

Inelastic Neutron Scattering Experiment on Dzyaloshinski-Moriya Interaction in $\text{SrCu}_2(\text{BO}_3)_2$

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Abstract

We report high energy-resolution inelastic neutron scattering results on the strongly frustrated, two-dimensional(2D) orthogonal-dimer system $\text{SrCu}_2(\text{BO}_3)_2$. Strongly localized first triplet excitations at around 3meV in zero and applied magnetic field perpendicular to the 2D plane are studied. The high energy-resolution measurement of the dispersion reveals interesting fine structure of the excited triplet state. The magnetic field dependence also shows unusual wave vector dependence. The spin anisotropies caused by the Dzyaloshinski-Moriya interaction with both the out-of-plane and in-plane components can explain the complex nature of the first excited state in this frustrated 2D quantum antiferromagnet.

1 Introduction

$\text{SrCu}_2(\text{BO}_3)_2$ (Strontium Copper Borate) is a new example of a magnetic oxide with a singlet ground state[1]. It was pointed out by Miyahara and Ueda[2], that this system can be mapped on the Shastry-Sutherland model[3] close to the quantum phase transition from a singlet dimer to a Néel state. The isotropic Hamiltonian of the frustrated Shastry-Sutherland

model

$$H = J \sum_{nn} \mathbf{S}_i \mathbf{S}_j + J' \sum_{nnn} \mathbf{S}_i \mathbf{S}_j$$

with nearest neighbour (nn) interaction J and next nearest neighbour (nnn) interaction J' being antiferromagnetic, has a ground state well described as simply a product of magnetic dimers in two dimensions on the bonds formed by the strongest nn magnetic exchange. The weaker nnn exchanges are frustrated by the geometry and the ground state of the Hamiltonian is independent of the value of the weaker exchange, up to a critical value, which is given by the critical ratio of $(J/J')_c=0.7$. For $\text{SrCu}_2(\text{BO}_3)_2$ Miyahara and Ueda estimated $J=85\text{K}$ and $J'=54\text{K}$ from the susceptibility and the gap using the above Hamiltonian. One of the remarkable features of its excited states is that a single triplet excitation can propagate only from the sixth order in the perturbation calculations. The first inelastic neutron scattering experiment using thermal neutron triple axis instrument with moderate energy resolution [4] indeed showed the lowest 3.0 meV excitation being almost dispersionless with a bandwidth of 0.2 meV at most. Recent ESR [5] and magnetization [1,6] experiments however indicate the existence of small spin anisotropy terms in the Hamiltonian of $\text{SrCu}_2(\text{BO}_3)_2$. Although the anisotropies are expected to be small they may be essential to the nature of this system, especially because it is believed to be close to a quantum critical point in parameter space. To clarify the origin of the spin anisotropies we performed high energy-resolution inelastic neutron scattering experiment on $\text{SrCu}_2(\text{BO}_3)_2$ in zero and applied magnetic field.

2. Experiments

The inelastic neutron scattering experiments were performed on the cold neutron triple axis instrument ISSP-HER at JRR-3M of Japan Atomic Energy Research Institute. The instrument is installed on a cold neutron guide and is equipped with vertically curved pyrolytic graphite PG(002) monochromator and horizontally curved PG(002) analyser. A combination of PG Bragg reflection filter for $E_i > 5.1\text{meV}$ and cooled beryllium filter for $E_i < 5.1\text{meV}$ before the sample is used to suppress higher-order contamination. The experimental conditions were k_f being fixed at 1.22\AA^{-1} ($E_f=3.10\text{meV}$) and collimations after the monochromator being open-radial collimator-open.

^{11}B -enriched (99.6%) bulk single crystal of 3.6g was used. The sample was oriented with its a- and b-axis in the scattering plane in the cryomagnet and the magnetic field was applied vertically along the c-axis.

Figure 1 shows series of energy scans at different wave vectors in zero field. It can be

recognized that the first excitation around 3meV energy transfer indeed consists out of at maximum three excitation branches. Figure 2 summarizes the observed excitation energies of the branches in different directions. The energies were determined by fitting gaussian line shapes with resolution limited line widths as indicated by the thin solid lines in the spectra of Fig.1. One recognizes three different types of spectra in zero field. At $Q=(1,0,0)$ one observes clearly three peak structure, while only two peaks are observable at $Q=(1.5,0,0)$. In contrast to these points one observes only one resolution limited peak at $Q=(1.5,0.5,0)$. The Zeeman

