Bose-glass state in one-dimensional random antiferromagnets

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One-dimensional random antiferromagnets $(CH_3)_2CHNH_3Cu(Cl_xBr_{1-x})_3$ [abbreviated as IPA-$Cu(Cl_xBr_{1-x})_3$] show three magnetic phases: Cl rich, intermediate, and Br rich. In this study, to confirm the presence of a gapped or gapless state in addition to a Bose-glass (BG) state of spin triplets at two magnetic phase boundaries, magnetic susceptibility, specific heat, and magnetization were measured. Contrary to our proposed model, the results suggest a gapless state and a BG state at the Br-rich phase boundary, and a spin-gap state at the Cl-rich phase boundary. From the viewpoints of the universality class, the BG state may be interpreted as a result of Anderson localization. Therefore, IPA-$Cu(Cl_xBr_{1-x})_3$ are intriguing compounds for studying the BG state because of good one dimensionality.

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In one-dimensional antiferromagnets (1dAFM) with a spin gap, the ground state is a spin singlet and the first-excited state is a spin triplet separated by a finite spin gap (Δ) from the ground state. The origin of the spin gap in the case of half-integer-spin is clearly different from that in the integer-spin case. In the former case, since uniform 1dAFM have gapless ground states, they necessarily exhibit bond alternation ($-J_1$-J_2-J_1-J_2-), i.e., each set of strongly AFM coupled spins is formed as a singlet dimer state. In the latter case, uniform 1dAFM shows Haldane gap state.[1] In spin-gap systems, the original spin language succeeded in transforming the boson picture, which offers an intuitive understanding for complex quantum properties of matter.[2] For example, the Bose-Einstein condensation (BEC) of spin triplets occurs above a first critical magnetic field (B_{c1}) that corresponds to Δ.[3] As the review of the BEC in Ref. 3 suggests, in spin-gap systems, the boson density can be controlled by magnetic fields (B). The BEC states in $TlCuCl_3$,[4] TlCuCl, and $BaCuSi_2O_6$ (Ref. 5) were studied in detail experimentally. A similar phenomenon occurs in ultracold atomic gases in optical lattices.[6]

Our previous paper[7] showed that the symmetry of Haldane state breaks down because of a different reason, sufficient randomness. Magnetization [$M(B)$] measurements at $T=0.5$ K show that the transition from the BEC to the Bose-glass (BG) state of spin triplets occurred at a third critical magnetic field $B_{c3}\simeq 40$ T. In disordered Bose systems such as ^{4}He in a random potential, both the BEC and superfluidity are suppressed and may disappear. In disordered Bose systems, Fisher *et al.*[8] theoretically show new three states: superfluid, Mott insulating, and BG. Phase transitions induced by randomness are very common in condensed-matter physics. In strongly correlated electron systems, randomness leads to a competition between Mott transition and Anderson localization.[9] Boson localization due to randomness plays a key role in the vortex dynamics of cuprate superconductors.[10] ^{4}He in nanoporous glass exhibits localized BEC, which does not correspond to a global coherent BEC.[11] From the viewpoints of the universality class, a BG state of spin triplets in spin-gap systems may be interpreted as a result of Anderson localization because Δ is expected to disappear despite boson localization due to randomness. In experiments, a BG state was reported in a mixtures of two spin-gap compounds, $Tl_{1-x}K_xCuCl_3$.[12,13] Both the spin-gap compounds are three-dimensional singlet dimer systems. From the viewpoints of Anderson localization, boson localization tends to occur in 1dAFM rather than three-dimensional AFM. Now, we investigate the BG state in 1dAFM with a mixture of the Haldane and singlet dimer states.

The isomorphous compounds $(CH_3)_2CHNH_3CuCl_3$ and $(CH_3)_2CHNH_3CuBr_3$ (abbreviated as IPA-$CuCl_3$ and IPA-$CuBr_3$) are spin-gap systems with $\Delta/k_B\simeq 14$ K (Refs. 14 and 15) and 98 K,[16] respectively. Inelastic neutron-scattering measurements show that IPA-$CuCl_3$ consists of a two-leg ladder system along the *a* axis, with strong ferromagnetic (FM) rungs and weak AFM legs.[15] The resulting pseudo $S=1$ 1dAFM is formed along the *a* axis because of good one dimensionality. Magnetic susceptibility [$\chi(T)$] measurements,[16] on the other hand, indicate that Δ is very large in IPA-$CuBr_3$, compared with IPA-$CuCl_3$, although the leg and rung exchange interactions are of almost equal strength because of their similar crystal structures.[17] We think that all the rungs change from FM to AFM in IPA-$CuBr_3$. Therefore, the ground state of IPA-$CuBr_3$ consists of a singlet dimer state of the AFM rungs.

