High field ESR measurements on the low dimensional $S=1/2$ Heisenberg antiferromagnet $(\text{CPA})_2\text{CuBr}_4$

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Abstract. ESR measurements have been performed on poly crystalline sample of an $S = 1/2$ Heisenberg antiferromagnetic ladder-like system $(\text{C}_5\text{H}_9\text{NH}_3)_2\text{CuBr}_4$. We estimated the $g$-value as $g = 2.20$ from analysis of a broad absorption line at high temperatures. Multiple ESR absorption lines were observed below 10 K. The ESR modes show non-linear frequency dependence of the resonance field at 4 K, and the temperature dependence of the line width with a thermal activation type formula. The ESR modes observed are well explained by the theoretical model of the breather excitation.

1. Introduction
Experimental and theoretical studies of the magnetic properties of quantum spin systems, in particular of $S = 1/2$ Heisenberg antiferromagnetic two-leg ladders, have attracted a lot of interest for a number of reasons. Antiferromagnetic two-leg ladder systems have a gap in the excitation spectrum and they reveal a rich behavior by quantum effects in the presence of the magnetic field [1]. These quantum phase transitions were intensively investigated both theoretically [2] and experimentally [3]. On the other hand, the discovery of a magnetic field induced gap in Cu-benzoate [4] and other spin chain materials [5, 6] have increased the interest for magnetic quantum phase transitions which are determined by the combined effects of the uniform and the staggered components of the effective magnetic field.

$(\text{C}_5\text{H}_9\text{NH}_3)_2\text{CuBr}_4$, which we abbreviate to $(\text{CPA})_2\text{CuBr}_4$ below, belongs to the orthorhombic space group $Pna2_1$ ($a = 24.111$ Å, $b = 8.089$ Å, $c = 18.449$ Å). The magnetic Cu$^{2+}$ ions, which are surrounded by the tetrahedron of Br$^-$ ions, form two leg ladders along the $b$ axis [7]. $(\text{CPA})_2\text{CuBr}_4$ has been expected to have strong exchange interaction among the rail direction in contrast with general two leg ladder materials reported with strong exchange interactions among the rung direction. The value of the exchange interactions $2J_{\text{rail}}/k = -11.6$ K, $2J_{\text{rung}}/k = -5.54$ K ($J_{\text{rung}}/J_{\text{rail}} = 0.47$) were estimated by the susceptibility measurements [7]. The spin gap $\Delta/k = 2.3$ K was also proposed by the analysis of the susceptibility and the magnetization $B_{\text{sat}} = 1.7$ T at low temperatures [8]. In the temperature and the magnetic field higher than the gapped state, Tomonaga-Luttinger liquid (TLL) state has been proposed by the power-law behavior of the spin relaxation time $T_1$ in NMR measurements [9], and compared to theoretical magnetic phase diagram in the spin ladder system. In this paper, we have investigated the magnetic state of $(\text{CPA})_2\text{CuBr}_4$ by ESR.
2. Experimental technique

(1,1-diptyl-2-picrylhydrazyl (DPPH) poly crystalline sample were prepared by hydro thermal synthesis method. ESR measurements were performed in the temperature from 4 K to 200 K, and in the frequency from 35 GHz to 100 GHz by using a Millimeter Wave Vector Network Analyzer (MVNA: ABmm), Gunn oscillator, and a small cavity. The external magnetic field was applied up to 16 T by using a superconducting magnet. The value of magnetic field was calibrated by a g-marker of 1, 1-diphenyl-2-picrylhydrazyl (DPPH) in all measurements.

3. Result and discussion

ESR measurements have been performed at 35 GHz in the temperature below 200 K down to 4 K. A broad electron paramagnetic resonance (EPR) absorption line (line-I) was observed at high temperatures. The g-value was estimated to be $g = 2.20$ from the analysis of the EPR absorption line measured at 188 K. Figure 1 shows the temperature dependence of the ESR spectrum from 4 K to 50 K. With decreasing temperature, the half line width increases gradually. With further decreasing temperature below 10 K, a new ESR absorption line (line-II) develops in addition to the resonance absorptions of line-I. Below 6 K, additional two small absorption lines (line-III and line-IV) were observed at the magnetic fields above line-II. We analyze the spectra by the calculation on the basis of the Lorentzian type absorption. In the analysis for the data at 4, 5, 6 K, we assumed that the spectra consists of several absorption lines that are indicated by filled circles, filled triangles, filled squares, and filled diamonds in Fig. 1. We will focus on the line-I and line-II below for simplicity. Figure 2 shows the temperature dependence of the half line width of line-I and line-II. The half line width of line-I increases with decreasing temperature toward 10 K, below which it cannot be extracted by the analysis because of the weak ESR intensity. On the other hand, the half line width of line-II rapidly decreases with decreasing temperature. Although the characteristic temperature corresponds to a broad maximum of the susceptibility as a function of temperature, any anomalies including the magnetic transition at 10 K have not been reported in (CPA)$_2$CuBr$_4$. This fact indicates that some crossover between different magnetic states exist around 10 K. It should be emphasized that the ESR absorptions observed below 6 K are in these measurements that are in the temperature and in the magnetic field above gapped state.