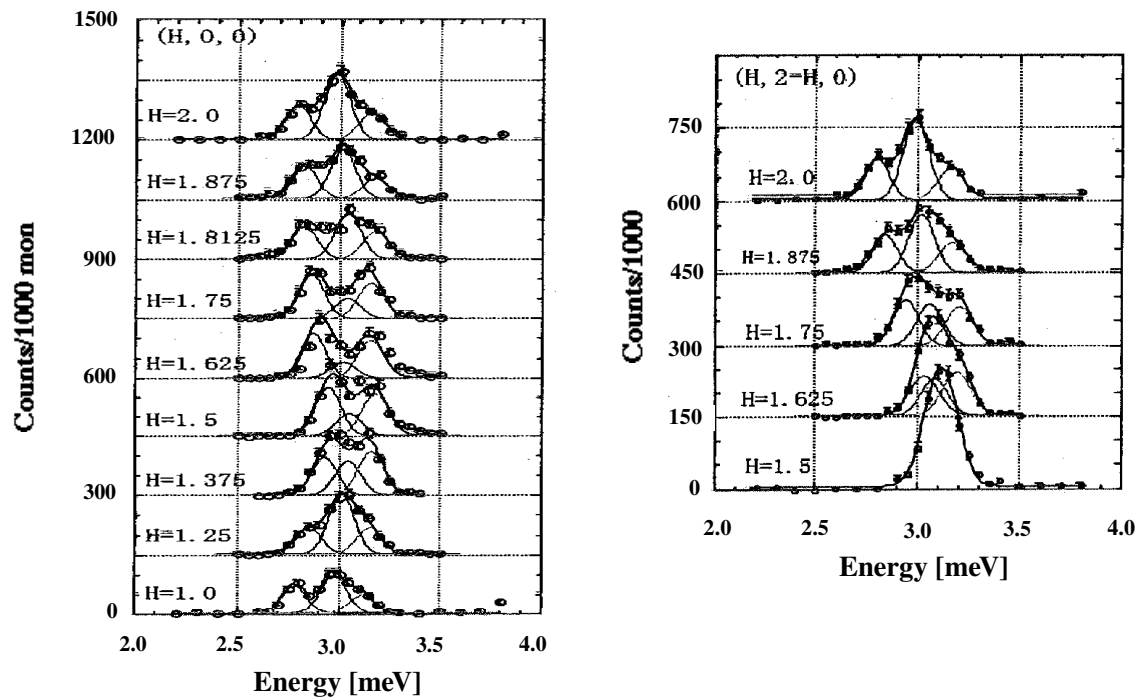


Fig. 1: Spectra at different Q-vectors.

splitting in the applied vertical field for these reciprocal points are shown in Fig.3. As has been already reported previously[7] the three branches at $Q=(1,0,0)$ in zero field split into five branches in the applied field. While the central peak remains unchanged in the field, the upper and lower branches each split into two modes. The resolution limited peak at

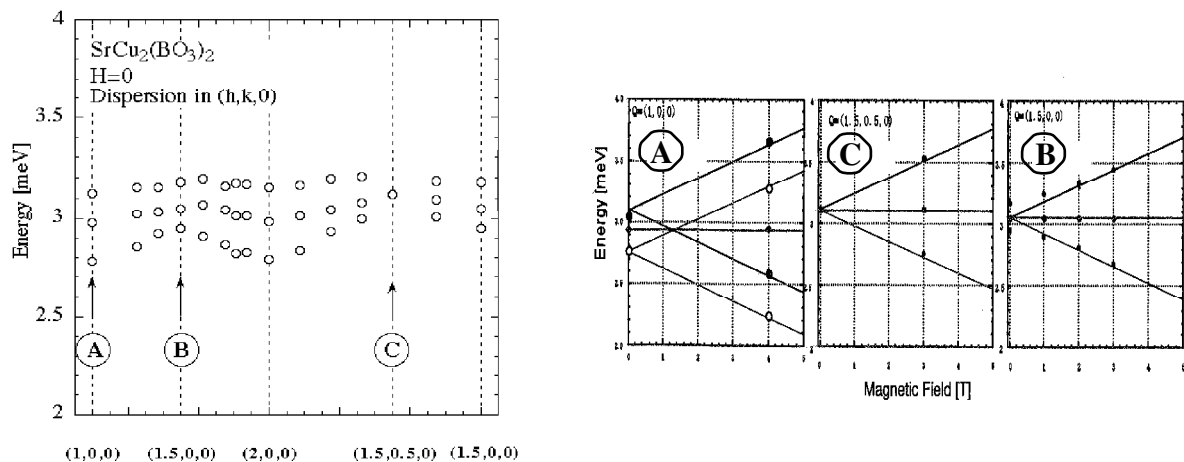


Fig.2: Dispersion of 3meV excitation.

Fig.3: Zeeman splitting at different Q-vectors.

