Anomalous muon-spin relaxation in the Zn-substituted $YBa_2Cu_{3-2y}Zn_{2y}O_{7-\delta}$ around the hole concentration of $\frac{1}{8}$ per Cu

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Muon-spin relaxation (μSR) measurements were carried out in the partially Zn-substituted Y-123 system, $YBa_2Cu_{3-2y}Zn_{2y}O_{7-\delta}$, to investigate the so-called 1/8 effect. The muon-spin depolarization rate has been found to be anomalously enhanced at low temperatures in the Zn-substituted samples around the hole concentration per Cu in the CuO_2 plane $p\sim1/8$, as observed in the case of the Zn-substituted $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ with $p\sim1/8$. This indicates slowing down of the Cu-spin fluctuations at $p\sim1/8$. It appears that a small amount of Zn ions are effective for the enhancement of the magnetic correlation between Cu spins at $p\sim1/8$. It has been concluded that the 1/8 effect exists in the Y-123 system and is common to all high-T_c cuprates.

I. INTRODUCTION

The so-called 1/8 problem, namely, the anomalous suppression of superconductivity at p (the hole concentration per Cu in the CuO_2 plane) $\sim1/8$ in the La-214 system of high-T_c cuprates is a long-standing one.[1,2] However, the stripe model proposed by Tranquada *et al.*[3] is a plausible one, which can explain the 1/8 effect. It has been proposed that the static order of spins and/or holes observed in the La-214 system with $p\sim1/8$ from the elastic neutron-scattering experiments[3–5] is due to pinning of the dynamical stripe correlations of spins and holes, which are guessed to exist from the observation of incommensurate peaks near $\boldsymbol{k}=(\pi,\pi)$ in the inelastic neutron-scattering experiments.[6–9] That is, the suppression of superconductivity has been interpreted as being due to the static stabilization of the dynamical stripe correlations. In the La-214 system, the tetragonal low-temperature structure (space group $P4_2/ncm$) is regarded as the pinning centers. A small amount of Zn ions substituted for Cu are also regarded as the pinning centers.[10–12]

Supposing the dynamical stripe correlations are characteristic of the CuO_2 plane, the 1/8 effect seems not to be peculiar to the La-214 system but common to all high-T_c cuprates with the CuO_2 plane when adequate pinning centers are introduced into a sample. In fact, the incommensurate peaks around $\boldsymbol{k}=(\pi,\pi)$, which are associated with those of the La-214 system, have been observed in the inelastic neutron-scattering measurements of the Bi-2212 and Y-123 systems also.[13–16] Moreover, in Zn-substituted ($y=0.025$) $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$, we have found the 1/8 effect, namely, the anomalous suppression of superconductivity at $p\sim1/8$ (Ref. 17) and also found anomalous slowing down of the Cu-spin fluctuations at $p\sim1/8$ from the muon-spin-relaxation (μSR) measurements.[18–21] According to the stripe model, these experimental results have been interpreted as being due to the pinning effect of substituted Zn ions on the dynamical stripe correlations, though the dynamical correlations have not been pinned so completely as to be statically stabilized.

As for the Y-123 system of $YBa_2Cu_3O_{7-\delta}$, it is well known that the superconducting transition temperature T_c exhibits the so-called 60-K plateau in the T_c vs oxygen content $7-\delta$ plot.[22–25] The 60-K plateau of T_c was explained as being due to ordering of oxygen vacancies in the $CuO_{1-\delta}$ chain.[25,26] That is to say, it was understood that oxygen vacancies in the $CuO_{1-\delta}$ chain were in order for samples around $7-\delta=6.6$ and that the ordering made the samples more conductive and enhanced T_c, leading to the appearance of the plateau of T_c around $7-\delta=6.6$. However, Tallon *et al.*[27] have pointed out from the thermoelectric power, NMR, and specific heat measurements that the 60-K plateau is related to the 1/8 effect. Moreover, we have found from the transport measurements of $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ ($x=0$ and 0.2) that the 60-K plateau is not correlated with $7-\delta$ but is correlated with p and proposed that the 60-K plateau is interpreted as being due to the suppression of superconductivity at $p\sim1/8$, namely, the 1/8 effect rather than the ordering of oxygen vacancies in the $CuO_{1-\delta}$ chain.[28] A similar result has also been reported by Tallon *et al.*[29]

