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Possibility of an ordered state of spins and holes in single-crystal $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ ($x=0.21$, $y=0$ and 0.01) studied by μSR

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Zero-field (ZF) and longitudinal-field (LF) muon spin relaxation (μSR) measurements have been carried out in the overdoped single crystals of $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ with $x=0.21$, $y=0$ and 0.01. In the case of the Zn-substituted crystal with $x=0.21$ and $y=0.01$ in which the high-T_c superconductivity is almost suppressed, the ZF- and LF-μSR measurements have revealed the existence of a static ordered state of Cu spins at low temperatures below 2 K. No clear coherent precession of the muon spin has been observed down to 20 mK. Slowing down of the Cu-spin fluctuations has also been observed in the non-Zn-substituted crystal with $x=0.21$ and $y=0$ at low temperatures below about 0.8 K in ZF, indicating that the observed magnetic anomaly is an intrinsic property at $x=0.21$. These μSR results suggest a strong possibility of the existence of a spin/charge ordered state at $x=0.21$ in the overdoped region.

The so-called stripe model of spins and holes[1–3] was originally suggested to explain a magnetically ordered state observed in $La_2NiO_{4+\delta}$ and $La_{2-x}Sr_xNiO_4$. In these nickelates, static stripe order of Ni spins and doped holes appears at various values of the hole concentration, $p=\frac{1}{2}$, $\frac{1}{3}$, and $\frac{1}{4}$ per Ni. Following this result, the static stripe order of spins and holes was suggested in $La_{1.48}Nd_{0.4}Sr_{0.12}CuO_4$ around $p=\frac{1}{8}$ per Cu by the neutron-scattering experiment.[4] Meanwhile, dynamical stripe correlations of spins and holes have been found to exist in a wide range of p from the underdoped to the overdoped region in $La_{2-x}Sr_xCuO_4$ (LSCO),[5] and tend to be statically stabilized around $p=\frac{1}{8}$ per Cu, which is well known as the so-called ‘‘$\frac{1}{8}$ effect.’’[4,6–11] Therefore, on analogy with the case of the nickelates, a sort of stripe order of Cu spins and doped holes is expected to be stabilized in the overdoped region of LSCO.

Actually, Kakinuma *et al.* reported anomalous behavior of the transport phenomena with a little suppression of the supeconductivity in Zn-substituted polycrystalline samples of $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ (LSCZO) around $x=0.22$, suggesting a possibility of the existence of an ordered state of spins and holes in the overdoped region.[12,13] Moreover, Kawamata *et al.*[14] recently developed the sample quality of LSCZO and successfully grew a homogeneous single crystal with $x=0.21$ and $y=0.01$, exhibiting no bulk superconductivity at low temperatures at least above 2 K. They confirmed that bulk superconductivity appeared again as the Sr-doping level shifted to the lighter or heavier side from $x=0.21$ in the Zn-substituted single crystals with $y=0.01$. This fact means that the suppression of superconductivity in the Zn-substituted single crystal occurs singularly at $x=0.21$. Taking into account that the static stabilization of the stripe correlations of spins and holes in the high-T_c cuprates seemed to compete with the superconductivity around $p=\frac{1}{8}$ per Cu in LSCO,[8,9,15–17] they argued that the dynamical stripe correlations of spins and holes tended to be pinned by the substituted Zn in the Zn-substituted single crystal with $x=0.21$. That is to say, a stripe ordered state of spins and holes has been expected in the present single crystals of LSCZO with $x=0.21$. Kawamata *et al.* also succeeded in growing a non-Zn-substituted single crystal with $x=0.21$ and $y=0$ which showed bulk superconductivity with a little suppressed superconducting transition temperature, T_c (T_c is about 25 K). This result suggested that the anomaly observed in LSCZO around $x=0.21$ is an intrinsic property of the La-214 system

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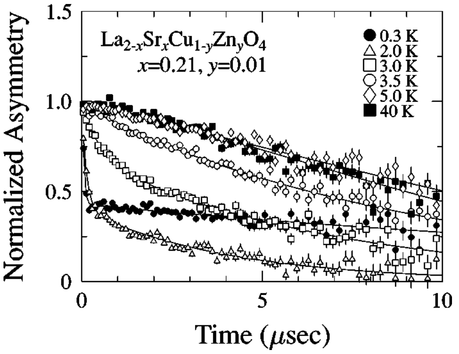

FIG. 1. Zero-field μSR time spectra of the Zn-substituted single crystal of La$_{2-x}$Sr$_y$Cu$_{1-z}$Zn$_y$O$_4$ with $x = 0.21$ and $y = 0.01$ at various temperatures. Fast depolarization behavior of the muon spin is observed at low temperatures below 3.5 K, which is due to slowing down of the Cu-spin fluctuations. Solid lines below 3.5 K are the best fit results using the two-exponential function of $A_1 e^{-\lambda_1 t} + A_0 e^{-\lambda_0 t}$.

