Low-energy spin dynamics in randomness-introduced spin-gap systems $Tl_{1-x}K_xCuCl_3$ ($0.40 \le x \le 0.65$) probed by zero- and longitudinal-field muon spin relaxation

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Zero- and longitudinal-field muon spin-relaxation (ZF- and LF-μSR) measurements were carried out on randomness-introduced quantum spin-gap systems $Tl_{1-x}K_xCuCl_3$ in the wide range of x from 0.40 to 0.65. The temperature changes in the muon spin-relaxation rate λ in longitudinal fields were deduced from LF-μSR measurements. Peak structure in the temperature change in λ in 3950 G, which suggests the soft mode of spin waves, is observed in the region of $x \ge 0.53$, although a finite frequency of spin fluctuations remains down to $T=0$, i.e., the ground state is paramagnetic. The temperature giving the peak in λ decreases with decreasing the concentration of x and the peak structure vanishes in $x=0.51$. In the case of $x=0.40$, however, λ shows the different behavior and λ in lower fields increases with decreasing temperature down to 0.28 K. The increase in λ is possibly interpreted as a precursor of the transition to the reported Bose-glass phase. These results suggest a possibility of the existence of the quantum critical point from the Bose-glass phase to a paramagnetic phase around $x=0.51$.

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I. INTRODUCTION

The isostructural materials $TlCuCl_3$ and $KCuCl_3$ are three-dimensionally coupled Cu 3d $S=1/2$ spin dimer systems, and their magnetic ground states are spin singlets with excitation gaps of $\Delta=7.5$ K and 31 K, respectively, which originate from strong intradimer antiferromagnetic interaction J.[1–5] The spin dimers couple with one another through interdimer exchange interactions. In these systems, field-induced magnetic ordering has been investigated extensively, and the results obtained are qualitatively well described by the magnon Bose-Einstein condensation (BEC) theory.[6–20]

When the randomness in the intradimer interaction is introduced, a new phase called Bose-glass (BG) phase was predicted to appear at $T=0$ in the lower magnetic field region adjacent to the magnon BEC phase.[21–23] According to theoretical predictions, the BG phase is produced between gapped and ordered phases corresponding to Mott insulating and superfluid phases. By the correspondence between the spin system and the bosonic-particle system, the uniform magnetization M and the magnetic susceptibility $\chi=\frac{\partial M}{\partial H}$ correspond to the number of the bosons N and the compressibility κ. In the BEC phase, i.e., the superfluid state of bosons, the system is characterized by a finite staggered magnetization perpendicular to the applied magnetic field whereas the BG phase is distinguished by the disappearance of the coherent staggered magnetization keeping the magnetic susceptibility finite. Bose-glass phenomena have also been studied intensively in other disordered quantum systems, vortex lattices[24,25] and trapped atoms.[26,27]

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.[28] Shindo *et al.*[29,30] carried out specific-heat measurements in magnetic fields and observed field-induced phase transitions which are described as the BEC of spin triplets of Cu 3d spins in the mixed system $Tl_{1-x}K_xCuCl_3$. They discussed the obtained phase diagram in connection with the appearance of the BG phase. Recently, Yamada *et al.*[31,32] performed electron-spin resonance (ESR) measurements on $Tl_{1-x}K_xCuCl_3$ with $x=0.22$ and 0.36, where the resonance fields are close to the critical field of BG-BEC transition and observed the change in the line shape from Lorentzian typical of narrowed spectrum by magnons motion to the intermediate shape between Gaussian and Lorentzian, which is indicative of the localization of magnons at sufficiently low temperatures. This ESR result also suggests the appearance of the BG phase. Yamada[31] reported that the BG phase would appear in the region at least up to $x=0.44$.

