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Muon-spin-relaxation study of the effect of nonmagnetic impurities on the Cu-spin fluctuations in $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ around $x=0.115$

I. Watanabe
Muon Science Laboratory, RIKEN (The Institute of Physical and Chemical Research), 2-1 Hirosawa, Wako, Saitama 351-0198, Japan

T. Adachi, K. Takahashi, S. Yairi, and Y. Koike
Department of Applied Physics, Graduate School of Engineering, Tohoku University, Aoba-yama 08, Aoba-ku, Sendai 980-8579, Japan

K. Nagamine
Meson Science Laboratory, Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK-MSL), 1-1 Oho, Tsukuba, Ibaraki 305-0801, Japan
and Muon Science Laboratory, RIKEN (The Institute of Physical and Chemical Research), 2-1 Hirosawa, Wako, Saitama 351-0198, Japan

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Zero-field muon-spin-relaxation (μSR) measurements have been carried out in order to investigate the effect of nonmagnetic impurities on the dynamics of the Cu-spin fluctuations in the Zn-substituted $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ with $x=0.115$ and 0.13, changing y from 0 to 0.10. A long-range-ordered state of Cu spins, which is observed in the Zn-free sample with $x=0.115$, disappears in the Zn-free sample with $x=0.13$, but appears again in the Zn-substituted samples with $x=0.13$ for $y \geqslant 0.0075$. The long-range-ordered state disappears for $y>0.03$ for both values of x, so that the Cu spins are in a paramagnetic state. The present μSR results support the suggestion that a small amount of nonmagnetic impurities produces a pinning of the dynamical stripe correlations of spins and holes and make them statically stabilized, while a large amount of nonmagnetic impurities destroy the stripe correlations themselves.

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I. INTRODUCTION

The stripe correlations of spins and holes were suggested by Tranquada *et al.* to be important for understanding the so-called $\frac{1}{8}$ effect observed in $La_{2-z-x}Nd_zSr_xCuO_4$ (LNSCO).[1] Based upon the stripe model, it is understood that the suppression of high-T_c superconductivity due to the $\frac{1}{8}$ effect around the hole concentration p of $\frac{1}{8}$ per Cu is caused by the static stabilization of the dynamical stripe correlations. Although no clear evidence of the existence of the dynamical stripe correlations has been obtained yet, the observation of the incommensurate magnetic peaks around (π,π) in $La_{2-x}Sr_xCuO_4$ (LSCO) from the neutron inelastic scattering experiments[2–5] leads us to argue about the dynamical stripe correlations around $p=\frac{1}{8}$ per Cu.

Recently, Koike *et al.*[6,7] and Adachi *et al.*[8] suggested that the dynamics of the dynamical stripe correlations was affected by nonmagnetic impurities such as Zn. They studied transport properties of $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ (LSCZO) by changing x and also the Zn concentration y, and argued that a small amount of Zn tended to statically stabilize the dynamical stripe correlations, while a large quantity of Zn, in contrast, destroyed them. As for the other studies on the Zn-substitution effect around $p=\frac{1}{8}$ per Cu in LSCZO, Hirota *et al.* performed a neutron-scattering experiment on LSCZO with $x=0.14$ and $y=0.012$, reporting the appearance of a quasistatic state of Cu spins.[9] Kimura *et al.* also reported the observation of a statically ordered state of Cu spins in LSCZO with $x=0.12$ and $y=0.03$.[10] Our previous zero-field muon-spin-relaxation (ZF-μSR) measurements on LSCZO with $x=0.115$ and $y=0.01$ revealed that the magnetic correlation between Cu spins seemed to be enhanced by the substituted Zn.[11] The detailed Zn dependence of the dynamics of Cu-spin fluctuations around $p=\frac{1}{8}$ per Cu, however, has not been established yet. Thus, in order to confirm the suggestion by Koike *et al.*,[6,7] and Adachi *et al.*,[8] we have carried out ZF-μSR measurements on LSCZO with $x=0.115$ and 0.13, changing y in fine steps.

II. EXPERIMENT

Polycrystalline samples of LSCZO with $y=0$, 0.0025, 0.005, 0.0075, 0.01, 0.02, 0.03, 0.05, 0.07, and 0.10 were prepared for both systems with $x=0.115$ and 0.13 by the ordinary solid-state-reaction method. The procedure of the sample preparation is the same as that used in the transport measurements.[8] All of the samples were checked to be single phase and their transport properties were also measured to check the quality of the samples before the μSR measurements.

