Disappearance of gapped Mott insulating phase neighboring Boseglass phase in $Tl_{1-x}K_xCuCl_3$ detected by longitudinal-field muon spin relaxation[†]

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$TlCuCl_3$ and $KCuCl_3$ are isostructural, and are three-dimensionally coupled Cu $3d$ S =1/2 spin dimer systems. The ground states are spin singlets with excitation gaps of Δ = 7.5 K in $TlCuCl_3$ and 31 K in $KCuCl_3$, originating from strong intradimer antiferromagnetic interaction J. Applying magnetic fields to the gapped state, the spin gap is collapsed, and a magnetically ordered state appears, which is qualitatively well described by the magnon Bose-Einstein condensation (BEC) theory[1)]. Describing the magnetic states by magnon motions, the spin singlet state corresponds to the gapped Mott Insulating (MI) phase.

By introducing randomness in the intradimer interaction, a new phase, Bose glass (BG) phase was theoretically predicted to appear at T = 0 neighboring the magnon BEC phase[2)]. In the mixed system $Tl_{1-x}K_xCuCl_3$, the randomness of the local chemical potential is introduced spatially, because the value of the dominant intradimer interaction J, which corresponds to the local potential of magnons, is different between $TlCuCl_3$ and $KCuCl_3$[3)]. Recently, Yamada *et al.* performed electron-spin resonance (ESR) measurements on $Tl_{1-x}K_xCuCl_3$ with x = 0.22 and 0.44 in resonance fields close to the critical field of BEC transition which is confirmed by specific heat measurements, and reported the change of the spectrum shape from Lorentzian shape to the intermediate shape between Gaussian and Lorentzian[4)]. This result suggests the localization of magnons at sufficiently low temperature, and suggest the appearance of the BG phase adjacent to the BEC phase. According to the theoretical prediction, the BG phase appears between the gapped MI phase and the BEC phase, and there exist a quantum phase transition point H_B from the BG phase to the gapped MI phase with decreasing the magnetic field. However, magnetization measurements suggest H_B =0. Thus, whether or not the gapped MI phase neighboring the Bose glass phase disappears in the zero-field limit is a controversial problem. The purpose of this study is to microscopically investigate this problem using LF - μSR technique.

Figure 1 shows temperarature dependence of LF - μSR time spectrum of $Tl_{1-x}K_xCuCl_3$ with x = 0.40 down to 25 mK in various longitudinal fields. All time spectra are analyzed using the stretched exponential function $A(t) = A_0 \exp(-\lambda t)^\beta$, and are well fitted as shown with solid lines. A_0 is the initial asymmetry and λ is the muon spin relaxation rate. Fitted results of β are almost constant for all spectra in a range of $\beta = 0.8 \pm 0.06$, which is consistent with the previous data for x = 0.40 in longitudinal fields. LF-μSR time spectra shows an exponential like decay, and the muon spin-relaxation rate does not has a significant temperature dependence down to 25 mK. As a fitted result, λ, which corresponds to a low frequency dynamical susceptibility, is almost constant down to 25 mK although slight changes in λ are observed. The low frequency spectrum seems to be a white spectrum, because λ in each field is finite below 500 gauss. These results mean that internal magnetic fields at the muon sites are fluctuating by low frequencies below $\sim$ 1 MHz down to 25 mK. When the spin system has a tendency toward a magnetic phase transition, λ is expected to increase with decreasing temperature in the zero-field limit below 500 gauss. Thus, in this case, a magnetic ordered state is not experimentally expected, and the ground state is a spin fluctuating state. It is suggested that the theoretically predicted quantum phase transition point from the Bose glass phase to the gapped Mott insulating phase disappears, i.e. H_B =0.

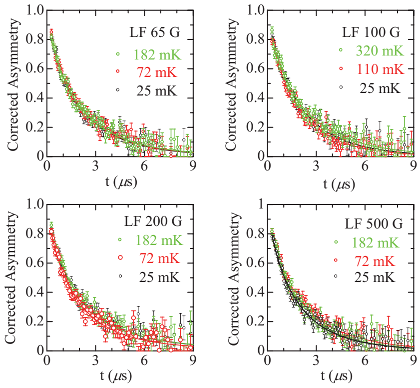

Fig. 1. Temperature dependence of LF-μSR time spectrum of $Tl_{1-x}K_xCuCl_3$ with x = 0.40.

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