For the case of a highly doped concentration of $x=0.60$, the soft mode of spin waves was observed by longitudinal-field muon spin-relaxation (LF-μSR) measurements.[33] The observed soft mode suggests that the system has strong spin fluctuations toward a magnetic phase transition. However, it is obscure at this stage whether or not this mode is headed for the BG phase, because the strong randomness could cause the transition to a magnetically ordered state. In another random bond one-dimensional Heisenberg chain system $(CH_3)_2CHNH_3Cu(Cl_xBr_{1-x})_3$, for example, two gapped phases are observed for $x<0.44$ and $x>0.87$ whereas a magnetically ordered phase at zero field is observed in the highly random region of $0.44 < x < 0.87$ by the magnetic susceptibility, specific heat, nuclear magnetic resonance, and μSR measurements.[34–37] Thus, in the highly doped $Tl_{1-x}K_xCuCl_3$, there is a possibility that spin fluctuations are headed toward not the BG phase but an ordered phase induced by randomness.

For further understanding of the randomness effect to ground states in quantum spin-gap systems, investigations of low-energy spin excitations are needed. The LF-μSR measurements are suitable for this purpose.[33,38–40] Thus, to obtain the microscopic information about the appearance of the BG phase and of other magnetic phases, we carried out ZF- and LF-μSR measurements in the wide region of x from 0.40 to 0.65.

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.[28] The concentration of x was determined by the inductively coupled plasma atomic emission spectrometry method. Measurements of μSR were carried out at the RIKEN-RAL Muon Facility in the U.K. using a spin-polarized double-pulsed positive surface-muon beam with an incident muon momentum of 27 MeV/c.[41] Used single crystals, which were cleaved to lots of small pieces in the helium gas just before each measurement, were mounted directly on a high-purity silver plate by an Apiezon N grease and were covered tightly by a high-purity silver film (thickness 50 μm) to ensure thermal contact. The sample temperature was controlled with an Oxford ^{3}He cryostat in the range from 0.28 to 8 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. Directions of crystal axis perpendicular to the incident muon spin direction were random on the silver plate. 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 $A(t)$ 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 α is a calibration factor reflecting relative counting efficiencies between the forward and backward counters and is determined by the muon spin rotation in the transverse field of 20 G at 8 K. The initial asymmetry is defined as $A(0)$. In this study, the calibration factor α and the background subtraction were taken into account for the data analysis. All μSR time spectra are plotted using the corrected asymmetry which is normalized by $A(0)$ in each concentration of x. Measured time spectra were analyzed using the WIMDA computer program.[42]

III. RESULTS

Figure 1 shows the temperature dependence of LF-μSR time spectrum of $Tl_{1-x}K_xCuCl_3$ $(x=0.51,0.53,0.60,0.65)$ in

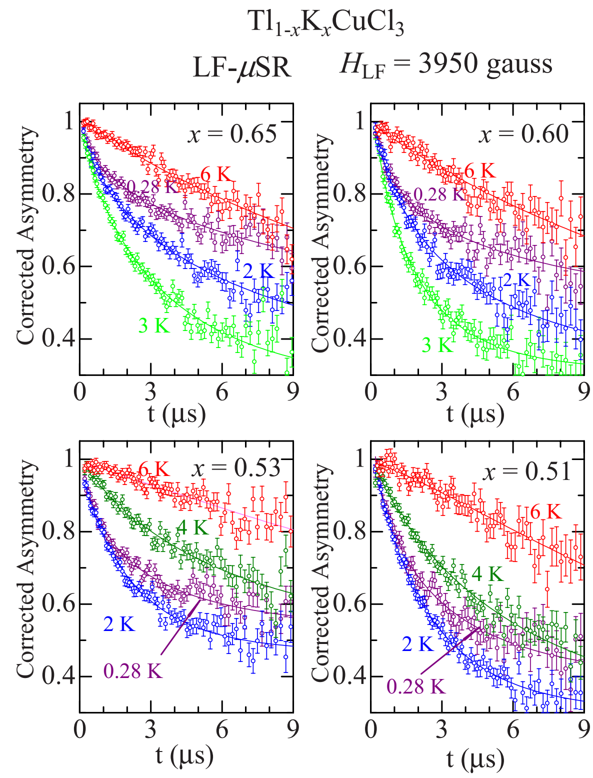

FIG. 1. (Color online) Temperature dependence of LF-μSR time spectrum of $Tl_{1-x}K_xCuCl_3$ $(x=0.51,0.53,0.60,0.65)$ in 3950 G. Solid lines are fitted results using the function $A(t)=A_0\exp(-\lambda t)^{\beta}$.

