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NMR studies of the partially disordered state in a triangular antiferromagnet UNi_4B

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Abstract

A triangular antiferromagnet UNi_4B experiences a partially disordered state, in which two-thirds of U 5f moments order in a vertex-like structure and one-third of U 5f moments remain paramagnetic. Magnetization and NMR measurements of UNi_4B have been performed to study the dynamical properties of U magnetic moments in the partially disordered state. The value of the effective moment above T_N derived from the fitting of the Curie–Weiss law suggests a significant crystal field splitting and the Kondo effect. The Weiss temperature was also estimated to be −28 K, which suggests that the exchange interaction is antiferromagnetic. Furthermore, the exchange interaction estimated from the relaxation rate T_1^{-1} above 50 K is 18 K, which is close to the Weiss temperature. These results suggest that the dominant interactions between U moments are antiferromagnetic in the basal plane. The relaxation rate T_1^{-1} decreases abruptly below T_N down to 15 K and is almost constant between 10 and 15 K. In the same temperature range, the magnetic susceptibility still increases with decreasing temperature. However, the effective moment estimated below T_N is almost the same as that above T_N. This shows that there are anomalous fluctuations below T_N which cannot be explained by the simple model.

1. Introduction

Geometrical frustration in f electron systems has attracted much interest recently because peculiar properties are expected due to the coexistence of itinerancy and frustration [1]. One of the most remarkable properties is a partially disordered state. The compound UNi_4B is a unique 5f electron system which shows a partially disordered state down to the lowest temperature. CePdAl, which is 4f electron system on a distorted kagome lattice, also shows a partially disordered state [2]. The magnetic structure of UNi_4B below T_N (20 K) was reported to be a

vortex-like structure of two-thirds of uranium 5f moments and one-third disordered uranium 5f moments [3]. It is believed that the peculiar magnetic structure originates from the geometrical frustration of the triangular arrangement of U atoms. Reported anomalous properties—a power law of the temperature dependence of the resistivity ($T^{0.11}$), an increase of the susceptibility below T_{N} and an increase of the value of the specific heat divided by temperature (C/T) at low temperature—were ascribed to a one-dimensional ferromagnetic chain [4]. Some experimental results were compared with the theoretical consequences of a one-dimensional ferromagnetic chain [5]. The resistivity and specific heat suggest that non-Fermi liquid states are realized at low temperature. The relation between the non-Fermi liquid states and frustration or the peculiar magnetic structure is attracting much attention.

In UNi_4B, uranium atoms form a triangular arrangement in the basal plane. The interplane distance between U atoms is much smaller than the intraplane distance. This suggests that there are strong exchange interactions between basal planes. Three theoretical models have been proposed to explain the magnetic structure. All models assume strong exchange interactions between basal planes. Lacroix *et al* proposed a model Hamiltonian with the Kondo effect and an anisotropy [6]. The other models can be regarded as localized models. Mentink *et al* proposed a model Hamiltonian with a strong ferromagnetic exchange interaction between the basal planes, second nearest neighbour antiferromagnetic interactions in the basal plane and an anisotropy in the basal plane with sixfold rotational symmetry [4]. Tejima *et al* proposed two kinds of nearest neighbour interactions: one is positive and the other is negative, without any anisotropies [7]. The origin of the magnetic structure is still controversial. In this paper we report magnetization and NMR measurements for investigating the dynamical properties of U 5f moments.

2. Experimental procedures

Powder samples were prepared by melting the starting materials in an arc furnace. Single crystals were grown by the Czochralski method using a tetra-arc furnace. The crystal structure of UNi_4B was determined with an x-ray diffraction spectrometer using the single crystal. Magnetization measurements of single crystals were performed by a SQUID magnetometer between 2 and 300 K. Furthermore the angle dependence of the magnetization was measured using a sample rotator with the single crystal. ^{11}B NMR measurements of powder samples were performed with a coherent pulsed spectrometer. Free powders in a epoxy resin tube were used so that the powders were expected to be aligned with the easy plane in the magnetic field. Actually the susceptibility of the free powder has almost the same temperature dependence as the susceptibility with the magnetic field applied along the a-axis. Spin echo spectra were measured by sweeping the magnetic field to determine the Knight shifts. The relaxation rates T_1^{-1} were determined by fitting the recovery curves of the integrated spin echo intensities to the equation for $I = 3/2$ as

$$\frac{M_0 - M(t)}{M_0} = a_1 \exp(-t/T_1) + a_2 \exp(-6t/T_1). \tag{1}$$

3. Results and discussions

First we briefly mention the crystal structure of UNi_4B. Mentink *et al* have reported that the crystal structure of this compound is of the hexagonal $CeCo_4B$-type with space group $P6/mmm$. However, it was also reported that there is a superstructure due to the small displacement of Ni and B atoms, which makes the unit cell nine times larger. Our x-ray

