Quantum magnetism of spin $S = \frac{1}{2}$ in low dimensions has attracted strong attention since the discovery of high $T_c$ superconductivity in hole-doped CuO$_2$ planes. Intensive experimental and theoretical studies have led to a reasonably good understanding of the statistical properties of the 2d square-lattice [1]. More recently, the successful synthesis of Sr$_2$CuO$_3$ and Sr$_2$Cu$_3$O$_5$ with two- and three-leg ladder structure, respectively [2], has made it possible to investigate $S = \frac{1}{2}$ quantum spins between 1d and 2d. Subsequent discovery of superconductivity in another two-leg ladder material (La,Ca,Sr)$_3$Cu$_2$O$_{4+\delta}$ [3] further enhanced interest in the statistical properties of quantum spin ladders. In addition, under-doped high $T_c$ superconductors show a stripe structure [4] which has been proposed as undoped three-leg spin ladders coupled across rivers of doped holes. Concentrated theoretical effort is underway to reproduce this phenomenon based on the anisotropic non-linear sigma model [5]. This extends the fundamental importance of spin ladders beyond the field of magnetism to the mechanism of high $T_c$ superconductivity.

Uniform susceptibility [6], NMR/NQR [7,12], and neutron scattering measurements [3,14] have confirmed the most basic prediction about spin ladders, namely that ladders with an even number of legs have a spin gap $\Delta$ in their spin excitation spectrum, while ladders with an odd number of legs do not [13]. For Sr$_2$CuO$_3$ and Sr$_2$Cu$_3$O$_5$, spin susceptibility by Azuma, et al. [3] and pioneering high field $^{63}$Cu NMR between 100 K and 300 K by Ishida, et al. [8] revealed the qualitative difference of magnetic properties between two- and three-leg ladders in the low temperature limit. However, the nature of spin correlations at finite temperatures remains largely untested, despite recent theoretical developments [10,21]. The large exchange interaction $J = 1300\sim1600$ K in copper-oxide ladders [6,12,13] makes measurements of spin-correlations at elevated temperatures essential to understanding the statistical properties at finite temperatures.

In this Letter, we report the first $^{63}$Cu NQR measurements of the $^{63}$Cu nuclear spin-lattice relaxation rate $1/T_1$ [21] and the Gaussian component of spin-spin relaxation rate $1/T_2G$ [22] for the three-leg ladder Sr$_2$Cu$_3$O$_5$ from 83 K up to 725 K. The Gaussian component of the $^{63}$Cu nuclear spin-spin relaxation rate, $1/T_2G$, probes the spin-spin correlation length, $\xi$, as demonstrated earlier for the 2d square-lattice [23,24] and the 1d spin-chain [26]. We present evidence that the spin-spin correlation length $\xi$ in the three-leg ladder follows the 1d form $\xi \sim 1/\sqrt{T}$ above 300 K. To our surprise, however, we found that weak inter-ladder coupling along the c axis results in dimensional crossover to a quasi-2d regime below 300 K, where $\xi$ diverges exponentially, $\xi \sim \exp(2\pi\rho/T)$ (2$\pi\rho$ = 290±30 K is the effective spin stiffness). The anisotropic non-linear sigma model [6] successfully describes the static and dynamic NQR/NMR properties in the quasi-2d regime. To the best of our knowledge, this is the first experimental demonstration of the validity of the anisotropic non-linear sigma model in a quantum Heisenberg antiferromagnet.

In Figure 2(a) and (b), we compare the temperature dependence of the nuclear spin relaxation rates for the three-leg ladder Sr$_2$Cu$_3$O$_5$, with other geometries between 1d and 2d: 1d spin-chain Sr$_2$CuO$_3$, two-leg ladders Sr$_2$CuO$_3$ and La$_6$Ca$_8$Cu$_{24}$O$_{41}$, and 2d square-lattice Sr$_2$Cu$_2$O$_2$Cl$_2$. The polycrystalline samples of the ladders, Sr$_2$Cu$_3$O$_5$ and Sr$_2$Cu$_2$O$_3$, were grown at Kyoto under high pressure [3], while the single crystal for 1d spin-chain Sr$_2$CuO$_3$ was grown at M.I.T. using an optical floating zone furnace. The results for 2d square-lattice Sr$_2$Cu$_2$O$_2$Cl$_2$ are from [25] and for two-leg ladder La$_6$Ca$_8$Cu$_{24}$O$_{41}$ from [13].
measurements at elevated temperatures limited only by sample decomposition. In addition, the three-leg ladder structure shown in figure 1(a) & 1(b) has two different copper sites, the atoms in the central chain and those in the two edge chains. Our experiments measure the relaxation times for the two sites separately without line superposition. Separation of the two signals is essential because $1/T_1$ differs by a factor of $2.3^{[27]}$.

