Magnetic Excitations in Strongly Correlated Electrons
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Thermodynamic Properties of Three-Dimensional Orthogonal Dimer Model for $SrCu_2(BO_3)_2$

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Effects of the inter-layer couplings for the orthogonal dimer system $SrCu_2(BO_3)_2$ are discussed. The spin-gap Δ of the three-dimensional model is independent of the inter-layer couplings when they are small. Therefore at low temperatures ($T < \Delta$) thermodynamic properties are described well by the two-dimensional model. On the other hand at high temperatures the mean-field type scaling ansatz is useful to discuss the magnetic susceptibility for week inter-layer couplings. From fit of the magnetic susceptibility, the estimated coupling constants are $J = 85$ K for the nearest-neighbor couplings, $J' = 54$ K for the next-nearest-neighbor couplings, and $J'' = 8$ K for the inter-layer couplings. These parameters are consistent with the temperature dependence of the specific heat at low temperatures.

KEYWORDS: $SrCu_2(BO_3)_2$, uniform susceptibility,spin-gap, specific heat, inter-layer coupling, orthogonal dimer state, Shastry-Sutherland model

§1. Introduction

The new spin-gap system $SrCu_2(BO_3)_2$, which was found by Kageyama *et al.*,[1] is a beautiful realization of the two-dimensional Shastry-Sutherland model (Fig. 1), which was studied by them almost twenty years ago.[2] The original model seems to be very artificial. In fact, it was constructed in such a way to realize an exact ground state on a two-dimensional model. The system for $SrCu_2(BO_3)_2$ is shown in Fig. 2(a), which is described as the orthogonal dimer model.[1,3] It is topologically equivalent to the two-dimensional Shastry-Sutherland model, where the nearest-neighbor bond in the former model corresponds to the next-nearest-neighbor bond of the latter and vice versa. The orthogonal dimer model shows a quantum phase transition from the gapful phase to the gapless phase as the next-nearest-neighbor interaction is increased.[4] In the gapful phase the ground state is the exact dimer ground state.

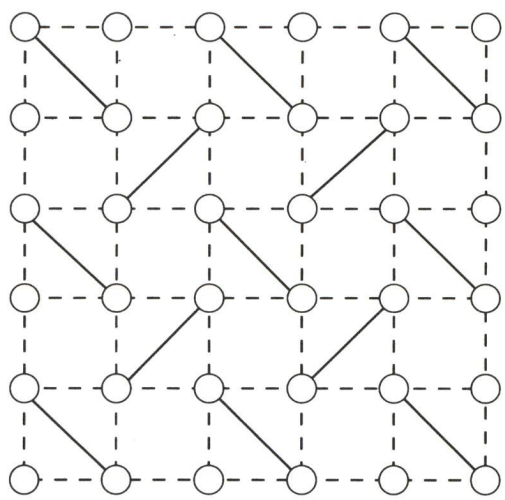

Fig. 1. The model discussed by Shastry and Sutherland, which has the dimer singlet ground state.

$SrCu_2(BO_3)_2$ consists of the layers of $CuBO_3$ and the layers of Sr. In the compounds Cu^{2+} ions occupy crystallographically equivalent sites and described by a spin-1/2 Heisenberg model. As the first approximation the compound may be treated as a two-dimensional system and the novel features of the $SrCu_2(BO_3)_2$, for example the magnetization plateaus, are explained by this model. In fact the 1/3-plateau predicted by theory[5–7] was observed in the recent experiment in high magnetic fields.[8] The coupling constants were estimated from the spin-gap and the Curie-Weiss constant. According to the estimation, $SrCu_2(BO_3)_2$ is regarded as a material which is near the quantum phase transition point.[3] Originally, the spin-gap observed in the powder sample 30 K was used. While, more recent experiments using the single crystal samples show that the gap is close to 35 K.[9–11]

In this paper we will study the thermodynamic properties of $SrCu_2(BO_3)_2$ in more detail and reestimate the coupling constants for this material taking also the inter layer couplings into consideration. First, one can show that the ground state and the first excited state are independent of the interlayer couplings. For that reason the properties of $SrCu_2(BO_3)_2$ at low temperatures can be described well by the two-dimensional model. Therefore we discuss the temperature dependence of the magnetic susceptibility based on the two-dimensional model at low temperatures and determine the coupling constants in the $CuBO_3$ plane. Then we show that these parameters are consistent with the specific heat. From the susceptibility at high temperatures the inter-layer couplings may be estimated.

