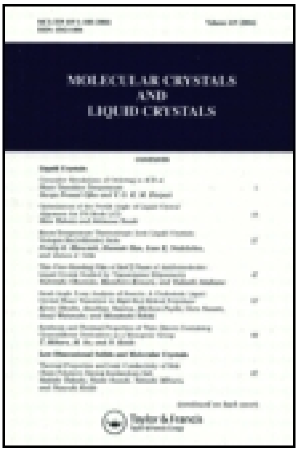

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Magnetic measurements have been performed to investigate random-bond effects on a spin-ladder system. We synthesize a random mixture compound $Cu_2(C_5H_{12}N_2)_2(Cl_{1-x}Br_x)_4$ which is expected to form a spin ladder with random-exchange legs and rungs. The magnetic properties of the compound are rather different from those of the pure systems. Paramagnetic divergence is significantly observed for the susceptibility at low temperatures and the magnetization appears even at low fields. The results suggest that the ground state of the random-bond ladder is a Griffiths phase.

Keywords: spin ladder; random-bond; susceptibility; magnetization; Griffiths phase; $Cu_2(C_5H_{12}N_2)_2(Cl_{1-x}Br_x)_4$

INTRODUCTION

New quantum antiferromagnetic chains, spin-ladder systems, have attracted a lot of theoretical and experimental interest. An even-leg ladder is believed to be a spin-gap system and to have a short-range resonating valence bond (RVB) ground state. The compound $Cu_2(C_5H_{12}N_2)_2Cl_4$ (hereafter, CHpC) is a typical two-leg ladder system with exchange interaction for the rungs ($J_{\perp}/k$=13.2K) and for the legs ($J_{\parallel}/k$=2.4K) of the ladder and a spin-gap energy of Δ/k=10.5K[1-3]. Recently, we reported experimental results of a new compound $Cu_2(C_5H_{12}N_2)_2Br_4$[4] (hereafter, CHpB) of which crystal structure is isostrucural with CHpC. The results suggest that the compound CHpB is a spin-ladder system with a spin-gap like CHpC. We estimated the exchange interactions $J_{\perp}/k$=24.8K, $J_{\parallel}/k$=3.5K and a spin-gap energy of Δ/k=21.3K. These compounds crystallize in a monoclinic $P2_1/a$ space group and the copper binuclear units stack up in the [101] direction forming an infinite ladder structure through the Cu-Cl···H-N-Cu hydrogen bond interaction. Therefore, the random mixture compound $Cu_2(C_5H_{12}N_2)_2(Cl_{1-x}Br_x)_4$ seems to be a ladder system with random antiferromagnetic bonds. Extensive theoretical work on a gapped quantum spin chain system shows that the system with random bonds is in a random singlet phase or a Griffiths phase which is a new type of quantum spin phase[5].

In this paper, we report our experimental results of susceptibility, magnetization and heat capacity measurements of a random bond ladder $Cu_2(C_5H_{12}N_2)_2(Cl_{1-x}Br_x)_4$ at low temperatures.

EXPERIMENTAL PROCEDURES

Powdered compounds $Cu_2(C_5H_{12}N_2)_2(Cl_{1-x}Br_x)_4$ were prepared by similar procedures as those used for the pure samples[1,4]. Namely, $Cu_2Cl_2 \cdot 2H_2O$ and Cu_2Br_2 (0.004mol total) were dissolved in a warm acetonitrile solution of $C_5H_{12}N_2$ (0.004mol) and the mixture were left at room temperature for two days. The content of Br^- ions, x, was determined by a fluorescence spectrometer. We think that isomorphous crystal structures of CHpC and CHpB and the small difference of the ionic radii between Cl^- and Br^- made easy to randomly replace Cl atoms by Br atoms. The *ac* susceptibility measurements at 15.9Hz were carried out using a SQUID magnetometer (Quantum Design MPMS-5S) above 1.7K and an *ac*-resistance bridge method below 1.7K. Magnetization measurements were performed under pulsed magnetic fields up to 27T at 1.6K. A conventional adiabatic calorimeter was employed to measure the heat capacity.

EXPERIMENTAL RESULTS AND DISCUSSION

The Magnetization curves of $Cu_2(C_5H_{12}N_2)_2(Cl_{1-x}Br_x)_4$ at 1.6K are shown in Fig.1. In the case of pure systems, CHpC and CHpB, the magnetization is close to zero up to the critical field H_{c1} and then increases almost linearly until around the saturation field H_{c2}, which exhibit the behavior that is typical for spin-gap systems. However, for

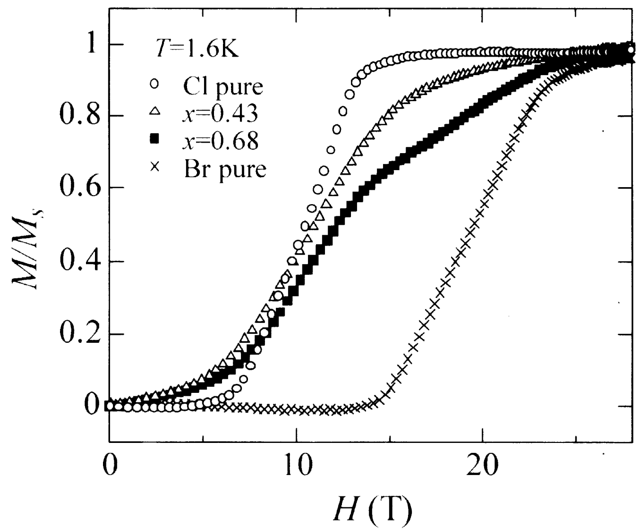

FIGURE 1. Magnetization curves of $Cu_2(C_5H_{12}N_2)_2(Cl_{1-x}Br_x)_4$ at 1.6K.

random systems, the magnetization is not zero even at lower fields and increases non-linearly until the saturated value. The curve seems to approaches to that of gapless systems.