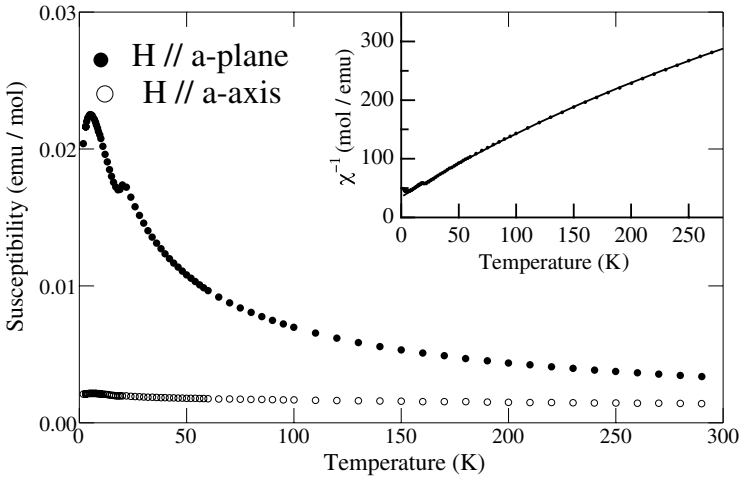

Figure 1. Temperature dependence of the susceptibilities of a single crystal with the applied field along the a-axis and a-plane between 1.5 and 280 K. The inset shows the temperature dependence of the inverse of the susceptibility with the applied field along the a-plane. The solid line in the inset is the fitting curve using a modified Curie–Weiss law.

diffraction studies revealed that the structure is actually C-centred orthorhombic with space group $Cmcm$. The crystal parameters are $a = 6.966$ Å, $b = 17.121$ Å and $c = 14.865$ Å and there are four non-equivalent U sites. Surprisingly, the triangular arrangement of U atoms is only slightly modified and an almost hexagonal structure still remains in the a-plane. Therefore we can regard the system as a triangular lattice.

Figure 1 shows the temperature dependence of the susceptibilities χ of a single crystal with applied field along the a-axis and the a-plane. A large anisotropy is easily seen and the easy axis is in the a-plane. There is a kink at $T_{\mathbf{N}} = 20$ K. The inset shows the temperature dependence of χ^{-1} in the applied field along the a-plane. The solid line shows the fitting curve using the modified Curie–Weiss law:

$$\chi = \chi_0 + C/(T - \theta), \tag{2}$$

where χ_0 a Van Vleck term, C the Curie constant and θ is the Weiss temperature. From the fitting, we estimate the parameters $\chi_0 = 9.7 \times 10^{-4}$ (emu mol^{-1}), $\theta = -28$ K and $\mu_{\text{eff}} = 2.48\ \mu_{\mathbf{B}}$, where μ_{eff} is the effective moment per uranium atom. The expected value of μ_{eff} is 3.58 $\mu_{\mathbf{B}}$ for the $5f^2$ or 3.62 $\mu_{\mathbf{B}}$ for the $5f^3$ configuration of U atoms. The smallness of the estimated μ_{eff} suggests significant crystal field splittings. The estimated θ is ascribed to the antiferromagnetic interaction between the spins in the ground states of the crystal field splitting levels. On the other hand, the magnitude of the ordered moments was reported to be 1.2 $\mu_{\mathbf{B}}$ through the neutron diffraction measurements [3], which is significantly smaller than the μ_{eff} estimated from the susceptibility in the paramagnetic region. This suggests a reduction of the magnetic moments due to the Kondo effect. Actually the temperature dependence of resistivity shows a logarithmic behaviour, which is typical behaviour for the Kondo effect. The estimated θ is comparable to the value of 35 K proposed by Mentink *et al*. However, the sign of the θ is different from that of the value proposed by Mentink. They proposed that there is strong ferromagnetic interaction along the a-axis, which may be responsible for the one-dimensional ferromagnetic chain along a-axis. The negative θ may be compatible with the ferromagnetic interactions if we ascribed the θ to the Kondo effect. However, the possibility that the dominant interaction is antiferromagnetic is not denied. If we perform the same fitting as equation (1) to

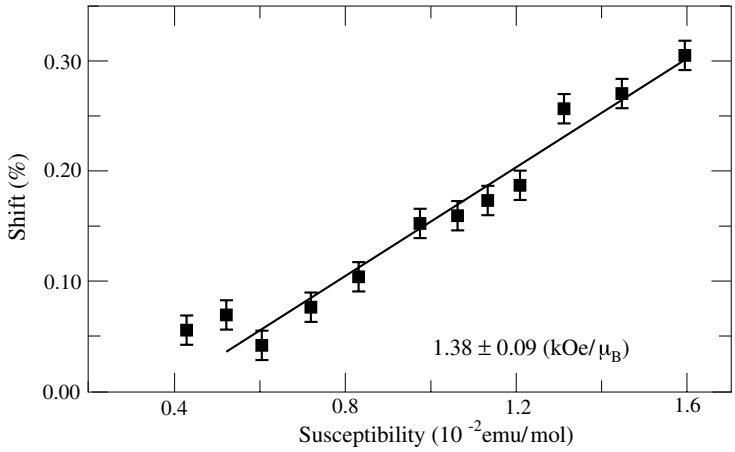

Figure 2. Knight shift versus susceptibility plot between 20 and 280 K.

