Spin correlation and spin gap in quasi-one-dimensional spin-1/2 cuprate oxides: A ^{63}Cu NMR study

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The Cu-NMR nuclear relaxation rate, $^{63}(1/T_1)$ and Gaussian spin-echo decay rate, $^{63}(1/T_{2G})$ have been measured in order to investigate the low-energy spin dynamics in the quasi-one-dimensional (1D) spin-1/2 antiferromagnetic (AF) cuprates such as Ca_2CuO_3 with single chain, $SrCuO_2$ at ambient pressure (AP) with zigzag chain coupled by weak ferromagnetic exchange interaction, $SrCu_2O_3$ and $Sr_2Cu_3O_5$ comprising of two- and three-leg ladder, respectively with isotropic AF exchange interaction along and between chains. In Ca_2CuO_3, $1/T_1$ stays constantly close to the 3D magnetic ordering temperature, $T_N \sim 10$ K, dominated by dissipation of overdamped spinons gas as shown theoretically by Sachdev. By contrast, in AP-$SrCuO_2$, $1/T_1$ below around 100 K decreases markedly down to the 3D magnetic ordering temperature, $T_N \sim 3$ K, providing a signature that the frustrated ferromagnetic interaction between CuO zigzag chains makes the spin gap open. In the spin-1/2 ladder compounds of $SrCu_2O_3$ and $Sr_2Cu_3O_5$ consisting of two- and three-leg, respectively, the former was demonstrated in the literatures to have the spin gap amounting to a half of AF exchange constant, $J/2$ where $J=1300$ K. For the latter, on the other hand, the magnetic coherence length, $\xi(T)$ grows up according to $\xi(T) \sim 1/(T-\theta)$ and as a result, the magnetic ordering emerges around 50–60 K accompanying the divergence of $1/T_1$. In the compounds composed of single and triple chains, it is shown that there is no analog of renormalized classical region identified in the 2D spin-1/2 cuprate such as La_2CuO_4, but the spin dynamics is in the quantum critical regime over wide T range due to the finite size of chains, even though the 3D magnetic ordering takes place at very low temperature. This is considered because the spin correlators have a power-law decay only along the chain, but between chains is limited to the number of chains in contrast to the 2D square lattice. For the compounds with two chains, the exchange coupling between chains leads to the formation of spin gap phase regardless of whether its sign is antiferromagnetic or ferromagnetic. A result on doping into the two-leg ladder, $SrCu_2O_3$ is argued in terms of the localization effect of carriers.

I. INTRODUCTION

Since the discovery of the high-temperature superconducting cuprate, the subject of spin-1/2 quantum antiferromagnets in low-dimensional systems has been a very active field, since the parent compounds of high-T_c cuprates such as La_2CuO_4 and $YBa_2Cu_3O_6$ were the spin-1/2 two-dimensional (2D) antiferromagnetic (AF) Heisenberg systems and Mott insulators.[1] The spin dynamics of La_2CuO_4 was extensively studied from theoretical and experimental points of view. Its ground state is a Néel order at $T=0$ and a very weak interlayer coupling enhances the 3D Néel temperature to ~ 300 K. Chakravarty *et al.*[2] showed that in the paramagnetic state of the 2D Heisenberg system, there are two regimes of spin correlation; namely, one is the renormalized classical regime where the correlation length $\xi(T)$ develops as $\exp(2\pi\rho_s/T)$ for $T<\rho_s$, and the other is the quantum critical regime where $\xi(T) \sim (T+\theta)^{-1}$ for $T>\rho_s$. Here, the zero temperature spin stiffness constant ρ_s is given by $\rho_s = 0.2J \sim 300$ K in La_2CuO_4. As a matter of fact, the theoretical description has well explained the spin dynamics investigated by the Cu nuclear quadrupole resonance (NQR) (Refs. 3,4) and inelastic neutron scattering experiments.[5]

Another novel class of spin-1/2 antiferromagnets is the 1D system, which is known to be generally critical in contrast to the 2D system possessing a Néel order at $T=0$. Focusing on the case of a finite range of couplings where the ground state is critical and hence in a lower-T region than J, the spin correlator has an analog of the quantum-critical regime of the 2D system. Sachdev has shown that the nuclear relaxation is dominated by dissipation from a gas of thermally activated, overdamped spinons and has led to the following low-T results of the nuclear-spin lattice relaxation rate $1/T_1$ and Gaussian spin-echo decay rate $1/T_{2G}$:[6]

$$\frac{1}{T_1} = A_\perp^2(\pi/a)\frac{\pi D}{\hbar^2 c}, \quad \frac{1}{T_{2G}} = A_\parallel^2(\pi/a)\frac{D}{2\hbar^2 c}\left(\frac{\hbar c}{k_B T a}\right)^{1/2} I, \tag{1}$$

where a is the lattice spacing and $A_\parallel$ ($A_\perp$) is the hyperfine coupling constant parallel (perpendicular) to the magnetic field. c is the $T=0$ spinon velocity, and D is a nonuniversal spin diffusion constant dependent on the choice of the microscopic exchange constants. These formulas were derived in such a context that since spinon excitations interact strongly with other thermally excited spinons, resulting in a very short lifetime, the NMR relaxation is dominated by these overdamped spinons. Upon considering the ratio of the above two rates (T_1T/T_{2G}), it was noted that the unknown

prefactor cancels and numerical factor is not important as far as the T dependence is concerned. Different from the (T_1T/T_{2G}) being constant for the 2D quantum critical regime, it is remarkable that its ratio in the 1D system is proportional to $\sqrt{T}$. Apparently, theoretical understandings of the ground state properties are in good shape in describing either 1D or 2D spin-1/2 antiferromagnets, and hence systematic experiments are desired.

