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High Energy-Resolution Inelastic Neutron Scattering Experiments on Triplet Bound State Excitations in $SrCu_2(BO_3)_2$

Naofumi ASO*, Hiroshi KAGEYAMA[1], Katsuyuki NUKUI†, Masakazu NISHI, Hiroaki KADOWAKI[2], Yutaka UEDA[3] and Kazuhisa KAKURAI[4]

Neutron Science Laboratory, Institute for Solid State Physics, University of Tokyo, Tokai, Ibaraki 319-1106
[1]*Graduate School of Science, Kyoto University, Kyoto 606-8502*
[2]*Department of Physics, Tokyo Metropolitan University, Hachioji, Tokyo 192-0397*
[3]*Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581*
[4]*Advanced Science Research Center, Japan Atomic Energy Research Institute, Tokai, Ibaraki 319-1195*

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In this letter we report on the high energy-resolution neutron scattering studies on the dynamic structure factor of the two-triplet bound states in the two-dimensional $S = 1/2$ Shastry–Sutherland system, $SrCu_2(BO_3)_2$, in the energy transfer range of 4 to 7 meV. In contrast to the earlier interpretation of the low energy-resolution results, these bound state excitations show hardly any dispersion, but do show a strong wave-vector-dependent structure factor. Although these findings are in overall agreement with the recent theoretical calculation of the 2-triplon dynamic structure factor by Knetter and Uhrig [Phys. Rev. Lett. **92** (2004) 027204], they also show distinct differences, which may be due to some symmetry-breaking terms in the Hamiltonian that are not included in the theory, such as the interdimer Dzyaloshinski–Moriya interactions recently discovered in this compound.

KEYWORDS: two-dimensional (2D), orthogonal-dimer system, Shastry–Sutherland, Dzyaloshinski–Moriya interaction, inelastic neutron scattering, spin gap
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After the discovery that $SrCu_2(BO_3)_2$ is a good realization of the two-dimensional Shastry–Sutherland model[1] by Kageyama *et al.*[2] and Miyahara and Ueda,[3] many experimental and theoretical studies have been undertaken to characterize and understand the spin liquid state of this quantum antiferromagnetic system. It is now widely accepted that $SrCu_2(BO_3)_2$ should be regarded as an $S = 1/2$, highly frustrated orthogonal dimer system with a singlet ground state with a gap energy of $\Delta = 34$ K.[3] The ratios of the intradimer exchange J and the interdimer exchange J' of $J'/J = 0.635$[4] and 0.603[5] have been proposed, being close to the critical value of $(J'/J_c) \sim 0.7$, where a quantum phase transition from a singlet dimer state to a Néel state is predicted.[3] The first inelastic neutron scattering (INS) experiment evidenced an almost localized first triplet excitation resulting in an almost wave-vector-independent spin excitation at around 3 meV.[6] The localized nature of the first triplet excitation has been explained to be a result of the frustration of the interdimer interaction due to the orthogonal arrangements of the dimers. This interaction, even to the fifth order in perturbation, prohibits the excited triplet excitations from hopping from one dimer to another.[7] The numerical exact diagonalization[8] results were consistent with this interpretation.

At the same time the first INS also clearly showed the existence of rather intense higher-energy excitations, i.e., around 5 and 9 meV.[6] These higher-energy excitations were also observed in ESR[9] and Raman scattering[10] experiments and attributed to the bound states theoretically.[5,7,8,11] Here again the low energy-resolution experimental results with a limited number of $\boldsymbol{Q}$-points for higher-energy excitations seemed to be in agreement with the finite-size exact diagonalization results, suggesting a more dispersive nature for these bound states. However, since the finite-size effect for an extended bound state is expected to cause severe restrictions and since the subsequent high energy-resolution experiments on the first triplet excitation clearly indicated the importance of the interdimer Dzyaloshinski–Moriya (DM) exchange interaction describing the $\boldsymbol{Q}$-dependent splitting of the triplet excitations and their field dependence,[12–14] we decided to extend the high energy-resolution INS experiment to the higher-energy excitation range of up to 7 meV to investigate the nature of the bound states in $SrCu_2(BO_3)_2$.