$1/T_1$ measures the imaginary part of the dynamical electron spin susceptibility $\chi''(q,\omega_n)$ at the NQR/NMR frequency $\omega_n(=11 \text{ (NQR)} \text{ or } 100 \text{ (NMR)} \text{ MHz})$, or low-frequency spin dynamics $^{[28]}$.

$$\frac{1}{T_1} = k_B T \frac{\gamma_n^2}{\mu_B^2} \sum_q |F(q)|^2 \frac{\chi''(q,\omega_n)}{\omega_n}$$  \hspace{1cm} (1)

where $\gamma_n$ is the nuclear gyromagnetic ratio. $F(q)$ is the wave vector dependent hyperfine form factor $^{[28]}$. On the other hand $1/T_{2G}$ measures the real part of the electron spin susceptibility at zero frequency, $\chi'(q)$ $^{[23]}$.

$$\left(\frac{1}{T_{2G}}\right)^2 \approx \frac{0.69(\gamma_n h)^4}{8\hbar^2} \sum_q |F(q)|^4 \chi'(q)^2$$  \hspace{1cm} (2)

where the factor 0.69 is the isotope abundance of $^{63}\text{Cu}$. $1/T_{2G}$ provides quantitative information of spin-spin correlation length $\xi$ in low dimensional systems; $1/T_{2G} \sim T \xi$ for 2d square-lattice $^{[13]}$, $\sim \sqrt{T}/T$ for ladders at elevated temperatures (see below), and $\sim 1/\sqrt{T}$ for 1d spin-chain $^{[24]}$.

The results in Fig.2 clearly show dramatic change of spin-spin correlations with dimension. In 2d square-lattice Sr$_2$CuO$_2$Cl$_2$, $1/T_1 \sim T^{3/2} \xi$ $^{[29]}$ and $1/T_{2G} \sim T \xi$ $^{[12]}$ diverge exponentially, because $\xi$ diverges exponentially in the renormalized classical low temperature regime $^{[47]}$. $^{[47]}$

$$\xi = \frac{c}{8 \sqrt{2} \pi \rho_s} \exp \left(\frac{2\pi \rho_s}{T} \right) \left[1 - \frac{T}{4\pi \rho_s} + O(T^2)\right]$$  \hspace{1cm} (3)

where $c$ is the spin wave velocity, and $2\pi \rho_s(=1.13J)$ is the spin stiffness. As shown in Fig.3(a), the semi-logarithm plot of $1/T_1 T^{3/2}$ and $1/T_{2G} T$ shows a linear behavior with slope $2\pi \rho_s = 1700 \pm 150$ K. In 1d spin-chain Sr$_2$CuO$_3$, $1/T_1$ and $1/T_{2G}$ show much weaker temperature dependence up to 800 K. This agrees with earlier confirmation of theoretical predictions below 300 K by Takigawa et al. $^{[24]}$, $1/T_1 \sim \text{constant}$ and $1/T_{2G} \propto 1/\sqrt{T}$. $^{[13]}$ In two-leg ladder SrCu$_2$O$_3$, the presence of spin-gap $\Delta \sim 450$ K $^{[17,20]}$, in the spin excitation spectrum results in exponential decrease of $1/T_1$ $^{[17,20]}$ below $\sim 450$ K, where $1/T_{2G} T$ (and thus $\xi$ $^{[17,20]}$) gradually saturates.

The results for three-leg ladder fall between 2d square-lattice and 1d spin-chain, as expected. The mild temperature dependence above 300 K is easy to understand. According to recent weak coupling continuum theory by Buragohain and Sachdev $^{[14]}$, which is applicable $25 \text{ K} \ll T \lesssim 500 \text{ K}$ for $J = 1500 \text{ K}$, the spin structure factor is given as $\chi(q) = S(q)/k_B T \propto (1/k_B T) \ln(T/\Lambda_{MS})^2 \xi/(1 + (q - \pi)^2 \xi^2)$ with $\Lambda_{MS}$ roughly predicted as $25 \text{ K}$. Inserting this expression into eq.(2), the leading order temperature dependence of $T_{2G}$ is given as $T_{2G} \propto T/\sqrt{\xi}$, with logarithmic corrections that become significant at low temperatures. Since the spin-spin correlation length in a three-leg ladder is given as $\xi \sim 1/\sqrt{3}T$, we expect $T_{2G} \sim T^{3/2}/\sqrt{3}$. Indeed, as shown in the inset to Fig. 2(b), we found power law behavior with exponent $1/2$ above $300 \text{ K}$. Comparison of $1/T_1$ with the analytic theory is more difficult because we cannot estimate the large diffusive (i.e. $q = 0$) contributions in the quasi-1d regime. Further numerical calculations are necessary to understand the observed constant behavior of $1/T_1$ at $T \sim 1/\sqrt{3}$. $^{[14]}$

