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Reentrant Double-Q Magnetic Structure in $HoRu_2Si_2$

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The reentrant two-step metamagnetic process of the tetragonal ternary compound $HoRu_2Si_2$ has been studied by neutron diffraction under magnetic fields on a single crystal. An antiferromagnetic structure at H=0 is characterized by the incommensurate propagation vector $\boldsymbol{Q}_1$=(0.206 0 0) below T_N. The field-induced phase is a double-$\boldsymbol{Q}$ structure with $\boldsymbol{Q}_1$ and $\boldsymbol{Q}_2$=(0 0.206 0). This double-$\boldsymbol{Q}$ structure appears in low and high temperature regions and disappears for intermediate temperature region, corresponding to the two-step metamagnetic process; a *reentrant double-$\boldsymbol{Q}$ structure* is observed.

KEYWORDS: $HoRu_2Si_2$ single crystal, neutron diffraction, metamagnetism, antiferromagnetism

§1. Introduction

The ternary rare earth compounds RRu_2Si_2 are one of the most interesting system among the RM_2X_2 (R = rare earth, M = 3d, 4d or 5d metal and X=Si, Ge) with the body-centered tetragonal structure of $ThCr_2Si_2$-type (space group *I4/mmm*) because of the great diversity of its magnetic properties. They show heavy fermion properties, ferromagnetism with giant magnetic anisotropy, a ferro-antiferromagnetic transition, multi-step metamagnetism, the existence of a mixed phase and so on, depending on the rare earth (R atom).[1-6] In particular, peculiar magnetic behavior of $HoRu_2Si_2$ has been reported recently from magnetic measurements on a single crystal;[7] 1) The compound orders antiferromagnetically below T_N=18.6 K with a huge magnetic anisotropy favoring the *c*-axis. 2) A metamagnetic process appears in the *c*-axis magnetization process, which changes with temperature; a two-step (with transition fields H_1=0.4 T and H_2=0.9 T), a one-step (H_1=0.7 T) and again a two-step (H_1=0.7 T and H_2=1.1 T) metamagnetic process appear for T<4 K, 4 K<T<6 K and 6 K<T<T_N, respectively. The temperature variation leads to a *reentrant two-step metamagnetic process* which is a peculiar behavior and has not been observed before. It was proposed that a magnetic instability should be responsible for this peculiar behavior. 3) There are at least three magnetic phases under the induced-ferromagnetic phase in the H_c-T phase magnetic diagram. Slaski *et al.* have reported that the antiferromagnetic structure is characterized by the commensurate propagation vector (1/5 0 0).[8]

In the present work, neutron diffraction study under H=0 and magnetic fields has been made on a $HoRu_2Si_2$ single crystal to determined the magnetic structures in the magnetic phase diagram and reveal the reentrant two-step metamagnetism.

§2. Experimental

Single crystals were grown by a tri-arc Czochralski method. The single phase nature was confirmed by the X-ray powder diffraction. The neutron diffraction experiments have been carried out in a double-axis mode with a triple-axis spectrometer, KUR-TAS, installed at the Research Reactor Institute, Kyoto University, using a wave length of 1.006 Å. The single crystal was mounted in a cryomagnetic system so that the c-axis was vertical. The magnetic field was applied along the *c*-axis with a superconducting magnet up to 5 T in the temperature range 2 K-20 K. Reciprocal lattice scans were performed in the a^*-b^* plane; reflections of (h k 0) type were collected.

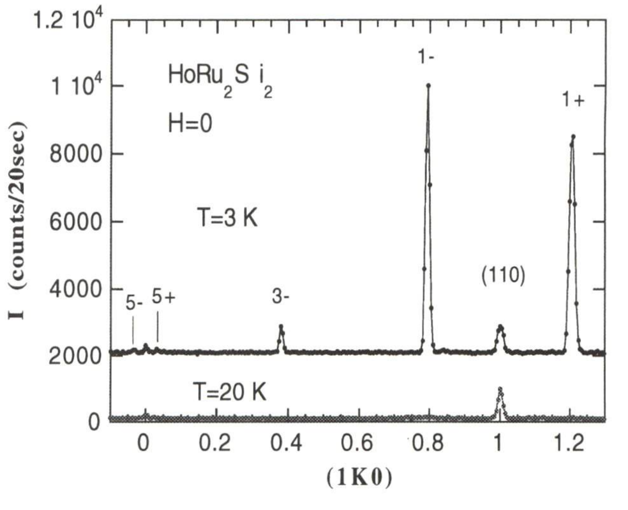

Fig. 1. Neutron diffraction patterns of reciprocal lattice k-scan along the [1 k 0] line around (1 1 0) under H=0 at 20 K, above T_N=18.6 K, and at 3 K, well below T_N on the $HoRu_2Si_2$ single crystal.

