Spin dynamics in a triangular antiferromagnet UNi$_4$B

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Abstract.
$^{11}$B NMR and magnetic susceptibility measurements have been performed in order to investigate the spin dynamics of a triangular-lattice XY antiferromagnet UNi$_4$B. In this compound, the partially ordered state appears below $T_N = 20$ K with a peculiar vortex-like structure. Magnetic susceptibility shows that $T_N$ is slightly lower than the round maximum of the magnetic susceptibility suggesting the two dimensional character of this compound. The temperature dependence of the relaxation rate $T^{-1}$ above $T_N$ is well reproduced by the theoretical model that were discussed in the Kosterlitz-Thouless-Berezinskii transition. Thus the temperature dependence of $T^{-1}$ suggests that the spin dynamics in UNi$_4$B above $T_N$ is dominated by the motion of the unbound vortices.

1. Introduction
Coexistence of frustration and itineracy is one of the most interesting issue recently. Such an example, a uranium compound UNi$_4$B, is a triangular-lattice antiferromagnet which has geometrical frustration, strong easy plane type anisotropy and Kondo effect.

The magnetic properties of this compound are characterized by the antiferromagnetic ordering at $T_N = 20$ K with a peculiar vortex-like structure [1]. In this magnetic ordered state, U moments are partially ordered and 1/3 of U moments remain paramagnetic. It is believed that the partially ordered state in this compound is originated from the geometrical frustration of the triangular arrangement of U atoms. The similar partially ordered state has been observed in a 4f electron system CePdAl [2] [3]. Recent X-ray diffraction studies have revealed that the crystal structure is actually C-centered orthorhombic with space group Cmcm [4]. The crystal parameters are $a = 6.966\AA$, $b = 17.121\AA$ and $c = 14.865\AA$ and the triangular-lattice plane is the $a$-plane. Slight distortion of the triangular lattice results in four non-equivalent U sites. The triangular arrangement of U atoms is only slightly modified and almost hexagonal structure still remains in the $a$-plane. Therefore we can regard the system as the triangular lattice.

Three theoretical models were proposed to explain the peculiar magnetic structure. All models assume the strong ferromagnetic interactions between $a$-planes as the interplane distance between U atoms (3.5 A) is much smaller than that of intraplane distance (5.0 A) and the magnetic structure has ferromagnetic arrangements along $a$-axis. The strong interaction along the $a$-axis leads to one dimensional ferromagnetically correlated chains. We can regard the compound as the two dimensional triangular lattice made of the correlated chains, which may offer two dimensional character of the magnetic properties. The first model consists of the first nearest neighbor interaction $J_1$, the second nearest neighbor interaction $J_2$, strong anisotropy
in the $a$-plane with the sixfold rotational symmetry and Kondo effect [5]. The second model consists of only $J_2$ and the similar strong anisotropy to the first model [6]. The third model consists of two kinds of $J_1$, which are positive and negative, and the strong ferromagnetic interaction between the $a$-plane, without any anisotropies [7]. These three models can reproduce the peculiar magnetic structure respectively. Therefore we can not conclude the origin of the magnetic structure. On the other hand, our magnetization measurements showed that there is no strong anisotropy in the $a$-plane [8]. Furthermore the specific heat measurements showed the existence of another phase transition at 0.3 K [9]. No model explain the second transition at lower temperature. Thus the origin of the magnetic structure is still controversial.

If the magnetic properties have two dimensional character, the strong spin correlations above $T_N$ are expected. Such spin fluctuations may affect the magnetic structure. We report the magnetization and NMR measurements to investigate the spin dynamics of U $5f$ moments.

2. Experimental Procedures

Magnetization measurements of the single crystal were performed by SQUID magnetometer between 2 K and 300 K. $^{11}$B NMR measurements of single crystals were performed by a coherent pulsed spectrometer. Eight pieces of single crystals were fixed in the sample holder so that the $a$-axis ,which is perpendicular to the triangular lattice plane, of each single crystal is aligned to the applied magnetic field. Spin echo spectra were measured by fast Fourier transformation of the spin echo signal. The relaxation rates $T_1^{-1}$ were determined by the fitting of the recovery curves with the same procedure as shown in ref. [8].

