Magnetization Process in Mixed Magnetic Chains $(CH_3)_2CHNH_3Cu(Cl_xBr_{1-x})_3$ over the Gapless Phase Induced by Bond Randomness

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Abstract

Mixed magnetic systems $(CH_3)_2CHNH_3Cu(Cl_xBr_{1-x})_3$ over the intermediate phase ($0.44 < x < 0.87$) are good candidate compounds for experimental studies on the gapless state induced by bond randomness. We performed measurements of the magnetization process at 1.7 K on these mixed compounds. As a result, a typical spin flop transition, which indicated absence of the energy gap, was detected. From the variation in the spin flop transition field with x, the expected value of spins $\langle S_i^z \rangle$ was found to reach its maximum at $x \simeq 0.71$, the point at which bond randomness is highest.

Key words: bond randomness; gapless state; $(CH_3)_2CHNH_3Cu(Cl_xBr_{1-x})_3$; magnetization process;

Recently, we found useful candidate compounds for the investigation of the gapless state induced by bond randomness; they are $(CH_3)_2CHNH_3Cu(Cl_xBr_{1-x})_3$ (abbreviated as IPACu$(Cl_xBr_{1-x})_3$)[1] obtained by mixing the almost isomorphous compounds $(CH_3)_2CHNH_3CuCl_3$ and $(CH_3)_2CHNH_3CuBr_3$[2,3]. As shown in the schematic drawing of the crystal structure of IPACuX$_3$ (X=Cl, Br), which is given in Fig. 1 in Ref. 4, the neighboring Cu ions both in each dimer and between the dimers are bibridged by X ions, which we express as Cu-(X,X)-Cu. The intradimer exchange interaction $J_{\rm intra}$(X, X) and the interdimer one $J_{\rm inter}$(X, X) were determined to be $J_{\rm intra}({\rm Cl, Cl})/k = +54.1$ K and $J_{\rm inter}({\rm Cl, Cl})/k = -23.5$ K from the analysis of the magnetic susceptibility $\chi(T)$ of IPACuCl$_3$[4], while $J_{\rm intra}({\rm Br, Br})/k = -61$ K and $J_{\rm inter}({\rm Br, Br})/k = -33$ K from $\chi(T)$ of IPACuBr$_3$[5]. Therefore, the ground states of these compounds are different from each other although both compounds have a finite energy gap, i.e., IPACuCl$_3$ realizes the Haldane state[6], while IPACuBr$_3$ is the singlet dimer state.

When Cl ions in IPACuCl$_3$ are randomly substituted by Br ions, we expect $J_{\rm inter}({\rm Cl, Br}) = J_{\rm inter}({\rm Br, Cl}) < 0$ because of $J_{\rm inter}({\rm Cl, Cl}) < 0$ and $J_{\rm inter}({\rm Br, Br}) < 0$. On the other hand, $J_{\rm intra}({\rm Cl, Br}) = J_{\rm intra}({\rm Br, Cl}) < 0$ is obtained from analysis of the $\chi(T)$ data, as will be reported elsewhere[7]. To express the degree of the bond randomness, we introduce a parameter P which indicates the probability of $J_{\rm intra} > 0$; $(1 - P)$ inevitably the probability of $J_{\rm intra} < 0$. Since $J_{\rm intra}({\rm Cl, Cl}) > 0$, $J_{\rm intra}({\rm Cl, Br}) < 0$ and $J_{\rm intra}({\rm Br, Br}) < 0$, we obtain the relation $P = x^2$[7].

From the $\chi(T)$ and magnetic specific heat measurements for $0 \le x \le 1$[1], we expected that the gapped state persists for $0 \le x \le 0.44$ and $0.87 \le x \le 1$, while the gapless state induced by bond randomness appears over the intermediate phase ($0.44 < x < 0.87$). As a result of realizing the gapless phase, the antiferromagnetic long-range order occurs at $T_{\rm N} \simeq 15$ K,

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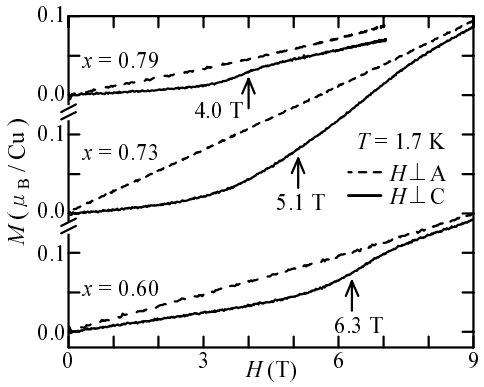

Fig. 1. The magnetization curves for $x = 0.79, 0.73$ and
The spin flop transition appears when $\boldsymbol{H}\perp$C plane. Eac
position of the $\mathrm{d}M/\mathrm{d}H$ curve is defined as the spin flo
sition field indicated by an arrow.