The related low-dimensional system is the Heisenberg AF ladder comprising spin-1/2 coupled chains. Johnston *et al.* suggested that the spin-1/2 ladder model might describe the magnetic properties of $(VO)_2P_2O_7$.[7] Although they analyzed their susceptibility data based on the dimerized Heisenberg chain model, Dagotto *et al.* applied extensive theoretical approaches to determine the ground state energy and the low-lying excitation spectrum of the spin-1/2 ladder and found a nonzero spin gap for all interchain AF couplings, $J_\perp > 0$.[8] Most remarkably, based on such a ladder model that consists of two t-J chains with t'-J' couplings forming the rungs between chains, they showed that for an insulating half filled ladder, the spin gap depends on J'/J, whereas for a doped system with $J \sim J'$, the added holes tend to make localized pairs at adjacent sites. In the scheme of numerical studies, this model at finite doping was argued to involve the possibility of either superconducting correlation or charge-density-wave (CDW) long range order at zero temperature.

Meanwhile, Rice *et al.*[9] theoretically investigated the nature of the ground state of a homologous series of cuprates compounds, $Sr_{n-1}Cu_{n+1}O_{2n}$ $(n=3,5,7,9,11,\ldots)$ which forms the spin-1/2 ladders with $J \sim J'$ comprising an even number of legs $(n=3,7,11,\ldots)$ and odd number of legs $(n=5,9,13,\ldots)$. They concluded that the compounds with $n=3,7,11,\ldots$, i.e., two, four, and six legs, should be a frustrated quantum spin liquid with a spin gap, whereas the phases with $n=5,9,13,\ldots$, i.e., three, five, and seven legs, should have a gapless ground state and hence a weak interlayer coupling leads to the 3D magnetic ordering at low T. Most remarkably, it was predicted that lightly doping into the former system leads to the onset of singlet superconductivity, maintaining the spin gap.[10] These novel magnetic properties of the undoped ladder systems have been evidenced by a subsequent neutron scattering experiment on $(VO)_2P_2O_7$[11] and susceptibility and NMR relaxation experiments on $SrCu_2O_3$ $(n=3)$ and $Sr_2Cu_3O_5$ $(n=5)$ with two and three legs, respectively.[12,13] In particular, the latter systems have provided an interesting opportunity to systematically investigate the nature of the spin correlation depending on the number of coupled chains.

In the light of experimental and theoretical efforts to look for new parent compounds for the novel superconductivity, it is important to clarify the nature of the spin correlation of quasi-1D systems and to be compared with that in the 2D system. The NMR experiments have been proved to be a powerful tool in studying the spin dynamics of the 2D antiferromagnetic and doped superconducting cuprates. In this paper, we report a systematic NMR relaxation study of the low-lying spin dynamics in quasi-1D spin-1/2 systems such as Ca_2CuO_3 with linear chains, $SrCuO_2$ at ambient pressure (AP) with zigzag chains, $SrCu_2O_3$ with two-leg ladders, $Sr_2Cu_3O_5$ with three-leg ladders and La- (electron-) doped $SrCu_2O_3$.

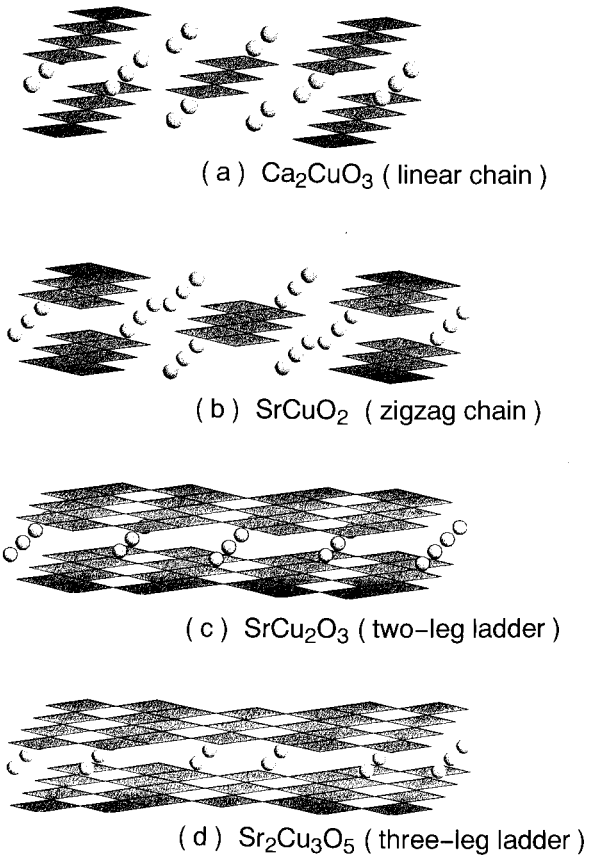

FIG. 1. Crystal structures of ambient pressure (AP) phases Ca_2CuO_3 (a) and $SrCuO_2$ (b) and high-pressure (HP) phases $SrCu_2O_3$ $(n=3)$ (c) and $Sr_2Cu_3O_5$ $(n=5)$ (d) from top to down. Squares and spheres represent CuO_4 and Sr or Ca atoms.

