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The Magnetic Excitations in $SrCu_2(BO_3)_2$

Shin MIYAHARA and Kazuo UEDA

Institute for Solid State Physics, University of Tokyo, Kashiwanoha, Kashiwa, Chiba 277-8581, Japan

The $SrCu_2(BO_3)_2$ is a unique two-dimensional spin gap system and the magnetic behavior of this compound at low temperature is explained well by the two-dimensional orthogonal dimer model. We discuss the character of the triplet excitations and show that the triplet excitations are almost localized. This almost localized nature is observed in the neutron scattering as a small dispersion for the lowest triplet branch. Recently the neutron scattering experiments and ESR experiments show that the lowest triplet branch splits. The splitting of the triplet excitation can be explained by Dzyaloshinsky-Moriya interactions.

KEYWORDS: $SrCu_2(BO_3)_2$, orthogonal dimer state, Shastry-Sutherland model, Dzyaloshinsky-Moriya interactions, ESR, neutron scattering.

§1. Introduction

$SrCu_2(BO_3)_2$ is a new spin gap system[1] and the behaviors of this material are studied intensively from both theoretical and experimental aspects. The magnetic behaviors of the $SrCu_2(BO_3)_2$ at low temperature are explained well by the two-dimensional orthogonal dimer Heisenberg model, which is equivalent to the Shastry-Sutherland model.[2,3] In this model a triplet excitation is almost localized and this feature explains the appearance of the magnetization plateaus.[1,3–6] The magnetic excitations in $SrCu_2(BO_3)_2$ have been studied by ESR,[7] inelastic neutron scattering,[8] and Raman scattering.[9] The results of the inelastic neutron scattering show the almost localized nature of the first excited states as their small dispersion.[8] The ESR and recent inelastic neutron scattering experiments report small splitting of the first magnetic excitations.[7,10]

In this paper, first we see the features of the first excited states in the two-dimensional orthogonal dimer model and show that this model can explain the small dispersion. But the splitting of the first excited states observed by the precise measurements, for example by the ESR and neutron scattering measurements, is not explained. Therefore it is necessary to consider the effects of the Dzyaloshinsky-Moriya (DM) interaction. We estimate the DM interaction to explain the splitting of the first excited states and then discuss the magnetic field dependence of the first excited states. We also comment on the effects of the anisotropic exchange (AE) interaction.

§2. First Excited State

The magnetic properties of $SrCu_2(BO_3)_2$ are described rather well by the two-dimensional Hamiltonian:

$$\mathcal{H} = J \sum_{\text{n.n.}} \mathbf{s}_i \cdot \mathbf{s}_j + J' \sum_{\text{n.n.n.}} \mathbf{s}_i \cdot \mathbf{s}_j \ . \qquad (2.1)$$

The system is shown in Fig. 1. The dimer bonds whose coupling constant is J are coupled with the inter dimer bonds J'. To describe the total spin of the system we use $S_\alpha (\alpha = x, y, z)$. In this model the neighboring dimers are orthogonal and the orthogonality is an important feature for the almost localized character of the triplet excitations. That nature may be seen clearly by the perturbation theory, as shown in ref. 3. The hopping of the triplet excitations starts from the sixth-order in the perturbation.[5] In this order only the hoppings along unit vectors a are possible. The hoppings from an A-dimer (black dumbbell in Fig. 1) to a B-dimer (white one) becomes possible considering the higher order process in the perturbation.[11]

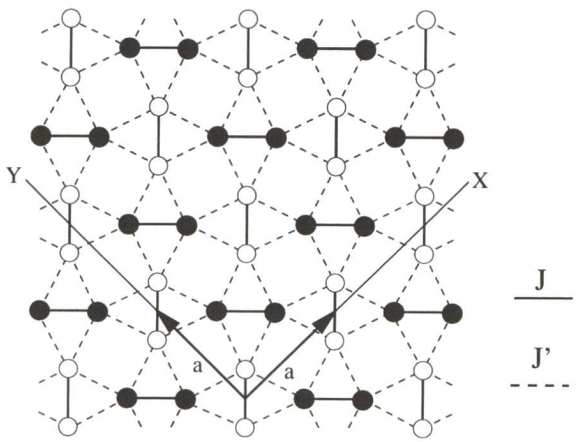

Fig. 1. The model for $SrCu_2(BO_3)_2$: Two-dimensional orthogonal dimer model. The solid lines represent the interaction J and the dashed lines J'. We call black dumbbells as A-dimers and white ones as B-dimers, for example. The unit vectors are shown by arrows and their length is a.