§2. Ground state and first excited state

2.1 Two-dimensional orthogonal dimer model

As shown in our previous analysis[3] the magnetic properties of $SrCu_2(BO_3)_2$ are described rather well by the two-dimensional Hamiltonian:

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

The system is shown in Fig. 2 (a). The model can be considered as a coupled dimer model. The dimers, where two spins are coupled with the nearest-neighbor coupling J, are connected by the next-nearest-neighbor bond J'. An elementary unit for the interaction between a pair of the dimer bonds is shown in Fig. 2 (b). It is convenient to use the dimer bases defined for each nearest-neighbor bond:

$$|s\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle - |\downarrow\uparrow\rangle), \tag{2.2}$$

$$|t_1\rangle = |\uparrow\uparrow\rangle, \tag{2.3}$$

$$|t_0\rangle = \frac{1}{\sqrt{2}}(|\uparrow\downarrow\rangle + |\downarrow\uparrow\rangle), \tag{2.4}$$

$$|t_{-1}\rangle = |\downarrow\downarrow\rangle. \tag{2.5}$$

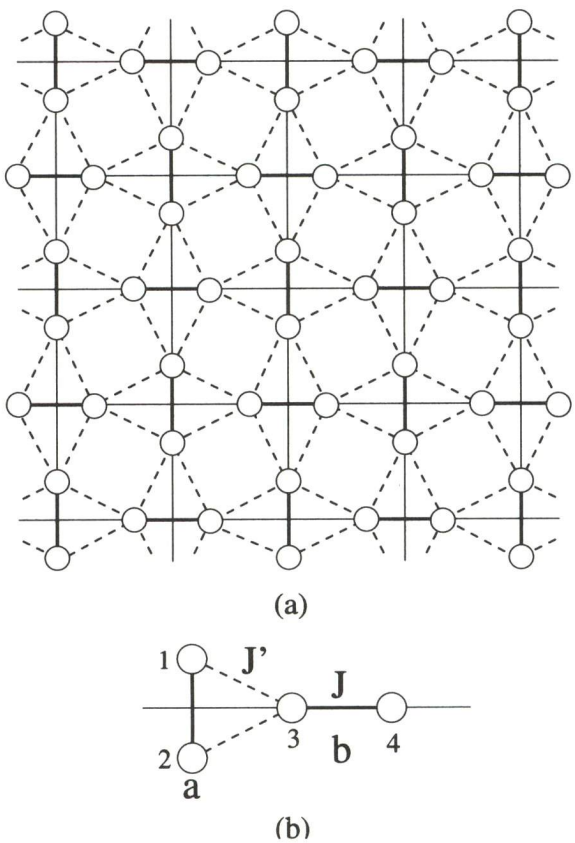

Fig. 2. (a)The model for $SrCu_2(BO_3)_2$: Two-dimensional orthogonal dimer model. (b)A configuration of two dimers which are orthogonal.

The direct product of the singlets on dimers defined by

$$|\Psi\rangle = \prod_a |s\rangle_a \tag{2.6}$$

is an exact eigenstate of the Hamiltonian (2.1).[2,3] Here the index a denotes each dimer bonds and runs over all dimer bonds. For coupling constants $J'/J < 0.69$ this eigenstate is the ground state.[3,12,13] This model shows a quantum phase transition at $J'/J = 0.69$ from the dimer singlet state to the Néel ordered state which is gapless.[4] Note that in the limit of $J'/J \to \infty$, the present model reduces to the square lattice Heisenberg model, whose coupling constant is J'.

