NMR Study of Magnetic Structures in NH_4CuCl_3

Yoshiyuki Shimaoka [a,1], Takao Goto [a], Katsuaki Kodama [b], Masashi Takigawa [b], Hidekazu Tanaka [c]

[a] *Graduate school of Human and Environmental Studies, Kyoto University, Sakyo-ku, Kyoto 606-8501, Japan*
[b] *Institute for Solid State Physics, The University of Tokyo, Kashiwa 277-8581*
[c] *Department of Physics, Tokyo Institute of Technology, Oh-okayama, Megro-ku, Tokyo 152-8551,*

Abstract

To study magnetization plateau in NH_4CuCl_3, nuclear magnetic resonance (NMR) experiments were performed under several magnetic fields which correspond to the region below, on, and above the first plateau. From the result of temperature dependence of ^{15}N-NMR spectra and spin-lattice relaxation rate T_1^{-1} around 3 T and 6 T, it is proposed that magnetic moments are induced on one Cu-dimer site per every four dimers along crystallographic a-axis at low temperature. This magnetic structure seems to remain up to about 8.5 K and then change to other structures with increasing temperature. It is also revealed that there exists 3D antiferromagnetic ordering below and above the first plateau region at low temperature. These results indicate that this sytem shows several magnetic structures depending on the external field and the temperature.

Key words: magnetization plateaus; NH_4CuCl_3; NMR

1. Introduction

Recentry, the problem of magnetization plateaus observed in high-field magnetization process of low-dimensional magnetic systems has been the subject of great concern in view of quantum effects.

Among typical candidate compounds for the study of this problem is monoclinic NH_4CuCl_3, in which the magnetization curve exhibits two plateaus with 1/4 and 3/4 of the saturated value at low temperature corresponding to five successive transitions at $H_{C1} = 5.0$ T, $H_{C2} = 12.8$ T, $H_{C3} = 17.9$ T, $H_{C4} = 24.7$ T, and $H_{C5} = 29.1$ T (for $H \parallel a$-axis)[1].

The crystallographic structure of NH_4CuCl_3 belongs to space group $P2_1/c$ as the isomorphas $KCuCl_3$ and $TlCuCl_3$ at room temperature[2]. It contains planar $Cu_2Cl_6^{2-}$ dimer which stacks and forms S=1/2 double spin chains along the crystallographic a-axis, and the cation NH_4^+ is situated between these spin chains. The proposed intra- and inter-chain interactions between each dimer have triangular configurations which may cause frustration effect. The inter-chain interactions form zig-zag ladder configuration. The intra-chain interactions, which are considered to be dominant along $(1, 0, \bar{2})$ plane as $TlCuCl_3$, form the configuration similar to the Shastry-Sutherland model[3].

We are interested in the magnetic structure and spin dynamics of this compound. First, we performed ^{14}N(I=1)-NMR measurement in high-field region. Next, by synthesizing ^{15}N(I=1/2)-enriched single crystal, we measured the temperature dependence of the NMR spectra and the longitudinal relaxation time T_1 from 1.4 K to 120 K and around $H = 3$ T and 6 T, which correspond to different field region below and on the first(1/4) plateau of the magnetization curve. The direction of the applied fields was always parallel to the b-axis in the measurement.

[1] Corresponding author. E-mail: k52952@sakura.kudpc.kyoto-u.ac.jp

2. Experimental Results and Discussions

Figure 1 shows the temperature dependence of center positions of the ^{15}N-NMR line at the resonance frequencies of 12.9 MHz and 25.9 MHz. The temperature dependence of the line splitting and the shift in two cases shows similar behavior down to 4 K. Single line observed above 70K splits into two branches referred to as A and B in Fig.1 and each branch splits into two branches (A1, A2) and (B1,B2). It is noted that A2 line changes the direction of its shift at about 15K. Below 4 K, temperature dependence in two cases shows different behavior. We have also measured T_1^{-1} for all of branches. Then several anomalies were observed. Figure 2 shows the typical temperature dependence of T_1^{-1} in different external fields including ^{14}N-NMR result.