In this paper, we have carried out the zero-field (ZF)

μSR measurements in the partially Zn-substituted ($y = 0.025$) and non-Zn-substituted ($y = 0$) samples of $YBa_2Cu_{3-2y}Zn_{2y}O_{7-\delta}$ with various values of $7-\delta$, in order to investigate the Cu-spin state around $p = 1/8$ and confirm the existence of the 1/8 effect in the Y-123 system. Here, Zn is introduced into a sample to pin the possible dynamical stripe correlations of spins and holes. As Zn is known to be mainly substituted for Cu in the CuO_2 plane, y in $YBa_2Cu_{3-2y}Zn_{2y}O_{7-\delta}$ is regarded as the Zn concentration in the CuO_2 plane.[30]

II. EXPERIMENT

Sintered samples of $YBa_2Cu_{3-2y}Zn_{2y}O_{7-\delta}$ with $y = 0$ and 0.025 were prepared by the conventional solid-state reaction method. Raw materials of Y_2O_3, $BaCO_3$, CuO, and ZnO powders were mixed and prefired at 900 °C for 12 h in air. The prefired materials were mixed, pelletized, and sintered in air at 930 °C for 24 h, following by furnace cooling. This sintering process was repeated. Post-annealing was performed in O_2 at 500°C for 24 h. In this stage, fully oxidized samples with small values of δ were prepared. The oxygen content was changed by subsequent annealing and quenching. That is, the fully oxidized samples were again heated in air at 900 °C for 12 h and cooled down to a temperature T_q, at a rate of 200 °C/h and kept at T_q for 12 h, following by quenching into liquid nitrogen. All products were characterized by powder x-ray diffraction to be of the single phase. The oxygen content was determined by iodometric titration. The p value was estimated from the value of the thermoelectric power at 290 K, $S_{290\,K}$, using the universal relation between p in the CuO_2 plane and $S_{290\,K}$, which is empirically known to hold good in most of the high-T_c cuprates including the Y-123 phase.[27,28,31] The electrical resistivity was measured by the dc four-point probe method to determine T_c.

ZF-μSR measurements were carried out at Muon Science Laboratory, Institute of Materials Structure Science, High Energy Accelerator Research Organization (KEK-MSL) in Japan. A spin-polarized pulsed surface-muon beam with a momentum of 27 MeV/c was used. A ^{4}He gas-flow-type cryostat was used for the measurements at temperatures above 2 K. The asymmetry parameter is defined as $A(t) = [F(t) - \alpha B(t)]/[F(t) + \alpha B(t)]$, where $F(t)$ and $B(t)$ are muon events counted by forward and backward counters at a time t, respectively. α is a calibration factor reflecting the relative counting efficiencies of the forward and backward counters.