The singlet dimer ground state has a spin-gap, which can be estimated by the perturbation theory. The spin-gap up to the fourth-order is given by

$$\frac{\Delta}{J} = 1 - (\frac{J'}{J})^2 - \frac{1}{2}(\frac{J'}{J})^3 - \frac{1}{8}(\frac{J'}{J})^4, \tag{2.7}$$

and the result up to the fifteenth-order is given in Ref. 12. The spin-gap for finite systems is shown in Fig. 3, where the number of the spins is 16, 20, and 24 with periodic boundary conditions. The finite size effects for $J'/J \lesssim 0.66$ are small. The results of eq. (2.7) and the numerical results for the finite size systems agree well for $J'/J < 0.5$.

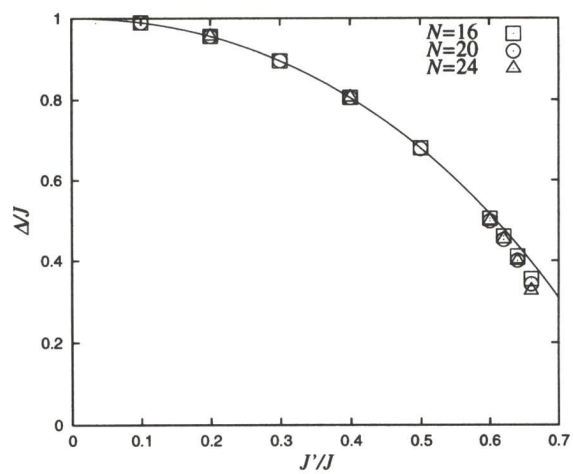

Fig. 3. Spin-gap for finite lattices: $N_s = 16$, 20, and 24 from the dimer singlet ground state. The solid line is the perturbation result up to the fourth order.

The perturbation theory predicts a novel character for the triplet excitation. The triplet excitation is completely localized up to the fifth-order, which leads to crystallization of the triplet excitations at certain magnetizations. At magnetizations where the crystallization occurs, the magnetization plateaus appear.[3,5-7] This feature of the triplet excitations can be understood from the matrix elements for one triplet excitation.

$$J'(\mathbf{s}_1 + \mathbf{s}_2) \cdot \mathbf{s}_3 |t_1\rangle_a |s\rangle_b = \frac{J'}{2}|t_1\rangle_a |t_0\rangle_b - \frac{J'}{2}|t_0\rangle_a |t_1\rangle_b \ , \tag{2.8}$$

and

$$J'(\mathbf{s}_1+\mathbf{s}_2)\cdot\mathbf{s}_3|s\rangle_a|t_m\rangle_b=0 \quad (m=0,\pm1)\ , \qquad (2.9)$$

where the site indices are specified in Fig. 2(b). Equation (2.8) means that when a triplet moves to one of neighboring bonds it leaves another triplet behind. From the symmetry reason, the parity with the reflection, the matrix element shown in eq. (2.9) vanishes. Equations. (2.8) and (2.9) make a hopping of a triplet excitation rather difficult. It becomes possible through a closed path of dimer bonds and thus the hopping processes start from the sixth-order in the perturbation. Recently this almost localized nature of the dispersion is directly observed in the neutron scattering experiment.[11]

2.2 Effects of the inter-layer couplings

As we noted before, $SrCu_2(BO_3)_2$ consists of $CuBO_3$ layers and Sr layers. The $CuBO_3$ layers stack alternately as is shown in Fig. 4(a). Along the c-axis dimers are coupled with inter-layer coupling J'' as shown in Fig. 4(b). It is obvious that the matrix element for the singlet dimers vanishes:

$$J''(\mathbf{s}_1+\mathbf{s}_2)\cdot(\mathbf{s}_3+\mathbf{s}_4)|s\rangle_a|s\rangle_b=0\ . \qquad (2.10)$$

Therefore the three-dimensional model for $SrCu_2(BO_3)_2$ has the exact orthogonal dimer ground state for small J' and J''.[14]

Next, let us consider the case where a triplet is excited. The matrix element from this state also vanishes:

$$J''(\mathbf{s}_1+\mathbf{s}_2)\cdot(\mathbf{s}_3+\mathbf{s}_4)|t_m\rangle_a|s\rangle_b=0 \qquad (m=0,\pm1). \qquad (2.11)$$

It is obvious that the triplet excitation cannot move along the c-axis at low temperatures. If the neighboring planes are filled with the dimer singlet states, the triplet excitation on a plane is completely confined in that plane. Thus the magnitude of the spin-gap for the three-dimensional model is the same as the two-dimensional one. The dispersion for the triplet excitation is not modified, either.

