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Magnetic Excitations in the Two-Dimensional Spin Gap System $SrCu_2(BO_3)_2$

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Neutron inelastic scattering has been performed on single crystals of 2D spin-gap system $SrCu_2(BO_3)_2$, which has an exact dimer ground state of the Shastry-Sutherland model. We observed three magnetic excitations around 3, 5 and 9 meV at 1.7 K. The lowest singlet-triplet excitation at 3 meV shows little wavevector dependence in all directions, indicating a localized character of the triplet state. The two triplet excitation at 5 meV exhibits more dispersive nature in the 2D *a*-*b* plane, but is dispersionless in the out-of-plane. This observation suggest that the correlated hopping of the two-triple excitations is confined to the 2D *a*-*b* plane.

KEYWORDS: $SrCu_2(BO_3)_2$, neutron scattering, 2D quantum spins, spin gap, Shastry-Sutherland model

§1. Introduction

Study on spin gap systems with the singlet ground state in low-dimensional quantum spin systems, which are realized by the strong quantum effect, is one of the hot topics in strongly correlated electron physics in recent years. $SrCu_2(BO_3)_2$ is a recently discovered two-dimensional (2D) spin-gap system which is actively studied by both theorists[1)] and experimental scientists.[2)]

$SrCu_2(BO_3)_2$ has a tetragonal structure of the space group of $I\bar{4}2m$ and lattice parameters $a = 8.995$ Å and $c = 6.649$ Å at room temperature. All Cu^{2+} ions with spin 1/2 are located crystallographically equivalent sites. The 2D layers containing the Cu^{2+} ions are separated by planes of Sr^{2+} ions. The magnetic susceptibility[2)] shows a broad maximum around 15 K and drops rapidly to zero with reducing the temperature, indicating a transition to a non-magnetic singlet ground state. A neighboring pair of the Cu^{2+} ions forms a spin dimer and the neighboring dimers are connected orthogonally. Miyahara and Ueda[1)] found that its unique 2D spin structure is equivalent to the Shastry-Sutherland model,[3)] which has an exact dimer ground state. Another interesting aspect is quantized plateaux in the magnetization, for example, at one-quarter and one-eighth of the full Cu^{2+} moment[2)] owing to the extremely localized nature of the excited triplet state. From previous experimental results[2)] and theoretical calculations,[1)] it has been recognized that $SrCu_2(BO_3)_2$ is an $S = 1/2$ highly frustrated spin gap system with an energy gap of $\Delta = 34$ K and with a ratio of intradimer (J) and interdimer (J') exchange of $J'/J = 0.68$ ($J = 100$ K, $J' = 68$ K) very close to the critical value of $(J'/J)_c \sim 0.7$, where Miyahara and Ueda[1)] also indicated that a phase transition from a singlet dimer to a Néel state occurs with increasing J'/J.

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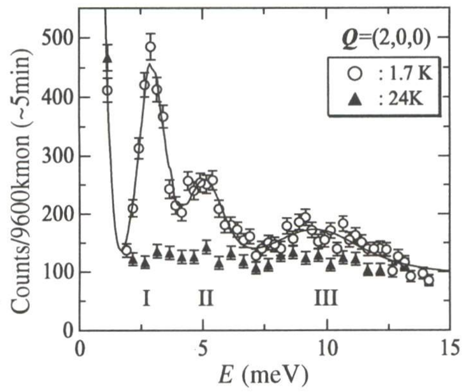

Fig. 1. Energy scans at $Q = (2,0,0)$ obtained at $T = 1.7$ K (circles) and 24 K (triangles). The excitations are labeled at the bottoms of the figure. The solid curve is the fit to the profile at 1.7 K. The abrupt upturn around energy zero is due to the incoherent elastic scattering.

In the present work, we have investigated magnetic excitation spectrum of single crystals of $SrCu_2(BO_3)_2$ by means of inelastic neutron scattering techniques. We discuss the features of the excited triplet states in connection with the orthogonality of the dimers.

§2. Experimental

Single crystals of $SrCu_2(^{11}BO_3)_2$ with a total mass of ~ 1.5 cm^3 were prepared by the traveling solvent floating zone method using $Li^{11}BO_2$ flux.[4)]

Inelastic neutron scattering experiments were performed on the ISSP-PONTA triple-axis spectrometer installed at JRR-3M reactor at Tokai establishment of the Japan Atomic Energy Research Institute. The spectrom-