Figure 1 indicates the crystal structures Ca_2CuO_3 (a), $SrCuO_2$ (b), $SrCu_2O_3$ (c), and $Sr_2Cu_3O_5$ (d) from top to bottom. In Ca_2CuO_3, the CuO_4 square forms a linear Cu-O-Cu bond aligned along the b axis, while in $SrCuO_2$, the interactions via 90° Cu-O-Cu bonds between two linear Cu-O-Cu chains must be much weaker than along linear chains and are even ferromagnetic and hence frustrated. In $SrCu_2O_3$ $(n=3)$, the zigzag chains in AP $SrCuO_2$ are coupled with each other by the 180° Cu-O-Cu bonds in the same plane which is stacked alternatively with Sr sheets along the c axis. Thus each linear CuO bond in the zigzag chains forms two-leg ladders which are coupled via the 180° Cu-O-Cu bond with one another. In $Sr_2Cu_3O_5$ $(n=5)$, one more Cu-O-Cu linear bond is inserted between two legs, forming a three-leg ladder. With an increasing number of n, i.e., number of legs, a dimensional crossover to the 2D system is expected since the $n=\infty$; e.g., the infinite layer compound is nothing but the 2D square lattice, which changes to the high-T_c superconductor by doping either holes or electrons.

II. EXPERIMENTAL PROCEDURES

The synthesizing process of the samples was described elsewhere.[14] In the NMR measurements, the Cu NMR spec-

trum was obtained by sweeping external field using the superconducting magnet (12 T at 4.2 K). T_1 of ^{63}Cu was measured by the saturation recovery method on the $(1/2 \leftrightarrow -1/2)$ central transition of the quadrupole split NMR spectrum in a magnetic field of 11 T. The nuclear relaxation function $R(t)$ for the central transition $(1/2 \leftrightarrow -1/2)$ is given by[15]

$$R(t)=[M_0-M(t)]/M_0=0.1\ \exp(-t/T_1) + 0.9\ \exp(-6t/T_1), \qquad (2)$$

where $M_z(t)$ is the nuclear magnetization for the central transition at time t after saturation pulses. The experimental results were fitted well by the above relaxation function. The spin-echo decay time T_{2G} was measured by changing time interval (τ) between first and second pulses. The experimental spin-echo decay data were fitted to the function[16]

$$M(2\tau)=M_0 \exp\left\{-\frac{1}{2}\left(\frac{2\tau}{T_{2G}}\right)^2-\frac{2\tau}{T_{2L}}\right\}. \qquad (3)$$

$1/T_{2G}$ is the spin-echo decay rate associated with the indirect coupling of the nuclear spins mediated by the electron spins, i.e., AF spin fluctuations argued extensively in the literature,[17] and $1/T_{2L}$ is the decay rate associated with the nuclear-spin-lattice relaxation (T_1) process.[18]

T_{2G} could be reliably determined provided that the linewidth of the Cu NMR spectrum is narrow enough to be saturated by an exciting rf field, since nuclear spins are needed to be uniformly flipped by the π pulse. The strong rf field H_1 with about 150 Oe allowed us to determine T_{2G} by virtue of the narrower linewidth of the Cu NMR spectrum than $H_1=150$ Oe.

III. EXPERIMENTAL RESULTS

A. Ca_2CuO_3 with a linear chain

The sample was not oriented even in a magnetic field of 11 T. The broad NMR spectrum of unoriented powder prevents the precise measurement of T_{2G}. By analyzing the powdered pattern of the Cu NMR spectrum, the Knight shifts parallel $(K_\parallel)$ and perpendicular $(K_\perp)$ to the c axis were estimated to be $K_\parallel=0.76(\%)$, $K_\perp=0.19(\%)$, which do not show any T dependence in a T range of 20–300 K. Upon lowering the temperature, the Cu NMR spectrum becomes broader below 50 K, and disappears below 12 K. The cause for the broadening of the NMR spectrum below 50 K is considered to be due to the dipole field from impurity spins, whereas the disappearance of the Cu NMR signal is due to the onset of magnetic order around 10 K.[19] $1/T_1$ was measured between 12 and 300 K at the peak of the NMR spectrum arising from grains of which the local symmetry axis is directed perpendicular to the magnetic field. The relaxation function of the magnetization recovery after saturation pulses, $R(t)$, is well fitted by Eq. (2) with a single component of (T_1) above 20 K. As shown in Fig. 2, $1/T_1$ stays constantly in the entire T range. The nearly constant value of $1/T_1$ is almost the same as that in the three-leg ladder $Sr_2Cu_3O_5$ as presented later. Unfortunately, T_{2G} is difficult to measure because the powder is not oriented. Accordingly, the theoretical formula of T_{2G} derived in Eq. (1) by Sachdev cannot be addressed here, but the result of $1/T_1$ seems to be consistent with the prediction, dominated by thermally activated spinons. We further note that the T independence of $1/T_1$ contrasts with the specific heat result which is proportional to the temperature due to the presence of a pseudo Fermi surface of spinons.[20] In order to complete the study of the 1D AF spin-1/2 Heisen-

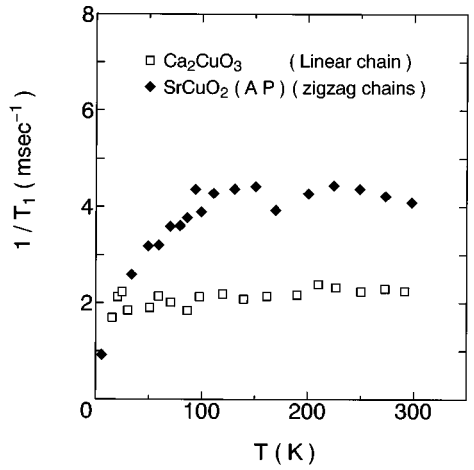

FIG. 2. T dependence of $1/T_1$ in Ca_2CuO_3 with linear chains and $SrCuO_2$ (AP) with zigzag chains.