III. RESULTS AND DISCUSSION

Figure 1 displays typical ZF-μSR time spectra of the non-Zn-substituted ($y = 0$) and Zn-substituted ($y = 0.025$) samples of $YBa_2Cu_{3-2y}Zn_{2y}O_{7-\delta}$ with $7-\delta = 6.75$ ($p > 1/8$), 6.65−6.67 ($p \sim 1/8$), and 6.41−6.56 ($p < 1/8$) at various temperatures. The time spectra at high temperatures are of the Gaussian type for every sample. This means that muon spins are depolarized mainly by randomly distributed nuclear dipole fields. For $7-\delta = 6.75$ ($p > 1/8$), it is found that the time spectrum does not change even at low temperatures in both of the non-Zn-substituted and Zn-substituted samples. For $7-\delta = 6.65-6.67$ ($p \sim 1/8$), on the other hand, it is found that the time spectrum of the Zn-substituted sample changes from the Gaussian type to the exponential type with decreasing temperature at low temperatures below ~5 K, while the time spectrum of the non-Zn-substituted sample does not change even at low temperatures. This is similar to the case of the Zn-substituted samples of $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ with $p \sim 1/8$,[18–21] and suggests that the Cu-spin fluctuations slow down at low temperatures also in the Zn-substituted sample of $YBa_2Cu_{3-2y}Zn_{2y}O_{7-\delta}$ with $p \sim 1/8$. For $7-\delta = 6.41-6.47$ ($p < 1/8$), it is found that the time spectrum changes to the exponential type at low temperatures in both of the non-Zn-substituted and Zn-substituted samples. This is interpreted as being due to static or nearly static Cu spins in the so-called spin-glass-like phase, which appears at low temperatures in the strongly underdoped region, as formerly reported in the Y-123 system.[32,33] Similar results have also been obtained at $p < 1/8$ in the Bi-2212 system.[20,21] What is remarkable in Fig. 1 is that the time spectrum of the Zn-substituted sample with $7-\delta = 6.56$ at 2 K is not so exponential as that of the Zn-substituted sample with $7-\delta = 6.65$ ($p \sim 1/8$) at 2 K.

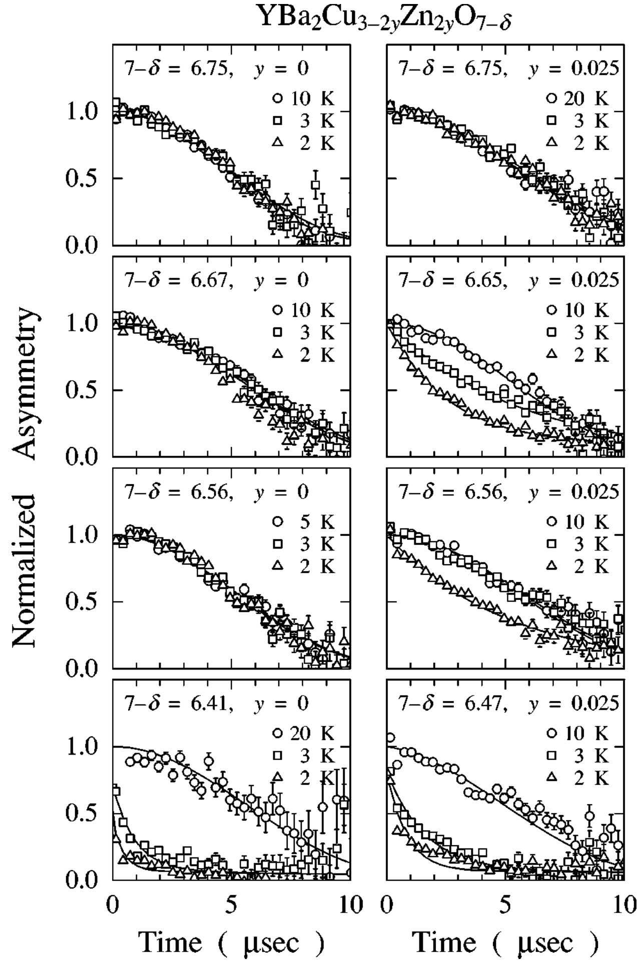

FIG. 1. ZF-μSR time spectra of the non-Zn-substituted ($y = 0$) and Zn-substituted ($y = 0.025$) samples of $YBa_2Cu_{3-2y}Zn_{2y}O_{7-\delta}$ with $7-\delta = 6.75$ ($p > 1/8$), 6.65−6.67 ($p \sim 1/8$), and 6.41−6.56 ($p < 1/8$) at various temperatures. Solid lines are the best fit of $A_1 e^{-\lambda_1 t} G_z(\Delta, t)$ or $A_1 e^{-\lambda_1 t} + A_2 e^{-\lambda_2 t}$ to the time spectra.

This suggests that the Cu-spin fluctuations exhibit slowing-down behavior singularly at $p\sim 1/8$ in the Zn-substituted samples.

