Partial magnetic order in the quantum spin system NH_4CuCl_3

T. Goto,[*1] K. Matsui,[*1] T. Suzuki,[*2] H. Guo,[*4] A. Oosawa,[*1] M. Fujisawa,[*3] H. Tanaka,[*3] I. Watanabe[*4]

The ground states of boson systems with randomness have been a puzzle for a long time. In a uniform system consisting of mobile bosons, the ground state is the Bose-Einstein condensed fluid. When the mobility of particles is reduced by introducing randomness or frustration, the system becomes solid-like, that is, the Bose glass or the Wigner crystal, details of which, however, are still unknown[1]. For studying the property of the boson system, NH_4CuCl_3 is suitable model compound, where both the density and mobility of the field-induced magnons can be tuned by the magnitude of applied field and the state of non-centrosymmetric NH_4 molecules, respectively.

The quantum spin magnet NH_4CuCl_3 is an S =1/2 three-dimensional dimer system with the crystal structure isomorphic to $TlCuCl_3$, which is a spin-gapped system and is known for the Bose-Einstein condensation of field-induced magnons. The ground state of NH_4CuCl_3, however, is different from that of $TlCuCl_3$, that is, gapless in zero field, showing a magnetic order at T_N = 1.25 K. When a high magnetic field is applied, two-step magnetization plateaus are observed[2]. The mechanism of the two-step plateaus attracts much interest, for it is related to the fundamental property of the field-induced magnons.

One of the models[3] proposed so far is the localized three sublattice dimer model that claims an existence of the three inequivalent dimers A, B and C with different critical fields H_c, one of which *i.e.* H_c^A is zero. This model elegantly explains the two-step plateaus: the two plateaus appear in the field regions where the dimer A and B saturates, and the net magnetization saturates when all three saturate. According to this model, only dimer A magnetically orders at zero field, indicating the partial magnetic order.

The elastic neutron scattering experiment[4] confirms the doubling of the unit cell along b, supporting the existence of three inequivalent dimers in an elongated unit cell. However, recent high-field NMR observation[5] disproves this model. That is, in the high field of the second plateau region, the two of the three inequivalent dimers are saturated and only C is paramagnetic, and hence the NMR signal corresponding to C is expected to be observed at nearly zero-shift position. However, in the high-field NMR spectra, a signal corresponding to either A or B is also observed with that for C. This contradiction gives us a good motivation to propose carrying out the μSR experiment on this compound. If the model is correct, one expects that only one site possesses a large hyperfine field while the other three quarter sites are expected to remain in the paramagnetic state, or that when the interaction among inequivalent three dimers is appreciable, three different hyperfine fields will appear at each dimer, similar to the case of $SrCu_2(BO_3)_2$.

So far, zero field (ZF) - μSR measurements on single crystals of NH_4CuCl_3 were carried out at the Riken-RAL Muon Facility using a pulsed surface-muon beam with a momentum of 27 MeV/c. In the ordered state below T_N, a clear muon spin rotation is observed at zero-field. At the lowest temperature 0.31 K, one can see in the depolarization curve a clear beat with two different frequencies and also an extremely fast term that monotonically decreases with time. This leads us to choose reasonably the three-component function $G_{KT}(\Delta;\tau)\cdot(A_1e^{-\lambda_1\tau}+A_2e^{-\lambda_2\tau}\cos\omega_2\tau+A_3e^{-\lambda_3\tau}\cos\omega_3\tau)$ to fit the data below T_N. Above T_N, the depolarization curves simply consist of one component, that is, the first term in the above function. Figure 1 shows the typical depolarization curves with the fitted function at various temperatures. The two hyperfine fields ω_1/γ and ω_2/γ increased monotonically below T_N and reached 150 and 90 Oe. The volume fraction of each component for A_1, A_2 and A_3 were 25, 70 and 5 % at 0.3 K.

Although the observed data clearly demonstrate that the ordered state consists of three different parts, the fraction of each part is not in accord with the model, which declares that A_1, A_2 and A_3 should be in the ratio 1:2:1. For further quantitative comparison of this result with the three-sublattice model, the detailed knowledge of the muon stopping site in the unit cell is indispensable.[6]

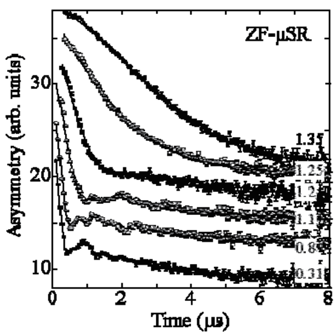

Fig. 1. Muon spin depolarization curves at various temperatures below and above T_N=1.25 K. Solid curves are fitted function with three components $G_{KT}(\Delta;\tau)\cdot(A_1e^{-\lambda_1\tau}+A_2e^{-\lambda_2\tau}\cos\omega_2\tau+A_3e^{-\lambda_3\tau}\cos\omega_3\tau)$.

*1 Department of Physics, Sophia University
*2 Department of Physics, Shibaura institute of Technology
*3 Department of Physics, Tokyo Institute of Technology
*4 RIKEN Nishina Center

References

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