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Specific heat study of $SrCu_2(BO_3)_2$

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Abstract

The specific heat measurement of a two-dimensional spin-gap material $SrCu_2(BO_3)_2$ with an exact dimer ground state was performed in various magnetic fields up to 12 T. Based on an isolated dimer model, we estimated the spin-gap size at zero field to be 35.0 K. The value of the spin gap decreases in proportion to the applied magnetic field, due to the Zeeman splitting of the excited triplet levels.

Keywords: $SrCu_2(BO_3)_2$; Exactly solvable model; Specific heat; Spin gap

Our experimental investigations on $SrCu_2(BO_3)_2$ with a spin-1/2 two-dimensional orthogonal dimer lattice [1–3] have established that this material is the very realization of the Shastry–Sutherland model with an exact dimer ground state [4,5]. From various measurements such as magnetic susceptibility and electron spin resonance (ESR), we estimated the value of spin gap to be about 34 K. Of equal importance is a rather strong interdimer interaction J' (comparable with an intradimer one J), which brings a spin frustration into the system [1]. In fact, J' and J were theoretically determined to be 68 and 100 K, respectively [4]. Interestingly, the ratio $J'/J = 0.68$ is just below the critical boundary, $(J'/J)_c = 0.70$, beyond which the system falls onto the Néel-ordered state [4]. Concerning magnetic excitations, quantized plateaus such as $\frac{1}{4}$ and $\frac{1}{8}$ of the Cu full moment were observed in the magnetization curves [1], which originate from considerable constraint upon a hopping of a single triplet [4].

In the present paper, we measured the magnetic field H dependence of the specific heat C of $SrCu_2(BO_3)_2$ to further understand both the ground and excited states, and obtained data were analyzed in terms of an isolated dimer model. The specific heat was measured by a heat-relaxation method in a temperature T range between 1.3 and 25 K in magnetic fields up to 12 T. A bulk single crystal of $SrCu_2(BO_3)_2$ was used, which was grown by the travelling solvent floating zone method [6]. The magnetic fields were applied perpendicular to the *ab* plane, i.e., the orthogonal dimer lattice.

A total specific heat divided by T, C/T measured at $H = 0$, 6, 9, 12 T is plotted against T in Fig. 1. For any magnetic field, C/T rises with decreasing T from 15 K, reaches a round maximum, and then falls rapidly toward naught. The round maximum, namely, the so-called Schottky anomaly is typical of spin-gapped system. A gradual increase in C/T with rising T above 15 K comes from the phonon term, which is, in general, known to vary as $C \propto T^3$. A prominent characteristic is that, with increasing H, a peak of C/T shifts to lower temperature: the temperatures where C/T reaches a maximum for $H = 0$, 6, 9, 12 T are, respectively, 7.5, 7.3, 6.9, 6.8 K. This behavior is related to a reduction in the spin-gap size $\Delta(H)$ with H, which will be confirmed later.

Because of a lack of an appropriate theory to describe the present experimental data, let us analyze the data only in lower-T region well below the spin gap utilizing the isolated dimer model, where J' is neglected and only J is taken into consideration [7]. In this simplest model, the magnetic contribution of the specific heat in a low-T limit is expressed as $C(H) \propto T^{-2} \exp(-\Delta(H)/T)$. Thus, CT^2 is plotted against $1/T$ in a logarithmic scale as shown in Fig. 2. One can see that all data well satisfy

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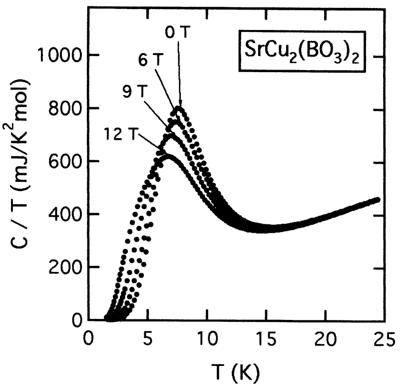

Fig. 1. C/T versus T measured at $H = 0$, 6, 9, and 12 T.

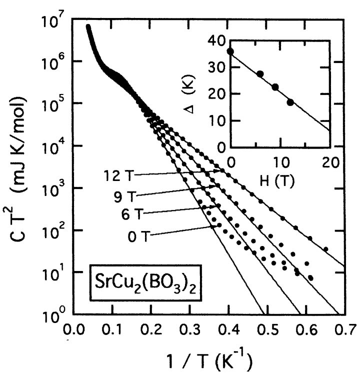

Fig. 2. Logarithmic plot of CT^2 as a function of $1/T$. Solid lines denote the calculations based on the isolated dimer model. Inset: magnetic field variation of $\Delta(H)$.

a linear field dependency. By the least-squares fittings in the low-T region, where the phonon contribution is negligible, we obtained $\Delta(0) = 35.9$ K, $\Delta(6) = 27.5$ K, $\Delta(9) = 22.5$ K and $\Delta(12) = 16.8$ K. As shown in the inset of Fig. 2, $\Delta(H)$ decreases in proportion to H. This can be explained by the Zeeman splitting of the excited triplet levels ($S = 1$). The fitting of the $\Delta(H)$–H data to the following relation, $\Delta(H) = \Delta(0) - g\mu_B H$ (g being the g factor of the Cu^{2+} electron spin), yielded $\Delta(0) = 35.0$ K. This value is quite consistent with the results of other measurements such as magnetic susceptibility (34 K) [2], ESR (34.7 K) [3].

As demonstrated above, the isolated dimer model seems to reproduce the experiment well, providing a reasonable value of spin gap. In higher temperature region, however, the deviation between our experiment and the theory is appreciable.[1] The magnetic entropy, for example, at 25 K is only about 74% of the expected value for the isolated dimer model with $\Delta(0) = 35.0$ K, indicating that the system is effectively correlated over much higher temperatures. It is noteworthy that the value of J is identical with that of $\Delta(0)$ for the isolated dimer model. Accordingly, $J = 35$ K ($= \Delta(0)$) obtained above is much smaller than $J = 100$ K (and also $J' = 68$ K) determined by Miyahara and Ueda [4]. We are looking forward to a theory based on the Shastry–Sutherland model with J and J' to reproduce our specific heat data over the whole T range.

References

[1] H. Kageyama, K. Yoshimura, R. Stern, N.V. Mushnikov, K. Onizuda, M. Kato, K. Kosuge, C.P. Slichter, T. Goto, Y. Ueda, Phys. Rev. Lett. 82 (1999) 3168.
[2] H. Kageyama, K. Onizuka, T. Yamauchi, Y. Ueda, S. Hane, H. Mitamura, T. Goto, K. Yoshimura, K. Kosuge, J. Phys. Soc. Japan 68 (1999) 1821.
[3] H. Nojiri, H. Kageyama, K. Onizuka, Y. Ueda, M. Motokawa, J. Phys. Soc. Japan 68 (1999) 2906.
[4] S. Miyahara, K. Ueda, Phys. Rev. Lett. 82 (1999) 3701.
[5] C.K. Majumdar, D.K. Ghosh, J. Math. Phys. 10 (1969) 1399.
[6] H. Kageyama, K. Onizuka, T. Yamauchi, Y. Ueda, J. Cryst. Growth 206 (1999) 65.
[7] R.L. Carlin, Magnetochemistry, Springer, Berlin, 1986 (Chap 5, p. 71).

[1] As space is limited, the detailed discussion in higher-T region will be given elsewhere.