3950 G. All spectra are analyzed using the stretched exponential function $A(t)=A_0\exp(-\lambda t)^{\beta}$ in order to discuss the temperature change in the spectrum and of the muon spin-relaxation rate. A_0 is the initial asymmetry and λ is the muon spin-relaxation rate.

Generally, when we discuss the spin dynamics in magnetic materials by the muon spin-relaxation technique using the pulsed muon beam, it is ideal to determine the absolute value of λ in longitudinal magnetic fields and to deduce the internal magnetic field and its fluctuation frequency at the muon sites using the Redfield formula.[43] Unfortunately, in this study, spectra are analyzed using the stretched exponential function in which decoupled spectra by longitudinal fields are not analyzed with keeping β constant, and we cannot apply the Redfield formula to obtained results. 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. As mentioned blow, in zero field, β decreases with decreasing temperature and saturates to 0.5–0.6. The root-exponential function could suggest that the muon spin relaxation originates from the spatially fixed dilute moments fluctuating in time,[44,45] which is one possible model but is not the direct evidence. In order to discuss the spin dynamics without the Redfield formula, we attach importance to extract the temperature change in λ in a constant magnetic field so that the effect of decoupling by longitudinal fields is not considered in this study, although discussions becomes to be rather qualitative a weak.

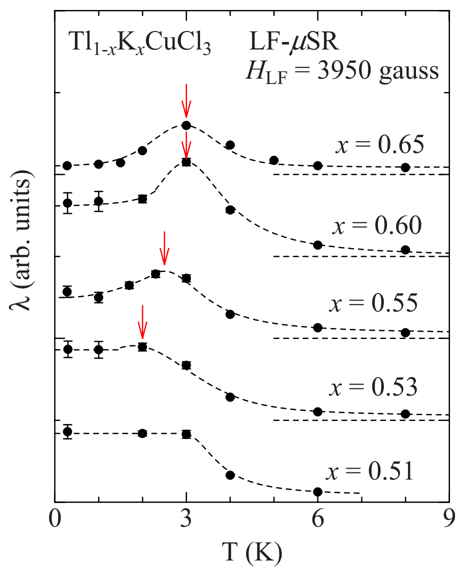

FIG. 2. (Color online) Temperature change in the muon spin-relaxation rate λ of $Tl_{1-x}K_xCuCl_3$ (x=0.51,0.53,0.60,0.65) in the longitudinal field of 3950 G. Dashed lines are guides for the eyes. Arrows indicate the temperature where the peak appears.

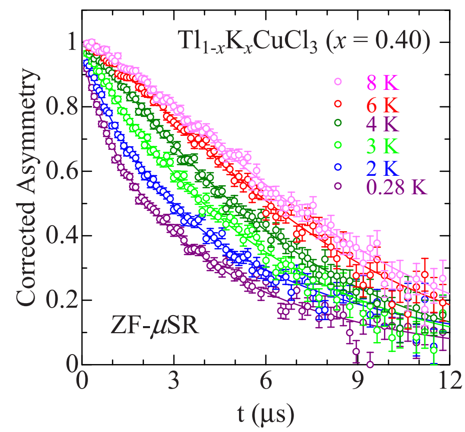

FIG. 4. (Color online) Temperature dependence of ZF-μSR time spectrum of $Tl_{1-x}K_xCuCl_3$ with x=0.40. Solid lines are fitted results using the function $A(t)=A_0 \exp(-\lambda t)^{\beta}$.

