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High-field ESR measurements of $SrCu_2(BO_3)_2$

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Abstract

High field ESR measurements of the $S=\frac{1}{2}$ two-dimensional spin gap system $SrCu_2(BO_3)_2$ have been performed in the frequency region from 526 to 1620 GHz using pulsed high magnetic fields up to 55 T. Besides the ESR signal which is observed near the EPR line, a number of new ESR signals are observed in the magnetic phase, where the plateau at $\frac{1}{3}$ of the saturated magnetization appears on the magnetization curve.

Keywords: Magnetization plateau; $S=\frac{1}{2}$ two-dimensional spin gap system; High field ESR measurement

1. Introduction

Recently, the two-dimensional spin gap system $SrCu_2(BO_3)_2$ has attracted much attention. This material has a stacked-layer structure, composed of alternating stacks of $CuBO_3$ and Sr planes along the *c*-axis [1]. In the $CuBO_3$ plane, Cu^{2+} ions with $S=\frac{1}{2}$, form dimer units, and an orthogonal arrangement of these dimers is realized in the plane perpendicular to the *c*-axis. Various experimental and theoretical studies have revealed that $SrCu_2(BO_3)_2$ shows interesting characteristics, reflecting this peculiar spin structure. This material has a spin singlet ground state with an energy gap, and the intra- and the inter-dimer exchange interaction, symbolized by J_0 and J', and the energy gap E_g were estimated to be $J_0 = 100\,K$, $J' = 68\,K$ and $E_g = 34.7\,K$, respectively [3,5]. It was theoretically pointed out that the ground state of this material can be exactly described by the direct product of the singlet dimers, and the triplet excitations are extremely localized [2,3]. One of the most interesting features of this material is the magnetization plateaus under an external magnetic field, which is considered to originate from the extremely localized nature of the triplet excitations [2–4,6–8]. In the high-field magnetization measurements, plateaus at $\frac{1}{8}$, $\frac{1}{4}$ and $\frac{1}{3}$ of the saturated magnetization were observed [2,4]. Theoretical studies suggested that the occurrence of the plateaus is a consequence of the crystallization of the triplet dimers [6–8]. Moreover, it was predicted that the superstructures of the triplet dimers of stripetype occur at the magnetization plateaus of $\frac{1}{3}$ and $\frac{1}{4}$ [6–8].

Recently, submillimeter wave ESR measurements of $SrCu_2(BO_3)_2$ using pulsed magnetic fields

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up to 30 T, were performed by Nojiri et al. [5]. These authors observed the direct transitions from the ground state to the first excited triplet state. Moreover, a second-gap excitation and a number of ESR absorption lines with zero field gap were observed in these measurements. In order to investigate the high-field phase of this material, we have performed high-field ESR measurements of $SrCu_2(BO_3)_2$ in the frequency region from 526 to 1620 GHz using the pulsed high magnetic field up to 55 T.

2. Experimental

The high-field ESR measurements of $SrCu_2(BO_3)_2$ single crystal were performed at KYOKUGEN Osaka university using a far-infrared laser and the pulsed magnet up to 55 T. A magnetically enhanced InSb detector made by QMC instruments was used. An external magnetic field was applied along the c-axis.

3. Results and discussion

Fig. 1 shows the frequency dependence of the ESR absorption lines observed for $H \| c$ at 1.4 K. The dashed lines show the critical fields observed by the previous high-field magnetization measurements at 1.4 K [4]. We define the critical fields as follows: a finite magnetization appears above H_0, and $\frac{1}{8}$, $\frac{1}{4}$ and $\frac{1}{3}$ plateaus of the magnetization occur in the region $H_1 < H < H_2$, $H_3 < H < H_4$, and $H_5 < H$, respectively. In this measurement, almost no ESR signal is observed below H_0 due to the limitation of the sensitivity of our ESR system. However, the ESR signal A_1, which appears above H_0, is observed near the resonance field of the electron paramagnetic resonance (EPR). As the frequency is increased, the resonance field of the ESR signal A_1 increases, and the intensity of this ESR signal increases with increasing resonance field. Reflecting the magnetic phase transition, the ESR signal A_1 suddenly disappears at H_1, while another ESR signal A_2 appears just above H_1. As in the case of the ESR signal A_1, the resonance field of ESR signal A_2 increases with increasing the frequency. The ESR signal splits into

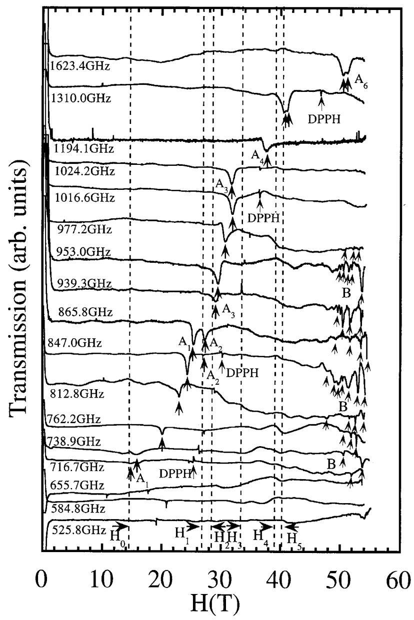

Fig. 1. Frequency dependence of the ESR absorption lines observed for $H \| c$ at 1.4 K. The dashed lines show the critical fields observed by the previous high-field magnetization measurements.