The mixtures of Haldane and singlet dimer state compounds, IPA-$Cu(Cl_xBr_{1-x})_3$, where the effect of bond randomness varies with *x*, were investigated by specific heat [$C(T)$],[18] $\chi(T)$,[18] and $M(B)$ (Refs. 7 and 19) measurements. We found three magnetic phases: $0<x<0.44$ (Br-rich phase), $0.56<x<0.83$ (intermediate phase), and $0.87<x<1$ (Cl-rich phase). The existence of a finite spin gap was confirmed in the Cl-rich phase,[7,18,19] and a similar spin-gap state may exist in the Br-rich phase.[18] On the other hand, gapless and ordered states corresponding to a quantum Griffith phase exist in the intermediate phase.[18–20]

There is a good possibility of observing the BG state at the phase boundaries, $x=0.87$ and 0.44, because at the

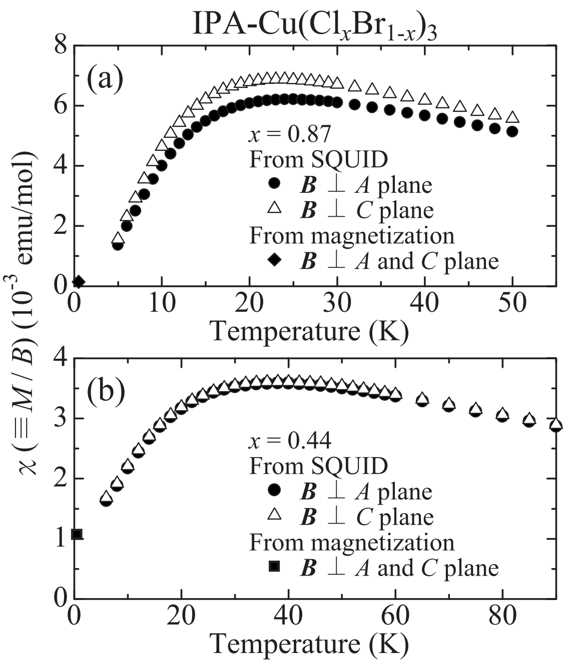

FIG. 1. Temperature dependence of magnetic susceptibility ($\chi \equiv M/B$) of IPA-Cu(Cl_xBr_{1-x})$_3$ for (a) x=0.87 and (b) 0.44, observed at B=0.1 T. $\chi(T)$ reaches its maximum at T_{max}=23–25 K for x=0.87 and T_{max}=38–40 K for x=0.44. At T=0.5 K, we estimate the value of $M(B)/B$ from the slope of the $M(B)$ below B=8 T. These values smoothly fit to the $\chi(T)$ curves.

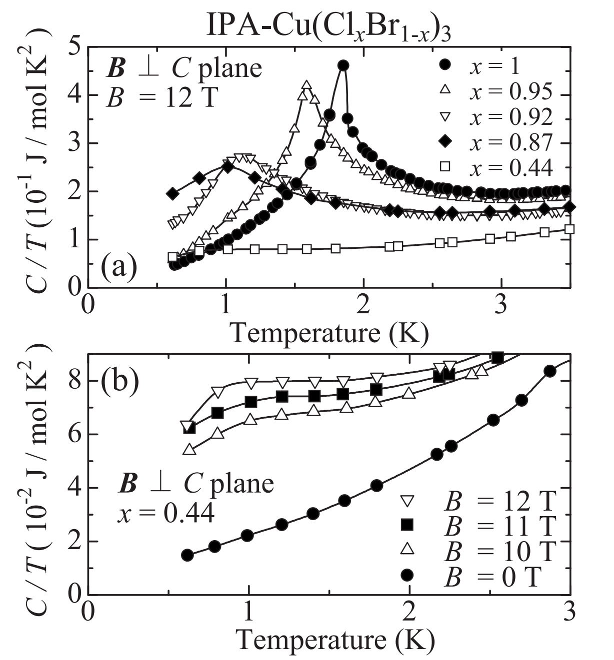

FIG. 2. (a) Temperature dependence of specific heat $[C(T)/T]$ for x=0.44 and the Cl-rich phase (x=1, 0.95, 0.92, and 0.87) obtained at B=12 T applied along the normal of the C plane. (b) $C(T)/T$ for x=0.44 obtained under various magnitudes of B. The ordinate is expanded from (a). The solid lines are guides for the eyes.