§3. Results and Discussion

Firstly, an antiferromagnetic structure is investigated; the results of neutron diffraction study under H=0 is presented. Figure 1 shows a neutron diffraction pattern of reciprocal lattice k-scan along the [1 k 0] line around (1 1 0) under H=0 at 20 K, above T_N=18.6 K, and at 3 K, well below T_N. In the pattern at 20 K, a nuclear reflection, the (1 1 0) reflection, is only observed. For low temperatures, antiferromagnetic reflections are observed, corresponding to the propagation vector $Q_1=(\tau\ 0\ 0)$ (τ=0.206) and their odd number higher harmonics. This result disagrees with the previous report by M. Slaski *et al.*[8] where the commensurate propagation vector (1/5 0 0) was proposed. It should be noted that τ, if Q_1 is commensurate, is 6/29. From comparison of observed and calculated neutron intensity (Table I, the reliability factor R=4.7 %), a long period commensurate anti-phase structure can be deduced at H=0 and low temperature; it is composed of (1 0 0) ferromagnetic planes with the " 0 4+ 5- 5+ 5- 5+ 4-" sequence along the [1 0 0] direction and full Ho^{3+} moments along the c-axis. Here 'n+ (n-)' stands for n successive ferromagnetic planes with up (down) moments and '0' stand for a zero moment plane. It should be emphasized that nonmagnetic planes appear at low temperature in the normal rare earth compound; the mixed phase, where magnetic and non-magnetic ions coexist, appears. Similar mixed phases have been previously evidenced in the isomorphous $TbRu_2Si_2$[6] and $TbRu_2Ge_2$.[9] The crucial role of the crystal field has been emphasized for the appearance of nonmagnetic ions. The nonmagnetic Ho could come from a singlet ground state due to the crystal-field effect, leading to a magnetic instability. The higher harmonics disappear around 13 K with increasing temperature; only fundamental reflections persist up to T_N, indicating that the antiferromagnetic structure becomes a sine-wave one above 13 K. This is a normal thermal behavior for a square-wave structure.

Next, neutron study under magnetic fields has been performed to make clear the two-step metamagnetic process at low temperature. Figures 2 and 3 show diffraction patterns of the reciprocal lattice k-scan along the [1 k 0] line and the q-scan along the [1 1 0] line around (1 1 0), respectively. No change of the fundamental propagation vector by application of fields is evidenced from Fig. 2; $Q_1=(\tau\ 0\ 0)$ (τ=0.206). In the field-induced intermediate phase (in the pattern of H=0.57 T), even number higher harmonics such as the (1 1-2τ 0) (1, 1-4τ, 0) peaks appear though very much weak intensity in addition to the odd ones. Ferromagnetic contribution, of course, is evidenced on the (1 1 0) nuclear peak. Along the [1 1 0] line (Fig. 3), new antiferromagnetic reflections appear above the transition field H_1, indexed by (1-τ 1-τ 0), (1-2τ 1-2τ 0) and (1-3τ 1-3τ 0) (τ=0.206) (Fig. 3). The magnetic field dependence of the (1 1-τ 0), (1-τ 1-τ 0) and (1 1-2τ 0) peak intensities are shown in Fig. 4. In the intermediate field region, the (1-τ 1-τ 0) reflection only develops . The (1-τ 1-τ 0) reflection

Table I. Comparision of the calculated and observed magnetic intensities at 3 K and H=0 T. τ=0.206

k in (1 k 0)	I_{obs} (barn)	I_{cal} (barn)
1+τ	32931	30862
1-τ	29247	31300
1-3τ	2533	3193
1-5τ	380	1064