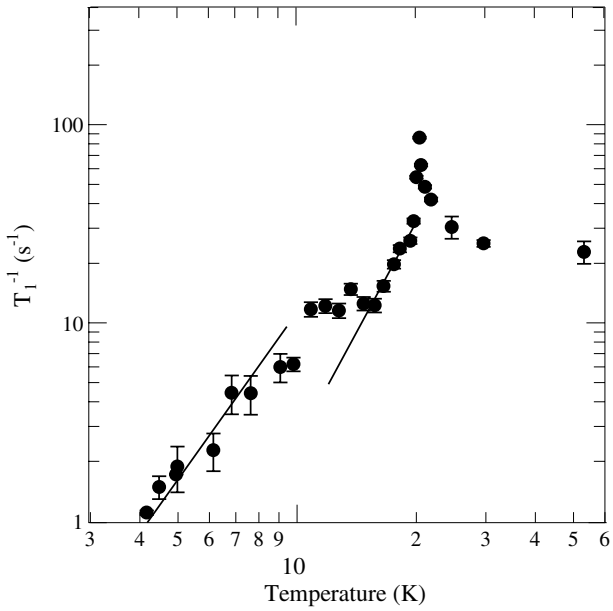

Figure 3. Temperature dependence of the relaxation rate between 4.2 and 50 K. The inset shows the Knight shift around its maximum.

the anomalous increase of χ below $T_{\mathbf{N}}$, the effective moment below $T_{\mathbf{N}}$ seems to be almost the same as that above $T_{\mathbf{N}}$. This result may be inconsistent with the interpretation that the system is composed of the two individual parts of the ordered and the paramagnetic U moments. We will discuss this point later.

Figure 2 shows the Knight shift versus susceptibility plot. By fitting the plot to a linear relation, we obtained the hyperfine coupling constant to be 1.38 kOe/$\mu_{\mathbf{B}}$. This value is a little larger than the value of 1.14 kOe/$\mu_{\mathbf{B}}$ reported by Liu *et al* [8].

Figure 3 shows the temperature dependence of the relaxation rate T_1^{-1}. The relaxation rate T_1^{-1} shows a remarkable peak at 20 K, demonstrating the critical slowing down of fluctuations

of U moments at the magnetic phase transition. Above 50 K, T_1^{-1} is almost constant. If the relaxation of nuclear spins is dominated by the fluctuations of localized U moments, the constant relaxation rate is expressed as

$$T_1^{-1} = \sqrt{2\pi}\, g_J^2\, \gamma_N^2 \frac{A_{hf}^2}{z_0} \frac{J(J+1)}{3\omega_{ex}}, \tag{3}$$

$$\omega_{ex}^2 = 8z|J_{ex}|^2 \frac{J(J+1)}{3\hbar^2}, \tag{4}$$

where g_J is the de Gennes factor, γ_N is the nuclear gyromagnetic ratio, A_{hf} is the hyperfine coupling constant, J is the total angular momentum of the U moment and J_{ex} is the exchange interaction between U moments. Furthermore, z_0 and z mean the number of the neighbouring U moments around a B atom and the number of neighbouring U moments around a U moment, respectively. As the interplane distance is much smaller, we assume $z = 2$. The number z_0 is 3 for the crystal structure of this compound. Using these relations we estimate that the exchange interaction between U moments along the a-axis is about 25 K. This value is close to the estimated θ, although the estimation from the T_1^{-1} cannot determine the sign of the interaction.

Measurements of the angle dependence of the magnetization showed that the anisotropy in the a-plane is rather small so that the stabilization of the vertex-like magnetic structure by the anisotropy, which was proposed by Mentink *et al*, is not plausible. Consequently, the strong ferromagnetic interaction between the basal planes is not definitive. Alternatively, we propose that the dominant interaction in the a-plane is antiferromagnetic. In this case strong frustration exists in the triangular lattice.

We pointed out that the effective moment derived from the anomalous increase of χ below T_N is almost the same as that of T_N. The relaxation rate T_1^{-1} shows an abrupt decrease between 20 and 15 K with decreasing temperature but is almost constant between 15 and 10 K. The temperature range between 15 and 10 K is where the anomalous increase of χ is observed. However, the simple explanation assuming the localized moments of the one-third U that are paramagnetic fails because the effective moment is anomalously large. Our results show that the magnetic properties of this compound should be reconsidered.

In summary, we measured the magnetization of the single crystals and the Knight shift and relaxation rates of the powder samples to study the dynamics of 5f moments in UNi_4B. The value of the effective moment above T_N suggests significant crystal field splitting and the Kondo effect. The sign of the Weiss temperature is negative, which is in contrast to the previous proposal. We propose that the dominant interaction is antiferromagnetic in the a-plane. The estimated exchange interaction from T_1^{-1} is roughly consistent with the Weiss temperature. The effective moment below T_N is almost the same as that above T_N. Furthermore, T_1^{-1} suggests the anomalous fluctuations of U moments in the same temperature range. Further investigations of the fluctuations below T_N and the interactions between U moments are required.

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