Characteristic metamagnetic transitions in NpRhGa$_5$ and UPtGa$_5$

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Abstract. We measured the high-field magnetization up to 55 T and constructed a magnetic phase diagram for a transuranium antiferromagnet NpRhGa$_5$ with the tetragonal structure. The magnetization at 4.2 K for an easy-axis $H // [100]$ indicates a metamagnetic transition with a sharp step at $H_c = 26$ T and saturates above $H_s = 38$ T, reaching $0.43 \mu_B$/Np at $H_s$. On the other hand, the magnetization saturates at $H_s = 28$ T for a hard-axis $H // [001]$, reaching $0.44 \mu_B$/Np at $H_s$. The magnetization indicated that the magnetic moment reduces from $0.96 \mu_B$/Np at zero field to $0.44 \mu_B$/Np at the saturation field as a function of magnetic field. These experimental results in NpRhGa$_5$ are compared with the simple magnetism in UPtGa$_5$ with the similar antiferromagnetic structure.

1. Introduction
An interesting magnetic phenomenon was recently observed in NpRhGa$_5$ with two successive antiferromagnetic transitions at $T_{N1} = 36$ K and $T_{N2} = 32$ K [1, 2, 3]. In the temperature region from $T_{N1}$ to $T_{N2}$, named the AN1 phase, the magnetic structure is the same as that in NpCoGa$_5$ [4], where the magnetic moment in the ferromagnetically ordered (001) plane of the tetragonal structure is oriented along the [001] direction and is stacked antiferromagnetically along the [001] direction in a sequence of $(\uparrow \downarrow)$, as shown in Fig. 1(a). Below $T_{N2}$, named the AN2 phase, the direction of the magnetic moment changes abruptly from [001] to [110], as shown in Fig. 1(b). The corresponding magnetic moment changes at $T_{N2}$ in magnitude. In the AN2 phase, the ordered moment is determined as $0.96 \mu_B$/Np from the Mössbauer and neutron scattering experiments, while the ordered moment is roughly estimated as about $0.6 \mu_B$/Np under the assumption that the AN1 phase persists down to 0 K without the magnetic transition at $T_{N2}$. From the magnetic susceptibility, specific heat and neutron scattering measurements, it was clarified that both reorientation and enhancement of the magnetic moment occur at $T_{N2}$, with the first-order transition [1, 2, 3]. It is also noted that the 5f-itinerant band model is applicable to the electronic state in NpRhGa$_5$ [5]. Four kinds of cylindrical Fermi surfaces, which were
detected by the de Haas-van Alphen experiment, were well explained by the results of spin- and orbital-polarized 5$f$-itinerant band calculations.

To clarify furthermore the magnetic properties, we measured the magnetization in pulsed magnetic fields up to 55 T and constructed the magnetic phase diagram. We found a considerable reduction of the magnetic moment with increasing the magnetic field. These experimental results are compared with the simple magnetism in UPtGa$_5$ of which the magnetic structure is the same as the AN1 phase in NpRhGa$_5$.

2. Experimental

The high-quality single crystals of NpRhGa$_5$ and UPtGa$_5$ were grown by the self-flux method. The high-field magnetization was measured by the standard pick-up coil system by using the pulsed magnet with the pulse duration of 10 msec at Center for Quantum Science and Technology under Extreme Conditions (KYOKUGEN), Osaka University.

3. Experimental results

Figure 2 shows the typical magnetization curve at 4.2 K in the magnetic field along the [100] direction. The magnetization increases linearly with increasing the field, but indicates a sharp metamagnetic transition at $H_c = 26$ T, reaching about $0.3 \mu_B/Np$. With further increasing the field, the magnetization increases linearly and bends at $H_s = 38$ T. The magnetization amounts $0.43 \mu_B/Np$ at $H_s$. It is noted that the magnetization in magnetic fields from $H_c$ to $H_s$ is extrapolated to zero, as shown by a thin straight line in Fig. 2, with a slope of $6.1 \times 10^{-3}$ emu/mol, which is exactly the same as the magnetic susceptibility in the temperature range from $T_{N1}$ to $T_{N2}$ [2].

