Random Quantum Chain System: Mixture of S=1/2 Antiferromagnetic Chains with Uniform and Alternating Couplings

Y. Ajiro [a,1], T. Wakisaka [a] H. Itoh [a], K. Watanabe [a], H. Satoh [b] Y. Inagaki [a] T. Asano [a], M. Mito [c], K. Takeda [c], H. Mitamura [d], T. Goto [d]

[a] *Department of Physics, Kyushu University, Fukuoka 812-8581, Japan*
[b] *Department of Applied Physics, Fukui University, Fukui 910-8507, Japan*
[c] *Department of Applied Physics, Kyushu University, Fukuoka 910-8507, Japan*
[d] *Institute for Solid State Physics, University of Tokyo, Kashiwa, Chiba 277-8581, Japan*

Abstract

$CuCl_{2x}Br_{2(1-x)}\cdot 2(\gamma\text{-pic})$ ($0 \leq x \leq 1$) [γ-pic=4-Methylpyridine] is a rare example of random mixture of one-dimensional S=1/2 Heisenberg system with uniform (x=0) and alternating (x=1) antiferromagnetic couplings. From a systematic study of susceptibility and high-field magnetization of the mixed system, we found that the alternating ratio as well as the intrachain interaction continuously change with x, and, therefore, tunable with mixture. The ground state properties are consistent with the prediction for the random dimer (RD) phase.

Key words: Random quantum spin chain; Susceptibility; High-field magnetization

Extensive theoretical work on random quantum magnetic systems has been carried out since the late 1970's and a related issue on spin chains that have an excitation gap in the absence of randomness is attracting a renewed interest owing to the occurrence of the topological order in the ground state of the most prominent examples such as S=1 Haldane chains and dimerized S=1/2 chains. Theoretically, it has been predicted the systems with criticality in the absence of randomness are unstable against weak randomness but the gapped phases with the topological order are extremely stable against randomness [1].

Experiments on the gapped chains, however, are few and not sufficient to reveal the exact nature of the ground state of the random gapped quantum spin chains. In the previous work [2], we investigated experimentally the topological stability in the Haldene and spin-Peierls (SP) systems and observed a remarkable difference of the effect of randomness. Without randomness, two systems exhibit similar physical properties. They both have a nonmagnetic singlet ground state with a finite excitation gap, and more importantly, they have topological order. However, it was found that the Haldane phase is quite stable but the SP phase is extremely unstable, reflecting the difference of the physical origins opening the gap. The Haldane state is a disordered phase with no criticality, while the SP state is realized by the SP cooperative transition.

Here we are interested in the random mixture of one-dimensional (1D) S=1/2 Heisenberg antiferromagnets (HAF) with uniform and alternating couplings. It is expected theoretically that the uniform chain with criticality is unstable, while the alternating chain with dimerization is stable against randomness.

Figs. 1 and 2 show the temperature dependence of the powder susceptibility $\chi(T)$ between 300 K and 2 K at 0.1 T and the high-field magnetization $M(H)$ up to 38 T at 1.5 K, respectively, for $CuCl_{2x}Br_{2(1-x)}\cdot 2(\gamma\text{-pic})$ ($0 \leq x \leq 1$) [γ-pic=4-Methylpyridine]. For the pure Br-compound (x=0), $\chi(T)$ exhibits a broad maximum

[1] Corresponding author. E-mail: aji6scp@mbox.nc.kyushu-u.ac.jp

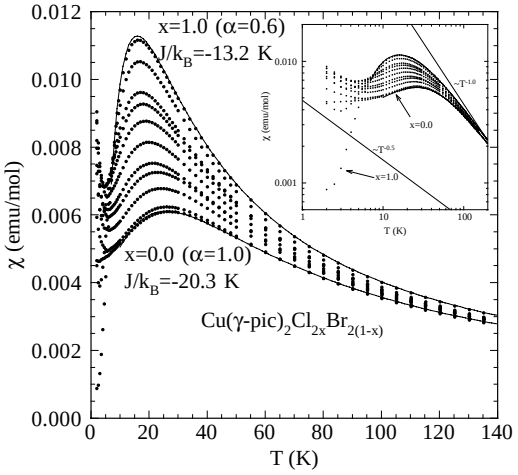

Fig. 1. Temperature dependence of the susceptibility $\chi(T)$ at 0.1 T.