In Fig.2 we show the temperature dependence of the *ac* susceptibility for $Cu_2(C_5H_{12}N_2)_2(Cl_{1-x}Br_x)_4$ in zero field. In the case of pure systems, the susceptibilities exhibit a broad maximum and decrease toward zero as the temperature is decreased, which are similar behavior observed for other spin-gap systems. For the random systems, the susceptibilities have a broad maximum around 8K but do not decrease toward zero as the temperature is decreased, which seems to approaches to that of gapless systems. Paramagnetic divergence is significantly observed at low temperatures. No anomaly corresponding to a long-range order is observed.

In order to investigate the behavior of the susceptibility at low temperatures, we show the log χ 'vs log$T$ plot in Fig.3. Below about 2K, the susceptibilities of pure systems increase almost linearly with decreasing temperature and follow the Curie law. Therefore, the susceptibilities seem to result from paramagnetic impurity spins.

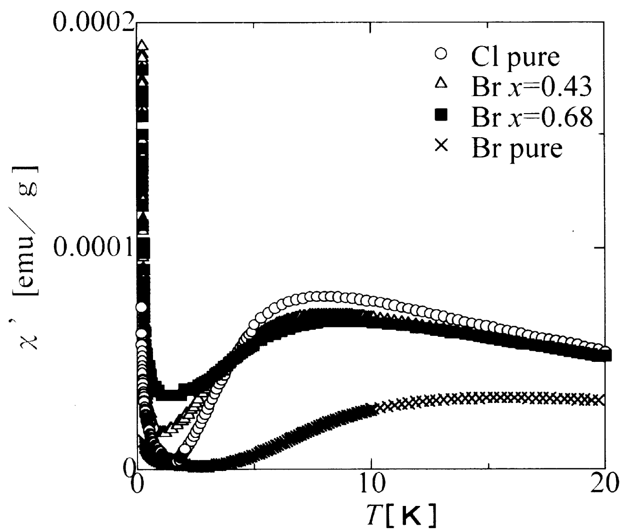

FIGURE 2. Temperature dependence of the ac susceptibility for $Cu_2(C_5H_{12}N_2)_2(Cl_{1-x}Br_x)_4$ in zero field.

However, for random systems, the susceptibilities do not follow the Curie law and diverge stronger than the Curie law. The systems with strong randomness are in a random singlet phase or are in a Griffice phase of which susceptibility diverges at low temperatures[5]. In the case of a random singlet phase, the susceptibility shows a weak divergence than that of the Curie law.[6] Therefore, the ground state of the random systems is not a random singlet phase.

The heat capacity of both pure and random systems was measured at low temperatures. In the cases of pure systems, the behavior of heat capacities is of the exponential type $T^{-2}\exp(-\Delta/kT)$, where Δ is the gap energy for the collective excitation. For random systems, the heat capacity at low temperatures shows similar behavior observed for pure systems in applied field[7], of which gap energy decreases with increasing H. There is no anomaly for pure and random systems down to 0.1K, which means the absence of the three-dimensional long range ordering. Therefore, the divergence of the susceptibility for the random systems is not due to the long range ordering.

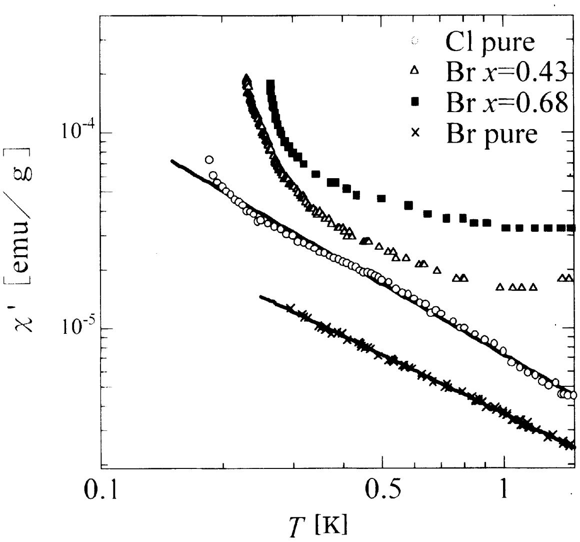

FIGURE 3. The low temperature region of the susceptibility for $Cu_2(C_5H_{12}N_2)_2(Cl_{1-x}Br_x)_4$ in zero field. Solid lines show the Curie-law susceptibilities.

CONCLUSIONS

In conclusion, the two-isostructural compounds, CHpC and CHpB, are typical spin-ladder systems with spin-gap. The magnetic properties of a random mixture compound $Cu_2(C_5H_{12}N_2)_2(Cl_{1-x}Br_x)_4$ are rather different from those of the pure systems. Paramagnetic divergence is significantly observed for the susceptibility at low temperatures and the magnetization appears even at low fields. The results suggest that the ground state of the random-bond ladder is a Griffiths phase.

Acknowledgments

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