All the time spectra are well fitted by the stretched exponential function, as shown in Fig. 1 with solid lines. For all samples in this study, the temperature dependence of β shows almost the same tendency as reported for x=0.51 and 0.60,[33] namely, β is ~1.5 at 8–10 K, decreases with decreasing temperature, and saturates to 0.5–0.6. In the case of x =0.60 and 0.65, the time spectrum becomes to show a faster relaxation down to 3 K, and below 3 K, the relaxation changes to be slower with decreasing temperature. In the case of x=0.51, however, the gradient of the time spectra in earlier time region does not show visible change below 2 K, which means the relaxation is not temperature dependent below 2 K. Temperature change in time spectra below 2 K is reflected from the change in the power β. Figure 2 shows obtained temperature changes in the muon spin-relaxation rate λ in 3950 G. In Fig. 2 which shows the concentration dependence of the temperature change, λ is plotted by an arbitrary unit because power β shows the large temperature dependence and the change in β affects the absolute value of λ. For x=0.65 and 0.60, peak structure appears in the temperature change in λ at 3 K. With decreasing the concentration of x, the temperature where the peak appears (T_{peak}) decreases and the peak structure collapses. The concentration x dependence of the peak temperature T_{peak} in 3950 G is shown in Fig. 3.

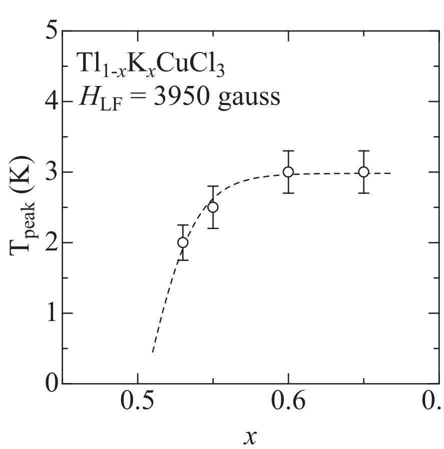

FIG. 3. Concentration x dependence of the temperature where a peak appears in the temperature change in the muon spin-relaxation rate shown in Fig. 2. Dashed line is guide for the eyes.

Figure 4 shows the temperature dependence of ZF-μSR time spectrum for $Tl_{1-x}K_xCuCl_3$ with x=0.40, where the ground state is expected to be in the Bose-glass phase from results of magnetization, neutron-scattering, specific-heat, and ESR measurements.[29–31] The shape of the time spectrum tends to change with decreasing temperature. ZF-μSR time spectra are analyzed using the stretched exponential function $A(t)=A_0 \exp(-\lambda t)^{\beta}$. Obtained temperature dependence of λ and of the power β is plotted in Fig. 5. The muon spin-relaxation rate λ in zero field increases with decreasing temperature and tends to saturate around 1.6 K. Below 1 K, λ begins to increase again down to 0.28 K. The power β decreases with decreasing temperature and tends to saturate to β=0.75. The characteristic behavior of λ, that the tempera-

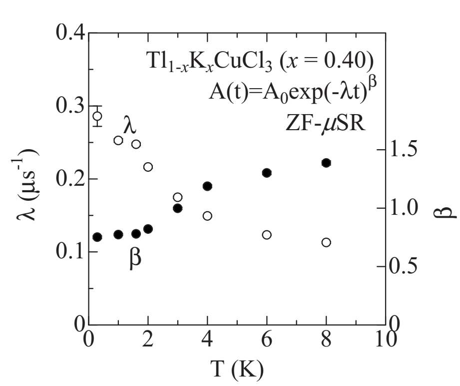

FIG. 5. Temperature dependence of the muon spin-relaxation rate λ (left-hand side vertical axis, open circles) and of the power β (right-hand side vertical axis, closed circles) in $Tl_{1-x}K_xCuCl_3$ with x=0.40.