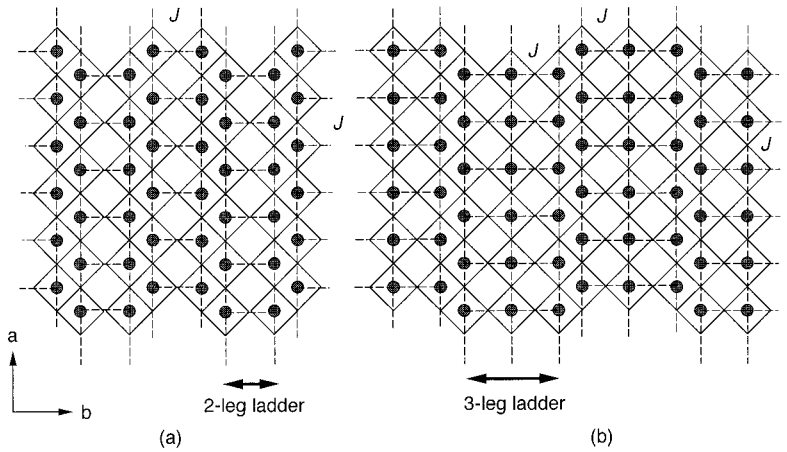

FIG. 3. $Cu_{n+1}O_{2n}$ plane in $SrCu_2O_3$ $(n=3)$ and $Sr_2Cu_3O_5$ $(n=5)$.

berg system, we plan to do the NMR experiment on the single crystal which enables us to measure T_{2G}.

B. $SrCuO_2$ with zigzag chain

The CuO_2 plane in AP $SrCuO_2$ consists of the one-dimensional zigzag chains as shown in Fig. 1(b). It should be noted that the interaction between zigzag chains must be weakly ferromagnetic, due to the 90° Cu-O-Cu, and frustrated. Since the powder was not oriented, the Cu NMR spectrum of the central transition $(1/2 \leftrightarrow -1/2)$ exhibited the typical powder pattern affected by the combined effects of the small nuclear quadrupole frequency ν_Q and the anisotropic Knight shift. From the anisotropic Cu NMR spectrum, $K_\parallel$ and $K_\perp$ are estimated to be $K_\parallel = 0.98(\%)$, $K_\perp = 0.19(\%)$, respectively, both of which are T independent as in Ca_2CuO_3. T_1 was measured at the main peak where the c axis of each grain is perpendicular to the external field. The experimental $R(t)$ is well fitted by Eq. (2) with a single component of T_1 from 50 K to 300 K. The T dependence of $1/T_1$ is also shown in Fig. 2 together with the results of Ca_2CuO_3. $1/T_1$ stays constantly above 100 K with a twice larger relaxation rate than that in Ca_2CuO_3, implying that the spinon velocity for the zigzag chain is somewhat lower than for the linear chain. Below 100 K, $1/T_1$, however, begins to decrease with lowering temperature. Although $1/T_1$ is not described by an activation type, it is considered that the low-energy weight of the spin correlation is significantly removed by weak ferromagnetic interactions between zigzag chains.

The Cu NMR spectrum disappears below 5 K, indicating that there exists some magnetic order as in Ca_2CuO_3. Actually 3D magnetic order was reported by a susceptibility measurement.[21] If the interchain interaction leading to the 3D Néel order were absent, the spin gap would be present. Since there are no data of T_{2G} which reflect to what extent the spin correlation is developed with temperature, we cannot give a definite conclusion about the spin gap.

C. Ladder with two- and three-leg $SrCu_2O_3$ and $Sr_2Cu_3O_5$

The recent progress of the high-pressure techniques made it possible to synthesize the ideal spin-1/2 ladder compounds which are suitable for theoretical study. In a homologous high-pressure phase, each $Cu_{n+1}O_{2n}$ $(n=3,5,7)$ sheet consists of a periodic CuO_2 plane separated by zigzag chains. The $Cu_{n+1}O_{2n}$ planes in $SrCu_2O_3$ and $Sr_2Cu_3O_5$ are illustrated in Fig. 3, respectively. As shown in Fig. 3, two- and three-leg ladders are coupled by way of 90° Cu-O-Cu bonds across the interface. The superexchange constant along the 90° Cu-O-Cu bond is considered to be one order of magnitude smaller than along the 180° Cu-O-Cu bonds, and to be even ferromagnetic.[22]

In a previous paper, we reported the Knight shift and the T_1 results of ^{63}Cu obtained by using the oriented powder of $SrCu_2O_3$. From an activated decrease of $^{63}(1/T_1)$ in a T range of 100–300 K, a spin gap Δ was evidenced to open with the value of 680 K.[13] Theoretically, since the spin gap is predicted to be the half of the intraladder AF exchange constant, $J/2$ and J should be 1300 K, judging from the similarity to the square CuO_2 plane,[23] the experimental value

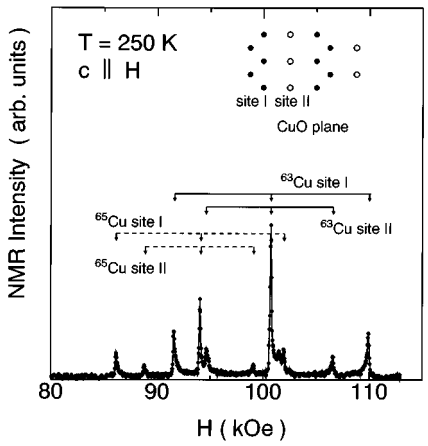

FIG. 4. Cu NMR spectrum of oriented powdered $Sr_2Cu_3O_5$ at 115.1 MHz, where the c axis of each grain is aligned parallel to the external field. The inset shows the coordination of two Cu sites.