3. Results and Discussions

Figure 1 shows the temperature dependence of the inverse of the susceptibility ($\chi^{-1}$) of a single crystal between 2 K and 300 K. The magnetic field is applied along the $a$-axis and the $a$-plane. The large anisotropy and the negative Weiss tempetarure are easily seen. We try to fit the data by the crystalline electric field (CEF) Hamiltonian. We assume $L - S$ coupling scheme and the hexagonal symmetry for simplicity. The specific heat measurements showed that the degeneracy of the ground state is doublet. Therefore we assume U$^{3+}$ that leads to 5f$^2$ state so that the system have Kramers doublet. The CEF Hamiltonian is expressed for the hexagonal symmetry as

$$H = B_{1}^{0}O_{1}^{0} + B_{2}^{0}O_{2}^{0} + B_{4}^{0}O_{4}^{0} + B_{6}^{0}O_{6}^{0},$$

where $O_{i}^{0}$ etc. are Steven’s operator equivalents and $B_{i}^{0}$ etc. are the fitting parameters. The lines in Fig.1 show the fitting curves using the Hamiltonian. The parameters are $B_{1}^{0} = 1$ K, $B_{4}^{0} = -0.135$ K, $B_{6}^{0} = 0.02$ K and $B_{6}^{0} = 0.3$ K and the molecular fields are $\lambda_x = -50$ emu/mol and $\lambda_z = -520$ emu/mol. This fitting demonstrates the U moments are localized at high temperatures and the strong easy plane type anisotropy is explained by the CEF and the anisotropic molecular fields. One problem is that the negative molecular field means that dominant interactions are antiferromagnetic although the strong ferromagnetic interactions between the $a$-plane were assumed. Mentink et al. proposed that the discrepancy may come from the CEF effect. However our calculation shows that CEF can not explain the discrepancy. One possibility is that the magnetic interaction between U moments are long range RKKY interactions. The oscillating nature of the RKKY interactions may originate the negative molecular field and the ferromagnetic interactions between the $a$-plane.

Figure 2 shows the temperature dependence of the susceptibility ($\chi$) and $d\chi/dT$ around $T_N$. The susceptibility $\chi$ shows the broad maximum around $T_N$ and decrease with decreasing temperature and start to increase again below 19 K. The transition temperature, which is determined by the peak position of $d\chi/dT$, is significantly lower than the temperature where $\chi$ shows a maximum. This suggests the low dimensional character of the transition. The similar behavior is also observed in CePdAl [3].
Figure 1. Temperature dependence of the inverse of the susceptibility of a single crystal. The solid lines are the fitting curve using the crystalline electric field Hamiltonian.

Figure 2. Temperature dependence of $\chi$ and $d\chi/dT$. Magnetic field was applied in the a-plane.

Here we discuss the temperature dependence of the relaxation rate ($T_1^{-1}$). The increase of $T_1^{-1}$ starts from the temperature which is much higher than $T_N$. The temperature dependence is roughly explained by the power law of $(T - T_N)/T_N$, which is the indication of the critical behavior. However, the temperature range is far beyond the critical region. In two dimensional Heisenberg model, the temperature dependence of the correlation length is exponential that leads to the exponential temperature dependence of $T_1^{-1}$ [10]. Figure 4 shows the $T_1^{-1}$ vs $1/T$ plot. The linear dependence is only seen at very close to $T_N$ meaning that the temperature range where $T_1^{-1}$ obeys the exponential temperature dependence is narrow. Alternatively, we note that the strong easy plane type anisotropy of this compound and two dimensional nature offered by the strong correlations between U moments. Furthermore recent NMR study revealed that the rate $T_1^{-1}$ in a triangular antiferromagnet LiCrO$_2$ is well reproduced by the theoretical model with unbound vortices, that were discussed in the Kosterlitz-Thouless-Berezinskii transition [11]. If the motion of the unbound vortices dominate the relaxation of nuclear spins, $T_1^{-1}$ for $T > T_N$ is expressed as follows [12], [13].

$$\frac{1}{T_1} = \alpha \left( \frac{\gamma g H_{hf}}{z'} \right)^2 \frac{z'}{\sqrt{\pi} n_\nu U} + \beta T \tag{2}$$

$$n_\nu = \left( \frac{1}{2\xi_0} \right)^2 exp\left( \frac{-2b}{\sqrt{\tau}} \right) \tag{3}$$

$$U = \left( \frac{\pi}{2} \frac{JS^2a^2}{\hbar} \right)^2 n_\nu \ln\left( \frac{k_BT_k}{JS^2n_\nu a^2} \right) \tag{4}$$

