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Quantum critical behavior in the highly random system $Tl_{1-x}K_xCuCl_3$ probed by zero- and longitudinal-field muon spin relaxation measurements

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Zero- and longitudinal-field muon-spin-relaxation (LF-μSR) measurements were carried out on the randomness-introduced quantum spin system $Tl_{1-x}K_xCuCl_3$. The relative temperature change in the muon-spin-relaxation rate λ in longitudinal-fields, which corresponds to the wave-vector integration of the generalized dynamical susceptibility, was deduced from LF-μSR measurements. Peak structure in the relative temperature change of λ for $x=0.60$ is observed at $T\sim 3$ K in 3950 G, and the temperature where the peak is observed decreases with decreasing the magnetic field. This behavior is interpreted as the soft mode of spin waves in the vicinity of the quantum critical point.

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I. INTRODUCTION

$TlCuCl_3$ and $KCuCl_3$, which are parent materials of the subject compound in this study $Tl_{1-x}K_xCuCl_3$, have the monoclinic structure (space group $P2_1/c$), and this crystal structure is composed of planar dimers of Cu_2Cl_6.[1,2] Magnetically, these isostructural materials are three-dimensionally coupled Cu 3d $S=1/2$ spin dimer systems, and their magnetic ground states are spin singlets with excitation gaps of $\Delta=7.5$ and 31 K, which originate from strong intradimer antiferromagnetic interaction J.[3–5] In these systems, field-induced magnetic ordering has been investigated extensively, and the obtained results are qualitatively well described by the magnon Bose-Einstein condensation theory.[6–21]

In the mixed system $Tl_{1-x}K_xCuCl_3$, the spatial randomness of the local chemical potential is introduced through the difference of the value of the dominant intradimer interaction J between $TlCuCl_3$ and $KCuCl_3$ because J corresponds to the local potential of magnons. Magnetization measurements suggest that the ground state is a magnetic state with finite susceptibility in the mixed system in zero field (ZF), although finite excitation gap remains.[22]

Recently, we have reported results of muon-spin-relaxation (μSR) measurements on $Tl_{1-x}K_xCuCl_3$ with $x=0.20$, 0.44, and 0.58. As for $x=0.20$, the increase and the saturation of the muon-spin-relaxation rate λ were observed at low temperatures, which is possibly a precursor to the Bose-glass phase at $T=0$.[23,24] These results are consistent with the theoretical and experimental predictions that the Bose-glass phase is expected to appear for $x>0$.[25–30] In the case of $x=0.44$ and 0.58, the increase of λ is observed, which suggests the slowing down of the frequency of the Cu 3d spin fluctuations toward a spin frozen state below 20 mK in contrast to the predicted Bose-glass phase, and the root-exponential-like behavior of the time spectrum indicates that the origin of the relaxation is possibly the spatially fixed fluctuating dilute moments.[31,32] The divergent increase of λ in $x=0.58$ suggests that the critical divergence around the phase boundary of the quantum critical point and the concentration dependence of λ suggests that the change toward the quantum critical point is of the second order.

However, the spin dynamics, which is the frequency spectrum of spin fluctuations, has not yet been clarified under the conditions that the randomness is enhanced with increasing the concentration of x and that the divergent increase of λ is observed. In order to investigate microscopic dynamical magnetic properties in highly random systems around $x=0.58$, we carried out the zero- and detailed longitudinal-field muon-spin-relaxation (ZF- and LF-μSR) measurements in $Tl_{1-x}K_xCuCl_3$ with $x=0.51$ and 0.60 single crystals.

II. EXPERIMENTS

Single crystals used in this study were grown from a melt by the Bridgman method. The details of crystal growth are given elsewhere.[22] The concentration of x was determined by the inductively coupled plasma atomic emission spectrometry (ICP-AES) method. Measurements of the μSR were made on $Tl_{1-x}K_xCuCl_3$ with $x=0.51$ and 0.60 at the RIKEN-RAL Muon Facility in the U. K. using a spin-polarized pulsed positive surface-muon beam with an incident muon momentum of 27 MeV/c. Used single crystals were cleaved in the helium gas just before each measurement and were mounted directly on a high-purity silver plate by an Apiezon N grease. The sample temperature was controlled with an Oxford ^{3}He cryostat in the range from 0.29 to 10 K. In μSR measurements, spin-polarized muons are implanted into samples. The incident muon-spin direction was perpendicular to the $(1,0,\bar{2})$ plane of single crystals. Forward and backward counters were located on the upstream and downstream sides of the beam direction, which was parallel to the initial muon-spin direction. The asymmetry parameter was defined as follows:

$$A(t)=\frac{F(t)-\alpha B(t)}{F(t)+\alpha B(t)}.$$

Here, $F(t)$ and $B(t)$ were total muon events counted by the forward and backward counters at a time t, respectively. The