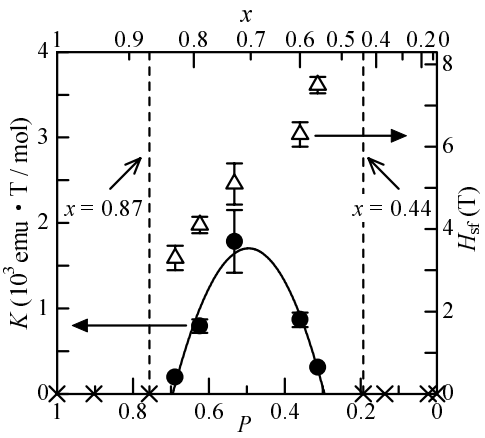

Fig. 2. The variations of the magnetic anisotropy constant K and the spin flop transition field $H_{\rm sf}$ with x and $P = x^2$, as indicated by closed circles and the open triangles, respectively. The crosses represent no observation of the spin flop transition. The solid line is a guide for the eyes.

which is due to the non-negligible interchain exc
interaction estimated to be less than 0.5 K[1]. T
tain more information about the intermediate p
we performed measurements of the magnetizatio
cess at 1.7 K ($\ll T_{\rm N}$). An external field $\boldsymbol{H}$ was a
along the normals of the three orthogonal surfa
each as-grown sample, A, B and C, as named i
4. According to the $\chi(T)$ data over the interm
phase[1], the $\chi(T)$ curves for $\boldsymbol{H}\perp$C plane, $\chi_{\rm C}$,
found to correspond to typical $\chi_{\parallel}(T)$, while the
curves for $\boldsymbol{H}\perp$A plane and B plane, $\chi_{\rm A}$ and $\chi_{\rm B}$
like $\chi_{\perp}(T)$. Therefore, a spin flop transition, whi... ...
dicates absence of the energy gap, is expected to ap-
pear when the $M(H)$ curves for $\boldsymbol{H}\perp$C plane.

Figure 1 shows the representative $M(H)$ curves over the intermediate phase. The $M(H)$ curves for $\boldsymbol{H}\perp$A plane show a straight line, but those for $\boldsymbol{H}\perp$C plane exhibit a typical spin flop transition. Therefore, the magnetic easy axis is found to be perpendicular to the C plane. When the values of the spin flop transition field $H_{\rm sf}$ are determined from the peak position of the derivative curve $\mathrm{d}M/\mathrm{d}H$ for $\boldsymbol{H}\perp$C plane, the variation of $H_{\rm sf}$ with x as well as $P = x^2$ are shown in Fig. 2. As can be seen in this figure, the values of $H_{\rm sf}$ monotonically increase with decreasing x. On the other hand, the $M(H)$ curves for $0 \le x \le 0.44$ and $0.87 \le x \le 1$ did not show a spin flop transition at all[7].

The appearance of magnetic long-range order means that the expected value of spins is not zero, i.e., $\langle S_i^z \rangle \neq 0$. We examine how $\langle S_i^z \rangle$ is affected by changing bond randomness. On the basis of the molecular field theory, $H_{\rm sf}$ is represented by using $\chi_{\parallel}$ and $\chi_{\perp}$ as

$$H_{\rm sf} = \sqrt{2K/(\chi_{\perp} - \chi_{\parallel})}, \qquad (1)$$

in which K is the magnetic anisotropy constant. In the present compounds, the magnetic anisotropy is due to the two-spin interactions such as the magnetic dipole-dipole interaction and the anisotropic exchange interaction, and consequently, $K \propto \langle S_i^z \rangle \langle S_j^z \rangle$. The variation in K with P suggests how $\langle S_i^z \rangle$ changes by varying the bond randomness. Employing $\chi_{\parallel} \equiv \chi_{\rm C}$ and $\chi_{\perp} \equiv (\chi_{\rm A} + \chi_{\rm B})/2$ and applying the values of $\chi_{\rm A}$, $\chi_{\rm B}$ and $\chi_{\rm C}$ observed at 1.8 K[1] into eq. (1), we calculate K at each value of x. The variation in the value of K with x as well as $P = x^2$ is added in Fig. 2. As can be seen in this figure, K versus P describes a parabolic curve with the maximum at $P \simeq 0.5$. We anticipate that $\langle S_i^z \rangle$ reaches its maximum at $P \simeq 0.5$, the point at which bond randomness is highest.

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