In the case of $SrCu_2(BO_3)_2$, the ratio $J'/J = 0.635$ is estimated.[12] To check the validity of the prediction for such a rather big J'/J, the dispersion is calculated by the exact diagonalization. The results are shown in Fig. 2. The number of the spins is 24. The almost flat dispersion is obtained and we conclude that the triplet excitations on $SrCu_2(BO_3)_2$ have the almost localized nature. As

shown in the inset in Fig. 2, the splitting of the excitations are observed at wave vectors at $(0,0)$ $((2\pi/a,0))$ and $(\pi/3a,0)$ $((2\pi/3a,0))$. The energy difference at $(0,0)$ ΔE_1 is $0.014J$. Assuming $J = 85$,[12] $\Delta E_1 \sim 1$K. On the other hand, only one mode is observed in the experiments, maybe due to poor resolution. The width of the instrumental resolution is typically a few Kelvin. The condition $J'/J = 0.635$ seems to reproduce the results of the neutron scattering experiments in ref. 8, if we consider a constant shift due to the difference of the spin gap determined from the thermodynamic measurements and the neutron scattering measurements.

It is observed by the recent neutron scattering measurements that the first excited states splits into the $Sz = 0$ mode (34 K) and two $Sz = \pm 1$ modes (31 K, 35 K).[10] The latter two modes are also observed by the ESR measurements at slightly different energies, 33 K and 36 K.[7] The energy difference of the two $Sz = \pm 1$ modes is about $4K$, which is lager than the estimated values by using the Heisenberg model. In addition to that, the splitting between $Sz = 0$ state and $Sz = 1$ state can not be explained in the Heisenberg model. Therefore we need consider other effects: the DM interactions proposed by Kakurai *et al.*[10] or the AE interaction of the J-bond proposed by Nojiri *et al.*[7]

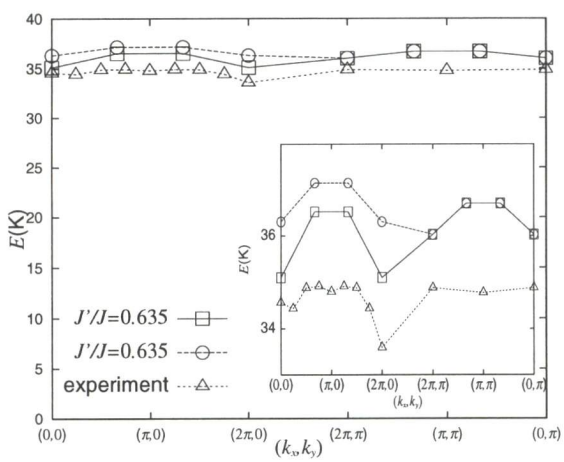

Fig. 2. The magnetic excitations for $SrCu_2(BO_3)_2$: $J'/J = 0.635$, $N_s = 24$ and $a = 1$. Inset is a magnification.

§3. The Splitting of the First Excited States

As discussed in the previous section, the splitting of the first excited states can not be explained by the Heisenberg model. Furthermore the ESR transition between the ground state and the excited triplet states is forbidden in the isotropic Heisenberg model. The ESR transition become possible for the system with the DM interaction, the AE interaction or non-equivalent g-tensors. Here we consider the effects of the DM interaction and the AE interaction.