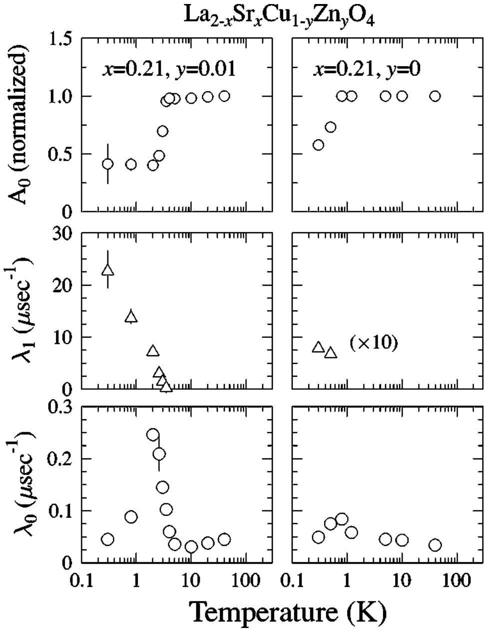

FIG. 2. Temperature dependences of the initial asymmetry of the slow depolarizing component (A_0) and the depolarization rates of the fast (λ_1) and slow (λ_0) depolarizing components in the Zn-substituted ($y = 0.01$) and non-Zn-substituted ($y = 0$) single crystals of La$_{2-x}$Sr$_x$Cu$_{1-y}$Zn$_y$O$_4$ with $x = 0.21$. The initial asymmetries are normalized by the data obtained at 40 K. Two-component analysis using the function of $A_1 e^{-\lambda_1 t} + A_0 e^{-\lambda_0 t}$ is applied at low temperatures where the fast depolarizing component appears.

in the overdoped region. Therefore, we have carried out the muon spin relaxation (μSR) measurements in order to investigate the Cu-spin state in La$_{2-x}$Sr$_x$Cu$_{1-y}$Zn$_y$O$_4$ with $x = 0.21$, $y = 0$ and 0.01.

Single-crystal-growth procedures and transport properties of the grown single crystals of La$_{2-x}$Sr$_x$Cu$_{1-y}$Zn$_y$O$_4$ with $x = 0.21$, $y = 0$ and 0.01 are reported in a separate paper.[14] Zero-field (ZF) and longitudinal-field (LF) μSR measurements were carried out at the RIKEN-RAL Muon Facility (RIKEN-RAL) at the Rutherford-Appleton Laboratory in the UK and at the Meson Science Laboratory at the Institute of Materials Structure Science in the High Energy Accelerator Research Organization (KEK-MSL) in Japan. A pulsed positive surface-muon beam with a momentum of 27 MeV/c was used in both facilities. Forward and backward counters were located on the upstream and downstream sides in the beam direction which was parallel to the initial muon-spin and LF directions. The asymmetry parameter, $A(t)$, was defined as $A(t) = [F(t) - \alpha B(t)]/[F(t) + \alpha B(t)]$, where $F(t)$ 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 the relative counting efficiencies of the forward and backward counters. The initial asymmetry was defined as $A(0)$. The time evolution of the asymmetry parameter (the μSR time spectrum) was measured in ZF and LF. The single crystals were aligned as the CuO$_2$ planes were perpendicular to the initial muon-spin polarization. Low-temperature measurements using a ^{3}He-^{4}He dilution refrigerator from 1 K down to 20 mK were carried out at KEK-MSL and high-temperature measurements above 0.3 K were carried out at RIKEN-RAL.

