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Anomalous slowing down of Cu-spin fluctuations observed by muon spin relaxation in the Zn-substituted $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ system around the hole concentration of $\frac{1}{8}$ per Cu

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Muon spin relaxation (μSR) measurements were applied to a partially Zn-substituted Bi-2212 system, $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$, to study a possibility of the existence of the so-called "$\frac{1}{8}$ effect" which has been established in La systems. The muon spin depolarization rate has been found to increase with decreasing temperature below 10 K in Zn-substituted samples around the hole concentration $p=\frac{1}{8}$ per Cu where the high-T_c superconductivity is anomalously suppressed. It has been revealed that the enhancement of the muon spin depolarization rate is attributed to the slowing down of the Cu-spin fluctuations and that both Zn substitution and $p=\frac{1}{8}$ per Cu are essential for the slowing down of the Cu-spin fluctuations. Although no three-dimensional long-range coherent ordering of the Cu spins appeared down to 0.30 K, the present μSR results support the existence of the $\frac{1}{8}$ effect in a Bi-2212 system also. [S0163-1829(99)50838-2]

Anomalous suppression of the high-T_c superconductivity (SC) within a narrow region of the hole concentration $p\sim\frac{1}{8}$ per Cu was first observed in $La_{2-x}Ba_xCuO_4$ (LBCO) around $x=0.125$.[1,2] This phenomenon is well known as the so-called $\frac{1}{8}$ effect. A structural phase transition from orthorhombic (space group: *Bmab*) to tetragonal (space group: $P4_2/ncm$) was discovered in this hole-concentration range, indicating a relationship between the anomalous suppression of the SC and the structural change.[3] A similar phenomenon was also found in $La_{2-x}Sr_xCuO_4$ (LSCO), although no structural transition was observed except for instability of a phonon mode.[4]

Muon spin relaxation (μSR) measurements using a positive muon beam were performed for both LBCO and LSCO and revealed the existence of an antiferromagnetically (AF) ordered state where the SC was anomalously suppressed.[5–7] The magnetic transition temperature showed a maximum at $x=0.125$ for LBCO and at $x=0.115$ for LSCO (Ref. 8). Subsequently, from neutron-diffraction measurements on Nd-substituted LSCO (LNSCO) undergoing a structural transition,[9] Tranquada *et al.* suggested the existence of a stripe structure of a spin-density wave (SDW) state and a charge-density wave (CDW) state.[10] However, the detailed mechanism of the $\frac{1}{8}$ effect and the ordered state of spins and charges are still in controversy.

At the moment, the $\frac{1}{8}$ effect has been established only in La systems. Thus, it is important to investigate it in other high-T_c cuprates which have different crystal structures from those of the La systems in order to clarify the relationship between the crystal structure and the $\frac{1}{8}$ effect.

Recently, Akoshima *et al.* showed one possibility. That is, they found anomalous suppression of the high-T_c SC in the partially Zn-substituted Bi-2212 system, $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$, around $p=\frac{1}{8}$ per Cu ($x=0.3125$), though a structural transition similar to those observed in LBCO (Ref. 3) and LNSCO (Ref. 9) did not take place.[11] They suggested that the substituted Zn atoms acted as charge and/or spin pinning centers, and that the SC was suppressed by the similar mechanism to the $\frac{1}{8}$ effect. The anomalous suppression of the SC has been confirmed by transport measurements on the Zn-substituted Bi-2212 system, while the existence of a magnetically ordered state, which is expected to be derived from the $\frac{1}{8}$ effect, has not yet been confirmed.

In this paper, we report on the results of zero field (ZF) and longitudinal field (LF) μSR measurements in a partially Zn-substituted Bi-2212 system. We have observed enhancement of the muon spin depolarization rate at low temperatures below 10 K in Zn-substituted samples around $p=\frac{1}{8}$ per Cu, indicating the slowing down of the Cu-spin fluctuations. It has been found that both Zn-substitution and $p=\frac{1}{8}$ per Cu are essential for the slowing down of the Cu-spin fluctuations. Our results support the existence of the $\frac{1}{8}$ effect in the Bi-2212 system.