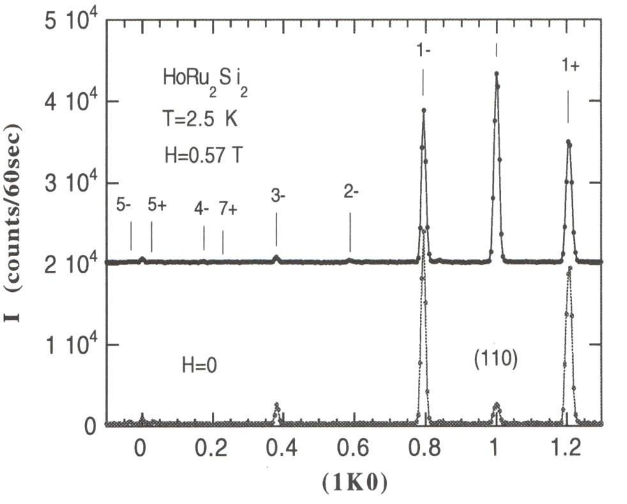

Fig. 2. Neutron diffraction patterns of reciprocal lattice k-scan along the [1 k 0] line around (1 1 0) under H=0 and H=0.57 T at T=2.5 K on the $HoRu_2Si_2$ single crystal. Number characters indicated, n, means the n-th order higher harmonics.

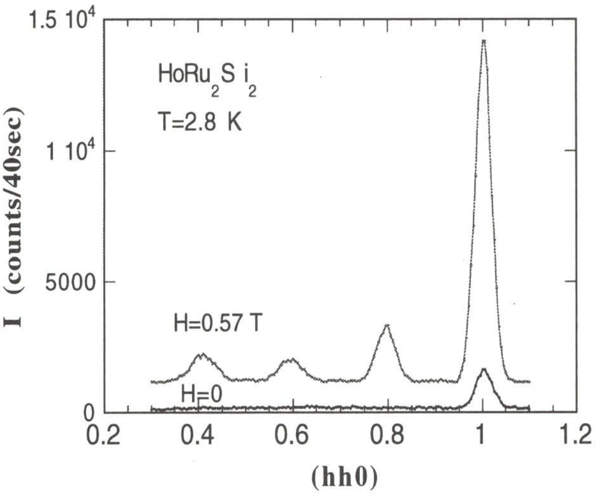

Fig. 3. Neutron diffraction patterns of reciprocal lattice q-scan along the [1 1 0] line around (1 1 0) under H=0 and H=0.57 T at T=2.8 K on the $HoRu_2Si_2$ single crystal.

is one of characteristic reflections for the field-induced intermediate phase. Moreover, other higher harmonics such as (1-$n\tau$ 1-$m\tau$ 0) (n, m=integer) develop, though the intensity is very much weak; magnetic reflections associated with the fundamental period τ distribute two-dimensionally all over the a^*-b^* plane. These results suggest that this intermediate phase is a ***double-Q structure*** with the propagation vector $Q_1=(\tau\ 0\ 0)$ and $Q_2=(0\ \tau\ 0)$ (τ=0.206). Unfortunately this magnetic structure has not been determined yet, because enough data to analyze the structure, precise intensity of all reflections, were not collected. In the antiferromagnetic region below H_1, the (1 1-2τ 0) intensity increases and the (1 1-τ 0) intensity decreases gradually with increasing temperature, meaning the square-wave structure is going to be modulate by magnetic field. Above H_2, all antiferromagnetic reflections disappear (as evidenced from Fig. 4) and ferromagnetic reflections only remain; a ferromagnetic phase is induced by the magnetic field. The two-step metam-

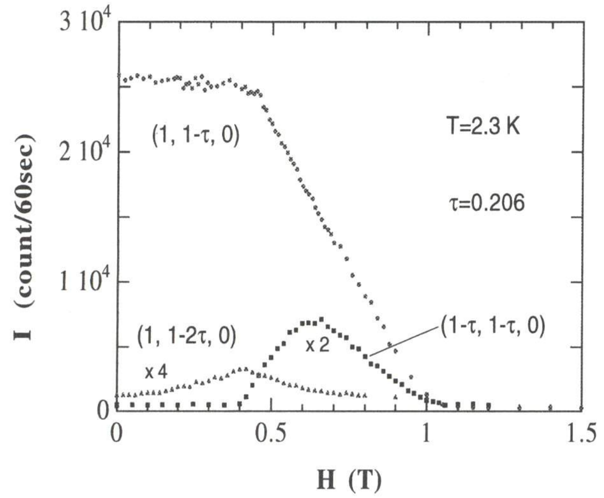

Fig. 4. Field dependence of peak intensities at antiferromagnetic reflections, (1 1-τ 0), (1-τ 1-τ 0) and (1 1-2τ 0) (τ=0.206), at T=2.3 K.

