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Magnetic Anisotropy of $SrCu_2(BO_3)_2$ with a Two-Dimensional Orthogonal Dimer Lattice

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Recently, we investigated the magnetic properties of $SrCu_2(BO_3)_2$[1)] in which Cu^{2+}–Cu^{2+} dimers connect orthogonally with each other to form a unique two-dimensional (2D) network as shown in Fig. 1.[2)] Together with several theoretical investigations,[3,4)] $SrCu_2(BO_3)_2$ was found to provide interesting physical phenomena, which are summarized as follows:

(1) This cuprate is a new 2D spin gap system with a gap of 30 K, and the ground state for this 2D spin network is solvable exactly, i.e., the direct product of singlet pairs is an eigenstate in spite of the existence of a second nearest (interdimer) interaction J'. This is due to the cancellation of the interdimer interactions.[3,4)] In contrast to theoretical works which have proved the exact dimer eigenstate for some 1-, 2- and 3D spin models,[5)] there was no experimental verification so far. $SrCu_2(BO_3)_2$ is the first case where this exact ground state is achieved.

(2) $SrCu_2(BO_3)_2$ is located close to the critical point between the dimer state and the Néel ordered state, and the theoretical calculations well reproduce the unusual temperature dependence of the magnetic susceptibility.[3)]

(3) The first observation of quantized magnetizations in 2D spin systems: 1/4 and 1/8 plateaus of the Cu saturation moment appear in the magnetization. Miyahara and Ueda discussed that the quantized plateaus should arise from the extremely localized character of excited triplets, and they predicted other plateaus at 1/2, 1/10, 1/16, etc.[3)]

As mentioned above, fundamental magnetic properties of $SrCu_2(BO_3)_2$ have been understood to a considerable extent. However, the previous results obtained using a polycrystal involve some ambiguities. For example, the anisotropy of the g-factor probably causes the magnetization plateaus to be obscured. In the present study, we will show the results of magnetic susceptibility

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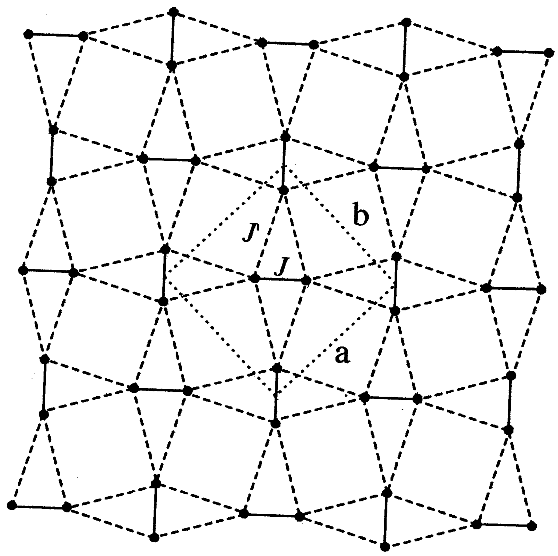

Fig. 1. The orthogonal Cu^{2+}–Cu^{2+} dimer lattice of $SrCu_2(BO_3)_2$ viewed along the c axis.

and high-field magnetization measurements for a field-oriented sample.

Powdered sample of $SrCu_2(BO_3)_2$ was synthesized by a conventional solid state reaction as described in ref. 2. The powdered sample was soaked into an epoxy, and then aligned magnetically in a magnetic field of $H = 9\,\mathrm{T}$. From an X-ray diffraction measurement, the alignment direction was found to be the c-axis (perpendicular to the 2D dimer lattice). The temperature T dependence of the magnetic susceptibility χ was measured using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design MPMS) in the T range 2 K and 400 K. The pulsed-high-field magnetization measurements were performed at 0.5 K by means of an induction method with well balanced pick-up coils. The duration time is about 20 ms.