3.1 Dzyaloshinsky-Moriya interaction

First we treat the DM interaction. Assuming both the A-dimers and B-dimers exist on the same plane, there is a reflection symmetry at the center of the J bonds and the DM interaction between the nearest neighbor sites vanishes. However concerning the J' bonds the DM interaction between the next nearest neighbor sites exists. The direction of DM interaction vectors is perpendicular to the ab plane, which is a mirror plane. Thus in this case the Hamiltonian is described as

$$\mathcal{H}_{\rm DM} = \sum_{\rm n.n.n.} D(s_i^x s_j^y - s_i^y s_j^x). \tag{3.1}$$

For a pair i, j, the direction from i to j is defined as in Fig. 3.

In fact at low temperatures (lower than 395K) there is no reflection symmetry concerning the ab plane in $SrCu_2(BO_3)_2$,[13] because the plane of the A-dimers is shifted slightly along the c axis from the plane of the B-dimers. But the magnitude of the shift is small and it is expected that the component discussed in eq.(3.1) is predominant and other component can be ignored approximately.

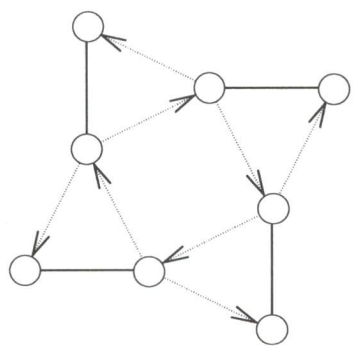

Fig. 3. The unit cell for $SrCu_2(BO_3)_2$. The arrows define the direction from i site to j site.

Assuming $J'/J = 0.635$,[12] we calculated the excited energy. The results are shown in Fig. 4. Here the number of the spins is 24 and the wave vector is fixed to $(0,0)$. There are two modes in the Heisenberg model. The effects of the DM interaction split each mode and there are four sates: two of them $Sz = 0$ and the others $Sz = \pm 1$. Inset in Fig. 4 shows the energy differences ΔE between the states with same S_z. From the experiments, the splitting ΔE for $Sz = 1$ is estimated to be about 4K. To satisfy that condition $D = (0.02 \pm 0.005)J$ is appropriate. In the following discussion we use $D = 0.02J$. The order of the DM interaction D may be estimated from the g value: $D \sim (g-2)/gJ' \sim 0.05J$. Therefore the order of the DM interaction $D = 0.02J$ is reasonable.

There are four modes for the fist excited state: 33.8 K (31 K) and 37.5 K (35 K) for $Sz = \pm 1$ and 35.1 K and 36.4 K (34 K) for $Sz = 0$, where the results of the neutron scattering measurements are shown in the round brackets. The energy differences of the state for $Sz = 0$ is about 1K and to observe the difference of the two modes is difficult by the neutron scattering measurements because of the instrumental resolution. The average values of the two states for $Sz = 0$ are 35.8K. The experimental values obtained by the ESR and the neutron scattering are consistent each other except for a constant shift.

Using the value $D = 0.02J$, the magnetic field dependence for the excitation energies is calculated. In the

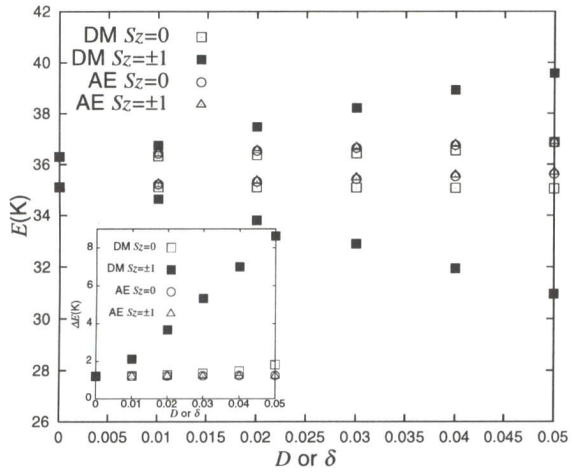

Fig. 4. The excite energy for $Sz = 0$ and $Sz = \pm 1$ with DM interaction and AE interaction. Inset shows the energy differences between the states with the same Sz. The x-coordinate represents D for DM interaction and δ for AE interaction.