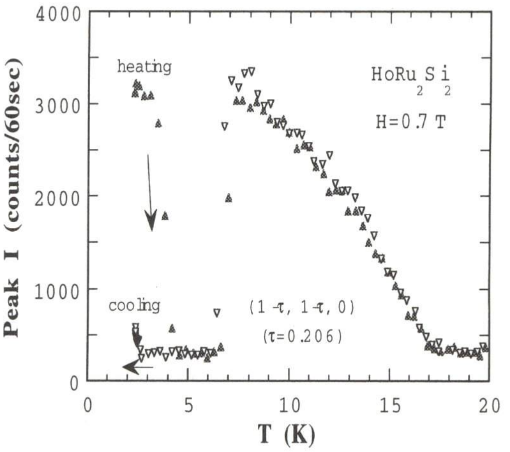

Fig. 5. Temperature dependence of the (1-τ 1-τ 0) (τ=0.206) peak intensity at H=0.7 T.

agnetic process at low temperature should be explained by change of magnetic structure from a single-$\boldsymbol{Q}$ antiferromagnetic one to a ferromagnetic one via a double-$\boldsymbol{Q}$ one.

Finally, thermal variation of magnetization process, the *reentrant two-step magnetization process*, was studied. Figure 5 shows the temperature dependence of the (1-τ 1-τ 0) peak intensity at H=0.7 T. At this applied magnetic field, the two-step metamagnetic processes appear at low (T<4 K) and high (6 K<T<T_N) temperatures. With increasing temperature, the (1-τ 1-τ 0) reflection, which characterizes a double-$\boldsymbol{Q}$ structure as mentioned above, disappears around 4 K and appears again around 6 K. On the other hand, no significant change in the (1 1-τ 0) peak intensity could be observed at even 4 K and 6 K though not shown in the figure; the intensity decreases continuously with increasing temperature. In the high temperature intermediate field region, any higher harmonics could not be detected. The magnetic structures of the intermediate field region are 1) in low temperatures, a double-$\boldsymbol{Q}$ one with $\boldsymbol{Q}_1$ and $\boldsymbol{Q}_2$ having higher harmonics, 2) in intermediate temperatures, a single-$\boldsymbol{Q}$ one with $\boldsymbol{Q}_1$ and 3) in high temperatures, a double-$\boldsymbol{Q}$ one without higher harmonics. A *reentrant double-$\boldsymbol{Q}$ structure*, therefore, could be concluded corresponding to the *reentrant two-step metamagnetic process*. It has been unknown yet whether there is a intrinsic difference between these double-$\boldsymbol{Q}$ ones or not. The reentrant behavior may come from a magnetic instability which could be attributed to the existence of nonmagnetic ion due to a singlet ground state.

In summary, the *reentrant two-step metamagnetic process* of $HoRu_2Si_2$ has been studied by neutron diffraction under magnetic fields. The following results are obtained; 1) In low magnetic fields, the antiferromagnetic structure is a *long period anti-phase* one with the $\boldsymbol{Q}_1$=(τ 0 0) (τ=0.206) at low temperature and a sine-wave one at high temperature. 2) In the intermediate field region, a double-$\boldsymbol{Q}$ structure with $\boldsymbol{Q}_1$ and Q_2=(0 τ 0), a single-$\boldsymbol{Q}$ one with $\boldsymbol{Q}_1$ and a double-$\boldsymbol{Q}$ one again appear with increasing temperature; a *reentrant double-$\boldsymbol{Q}$ structure*, very peculiar behavior which has been unknown yet. To understand this reentrant double-$\boldsymbol{Q}$ structure more clearly, the knowledge of crystal field effects, exchange interactions, the magnetic structures and so on, should be needed. Further measurements of neutron diffraction under magnetic fields and inelastic neutron scattering are planning.

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