Magnetic Behavior of the 2-Leg and 3-Leg Spin Ladder Cuprates $Sr_{n-1}Cu_{n+1}O_{2n}$

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We have performed muon spin relaxation measurements of the spin-ladder cuprates $Sr_{n-1}Cu_{n+1}O_{2n}$ ($n = 3,5$). The 3-leg ladder system ($n = 5$) shows static magnetic order at $T \sim 52$ K, which is between the ordering temperatures of the one-dimensional chain system Sr_2CuO_3 ($T_N \sim 5$ K) and of the two-dimensional layer system $Ca_{0.86}Sr_{0.14}CuO_2$ ($T_N \sim 540$ K). The 2-leg ladder system ($n = 3$) does not show any magnetic order down to 20 mK, in agreement with the theoretical prediction of a nonmagnetic ground state with a spin gap for even-leg-number ladder systems.

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Recently, a characteristic series of structures known as "spin ladders" was found in the high pressure phase of $Sr_{n-1}Cu_{n+1}O_{2n}$ ($n = 3,5,\ldots$) [1]. The lattice structure is composed of $(n+1)/2$-leg spin ladders, namely, strips of CuO_2 square lattice which have $(n+1)/2$ Cu^{2+} ions across their width (Fig. 1). Each Cu^{2+} ion has spin $S = 1/2$ with antiferromagnetic couplings within a ladder (strength J), both in the "rung" and the "leg" directions. In the two directions, differences of the coupling strengths are presumably small, because the Cu-O-Cu bond lengths are almost equal for both directions [1]. Neighboring ladders are displaced by half the lattice constant, making the interladder interactions small and ferromagnetic ($-J'$; $J'/J \approx 0.1-0.2$) [2,3]. The spins at the edge of the ladders are frustrated because of the triangular structure constituent from two ferromagnetic interactions ($-J'$) and one antiferromagnetic interaction (J) [2,3].

Theoretical investigations of an isolated 2-leg spin ladder [2–4] predict a ground state with a many-body singlet nature: The ground state energy is separated from the triplet first excited state with a large "spin gap" ($\approx 0.5J$). The spin gap is explained as an effect of singlet pair formations on the rungs of the ladder [3,4]. If this is the case, magnetic order will be absent in the 2-leg ladder system. The 3-leg ladder system, on the other hand, may have a magnetically ordered ground state, because singlet pair formation on its rungs is not possible [3].

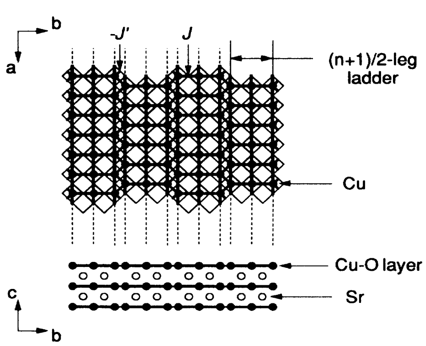

FIG. 1. The "spin ladder" structure. Oxygen ions locate at each corner of the squares. The figure shows the 3-leg ladder structure.

These theoretical predictions of the magnetic behavior of the spin ladder systems motivated our experimental investigations of $Sr_{n-1}Cu_{n+1}O_{2n}$, which is a 2-leg ladder system for $n = 3$, and a 3-leg ladder system for $n = 5$. Previous experiments on $Sr_{n-1}Cu_{n+1}O_{2n}$ have investigated magnetic susceptibility [5] and ^{63}Cu NMR [6]. In the 2-leg ladder system ($n = 3$), the temperature dependence of the susceptibility and the T_1 relaxation rate are well described by thermal excitations over a gap, which may correspond to the spin gap between the nonmagnetic ground state and magnetic excited states. The 3-leg ladder system ($n = 5$), in contrast, has a finite susceptibility in the $T \rightarrow 0$ limit, demonstrating that the ground state of this system can respond to the external magnetic field. The ground state may, therefore, be magnetically ordered. In the 3-leg ladder system, the T_1 relaxation rate of ^{63}Cu nuclear moment is so large that it is hardly measurable with the NMR method. This result implies the existence of strong magnetic correlations in the 3-leg system [6].