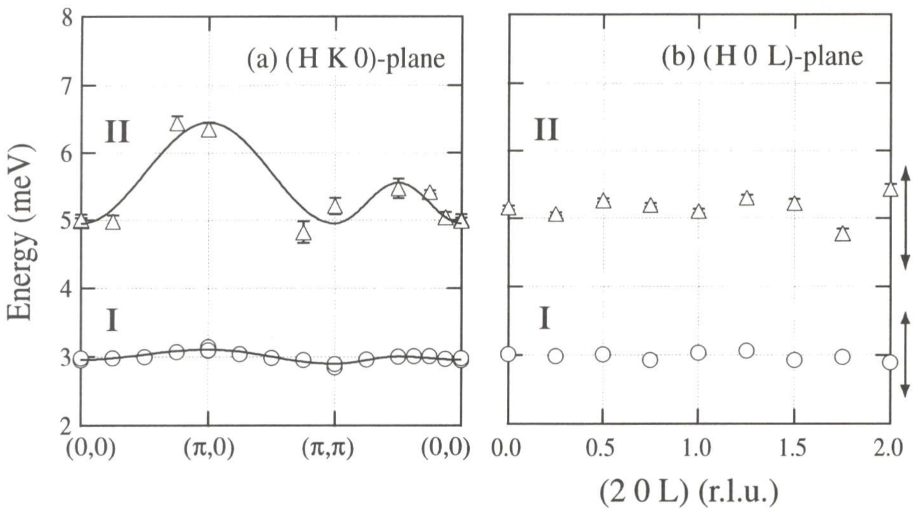

Fig. 2. Q-dependence of the excitation energies in the $(H\ K\ 0)$ plane (left) and $(H\ 0\ L)$ plane (fight) obtained at 1.7 K. The solid curves are guides to the eye. The arrows represent the energy resolutions of the instrument (FWHM). The unit cell in the $(H\ K\ 0)$ plane (left) by the notation of Weihong *et al.*[5,6] is used. $Q = (2,\ 0,\ L)$ in the right panel corresponds to the (0, 0) point in the left one.

eter was operated with fixed final energy $E_f = 14.7$ meV ($k_f = 2.67$ Å^{-1}) and collimations of open(40')-40'-80'-80'. A PG filter was placed after the sample to reduce the higher order contaminations. The crystals are oriented so as the scattering plane contains a^*- and b^*-axes or a^*- and c^*-axes.

§3. Results and Discussion

Figure 1 shows typical energy scans obtained at 1.7 K and 24 K for a wave vector of $Q = (2,0,0)$. Three magnetic excitations at 1.7 K ($T \ll \Delta$) are observed around 3, 5 and 9 meV (excitations I, II and III, respectively), while no distinct peaks are observed at 24 K. The excitation I centered at $\Delta = 3.0$ meV corresponds to the transition of a single triplet from the singlet ground state, as indicated by other experiments.[2,7,8] The solid curve is a fit to the 1.7 K profile convoluted with instrumental resolution. It should be noted that the excitation I showed almost energy resolution limited behavior, while the excitations II and III tend to have intrinsic energy widths. Precise temperature dependence of these excitations I, II and III (not presented here) shows that all of these three excitations apparently vanish at about 13 K,[6] implying that the excitations II and III consist of multi-triplet excitations. The temperature of 13 K is only one-third of spin gap energy ($\Delta = 3.0$ meV). A similar behavior has also been seen in Raman scattering study.[8] We suppose that this anomalous temperature dependence of intensity possibly comes from a strong spin frustration due to the orthogonality of the dimers.

The dispersion relation of the excitation I and II for the $(H\ K\ 0)$ and $(H\ 0\ L)$ planes are shown in Fig. 2 (a) and (b), respectively. It is noted that the energies of the excitation I in both the planes are almost dispersionless and change within ± 0.2 meV at most, indicating the single triplet hardly moves both in the 2D plane and along the c-axis. Miyahara and Ueda[1] theoretically showed that the dispersionless behavior in $SrCu_2(BO_3)_2$ is not due to the spatial isolation but due to the orthogonality of the neighboring dimers. According to their perturbation calculations,[9] a hopping of the single triplet from one site to another within the 2D a-b plane is possible only from the sixth order, leading to an extremely weak dispersion of the excitation I in the $(H\ K\ 0)$ plane, in excellent agreement with the present result shown in Fig. 2 (a).

They also argured the effects of the interlayer coupling J'' and proved that the single triplet excitation cannot move along the c-axis,[10] indicating the complete flat dispersion along the c^* direction. As shown in Fig. 2. (b), the obtained dispersion relation of the excitation I is almost flat with scatter of 0.2 meV, which is similar to the dispersion energy in the 2D plane. To clarity the above argument, we need to perform experiments with a better energy resolution.

Then let us look at the second triplet excitation II at 5 meV. This excitation II can be interpreted as correlated two-triplet excitation, discussed in the reference,[6] and has been already observed by ESR[7] and Raman Scattering.[8] In Fig. 2 (a), the dispersive excitation II has been observed between 5 meV and 6.5 meV. This means that the correlated two triplets can propagate more easily than the single triplet in accord with the thoeretical prediction.[11] Considering the instrumental energy resolution, the intrinsic energy width of the excitation II for any Q points is obtained to be about 0.7 meV. The finite width possibly indicates a short life time of correlated two triplets and/or the existance of several excited state with closely degenerate discrete energies. The striking difference between the two-triplet excitation in the 2D plane and that along the c^* direction is that the energy of the excitation II is almost flat as illustrated in Fig. 2 (b). This observation indicates that the correlated two triplets can only move within the 2D plane.

The higher-energy excitation in the energy range of 8-12 meV would possibly arise from the correlated three-triplet or multi-triplet excitations, due to the highly frustrated spin dimers.

§4. Summary

We performed inelastic neutron scattering experiment on $SrCu_2(^{11}BO_3)_2$ to find three magnetic excitations. The lowest single triplet excitation from the ground state was observed at 3.0 meV, in excellent agreement with the previous measurements.[2,7,8] The almost dispersionless behavior arises from the orthogonality of the neighboring dimers. The second triplet excitation around 5 meV can be interpreted as correlated two-triplet excitation. It moves more easily in the *a*-*b* plane than the single triplet does, but hardly does along the *c*-axis.

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