boundaries, the effect of bond randomness is maximum in each spin-gap phase. In the Cl-rich phase, the BEC was found to occur above $B_{c1} \simeq 10$–12 T with almost the same values of Δ, despite bond randomness.[7,18,19] In this study, to clarify the bond-randomness-induced BG state, $\chi(T)$, $C(T)$, and $M(B)$ were measured, mainly in the Br-rich phase boundary, because muon spin-relaxation results for x=0.35 strongly suggest a transition to the BG state at T=0 K.[21]

The preparation of IPA-Cu(Cl_xBr_{1-x})$_3$ is described elsewhere.[18] Here, we mention only that the A, B, and C planes refer to natural cleavage planes, which are close to, but do not coincide with, the corresponding crystallographic planes.[14] $\chi(T)$ was measured down to T=5 K using a superconducting quantum interference device (SQUID) magnetometer (Quantum Design, MPMS2). $C(T)$ was measured up to B=12 T over T=0.6–40 K using a Mag Lab^{HC} microcalorimeter (Oxford Instruments) by the relaxation method. In a previous study, $M(B)$ was measured at T=1.7 K (Ref. 19) and the spin-gap $M(B)$ behavior appeared for x=0.87, but the existence of the spin gap could not be determined for x=0.15–0.44.[19] Thus, $M(B)$ was measured at T=0.5 K. $\chi(T)$ and $M(B)$ were measured in the same sample as that used in Ref. 19.

Figure 1 shows $\chi(T)$ curves for x=0.87 and 0.44 down to T=5 K. These data agree well with a previous report:[18] $\chi(T)$ curves exhibit broad peaks at T_{max}=23–25 K for x=0.87 and T_{max}=38–40 K for x=0.44, and then show steep monotonic decreases and tend to zero with T decreasing to 0 K, which indicates that the ground state is a spin singlet state. We think that these phenomena strongly suggest a finite spin gap at T=0 K.[18] Figure 2(a) shows $C(T)/T$ curves at B=12 T for x=0.44, in addition to those for the Cl-rich phase (x=1, 0.95, 0.92, and 0.87). As shown in this figure, $C(T)/T$ curves in the Cl-rich phase exhibit sharp or cusplike peaks when the BEC occurs. As shown in Fig. 6 of Ref. 18, in the Cl-rich phase, the peak positions of $C(T)/T$ shift higher on the T side when B becomes higher. On the other hand, Fig. 2(b) shows the $C(T)/T$ curves for x=0.44 and these curves do not resemble those for the BEC state down to T=0.6 K and up to B=12 T. Broad and small peaks appear at $T \simeq 1$ K and the peak positions depend weakly on B, although the lattice contribution is not subtracted from the experimental values.

Figure 3(a) shows the $M(B)$ curves for x=0.87 when $\boldsymbol{B}$ is applied along the normal of the A and C planes. We obtain $B_{c1} \simeq 9$ T, which corresponds to Δ, and $B_{c3} \sim 35$ T, which is a transition from the BEC to BG states. This curvature is almost the same as $M(B)$ for x=0.95 and 0.92.[7] As already discussed in Ref. 7, these bending curves at the high-field region strongly resemble theoretical $M(B)$ curve in the BG state shown in Fig. 3(a) of Ref. 22. This theoretical model is AFM dimers with slight and random changes from weak AFM intradimer interactions to strong AFM ones.[22] The bond-randomness system in IPA-Cu(Cl_xBr_{1-x})$_3$ is essentially similar to this theoretical model.[7] Figure 3(b) shows the $M(B)$ curves for x=0.44 when $\boldsymbol{B}$ is applied along the normal of the A and C planes. As shown in the inset of Fig. 3(b), the slope of $M(B)$ below B=8 T, indicated by a dashed straight line, is large. This tendency is not shown in the inset of Fig. 3(a) for x=0.87. This slope strongly indicates a gapless ground state at B=0 T. We estimate the value of $M(B)/B$ at