The inset of Fig. 2 shows the magnetic susceptibilities of $SrCu_2(BO_3)_2$, $\chi_{/\!/}$ (along the c-axis) and $\chi_{\perp}$ (perpendicular to the same axis) as a function of T. For a comparison, the previous data for a nonoriented sample are also presented by a solid line.[1)] We fitted $\chi_{/\!/}$ and $\chi_{\perp}$ above 200 K to the Curie-Weiss model, $\chi = Ng^2\mu_B^2/4k_B(T-\theta)+\chi_0$, where N, μ_B and k_B represent Avogadro number, the Bohr magneton and Boltzmann constant, respectively, and obtained $g_{/\!/} = 2.23$, $g_{\perp} = 2.05$, $\theta = -97.8\,\mathrm{K}$ and $\chi_0 = -2.46\times10^{-5}$ emu/Cu mol. The obtained g-values agree well with $g_{/\!/} = 2.28$ and $g_{\perp} = 2.07$ determined by ESR[6)] and the slight deviation should be attributable to the incompleteness of sample orientation. The data below 4 K were fitted to the Curie-Weiss law, $C'/(T-\theta')$, and we estimated $C' = 3.3\times10^{-3}$ emu K/Cu mol and $\theta' = -2.5\,\mathrm{K}$, corresponding to 0.88% of nearly free $S = 1/2$ impurities. Finally, spin susceptibilities were obtained by subtracting this extrinsic Curie-Weiss term and χ_0 from the raw data. The theoretical susceptibilities were calculated by the quantum transfer matrix method for a finite system $N = 16$ with periodic boundary conditions,[7)] where $g_{/\!/} = 2.23$, $g_{\perp} = 2.05$ and $\theta = -97.8\,\mathrm{K}$ obtained above and $\Delta = 30\,\mathrm{K}$ determined by Cu-NQR measurement

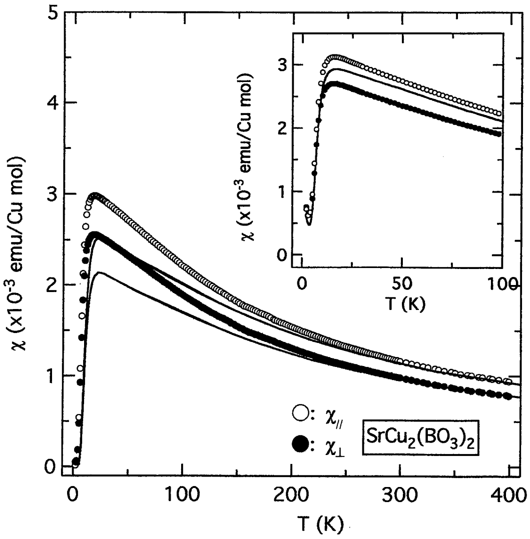

Fig. 2. Spin susceptibilities, $\chi_{/\!/}$ and $\chi_{\perp}$, as a function of temperature for $H /\!/ c$ and $H \perp c$, respectively. Solid lines are the theoretical susceptibility by Miyahara and Ueda. Inset shows the raw data of $\chi_{/\!/}$ and $\chi_{\perp}$. The solid curve represents the susceptibility for the nonoriented sample.