Figure 1 shows the ZF-μSR time spectra of the Zn-substituted single crystal obtained at various temperatures down to 0.3 K. The time spectrum shows a Gaussian type depolarization behavior above 5 K, indicating that only static nuclear-dipole fields exist at the muon site. The depolarization behavior deviates from the Gaussian-type at low temperatures below 3.5 K, indicating the appearance of a fast depolarizing component. At low temperatures below 2 K, the asymmetry of the muon spin polarization decreases quickly by nearly two thirds within a time range of 0.5 μsec after the arrival of muons in the sample, while the rest one third of the asymmetry depolarize slowly afterwards. The time spectrum has not changed at low temperatures between 0.3 K and 20 mK and clear coherent precession of the muon spin,[6–9] which is similar to that observed in LSCO around $p = \frac{1}{8}$ per Cu, has not been observed even at 20 mK.

The time spectra at high temperatures above 5 K are analyzed using $A_0 e^{-\lambda_0 t} G_Z(\Delta, t)$, where λ_0 is the depolarization rate and A_0 is the initial asymmetry. The term of $G_Z(\Delta, t)$ is the Kubo-Toyabe function[18] which represents the effect of nuclear-dipole fields distributed at the muon site with a distribution width of Δ. After the fast depolarizing component appears below 3.5 K, the analysis using $G_Z(\Delta, t)$ is no longer effective, so that we put $\Delta = 0$ making $G_Z(\Delta, t) = 1$. Consequently, the two-exponential function of $A_1 e^{-\lambda_1 t} + A_0 e^{-\lambda_0 t}$ was used to describe simply the time spectra below 3.5 K, where A_1 and λ_1 are the initial asymmetry and depolarization rate of the fast depolarizing component, respectively. Accordingly, A_0 and λ_0 are regarded as the initial asymmetry and the depolarization rate of the slow depolarizing one, respectively.

Results of the analysis are summarized in Fig. 2. The A_0 decreases suddenly around 2 K with decreasing temperature for the Zn-substituted crystal, which correlates with the increase of λ_1. The λ_0 is enhanced with decreasing temperature and exhibits divergence at 2 K. These are typical characteristics of the case where a static magnetically ordered state of Cu spins appears in the La-214 systems as observed in the previous μSR studies.[8,9]

The LF has been applied to investigate whether the Cu-spin state is dynamic or static at 0.3 K. Figure 3 shows the

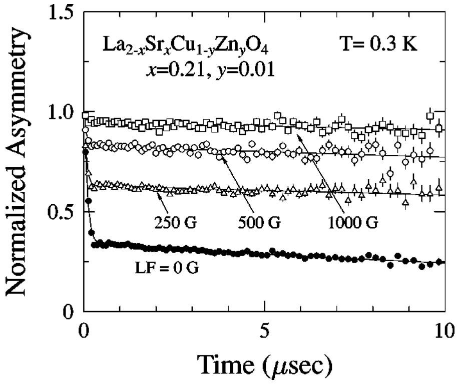

FIG. 3. Variation with the longitudinal field of the μSR time spectra of the Zn-substituted single crystal of $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ with $x=0.21$ and $y=0.01$ at 0.3 K. Solid lines are the best fit results using the two-exponential function of $A_1e^{-\lambda_1 t}+A_0e^{-\lambda_0 t}$.

variation with LF of the time spectrum of the Zn-substituted single crystal at low temperatures. The fast depolarizing component is recovered with increasing LF and almost quenched by the application of LF of 1 kG. In addition, it is found that the time spectrum in LF is nearly flat. These results mean that the observed fast depolarization of the muon spin is caused by a static internal field and that the Cu-spin fluctuations are almost suppressed at 0.3 K. Therefore, it is concluded that a static magnetically ordered state of Cu spins appears in the Zn-substituted single crystal at low temperatures. The magnetic transition temperature is determined from the results shown in Fig. 2 to be about 2 K. The absence of the coherent precession of the muon spin is corresponding to a disordered arrangement of the Cu spins like a spin-glass state.

It has been reported by Kawamata *et al.*[14] that the resistivity of the present Zn-substituted single crystal with $x=0.21$ is metallic at high temperatures above about 80 K, but exhibits upturn below about 80 K in every direction of the crystal axes, showing an insulating behavior. In addition, no Meissner effect has been confirmed at temperatures above 2 K. These results suggest that doped holes are mobile at high temperatures above about 80 K but tend to be localized at lower temperatures. Since a static magnetically ordered state of Cu spins appears at low temperatures below 2 K in the same single crystal, it is suggested that the ground state of this Zn-substituted single crystal is a sort of coexisting state of statically ordered Cu spins and localized holes. Therefore, a plausible explanation of the observed disordered state can be that a stripe ordered state of spins and holes exists in the overdoped region of LSCZO as suggested from the transport measurements by Kawamata *et al.*[14] At the moment, it is hard to discuss the detailed structure of this ordered state of spins and holes from the present μSR study. More μSR measurements of the single crystals around $x=0.21$ are being planned.