The Gaussian-type behavior of the time spectrum is fitted using a single-component function $A_1 e^{-\lambda_1 t} G_z(\Delta,t)$, where A_1 and λ_1 are the initial asymmetry at $t=0$ and the depolarization rate of muon spins, respectively. $G_z(\Delta,t)$ is the static Kubo-Toyabe function with a field-distribution width of Δ/γ_μ at the muon sites, where γ_μ is the gyromagnetic ratio of the muon spin ($2\pi\times 13.554$ MHz/kG). As for the exponential-type behavior of the time spectrum, the time spectrum of the Zn-substituted sample with $7-\delta=6.65$ at 2 K cannot be analyzed using a simple exponential function. Therefore, a two-exponential-component function, $A_1 e^{-\lambda_1 t} + A_2 e^{-\lambda_2 t}$, is used for the analysis. Here, A_1 and A_2 are initial asymmetries at $t=0$ and λ_1 and λ_2 are depolarization rates of the fast- and slow-depolarization components, respectively. This analysis means that two kinds of muon sites are taken into account.

It is known that muons injected into a sample of a high-T_c cuprate sit adjacent to oxygen ions.[34] From the previous μSR studies, there are expected to be roughly two kinds of muon sites in the Y-123 system; one is adjacent to the in-plane and apical oxygen ions surrounding Cu in the CuO_2 plane, and the other is adjacent to the oxygen ions in the $CuO_{1-\delta}$ chain.[35,36] Muons sitting adjacent to the in-plane and apical oxygen ions feel strong internal fields from the Cu spins in the CuO_2 plane, and muons sitting adjacent to the oxygen ions in the $CuO_{1-\delta}$ chain that are far away from the Cu spins in the CuO_2 plane feel weaker internal fields, because the internal fields at the muon sites are dominated by the rapidly decaying dipolar fields of the Cu spins in the CuO_2 plane. Because of this difference in the internal field, the depolarization of the muon spins near the in-plane and apical oxygen ions surrounding Cu in the CuO_2 plane is expected to be faster than that of the muon spins near the oxygen ions in the $CuO_{1-\delta}$ chain. Accordingly, the ratio of A_1:A_2 is taken as $6/(7-\delta)$:$(1-\delta)/(7-\delta)$ on the assumption that a muon sits adjacent to each oxygen ion equally. Solid lines in Fig. 1 show the best fit results. It appears that the two-component analysis is reasonable.[36] This kind of two-component analysis also proved successful in the previous μSR study on the Zn-substituted Bi-2212 system.[18–21]

Figure 2 shows the dependences on the oxygen content $7-\delta$ of T_c and the muon-spin depolarization rate λ_1 at 2 and 3 K. T_c is defined as the midpoint of the superconducting transition curve in the resistivity vs T plot. It is clearly found that the depolarization rate shows a local maximum around $7-\delta=6.65$, namely, $p\sim 1/8$, in the Zn-substituted samples with $y=0.025$. This local maximum indicates drastic slowing down of the Cu-spin fluctuations in the Zn-substituted samples with $p\sim 1/8$. This is the same anomaly as observed at $p\sim 1/8$ in the Zn-substituted samples of $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ with $y=0.025$.[19–21] The local maximum of λ_1 at $p\sim 1/8$ is roughly in correspondence to the local suppression of superconductivity, as seen in Fig. 2. In the non-Zn-substituted samples, no enhancement of λ_1 is observed at $p\sim 1/8$. Therefore, it is found that both Zn-substitution and $p\sim 1/8$ are essential for the slowing down of the Cu-spin fluctuations. These results are analogous to the 1/8 effect in the La-214 system and very similar to the 1/8 effect in the Bi-2212 system. Therefore, it is concluded that the same 1/8 effect exists in the Y-123 system as well.