The inter-layer coupling J'' does not affect the spin-gap. It means that the properties of $SrCu_2(BO_3)_2$ can be described by using the two-dimensional orthogonal dimer model at low temperatures: $T<\Delta$, even if the inter-layer couplings exist. Notice that the theoretical predictions discussed so far[3,5-7] are hardly changed at low temperatures by the inclusion of finite inter-layer couplings, which are smaller than J'.

§3. Thermodynamic properties of $SrCu_2(BO_3)_2$

3.1 Magnetic susceptibility

Here we discuss the temperature dependence of the magnetic susceptibility of $SrCu_2(BO_3)_2$ and determine an optimal set of the parameters J, J', and J''.

Let us start from low temperatures: $T<\Delta$. The magnetic susceptibility at $T<\Delta$ may be described well by the susceptibility of the two-dimensional model χ_{2d}. Therefore we calculate χ_{2d} with the transfer matrix method and compare the results with experimental ones at $T<30$ K. The results are shown in Fig. 5. The results for

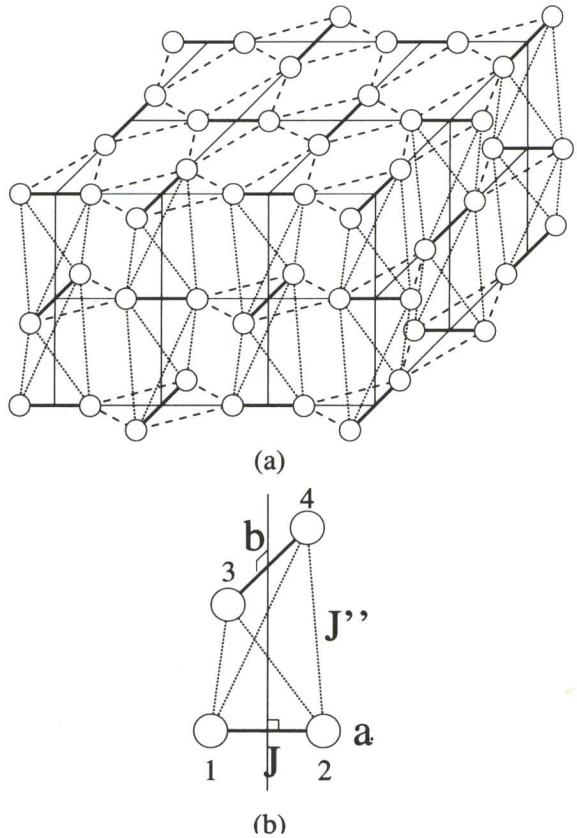

Fig. 4. (a) The model for $SrCu_2(BO_3)_2$: Three-dimensional orthogonal dimer model. (b) The configurations of two dimers along the c-axis are also orthogonal in a different way from Fig. 2(b).

$J'/J=0.62, 0.64, 0.66$ are shown by the filled symbols. The spin-gap $\Delta\sim35$ K is obtained by various experiments,[9-11] and J is determined so that the spin-gap obtained from the exact diagonalization for $N_s=24$ is 35 K: $J=72$ K for $J'/J=0.62$, $J=88$ K for $J'/J=0.64$, and $J=104$ K for $J'/J=0.66$. Here we note that if we determine the parameters with which both the Curie-Weiss constant and the spin-gap is satisfied by the two-dimensional model, we get $J'/J=0.66$ and $J=104$ K. The system size $N_s=16$ is used for the calculations at finite temperatures. In the present system the finite size effects is not so important because the triplet excitation is almost localize. In Fig. 5, to show finite size effects, the results for $N_s=16$ and 20 are shown with open symbols for $J'/J=0.635$, which is considered to be an optimal choice. The finite size effects are observed but small. Considering the finite size effects, we may conclude that the parameters $J'/J=0.635\pm0.01$ explain the experiment well. In the following, we use the parameters $J'/J=0.635$ and $J=85$K as the optimal set for $SrCu_2(BO_3)_2$.