Figure 1. Temperature dependence of the ESR spectrum at 35 GHz. A sharp absorption line around 1.25 T is a DPPH absorption ($g = 2.00$).

Figure 2. Temperature dependence of the half line widths of line-I ($\bullet$), line-II ($\triangle$). The solid line and the dashed line are referred to the text.
We measured the multiple frequency ESR measurements at 4 K in order to obtain the properties of line-II. A sharp absorption of line-II was observed with small absorption line-III and line-IV in each spectrum. Figure 3 shows frequency-resonance field relation of ESR mode of line-II. The non-linear relation of the mode in line-II cannot be explained by the EPR mode, antiferromagnetic resonance mode, and even by the direct transition between singlet and triplet state that is expected below 1.7 K in this system. By the double logarithmic plot of the data shown in inset of Fig. 3, we find that the relation of the resonance field and frequency in absorption line-II obeys a power-law \( f_{\text{res}} \propto B \) with an index \( \gamma = 0.67 \). Although the data of the line-III and line-IV are not shown in Fig. 3, the ESR modes also obey a power-law with an index approximately 0.67.

The similar behavior of the ESR spectrum has been observed on the breather excitation in quasi-one-dimensional spin chain materials [4-6]. The breather excitation with the field-induced gap is represented by solitons, antisolitons and multiple soliton-antisoliton bound states. On the basis of a quantum sine-Gordon (SG) model, the particle-like soliton and breather excitations are expected. A theoretical model by Oshikawa and Affleck (OA) has predicted the power-law behavior among the resonance field and the frequency with the index \( \gamma = 2/3 \) for the breather excitation [10, 11], and successfully explain the experimental results of Cu-benzoate [4]. The relation of the formula is in good agreement with the relation obtained in \((\text{CPA})_2\text{CuBr}_4\). Based on the OA model, the temperature dependence of the half line width in the breather excitation should obey the thermal activation type formula as 
\[
B \propto \exp(-E_g/T),
\]
where \( E_g \) is a energy of the field-induced gap [4]. The value of \( E_g \) is not an adjustable parameter, but be able to obtain from the resonance frequency-field relation of the ESR experiment: 
\[
E_g \propto B.
\]
By using the index \( \gamma = 2/3 \), the temperature dependence of the absorption line-II is compared with the OA model by the dashed line in Fig. 2. We can find that the theoretical model also explains the observed behavior of the half line width below 10 K.

For an \( S = 1/2 \) one-dimensional spin chain system, the gapless excitation at the temperature higher than breather regime is represented by spinon excitation. Figure 4 shows the shift of the resonance field of line-I below 50 K down to 10 K. The dashed line and solid line indicate a power-law behavior \( T : \gamma = 2.5 \) and 3, respectively.

The OA theory describes a breather excitation in the field-induced gap for gapless one-dimensional chain system. Subsequent theoretical calculation by Zhao et al. [12], recently, suggests that the spin ladder system also has the breather excitation with the field-induced gap. The breather excitation in both systems requires the existence of an effective staggered magnetic field, which arises from the alternating \( g \)-tensor or Dzyaloshinskii-Moriya interaction in the crystal structure [10, 11]. It should be noted that the alternation seems not to exist in the crystal structure of \((\text{CPA})_2\text{CuBr}_4\) in the zero magnetic field [7]. Although we cannot explain the microscopic mechanism of the present breather-
like behavior, some new mechanism or the effect of finite magnetic field for the crystal structure may be required for our observation.

4. Conclusion

We performed the ESR measurements on poly crystalline sample of (CPA)$_2$CuBr$_4$, in the temperature region from 4 K to 200 K and in the frequency region from 35 GHz to 110 GHz. A broad absorption line (line-I) was observed at high temperatures. Multiple ESR absorptions (line-II, line-III and line-IV) were observed below 10 K. The frequency dependence of the resonance field of line-II shows a power-law behavior: $B \sim 0.67$, at 4 K. The temperature dependence of the half line width shows thermal activation type relation: $\exp(-E_g/T)$. Above 10 K, the temperature dependence of the resonance shift and the half line width of line-I are almost proportional to $(B/T)^3$ and $(B/T)^2$, respectively. These properties of ESR spectrum are well described by the theoretical prediction on the spinon and the breather excitation modes.

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