Muon spin relaxation (μSR) is a NMR-like local magnetic probe, but with much higher timing resolution ($\lesssim 1$ ns) than typical NMR methods ($\sim 1\ \mu$s). Consequently, μSR is a powerful technique to study the 3-leg ladder system, in which the NMR relaxation rate is beyond its time resolution. Another advantage of μSR is its high sensitivity to small and/or dilute static moments. μSR is the most sensitive technique to investigate the absence of static magnetic order in the ground state of the 2-leg ladder system.

Polycrystalline specimens of the spin ladder cuprates ($Sr_{n-1}Cu_{n+1}O_{2n}$; $n = 3,5$) were prepared at the Institute for Chemical Research, Kyoto University, using a cubic anvil-type high pressure apparatus [7]. Powder x-ray analysis of our samples showed the stoichiometric ladder structure, except for small amounts ($\sim$10 Cu at.%) of the

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CuO impurity phase [5]. Since CuO is an antiferromagnet ($T_N \sim 230$ K [8]), an impurity cluster should not have a long range effect on the muons which did *not* land within the cluster. Thus, in our μSR measurements, ~90% of the signal amplitude comes from the pure ladder structure. We performed μSR measurements at the M15 and M13 beam lines, at TRIUMF (Vancouver). Longitudinally polarized muons (spin || beam axis) were implanted in the polycrystalline pellets, which had been placed on a 99.99% silver backing plate. We evaluated the time evolution of muon spins, using the conventional μSR technique [9–11].

Figure 2(a) shows typical zero-field μSR spectra in the 3-leg ladder system. Below ~55 K, spin relaxation was observed, which suggests magnetic ordering of the system. The signal amplitude was large enough to conclude that it comes from the ladder structure, rather than from the CuO impurity phase. The time spectra taken below 30 K fit well to the Gaussian Kubo-Toyabe function, which is appropriate to randomly oriented, frozen dense spin systems [10]. In this kind of frozen spin system, the sum of dipolar fields from the frozen moments makes a Gaussian field distribution (width Δ) at the muon site. The corresponding muon spin relaxation initially decays as a Gaussian with the characteristic Gaussian relaxation

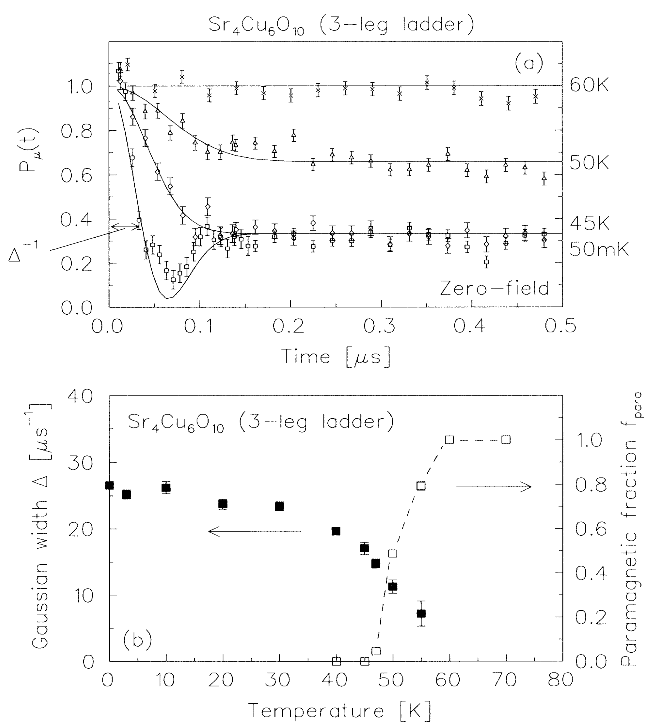

FIG. 2. (a) Zero-field μSR spectra in the 3-leg ladder system. The solid lines are the fit with the model function, Eq. (1). (b) Temperature dependence of the Gaussian field-distribution width (Δ) and the paramagnetic volume fraction (f_{para}). The broken line is a guide to the eye.