Now we consider the magnetic susceptibility at temperatures $T>\Delta$. As is shown in Fig. 7, the calculated susceptibility for $J'/J=0.635$ and $J=85$K, which agrees with experiments well at $T<\Delta$, does not fit to the experiment in the temperature range: $20<T<350$. The system size effects at the temperatures $T>50$ K is so small that the results for $N=20$ are considered as the

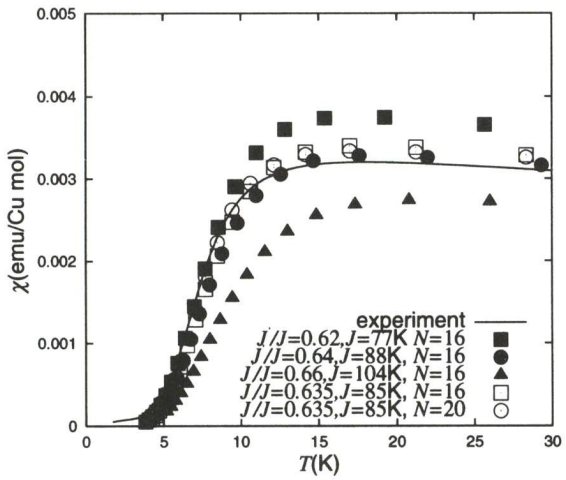

Fig. 5. The temperature dependence of the magnetic susceptibility on the two-dimensional model. $J'/J = 0.62$, $J'/J = 0.64$, $J'/J = 0.635$, and $J'/J = 0.66$.

bulk limit for such temperature range. This fact shows that the effects of the inter-layer couplings J'' on the susceptibility cannot be ignored in such a temperature range. To estimate the magnitude of J'', we follow the mean-field type scaling ansatz used in Ref. 15:

$$\chi(J'/J, J''/J) = \frac{\chi_{2d}(J'/J)}{1 + 4J''\chi_{2d}(J'/J)}. \qquad (3.1)$$

The coefficient $4J''$ in the denominator reproduces the correct high-temperature Curie-Weiss constant of the three-dimensional model. At low temperatures this ansatz gives the same spin-gap as that given by the two-dimensional model, which is reasonable because the spin-gap is not modified by a small J''.

To check the quality of eq. (3.1) we have calculated the temperature dependence of the susceptibility using a bilayer model, where there are 8 spins in each layer. We assume the periodic boundary conditions. The used parameters are $J'/J = 0.4$ for the in-plane couplings and $J''/J = 0.05$, 0.1, 0.15 for the inter-layer couplings. The results are shown in Fig. 6(a). For this system, the spin-gap is J because of the periodic boundary condition. We select small J''/J so that inter-layer couplings do not change the magnitude of the spin-gap and the ground state. The ground state and the first excited state for these systems are checked by the exact diagonalization calculations. We scale the obtained susceptibility onto the effective two-dimensional one $\tilde{\chi}_{2d}$ through the inverse relation:

$$\tilde{\chi}_{2d}(J'/J, J''/J)) = \frac{\chi_{3d}(J'/J, J''/J)}{1 - 4J''\chi_{3d}(J'/J, J''/J)}. \qquad (3.2)$$

The scaling plots are shown in Fig 6(b). The results form approximately a single line, supporting the mean-field type ansatz.

