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Quantum phase transitions in the orthogonal dimer system $SrCu_2(BO_3)_2$

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Abstract

Inspired by recent enthusiastic theoretical works on the phase diagram of the (generalized) Shastry–Sutherland model, we measured the temperature dependence of magnetic susceptibility of $SrCu_2(BO_3)_2$ at high pressures up to 7 kbar. We employed a clumping type of a pressure cell made by CuTi alloys, which allows us to obtain the sample magnetization with a high sensitivity. We found that the temperature at maximum susceptibility decreases with increasing pressure, implying the reduction of the spin gap. This result strongly suggests that the system nears the phase boundary to an antiferromagnetically ordered or a plaquette singlet state.

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Keywords: $SrCu_2(BO_3)_2$; Shastry–Sutherland model; Spin gap; High pressure

1. Introduction

Over the last three decades, geometrical spin frustration has been one of the most interesting and challenging issues in the condensed matter physics. Three spins sitting on the corners of one rigid triangle, coupled antiferromagnetically with respect to each bond, is the most fundamental unit for conventional frustrated models. By arranging rigid triangles in some space, one can tailor various lattices with geometrical frustration, as exemplified in Fig. 1. Figs. 1(a) and (b) show the one-dimensional (1D) cases, a delta chain [1] and a zigzag chain [2], respectively. If the rigid triangles are packed completely on a plane, one obtains a 2D triangular lattice as shown in Fig. 1(c) [3], while one quarter of periodic vacancies in the triangular lattice results in the so-called kagomé lattice (Fig. 1(d)) [4]. Shown in Fig. 1(e) is one of the 3D versions, called the pyrochlore lattice, consisting of the alternate stack of the dilute triangular and dense kagomé layers along the cubic [1 1 1] [5].

In contrast, antiferromagnetically coupled spins on a 2D square lattice are not at all frustrated because of the absence of such triangles. Nevertheless, geometrical frustration can be induced into this lattice by introducing some regular depletion (Fig. 2(a)) [6] or by adding some diagonal interactions (Figs. 2(b)–(d)) [7–9]. Compared with the former models based on the rigid triangles, only little attention had been paid for the latter models so far presumably due to the lack of experimental correspondences. But recent discovery of materials such as CaV_4O_9 [10], Li_2VOSiO_4 [11], $SrCu_2(BO_3)_2$ [12] sparked experimental and theoretical investigations on the respective models of square lattice origin.

The Shastry–Sutherland lattice with the intradimer (J) and interdimer (J') bonds is characterized by the orthogonality of the neighboring dimers. Because of this peculiar topology, many intriguing phenomena have been found for the Shastry–Sutherland model and the corresponding material $SrCu_2(BO_3)_2$, including an exact

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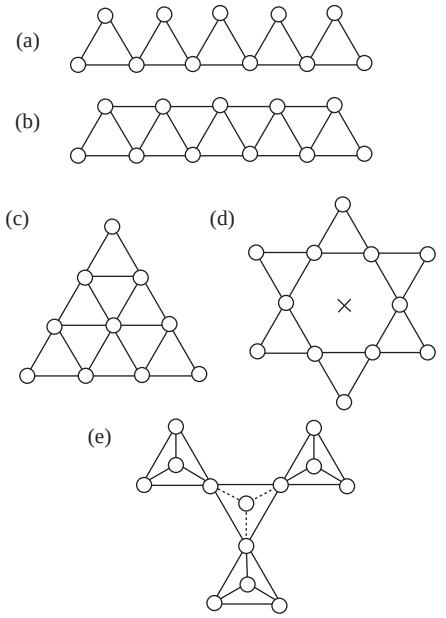

Fig. 1. Geometrically spin frustrated models consisting only of rigid triangles. (a) delta chain, (b) zigzag chain, (c) triangular lattice, (d) $\frac{1}{4}$ regularly depleted triangular lattice (kagomé lattice), (e) pyrochlore lattice.

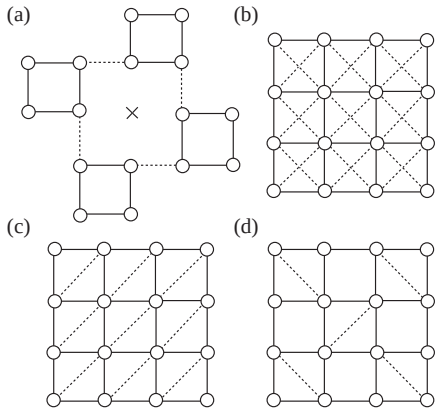

Fig. 2. Geometrically frustrated models derived from the 2D square lattice. (a) $\frac{1}{5}$ regularly depleted square lattice realized by CaV_4O_9, (b) J_1-J_2 model realized by Li_2VOSiO_4, (c) tilted triangular lattice, (d) Shastry–Sutherland lattice realized by $SrCu_2(BO_3)_2$.