ZF-μSR measurements were carried out at the RIKEN-RAL Muon Facility at the Rutherford-Appleton Laboratory in the UK using a pulsed positive surface-muon beam. The asymmetry parameter at a time t was given by $A(t)=[F(t)-\alpha B(t)]/[F(t)+\alpha B(t)]$, where $F(t)$ and $B(t)$ were total muon events of the forward and backward counters, which were aligned in the beam line, respectively. The α is a calibration factor reflecting the relative counting efficiencies between the forward and backward counters. The asymmetry at $t=0$ is the initial asymmetry A_0. Time evolution of $A(t)$ (μSR time spectrum) was measured down to 2 K to detect the appearance of a magnetically ordered state and slowing-down behavior of the Cu-spin fluctuations.

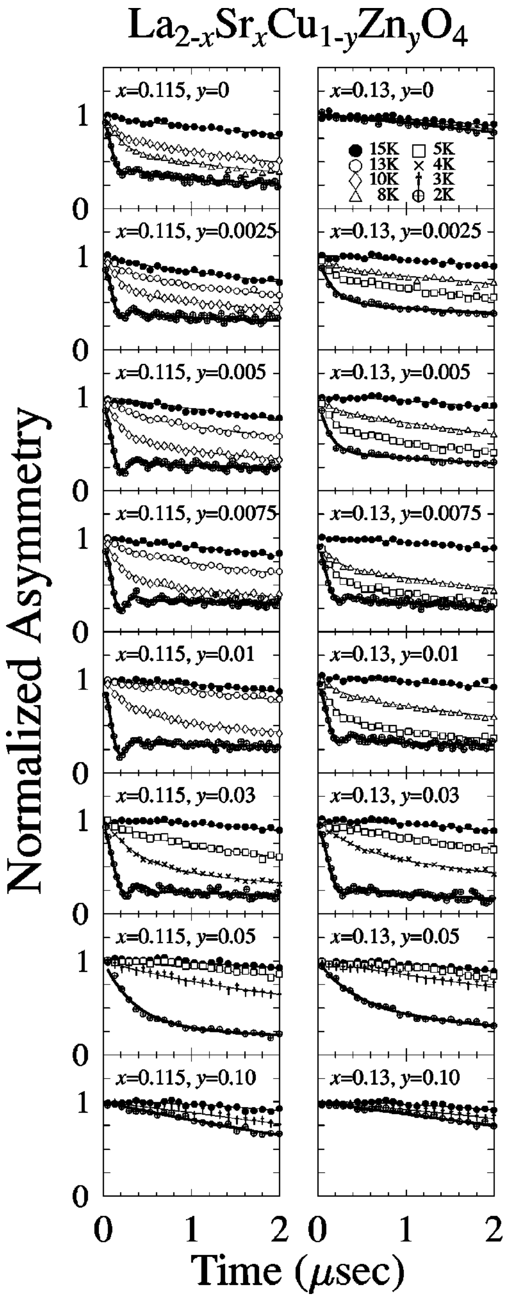

FIG. 1. ZF-μSR time spectra of the Zn-free and Zn-substituted $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ with $x=0.115$ and 0.13 obtained at values of y from 0 to 0.10 and at various temperatures down to 2 K. Time spectra in the early time region from 0 to 2 μsec are displayed. Solid lines indicate the best-fit results using $A_0e^{-\lambda t}G_Z(\Delta,t)$ or $A_0e^{-\lambda_0 t}+A_1e^{-\lambda_1 t}+A_2e^{-\lambda_2 t}\cos(\omega t+\phi)$.