Next, using eq. (3.1), we estimate the magnitude of the inter-layer coupling J''. Here we use $\chi_{2d}(J'/J = 0.635)$ with $J = 85$ K because it agrees with the experiments well at low temperatures. We fit the susceptibility data

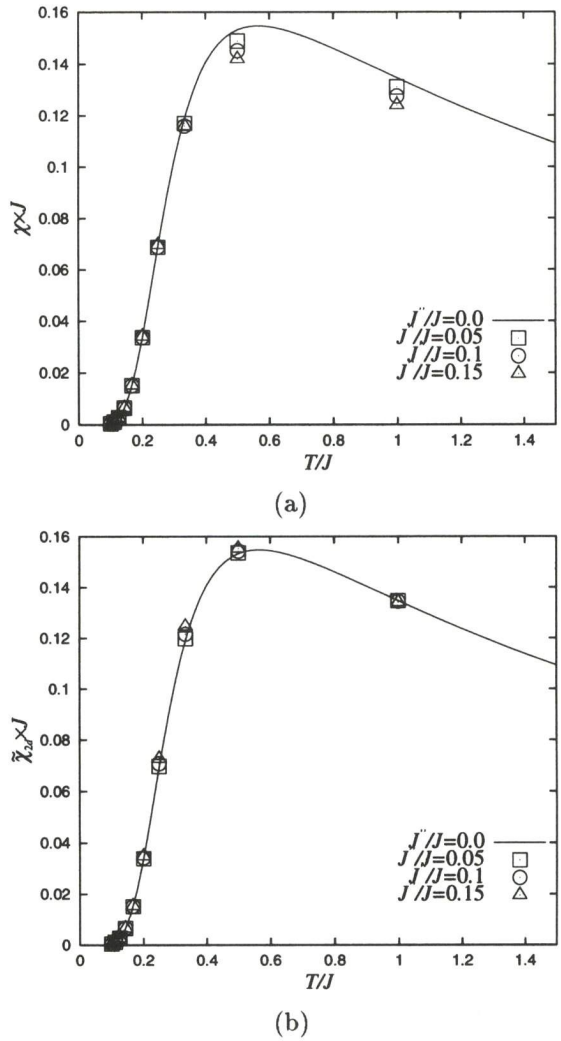

Fig. 6. (a) The temperature dependence of the magnetic susceptibility of the bilayer model. $J'/J = 0.4$ and $J''/J = 0.05$, $J'/J = 0.1$, and $J'/J = 0.15$. The solid line is the results of the two-dimensional model. (b) Scaling plot of the mean-field type ansatz.

in the range 100 K $< T <$ 350 K to eq. (3.1) and determine J''. Good agreement is obtained for $J''/J = 0.09$. This fit is shown in Fig. 7: $J = 85$ K, $J' = 54$ K, and $J'' = 8$ K. The inter-layer coupling J'' is about 10% of the intra-dimer coupling J. The inter-layer coupling is not important at low temperatures but is important to fix the energy scale of the coupling constants. One of the origin of the rather big Curie-Weiss constant $\sim$ 83 K may be attributed to the existence of the inter-layer couplings.

3.2 Specific heat

Next we discuss the specific heat. The temperature dependence of the specific heat of $SrCu_2(BO_3)_2$ at low temperatures $T < \Delta$ may be also explained well by using the two-dimensional model. The specific heat is calculated by the transfer matrix methods again. We compare the theoretical results for $J'/J = 0.635$ and $J = 85$ K, which explained the temperature dependence of the sus-

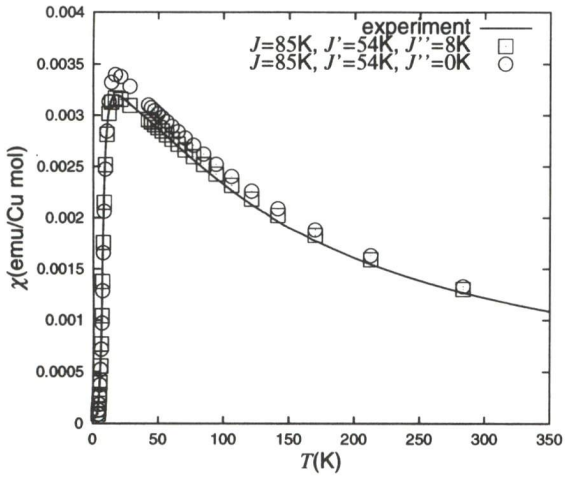

Fig. 7. Fit of the uniform magnetic susceptibility for $SrCu_2(BO_3)_2$. The parameters used are $J = 85$ K, $J' = 54$ K, and $J'' = 8$ K.