two absorption lines above H_5, where the $\frac{1}{3}$ plateau of the magnetization is observed. Moreover, in this magnetic phase, ESR signals B accompanied by a number of sharp structures are observed in the frequency region from 716 to 977 GHz. These ESR signals appear only well above the critical field H_5. Fig. 2 shows the temperature dependence of the ESR absorption lines observed at 847 GHz for $H \| c$. The observed temperature dependences are different between the ESR signals A_i ($i = 1, 2$) and B. As the temperature is increased, the intensities of the ESR signals A_1 and A_2 decrease, suggesting that these ESR signals correspond to the excitations from the ground state, while the ESR signals B broaden rapidly. Thus, the temperature dependence of the intensity of the ESR signals B is not clear. The ESR signal P, which appears above 2.2 K, is considered to come from the transition between the excited states. Fig. 3 shows the frequency–field relation of the ESR signals observed at 1.4 K for $H \| c$. The ESR signals A_i's and B are shown by the open circles and the open diamonds, respectively. The magnetization curve observed at 1.4 K for $H \| c$ [4], is shown by the solid line in

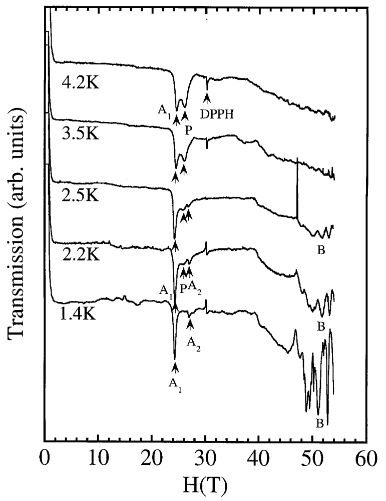

Fig. 2. Temperature dependence of the ESR absorption lines observed at 847 GHz for $H \| c$.

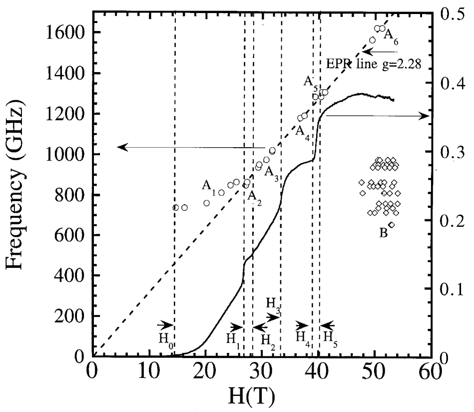

Fig. 3. The frequency–field relation of the ESR signals observed at 1.4 K for $H \| c$. The ESR signals A_i's and B are shown by the open circles and the open diamonds, respectively. The magnetization curve observed at 1.4 K for $H \| c$ are shown by the solid line.

Fig. 3. The ESR branch A_1 strongly deviates from the EPR line of $g = 2.28$, which was determined by the EPR measurement at 25 K [5], just above H_0. This ESR branch was also observed by the submillimeter wave ESR measurement of Nojiri et al. [5]. The magnetization process shows an anomalous behavior in that the finite magnetization appears at $H_0 = 14.5\,\mathrm{T}$ which is much lower than the critical field $H_c = 22.6\,\mathrm{T}$, calculated from $E_g = g\mu_B H_c$. The observed frequency dependence of the ESR branch A_1 may reflect the finite mixing between the initial spin-singlet ground state and the excited states, which is inferred from the magnetization process [2,4], or it may suggest the possibility of field induced antiferromagnetic order around H_0. An anomaly of the ESR branch can be seen at H_1, suggesting the discontinuous change of the spin structure at H_1. In the magnetic phase $H_1 < H < H_2$ of the $\frac{1}{8}$ plateau, the ESR branch A_2 slightly deviates from the EPR line to higher magnetic field. In the range $H_2 < H < H_4$, however, the ESR branches A_3 and A_4 almost coincide with the EPR line. The origin of the ESR branch shown by the open circle is not clear at the moment, but we consider that this ESR branch may be due to the spin flip of the triplet dimer. Next we discuss the ESR signals B observed in the magnetic phase of the $\frac{1}{3}$ plateau. We speculate that these ESR signals come from the excitations of the dimers such as the transitions of the triplet to the singlet dimer state or those of the singlet to the triplet dimer state. However, it is difficult to calculate the energies of such excitations at the moment; in order to explain the ESR signals, the theory of the excitations in the magnetic phase of $\frac{1}{3}$-plateau is required.

In conclusion, high-field ESR measurements of $SrCu(BO_3)_2$ using pulsed magnetic fields up to 55 T have been performed. The ESR signal is observed near the resonance field of EPR in the magnetic phase above H_0. Moreover, a number of new ESR signals are observed in the magnetic phase of the $\frac{1}{3}$-plateau for the first time. These ESR signals suggest the presence of magnetic excitations peculiar to the magnetic phase of the $\frac{1}{3}$-plateau.

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