$\Delta = 680$ K deduced from the T_1 experiment was approximately in agreement with the theoretical estimation, $J/2 = 650$ K, although the gap, 420 K, estimated by fitting the magnetic susceptibility data, is considerably smaller than the values mentioned above. The reason for the disagreement is not yet clear at the moment.

By contrast, the susceptibility data of the three-leg $Sr_2Cu_3O_5$ suggested to have no spin gap.[12] Then the disappearance of the NMR signal below around 100 K may be due to the appearance of 3D magnetic ordering since the magnetic coherence length develops with lowering temperature. Actually, a recent muon-spin-resonance (μSR) experiment reported that $Sr_2Cu_3O_5$ exhibited unusual magnetic ordering around 50–60 K.[24]

Here, we collect more extensive ^{63}Cu NMR results of $Sr_2Cu_3O_5$ and prove that the character of the magnetic correlation ξ is quite different between two- and three-leg ladders from the T_{2G} measurement. Figure 4 shows a Cu NMR spectrum of oriented powdered $Sr_2Cu_3O_5$ at 115.1 MHz. As denoted in the inset of Fig. 4, there are present two crystallographic inequivalent Cu sites; i.e., the coordination of site I

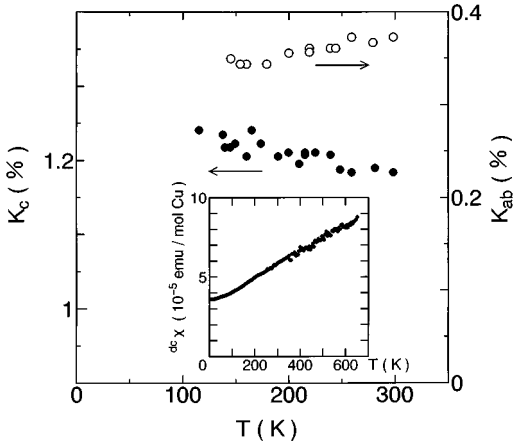

FIG. 5. T dependence of K_c and K_{ab} in $Sr_2Cu_3O_5$. The inset is the T dependence of χ_s where the Curie term is subtracted from the raw data.

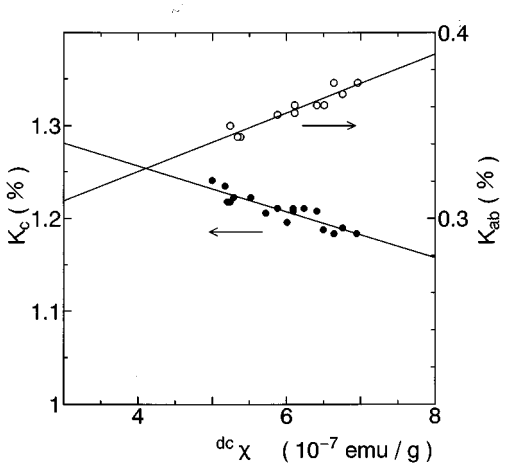

FIG. 6. K-χ plot in $Sr_2Cu_3O_5$.

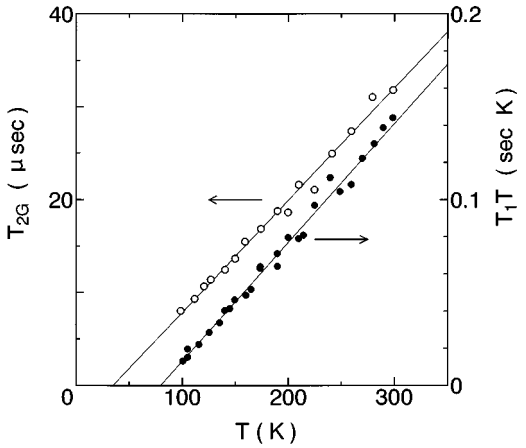

FIG. 8. T dependence of T_{2G} and T_1T in $Sr_2Cu_3O_5$.

is the same as that in $SrCu_2O_3$ and the coordination of site II is the same as that in the CuO_2 square lattice. Corresponding to two Cu sites, the Cu NMR spectra are well articulated by the electric quadrupole interaction as marked by arrows in the figure where each ^{63}Cu and ^{65}Cu ($I=3/2$) site consists of three peaks due to the first order nuclear quadrupole effect. From an analysis of the Cu NMR spectrum, the quadrupole frequency (ν_Q's) (MHz) of each Cu site is estimated as

$$\nu_a=-1.93, \quad \nu_b=-8.43, \quad \nu_c=10.36 \text{ (MHz) (site I)},$$

$$\nu_a=-2.79, \quad \nu_b=-3.85, \quad \nu_c=6.64 \text{ (MHz) (site II)}.$$

ν_Q of site I is almost the same as that in $SrCu_2O_3$,[13] and ν_Q of site II is nearly equal to that of CuO_2 plane in the infinite-layer compound.[25] This means that the local electronic state is determined by the local coordination of oxygens through the $3d$-$2p_\sigma$ covalent bonds.