We thus report in the present letter the high-resolution INS experiment using a cold-neutron triple-axis instrument to reveal the wave-vector-dependent excitation spectra of the bound state in the strongly frustrated two-dimensional $S = 1/2$ Shastry–Sutherland system $SrCu_2(BO_3)_2$.

^{11}B-enriched (99.6%) bulk single crystals of $SrCu_2(^{11}BO_3)_2$ have been prepared by the traveling solvent floating zone method using $Li^{11}BO_2$ flux.[15] Inelastic neutron scattering experiments were performed on the cold-neutron triple-axis spectrometer ISSP-HER installed at the JRR-3M reactor at the Tokai establishment of the Japan Atomic Energy Research Institute. The spectrometer is equipped with a vertically focused PG (002) monochromator and a horizontally focused PG (002) analyzer and is operated with fixed final energy $E_f = 3.1$ meV ($k_f = 1.22$ Å^{-1}) and collimations after the monochromator of open–radial collimator–open. A combination of a PG Bragg reflection filter for $E_i \geq 5.1$ meV and another Be filter for $E_i < 5.1$ meV before the sample and Be filter after the sample is used to reduce higher-order contaminations. The sample consisted of four aligned single crystals with a total mass of ~ 9 g and was oriented with its a^*- and b^*-axes. The notation of the unit cell in the ($HK0$) plane as used by Zheng *et al.*[16] is

*E-mail: aso@issp.u-tokyo.ac.jp
†Present address: Fuji Photo Film Co., Ltd.