In order to study the Zn-substitution effect, ZF-μSR measurements have also been applied to the non-Zn-substituted single crystal of $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ with $x=0.21$ and $y=0$ in which bulk superconductivity is a little suppressed.[14] Figure 4 shows the ZF-μSR time spectra obtained at various temperatures down to 0.3 K. The time spectrum does not change so much at high temperatures above 2 K, exhibiting a Gaussian-type depolarization behavior. This means that the Cu spins are still dynamically fluctuating at higher frequencies outside the μSR time window (typically from 10^{-6} to 10^{-11} sec), while the Cu-spin fluctuations slow down in the Zn-substituted single crystal in the same temperature region. The time spectrum changes at low temperatures below 0.8 K and exhibits an exponential-type deplarization behavior, indicating the slowing down of the Cu-spin fluctuations.

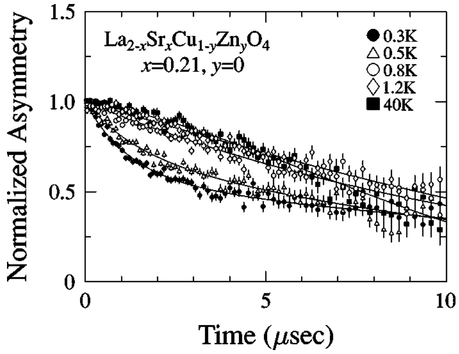

FIG. 4. Zero-field μSR time spectra of the non-Zn-substituted single crystal of $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ with $x=0.21$ and $y=0$ at various temperatures. Fast deplarization behavior of the muon spin appears at low temperatures below 0.8 K, which is due to slowing down of the Cu-spin fluctuations as well as in the Zn-substituted single crystal with $x=0.21$ and $y=0.01$. Solid lines below 0.8 K are the best fit results using the two-exponential function of $A_1e^{-\lambda_1 t}+A_0e^{-\lambda_0 t}$.

The time spectra are analyzed in the same way as those of the Zn-substituted single crystal. The temperature dependence of each parameter is summarized in Fig. 2. The initial asymmetry of the slow component, A_0, starts to decrease below 0.8 K with decreasing temperature and simultaneously the fast depolarizing component starts to appear. The depolarization rate of the slow component, λ_0, seems to exhibit a small peak around 0.8 K.

The tendency of the temperature dependence of each parameter of the non-Zn-substituted single crystal is the same as that observed in the Zn-substituted one. The difference is only the temperature at which the slowing down of the Cu-spin fluctuations starts to be observable by μSR. This difference can be reasonably explained by the model that the dynamical stripe correlations of spins and holes are pinned by the substituted Zn and stabilized being accompanied by the suppression of the Cu-spin fluctuations.[11–14,16] As a result, it is concluded that the slowing-down behavior of the Cu-spin fluctuations is not due to the Zn-substitution effect but to the intrinsic behavior at $x=0.21$ in LSCO. That is to say, the Cu-spin correlation is enhanced at $x=0.21$ and the Zn substitution leads to a static order of Cu spins.

In summary, ZF- and LF-μSR measurements have been carried out in the single crystals of $La_{2-x}Sr_xCu_{1-y}Zn_yO_4$ with $x=0.21$, $y=0$ and 0.01. Fast depolarization of the muon spin has been observed at low temperatures below 3.5 K in the Zn-substituted crystal and below 0.8 K in the non-Zn-substituted one. This means that slowing down of the Cu-spin fluctuations appears at low temperatures in both single crystals and that the slowing down of the Cu-spin fluctuations is an intrinsic behavior of LSCO with $x=0.21$. A static magnetically ordered state of Cu spins has been confirmed at low temperatures below 2 K in the Zn-substituted single-crystal. Taking into account the results of transport measurements,[14] a coexisting state of statically ordered Cu spins and localized holes is expected to exist below 2 K in the Zn-substituted single crystal. A plausible explanation of this coexisting state is suggested to be that a stripe ordered state of spins and holes exists at low temperatures at $x=0.21$ in LSCO in the overdoped region also.

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