The central lines arising from the $(1/2 \leftrightarrow -1/2)$ transition of Cu (I) and (II) sites are overlapped with one another, indicating that the magnetic shifts are almost the same even if ν_Q is different between two sites. The T dependences of the Knight shifts parallel, $K_c(T)$, and perpendicular, $K_{ab}(T)$, to the external field are collected in Fig. 5 together with the data of the magnetic susceptibility, where the Curie tail at low T was subtracted from the raw data, since the NMR shift shows no Curie tail. The corrected susceptibility $\chi_s(T)$ in $Sr_2Cu_3O_5$ gradually decreases with lowering temperature, in contrast to the susceptibility of $SrCu_2O_3$ that was fitted with the spin gap.

In order to estimate the hyperfine coupling constants parallel, A_c, and perpendicular, A_{ab}, to the c axis, K_c and K_{ab} are plotted with respect to the corrected susceptibility χ_s with temperature as an implicit parameter as indicated in Fig. 6. A linear relation holds in both components of the shift. A_c and A_{ab} are estimated from the slope of linear lines to be $A_c=-92.4$ (kOe/μ_B) and $A_{ab}=59$ (kOe/μ_B). It should be noted that these values are dominated by the on-site hyperfine values without an appreciable supertransferred hyperfine coupling B which was very large in the CuO_2 square lattice with a range of 40–100 kOe/μ_B.[26] This novel difference of the NMR hyperfine parameter was also reported for two-leg ladder $SrCu_2O_3$ in the previous paper.[13] Accordingly, the electronic structure of ladders is different from the CuO_2 square lattice in such a context that the Cu($3d$)-O($2p_\sigma$) covalent bond has no admixture with the $4s$ states in the four nearest neighbor Cu sites.

Figure 7 shows the T dependence of $1/T_1$ and $1/T_{2G}$ in $Sr_2Cu_3O_5$, whereas in Fig. 8, the T dependences of T_{2G} and

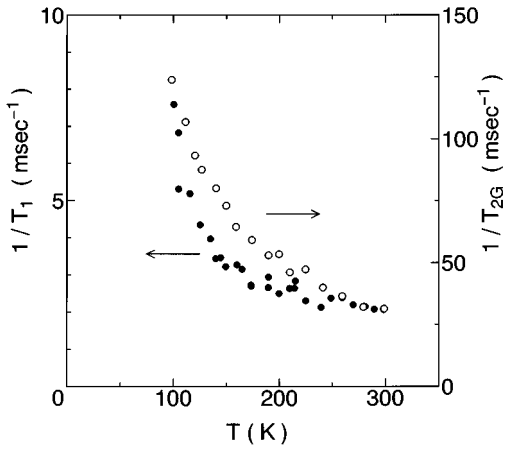

FIG. 7. T dependence of $1/T_1$ and $1/T_{2G}$ in $Sr_2Cu_3O_5$.

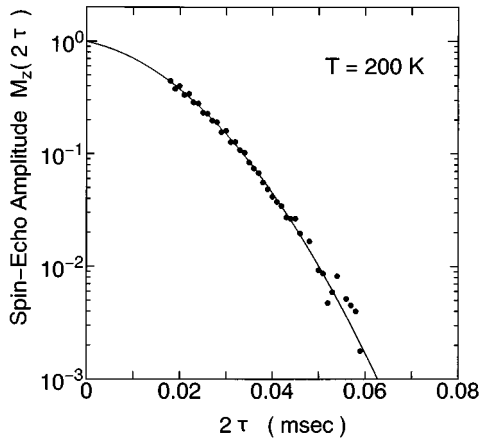

FIG. 9. Typical fitting of spin-echo decay in $Sr_2Cu_3O_5$ to estimate T_{2G}.

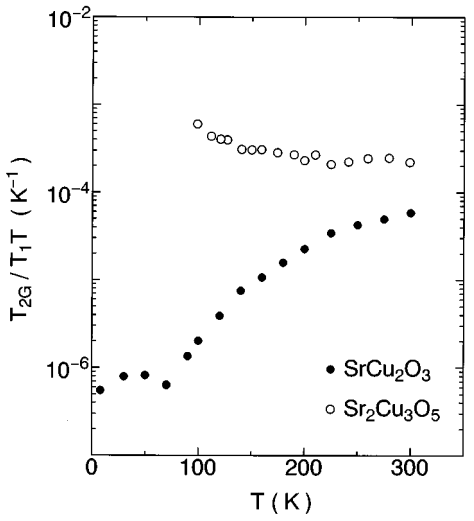

FIG. 10. T dependence of $T_{2G}/(T_1T)$ in $SrCu_2O_3$ ($n=3$) and $Sr_2Cu_3O_5$ ($n=5$).

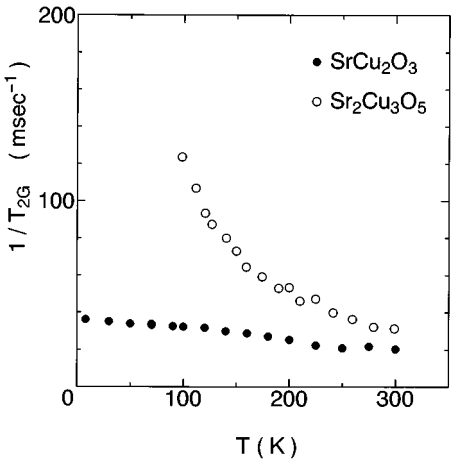

FIG. 11. T dependence of $1/T_{2G}$ in $SrCu_2O_3$ ($n=3$) and $Sr_2Cu_3O_5$ ($n=5$).