$Q=(1.5,0.5,0)$ splits into three peaks, as one would expect of an isotropic triplet excitation. At $Q=(1.5,0,0)$ the two peak structure in zero field changes over to a three peak structure under applied field same as observed at $Q=(1.5,0.5,0)$. It looks as the central peak at $Q=(1.5,0,0)$ has almost no spectral weight in zero field, but its intensity grows with the application of the magnetic field, as shown in Fig. 4.

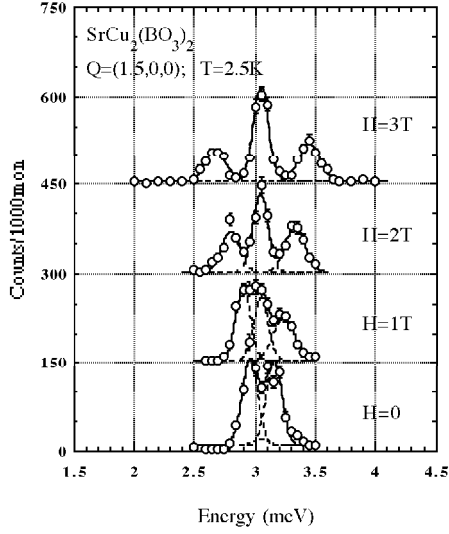


Fig.4: Field dependence of the spectrum at $Q=(1.5,0,0)$.

3. Discussion

small J' and $D_{OP}=0$ a perturbative approach gives to linear order in J' and D_{OP} the following dispersions for the two modes each twice degenerate with $S_z=\pm 1$;

$$\omega_q^\pm(S^z=+1) = \omega_q^\pm(S^z=-1) = J \pm 2D_{OP} \cos(q_a a/2) \cos(q_b a/2).$$

The Dzyaloshinski-Moriya interaction has no effect on the $S_z=0$ component of the triplet, so that its energy remains unchanged and dispersionless.

$$\omega_q(S^z=0) = J$$

In particular at $q=2\pi/a$, corresponding to $Q=(1,0,0)$, we have an upper and lower modes $S_z=\pm 1$ with $\omega(S^z=\pm 1) = J+2D_{OP}$ and $\omega(S^z=\pm 1) = J-2D_{OP}$, respectively. A magnetic field parallel to D_{OP} splits these modes in four branches. The energy of the mode $S_z=0$ remains exactly at the middle of the four. This is in agreement with the above neutron scattering

In the previous paper[7] we have shown that the Zeeman splitting observed at $Q=(1,0,0)$ can be understood when the anisotropic Dzyaloshinski-Moriya (DM) terms in the interdimer interactions nnn

$$H_{DM} = \sum_{nnn} \pm D_{OP} \mathbf{e}_c (\mathbf{S}_i \times \mathbf{S}_j)$$

are assumed whose vectors are perpendicular to the plane (\mathbf{e}_c : unit vector in c -direction) with an alternation of the DM vector direction from bond to bond. This DM interaction is not frustrated because of the alternation of its vector. Hence the degeneracy of the localized triplets is lifted to first order in D_{OP} and small but appreciable dispersion is introduced. Starting from the exact ground state for

result. Furthermore the theory predicts a specific q -dependence, namely the degeneracy restored at π . The experimental observation of the single peak structure in zero field at $Q=(1.5,0.5,0)$ and the ordinary triplet splitting observed under applied field is in accord with this prediction. Thus the introduction of the DM interaction perpendicular to the a - c plane qualitatively accounts for the detailed q -dependent behaviour of the first excitations in $(1,1,0)$ direction in zero and applied field. However the above theory would predict the same behaviour in $(1,0,0)$ direction, i.e. a single peak structure at $Q=(1.5,0,0)$. The experiment in contrast shows a double peak structure at zero field and a triplet Zeeman splitting under applied field. How can one understand this unexpected feature? The result of ESR measurement under applied field in the plane may be regarded as a hint to the solution. There the field is perpendicular to the above DM interaction and the energies remain twice degenerate and are given by

$$\omega_q^\pm = J \pm \sqrt{(4D_{OP}^2 f^2(q) + (g_\perp \mu_B H_\perp)^2)} \quad \text{and} \quad \omega_q^0 = J \quad \text{with} \quad f(q) = \cos(q_a a/2) \cos(q_b a/2).$$

As you can see, the neutron scattering result at $Q=(1.5,0,0)$ under applied field surprisingly resemble this situation, indicating that there may be additional DM component perpendicular to the vertically applied field, i.e. a in-plane DM component. A closer look at the structure of $\text{SrCu}_2(\text{BO}_3)_2$ then justifies the existence of such an in-plane component of DM vector due to