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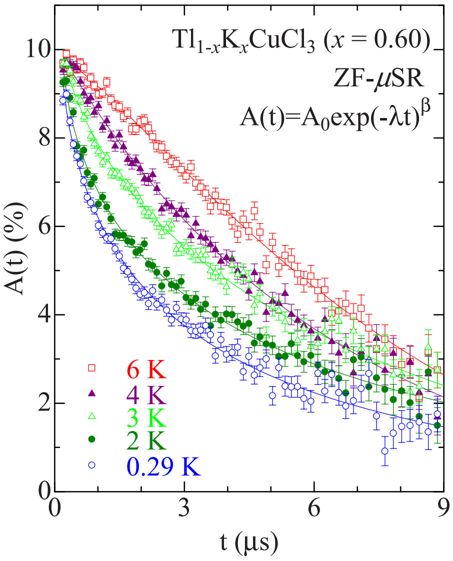

FIG. 1. (Color online) ZF-μSR time spectrum in $Tl_{1-x}K_xCuCl_3$ with $x=0.60$ at each temperature. Solid lines are fitted results using the function $A(t)=A_0\exp(-\lambda t)^\beta$.

α is a calibration factor reflecting relative counting efficiencies between the forward and backward counters. The initial asymmetry is defined as $A(0)$. Measured time spectra were analyzed using the WIMDA computer program.[33]

III. RESULTS AND DISCUSSIONS

Figure 1 shows ZF-μSR time spectrum in $Tl_{1-x}K_xCuCl_3$ with $x=0.60$ at each temperature. The shape of the time spectrum tends to change with deceasing temperature, and no rotational signal is observed down to 0.29 K. The μSR time spectra are analyzed using the stretched exponential function $A(t)=A_0\exp(-\lambda t)^\beta$. A_0 is the initial asymmetry and λ is the muon-spin-relaxation rate. All the time spectra are well fitted by the above function, as shown in Fig. 1 with solid lines. Obtained temperature dependence of the muon-spin-relaxation rate λ and of the power β is plotted in Fig. 2. The muon-spin-relaxation rate λ in zero-field increases below 3 K and saturates below 1 K. The increase of λ suggests the slowing down of the spin-fluctuation frequency of the Cu 3d spins. The power β of the stretched exponential function decreases with decreasing temperature and tends to saturate to $\beta=0.5$. These results are similar with the case in $x=0.44$ and 0.58.[31,32] As mentioned and discussed in our previous works, the root-exponential function suggests that the muon-spin-relaxation originates from the spatially fixed dilute moments fluctuating in time by the analogy of the nuclear-spin-lattice relaxation by spatially localized impurity spins in intermetallic compounds, which is discussed by McHenry *et al.*[34]

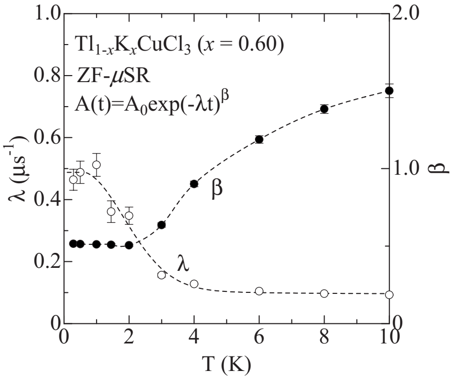

FIG. 2. Temperature dependence of the muon-spin-relaxation rate λ (left-hand side vertical axis, open circles) and of the power β (right-hand side vertical axis, closed circles). Dashed lines are guides for the eyes.

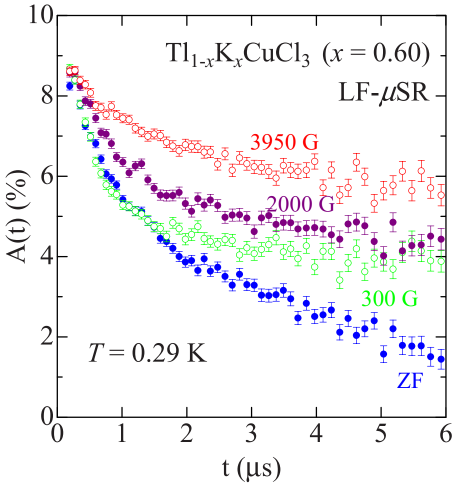

FIG. 3. (Color online) LF-μSR time spectrum in $Tl_{1-x}K_xCuCl_3$ with $x=0.60$ at 0.29 K in 0, 300, 2000, and 3950 G.