time (Δ^{-1}). In case of static order in polycrystalline pellets, the muon spin relaxation at large times comes back up to 1/3 of the total amplitude, because 1/3 of the internal fields are parallel to the initial muon polarization, and hence do not contribute to the depolarization. This nonrelaxing amplitude of 1/3 is called the "1/3 component" in the static Gaussian Kubo-Toyabe function and is a hallmark of static order. The static Gaussian Kubo-Toyabe function shows a "dip" between the initial Gaussian decay and the 1/3 component, which is characteristic of static order with a single Gaussian field width (Δ).

The spectra near the transition temperature [Fig. 2(a)] behave as if there is a distribution of ordering temperatures; (1) the relaxation amplitude ($\propto$ ordered volume fraction) decreases as temperature increases, and (2) the dip in the spectra disappears above ~40 K, although the relaxation is static, as shown by the decay to the 1/3 component. The lack of such a dip in the static relaxation suggests a distribution of field widths, which would result from a distribution in the ordering temperatures. We analyzed the μSR spectra with a functional form of

$$P_\mu(t) = f_{\text{para}} + (1 - f_{\text{para}})G_{\text{static}}(t;\Delta), \qquad (1)$$

where f_{para} is the paramagnetic volume fraction in the sample and $G_{\text{static}}(t;\Delta)$ is the static Gaussian Kubo-Toyabe function at $T \leq 30$ K or a static Gaussian function $\{1/3 + 2/3\exp[-(\Delta t)^2]\}$ at $T \geq 40$ K. The parameter Δ is the Gaussian field distribution width, which is proportional to the size of the static component of Cu moments. At $T \geq 40$ K, Δ represents an "average" of the field widths at the muon sites. We show the temperature dependence of f_{para} and Δ in Fig. 2(b). The ordering temperature is around 52 K with a distribution of ± 5 K, as deduced from the temperature dependence of f_{para}.

The absence of muon spin precession in Fig. 2(a) suggests that the magnetic order of the 3-leg ladder system is a random freezing of moments, rather than true Néel order. A possible source for the randomness is the frustration at the edge of the ladder. Another characteristic of the magnetic order is that critical slowing down of the Cu moments was hardly observable around the transition temperatures; at 50 and 55 K, the muon spin relaxation rate at long times (0.5–9 μs, not shown) was less than 0.02 μs^{-1}, which corresponds to a fast field fluctuation rate of $\nu \sim 3 \times 10^4$ MHz, as estimated by the T_1 relaxation theory in paramagnets [11]. At 45 K, the muon spin relaxation exhibited the 1/3 component [Fig. 2(a)], the relaxation of which was very slow (~0.04 μs^{-1}). This result shows that the Gaussian relaxation at 45 K is already almost static ($\nu \sim 0.06$ MHz, as estimated from Kubo-Toyabe theory with slow dynamics [10]). This "abrupt" onset of the static magnetic order may reflect the low dimensionality of the system, because similar behavior has been observed in a one-dimensional antiferromagnet Ca_2CuO_3 [12] and a spin-density-wave system $(TMTSF)_2$-PF_6 [13].

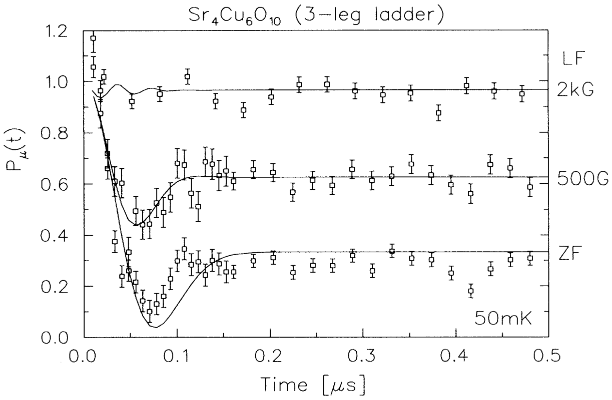

FIG. 3. Longitudinal field (LF) decoupling measurements on the 3-leg ladder system at 50 mK. The solid lines are the fit with static Kubo-Toyabe functions.