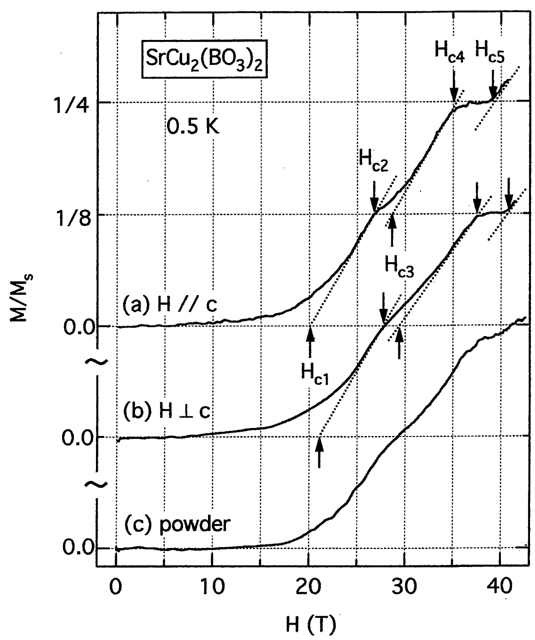

Fig. 3. Magnetization curves of $SrCu_2(BO_3)_2$ measured at 0.5 K for (a) $H /\!/ c$, (b) $H \perp c$, and (c) nonoriented sample.

were used. As seen in Fig. 2, agreement between the theory and the experiment is fairly good. The deviation in the low T range would be due to the small cluster size used in the theory. The intra- and interdimer exchange coupling constants were estimated to be $J = 105\,\mathrm{K}$ and $J' = 72\,\mathrm{K}$, respectively,[7] and the ratio $J'/J = 0.68$ is close to the critical boundary $(J'/J)_c = 0.70$ between the dimer state and the antiferromagnetic state.

Shown in Fig. 3 are the parallel and perpendicular magnetization curves, $M_{/\!/}$ (a) and $M_{\perp}$ (b), respectively, obtained at 0.5 K together with that for the nonoriented sample (c). Because the amount of the sample for the present measurement had to be small in order to make the sample align in the epoxy, the error in the absolute value of the magnetization is of the order of 10%. Accordingly, the saturation magnetization M_s was assumed in such a manner that the observed plateau for $35\,\mathrm{T} < H < 40\,\mathrm{T}$ corresponds to the 1/4 plateau. It is obvious that $M_{/\!/}$ and $M_{\perp}$ have more distinct plateaus (especially 1/4 plateau) compared with the data for the nonoriented sample.

The critical fields were evaluated by extrapolating the magnetization curves in the slope regions to the zero, 1/8- and 1/4-plateau lines in Fig. 3. We obtained $H_{c1} = 20.2\,\mathrm{T}$, $H_{c2} = 26.7\,\mathrm{T}$, $H_{c3} = 28.6\,\mathrm{T}$, $H_{c4} = 35.0\,\mathrm{T}$, $H_{c5} = 39.0\,\mathrm{T}$ for $H /\!/ c$ and $H_{c1} = 21.1\,\mathrm{T}$, $H_{c2} = 27.7\,\mathrm{T}$, $H_{c3} = 29.4\,\mathrm{T}$, $H_{c4} = 37.7\,\mathrm{T}$, $H_{c5} = 40.6\,\mathrm{T}$ for $H \perp c$. In the gap full regions $H < H_{c1}$, $H_{c2} < H < H_{c3}$, and $H_{c4} < H < H_{c5}$, $SrCu_2(BO_3)_2$ has the finite energy gap between the ground and the lowest exited states, and in the gapless regions $H_{c1} < H < H_{c2}$, $H_{c3} < H < H_{c4}$ and $H_{c5} < H$, the system has no excitation gap and the magnetization increases continuously. The critical field H_{ci} $(i = 1 \sim 5)$ for $H \perp c$ is larger than that for $H /\!/ c$, which comes from the difference of the g-factor.

Unfortunately, the predicted plateaus such as 1/10 and 1/16 were not observed in the present measurement. The difficulty lies in the fact that, when excited triplets form a regular lattice structure in a gapful region, less energy gain is expected with descending the plateaus as 1/2, 1/4, 1/8, 1/10, 1/16, $\cdots$.[7] Measurement using a single crystal as well as low temperatures should be needed in order to observe these plateaus.

In summary, we have measured the magnetic susceptibility as a function of T and the pulsed-field-magnetization for the oriented sample of $SrCu_2(BO_3)_2$. The theoretical susceptibility has succeeded to reproduce the experimental one. In the magnetization, more distinct plateaus were observed compared with the previous measurement for the nonoriented sample.

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