Proton NMR Study of a Random-Bond Ladder $Cu_2(C_5H_{12}N_2)_2(Cl_{1-x}Br_x)_4$

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Abstract

Proton NMR measurements have been performed to investigate random-bond effects in a spin-ladder system. We synthesized a random-bond ladder $Cu_2(C_5H_{12}N_2)_2(Cl_{1-x}Br_x)_4$ and measured the temperature dependence of ^1H spin-lattice relaxation time. The spin-gap energy for a random mixture compound is smaller than those for pure systems. The result suggests that the ground state of the random-bond ladder approaches to a gapless state.

Key words: spin ladder; random-bond; NMR; spin-gap

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1. Introduction

Spin-ladder systems, which are new quantum antiferromagnetic chains, have attracted a lot of theoretical and experimental interest. Extensive theoretical work on random quantum magnetic systems has been carried out since the late 1970s. However, there are few experimental studies on random-bond effects in the spin-ladder systems. Both of the compounds, $Cu_2(C_5H_{12}N_2)_2Cl_4$ (hereafter, CHpC) [1] and $Cu_2(C_5H_{12}N_2)_2Br_4$ (hereafter, CHpB) [2], are two-leg ladder systems with a spin gap. In particular, CHpC is known as a typical two-leg ladder and has been studied by many experimental measurements. Recently, we synthesized a random-bond ladder $Cu_2(C_5H_{12}N_2)_2(Cl_{1-x}Br_x)_4$ (hereafter, CHpCB) and studied it's magnetic properties [3]. For CHpCB, the paramagnetic divergence of the susceptibility is significantly observed at low temperatures and the magnetization appears even at low fields, which is rather different from those of the pure systems. The results suggest that the ground state of the random-bond ladder turns into a Griffiths phase or a gapless one due to the randomness. In the present work, we measured the proton spin-lattice relaxation time T_1 for CHpC, CHpB and CHpCB, and estimated their spin-gap energy Δ from its temperature dependence.

2. Experiment

Powdered compounds CHpCB were prepared by similar procedures used for CHpC and CHpB. Namely, $CuCl_22H_2O$ and $CuBr_2$ (0.004mol total) were dissolved in a warm acetonitrile solution of $C_5H_{12}N_2$ (0.004mol) and the mixture was left at room temperature for two days. The content of Br^- ions, x, was determined by a fluorescence spectrometer. We think that the isomorphous crystal structures of CHpC and CHpB and the small difference of the ionic radii between Cl^- and Br^- make it easy to replace Cl^- ions and Br^- ions randomly. The measurements of NMR were performed using the conventional phase-coherent pulsed-NMR spectrometer (PROT3001MR) produced by THAMWAY Inc.

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3. Results and discussion

First, we have measured the proton NMR spectra by observing a spin echo signal as a function of an external field at the frequency of 53MHz. For powdered pure systems, CHpC and CHpB, the proton NMR spectra at 4.2K have two lines. Only a broad line is observed in the proton NMR spectra for a powdered random system. Chaboussant *et al* [4] performed the proton NMR measurement using single crystals of CHpC in the fields higher than 4.5T. They obtained the complicated spectra which suggest two different proton sites with each different spin lattice relaxation time T_1. We measured recoveries of a spin echo signal at the field of the lines of the spectra. T_1 at the two peaks is same each other for both pure systems CHpC and CHpB. Figure 1 shows the recoveries of the intensity of a spin echo signal of proton at 4.2K. Surprisingly, the recovery curve for CHpCB is single-exponential as well as for pure systems. Therefore, T_1 is determined with the single-exponential recovery equation. Figure 2 shows the temperature dependence of $1/T_1$ for CHpC, CHpB and CHpCB(x=0.43). The dependence shows an activated behavior, $1/T_1 \propto \exp(-\Delta_h/kT)$, where Δ_h is an effective gap in applied field. We obtain $\Delta_h/k = 7.5$K, 15.4K and 3.3K for CHpC, CHpB and CHpCB(x=0.43), respectively. The spin gap at zero-field Δ is estimated Δ/k=9.3K, 17.2K and 5.0K for CHpC, CHpB and CHpCB(x=0.43), using the relation $\Delta_h = \Delta - g\mu H$. The values of Δ for CHpC and CHpB obtained by our NMR study are close to those obtained from the magnetization curves [1,2]. We emphasize that Δ for CHpCB (x=0.43), the random-bond ladder, is smaller than those for the pure systems, CHpC and CHpB. The result suggests that the ground state of the random-bond ladder approaches to the gapless state. It is well consistent with the results of the susceptibility and the magnetization [3]. Mélin *et al* [5] studied the random spin-ladder models using a numerical strong disorder RG method. They predicted that some systems with a spin-gap turn into a gapless Griffiths-type phase when the systems are strongly disordered. Our experimental results support the theoretical prediction.

4. Summary

We measured the temperature dependence of T_1 for the pure ladder and random-bond ladder systems. The gap Δ for the random-bond ladder is smaller than those for the pure ladders. This means that the ground state of the random-bond ladder approaches to the gapless state.

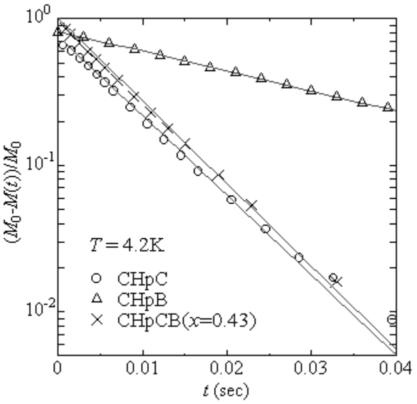

Fig. 1. Recoveries of the intensity of a spin echo signal at 4.2K

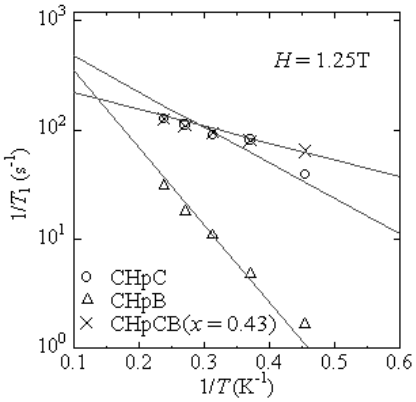

Fig. 2. Temperature dependence of $1/T_1$ at 1.25T

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