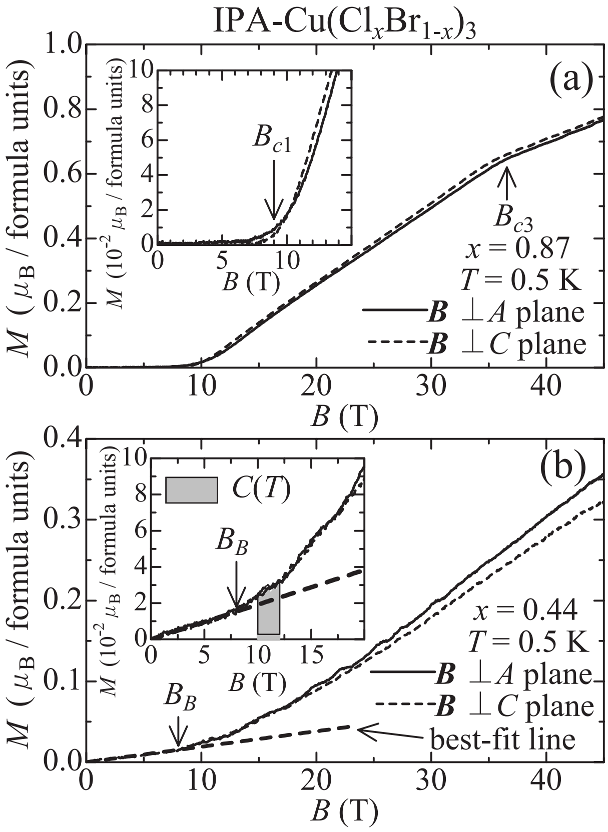

FIG. 3. Magnetization (M) versus magnetic field (B) at $T=0.5$ K, up to $B=45$ T for (a) $x=0.87$ and (b) 0.44. Each inset is the lower field portion. For $x=0.87$, the $M(B)$ curves show spin gap behavior. The first and third critical fields are $B_{c1}\simeq 9$ T and $B_{c3}\sim 35$ T. In the inset of (b), the gray hatch corresponds to magnetic fields for which the specific-heat measurements were performed. The dashed straight line is the best-fit line below $B=8$ T and $M(B)$ deviates from this line at a field defined as $B_B\simeq 8$ T.

$T=0.5$ K, which corresponds to $\chi(T=0.5\ \text{K})$, from the slope of $M(B)$ below $B=8$ T. This yields an $M(B)/B$ of 1.07×10^{-3} emu/mol for $x=0.44$, for the $\boldsymbol{B}\perp A$ and C planes. This value is plotted in Fig. 1(b). The $M(B)/B$ data at $T=0.5$ K correspond well with the $\chi(T)$ curves for $x=0.44$. In the case of $x=0.87$, we estimated $M(B)/B$ to be 1.44×10^{-4} emu/mol; this value is 1 order of magnitude smaller than the value for $x=0.44$. This value is plotted in Fig. 1(a), and the $M(B)/B$ data at $T=0.5$ K smoothly fit the $\chi(T)$ curves. If the value of 1.07×10^{-3} emu/mol for $x=0.44$ includes contribution from magnetic impurity ions, the $M(B)$ curves expect to show Brillouin functions at the lower B, but such $M(B)$ curves were not observed at all. As a result, we conclude that at $B=0$ T, $\chi(0)\neq 0$ and a gapless ground state exists for $x=0.44$. This conclusion is opposite to our proposed model, which derived from the exponential decay of $\chi(T)$ curves. Since we neglected the residual moments of 1×10^{-3} emu/mol$\simeq 3\times 10^{-6}$ emu/g for $x=0.44$, our assumption that the ground state is in the Br-rich phase was incorrect.

As shown in the inset of Fig. 3(b), the $M(B)$ curves begin to further increase from the best-fit line at $B\simeq 8$ T which is defined as B_B. Thus we measured $C(T)$ at $B=10-12$ T (gray hatch). If the concave curvature above $B>B_B$ indicates the BEC, sharp- or cusplike $C(T)/T$ curves are expected; however, this tendency is not shown in Fig. 2(b). Figure 2(a), furthermore, shows that $C(T)/T$ for $x=0.44$ should become larger more than $0.15-0.20$ J/(mol K^2) because the magnetic entropy is largely spent on the BEC.[23] The B dependence of the peak position for $x=0.44$ is not consistent with that for the Cl-rich phase. As a result, the BEC is absent because of boson localization due to randomness although M begins to further increase at $B>B_B$. As discussed by Fisher *et al.*,[8] these properties are consistent with the characteristics of the BG state. But the BG state is realized at $T=0$ K and $B\neq 0$ T,[8] and then we observed its permeation at a finite T. The broad and small peak of the $C(T)/T$ curve at $B=12$ T [Fig. 2(b)] resembles spin-glass behavior. But since a typical spin-glass state is weak against B, the $M(B)$ curvature at around B_B [the inset of Fig. 3(b)] cannot be explain by a typical spin-glass state. According to the boson picture, on the other hand, the BG state consists of quenched disorder that disrupts BEC via a mechanism similar to Anderson localization. Therefore, Fig. 2(b) strongly suggests a BG state of spin triplets, and Fig. 3(b) shows that a crossover from a gapless state without the BG state to a gapless state with the BG state may occur at around $B_B\simeq 8$ T. We found a field-induced BG state of spin triplets in 1dAFM with a mixture of the Haldane and singlet dimer states. The similar $M(B)$ gapless behavior is observed for $x=0.37$. We found that a gapless ground state exists for $0.37\leq x\leq 0.44$.