Samples of $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ were prepared by the conventional solid-state reaction method.[11] We prepared three sorts of polycrystalline samples with different hole concentrations: $x=0.20$ ($p>\frac{1}{8}$ per Cu), $x=0.3125$ ($p\sim\frac{1}{8}$ per Cu), and $x=0.40$ ($p<\frac{1}{8}$ per Cu). Zn concentrations

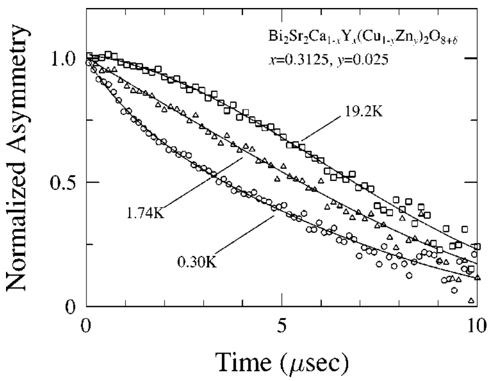

FIG. 1. Zero-field μSR time spectra of Zn-substituted $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ with $x=0.3125$ and $y=0.025$ at 19.2, 1.74, and 0.30 K. The solid lines are the best fit of Eq. (1). The high-T_c superconductivity of this sample is anomalously suppressed (Ref. 11).

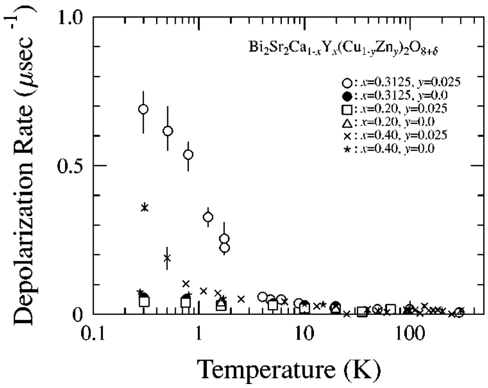

FIG. 2. Temperature dependence of zero-field muon spin depolarization rate of $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ with $x=0.20$, 0.3125, 0.40 and $y=0$, 0.025. The anomalous enhancement below 4 K of Zn-substituted samples with $x=0.3125$ and $x=0.40$ is due to a temperature dependence of the fast depolarization rate, λ_1. In these cases, the slow depolarization rates, λ_2, show similar small values with those obtained at higher temperatures.

were $y=0$ and $y=0.025$ for each x value. All of the samples were checked to be of the single phase, and the characterization of the samples was performed by resistivity and susceptibility measurements.

μSR measurements were carried out at the RIKEN-RAL Muon Facility at the Rutherford-Appleton Laboratory in the United Kingdom. A spin-polarized pulsed surface muon beam with a momentum of 27 MeV/c was used. Forward and backward counters were located at the upstream and downstream sides of the sample in a beam direction. The muon spin polarization was parallel to the beam. The asymmetry parameter was defined as $A(t)=[F(t)-B(t)]/[F(t)+B(t)]$, where $F(t)$ and $B(t)$ were the total muon events counted by the forward and backward counters at time t, respectively. A LF was applied along the muon spin polarization. The time evolution of the asymmetry parameter (μSR time spectrum) was measured.

Figure 1 shows ZF-μSR time spectra of a Zn-substituted sample with $x=0.3125$ and $y=0.025$ in which the high-T_c SC is anomalously suppressed. The time spectrum above 10 K shows a Gaussian-type depolarization behavior, indicating that no significant magnetic effect exists at the muon sites except for randomly distributed nuclear dipolar fields. The time spectrum deviates from a Gaussian type below 10 K. At low temperatures, it shows an exponential-type depolarization behavior, as reported from a preliminary experiment.[12] This change in the time spectrum suggests the appearance of an additional effect due to local fields. Since the magnetic properties of this system are ruled by the Cu spins, it is inferred that this additional effect reflects their static or dynamical properties.

