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Magnetic excitations at magnetization plateaux: ESR study on $SrCu_2\ (BO_3)_2$

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Abstract

Magnetic excitations of $SrCu_2(BO_3)_2$ have been studied in very high fields including $\frac{1}{4}$-plateau and $\frac{1}{8}$-plateau phases by means of electron spin resonance. Below the $\frac{1}{8}$-plateau, a bending of the $S_z = -1$ branch of the lowest excited triplet is observed, which is caused by the strong mixing between the ground state and the excited state. Besides the triplet, a strong main mode is observed which is apart from the resonance field expected for the conventional paramagnetic resonance. At magnetization plateaux, the main mode appears around the paramagnetic resonance position and several broad absorption peaks are observed at lower frequencies. © 2001 Elsevier Science B.V. All rights reserved.

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1. Introduction

In a classical antiferromagnet, a magnetization plateau appears mostly when there is an Ising-type anisotropy. However, the mechanism is different for the plateau appearing in quantum spin systems such as coupled dimer systems. When there is an effective repulsive force acting among triplets, a regular lattice made up of triplets is formed at a magnetization plateau. This new type of plateau has been found in $SrCu_2(BO_3)_2$ [1] and NH_4CuCl_3 [2]. The necessary condition for the plateau formation and the possible spatial arrangements of triplets such as stripe or checker have been discussed theoretically [3]. However, much less is known for the magnetic excitation in these systems because neutron scattering experiments are difficult in such high fields. On the other hand, ESR can be performed in very high fields by using pulsed magnetic fields. Hence, ESR is considered as a unique and powerful tool to study the magnetic excitation at a magnetization plateau. In the present work, ESR measurements have been performed up to 40 T for $SrCu_2(BO_3)_2$. The characteristic features of the magnetic excitation of the system are presented in the following.

2. Results and discussion

Submillimeter wave ESR measurements have been performed up to 40 T. A far-infrared laser, backward-travelling wave tubes and Gunn oscillators have been employed as the radiation source. A single crystal is grown by the travelling-solvent-floating-zone method.

Fig. 1 shows the frequency-field diagram measured at 1.6 K for $H \| c$. The magnetization curve taken from the reference is plotted together, where distinct magnetization plateaux are shown at $\frac{1}{8}$ and $\frac{1}{4}$ of the saturation magnetization [4]. At zero field, the magnetization is zero for the existence of the spin gap. The transition between the ground state and the lowest excited triplet state is observed by ESR as indicated by the closed circles. The small splitting of the lowest spin gap excitation may be caused by the inter-dimer coupling [5]. In each triplet, the $S_z = 0$ branch is not observed because ESR absorption spectra are measured by scanning magnetic fields at fixed frequencies. As the frequency is decreased, the $S_z = -1$ branches of the lowest triplets show a bending above 17 T. This bending indicates the

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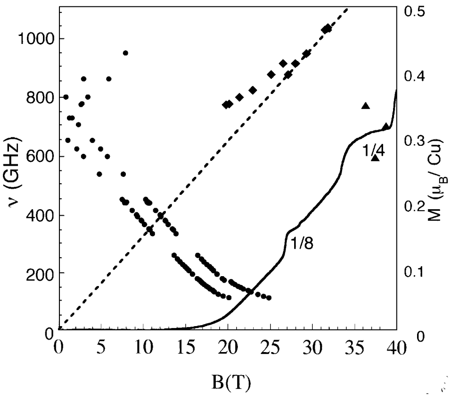

Fig. 1. Frequency-field diagram for $H \| c$. Closed circles, closed diamonds and closed triangles indicate the lowest spin gap excitation, the main mode and the low-frequency mode found at plateau, respectively. The dashed line indicates the position of paramagnetic resonance. The magnetization curve taken from Ref. [4] is also plotted.

strong mixing between the ground state and the excited state. It is noticed that the magnetization becomes finite above 17 T. As the origin of this finite magnetization, the temperature effect is ruled out. It is because this behavior appears even at 0.1 K [4]. Therefore, we speculate that the ground state of the system is changed although the finite energy gap remains between the ground state and the excited triplet state.

In a spin gap system, a disordered state or a three-dimensional (3D) ordered state appears when the gap is collapsed by a strong magnetic field. In a disordered state, the strong ESR absorption peak is observed at the resonance field identical with that of the conventional paramagnetic resonance. In a 3D-ordered state, an antiferromagnetic resonance (AFMR) is observed. In Fig. 1, the strong ESR lines are indicated by closed diamonds. It is noticed that these ESR lines show large deviation, below the $\frac{1}{8}$-plateau, from the positions expected for a paramagnetic resonance, which is shown by the dashed line. Another interesting point is that the frequency of ESR mode exhibits a jump at the jump of the magnetization just below the $\frac{1}{8}$-plateau. As is well known, a single ion-type anisotropy is not responsible for such a deviation in the case of $S = \frac{1}{2}$ Cu^{2+} ion. Hence, it is difficult to explain the origin of the signals as the ESR lines of a disordered state. If a collective spin wave mode arises due to the 3D-ordering, the deviation can be attributed to the observation of AFMR. Therefore, we speculate that a 3D-ordered state may exist just below the $\frac{1}{8}$-plateau. It should also be noted that the finite energy gap remains between the ground state and the excited triplet state. Hence, the ordered state cannot be a simple Neel state.

At the $\frac{1}{8}$ and $\frac{1}{4}$ plateaux, two types of magnetic excitations are found. The strongest main mode is located beside the paramagnetic mode. A small deviation is found between the main mode and the paramagnetic resonance position and the deviation slightly changes between these two plateaux. At $\frac{1}{4}$-plateau, broad absorption peaks are found at lower frequencies as depicted by the closed triangles in Fig. 1. Such a low-energy mode was also found in NH_4CuCl_3 at magnetization plateaux and the mode was considered as the manifestation of the opening of the excitation gap at a magnetization plateau. This gap is related to the energy required for the creation or annihilation of a new triplet in the lattice made up of triplets. The energy needed for the creation of a new triplet is roughly given by the sum of the one-triplet excitation energy, the interaction energy with the surrounding triplets and the Zeeman energy. It is noticed that the Zeeman energy is very large in the field range of the $\frac{1}{4}$-plateau and it exceeds the one-triplet excitation energy at zero field. Therefore, we can expect the excitation energy of a new triplet to be much lower than that of the main mode. Hence, it is natural to attribute the low-energy mode found in the present experiments to the creation or annihilation of a new triplet. The estimate of the energy between a new triplet and the surrounding triplets is a very difficult task and thus the quantitative analysis remains for future theoretical investigations.

As a conclusion, two types of ESR modes are found at the magnetization plateaux in $SrCu_2(BO_3)_2$. A gapped mode found in low frequency can be attributed to the creation or annihilation of a new triplet. Below the $\frac{1}{8}$-plateau, several anomalies are found in ESR. It has been suggested that the ground state of system is not a simple singlet state above 17 T.

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