In contrast with the mild temperature dependence at elevated temperatures, the divergent behavior below 300 K is quite surprising. As shown in Fig.3, the temperature dependence is exponential, similar to the case of the 2d square-lattice. The linearity in the semi-logarithmic plot extends for an order of magnitude. Within the framework of an isolated three-leg ladder, we expect that at $T \approx J$ the exchange interaction along a rung couples the three $S = 1/2$ spins into an effective $S_{eff} = 1/2$, forming a $S_{eff} = 1/2$ chain. Therefore, an isolated three-leg ladder would exhibit $1/T_{2G} \propto 1/\sqrt{T}$ and $1/T_1 \sim \text{constant}$ at low temperatures $^{[22]}$ as observed for 1d spin-chain Sr$_2$CuO$_3$ $^{[24]}$. This clearly contradicts with the exponential divergence we find. Three-dimensional spin freezing observed at $T_{SF} = 52 \text{ K} ^{[32]}$ is unlikely to be the origin of the observed divergence, either, because the onset of the exponential divergence ($\sim 300$ K) is nearly a factor of 6 higher than $T_{SF} = 52 \text{ K}$. Furthermore, the temperature dependence is exponential rather than the ordinary power law divergence of $1/T_1$ and $1/T_{2G}$ expected near 3d orderings. The lack of 3d character is consistent with the fact that the exchange coupling along the b axis is frustrated due to opposing pairs of $90^\circ$ Cu-O-Cu bonds, which suppresses 3d correlations $^{[14]}$.

The key point to note is that, along the c axis, the three-leg ladders are stacked directly on top of one another as shown in Fig.1(b). We recall that the so-called infinite layer compound Ca$_{0.85}$Sr$_{0.15}$CuO$_2$ has 2d square-lattice layers with a similar c-axis stacking, and has an equivalent structure to Sr$_2$Cu$_2$O$_5$ except for the line defect between adjacent three-leg ladders. In Ca$_{0.85}$Sr$_{0.15}$CuO$_2$, Néel ordering driven by the large c axis coupling, $J_c$, occurs at extremely high temperature, $539 \text{ K}$, and the dimensional crossover from isolated 2d square-lattice behavior to 3d behavior occurs as high as $600 \text{ K} ^{[34]}$. This suggests that the three-leg ladders in Sr$_3$Cu$_3$O$_5$ will also couple strongly along the c-axis. At low temperatures, the stacked three-leg ladders form a
2d plane of $S_{\ell f} = \frac{1}{2}$ with anisotropic exchange interactions, $J$ along the a axis and the effective c axis coupling for the 2d model, $J_{c}^{\ell f} \approx 3J_c$, as shown in figure 1(c).

Our viewpoint is supported by two sets of recent Monte Carlo simulations. First, Greven and Birgeneau showed that inter-layer coupling $J_c$ along c-axis was essential to understand the 3d long-range order of Zn-doped two-leg ladder SrCu$_2$O$_3$ [33,34]. Second, more recent Monte-Carlo simulations by Y.J. Kim et al. showed that inter-layer coupling two-orders of magnitude smaller than $J$ is sufficient to induce dimensional crossover from quasi-1d behavior of an isolated three-leg ladder to anisotropic 2d behavior of coupled three-leg ladders [35]. In the present case, the inter-ladder coupling along c-axis is large, $J_{c}^{\ell f}/J \approx (0.15 - 0.22)$ using estimates $J_c \sim 75 - 110$ K [35,36], and $J \sim 1500$ K.