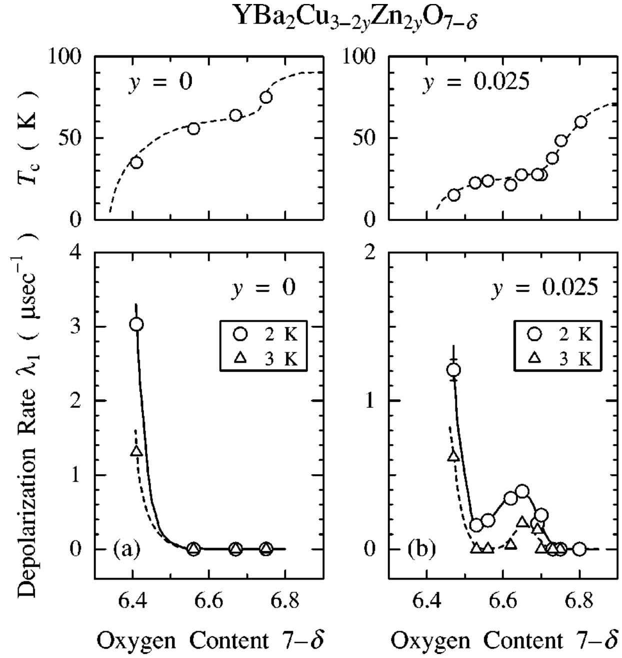

FIG. 2. Dependences on the oxygen content $7-\delta$ of T_c and the zero-field muon-spin depolarization rate λ_1 at 2 and 3 K for (a) the non-Zn-substituted ($y=0$) and (b) the Zn-substituted ($y=0.025$) samples of $YBa_2Cu_{3-2y}Zn_{2y}O_{7-\delta}$. T_c is defined as the midpoint of the superconducting transition curve in the resistivity vs T plot. Lines are guides to the eye.

Figure 3 shows the temperature dependence of the muon-spin depolarization rate λ_1 of all of the measured samples. The depolarization rate of the non-Zn-substituted samples with $7-\delta\geqslant 6.56$ shows a very weak temperature dependence above 2 K. The Zn-substituted samples with $7-\delta\geqslant 6.73$ also shows a similar weak temperature dependence. These results indicate that the static and dynamical properties of Cu spins do not change in the case of $7-\delta\geqslant 6.73$, even in the Zn-substituted samples. The depolarization rates of the non-Zn-substituted sample with $7-\delta=6.41$ and the Zn-substituted sample with $7-\delta=6.47$ in the strongly underdoped region increase with decreasing temperature at low temperatures below 3–5 K, which is due to static or nearly static Cu-spins in the spin-glass-like phase, as mentioned before. It is found that the depolarization rate of the Zn-substituted sample with $7-\delta=6.65$ ($p\sim 1/8$) increases with decreasing temperature at low temperatures below 3–5 K. In the Zn-substituted $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ with $y=0.025$ and $p\sim 1/8$, the similar increase of the depolarization rate with decreasing temperature was observed below about 10 K.[19]

Previously, we have suggested the existence of the 1/8 effect in the non-Zn-substituted Y-123 system.[28] So, the muon-spin depolarization was expected to be enhanced also in the non-Zn-substituted sample with $p\sim 1/8$, but no anomaly has been observed for the sample by the present μSR measurements. The time window of μSR is typically 10^{-6}–10^{-11} sec. That is to say, when the characteristic time of the Cu-spin fluctuations is much longer than $\sim 10^{-6}$ sec, muons feel almost static fields of the Cu spins in their lifetime. When the time is much shorter than $\sim 10^{-11}$ sec, on the