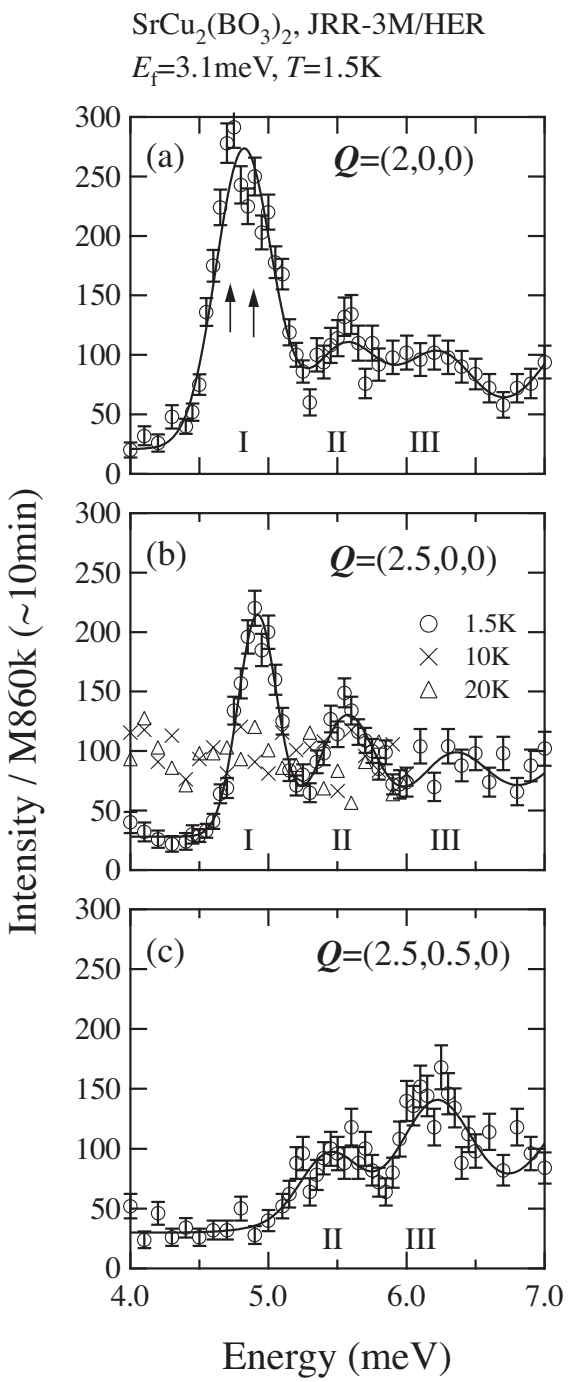

Fig. 1. Examples of INS spectra at (a) $\boldsymbol{Q} = (2, 0, 0)$, (b) $\boldsymbol{Q} = (2.5, 0, 0)$ and (c) $\boldsymbol{Q} = (2.5, 0.5, 0)$ obtained at $T = 1.5$ K (circles). Spectra at $\boldsymbol{Q} = (2.5, 0, 0)$ contain the data at $T = 10$ K (crosses) and 20 K (triangles). The peaks are labeled at the bottom of the figure. The solid curve is the fit to the data, as illustrated in the text.

employed. An ILL-type orange cryostat was used to cool the sample down to 1.5 K.

Constant-$\boldsymbol{Q}$ energy scans at $T = 1.5$ K were performed at different $\boldsymbol{Q}$'s in different directions of the reciprocal plane. Figure 1 shows typical INS spectra in the energy range of 4 to 7 meV at characteristic $\boldsymbol{Q}$ points. In Fig. 1(a) the spectrum at $\boldsymbol{Q} = (2, 0, 0)$ corresponding to the $(0, 0)$ position is depicted. In contrast to the previous thermal neutron scattering experiment with moderate energy-resolution (~1.5 meV) where one broad peak at around 5 meV with an intrinsic line width of ~ 0.7 meV was observed,[6] one clearly recognizes three excitation peaks centered at about 4.8, 5.5 and 6.2 meV in the present experiment with higher energy-resolution. The solid line in the spectrum is a tentative fit using three Gaussian peaks convoluted with the instrumental resolution. (Note that we have assumed higher-energy excitations above 7 meV, as has been shown in the previous low energy-resolution experiment, to account for the increase in the intensities towards 7 meV.) The three excitations, which will be referred to as excitations I, II and III, all have finite intrinsic energy widths (full width at half maximum (FWHM)) of 0.42, 0.51 and 0.77 meV, respectively. The finite intrinsic energy widths may arise from the finite lifetime of the excitations or the existence of numerous discrete excitations. A closer look at excitation I in Fig. 1(a) suggests an intrinsic peak structure pointing to a two-peak structure centered at 4.7 and 4.9 meV as marked by the arrows. The fact that the recent ESR measurement by Nojiri *et al.*[17] shows at least five lines between 4.7 and 5.6 meV strongly suggests that the latter is the case at this low temperature. However, to clarify how many peaks the spectrum in this energy range is composed of, we would need an even higher energy-resolution that is not yet available in neutron scattering. We therefore continue to fit the spectra at different $\boldsymbol{Q}$ points using the groupings of (I), (II) and (III) as introduced above for $\boldsymbol{Q} = (2, 0, 0)$.

Figure 1(b) depicts the spectrum for $\boldsymbol{Q} = (2.5, 0, 0)$. The overall features of this spectrum are similar to those of $\boldsymbol{Q} = (2, 0, 0)$. In this graph the spectra taken at 10 and 20 K are also shown. The temperature dependence of the intensities shows that the well-structured spectrum at 1.5 K becomes flat and featureless when increasing the temperature up to 10 and 20 K. This clearly shows that the excitations in this energy range are of magnetic origin.

Figure 1(c) shows the spectrum at $\boldsymbol{Q} = (2.5, 0.5, 0)$. In contrast to the spectra at the previous two wave vectors, it is obvious that excitation (I) has no spectral weight. The two broad peaks associated with (II) and (III) excitations are observed to be centered at 5.5 and 6.2 meV. These results are consistent with the previous low energy-resolution results assigning one broad peak at around 6 meV with an intrinsic energy width of 0.7 meV.[6]