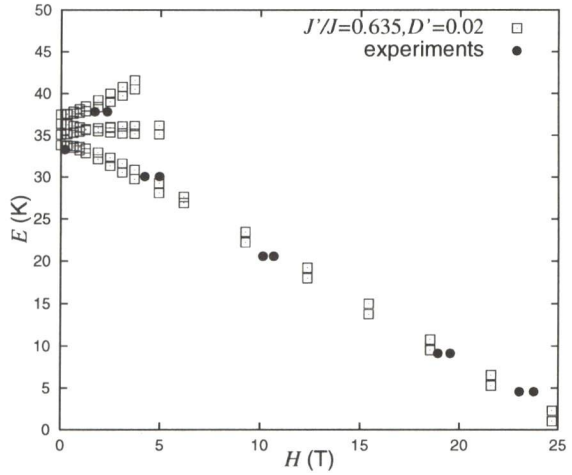

Fig. 5. The excited state energy in the magnetic fields perpendicular to the c axis. The results of the ESR experiments are also shown. Only the lowest two modes are shown in the high magnetic fields range.

magnetic fields along c axis H_z, S_z commutes with the Hamiltonian and the change of the energies concerning the magnetic fields simply is described as $-H_z S_z$. Therefore the energies for $S_z = \pm 1$ splits into two state: one for $S_z = 1$ and the other for $S_z = -1$. Those for $S_z = 0$ do not depend on the fields H_z. These results are consistent with the experiments.[7,10]

On the other hand, in the case of the magnetic fields perpendicular to the c axis H_x, S_x does not commute with Hamiltonian. Therefore the energies do not change linearly. The magnetic field dependence on the fields H_x is shown in Fig. 5. The energies of the lower $S_z = \pm 1$ states split into two modes: they decrease with H_x and those of the higher $S_z = \pm 1$ states also split and they increase with H_x. The energies of the $S_z = 0$ states depends on the magnetic fields only weakly. The results of ESR experiments are also shown in Fig. 5. As shown in the figure, the energies observed in the ESR experiments are for $S_z = \pm 1$ states in the zero fields. Because of the nonlinearity in the small magnetic fields, estimated energies fitted linearly in high magnetic field region are different from the energies with no magnetic fields.

3.2 Effects of the anisotropic exchange interaction

As pointed out in ref. 7, the anisotropic exchange Hamiltonian in a tetragonal crystal field may be written as,

$$\mathcal{H}_{AE} = J \sum_{\text{n.n.}} (s_i^x s_j^x + s_i^y s_j^y + (1+\delta)\, s_i^z s_j^z) + J' \sum_{\text{n.n.n.}} (s_i^x s_j^x + s_i^y s_j^y + (1+\delta')\, s_i^z s_j^z). \quad (3.2)$$

For simplicity we assume $\delta = \delta'$ in the following discussion.

In the same way as for the DM interactions we calculate the energy differences as a function of δ in this case. The order of the anisotropy δ is estimated by g value: $\delta \sim 0.01$ and we calculated the case from $\delta = 0.01$ to $\delta = 0.05$. As shown in Fig. 4 the change introduced by the anisotropy δ is much smaller than the experimental values. Therefore we conclude that the effects of the AE interaction can not explain the splitting of the first excite energies neither the energy difference between two $S_z = \pm 1$ states nor that between $S_z = 0$ and $S_z = \pm 1$.

§4. Conclusion

The splittings of the first excited states are explained by including the DM interaction, but not by the AE interaction. From the first excited state, we estimated the value of the DM interaction $D \sim 0.02J$. In the Raman scattering, intensity depends on the direction of the magnetic fields. This aspect will be discussed in a future publication.

Although the DM interaction is important to explain the splitting of the first excited state, to discuss thermodynamic properties of $SrCu_2(BO_3)_2$ the two-dimensional Heisenberg model may be sufficient except for discussing very small energy scales (a few Kelvin).

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