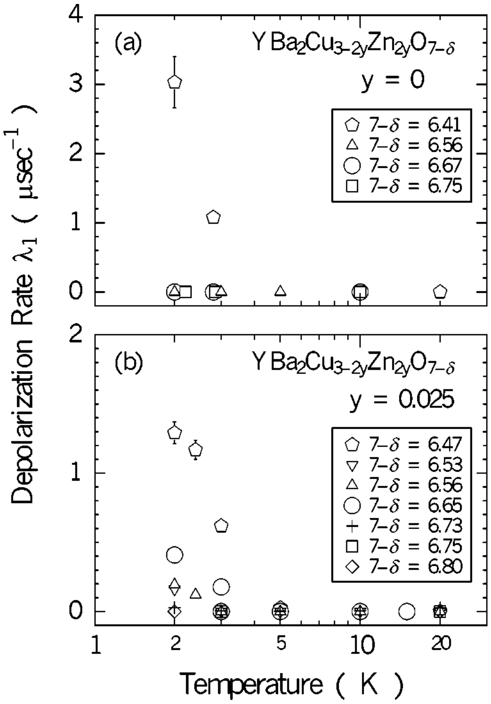

FIG. 3. Temperature dependence of the zero-field muon-spin depolarization rate λ_1 for (a) the non-Zn-substituted ($y=0$) and (b) the Zn-substituted ($y=0.025$) samples of $YBa_2Cu_{3-2y}Zn_{2y}O_{7-\delta}$.

other hand, muons feel almost zero averaged field due to the Cu spins, leading to no depolarization due to the Cu spins. Considering that the superconductivity is suppressed in the non-Zn-substituted samples with $p\sim1/8$, the Cu-spin fluctuations are expected to slow down to some extent. Therefore, we suppose that the slowing down has not been detected by the μSR measurements because their characteristic time remains shorter than $\sim10^{-11}$ sec.

Accordingly, it appears that a small amount of Zn ions are effective for the enhancement of the magnetic correlation between Cu spins at $p\sim1/8$ as in the case of the Bi-2212 system.[17–21] On the other hand, we suppose that a large amount of Zn ions destroy the magnetic correlation between Cu spins and hence the Cu-spin fluctuations remain fast. In the former μSR measurements on the heavily Zn-substituted samples of the Y-123 system, in fact, no slowing down behavior at $p\sim1/8$ was reported.[33] These results are consistent with the stripe model. That is, a small amount of Zn ions are regarded as operating to pin the dynamical stripe correlations, leading to the slowing down of the Cu-spin fluctuations.

In the Zn-substituted Y-123 system with $p\sim1/8$, three-dimensional long-range ordering of the Cu spins, as observed in the La-214 system,[37] has not been observed at temperatures above 2 K. The same was found for the Zn-substituted Bi-2212 system with $p\sim1/8$.[18–21] This may mean that a coherent long-range-ordered state of the Cu spins is hard to be stabilized in the Zn-substituted Y-123 and Bi-2212 systems, because the pinning force of one Zn ion is much more short-range and weaker than that of the tetragonal low-temperature structure in the La-214 system or because Zn ions pinning the stripes within a plane are not correlated from plane to plane so that there is no correlation of the stripes plane to plane. In order to study the state of the Cu spins in detail, further μSR measurements in longitudinal magnetic fields at lower temperatures will be necessary.

IV. SUMMARY

μSR measurements were carried out in the partially Zn-substituted ($y=0.025$) and non-Zn-substituted ($y=0$) samples of $YBa_2Cu_{3-2y}Zn_{2y}O_{7-\delta}$. Anomalous enhancement of the muon-spin depolarization rate has been found at low temperatures in the Zn-substituted samples with $p\sim1/8$, as previously observed in Zn-substituted samples of $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ with $y=0.025$ and $p\sim1/8$. This indicates that the Cu-spin fluctuations slow down into the μSR time window at low temperatures. It appears that a small amount of Zn ions are effective for the enhancement of the magnetic correlation between Cu spins at $p\sim1/8$. On the analogy with the 1/8 effect in the La-214 and Bi-2212 systems, there is a possibility that a kind of dynamical order of holes and/or spins, such as the so-called stripe order, exists also in the Y-123 system and that it tends to be pinned by Zn at $p\sim1/8$, leading to the anomalous enhancement of the muon-spin depolarization rate and the suppression of superconductivity at $p\sim1/8$. It is concluded that the 1/8 effect probably exists in the Y-123 system and is common to all high-T_c cuprates.

To be more conclusive, further experiments, such as direct observation of the possible order of spins and/or holes by means of electron diffraction and neutron diffraction, are necessary in the Zn-substituted $YBa_2Cu_{3-2y}Zn_{2y}O_{7-\delta}$ with $p\sim1/8$.

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