Figure 2 summarizes the fit results, as described above, of all the spectra in different directions. Figure 2(a) displays the energies and widths, indicated by the vertical bars, of the three excitation groups and Fig. 2(b) depicts their integrated intensities. It is clear from these results that all the excitation groups have almost constant $\boldsymbol{Q}$-independent energies and that the spectral weights of the groups strongly vary with $\boldsymbol{Q}$. In particular, the intensity of excitation group (I) acquires its maximum at $\boldsymbol{Q} = (2, 0, 0)$ and decreases gradually by moving away from this point. At around $\boldsymbol{Q} = (2.5, 0.5, 0)$ the spectral weight of (I) vanishes and the intensities of (II) and (III) dominate the spectra. These characteristic structure factors of the dispersionless excitation groups led to the energy points at around 5 and 6 meV in the previous coarse energy-resolution results (Fig. 3 of ref. 6), which at that time were explained by the strongly dispersing character of the bound triplets excitations. These high energy-resolution results clearly demonstrate that there are many isolated energy levels of bound states in the energy range of 5 to

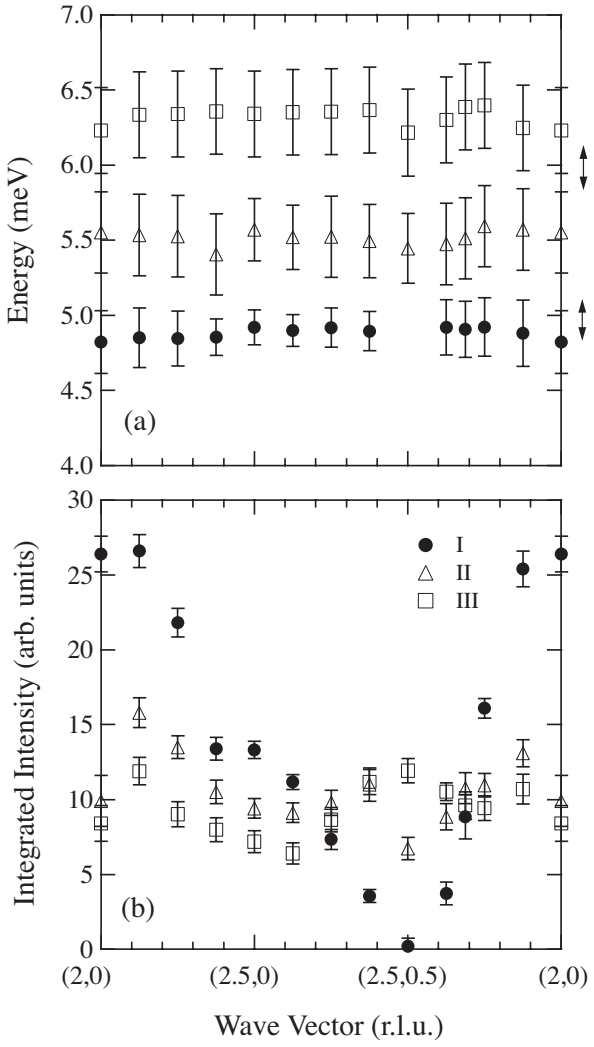

Fig. 2. (a) Q dependence of the energy of excitations I (circles), II (triangles) and III (squares), obtained at 1.5 K. The bars represent the intrinsic linewidth (FWHM) of excitations I, II and III. The arrows represent the energy resolution of the instrument (FWHM) at 5.0 and 6.0 meV. (b) Q dependence of the integrated intensity of excitations I, II and III.

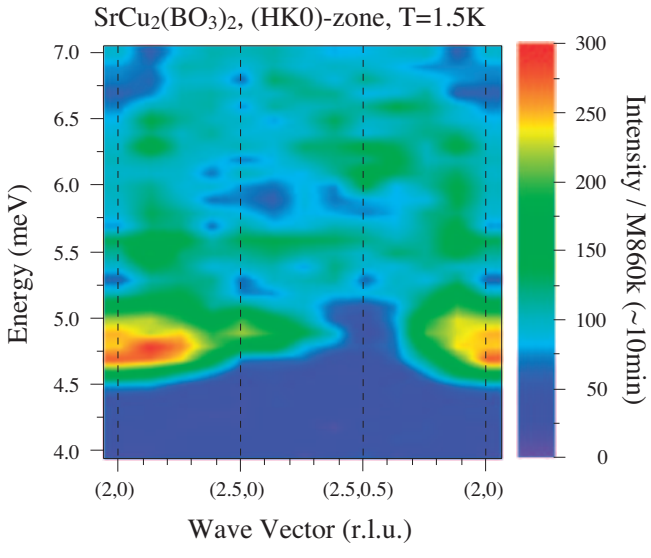

Fig. 3. Image plot of the scattering intensities in the energy–wave vector plane. Intensities are indicated by the scale on the right hand side.