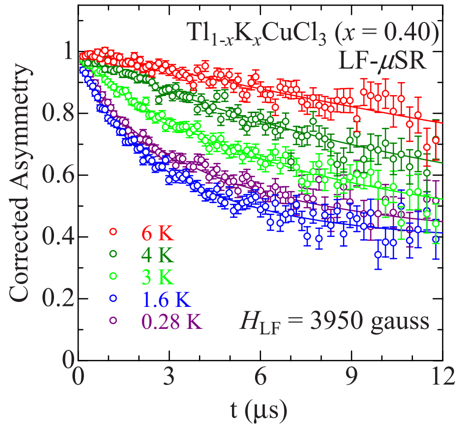

FIG. 6. (Color online) Temperature dependence of LF-μSR time spectrum of $Tl_{1-x}K_xCuCl_3$ with $x=0.40$ in the longitudinal field of 3950 G. Solid lines are fitted results using the function $A(t)=A_0\exp(-\lambda t)^{\beta}$.

ture dependence shows a shoulderlike change, is similar to that in the case of $x=0.20$ (Ref. 46) and is different from the case in $x\geq 0.51$ where λ in ZF saturates at lower temperatures.[33]

In order to investigate low-energy spin excitations in the region where the ground state is expected to be in the Bose-glass phase, we carried out detailed LF-μSR measurements for $x=0.40$. Figure 6 shows the temperature dependence of LF-μSR time spectrum in 3950 G. All time spectra in longitudinal fields are analyzed using the stretched exponential function. With decreasing temperature, the time spectrum becomes to show a faster relaxation down to 1.6 K and the relaxation becomes slower. Figure 7 shows the obtained temperature change in λ in various fields above 100 G. Above 1000 G, the peak structure appears in the temperature change in λ, and with decreasing the longitudinal field, the peak temperature slightly decreases. In 100 G, however, λ tends to saturate around 1.6 K and begins to increase again below 1 K with decreasing temperature. The shape of the temperature change curve, which shows a shoulder around 1.6 K, is similar to that obtained by the ZF-μSR mentioned above. Figure 8 shows the temperature dependence of LF-μSR time spectrum for $x=0.40$ in 10 G, which is a typical behavior in lower longitudinal fields. The gradient of the time spectrum in earlier time region becomes larger with decreasing temperature down to 0.28 K. Figure 9 shows the obtained temperature change in λ in lower longitudinal fields of 10, 20, and 100 G. In all the fields below 100 G in this study, λ shows the similar temperature change.

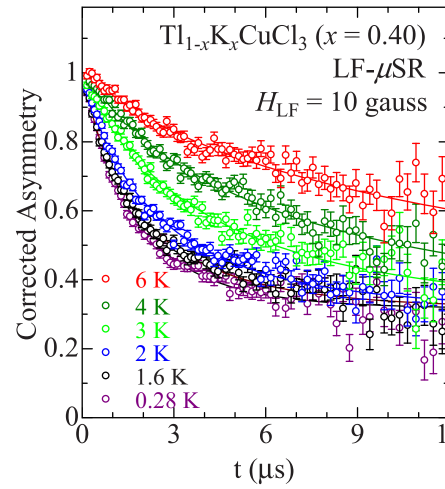

FIG. 8. (Color online) Temperature dependence of LF-μSR time spectrum of $Tl_{1-x}K_xCuCl_3$ with $x=0.40$ in the longitudinal field of 10 G. Solid lines are fitted results using the function $A(t)=A_0\exp(-\lambda t)^{\beta}$.

The important fact is that with decreasing the concentration of x, the peak temperature T_{peak} in 3950 G decreases and the peak structure vanishes in $x=0.51$. These results strongly suggest that the ground states are different between $x>0.51$ and $x<0.51$ region. We have a detailed discussion about the low-energy spin dynamics in the next section.

IV. DISCUSSION

As discussed in our previous report,[33] the muon spin-relaxation rate λ in the longitudinal field corresponds to the

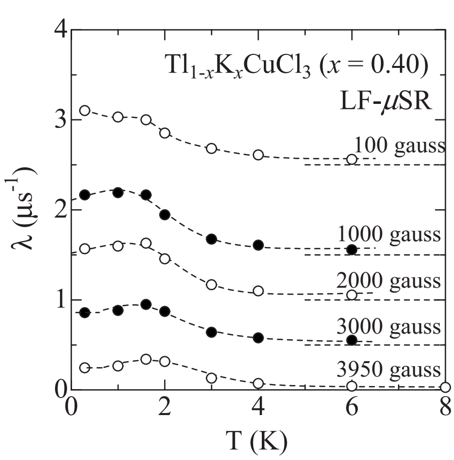

FIG. 7. Temperature change in the muon spin-relaxation rate λ of $Tl_{1-x}K_xCuCl_3$ with $x=0.40$ in various longitudinal fields above 100 G. Error bars are within symbols. Dashed lines are guides for the eyes. Each plot is shifted upward consecutively for clarity.