Theoretically, the anisotropy $\alpha = J_{c}^{\ell f}/J$ of exchange interaction introduces anisotropy in the spin wave velocity $c_0$ and spin-spin correlation length $\xi$ for two orthogonal directions, and reduces the isotropic spin stiffness $2\pi \rho_s$ to an effective $2\pi \rho_s^{\ell f}$ [35]. That is, $c_{\parallel}(\alpha) = (1 + \alpha)/2c_0$, $c_{\perp}(\alpha) = (\sqrt{\pi}c_0)(\alpha)$, and $2\pi \rho_s^{\ell f} = (1-g_0(1)g_\alpha(\alpha))/\sqrt{\pi}2\pi \rho_s$, where $g_\alpha(\alpha)$ is the critical coupling constant, and $g_0(1)$ is the bare coupling constant for $\alpha = 1$ [35]. Otherwise the theoretical framework of the renormalized classical regime in isotropic 2d square lattice [29,31], which was successfully employed to analyze $^{63}$Cu NQR/NMR relaxation rates in 2d square-lattice La$_2$CuO$_4$ [32,34] and Sr$_2$CuO$_2$Cl$_2$ [21], is applicable to the anisotropic case. By fitting $1/T_1\ell f^2$ and $1/T_2G T$ in Fig.3 to exponential form, we obtain $2\pi \rho_s^{\ell f} = 290 \pm 30$ K. This implies an anisotropy, $\alpha = 0.16(0.17) \pm 0.02$ for $J = 1500(1300)$ K, hence $J_{c}^{\ell f} = 230 \pm 30$ K. The obtained value of $\alpha = J_{c}^{\ell f}/J = 0.16 \pm 0.02$ is consistent with the estimate of 0.15 - 0.22 in the previous paragraph.

We can test the consistency of the preceding renormalized classical analysis by estimating $\alpha$ based on a different method without knowing $J$. In the low-temperature renormalized classical limit, we expect the ratio $R(\alpha, 2\pi \rho_s^{\ell f}) = (T_1T_2^{\ell f})/(T_2G T) \cdot (F_{ab}(\pi)/F_{\pi}(\pi))^2 \propto \sqrt{(h^2c_\parallel(\alpha)c_{\perp}(\alpha)/2\pi \rho_s^{\ell f}) \cdot (F_{ab}(\pi)/F_{\pi}(\pi))^2}$, to be independent of temperature. The ratio of hyperfine form factors, $F_{ab}(\pi)/F_{\pi}(\pi)$, can be determined experimentally as 0.42 ± 0.02 from $T_{1c}/T_{1ab} = 3.4 \pm 0.2$, and $2\pi \rho_s^{\ell f} = 290 \pm 30$ K from the fit in Figure 3. This leaves $\alpha$ as the only unknown parameter in $R(\alpha, 2\pi \rho_s^{\ell f})$. Shown in the inset to Fig.3, the ratio $R(\alpha, 2\pi \rho_s^{\ell f})$ indeed approaches a constant 61 ± 5 K at low temperatures, which implies $\alpha = 0.15 \pm 0.03$, in agreement with our earlier estimate, 0.16 ± 0.02 [35].

To conclude, we demonstrated that the temperature dependence of the spin-spin correlation length $\xi$ in Sr$_2$Cu$_3$O$_5$ is consistent with the isolated three-leg ladder behavior $\xi \sim \frac{1}{T}$ from 300 K to 725 K ($\sim J/2$). Below 300 K, we discovered dimensional crossover to anisotropic 2d regime, where spin correlations diverge exponentially. Our result is the first experimental demonstration of the validity of the anisotropic non-linear sigma model, which was recently proposed for the stripe phase of high $T_c$ cuprates, in a model $S = \frac{1}{2}$ quantum Heisenberg antiferromagnet. This should encourage further theoretical analysis of the stripe physics of high $T_c$ cuprates based on the anisotropic non-linear sigma model.

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[20] A. W. Sandvik et al., Phys. Rev. B 53, R2934 (1996).
[21] The nuclear quadrupole interaction tensor $\eta_{ab}$ at the copper sites in ladders is not axially symmetric with respect to the c axis, $\eta = (\eta_{ab} - \eta_{ac})/\eta_{bc}=0.64$ and 0.17 for the edge and central chain of ladders, respectively [8]. It is straightforward to show that, based on the standard Redfield approach, $1/T_1$ measured by NQR is a combination of $1/T_1$ measured by high field NMR for different magnetic field directions,

$$\frac{1}{T_1}^{NQR} = \left \{ 1 + \frac{1}{9 + 3\eta^2} \right \} \left \{ \frac{1}{T_1}^{ab} + (9 - \eta^2) \frac{1}{T_1}^{ac} \right \} \quad (4)$$

where subscript $ab$ and $c$ denote the direction of magnetic field. For the edge chain site, we measured $1/T_1$ with both NQR and high field NMR, and confirmed the con-
sistency of the experimental ratios, $T_{1c}/T_{1ab} = 3.4 \pm 0.2$
and $T_{1c}/T_{1\text{NQR}} = 0.70 \pm 0.03$ with the theoretical expression, $T_{1c}/T_{1\text{NQR}} = 0.72 \pm 0.02$. For the purpose of systematic comparison with other materials, we multiplied a factor 0.70 to the NQR results to deduce $\frac{1}{T_{1c}}$.