Figure 3 shows LF-μSR time spectrum in $Tl_{1-x}K_xCuCl_3$ with $x=0.60$ at 0.29 K. The fast relaxation in earlier time region is almost unchanged from zero-field to 300 G, becomes slower, and a finite relaxation exists at 3950 G. This means that there exists a fluctuating internal magnetic field which cannot be fully decoupled in 3950 G. Unfortunately, decoupled spectra are not analyzed using the function of the stretched exponential $A_0\exp[-(\lambda t)^\beta]$ with keeping β constant because the total internal magnetic field at muon sites is summation of magnetic fields generated by each relaxation center and the tendency of the decoupling of the muon spin from an internal field generated by each relaxation center is different. It leads to the drastic change in the time spectrum formula with increasing the external magnetic field. For this reason, we could not determine the absolute value of λ in magnetic fields and could not deduce the internal magnetic field and its fluctuation frequency at the muon sites using the Redfield fomula.[35] We can only conclude here that the Cu 3d spins are fluctuating at 0.29 K.

Generally, the two component function could be used to analyze distorted time spectra. When two kinds of homogeneous magnetic domains exist, time spectra can be well described by the two component function.[36,37] In the highly random system $Tl_{1-x}K_xCuCl_3$, however, measured time spectra are not fitted by the two component function at all, especially in the longitudinal-field dependence. This means that the two domains model cannot be applied in this system and is consistent with the model of spatially fixed dilute moments fluctuating in time as mentioned above.

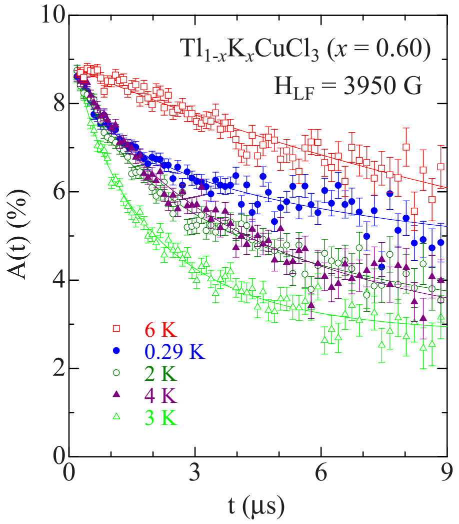

FIG. 4. (Color online) Time spectrum of the LF-μSR of $Tl_{1-x}K_xCuCl_3$ with $x=0.60$ in 3950 G at each temperature. Solid lines are fitted results using the function $A(t)=A_0\exp(-\lambda t)^{\beta}$.

In order to obtain the information about the frequency spectrum of spin fluctuations at the muon sites, the temperature dependence of the LF-μSR time spectrum is measured in constant fields. Figure 4 shows the LF-μSR time spectrum at each temperature in the longitudinal field of 3950 G. With decreasing temperature, the time spectrum comes to show a faster relaxation down to 3 K; however, below 3 K, the spectrum shape goes back to those of higher temperature, i.e., the relaxation becomes slower. To discuss the temperature change in the spectrum and the muon-spin-relaxation rate λ, the LF-μSR time spectra are analyzed using the stretched exponential function $A(t)=A_0\exp(-\lambda t)^{\beta}$. Figure 5 shows the deduced relative temperature change in the muon-spin-relaxation rate λ and of the power β in the longitudinal field of 3950 G. Peak structure in the relative temperature change of λ is observed at $T\sim3$ K. We have to notice again that a unit of λ is practically an arbitrary unit because power β shows the large temperature dependence and the change of β affects the absolute value of λ. Thus, we can only discuss the relative change of λ in this case.

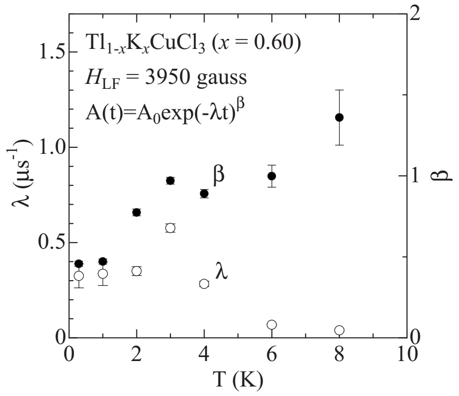

FIG. 5. Relative temperature change in the muon-spin-relaxation rate λ (left-hand side vertical axis, open circles) and of the power β (right-hand side vertical axis, closed circles) of $Tl_{1-x}K_xCuCl_3$ with $x=0.60$ in the longitudinal field of 3950 G.