III. RESULTS

Figure 1 shows the ZF-μSR time spectra of LSCZO with $x=0.115$ and 0.13 in the early time region from 0 to 2 μsec obtained at various temperatures. In the Zn-free sample with $x=0.115$, no influence of the Cu-spin fluctuations is observed at temperatures higher than about 15 K. In such a case, the time spectrum is analyzed using the simple function $A_0e^{-\lambda t}G_Z(\Delta,t)$, where A_0 is the initial asymmetry and λ is the depolarization rate of the muon spin. The $G_Z(\Delta,t)$ is the static Kubo-Toyabe function with a half-width of Δ describing the distribution of the nuclear-dipole field at the muon site.[12] With decreasing temperature, a fast depolarizing component appears and muon-spin precession is observed at 2 K in the Zn-free sample with $x=0.115$, indicating the appearance of a long-range-ordered state of Cu spins. Since the fast decay of the muon-spin precession means large distribution of the internal field at the muon site, the correlation length of the observed long-range-ordered state might not be so long. In this case, the analysis using the Kubo-Toyabe function is no longer valid and the multicomponent function, $A_0e^{-\lambda_0 t}+A_1e^{-\lambda_1 t}+A_2e^{-\lambda_2 t}\cos(\omega t+\phi)$ is used, as in the previous study.[13] The first and second terms indicate the slow and fast depolarization, respectively. The third term expresses the muon-spin precession. The A_1 and A_2 are the initial asymmetries. The λ_1 and λ_2 are the depolarization rates. The ω and ϕ are the frequency and phase of the muon-spin precession, respectively. Solid lines in Fig. 1 indicate the best-fit results.

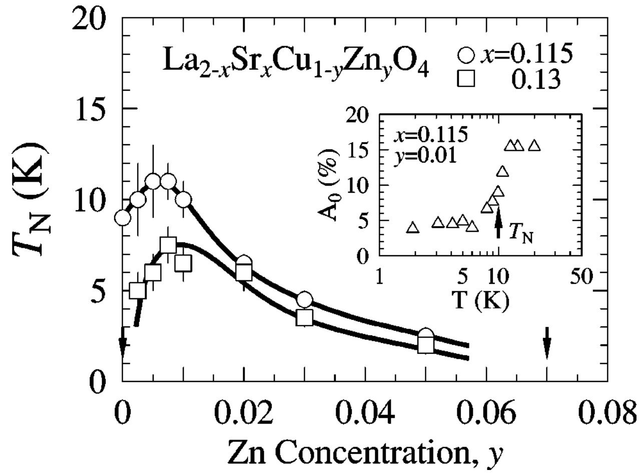

FIG. 2. Zn-concentration dependence of the magnetic transition temperature T_N defined at the midpoint of the temperature dependence of the initial asymmetry of the slow-depolarization component A_0. Circles and squares show T_N's of the samples with $x=0.115$ and 0.13, respectively. Solid lines are guides to the eye. Arrows at $y=0$ and 0.07 mean that no sign of the appearance of the magnetic transition was observed down to 2 K for $x=0.13$. As for $x=0.115$, no sign of the appearance of the magnetic transition was observed down to 2 K for $y\geqslant 0.07$. The inset shows the temperature dependence of A_0 in the case of $x=0.115$ and $y=0.01$. The T_N is indicated by an arrow in the inset as the midpoint of the transition in A_0.

The muon-spin precession in the Zn-free sample with $x=0.115$ observed at 2 K becomes obvious with increasing y, and its amplitude is the largest at $y=0.01$. The muon-spin precession disappears for $y>0.03$. Almost no fast depolarization of the muon spin is observed at $y=0.10$, meaning less influence of the Cu spins on the muon spins.

As for the ZF-μSR time spectra of LSCZO with $x=0.13$, the same functions as described above are used for the analysis. In contrast to the case of $x=0.115$, no fast depolarization is observed down to 2 K in the Zn-free sample with $x=0.13$, indicating the absence of both the long-range

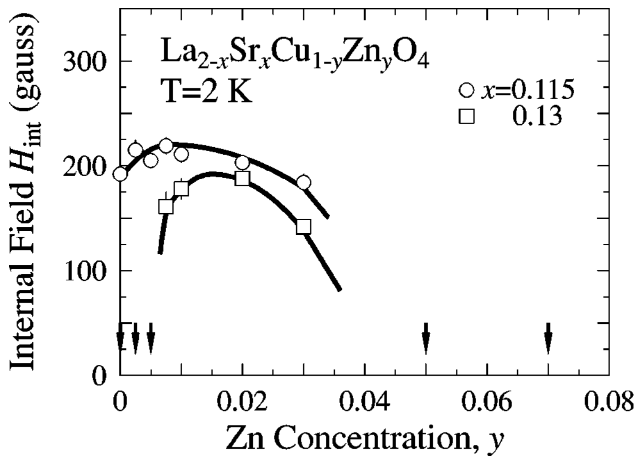

FIG. 3. Zn-concentration dependence of the internal field H_{int} at the muon site obtained at 2 K. Circles and squares show H_{int}'s of the samples with $x=0.115$ and 0.13, respectively. Solid lines are guides to the eye. Arrows mean that no clear muon-spin precession was observed down to 2 K for $x=0.13$. For $x=0.115$, no clear muon-spin precession was observed down to 2 K for $y \geq 0.05$.

