High-field and high-pressure ESR measurements of SrCu$_2$(BO$_3$)$_2$

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Abstract. High-field and high-pressure ESR measurements of SrCu$_2$(BO$_3$)$_2$ have been performed at 1.6 K in the field region up to 30 T, the frequency region from 70 to 360 GHz and the pressure region up to 12 kbar. The direct ESR transition mode between the singlet ground state and the triplet excited states was observed at various pressures. We have succeeded in observing the decrease of the spin gap directly upon applying the pressure. The origin of the decrease of the gap energy is discussed.

1. Introduction

SrCu$_2$(BO$_3$)$_2$ is known as a quasi two-dimensional quantum spin system in which $S = 1/2$ dimers realize the Shustally-Sutherland model [1, 2]. The system shows a variety of interesting phenomena such as the magnetization plateaus in the magnetization process. The ground state is the singlet state with a finite gap below the excited triplet states due to the stronger intradimer antiferromagnetic interaction $J$ than the interdimer interaction $J'$. The gap energy is determined to be 34 K from the inelastic neutron scattering measurements [3, 4] and the high field ESR measurements [5, 6]. The ratio $J'/J$ is estimated to be $\sim 0.67$ and it is considered to be close to the critical point $(J'/J)_c = 0.69$, where the Néel state becomes stable above $(J'/J)_c$ [2, 7]. If the ratio can be changed continuously, a novel phenomenon around the quantum critical point may be discovered. The pressure is the most effective method to explore such new phase. The magnetic susceptibility measurement up to 7 kbar was performed and it suggested that the gap energy decreases upon applying the pressure [8]. Recently, the magnetic susceptibility measurement and the nuclear magnetic resonance measurement have revealed that a novel high pressure phase appears above 14 kbar and magnetic and non-magnetic dimers coexist in this phase [9].

We have developed the high field and high pressure ESR apparatus so far, and it turned out that this apparatus is a powerful means to study magnetic properties of materials under pressure from the microscopic point of view [10, 11, 12, 13]. Therefore, we have applied this high pressure ESR technique to SrCu$_2$(BO$_3$)$_2$ in order to investigate its pressure dependence in detail. In this paper, we report that the decrease of the gap energy between the ground state and the first excited state was observed directly by our high pressure ESR measurements.
2. Experimental

The apparatus consists of the pulsed high-magnetic field ESR setup and a clamped type piston-cylinder type pressure cell. The transmission ESR measurement can be performed by using zirconia or sapphire as inner parts of the pressure cell, which can transmit millimeter or submillimeter wave. The detailed setup of the pressure cell can be found in ref. [12]. We extended the maximum magnetic field from 16 T to 55 T this time by using the newly developed ESR apparatus in our laboratory [14]. Figure 1 shows the schematic diagram of the high-pressure ESR apparatus whose maximum field is 55 T. The pressure cell is connected to the end of the light pipe and it is inserted to the magnet. Multi-frequency ESR measurement at 4.2 K or 1.6 K can be performed in quasi-continuous frequency region from 70 to several hundred GHz using Gunn oscillators and backward traveling wave oscillators as the light sources.

Frequency dependence ESR measurements at several pressure points have been done on SrCu$_2$(BO$_3$)$_2$ single crystal at 1.6 K up to 30 T. The magnetic field was applied parallel to the $a$-axis. The pressure was estimated from the relation between the load at room temperature and the pressure around 3 K determined by the change of the superconducting transition temperature of Sn [12].

3. Results and discussion

Figure 2 shows the pressure dependence ESR spectra obtained at around 230 GHz. The solid triangle indicates the signal due to the transition between the singlet ground state and the lower branch of the triplet excited states ($S_z = −1$). Although the transition between the singlet state and the triplet states is forbidden in general, such direct ESR transition mode was observed at ambient pressure by Nojiri et al. [5, 6] due to the existence of Dzyaloshinsky-Moriya interaction. It is clearly seen that the resonance field shifts to the lower field side as the pressure is applied. This is caused by the decrease of the gap energy as is clarified later.

The frequency-field diagram is shown in Fig. 3. The solid line is the fitting line expressed as $hν = −gμ_BB + Δ$ where the $g$-value is fixed to be $g = 2.05$ [5] and the gap energy $Δ$ is the fitting parameter. The fitting is done by eliminating the obtained data at low frequency region because they deviate from a straight line [5, 6]. Thus, all the obtained data are well interpreted as the direct ESR transition mode between the singlet state and the lower branch of the triplet
Figure 2. Pressure dependence ESR spectra of $\text{SrCu}_2(\text{BO}_3)_2$ at 1.6 K for $B \parallel a$ obtained at around 230 GHz. DPPH was used as a field marker ($g = 2.00$).

states as shown in Fig. 3.