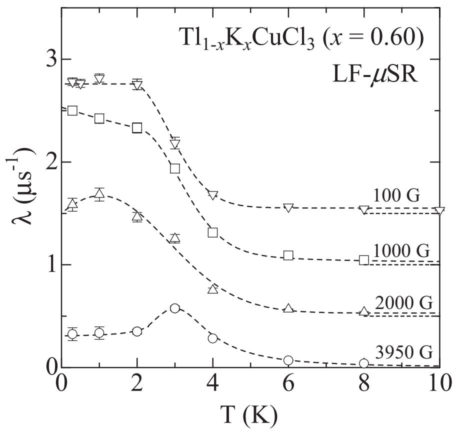

FIG. 6. Relative temperature change in the muon-spin-relaxation rate λ of $Tl_{1-x}K_xCuCl_3$ with $x=0.60$ in each longitudinal field. Dashed lines are guides for the eyes. Each plot is shifted upward consecutively by 0.5 μs^{-1} for clarity.

The same analysis procedure is applied to LF-μSR time spectra in other longitudinal fields, and we successfully analyzed all time spectra. Deduced temperature dependence of β in other longitudinal fields is similar with the case in 3950 G. The relative temperature change in the muon-spin-relaxation rate λ in each longitudinal field is summarized in Fig. 6. The temperature where the peak is observed decreases with decreasing magnetic field. The muon-spin-relaxation rate λ in the longitudinal field corresponds to the wave-vector (q) integration of the generalized dynamical susceptibility. In other words, the longitudinal field (H_{LF}) dependence of λ corresponds to the frequency ($\omega_{LF}=\gamma_{\mu}H_{LF}$) spectrum of spin fluctuations, where γ_{μ} is the gyromagnetic ratio of the muon spin ($2\pi\times13.5534$ kHz/G). Therefore, observed peak shift to lower temperatures with decreasing H_{LF} is the observation of the slowing down of Cu 3d spins fluctuation frequency and is interpreted as the soft mode of spin waves toward a possible magnetic phase transition. In the case of $x=0.60$, the change of λ saturates in low magnetic fields. This result in-

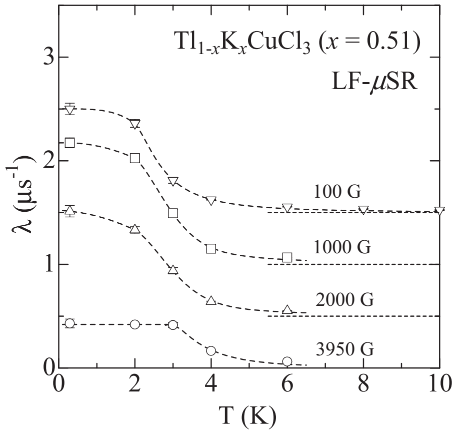

FIG. 7. Relative temperature change in the muon-spin-relaxation rate λ of $Tl_{1-x}K_xCuCl_3$ with $x=0.51$ in each longitudinal field, which is evaluated by the same procedure with the case of $x=0.60$. Dashed lines are guides for the eyes. Each plot is shifted upward consecutively by 0.5 μs^{-1} for clarity.

dicates that the ground state at the zero temperature is not magnetic, and the system is in the vicinity of the quantum critical point.

In another bond-randomness-introduced quantum spin system $(CH_3)_2CHNH_3Cu(Cl_xBr_{1-x})_3$, the similar behavior is reported in the case of $x=0.35$ which is the vicinity of the ordered state observed in the region of $0.44<x<0.87$.[38–41] In this case, the change of λ continues to increase in low magnetic fields, which indicates a magnetic ground state at the absolute zero.

Figure 7 shows the relative temperature change in the muon-spin-relaxation rate λ of $Tl_{1-x}K_xCuCl_3$ with $x=0.51$ in each longitudinal field, which is evaluated by the same procedure with the case of $x=0.60$. In contrast to the case of $x=0.60$, the peak structure is not seen in the relative temperature change of λ. With decreasing temperature, the muon-spin-relaxation rate λ increases and tends to saturate similarly in the wide region of the magnetic field. This means that the soft mode is not observed and that the frequency spectrum is nearly white in the case of $x=0.51$. This result indicates that in the case of $x=0.51$, the system is far from the quantum critical point. It is suggested that an ordered phase would appear in a narrow region of x around $x=0.6$ if it exists. The investigation of the appearance of the magnetically ordered state is in progress.

IV. SUMMARY

We carried out zero- and detailed longitudinal-field muon-spin-relaxation measurements on the randomness-introduced quantum spin system $Tl_{1-x}K_xCuCl_3$ with $x=0.51$ and 0.60. The relative temperature change in the muon-spin-relaxation rate λ in the longitudinal-fields was deduced from LF-μSR measurements. In the case of $x=0.60$, the peak structure in the relative temperature change of λ is observed at $T\sim3$ K in 3950 G, and the temperature where the peak is observed decreases with decreasing the magnetic field. This behavior is interpreted as the soft mode of spin waves in the vicinity of the quantum critical point.

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