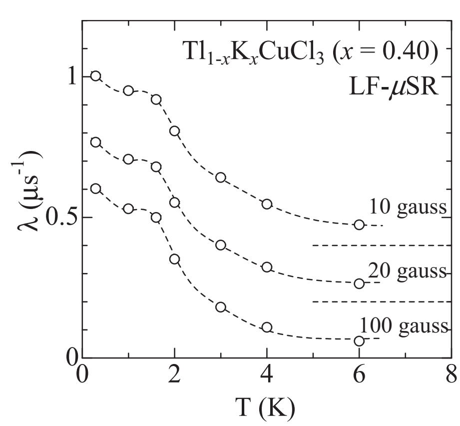

FIG. 9. Temperature change in the muon spin-relaxation rate λ of $Tl_{1-x}K_xCuCl_3$ with $x=0.40$ in lower longitudinal fields of 10, 20, and 100 G. Error bars are within symbols. Dashed lines are guides for the eyes. Each plot is shifted upward consecutively for clarity.

wave-vector (q) integration of the generalized dynamical susceptibility. In other word, the longitudinal field (H_{LF}) dependence of λ corresponds to the frequency spectrum of spin fluctuations. Thus, the peak structure in the change in λ (Fig. 2) means that with decreasing temperature, Cu 3d spins fluctuation frequency is slowing down and that a frequency of the maximum intensity in the spectrum passes through around $\omega_{LF}=\gamma_{\mu}H_{LF}$, where γ_{μ} is the gyromagnetic ratio of the muon spin ($2\pi\times 13.5534$ kHz/G). This behavior is interpreted as the soft mode of spin waves toward a possible magnetic phase transition. Here, it is emphasized that the ground state for $x\geq 0.51$ is a paramagnetic state because the change in λ in lower magnetic fields below 100 G does not show a divergence behavior but saturates at low temperatures with decreasing temperature.[33] This behavior of λ means that a magnetic phase transition does not occur, and that a finite frequency of spin fluctuation remains down to $T=0$. Thus, we conclude that the ground state for $x\geq 0.51$ is a paramagnetic state. Speculatively, it is expected that an ordered state will appear when the remained spin gap collapses by magnetic fields and by pressures, as reported in mixed systems and in pure parent systems by magnetization, neutron-scattering, specific-heat measurements.[10,12,28,47–50] As shown in Figs. 2 and 3, T_{peak} decreases and the peak structure vanishes with decreasing the concentration of x, which indicate the suppression of the slowing down of Cu 3d spin fluctuations and the disappearance of the soft mode.

The change in magnetic states across $x=0.51$ is considered below. The ground state for $x>0.51$ is a paramagnetic state which has spin fluctuations toward a possible magnetic phase transition as discussed above. If an ordered state, for which the paramagnetic state in $x>0.51$ is headed, is the same with the ground state in $x<0.51$, a magnetic state will continuously change pass through $x=0.51$ with decreasing x and λ in longitudinal fields will show a divergent increase at low temperatures around $x=0.51$ with the development of the soft mode toward the magnetic phase transition. However, the temperature change in λ becomes to be less significant with decreasing x and λ in 3950 G is almost constant below 3 K in $x=0.51$. In other words, the frequency spectrum becomes to be nearly white in the case of $x=0.51$, because at lower temperatures, λ in constant fields saturates and shows no peak structure down to 100 G.[33] Therefore, it is suggested that the ground state in $x<0.51$ is different from the magnetic state for which the paramagnetic ground state in $x>0.51$ is headed.