First, we discuss magnetic structure of the 1/4 plateau on the basis of the NMR spectra observed at 1.4 K around 6 T. We calculated internal fields at ^{15}N sites by using a point dipole approximation with respect to several candidates of the structure. Then, we would like to propose one candidate for the 1/4 magnetic structure, which is most plausible in the present time from a qualitative view point. The magnetic moments are induced only at one quarter of whole Cu^{2+} dimer sites and the dimers at other sites are singlet state. The magnetic dimer sites are arranged per four dimers along crystallographic *a*-axis and they also satisfy translational symmetry along crystallographic *b* and *c*-axis. In the double spin chain, this structure satisfies the quantum condition for magnetization plateau in spin chain, $n(S-m) = integer$, where n is the period of the spin state, S the magnitude of the spin and m the magnetization per site[4]. In $(1, 0, \bar{2})$ plane, on the other hand, this structure is similar to the 1/4-stripe state proposed for modified Shastry-Sutherland model[5].

Next, we discuss the temperature depemdence of the magnetic structure by taking account of the behavior of T_1^{-1} and the line splitting together. The 1/4 magnetic structure remains up to 8.5 K, then singlet dimers seem to become triplet and other phases of magnetic structure emarge with increasing temperature. It is noted that each line splitting accompanies anomalous behavior of T_1^{-1}. Such situations are almost the same in the measurement around 3 T and 6 T from 8.5 K to 120 K. This fact means that the same magnetic structure exists in two magnetic field regions.

We mention the 3D-ordering at about 1.7 K in the field region outside the plateau. In this region, magnetic moments, which are not fully polarized along external field even in the ground state, is considered to exhibit the antiferromagnetic ordering of transeverse components from the line splitting and the anormaly of T_1^{-1} for 3 T and 15.9 T as shown in Fig. 1 and Fig. 2.

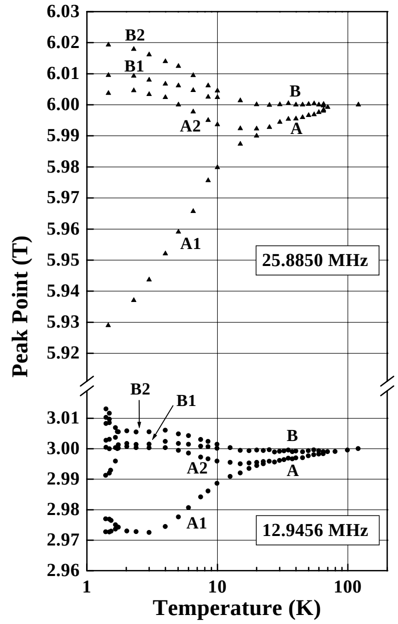

Fig. 1. Temperature dependence of resonance fields of ^{15}N at the operating frequencies of 12.9 MHz and 25.9 MHz

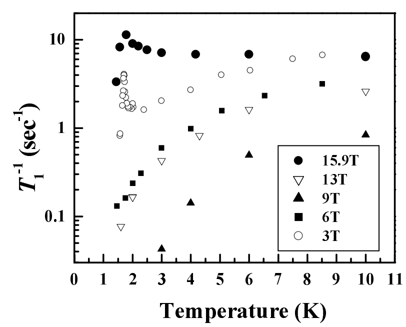

Fig. 2. Temperature dependence of T_1^{-1} of ^{15}N (H=3, 6 T) and ^{14}N (H=9, 13, 15.9 T)

In the plateau region, T_1^{-1} shows gapped-like behavior.

As a concluding remark, we propose that this sytem shows several phases of magnetic structure depending on the external field and the temperature.

References

[1] W. Shiramura, K. Takatsu, B. Kurniawan, H. Tanaka, H. Uekusa, Y. Ohashi, K. Takizawa, H. Mitamura, T. Goto, J. Phys. Soc. Jpn. **67** (1998) 1548.

[2] R.D. Willet, C. Dwiggins, R.F. Kruh, R.E. Rundle, J. Chem. Phys **38** (1963) 2429.

[3] S. Shastry, B. Sutherland Physica B **108** (1981), 1069.

[4] M. Oshikawa, M. Yamanaka,and I. Affleck, Phys. Rev. Lett. **78** (1997) 1984.

[5] Y. Fukumoto J. Phys. Soc. Jpn. **70** (2001) 1397.