6.5 meV. This energy range corresponds to the previously discussed bandwidth of enhanced correlated hopping based on the coarse energy-resolution results. At the same time the results clearly show that the dispersion of the individual bound-state mode in this energy range is at most 0.5 meV and hence the bandwidth due to the correlated hopping of the bound triplets excitations is experimentally determined to be of the order of 0.5 meV in $SrCu_2(BO_3)_2$. One should note that the finite-size exact diagonalization results[8] at limited wave-vector positions indicated a bound-state excitation level at around 5 meV with a bandwidth of 0.5 meV.

Figure 3 is an image plot of the scattering intensities in the energy–wave vector plane. The above conclusions drawn from the fit results can be reconciled and the almost dispersionless nature and the $\boldsymbol{Q}$-dependent structure factor of the excitations are clearly visible.

Very recently Knetter and Uhrig calculated the dynamical structure factor of two-triplet bound-state excitation, which they call 2-triplon, in $SrCu_2(BO_3)_2$ by means of perturbative continuous unitary transformations.[18] In contrast to the numerical exact diagonalization calculation, this perturbative method can provide continuous distribution in the higher-order bound states' spectra and hence can be directly compared with the INS results. They could well reproduce the spectra at $\boldsymbol{Q} = (2, 0, 0)$ and $(2.5, 0.5, 0)$ [Figs. 2(a) and 2(c)] using $J'/J = 0.603$ and $J = 6.16$ meV (see Fig. 2 of ref. 18). The spectral density of the 2-triplon channel (Fig. 3 of ref. 18) calculated with the same set of the parameters also well reproduces the essential features of our results represented in Fig. 3. They interpret the nonsignificant momentum dependence of the 2-triplon eigenenergies as evidence for level repulsion. Due to the negligible kinetic energy of the single-triplet state, there are a relatively large number of individual bound states involved and their energetic repulsion causes each individual band to be very flat.

Although the overall agreement seems to be satisfactory, particularly the momentum dependence of the spectral weight at an energy transfer of 4.8 meV and the rather strong intensity at around 6 meV at $\boldsymbol{Q} = (2.5, 0.5, 0)$, there are some features in the observed spectra which are not accounted for by the calculation. First of all the spectral weight of excitation group (II) at 5.5 meV is much less pronounced in the calculation, although the theory predicts eigenenergy states at that energy. Secondly the observed intensities at around 6.5 meV are absent in the calculation. Furthermore, experimentally we do see a finite width in the excitation groups, which probably indicates that each mode consists of numerous split states, as discussed above. These discrepancies may be due to the interdimer DM interactions in $SrCu_2(BO_3)_2$ not included in the calculation. The high energy-resolution INS measurements of the first triplet excitation at around 3 meV revealed the $\boldsymbol{Q}$-dependent splitting of the modes in zero field and Zeeman splitting in a field, which can be only explained by assuming the interdimer DM interactions.[12,13] The qualitative changes in the kinetic energies of the first triplet excitation due to this term may show up in the subtle modifications of the bound-state spectra, as has been observed in the experiment.

To summarize, we have performed high energy-resolution INS experiments on the two-triplet excitations in the

Shastry–Sutherland model substance $SrCu_2(BO_3)_2$. In contrast to the earlier interpretation of the low energy-resolution results, there are a relatively large number of isolated energy levels of two-triplet bound states, which show hardly any dispersion, but show a strong wave-vector-dependent structure factor. These findings are in overall agreement with the recent theoretical calculation of the "2-triplon", i.e., the two-triplet bound state, dynamic structure factor obtained by Knetter and Uhrig,[18] but show also distinct differences, which may be due to some symmetry-breaking terms in the Hamiltonian that are not included in the theory, such as the interdimer Dzyaloshinski–Moriya interactions recently discovered in this compound.

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