From the fitting results, we obtained the gap energy $\Delta$ at each pressure. At ambient pressure $\Delta$ is determined to be 723 GHz (34.7 K) and this value agrees well with the previous result by Nojiri et al. [5, 6]. Figure 4 shows the pressure dependence of the gap energy $\Delta$. It is obvious that the gap energy decreases when the pressure is applied and this is the direct evidence of the decrease of the gap energy by pressure for this compound. In ref. [8], it is reported that the temperature at maximum susceptibility ($T_{\text{max}}$) decreases as the pressure is applied and the linear extrapolation of $T_{\text{max}}$ to zero seems to cross 25~35 kbar. On the other hand, our result revealed that the gap energy changes more gradually to the pressure. From the obtained data in this study, the pressure where the gap is closed is estimated to be around 55 kbar by a linear extrapolation.

From the perturbation theory, the gap energy in the singlet dimer phase is obtained up to the forth order of $(J'/J)$ as follows [2],

$$
\Delta = J \left\{ 1 - \left( \frac{J'}{J} \right)^2 - \frac{1}{2} \left( \frac{J'}{J} \right)^3 - \frac{1}{8} \left( \frac{J'}{J} \right)^4 \right\}.
$$

(1)

From this relation, it is concluded that the ratio $(J'/J)$ increases and $\Delta$ decreases when the pressure is increased. It is a natural assumption that the intradimer interaction $J$ is less sensitive to the pressure than the interdimer interaction $J'$ because the intradimer packing seems more rigid than that between dimers. Therefore, we conclude that the decrease of the gap energy under pressure can be attributed mainly to the increase of the interdimer interaction $J'$. Any anomaly related to the high pressure phase, in which the magnetic and non-magnetic dimer coexist, was not found up to 12 kbar. We have not reached the pressure region above 14 kbar at this moment where the high pressure phase is expected to appear [9]. We plan to extend the pressure region up to 20 kbar and to clarify the high pressure phase.

4. Conclusions
We have applied our new high-field and high-pressure ESR apparatus to $\text{SrCu}_2(\text{BO}_3)_2$. The pressure dependence measurements up to 12 kbar are performed at 1.6 K in the field range up to 30 T. The direct ESR transition mode due to the transition between the singlet ground state and the triplet excited states is observed at various pressure points. Moreover, direct observation
of the decrease of the energy gap was successfully achieved by applying the pressure. We can conclude that the decrease of the gap energy is mainly caused by the increase of the interdimer interaction.

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References
[1] Kageyama H, Yoshimura K, Stern R, Mushnikov N V, Onizuka K, Kato M, Kosuge K, Slichter C P, Goto T and Ueda Y 1999 Phys. Rev. Lett. 82 3168.
[2] Miyahara S and Ueda K 1999 Phys. Rev. Lett. 82 3701
[3] Kageyama H, Nishi M, Aso N, Onizuka K, Yoshitama T, Nukui K, Kodama K, Kakurai K and Ueda Y (2000) Phys. Rev. Lett 84 5876
[4] Gaulin B D, Lee S H, Haravifard S, Castellan J P, Berlinsky A J, Dabkowska H A, Qiu Y and Copley J R D (2004) Phys. Rev. Lett. 93 267202
[5] Nojiri H, Kageyama H, Onizuka K, Ueda Y and Motokawa M (1999) J. Phys. Soc. Jpn. 68 2906
[6] Nojiri H, Kageyama H, Ueda Y, Motokawa M (2003) J. Phys. Soc. Jpn. 72 3243
[7] Weihong Z, Hamer C J and Ottmaa J (1999) Phys. Rev. B 60 6608
[8] Kageyama H, Mushnikov N V, Yamada M, Goto T and Ueda Y 2003 Physica B 329-323 1020.
[9] Waki T, Arai K, Takigawa M, Saiga Y, Uwatoko Y, Kageyama H and Ueda Y (2007) J. Phys. Soc. Jpn. 76 073710
[10] Ohta H, Sakurai T, Okubo S, Saruhashi M, Kunimoto T, Uwatoko Y and Akimitsu J (2002) J. Phys.: Condens. Matter 14 10637.
[11] Sakurai T, Saruhashi M, Hirano T, Inagaki Y, Okubo S, Kunimoto T, Ohta H, Tanaka H and Uwatoko Y (2003) J. Phys. Soc. Jpn. 72 Suppl. B 156
[12] Sakurai T, Taketani A, Tomita T, Okubo S, Ohta H and Uwatoko Y (2007) Rev. Sci. Instrum. 10 1237
[13] Sakurai T, Taketani A, Tomita T, Okubo S, Ohta H, Tanaka H and Uwatoko Y (2006) J. Phys.: Conf. Ser. 51 565.
[14] Ohta H, Tomoo M, Okubo S, Sakurai T, Fujisawa M, Tomita T, Kimata M, Yamamoto T, Kawauchi M and Kindo K (2006) J. Phys. Conf. Ser. 51 611.