We measured the magnetization at various temperatures and constructed the magnetic phase diagram, as shown in Fig. 3. The data shown by squares are obtained by the SQUID magnetometer [2]. A curve connecting the $H_c$ data is smoothly connected the transition temperature $T_{N2} = 32$ K at zero field. Furthermore, a curve connecting the $H_s$ data also reaches the Néel temperature $T_{N1} = 36$ K at zero field. It is thus concluded that the bending field $H_s$ corresponds to the saturation field. This means that the antiferromagnetic AN1 state is changed into the paramagnetic (field-induced ferromagnetic) state above $H_s$. An increase of the magnetization above $H_s$ is most likely due to a Zeeman splitting of conduction bands with a relatively large density of states. The similar results is obtained in UPtGa$_5$, as shown later in Fig. 6.

Figure 4 shows the magnetization of NpRhGa$_5$ in the field along the [001] directions at 4.2 K. The magnetization increases linearly as a function of magnetic field and saturates above $H_s=28$ T, indicating $0.44 \mu_B/Np$ at $H_s$. This value is the same as the magnetic moment of $0.43 \mu_B/Np$ at $H_s$ for $H//[100]$.

We measured the magnetization at various temperatures and constructed the magnetic phase diagram for $H//[001]$, as shown in Fig. 5. The data below 7 T were also obtained by the SQUID
magnetometer [2]. It is noted that the AN1 phase is closed in a small $H$-$T$ region.

On the other hand, the magnetization is very simple in UPtGa$_5$, as shown in Fig. 6. The magnetization at 4.2 K indicates a metamagnetic transition for $H//[001]$ at $H_c=52$ T, reaching an ordered moment of 0.24 $\mu_B$/U at $H_c$. The corresponding magnetic phase diagram is shown in Fig. 7. An increase of the magnetization is observed above $H_c$, as in NpRhGa$_5$.

4. Discussions

We considered the magnetization process in the AN1 and AN2 phases on the basis of the magnetic structure mentioned above and the present magnetic phase diagram in NpRhGa$_5$. When a very small magnetic field is applied along [100], the magnetic moment with 0.96 $\mu_B$/Np, which is in the (001) plane and is oriented along [110] at zero field, is oriented perpendicularly to the direction of magnetic field. With increasing the magnetic field, the magnetic moment is tilted along the field direction. When the magnetic field reaches $H_c$, the orientation of the magnetic moment is changed from the (001) plane to the (100) plane. It is expected that
the magnitude of the magnetic moment is reduced into the AN1 phase above \( H_c \). Finally, the magnetic moment is oriented along the field direction above the saturation field \( H_s \). The present magnetic moment of 0.43 \( \mu_B/N_p \) at \( H_s \) is close to about 0.6 \( \mu_B/N_p \) estimated from the neutron scattering experiment under the assumption that the AN1 phase persists down to 0 K without the magnetic transition at \( T_{N1} \).

On the other hand, the magnetic moment, which is oriented along [110] at zero field, is tilted along the field direction of \( H/[001] \) as a function of the magnetic field and is oriented along [001] at the saturation field \( H_s=28 \) T. It is noticed that the magnetic moment reduces from 0.96 \( \mu_B/N_p \) at zero field to 0.44 \( \mu_B/N_p \) at \( H_s \).

The present reorientation and reduction of the magnetic moment in magnetic fields are thus closely related to the origin of the first-order phase transition at \( T_{N2} \). Recently, the magnetic transitions in NpTGa\(_5\) (T: Co, Rh, Ni) were discussed on the basis of the crystalline electric field (CEF) model based on the non-Kramers doublet ground state and a singlet excited state in the 5\( f^4 \) configuration, including dipolar and quadrupolar intersite interactions [6]. The quadrupolar interaction might play an important role of the present magnetization in high magnetic fields.

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