In the analysis of the time spectra, the difference of the muon sites should be taken into account. It is known that muons injected in a high-T_c oxide stop near oxygen ions.[13] From the comparison with previous μSR studies in Y (Ref. 14) and Tl systems,[15] there are expected to be roughly two kinds of muon sites in a Bi-2212 system; sites near the oxygen ions constituting the CuO_5 pyramid and sites near the oxygen ions in the Bi-O plane. Muons stopping near the CuO_5 pyramid feel strong internal fields from the Cu spins, and muons stopping near the Bi-O plane which are far away from the CuO_5 pyramid feel weaker internal fields, because the internal fields at the muon sites are dominated by the dipolar fields of the Cu spins.[16] Because of this difference of internal fields, the depolarization of the muon spins near the CuO_5 pyramid is expected to be faster than that of the muon spins near the Bi-O plane. In fact, two components (fast and slow) of the depolarization behavior were already observed in a magnetically ordered state of an insulating Bi-2212 system by Nishida *et al.*[16] Therefore, we take a two-component function to describe the observed exponential-type time spectrum, as given by

$$A_1 e^{-\lambda_1 t} + A_2 e^{-\lambda_2 t} \times G_Z(\Delta, H_{LF}, t). \qquad (1)$$

The first term describes muons which are close to the CuO_5 pyramid and depolarize mainly by the additional fields from the Cu spins. The second term describes the other muons which are far away from the CuO_5 pyramid and depolarize mainly by the randomly distributed nuclear dipolar fields with a weak effect of the Cu spins. $G_Z(\Delta, H_{LF}, t)$ indicates the static Kubo-Toyabe function, where Δ is the distribution width of the nuclear dipolar fields at the muon sites.[17] H_{LF} is the LF. A_1 and A_2 are the initial asymmetries of each component at $t=0$. In the case of ZF-μSR, we put zero in H_{LF}.

The solid lines in Fig. 1 show the best fit of Eq. (1) to the time spectra. λ_1 is close to λ_2 at high temperatures, which means that the effect of the internal fields from the Cu spins becomes small. Therefore, it is difficult to apply the two-component fitting. In this case, a single-component analysis using only the second term is adopted for convenience.

Figure 2 shows the temperature dependence of the depolarization rate of all of the measured samples down to 0.30 K. The depolarization rate of the non-Zn-substituted samples shows a very weak temperature dependence within the measured temperature range. The Zn-substituted sample with $x=0.20$ also shows a similar weak temperature dependence. These results indicate that the static and dynamical properties of the Cu spins do not change in the case of $p>\frac{1}{8}$ per Cu even in a Zn-substituted sample. However, it is found that

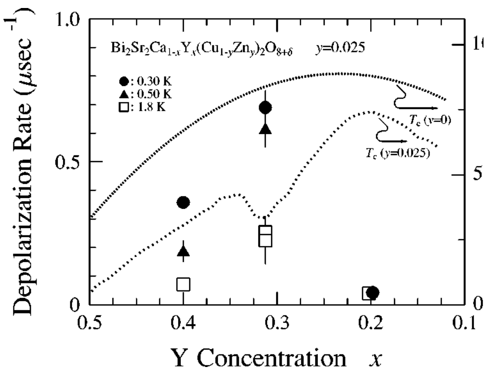

FIG. 3. x dependence, namely, the hole-concentration dependence of the zero-field muon spin depolarization rate, λ_1, of Zn-substituted $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ with $y=0.025$ at 0.30, 0.50, and 1.8 K. The depolarization rate is anomalously enhanced at $x=0.3125$ ($p\sim\frac{1}{8}$ per Cu). Dotted lines show the x dependence of T_c determined by Akoshima *et al.* from transport measurements with $y=0$ and 0.025 (Ref. 11).

