Ground state of quasi-one dimensional competing spin chain $Cs_2Cu_2Mo_3O_{12}$

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The ground state of the competing-spin chain $R_2Cu_2Mo3O_{12}^{(1,2)}$ has attracted much interest, not only for the possibility of spin nematic state,³⁻⁷⁾ but also because the two isomorphic compounds with $\mathbf{R}=\mathbf{C}\mathbf{s}$ and Rb show different ground states.^{8–11)} While the former shows a magnetic order at $T_{\rm N}=1.8$ K, the latter shows a small spin gap of $\Delta/k_{\rm B} = 1.6$ K. Within a classical spin model, the competing spin chain with the ferromagnetic ${\cal J}_1$ for nearest neighboring spins and the antiferromagnetic J_2 for next nearest spins, the helical order is expected for $|J_2/J_1| > 1/4$. For both the two compounds satisfy this condition, that is $J_1 = -93$ and $J_2 = +33$ K for Cs, and -138 and +51 K for Rb, the difference in their ground states may come from the quantum effect or a tiny inter-chain interaction. As for the Cs-system, the existence of magnetic order has so far been confirmed by the specific heat⁷⁾ and μSR^{8-11} at zero field, and by $NMR^{\hat{8}-11)}$ under finite fields. However, the observed increase in NMR line width below $T_{\rm N}$ becomes quite small when the applied field is lower than 2 T. That is, the increase in FWHM of NMR spectrum below $T_{\rm N}$ is decreased from 400 Oe for the measurements above 3 Tto only 120 Oe for below 2 T. In order to investigate the spin state under low fields, we have performed LF- μ SR experiments on this compound.

Zero (ZF) and longitudinal (LF) field- μ SR measurements in the ³He temperature range were performed on a powder sample at Riken-RAL Muon facility using a spin-polarized pulsed surface-muon (μ^+) beam with a momentum of 27 MeV/c. The zero-field muon spin depolarization data were analyzed with the function $G_{\rm KT}(\tau; \sigma) e^{-\lambda \tau}$, where $G_{\rm KT}$ is Kubo-Toyabe function, $\sigma \approx 0.068 \ \mu {\rm sec}^{-1}$, the temperature independent quasi-static field distribution contributed from nuclear moments and λ , the depolarization rate due to the dynamical spin fluctuation.

With decreasing temperature, ZF- λ showed an abrupt increase below $T_{\rm N} = 1.8$ K, supporting the results of specific heat and NMR.^{7,8)} Under finite LF's, the depolarization curves obeyed $G_{\rm KT}(\tau; \sigma)e^{-\lambda\tau} + P(\infty)$, where the constant $P(\infty)$ is the asymptotic value of muon spin polarization at $\tau \to \infty$. $P(\infty)$ was dependent on $H_{\rm LF}$, and for example, at 0.3 K, $P(\infty)$ increased monotonically from zero with increasing $H_{\rm LF}$ and reached 0.8 above 3 kOe.¹²⁾ There observed no initial drop in the depolarization function even at the lowest temperature, indicating that the relaxing amplitude was decreased under higher $H_{\rm LF}$.

The temperature dependence of λ under various $H_{\rm LF}$ is shown in Fig. 1. Note here that $\lambda(H_{\rm LF})$ is a measure of frequency component of $\gamma H_{\rm LF}$ in the electron spin fluctuation spectrum. One can see that in higher temperature region above $T_{\rm N}$, λ is small and nearly independent of $H_{\rm LF}$, indicating that the spin fluctuation has a white spectrum.¹³⁾ With decreasing temperatures, the weight of low frequency part of the spectrum increases significantly, showing the freezing of spin fluctuation due to the magnetic order. This is just analogous to our previous report where the slow down toward the Bose-glass is shown.¹³⁾ However, in the present case, the dynamical spin fluctuation persists even at lower temperatures far below $T_{\rm N}$. At this stage we can only speculate that the spins are in the quasi static ordered state, where only a small static hyperfine field is observed by NMR, and are gradually stabilized with increasing applied field. The latter is in accordance with the increase in $P(\infty)$ and in the hyperfine field at higher field.¹¹ In order to confirm this speculation, μ SR and NMR measurements under the widerange of applied field are necessary, and it is now on the progress.



Fig. 1. Temperature dependence of dynamical component of relaxation rate λ under various longitudinal fields $H_{\rm LF}$.

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