Abstract. (CH$_3$)$_2$CHNH$_3$Cu(Cl$_x$Br$_{1-x}$)$_3$ abbreviated IPACu(Cl$_x$Br$_{1-x}$)$_3$ is a mixed system of spin-gap compounds IPACuCl$_3$ and IPACuBr$_3$. This mixed system is reported by macroscopic measurements to show a magnetic order when the value of $x$ is between the quantum critical points (QCPs) 0.44 and 0.87. We have investigated microscopically the ground state of the samples $x = 0.40$, 0.45 and 0.50 which are located near the QCP of $x_c = 0.44$, by Muon Spin Relaxation ($\mu$SR) and NMR measurements. For $x = 0.40$, $\mu$SR measurements in zero and longitudinal fields have shown that the spin fluctuation shows a significant slowing down with decreasing temperature, although the magnetic order does not appear down to $T = 15$ mK, indicating that the ground state is an exotic one rather than a gapped one. For $x = 0.45$ and 0.50, $\mu$SR and NMR measurements do not show any magnetic order at low temperatures, indicating that the QCP is larger than the reported value of $x_c = 0.44$.

1. Introduction

(CH$_3$)$_2$CHNH$_3$Cu(Cl$_x$Br$_{1-x}$)$_3$ abbreviated IPACu(Cl$_x$Br$_{1-x}$)$_3$ is a mixed system of spin-gap compounds IPACuCl$_3$ and IPACuBr$_3$. IPACuCl$_3$ is characterized by a two-legged spin ladder along the $a$-axis with strongly coupled ferromagnetic (F) rungs, namely, an antiferromagnetic (AF) chain with an effective $S = 1$ composite Haldane chain[1]; its excitation gap is estimated to be 13.6 K by neutron inelastic scattering experiments[2]. As for IPACuBr$_3$, recent report shows that it is also characterized by a ladder and that the interaction between spins on a rung is AF[3]; its ground state is a singlet dimer state of the AF rungs with an excitation gap of 98 K[4].

The mixed system IPACu(Cl$_x$Br$_{1-x}$)$_3$ has been studied by magnetization and specific heat measurements[4], and a bond-randomness-induced AF ordered phase is reported to appear in the region $0.44 < x < 0.87$ with $T_N = 13 - 20$ K, depending on the value of $x[4]$. This phase, microscopically investigated on a sample with $x = 0.85$ by Muon Spin Relaxation ($\mu$SR) and Nuclear
Magnetic Resonance (NMR)[5,6], exhibited a three-dimensional AF long-range order. However, theoretical study by the quantum Monte Carlo method suggests that the system with \( x < 0.44 \) is also in the gapless state[7]. The purpose of this work is to investigate the ground state in the vicinity of the quantum critical point (QCP) \( x_c = 0.44 \) from a microscopic viewpoint by \( \mu \)SR and NMR.

2. Experimental

Single crystals of IPACu(Cl,Br\(_{1-x}\)) with \( x = 0.40, 0.45 \) and 0.50 have been grown by a solvent evaporation method from an isopropylalcohol solution of (CH\(_3\))\(_2\)CHNH\(_2\)-HX and CuX\(_2\) (X = Cl, Br). As-grown crystals are black in color, flat and rectangular in shape[3], and typically \( 1 \times 3 \times 2 \) mm\(^3\) in size.

\( \mu \)SR measurements were performed for about 15 pieces of single crystal with \( x = 0.40 \) and 0.45, so as to straddle the QCP at \( x_c = 0.44 \). \( \mu \)SR measurements were performed at the RIKEN-RAL Muon Facility in the UK and the Swiss Muon Source (S\(_\mu\)S), PSI in Switzerland. The incident muon-spin direction was parallel to the \( b^\ast \)-axis[3]. The decay of muon spin polarization, which is proportional to the spin polarization of the muon ensemble, was obtained from the ratio of the numbers of muon events counted by forward and backward counters; it is represented by the asymmetry parameter \( A(t) \).