magnetically ordered state and the slowing-down behavior of the Cu-spin fluctuations. However, once Zn is substituted for Cu, a fast depolarization appears at low temperatures. For $y \geq 0.0075$, moreover, clear muon-spin precession is observed at 2 K. The amplitude of the precession is the largest at $y=0.01$ as in the case of $x=0.115$. The precession pattern of the time spectrum is similar to that obtained in the Zn-free sample with $x=0.115$. The muon-spin precession is observed up to $y=0.03$ at 2 K. Almost no fast-depolarization behavior is observed at $y=0.10$, as in the case of $x=0.115$.

The magnetic transition temperature T_N of each sample can be estimated from the midpoint of the temperature dependence of A_0, as typically shown in the inset of Fig. 2 for the sample with $x=0.115$ and $y=0.01$.[13] Figure 2 shows the Zn-concentration dependence of T_N in both systems with $x=0.115$ and 0.13. In the case of $x=0.115$, T_N slightly increases with increasing y and shows a peak at $y=0.0075$. In the case of $x=0.13$, on the other hand, the change of the magnetic state is very drastic. The magnetically ordered state appears at $y=0.0025$. T_N increases with increasing y and exhibits a peak at $y=0.0075$, as in the case of $x=0.115$. The T_N decreases slowly with increasing y for $y>0.0075$ in both systems with $x=0.115$ and 0.13. The y dependence of T_N for $y \geq 0.01$ is similar for both x values. No sign of the appearance of the static magnetically ordered state is observed down to 2 K for $y \geq 0.07$ in either system.

The internal field H_{int} at the muon site is estimated from ω, as $H_{int}=\omega/\gamma_\mu$. Here, γ_μ is the gyromagnetic ratio of the muon spin ($\gamma_\mu/2\pi=13.55$ MHz/kOe). Figure 3 exhibits the Zn-concentration dependence of H_{int} in both systems with $x=0.115$ and 0.13. In the case of $x=0.115$, H_{int} increases slightly with increasing y and shows a peak at $y=0.01$. In the case of $x=0.13$, on the other hand, H_{int} becomes well defined for $y \geq 0.0075$ and increases with increasing y, followed by a peak at $y=0.015$. After showing the peak, H_{int} decreases with increasing y in both systems, and no well defined H_{int} is obtained for $y \geq 0.05$.

IV. DISCUSSION

First, we discuss the Cu-spin state in the small Zn-concentration region $y \leq 0.01$. The pattern of the muon-spin precession observed in the Zn-free sample with $x=0.115$ is quite similar to that obtained around $p=\frac{1}{8}$ per Cu in LNSCO, where the static stripe order of spins and holes is formed.[14] This fact means that the alignment of Cu spins surrounding the muon in the Zn-free sample with $x=0.115$ is similar to that around $p=\frac{1}{8}$ per Cu in LNSCO. Although no information on the alignment of holes can be obtained from the μSR measurements, the long-range-ordered state of Cu spins observed in the present study is probably to be regarded as the statically stabilized state of the dynamical stripe correlations. This result is consistent with those obtained by the neutron-scattering experiments suggesting the existence of the modulated static order of Cu spins in LSCZO with $x=0.12$ and $y=0$ (Refs. 10,15) and with $x=0.12$ and $y=0.03$.[10]

Accordingly, the disappearance of the muon-spin precession even at the lowest temperature of 2 K in the Zn-free sample with $x=0.13$ means that a small amount of excess holes compared with those at $x=0.115$ destabilizes the static stripe ordered state. The Cu spins are apparently dynamically fluctuating with shorter periods than the typical μSR time window (10^{-6}–10^{-11} sec). Based upon the stripe model, therefore, the appearance of the muon-spin precession, that is, the appearance of a static long-range-ordered state of the Cu spins in the samples with $x=0.13$ and $y \geq 0.0075$ and the rapid increase of T_N with increasing y, strongly suggest that a small amount of Zn leads to the static stabilization of the dynamical stripe correlations. This result is also supported by the fact that the precession patterns of $x=0.13$ are similar to those obtained in the samples with $x=0.115$ and also to that observed around $p=\frac{1}{8}$ per Cu in LNSCO.[14]