T_1T are plotted. Here, T_{2G} was deduced from the fitting with Eq. (3) as seen in Fig. 9. $1/T_1$ and $1/T_{2G}$ were uniquely and precisely determined, indicating that the magnetic properties are not different between two Cu sites, although the central transitions from two inequivalent Cu sites are overlapped with one another.

Below 100 K, we could not observe the Cu NMR signal due to the extremely short relaxation time, which suggests that the magnetic coherence length ξ rapidly grows up.

In general, $1/(T_1T)$ is given in terms of the imaginary part of the dynamical susceptibility as[27]

$$\frac{1}{T_1T}=\frac{\gamma_n^2 k_B}{2\mu_B^2}\sum_q A(q)^2\frac{\chi''(q,\omega)}{\omega_n},$$

where γ_n is the nuclear gyromagnetic ratio of ^{63}Cu and $A(q)$ is the q-dependent hyperfine field called the form factor. ω_n is the nuclear Larmor frequency. In the ladder system, $A(q)$ does not depend on q with no supertransferred hyperfine field in the Mila-Rice nuclear spin Hamiltonian for the CuO_2 square lattice. In the case that a Lorentzian spectrum having a q-dependent spin relaxation rate Γ_q is assumed for $\chi''(q,\omega)/\omega_n$, $(1/T_1T)$ is expressed as

$$\frac{1}{T_1T}\sim A_\perp^2\frac{\chi_\perp''(Q,\omega)}{\omega_n}\propto\frac{\pi\chi(Q)}{\Gamma_Q},$$

where $\chi(Q)$ is the static q-dependent susceptibility around $(Q=\pi/a)$ for a quasi-1D AF system. T_{2G} is dominated by the indirect nuclear spin-spin interaction mediated by the AF spin correlation as extensively argued in the literature on the high-T_c cuprates and is described by[17]

$$\left(\frac{1}{T_{2G}}\right)^2\sim\sum_q A(q)^4\chi(q)^2-\left(\sum_q A(q)^2\chi(q)\right)^2.$$

From Fig. 8, T_{2G} for $Sr_2Cu_3O_5$ is proportional to $(T-\theta)$ with $\theta=40$ K, which is not consistent with the expectation from the 1D quantum critical regime where T_{2G} follows a $\sqrt{T}$ dependence from Eq. (1). Interestingly, the ratio of (T_{2G}/T_1T) becomes T independent above 200 K as indicated in Fig. 10, which is not expected for the 1D system,[6] but for the 2D quantum critical regime.[3] $Sr_2Cu_3O_5$ is considered to exhibit the magnetic ordering around 50–60 K as suggested from the disappearance of the NMR signal and μSR experiment,[24] while the renormalized classical regime argued in the 2D Heisenberg square lattice where ξ shows an exponential T dependence is absent in $Sr_2Cu_3O_5$ down to 100 K. Thus, the nature of the spin correlation in $Sr_2Cu_3O_5$ with a three-leg ladder is different from either the 1D or 2D system. At this point, it should be noted that from the T dependence of (T_1T) indicated in Fig. 8, $T_1T\sim\Gamma_Q/\chi_Q$ is proportional to $(T-\theta')$ with $\theta'=80$ K, which is twice as large as $\theta=40$ K from the T_{2G} vs T plot. Namely, ξ and Γ_Q seemingly reach an infinite value and zero, respectively, as if approaching different magnetic ordering temperatures. These novel differences of the spin correlation in $Sr_2Cu_3O_5$ may be considered to be due to finite size effects of legs which means that ξ in the direction of the rung is limited by the number of legs and/or the presence of the 3D interaction along the c axis which is responsible for the magnetic order.

Next, we compare the results in the three-leg ladder with the two-leg ladder. As shown in Fig. 11, $1/T_{2G}$ in the two-leg ladder $SrCu_2O_3$ reveals a much weaker T dependence than that in the three-leg ladder $Sr_2Cu_3O_5$, since ξ remains short range due to the opening of the spin gap.[28] Therefore, the ratio of T_{2G}/T_1T is dominated by the spin gap formation as indicated in Fig. 10. The ξ, i.e., $\chi(Q)$, stays constantly in the spin gap state because χ_Q is relevant to the energy integration of the imaginary part of the dynamical susceptibility from the Kramers-Kronig transformation. Namely, even in the spin gap state, the instantaneous spatial spin correlation is considered to be finite over $\xi/a\sim$ 3–4 as calculated theoretically.

D. Electron doping into a two-leg ladder $SrCu_2O_3$ ($n=3$)

In order to investigate the effect of the electron doping on the spin-gap state, the Knight shift and T_1 in 5% doped

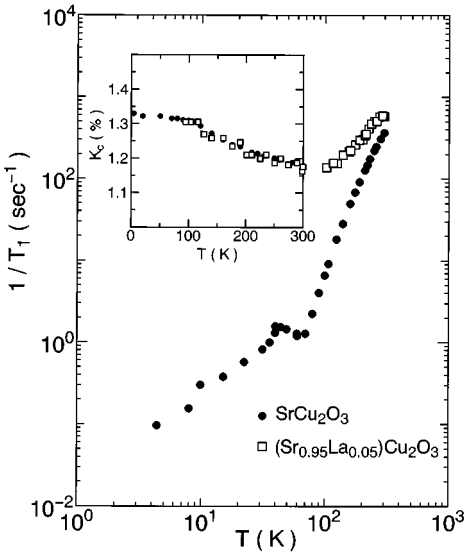

FIG. 12. T dependence of $1/T_1$ in $(Sr_{0.95}La_{0.05})Cu_2O_3$ together with $SrCu_2O_3$. The inset is the K_c vs T plot.