We separated the contribution of spin-lattice relaxation process $(1/T_{2R})_{NQR}$ to NQR spin echo decay using theoretical calculations based on Redfield theory,

$$
\left( \frac{1}{T_{2R}} \right)_{NQR} = \left( \frac{1}{3 + \eta^2} \right) \left[ \frac{1}{T_{1c}} + (3 + 3\eta^2) \frac{1}{T_{1a,b}} \right]
$$

The effects of finite value of $T_1$ (N. J. Curro et al., Phys. Rev. B 56, 877 (1997)) is estimated to be small (~5%). For systematic comparison, we converted the NMR results for Sr$_2$CuO$_3$ to the powder NQR results using $(1/T_{2G})_{NQR} = \frac{T_{2G}}{10}(1/T_{2G})_{NMR}$. We confirmed the validity of the theoretical conversion at 300 K experimentally, and that the observed value of $1/T_{2G}$ does not depend on the strength of R.F. pulses.

[22] We separated the contribution of spin-lattice relaxation process $(1/T_{2R})_{NQR}$ to NQR spin echo decay using theoretical calculations based on Redfield theory.

FIG. 1. Structure of three leg ladder material, Sr$_2$Cu$_3$O$_5$, (a) top view, (b) side view, and (c) effective structure in anisotropic 2d regime below 300 K.

FIG. 2. (a) $1/T_{1c}$ and (b) $(1/T_{2G})_{NQR}$ for the 3-leg ladder Sr$_2$Cu$_3$O$_5$ (edge chain Cu site) [1], in comparison to two-leg ladders SrCu$_2$O$_3$ [2] and La$_6$Ca$_8$Cu$_{22}$O$_{41}$ [3], 2d square-lattice Sr$_2$CuO$_2$Cl$_2$ [4], and 1d spin chain Sr$_2$CuO$_3$ [5]. Inset to Fig.2(b): $T_{2G}$ at edge chain Cu site in Sr$_2$Cu$_3$O$_5$ and SrCu$_2$O$_3$. Solid line shows fit above 300 K to predicted power law, $T_{2G} \propto T^{3/2}$ in the quasi-1d regime of an isolated three-leg ladder.

FIG. 3. $1/(T_{1c}T^{3/2})$ (circles) and $1/(T_{2G}T)$ (triangles) versus $1/T$ for three-leg Sr$_2$Cu$_3$O$_5$ (solid symbols) and 2-d Sr$_2$CuO$_2$Cl$_2$ (open symbols). The fit to renormalized classical (exponential) behavior for Sr$_2$Cu$_3$O$_5$ $T < 225$ K gives $2\pi\rho_{eff}^c = 290 \pm 30$ K, implying anisotropy $\alpha = 0.16 \pm 0.02$ for $J = 1500$ K. Inset: Ratio $R(\alpha, 2\pi\rho_{eff}^c) = (T_{1c}T^{3/2}/T_{2G}T) \cdot (F_2^a/F_2^b)$ for three-leg Sr$_2$Cu$_3$O$_5$. The ratio should be constant in the low temperature limit deep inside the renormalized classical regime. The solid line shows the value of $R(\alpha, 2\pi\rho_{eff}^c) = 290$ K.
\[ \frac{1}{T_1 c} \text{[sec}^{-1}] \]

\[ \frac{1}{T_2 G} \text{[msec}^{-1}] \]

\( T \) [K]

\( T \) [msec]
\[ \frac{1}{T_{1C}} \left[ \text{sec}^{-1} \text{K}^{3/2} \right] \]

\[ \frac{1}{T_{2G}} \left[ \text{msec}^{-1} \text{K}^{-1} \right] \]

\[ \frac{250}{T} \left[ \text{K}^{-1} \right] \]

\[ \frac{125}{T} \left[ \text{K}^{-1} \right] \]

\[ \frac{83}{T} \left[ \text{K}^{-1} \right] \]