The Zn-substitution effect on the magnetically ordered state in LSCZO was investigated by Hücker *et al.* in the insulating region $x<0.02$. They pointed out from susceptibility measurements that the localization of holes around the substituted Zn restored the antiferromagnetic correlation between Cu spins, resulting in the increase in T_N[16]. Such an effect may be expected to exist around $x=0.115$ as well, but this effect seems not to be suitable in order to explain the rapid change in T_N as a result of a 1% substitution of Zn for Cu at $x=0.13$. This is because the drastic change in T_N was not observed in $x=0.115$ but was observed at $x=0.13$, though the hole concentration of LSCZO is higher and doped holes are more mobile at $x=0.13$ than at $x=0.115$.

A possible explanation of the rapid change in T_N for $y \leq 0.01$ at $x=0.13$ is given in terms of the pinning effect of Zn on the dynamical stripe correlations, as proposed by Koike *et al.*[6,7] and Adachi *et al.*[8] That is, the substituted Zn pins a spin domain or a hole domain of the dynamical stripe and makes it statically stabilized. Following this suggestion, the present results for $y \leq 0.01$ in both systems with $x=0.115$ and 0.13 can be understood as follows. Since the period of the stripe pattern of spins and holes at $x=0.115$ is nearly commensurate with that of the crystal lattice,[1] the dynamical stripe correlations tend to be stabilized forming the static stripe order, even in the Zn-free sample. The small increase in T_N as a result of a 1% Zn substitution in the $x = 0.115$ series of samples can be explained as being due to slight enhancement of the static stabilization of the dynamical stripe correlations owing to the pinning force of the substituted Zn. When the hole concentration increases from $x = 0.115$ to 0.13, excess holes destroy the commensurability between the periods of the stripe pattern and the crystal lattice, leading to the destabilization of the static stripe order. As the value of y increases for $x = 0.13$, the substituted Zn tends to pin a spin or hole domain of the dynamical stripe so as to suppress the Cu-spin fluctuations, and finally forces them to be statically stabilized, leading to the formation of the static stripe order and the increase in T_N.

Next, we discuss the Cu-spin state in the large Zn-concentration region for $y \geqslant 0.01$. When the Zn concentration exceeds more than $y = 0.01$, the Zn-substitution effect on T_N changes its sign, because the muon-spin precession observed at 2 K disappears for $y > 0.03$ and no clear decrease in A_0 is observed for $y \geqslant 0.07$. That is, T_N decreases with increasing y. This result also means that even a spin-glass state is not realized. No big difference in the y dependences of T_N and H_{int} between $x = 0.115$ and 0.13 suggests that the magnetic correlation between Cu spins is mainly influenced by the substituted Zn.

The gradual decrease in T_N by a large amount of Zn is also known in the insulating region for $x < 0.02$ in LSCZO.[16,17] In this case, the decrease in T_N is explained as being due to the spin dilution through the Zn substitution. That is, the substituted Zn is regarded as destroying the magnetic correlation between Cu spins. The Zn-concentration dependence of T_N observed in the present μSR study for $y > 0.01$ in both systems with $x = 0.115$ and 0.13 shows a quite similar tendency to that observed in the insulating region for $x < 0.02$. Thus, the decrease in T_N for $y > 0.01$ shown in Fig. 2 is also simply understood to be due to the spin dilution through the Zn substitution. This is evidence that the stripe correlations themselves are being destroyed by the substituted Zn. Accordingly, it is concluded that the magnetic correlation between Cu spins is weakened through the Zn substitution for $y \geqslant 0.01$ and that the Cu-spin state turns into a paramagnetic state rather than a spin-glass state with increasing y.

V. CONCLUSION

As a result, it is concluded from the present μSR study that a small concentration of nonmagnetic impurities tends to statically stabilize the dynamical stripe correlations forming the static long-range-ordered state, whereas a large concentration destroys the stripe correlations themselves. The present results strongly support the conclusion obtained from transport measurements by Koike *et al.*[6,7] and Adachi *et al.*[8]

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