ceptibility well, with the experiments.[16] Here $\beta \times T^3$ is used as the specific heat for the phonon term. $\beta = 0.4$ mJ/K^4 is used. The results are shown in Fig. 8. We see that the specific heat for $J'/J = 0.635$ and $J = 85$ K agrees well with the experiments and the difference shown around the peak may be explained by the finite size effects. In Fig. 8 the specific heat for $J'/J = 0.62$, $J'/J = 0.64$ and $J'/J = 0.66$ are also shown for comparison.

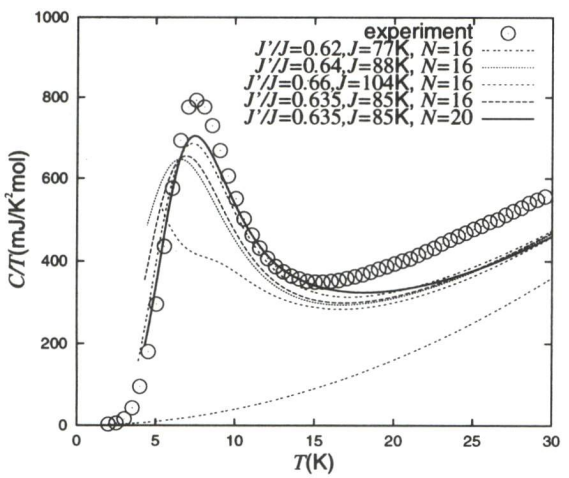

Fig. 8. The temperature dependence of the specific heat of $SrCu_2(BO_3)_2$. The fit parameters are $J = 85$ K, $J' = 54$ K. $0.4 \times T^3$ is used as the contribution from phonon degrees of freedom.

At temperatures $T \gtrsim 15$ K good quality of the fits is not obtained and the differences cannot be attributed only to the system size effects. Possible reasons of this difference may be the followings. First, in this temperature range the simple $\beta \times T^3$ expression is not sufficient for the lattice contribution. Second, the effects of the spin-phonon coupling may be important. In the orthogonal dimer model, the small kinetic energies of the excited triplets originate from the geometrical constraint. When the orthogonality is broken by some distortion of dimer bonds, a finite matrix element for a hopping of a triplet arises. Therefore a triplet excitation is expected to be a strongly coupled with phonons. Effects of the spin-phonon couplings will play an important role to understand the behavior of the specific heat, which is an interesting future problem.

§4. Conclusion

In the present study we have discussed thermodynamic properties of $SrCu_2(BO_3)_2$ with the three-dimensional orthogonal dimer model. At low temperatures this three-dimensional model is effectively equivalent to the two-dimensional model because the spin-gap is not affected by the inter-layer coupling J''. Therefore $SrCu_2(BO_3)_2$ can be described well by using the two-dimensional model as the first approximation, not because of the weakness of the inter-layer coupling J'' but by the geometrical reason.

Even if many triplet excitations exist in one plane, the triplets move only in the plane in spite of the sizable inter-layer couplings if the dimer bonds on the neighboring planes are occupied by the singlets. Therefore the two-dimensional spin dynamics are expected in $SrCu_2(BO_3)_2$ even if there are sizable inter-layer couplings. In fact unusual diffusive spin dynamics of two-dimensional character is observed by the NMR measurements.[17]

Concerning the coupling constant: J, J', and J'', the best fit is given by $J = 85$ K, $J' = 54$ K, and $J'' = 8$ K. In the real system the horizontal and vertical bonds in each plane are shifted slightly, which means that the one type of dimers, for example horizontal dimers, are on a plane and the other dimers, vertical dimers, are on the other plane. Therefore the inter-layer couplings along the c-axis consist of the two coupling constants. Here we treat J'' by the mean-field type approximation so that the average of the two slightly different coupling constants just corresponds to J''. The magnitude of each coupling constant can not be determined separately from the present analysis.

Acknowledgments

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