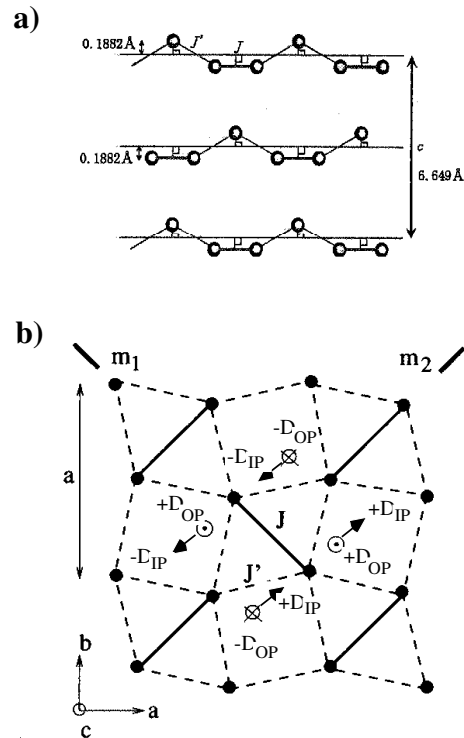


Fig.5:a) Buckling of the dimer plane.

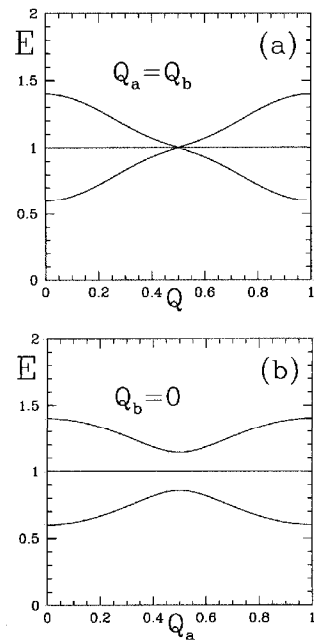


Fig.6: Dispersion of the triplet excitation for (a) $Q=Q_a=Q_b$

b) Scheme of the in-plane component D_{IP} and (b) Q_a with $Q_b=0$. $D_{IP}=1/2 D_{OP}$.

the fact that the (a,b) plane is not exactly a mirror plane. The orthogonal dimers are located not exactly on a single plane, but are alternately located slightly above and below a plane, as shown in Figure 5a). Introducing in-plane component partially lifting the frustration in the triangular interdimer configuration, as indicated in Fig. 5b), one can derive in the same perturbative approach as above the Hamiltonian to linear order in J' and $D_{OP,IP}$ for small J' . Diagonalizing the Hamiltonian then gives dispersion relations in zero field as shown in Fig. 6. It can be seen that the previous results for (1,1,0) direction is unchanged, but the dispersion in (1,0,0) direction is modified in accordance with the experimental result (see Fig.2). The effect of the magnetic field perpendicular to the plane is predicted to be also in accord with the experimental findings. The neutron spectral weight for $S_z=0$ is predicted to be zero in zero applied field and to grow with applied field, corresponding to the observed double peak structure in zero field and the triplet Zeeman splitting under applied field.(see Fig.4) The explicit value of the in-plane component estimated from the splitting at $Q=(1.5,0,0)$ would be $D_{IP}\approx 0.07\text{meV}$ and would be more than 1/3 of the perpendicular component ($D_{op}=0.18\text{meV}$). This would correspond to almost 21 degrees 'buckling' of the DM vector. But the structural buckling due to the small deviation of the dimers from the plane is only about 1 degree and it is an open question whether the splitting observed at $Q=(1.5,0,0)$ is solely due to the in-plane DM component. A higher order correction term of the frustrated interdimer interaction J' may also contribute to the splitting at $Q=(1.5,0,0)$.

4. Conclusion

The high energy-resolution inelastic neutron scattering experiments on $\text{SrCu}_2(\text{BO}_3)_2$ under applied field clearly demonstrate that the q -dependent behaviour of the lowest excitation in this highly frustrated 2-D system is mainly determined by the Dzyaloshinski-Moriya interaction. The unusual q -dependent features of the lowest excited states in zero field and their Zeeman splitting can be well described by assuming the Dzyaloshinski-Moriya interaction with both the out-of-plane and in-plane components. These anisotropic terms, although small, may play important role when comparing the theoretical predictions for the Shastry-Sutherland model with the experimental results on $\text{SrCu}_2(\text{BO}_3)_2$ because of its near quantum criticality. Another interesting consequence of the above findings is the possibility of 'dynamical' Dzyaloshinski-Moriya interaction. Since the existence of Dzyaloshinski-

Moriya interaction strongly depends on the symmetry of the lattice, the interaction is expected to be strongly coupled to a specific dynamical distortions caused by lattice vibrations. Selection rules for optical transitions as observed by ESR and infrared absorption are already suggesting the existence of such a dynamical term[5,8,9]. Hence $\text{SrCu}_2(\text{BO}_3)_2$ may be an excellent model system to study such a novel dynamical spin-lattice coupling.

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