dimer ground state [9,12–15], fractional plateaux in the magnetization curve [12,16–19], and multiple triplets excitations [20–23]. Theoretically, the possibility of quantum phase transitions from the dimer singlet state to other states has been discussed intensively [9,15,21,24–26]. For instance, Koga considered the generalized Shastry–Sutherland model adding the coupling between the layers (J''). As illustrated in Fig. 3,

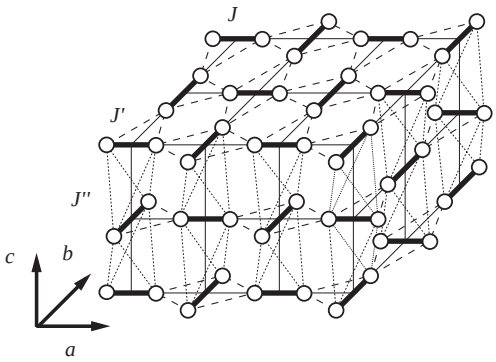

Fig. 3. The 3D lattice of $SrCu_2(BO_3)_2$.

neighboring dimers along the c-axis are also connected orthogonally, thus making the grand state of the 3D model still solvable exactly [27]. Considering the estimated coupling constants ($J=84$ K, $J'=54$ K, $J''=8$ K [28]; $J=70$ K, $J'=42$ K, $J''=15$ K [21]), this material must be located in the vicinity of the phase boundaries to an antiferromagnetically ordered state as well as a resonating plaquette singlet state.

In spite of these vigorous theoretical activities, however, there has been no experimental observation of the quantum phase transitions. Although the solid solution $Sr_{1-x}A_xCu_2(BO_3)_2$ ($A=$ Ca, Ba; $0<x<0.3$) was prepared to investigate the chemical pressure effect, the obtained susceptibility did not exhibit systematic change with x and A due to marked difference of sample quality [29]. This makes it difficult to estimate the exchange couplings reliably in this solid solution. On the other hand, application of physical pressure is considered to be a clean tool to tune exchange couplings. In this paper, we will report on the susceptibility measurement at various pressures up to 7 kbar.

2. Experimental

We recently developed a high-pressure clamp cell consisting of CuTi alloys with 3 wt% Ti [30]. The magnetization of the CuTi pressure cell is extremely small, namely, of the order of 10^{-7} emu. Furthermore, it is almost independent of temperature (T) and pressure (P). These characteristics are based on the fact that the paramagnetic susceptibility of Ti almost compensates the diamagnetic susceptibility of Cu. Therefore, we can precisely measure even weak magnetizations of materials like the present compound. Sample magnetizations were measured under pressure up to 7 kbar using the CuTi cell with the outer diameter of 8.8 mm, which was installed in a SQUID magnetometer. Single crystals of $SrCu_2(BO_3)_2$ of high quality were grown by the travelling solvent floating zone method using a solvent

$LiBO_2$ [31]. One piece of grown crystals with an appropriate dimensions was compressed in a Teflon capsule 3 mm in diameter and 10 mm in length, filled with a liquid pressure-medium, Fluorinert (CF 70: CF77 = 1:1). The magnetic field (H) of 1 T was applied perpendicular to the c-axis and temperature was scanned between 3 and 30 K.

3. Results and discussion

For magnetization measurements under high pressures, the pressure cell made by Co-contaminated CuBe alloys has been usually employed [32]. However, this pressure cell itself has finite magnetization because of the Co impurities in the commercially available alloys. To make matters worse, the magnetization of the pressure cell varies as a function of both P and T. The susceptibility measurement for $SrCu_2(BO_3)_2$ using this type of pressure cell was previously performed between $5<T<30$ K [33]. As anticipated, the total magnetization ($SrCu_2(BO_3)_2$ + pressure cell) even at ambient pressure does not approach to zero as $T\to 0$. Moreover, at a given T, it remarkably increases with P.

The use of the new type of pressure cell has dramatically changed the situation into better. Fig. 4 demonstrates the result of temperature dependence of the magnetic susceptibilities for $3<T<30$ K at 0, 2.0, 4.0, 6.0, and 7.0 kbar, using the CuTi pressure cell. At higher temperatures, the obtained susceptibilities are almost independent of pressure, significantly in contrast to the result of previous magnetization as mentioned above. With decreasing T, however, marked pressure dependence appears. The temperature at the maximum susceptibility T_{Max} and the temperature at the half-maximum susceptibility T_{HM} decrease with the application of pressure. With further decreasing T, all the susceptibilities decrease to zero as $T\to 0$. This means that the ground state remains spin singlet at any pressure applied in this study. It should be pointed out that the plaquette singlet state also has nonmagnetic ground state. However, we believe that the dimer singlet phase still survives at least until 7 kbar since the susceptibility changes quite systematically with P.