\(^1\)H-NMR spectra were measured on sample with \( x = 0.45 \) and 0.50 by recording spin-echo amplitude, while ramping the magnetic field. The direction of the applied magnetic field was set parallel to the \( b^\ast \)-axis.

3. Results and Discussion

First, we consider the sample with \( x = 0.40 \). Figures 1 (a) and (b) show \( \mu \)SR time spectra in zero field (ZF) and various longitudinal fields (LFs). Muon spin rotation, indicating the presence of a magnetically ordered phase, is not observed down to 15 mK in the ZF. The \( \mu \)SR time spectrum over the entire temperature and field region is represented by the following function, containing two components:

\[
A(t) = A_1 G_{K_T}(t, \Delta) \exp(-\lambda_1 t) + A_2 \exp(-\lambda_2 t)
\]  

(1)

where \( G_{K_T}(t, \Delta) \) is the Kubo-Toyabe function whose parameter is the static field distribution width at muon site \( \Delta \), \( \lambda_1 \) and \( \lambda_2 \) are muon relaxation rates, and \( A_1 \) and \( A_2 \) are component fractions. The value \( \Delta = 0.23 \) \( \mu \)s\(^{-1} \) is constant within experimental precision, and is considered to be of nuclear spin contribution. The values of the component fractions are such that \( A_2/(A_1+A_2) = 0.14 \) at 6.5 K and reaches 0.4 at 15 mK. The details of parameter fitting are described in Ref. 8. The LF dependence of

![Figure 1. \( \mu \)SR time spectra for \( x = 0.40 \). (a) Temperature dependence in ZF. (b) LF dependence at the 15 mK. The dashed lines are the fitting results by Eq. (1).](image1)

![Figure 2. LF dependence of the relaxation rates \( \lambda_1 \) and \( \lambda_2 \) at \( T = 15 \) mK.](image2)
Muon spin relaxation rates $\lambda_1$ and $\lambda_2$ is shown in Fig. 2. This is interpreted as the Fourier spectrum of electron spin fluctuation, because muon spin relaxation in $H_{LF}$ probes selectively the Fourier component of spin fluctuation with muon spin Larmor frequency $\gamma_m H_{LF}$ (where $\gamma_m$ is the muon spin gyromagnetic ratio $13.5534 \text{ kHz/Oe}$). The observed spectrum obeys Redfield formula, which describes the spectrum of paramagnetically fluctuating spins, and is expressed as

$$\lambda(H_{LF}) = \frac{2(\gamma_m^2 \delta H_{loc})^2 \tau_c}{1 + (\gamma_m^2 H_{LF} \tau_c)^2}$$  \hspace{1cm} (2)

where $\delta H_{loc}$ is the local-field fluctuation amplitude at the muon sites, and $\tau_c$ is the characteristic fluctuation time constant. By fitting the observed $\lambda_1(H_{LF})$ and $\lambda_2(H_{LF})$ to Eq. (2), we obtain parameters $\delta H_{loc} = 8.86 \text{ Oe}$ and $\tau_c = 3.51 \mu\text{s}$ for the $A_1$ component and $\delta H_{loc} = 50.8 \text{ Oe}$ and $\tau_c = 1.42 \mu\text{s}$ for the $A_2$ component, respectively. The existence of the two components with different frequencies suggests that the system is microscopically separated into two phases[8], the magnetic phase corresponding to the $A_2$ component with larger $\delta H_{loc}$ and the nonmagnetic phase corresponding to the $A_1$ component with smaller $\delta H_{loc}$.

Figure 3 shows the temperature dependence of $\lambda_2$ in various LFs. Figure 4 shows the temperature dependence of the characteristic frequency of the spin fluctuation corresponding to the magnetic phase $\lambda_2$.

![Figure 3](image3.png)  \hspace{1cm} ![Figure 4](image4.png)

**Figure 3.** Temperature dependence of $\lambda_2$ for $x=0.40$ in various LFs. Solid curves are for a guide to the eye.

**Figure 4.** Temperature dependence of the characteristic frequency of the spin fluctuation corresponding to the magnetic phase $\lambda_2$.