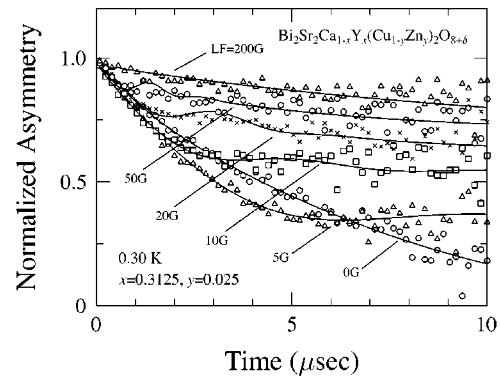

FIG. 4. Longitudinal-field dependence of μSR time spectra of the Zn-substituted $Bi_2Sr_2Ca_{1-x}Y_x(Cu_{1-y}Zn_y)_2O_{8+\delta}$ with $x=0.3125$ and $y=0.025$ at 0.30 K. The solid lines are the best fit of Eq. (1).

the depolarization rate of the Zn-substituted samples with $x=0.3125$ and $x=0.40$ increases with decreasing temperature below about 10 K.

If the muon spins depolarize by dynamical effects of the Cu spins, the enhancement of the depolarization rate means that the Cu-spin fluctuations are suppressed with decreasing temperature, exhibiting the so-called slowing-down behavior. On the other hand, if they depolarize by static effects of the Cu spins, the enhancement of the depolarization rate means that some Cu spins freeze below 10 K and make additional static internal fields at the muon sites which cause this exponential type depolarization of the muon spins. This static depolarization process can be realized in the case that the distribution of the static internal fields is of an exponential type. Actually, such an exponential depolarization behavior by the static internal fields was already observed in other materials.[17,18] In any case, the enhancement of the muon spin depolarization rate suggests that the Cu-spin fluctuations slow down with decreasing temperature in Zn-substituted samples. This fact implies that the magnetic correlation between Cu spins is expected to be enhanced in Zn-substituted samples resulting in the slowing down of the Cu-spin fluctuations.

Figure 3 shows the x dependence, namely, the hole-concentration dependence of the depolarization rate of the Zn-substituted samples at 0.30, 0.50, and 1.8 K. The depolarization rate exhibits a maximum at $x=0.3125$ where $p\sim\frac{1}{8}$ per Cu and the high-T_c SC is anomalously suppressed.[11,19] Since no enhancement in the depolarization rate is observed in the non-Zn-substituted samples, it is found that both Zn substitution and $p\sim\frac{1}{8}$ per Cu are essential for the slowing down of the Cu-spin fluctuations. This relationship between the slowing down of the Cu-spin fluctuations and the SC, revealed in the present μSR studies, is analogous to that of the $\frac{1}{8}$ effect which has been established in La systems.[6,8] Therefore, in conjunction with the results of the transport measurements,[11] we can conclude that the $\frac{1}{8}$ effect is likely to exist in the Bi-2212 system also. This conclusion suggests that the $\frac{1}{8}$ effect is characteristic of the CuO_2 plane of high-T_c cuprates.

A three-dimensional (3D) long-range ordered state, as observed in La systems,[6,8,10] has not been observed down to 0.30 K in a Zn-substituted Bi-2212 system. This is a big difference in the state of the Cu spins between the La systems and the Bi-2212 system around $p=\frac{1}{8}$ per Cu. This result shows that the coherent long-range ordered state of the Cu-spins is difficult to be stabilized in the Zn-substituted Bi-2212 system. In the case of LNSCO, it is suggested that the coherent long-range ordered state (the stripe structure of SDW and CDW states) is derived from the pinning of the dynamical stripe correlations of spins and holes.[10] Assuming that the $\frac{1}{8}$ effect is due to the stripe structure, no observation of the long-range ordered state indicates that the pinning force due to Zn atoms in a Zn-substituted Bi-2212 system is much weaker than that in LNSCO. A 3D ordering may appear at lower temperatures. μSR measurements at lower temperatures using a ^{3}He-^{4}He dilution refrigerator are now planned to search for the long-range ordered state.