Figure 3 shows μSR spectra from longitudinal field (LF) decoupling measurements, which investigate spin fluctuations by applying the external magnetic field parallel to the initial muon polarization [10]. In the case of static order, the longitudinal field increases the amplitude of the $1/3$ component, without changing the initial decay rate. This is because the longitudinal field is parallel to the initial muon polarization, and does not affect the distribution width (Δ) of the random fields. In the static case, the typical longitudinal field which decouples the muon relaxation is in the order of the field distribution width (Δ), which may be estimated from the relaxation rate in a zero-field measurement. In the case of dynamical muon spin relaxation, on the other hand, the longitudinal field suppresses the muon relaxation rate rather than changing the relaxation amplitude [11]. In this case, the magnitude of the decoupling field is much larger than the zero-field relaxation rate suggests. One can thus distinguish static and dynamic muon relaxation from the response to longitudinal fields.

As shown in Fig. 3, the longitudinal field measurements on the 3-leg ladder system clearly display the static nature of the muon relaxation, which is consistent with the previously observed "1/3 component" and the "dip" in the zero-field measurements. The recoveries of the muon spin polarization in external fields are well described by the static Gaussian Kubo-Toyabe theory (solid lines in Fig. 3). This series of longitudinal field measurements at 50 mK indicates that the ground state of the 3-leg ladder system is a conventional static ordered state, rather than the "spin liquid" state, which has been proposed for the 2-leg ladder system both theoretically and experimentally [2–6]. The conventional μSR relaxation in the 3-leg ladder system also contrasts with our μSR results from other spin liquid materials, such as the frustrated Kagomé lattice system $SrCr_zGa_{12-z}O_{19}$ [14] and the charge-doped Haldane system $(Y_{2-x}Ca_x)BaNiO_5$ [15].

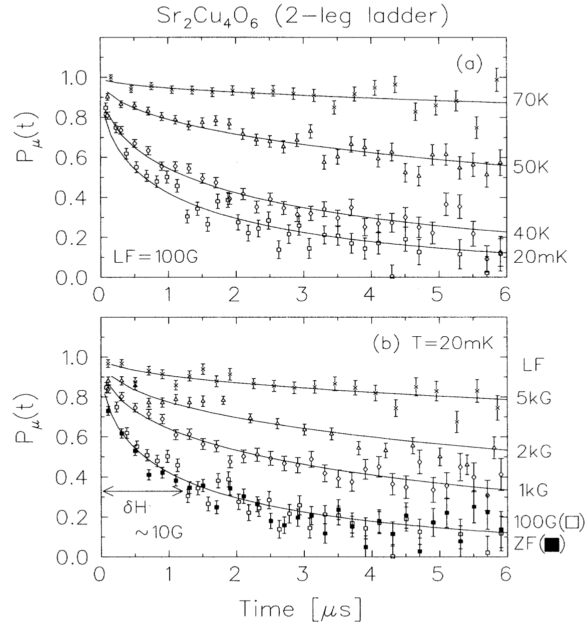

FIG. 4. (a) LF = 100 G μSR spectra in the 2-leg ladder system. (b) Longitudinal field decoupling measurements at 20 mK. For both panels the solid lines are fits with a square-root exponential function. Note that the horizontal scale is ~10 times larger than that in Figs. 2(a) and 3.

The magnetic behavior of the 2-leg ladder system differs remarkably from that of the 3-leg system. In Fig. 4(a), we show typical μSR spectra in the 2-leg ladder system. Note that Fig. 4 has a horizontal scale about 10 times larger than that for Figs. 2(a) and 3. In the 2-leg system, weak relaxation below ~50 K is apparent. However, as we discuss below, the source of this relaxation is fluctuating rather than static internal fields. In Fig. 4(b), we show LF decoupling measurements, which demonstrate the dynamical nature of the spectra: If the slow relaxation in the zero-field measurement were due to a static field distribution, the μSR spectrum would be flattened in a LF ~ 100 G. This decoupling field has been estimated from the relaxation rate in zero field. As shown in Fig. 4(b), the relaxation persists in longitudinal fields up to 2 kG, proving that this slow relaxation is a purely T_1-like dynamical one.