Hereafter, we discuss the spin dynamics in the case of $x=0.40$, where the ground state is expected to be in the Bose-glass phase from results of magnetization, neutron-scattering, specific-heat, and ESR measurements.[29–32] As mentioned above, the peak temperature T_{peak} in 3950 G decreases and the peak structure vanishes in $x=0.51$ with decreasing the concentration of x (Fig. 3), which suggests that the ground state for $x>0.51$ is different from that for $x<0.51$. In addition, as shown in Fig. 9, λ in lower magnetic fields below 100 G does not saturates and increases down to 0.28 K, and this behavior is different from the case for the region of $x>0.51$ where λ in constant fields saturates with decreasing temperature.[33] This result, which suggests that the ground state in $x=0.40$ is a different magnetic state compared with a paramagnetic state in the region of $x>0.51$, is consistent with the suggestion from the result in Fig. 3. The increase in λ in $x=0.40$ suggests the critical divergence toward a magnetic phase transition at a quite lower temperature, because it means that the Cu 3d spins-fluctuation frequency is slowing down, and that a frequency of the maximum intensity in the frequency spectrum continues decreasing down to 0.28 K with decreasing temperature. Thus, the increase in λ in $x=0.40$ is possibly interpreted as a precursor of the transition to the reported Bose-glass phase. These results suggest the possibility of the existence of the quantum critical point from the Bose-glass phase to a paramagnetic phase which has spin fluctuations toward an ordered phase except the Bose-glass phase around $x=0.51$.

In the last, we speculate an excited state of the Bose-glass phase in connection with the temperature change in λ in lower magnetic fields which shows a shoulderlike shape as shown in Fig. 9. Depending how we look at the shoulderlike shape, it seems that the part of shoulder is a remained peak of the soft mode in the temperature change in λ observed in higher magnetic fields above 1000 G as shown in Fig. 7, and that another part of increased λ at lower temperatures in lower fields corresponds to another mode. The behavior in the former part is similar to the case in $x=0.60$. If this diagnosis is correct, the two soft modes are superposed, one is a mode toward the ordered state and another is a mode toward the Bose-glass state. It seems to be consistent with the reported field-induced magnetic phase transition in the lower concentration of x,[28–32] because in the mixed system, a finite spin gap remains and the ordered state appears when the spin gap collapses. The detailed LF-μSR investigations of the Bose-glass phase down to the dilution refrigerator temperature is in progress.

V. SUMMARY

Zero- and longitudinal-field muon spin-relaxation measurements were carried out on $Tl_{1-x}K_xCuCl_3$ in the wide range of x from 0.40 to 0.65 in order to investigate the randomness effect for ground states. In the region of $x\geq 0.53$, the temperature change in the muon spin-relaxation rate λ in longitudinal fields shows a peak structure, which suggests the soft mode of spin waves, although a finite frequency of spin fluctuations remains down to $T=0$, i.e., the ground state is paramagnetic. With decreasing x, the temperature giving the peak decreases, and the peak structure vanishes in $x=0.51$. In the case of $x=0.40$, however, the temperature change in λ in longitudinal fields shows the different behavior, and λ in lower fields below 100 G increases down to 0.28 K. The increase in λ is possibly interpreted as a precursor of the transition to the reported Bose-glass phase. These results suggest that around $x=0.51$, there is a possibility of the existence of the quantum critical point from the Bose-glass phase to a paramagnetic phase which has spin fluctuations toward an ordered phase except the Bose-glass phase.

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[50] By μSR measurements, which is a local probe, it is hard to distinguish between a magnetically ordered state and a randomly

spin frozen state when the internal magnetic field at muon sites exceeds the time-resolution limit of detection of muon spin rotation. However, in this spin-gap system, it has been confirmed that the collapse of the spin gap leads to the appearance of a magnetically ordered state by magnetization, neutron-scattering, and specific-heat measurements. Thus, we use the word "ordered state" and should emphasize that it is not suggested by μSR measurements but by other macroscopic probes. Indeed, no evidence for a static field is seen in μSR time spectra in this study.