By fitting the observed $\lambda_1(H_{LF})$ and $\lambda_2(H_{LF})$ to Eq. (2), we obtain parameters $\delta H_{loc} = 8.86 \text{ Oe}$ and $\tau_c = 3.51 \mu\text{s}$ for the $A_1$ component and $\delta H_{loc} = 50.8 \text{ Oe}$ and $\tau_c = 1.42 \mu\text{s}$ for the $A_2$ component, respectively. The existence of the two components with different frequencies suggests that the system is microscopically separated into two phases[8], the magnetic phase corresponding to the $A_2$ component with larger $\delta H_{loc}$ and the nonmagnetic phase corresponding to the $A_1$ component with smaller $\delta H_{loc}$.

Figure 5 shows the temperature dependence of $\lambda_2$ in various LFs. Figure 4 shows the temperature dependence of the electron-spin fluctuation frequency $\omega$, where $\lambda_2$ takes a maximum for each $H_{LF}$. At each temperature, $\omega$ coincides with the Larmor frequency of muon spins. It is evident that $\omega$ decreases monotonically to zero with the temperature decreasing to $T = 0$. This behavior is the same as that

![Figure 5](image5.png)  \hspace{1cm} ![Figure 6](image6.png)

**Figure 5.** Temperature dependence of time spectra for $x=0.45$ in ZF. The dashed lines are the fitting results of Eq. (1).

**Figure 6.** Temperature dependence of $\lambda_2$ in $H_{LF}=100 \text{ Oe}$. (Inset) LF dependence of $\lambda_1$ and $\lambda_2$ at $T = 2 \text{ K}$.
observed for $x = 0.35[8]$, and indicates the existence of a phase transition at absolute zero. Hence, the system has an exotic ground state.

Next, we consider the sample with $x = 0.45$. Figure 5 shows typical time spectra at various temperatures in ZF. No muon spin rotation is observed down to 2 K where the time spectra are represented by Eq. (1). The inset of Fig. 6 shows the LF dependence of relaxation rates $\lambda_1$ and $\lambda_2$. By fitting $\lambda_1(H_{LF})$ and $\lambda_2(H_{LF})$ to Eq. (2), we obtain parameters $\delta H_{loc} = 9.83$ Oe and $\tau_c = 5.75 \mu$s for the $A_1$ component and $\delta H_{loc} = 55.4$ Oe and $\tau_c = 0.98 \mu$s for the $A_2$ component, respectively. The values of the component fractions are such that $A_2/(A_1+A_2) = 0.36$ at 2 K. The existence of different fluctuation frequencies for the two components suggests that the system is microscopically separated into two phases for this sample as well. Above 8 K, time spectra are represented by a single-component equation $A(t) = AG_{k+}(t, \Delta) \exp(-\lambda t)$, indicating that the relaxation rates $\lambda_1$ and $\lambda_2$ are identical. Therefore, microscopic phase separation occurs at only low temperatures. Figure 6 shows the temperature dependence of $\lambda_2$ in $H_{LF} = 100$ Oe, where $\lambda_1$ is suppressed to be zero at 2 K. With decreasing temperature, $\lambda_2$ rapidly increases below 5 K and reached a high value of 0.46 $\mu$s$^{-1}$ at 2 K. The absence of magnetic order at finite temperature and the similarity of the temperature dependence of $\lambda_2$ with that for $x = 0.35[8]$ and 0.40 suggest an exotic ground state for $x = 0.45$.

NMR spectra for $x = 0.45$ and 0.50 show a sharp paramagnetic resonance line over the entire temperature range. Figure 7 shows typical spectra for $x = 0.45$ in field around 2 T, with peak widths as indicated. Peak widths for $x = 0.45$ and 0.50 (Fig. 8) are temperature independent within experimental accuracy. These observations indicate the absence of magnetic order and are consistent with the present $\mu$SR results. We thus conclude that the value of $x_c$ should be shifted from the reported $x_c = 0.44[4]$ to a value larger than 0.50.

Acknowledgement

We wish to thank Christopher Baines for his kind assistance with $\mu$SR experiments at PSI.

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