We plotted T_{Max} and T_{HM} as a function of P in Fig. 5. Both values decrease in proportion to P, which would be attributable to the reduction of the energy gap Δ from the singlet ground state to the first (triplet) excited state. It is noted, however, that in spin-gap materials it is in principle possible to evaluate the value of Δ by fitting the susceptibility to a function assuming an activation-type of T dependence. In $SrCu_2(BO_3)_2$, however, such a fitting using the data even without a pressure cell (i.e., at the ambient pressure) gave much smaller value of 19 K [12] than the actual value ($\Delta = 34$ K) obtained by e.g., ESR [13]. Therefore, the pressure variation of Δ cannot be estimated from the present experiment. Even then, as seen in Fig. 5, extrapolation of T_{Max} and T_{HM} to higher P may predict the critical pressure P_c to be $25\sim 35$ kbar. Possibly, P_c can be smaller because the transitions either to the ordered state or to the plaquette singlet state are considered to be of first order [25]. This result has further motivated us to investigate the pressure effect. Actually, various experiments at high(er) pressure (magnetization, nuclear magnetic resonance, neutron

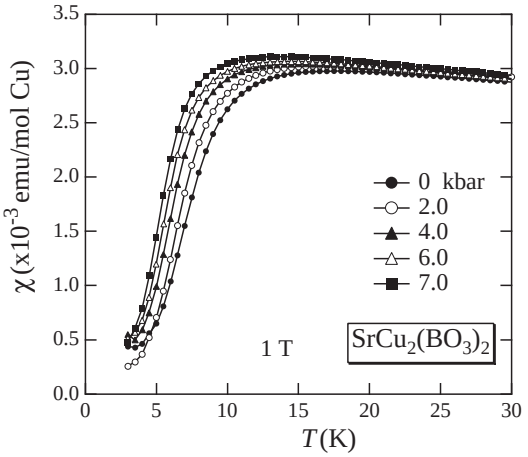

Fig. 4. Temperature dependence of the magnetic susceptibility of $SrCu_2(BO_3)_2$ at high pressures up to 7.0 kbar.

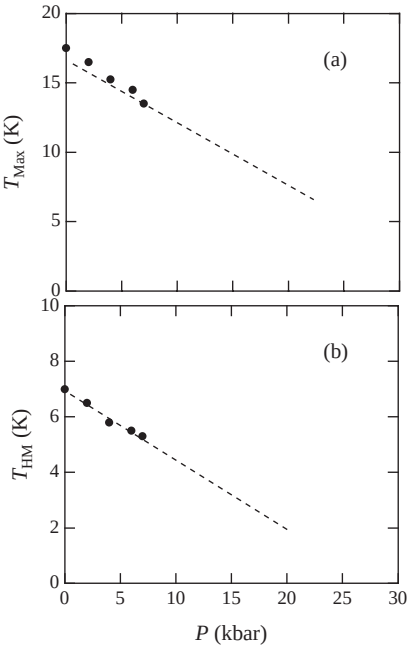

Fig. 5. Pressure dependence of (a) T_{Max} and (b) T_{HM}. Broken lines are the guides to the eyes.

scattering/diffraction, Raman scattering, specific heat) are in progress.

Finally, we would like to discuss the origin of the pressure effect. Since the structure of $SrCu_2(BO_3)_2$ consists of the $CuBO_3$ (Shastry–Sutherland) layers, intercalated by nonmagnetic Sr^{2+} ions, the application of P would affect only interlayer distance, thus changing J'' while keeping J and J' unchanged. However, this is not the case. Theoretical considerations show that Δ is independent of J'' for a given set of J'/J, while it largely depends on J'/J [15,25]. In fact, the theoretical susceptibility as a function of J'/J is quite similar to the experimental susceptibility as a function of P [15]. Considering these facts, it is natural to think that with increasing P both interlayer and intralayer coupling constants change toward the direction of the phase boundary.

4. Conclusion

We performed the magnetic susceptibility experiments for $SrCu_2(BO_3)_2$ at high pressures using a new type of pressure cell made by CuTi alloys with almost non-magnetic susceptibility. Although the pressure in our hand is not enough to induce the quantum phase transition, we obtain a strong piece of evidence that the system gets closer to the phase boundary. Higher P experiments will be conducted soon.

Acknowledgements

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