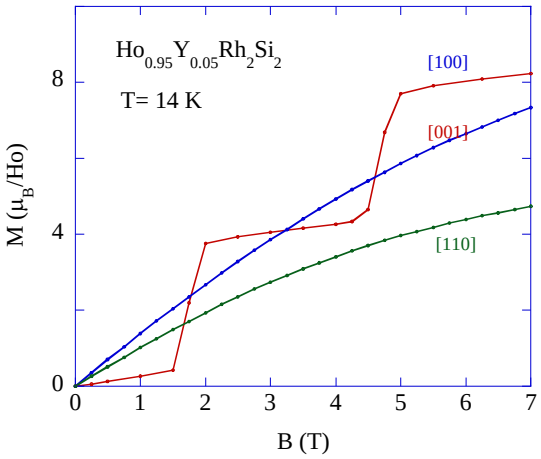

Figure. 7 Magnetization curves at 14 K along the main symmetry axes on the $Ho_{0.95}Y_{0.05}Rh_2Si_2$ (*x*=0.05) single crystal.

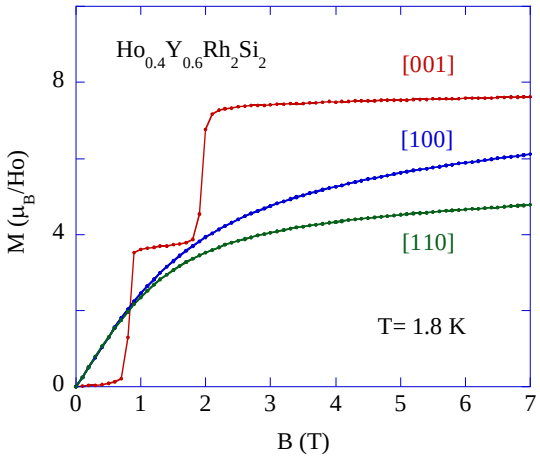

Figure. 8 Magnetization curves at 1.8 K along the main symmetry axes on the $Ho_{0.4}Y_{0.6}Rh_2Si_2$ (*x*=0.6) single crystal.

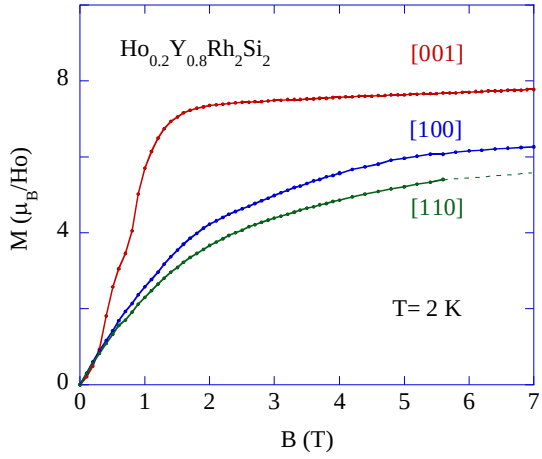

Figure. 9 Magnetization curves at 2 K along the main symmetry axes on the $Ho_{0.2}Y_{0.8}Rh_2Si_2$ (*x*=0.8) single crystal.

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 $Ho_{0.95}Y_{0.05}Rh_2Si_2$ 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 $HoRh_2Si_2$ [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 μ_B/Ho and M_2=8.0 μ_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 μ_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 μ_B/Ho by higher magnetic fields as the case of $HoRh_2Si_2$ [6]. A clear step-like metamagnetic transition also appears in the basal plan 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 μ_B/Ho and M_{110}=3.5 μ_B/Ho, respectively. The higher temperature behaviours shown in Fig. 7 are also quite similar to that of $HoRh_2Si_2$; 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 $Ho_{1-x}Y_xRh_2Si_2$ (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 μ_B/Ho and 7.8 μ_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 μ_B as same to that of $HoRh_2Si_2$. This means that the Ho magnetic moment keeps the theoretical Ho^{3+} magnetic moment (gJ=10 μ_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 $Ho_{1-x}Y_xRh_2Si_2$ compounds are shown in Fig. 10. The transitions were determined from measurements of χ-T above mentioned. The magnetic ordered state is divided to 3 states by the transition lines, which are

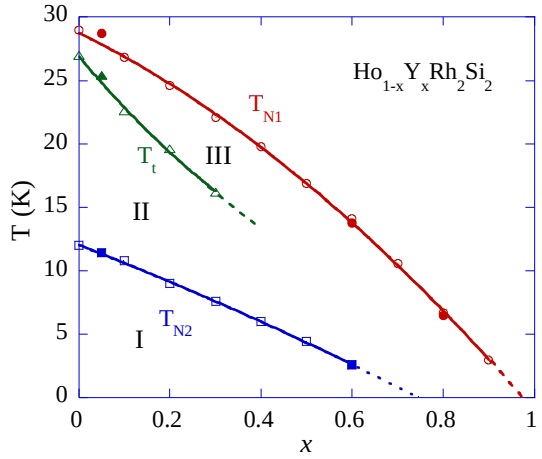

Figure. 10 Y composition x dependences of magnetic transition temperatures T_{N1}, T_t, and T_{N2} on the $Ho_{1-x}Y_xRh_2Si_2$ compounds. Closed and open symbols are transition temperatures determined from single crystal and polycrystalline samples, respectively. The lines are drawn by fitting to quadratic 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 $\boldsymbol{k}$ = (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_t, 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 Y) 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_t 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_t. 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: $(g\text{-}1)^2J(J+1)$. The *x*-dependency of T_t 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_t.

3.5. Summery

Magnetic studies on the pseudo-ternary compounds $Ho_{1-x}Y_xRh_2Si_2$, which substitute non-magnetic Y for Ho, were performed using either single crystal (x= 0.05, 0.6 and 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 $Ho_{0.95}Y_{0.05}Rh_2Si_2$ (x= 0.05) exhibit quite similar magnetic features to those of $HoRh_2Si_2$. 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 $Ho_{1-x}Y_xRh_2Si_2$ (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 $HoRh_2Si_2$; 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_t, 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; μ_{eff}=10.6 $\pm$0.15 μ_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. The successive component-separated magnetic transitions appear for wide *x* range up to x_{N2}=0.75. The *x*-dependency of T_t indicates the RKKY interaction may take a part of the cause for the transition of T_t. 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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