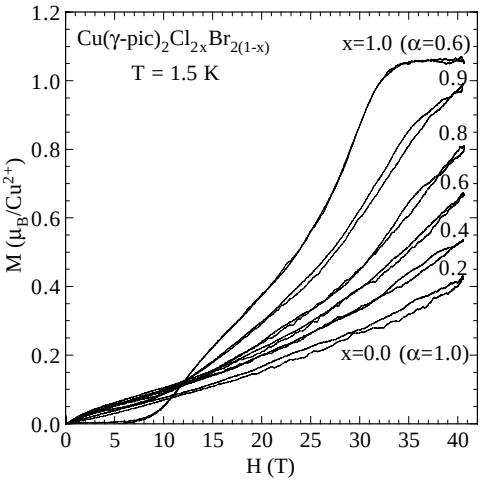

Fig. 2. High-field magnetization $M(H)$ at 1.5 K.

and approaches a finite value as $T\rightarrow$O and $M(H)$ increases nonlinearly with up-turn increase toward the expected saturation value of 1 $\mu_{\rm B}$, although the saturation field is unfortunately too high. The both features of $\chi(T)$ and $M(H)$ are typical for the gapless 1D HAF uniform chain and can be explained quantitatively with the intrachain AF coupling of $J/k_{\rm B}$=-20.3 K. In contrast, $\chi(T)$ of the pure Cl-compound (x=1) shows an exponential drop toward zero as $T\rightarrow$O, after passing a relatively sharp maximum. $M(H)$ is practically zero up to the critical field 10 T reflecting the nonmagnetic ground state with the energy gap of $\Delta/k_{\rm B}$=14.4 K and rapidly increases up to the saturation value at 33 T. The behaviors of $\chi(T)$ and $M(H)$ are also characteristic of the gapful 1D HAF alternating chain and can be explained quantitatively with the dominant AF coupling of $J/k_{\rm B}$=-13.2 K and the alternating ratio of α=0.6. The derived values are in good agreement with the existing data [3]. With the above observation in mind, first of all, we can see a systematic change in the overall shapes for mixed compounds in Figs. 1 and 2, featuring from one end representative of gapless uniform chain system (x=0) to the other end representative of gapful dimerized chain system (x=1). The characteristic peak in $\chi(T)$ becomes progressively sharper with down-shift of the maximum temperature, as x increases. The signature of bending of $M(H)$ around 10 T survives with down-shift of the critical field and the saturation field progressively increases, as x decreases from 1. These systematic behaviors suggest that the alternating ratio as well as the intrachain interaction continuously change, and, therefore, they are tunable with mixture. This is the first observation of tunable change of an alternating ratio α with mixture and may have an important significance for studying the predicted singular behavior at the critical ratio $\alpha_{\rm c}$ in the case of $S\geq$1, though $\alpha_{\rm c}$=0 in S=1/2 chain system.

Now we proceed to the interpretation of the observed low-T susceptibility divergence of the mixed compounds. The power law behavior of divergent contributions is demonstrated by log$\chi(T)$ vs. log$T$ plot in the inset of Fig. 1. As is easily seen, the susceptibility diverges like $\chi(T)\sim T^{\beta-1}$ with β=0.5 in agreement with the prediction for the random dimer (RD) phase [1]. In the RD phase, the system can be viewed as a collection of uncoupled spin pairs and the leading temperature dependence of $\chi(T)$ is physically easy to understand: all spins connected by bonds with energy greater than the temperature form singlets, and all spins connected by bonds with energy less than the temperature are essentially free but the amplitude of the $1/T$ divergence decreases as $T\rightarrow$0 due to the progressive dimerization in these free spins, giving rise to a mild divergence of $\chi(T)\sim T^{\beta-1}$. This scenario is consistent with the magnetization behavior at low field: Since an energy gap always exists for all α <1 in the dimerized chain and the energy gap is relatively small for the weakely dimerized chain ($\alpha\sim$1), the bending of $M(H)$ around 10 T survives with down-shift of the critical field and, at the same time, $M(H)$ contains a contribution of free spins which manifests as an initial humpt shoulder in the mixed compounds.

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