We analyzed the muon spin relaxation in the 2-leg ladder system, using a square-root exponential function $[P_\mu(t) = \exp(-\sqrt{\lambda t})]$, which is appropriate for dilute fluctuating moments [11]. The temperature dependence of the relaxation rate (λ) is shown in Fig. 5. The increase of λ at temperatures down to ~40 K indicates a slowing down of field fluctuations, yet the fluctuations never die even in the millikelvin regime. The general T and LF dependence of λ, as well as the square-root behavior of the spectrum itself, is very similar to the results in a Haldane system, which has a many-body singlet ground

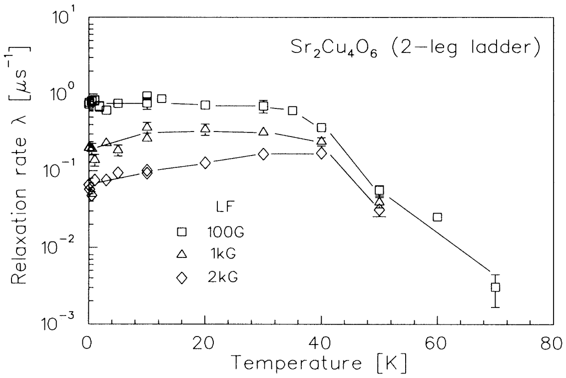

FIG. 5. Temperature dependence of muon spin relaxation rate (λ) in the 2-leg ladder system. The solid lines are a guide for the eyes.

state [16]. In our previous measurements on a Haldane material (Y_2BaNiO_5), the T and LF dependence of λ was ascribed to unpaired spins which originate from native chain breaks [15,17]. Considering that our 2-leg ladder sample has a small Curie component in its susceptibility [5], the observed slow dynamic muon spin relaxation is probably due to the imperfections.

In addition to the spin ladder structures, the Sr(Ca)-Cu-O series of compounds has a variety of stable composites, such as the "infinite chain" (Sr_2CuO_3) [18] and "infinite layer" system ($Ca_{0.86}Sr_{0.14}CuO_2$; $n \to \infty$ structure of the spin-ladder series) [19]. We summarize their structure and magnetic behavior in Table I. The ordering temperature of the 3-leg ladder system is between those of the chain system (Sr_2CuO_3) and the layer system ($Ca_{0.86}Sr_{0.14}CuO_2$). Suppression of T_N in the chain system has been explained as an effect of the low dimensionality [18]; the intermediate ordering temperature for the 3-leg system is consistent with this idea, as the 3-leg ladder structure is the smallest extension of the one-dimensional chain toward two dimensionality.

TABLE I. Structure and magnetic behavior of Sr(Ca)-Cu-O compounds.

Compound	Structure	Magnetic behavior
Sr_2CuO_3	CuO chain (1D)	$T_N \sim 5$ K[a]
$Sr_2Cu_6O_{10}$	3-leg ladder	$T \sim 52 \pm 5$ K[b]
$Ca_{0.86}Sr_{0.14}CuO_2$	CuO_2 plain (2D)	$T_N \sim 540$ K[c]

[a]Obtained by μSR [19].
[b]Obtained by μSR (this work)
[c]Obtained by neutron scattering [20] and μSR [19].

In conclusion, using μSR, we observed static magnetic order for the 3-leg ladder system at $T \sim 52$ K, while no magnetic order was observed in the 2-leg ladder system down to 20 mK. The slow and dynamic relaxation in the 2-leg ladder system originates from dilute unpaired spins, which may be associated with defects in the sample. Our results confirmed theoretical expectations for the magnetic behavior of the 2- and 3-leg ladder systems in $Sr_{n-1}Cu_{n+1}O_{2n}$ compounds.

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