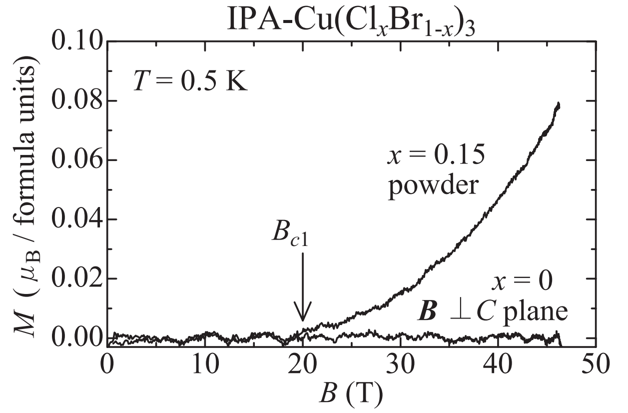

FIG. 4. Magnetization (M) versus magnetic field (B) at $T=0.5$ K for the $x=0.15$ powder sample and $x=0$ single crystal. For $x=0$, the spin gap is $B>45$ T, which agrees with the data for Ref. 19. For the $x=0.15$ powder sample, on the other hand, the $M(B)$ curve clearly shows spin-gap behavior and the spin gap is $B_{c1}\sim 20$ T.

Figure 4 shows the $M(B)$ curves at $T=0.5$ K for the $x=0.15$ powder sample and $x=0$ single crystal. The two curves clearly show spin-gap behavior, i.e., $B_{c1}\sim 20$ T for $x=0.15$ and $B_{c1}>45$ T for $x=0$. The concave curvature of rising $M(B)$ for $x=0.15$ is similar to those for $x=0.87$ [the inset of Fig. 3(a)] despite convex curvature in an ideal 1dAFM with spin gap, i.e., the square-root dependence.[24] According to Ref. 19, the probability of the FM rungs expressed as P in IPA-Cu(Cl$_x$Br$_{1-x}$)$_3$ is expressed as x^2 valid for the Cl-rich and Br-rich phases because the rungs form bibridged paths. Thus the value of $p\equiv(1-P)$ shows magnitude of the singlet dimer state. Since we estimate $B_{c1} \simeq 73$ T for $x=0$,[16] the value of the spin gap decreases rapidly over $0 \le x \le 0.15 (0.98 \le p \le 1)$. This tendency strongly suggests that the singlet dimer state is very weak against bond randomness, contrary to the Haldane state.[25] Furthermore, the critical point, or crossover from gapped to gapless states, should exist for $0.15 < x < 0.37 (0.86 < p < 0.98)$.

In conclusion, $\chi(T)$, $C(T)$, and $M(B)$ were measured for mixtures of Haldane and singlet dimer state compounds, IPA-Cu$(Cl_xBr_{1-x})_3$, for $x=0.87$ and 0.44, corresponding to the magnetic phase boundaries. For $x=0.87$, the existence of the spin gap and BEC states was confirmed, as expected. On the other hand, contrary to our proposed model, a gapless ground state was observed for $x=0.44$ at $B=0$ T. We think that the crossover from a gapless state without the BG state to a gapless state with the BG state occurs at around $B_B \simeq 8$ T. From the viewpoints of Anderson localization, dimensionality is very important. In $Tl_{1-x}K_xCuCl_3$, a gapless state is realized even for $x=0.05$.[13] As already pointed out by Shindo and Tanaka,[13] the effects of reduction in the spin gap did not make a distinction between three-dimensional exchange interactions and randomness. On the other hand, IPA-Cu$(Cl_xBr_{1-x})_3$ are suitable for studying the BG state because of good one dimensionality and existence of the gapped state $(0 < x \le 0.15)$. Furthermore, the intermediate phase only appears in IPA-Cu$(Cl_xBr_{1-x})_3$ for $0.56 \le x \le 0.83$. In the future, we will investigate the intermediate phase in more detail. Another interesting question remains: why there is a large difference between $T_N \le 2$ K at $B=12$ T in the Cl-rich phase and $T_N = 12-17$ K at $B=0$ T in the intermediate phase.[18]

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