The first term of eq.(2) means the contribution from the unbound vortices. $\gamma$ is the gyromagnetic ratio for $^{11}$B nucleus, $H_{hf} = 0.7$ kOe/$\mu_B$ is the hyperfine field and $z' = 3$ is the number of the magnetic moments around a $^{11}$B nucleus. $n_\nu$ in eq.(3) is the density of unbound vortices. $\tau$ is $(T - T_{KB})/T_{KB}$, $\xi_0$ is the lattice parameter and $b$ is the scaling parameter less than $\pi/2$. $U$ in eq.(4) is the velocity of the vortices and $J$ is the exchange interaction between U moments. The temperature dependence of $T_1^{-1}$ is well reproduced by the theoretical model as shown in Fig.3. The solid line is the theoretical curve with $J = -2.5$ K, $T_{KB} = 20.04$ K and
$\beta = 0$. Thus the temperature dependence of $T^{-1}_1$ suggests that the spin dynamics above $T_N$ is dominated by the motion of the unbound vortices.

In the case of the XY antiferromagnet on a triangular lattice, the relevant symmetries of the order parameter are $Z_2$, which is ascribed to the frustration and $SO(2)$. Whether the phase transition of $Z_2$ vortex and KBT transition occur at the same temperature or not is controversial [14]. The typical compound CsMnBr$_3$ has only one phase transition and the new universality class was proposed [15]. In the case of UNi$_4$B, the frustration may be released partially because of the slight distortion of the triangular lattice and the temperature of the phase transition of $Z_2$ vortex becomes lower. Then spin dynamics is dominated by vortices associated with the KTB transition. As the correlated chains along the $a$-axis form a triangular lattice, three dimensional magnetic order occurs at $T_N$. The study on the second phase transition may clarify the situation of the frustration in UNi$_4$B.

![Figure 3. $T^{-1}_1$ vs $1/T$ plot.](image3)

![Figure 4. $T^{-1}_1$ vs $(T - T_{KTB})/T_{KBT}$ plot.](image4)

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**References**

[1] Mentink S, Drost A, Nieuwenhuys G, Frikkee E, Menovsky A and Mydosh J 1994 *Phys. Rev. Lett.* 73 1031
[2] Dönni A, Ehlers G, Maletta H, Fischer P, Kitazawa H and Zolliker M 1996 *J. Phys.: Condens. Mat.* 8 11213
[3] Oyamada A, Maegawa S, Nishiyama M, Kitazawa H and Isikawa Y 2008 *Phys. Rev. B* 77 064432
[4] Haga Y, Oyamada A, Matsuda T, Ikeda S and Onuki Y 2008 to be published in Physica B
[5] Lacroix C, Canals B and Nunez-Regueiro M 1996 *Phys. Rev. Lett.* 77 5126
[6] Mentink S, Nieuwenhuys G, Nakotte G, Menovsky A, Drost A, Frikkee E and Mydosh J 1995 *Phys. Rev. B* 51 11567
[7] Tejima S and Oguchi A 1997 *J. Phys. Soc. Jpn.* 66 3611
[8] Oyamada A, Kondo M, Fukunaka K, Itou T, Maegawa S, Li D X and Haga Y 2007 *J. Phys.: Condens. Matter* 19 145246
[9] Movshovich R, Jaime M, Menovsky S M A and Mydosh J 1999 *Phys. Rev. Lett.* 83 2065
[10] Chakravarty S and Orbach R 1999 *Phys. Rev. Lett.* 64 234
[11] Alexander L, Buttgen N, Nath R, Mahajan A and Loi dl A 2007 *Phys. Rev. B* 76 064429
[12] Mertens F, Bishop A and Wysin G 1989 *Phys. Rev. B* 39 591
[13] Huber D 1982 *Phys. Rev. B* 26 3758
[14] Miyashita S and Shiba H 1984 *J. Phys. Soc. Jpn.* 53 1145
[15] Kawanura H 1986 *J. Phys. Soc. Jpn.* 55 2095