



Study of spin gap excitations in $\text{SrCu}_2(\text{BO}_3)_2$ by submillimeter wave ESR

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Abstract

Spin gap excitations have been observed directly in two-dimensional dimer system $\text{SrCu}_2(\text{BO}_3)_2$ by means of submillimeter wave ESR. The zero-field energy gap shows an unexpected splitting into two branches due to the anisotropy of the inter-dimer coupling. The average coincides with the energy gap value estimated from the high field magnetization process. © 2000 Elsevier Science B.V. All rights reserved.

Keywords: ESR; Quantum spin system; Spin gap

A quantized plateau of magnetization in low-dimensional quantum spin system is a subject of current interest. Such plateau is expected to be realized in a system with localized excited states. In this case, triplets hardly propagate in the system and thus a spin gap coexists with excited triplets. Hence it is very interesting to observe such localized and gapped magnetic excitations around magnetization plateau. Recently, the quantized plateaus of magnetization were observed for $\text{SrCu}_2(\text{BO}_3)_2$ [1]. A unique two-dimensional (2D) network of Cu^{2+} causes a special situation, where nearest-neighbor dimers are arranged orthogonally to each other as shown schematically in the inset of Fig. 1. The theoretical investigations have shown that the hopping of a triplet is impossible within the fifth order in the perturbation [2]. The main issue of the present article is the investigation of the spin gap excitations of the system by means of ESR.

Fig. 1 shows the temperature dependence of ESR spectra at 428.9 GHz for $H||c$. At low temperatures, a set of two resonance peaks marked by arrows are observed. The temperature dependence shows that these two peaks are the excitations from the singlet ground state. We have

measured the frequency dependence of these peaks up to 1 THz and found that the two peaks show a characteristic frequency dependence as schematically depicted in the inset of Fig. 1. It clearly indicates that the two modes correspond to the transitions between the ground singlet state and the first excited triplet state. The zero-field energy gaps are evaluated as 708 and 738 GHz. The average of these two gaps are found to be 722 GHz and it is close to the gap estimated so far by different methods [1,3,4].

A possible candidate for the zero-field splitting between the two triplets is the finite inter-dimer interaction J' . For finite J' , we expect the two excited states to be separated from each other by the order of J' . Since the next-nearest neighbor coupling J_2 is estimated to be 68 K, it is not reasonable to attribute the experimentally observed small splitting to the inter-dimer coupling even if we take the significant frustration effect into account [2]. To investigate the origin of the splitting, we measured the field orientation dependence of the two spin gap excitations as shown in Fig. 2. A large anisotropy of the splitting is found and thus we speculate that the anisotropy of exchange interaction is the origin of this splitting. As is well known, the anisotropic exchange is caused by the spin-orbit coupling λ and the λ is also related to the anisotropy of g -value. The angular dependence of the g -value shown in Fig. 2 indicates that a large anisotropy exists between $H||c$ and $H||a$. Thus we can

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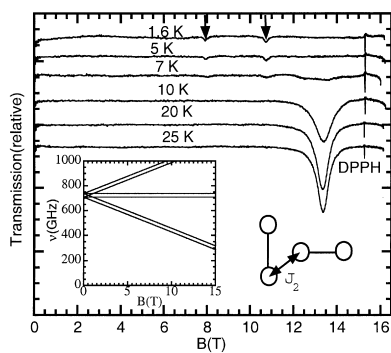


Fig. 1. Temperature dependence of ESR spectra at 428.9 GHz. Two arrows indicate the singlet-triplet transitions. The energy diagram and the configuration of two dimers in the c -plane are depicted schematically in the inset. Open circles denotes the Cu^{2+} ions.

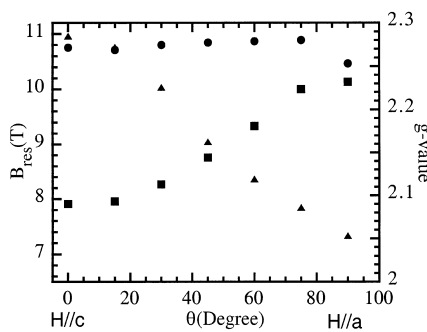


Fig. 2. The angular dependence of resonance fields B_{res} of the two singlet-triplet transitions (closed rectangles and closed circles) and the g -value (closed triangles) in the ac -plane.

expect that the J_{2aa} deviates from J_{2cc} , where J_{2cc} and J_{2aa} are the components of the inter-dimer exchange coupling along the c -axis and a -axis, respectively. Hence the small splitting for $H\parallel c$ can be attributed to the difference between J_{2cc} and J_{2aa} .

Finally we mention the line shape of paramagnetic signals. Above 10 K, only one broad ESR signal can be observed at around 13.4 T as shown in Fig. 1. The resonance field shows no clear temperature dependence between 10–25 K. On the other hand, the line shape deviates from the Lorentzian below 15 K and shows an intermediate shape between the Lorentzian and Gaussian. As is well known, such behavior appears when spin diffusion is important in the 2D system. However, for the present system the line shape can be fitted by the 2D spin diffusion theory only in a very narrow temperature region. Moreover, at around 10 K, the signal can be fitted as the superposition of two components with different linewidths possibly due to the existence of multiple excited states [5].

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