LF was applied to the Zn-substituted sample with $x=0.3125$ and $y=0.025$ at 0.30 K to investigate the magnetic state of the Cu spins. Figure 4 shows the LF dependence of the time spectrum at 0.30 K. The LF-μSR time spectra are analyzed with Eq. (1). Solid lines in the figure are the best fit. The depolarization is found to be suppressed by LF. This means that the additional effect from the Cu spins is quenched by LF.

It is found from this analysis that the distribution width of the static internal fields, Δ, increases linearly up to a few tens Gauss with increasing LF. When a polycrystalline sample is used, the distribution of the applied fields in the sample is usually inhomogeneous and complicate in the bulk SC state because of intricate giometrical conditions among grains and large anisotropy of the electronic state. Since the contribution of the nuclear dipolar fields is at most 1 G in this system, the wider field distribution seen at the muon sites is expected to be due to the inhomogeneity of the applied fields in the sample. This result may suggest that the bulk SC is still left in the sample.

The decoupling behavior in LF shown in Fig. 4 indicates a possibility that the additional internal fields which induce an exponential-type depolarization behavior are static. In this case, the Cu spins are expected to be frozen at low temperatures and strongly disordered, because no coherent precession of the muon spins is observed. On the other hand, the effect of additional fields can be also understood assuming a dynamical reason, namely, the slowing down of the Cu-spin fluctuations. This is because a similar change in the depolarization behavior from a Gaussian type to an exponential type was observed in La systems just above the 3D ordering temperature, and this exponential-type depolarization behavior was concluded to be due to dynamically fluctuating Cu spins.[6]

Although more detailed measurements are needed to obtain results on the depolarization process in LF, if the dynamical depolarization process is assumed, the fitting result of the LF-μSR shows an interesting feature. That is, the dynamical depolarization rate, λ_1, decreases with increasing LF with roughly a square-root field dependence. Such suppression of the depolarization behavior by LF has also been observed in one-dimensional materials.[20] In those cases, the depolarization of the muon spins is dominated by a magnetically excited state moving along one direction. Although no clear evidence has been obtained yet, there may be two possibilities for a one-dimensional excitation. One is a magnetic excitation of the in-plane short-range ordered state along the direction perpendicular to the CuO_2 plane through the weak interplane AF interaction. The other is based on the stripe structure. That is, the stripe structure is periodic along one direction in the CuO_2 plane. Therefore, one-dimensional excitation in the CuO_2 plane may be possible. More detailed LF-μSR measurements are necessary to be conclusive.

In summary, ZF- and LF-μSR measurements have been carried out to study magnetic properties of a Zn-substituted Bi-2212 system. The enhancement of the depolarization rate of the muon spins, which corresponds to the slowing down of the Cu-spin fluctuations, has been observed below 10 K around $p=\frac{1}{8}$ per Cu in the Zn-substituted samples. Both Zn substitution and $p=\frac{1}{8}$ per Cu are essential to induce the slowing down of the Cu-spin fluctuations. The relationship between the slowing down of the Cu-spin fluctuations and the SC, revealed by the present μSR measurements, is analogous to that of the $\frac{1}{8}$ effect which has been established in La systems. Thus, taking into account the results of the transport measurements,[11] it is concluded that the $\frac{1}{8}$ effect is likely to exist in the Bi-2212 system as well as in La systems. Our μSR results suggest that the $\frac{1}{8}$ effect is characteristic of the CuO_2 plane of high-T_c cuprates.

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