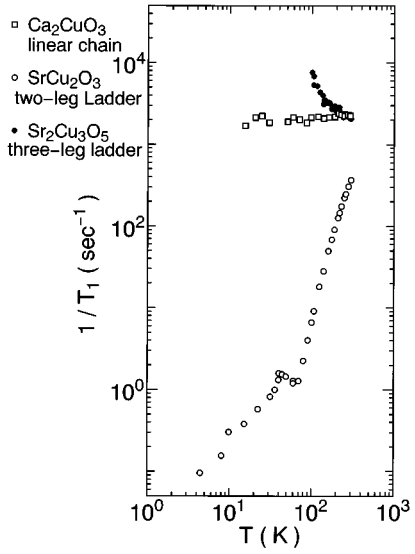

FIG. 13. T dependence of $1/T_1$ in Ca_2CuO_3 with linear chains, $SrCu_2O_3$ with two legs, and $Sr_2Cu_3O_5$ with three legs in logarithmic scale.

$SrCu_2O_3$ have been measured. Below 160 K, the Cu NMR spectrum for the oriented powder with the c axis parallel to the magnetic field at 11 T became broader and was difficult to observe below 80 K. The cause of the broadening of the NMR spectrum is ascribed to the randomly oriented dipole field from local moments induced by doping La into Sr sites. Figure 12 shows the T dependence of $1/T_1$ in $(Sr_{0.95}La_{0.05})Cu_2O_3$ together with the data for undoped $SrCu_2O_3$. T_1 was uniquely determined from the fitting to Eq. (2) above 100 K. The inset in Fig. 12 indicates the T dependence of the Knight shift parallel to the c axis. As seen in the inset, the T dependence of the Knight shift is almost invariant, whereas $1/T_1$ is enhanced at low T by doping La.

In general, the presence of local moments behaving as paramagnetic makes the linewidth of the NMR spectrum broaden by the randomly oriented dipole field for local moments, whereas the Knight shifts are not significantly affected, since the gravity of the NMR spectrum does not shift on average by such a dipole field. By contrast, the relaxation rate is dominated by a fluctuation of moments acting as the relaxation center, masking an intrinsic relaxtion channel as actually observed in La-doped $SrCu_2O_3$.

From the Knight shift measurement, since the spin gap is not seemingly changed by 5% La doping, an enhancement of $1/T_1$ by doping La is possibly caused by the spin fluctuation of local moments produced by breaking singlet states by electron doping associated with the localization of doping electrons.

From the Knight shift measurement in $SrCu_2O_3$, the supertransferred hyperfine coupling (B), arising from the covalent bond between Cu($3d_{x^2-y^2}$)-O($2p_\sigma$) and Cu($4s$) in the four nearest neighbor Cu sites, is considered to be small, as reported in the previous paper.[13]

Therefore the doped carrier is expected not to have the conductive character so much. In fact, Azuma *et al.*[29] reported that the doped carriers in La-doped $SrCu_2O_3$ seemed to remain localized; nevertheless, the electron carriers were added into the Cu_2O_3 sheet, in agreement with our experimental results. The experimental result in $(Sr_{1-x}La_x)Cu_2O_3$ is quite in contrast with in La-doped $SrCuO_2$ (infinite-layer compound) which changes from an AF magnet to a superconductor by only 5% La doping. In the infinite-layer compound, the hybridization between two Cu sites is considered to be large, since the supertransferred hyperfine coupling (B) is about 50 kOe/μ_B.

We suggest that the high mobility of the doped carriers ascribed to the strong hybridization may be indispensable for attaining superconductivity.

IV. SUMMARY

In Fig. 13 are collected the T dependences of $1/T_1$ for linear chain Ca_2CuO_3, two-leg ladder $SrCu_2O_3$, and three-leg ladder $Sr_2Cu_3O_5$. The NMR relaxation studies have clarified the novel nature of the spin correlation depending on the number of chains. In Ca_2CuO_3 with a single linear chain, even though the weak 3D interaction brings about magnetic order at $T_N \sim 10$ K, the spin correlator seems to be in a quantum critical regime, $(1/T_1)$ staying constantly down to T_N. In AP $SrCuO_2$ with a zigzag chain, the weak ferromagnetic exchange passing through the 90° Cu-O-Cu bond tends to depress the low-energy spectral weight of the spin correlation around $Q=(\pi/a)$. If the 3D magnetic order were absent, the spin gap would be present. In $SrCu_2O_3$ with a two-leg ladder, the spin gap formation was evidenced from the susceptibility and NMR T_1 measurements in the previous paper. Furthermore, from the weak T dependence of T_{2G}, it was ensured that the magnetic coherence length ξ remains short range, pointing to the exponential decay of the spatial spin correlator.[28] In contrast, from the T dependences of T_1 and T_{2G} in $Sr_2Cu_3O_5$, the marked decrease of the magnetic relaxation rate Γ_Q and the increase of the magnetic coherence length ξ below 200 K are consistent with the gapless nature of the spin correlators. Triggered by the weak 3D magnetic interaction, the magnetic order is considered to

take place around 50–60 K. Above 200 K, the ratio of T_{2G}/T_1T is nearly the constant, meaning that the quantum critical regime similar to the 2D Heisenberg square lattice sets in the high-T region.

In La-doped $SrCu_2O_3$, the electronlike carriers were shown to be localized, dominating the low-energy excitation by local moments induced around localized carriers.

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