NMR study of spin ladder compound (CPA)$_2$CuBr$_4$

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Abstract

We performed a nuclear magnetic resonance experiment on $^1$H in (CPA)$_2$CuBr$_4$ (CPA = cyclopentylammonium) which is considered as two-leg spin ladder system antiferromagnetic compound. Applied field range up to 8.7 T covers the critical field $H_c$ (= 1.7 T) where the energy gap disappears. In the field range $H < H_c$, the nuclear spin-lattice relaxation rate $T_1^{-1}$ showed a thermal activation-type behavior, reflecting the presence of the finite spin gap at low temperature. As increasing the magnetic field across the $H_c$, the temperature dependence of $T_1^{-1}$ gradually changed from the thermal activation-type to the power-law type in the temperature range between 5 and 2 K. Further, we found a maximum of $T_1^{-1}$ around 2 K for $H > H_c$.

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1. Introduction

(CPA)$_2$CuBr$_4$ (CPA = cyclopentylammonium) is characterized as an $S = \frac{1}{2}$ two-leg ladder Heisenberg spin system whose exchange interaction along the rail direction $J_{\text{rail}}(2J_{\text{rail}}/k_B = -11.6 \text{ K})$ is about twice as large as that along the rung direction $J_{\text{rung}}(2J_{\text{rung}}/k_B = -5.54 \text{ K})$ [1]. The spin gap energy $\Delta/k_B$ has been determined to be 2.3 K from the parameters, which is consistent with the magnetization data [2]. (CPA)$_2$CuBr$_4$ attracts much interest as the first real compound of the two-leg ladder system with $|J_{\text{rail}}| > |J_{\text{rung}}|$. In one-dimensional spin gap systems such as two-leg ladder, bond alternating chain, Haldane-gap system etc., it is known that the system experiences the Tomonaga–Luttinger liquid (TLL) state at low temperature under the magnetic field in the critical magnetic field region defined as the region between the magnetic fields $H_c$ and $H_q$ at which the energy gap disappears and the magnetization saturates, respectively [3]. Theoretical studies have shown that temperature dependence of the nuclear spin-lattice relaxation rate $T_1^{-1}$ is power-law like, namely, $T_1^{-1} \sim T^{-\alpha}$ in the TLL state. This material has been studied by several experimental methods, such as the magnetic susceptibility and magnetization [1], magneto-optical response [2] etc. In order to clarify the dynamic nature of the spin system in (CPA)$_2$CuBr$_4$ in the critical field region, we measured temperature and field dependence of $T_1^{-1}$ using spin echo technique of $^1$H-NMR down to liq-$^3$He temperature. NMR spectrum and $T_1^{-1}$ were measured in the field range up to 8.7 T across the critical field 1.7 T (= $H_c$).

2. Experimental results and discussion

The experiments were carried out on a polycrystalline sample of (CPA)$_2$CuBr$_4$. At first the susceptibility was measured under the temperature range between 2 and 250 K. Obtained results showed broad maximum around 7 K, which is consistent with the reported data [1].

Fig. 1 shows $^1$H-NMR spectra measured in the temperature between 0.4 and 70 K with an operating frequency 230.6 MHz. The resonance line shape is symmetric and sharp at high temperatures above about 50 K,
and becomes broad and rather asymmetric with decreasing temperature. Note that the line shape becomes symmetric again below about 2 K.

Fig. 2 shows the temperature dependence of the nuclear spin-lattice relaxation rate $T_1^{-1}$ measured in the field range 1.0–8.7 T. Let us discuss the obtained results for $H < H_c$ at first and then $H > H_c$.

Under the applied field of 1.0 T ($< H_c$), $T_1^{-1}$ showed a thermal activation type behavior $T_1^{-1} \sim \exp(-\Delta(H)/k_B T)$, where $\Delta(H)$ is the energy gap under the applied field. $\Delta(H)/k_B$ is estimated to be 0.98 K for 1.0 T (see Fig. 3). This value of $\Delta(H)$ is consistent with the zero field spin gap $\Delta/k_B = 2.3$ K because the energy gap is reduced as $\Delta(H) = \Delta - g \mu_B H$.

As the field was increased across the $H_c$, the temperature dependence of $T_1^{-1}$ gradually changed from the thermal activation type to the power-law type in the temperature range between 5 and 2 K. Such a power-law type behavior of $T_1^{-1}$ seems to be the evidence of the TLL state. However, when assuming TLL like behavior $T_1^{-1} \sim T^{-\alpha}$, we obtained $\alpha = 0.2–0.3$, which is somewhat different from the value $\alpha > 0.5$ required by the calculation [4] for spin ladder with $J_{\text{rung}}/J_{\text{rail}} = 0.5$. Further detailed discussion will be necessary, which is beyond this paper.

Below about 2 K in the critical field region above $H_c$, we found that $T_1^{-1}$ turned to decrease with decreasing temperature. Simultaneously, the spectrum pattern changed from an asymmetric to a symmetric shape as shown in Fig. 1. Because this anomaly does not accompany a divergent behavior of $T_1^{-1}$, there seems to be no long range order. One may note that the field dependence of $T_1^{-1}$ is not monotonous, since $T_1^{-1}$ for 5.4 T is larger than those for 6.3 and 5.7 T below about 5 K. As a possible reason for these behaviors, it is noteworthy that rotation of CuBr$_2^{2-}$ ions is suggested by the magneto-optical response measurements.
If so, the energy gap can be affected because the exchange interactions are modified by the rotation. Then, the behavior of $T_1^{-1}$ will be changed. However, they have shown only the data of the magnetic field dependence at 4.2 K, and then the data of temperature dependence are desired.

We note here that similar behaviors of $T_1^{-1}$ have been reported for a dimer-like ladder system CuHpCl [5] and a Haldene-gap system TMNIN [6]. For both cases, as the temperature is decreased in the critical field region, $T_1^{-1}$ shows a TLL-like behavior $T_1^{-1} \sim T^{-2}$ followed by a rapid decrease. At low temperatures below TLL phase, the former shows a long range order, and the latter does not. Such a critical state as the TLL state will be modified or destroyed by a small perturbation. Therefore, the dynamic behavior below the TLL state will provide a rich variety of phase changes.

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References