μSR study of the Cu-spin dynamics in the electron-doped high-
Tc cuprate of Pr$_{0.86}$LaCe$_{0.14}$Cu$_{1-y}$(Zn,Ni)$_{y}$O$_{4+\delta}$

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Abstract

Effects of the Zn- and Ni-substitution on the Cu-spin dynamics in the electron-doped Pr$_{0.86}$LaCe$_{0.14}$Cu$_{1-y}$(Zn,Ni)$_{y}$O$_{4+\delta}$ with $y = 0, 0.01, 0.02, 0.05$ and different values of the reduced oxygen content $\delta$ have been studied using zero-field muon-spin-relaxation ($\mu$SR) measurements at temperatures down to 2 K. For the as-grown sample ($\delta = 0, y = 0$) and the sample with a very small $\delta$ value ($\delta < 0.01, y = 0$), a muon-spin precession due to long-range antiferromagnetic order has been observed. On the other hand, no precession has been observed for moderately oxygen-reduced samples ($0.01 \leq \delta \leq 0.09$). It has been found that for all the samples of $0.01 \leq \delta \leq 0.09$ the asymmetry A(t) ($\mu$SR time spectrum) in the long-time region increases with decreasing temperature at low temperatures, suggesting possible slowing-down of the Cu-spin fluctuations. On the other hand, no significant difference between Zn- and Ni-substitution effects on the slowing down of the Cu-spin fluctuations has been observed.

Keywords: Muon spin relaxation; Electron-doped high-Tc cuprate; Cu-spin dynamics; Impurity effects

1. Introduction

The study of the impurity effect, namely, the effect of the partial substitution of Zn or Ni for Cu on the Cu-spin dynamics in the high-Tc cuprates has attracted much attention in relation to the mechanism of superconductivity. A lot of experimental results on the impurity effect have been reported for the hole-doped high-Tc cuprates [1-3], in contrast to those for the electron-doped ones. The difficulty in preparing superconducting samples of good quality in the electron-doped cuprates is one of the reasons. In the electron-doped cuprate with the so-called T' structure, superconducting samples are obtained after the heat treatment of as-grown samples in a reducing atmosphere [4]. The superconducting properties such as the superconducting transition temperature, $T_c$, are affected by the reduced oxygen content, $\delta$, as well as by the impurity concentration [5].

Here, we have investigated the Zn- and Ni-substitution effects on the Cu-spin dynamics in the electron-doped Pr$_{0.86}$LaCe$_{0.14}$Cu$_{1-y}$(Zn,Ni)$_{y}$O$_{4+\delta}$ from muon-spin relaxation ($\mu$SR) measurements, changing $y$ up to 0.05 and $\delta$ up to 0.09 [6].

2. Experimental

Polycrystalline samples of Pr$_{0.86}$LaCe$_{0.14}$Cu$_{1-y}$(Zn,Ni)$_{y}$O$_{4+\delta}$ with $y = 0, 0.01, 0.02$ and $0.05$ were prepared by the ordinary solid-state reaction method from La$_2$O$_3$, Pr$_6$O$_{11}$, CeO$_2$, CuO and ZnO or NiO

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powders of high purity [6,7]. As-grown samples were post-annealed in flowing Ar gas of high purity (6N) at 950 °C for 10 h in order to remove the excess oxygen. The δ value was estimated from the weight change before and after post-annealing. To check the quality of the obtained samples and to determine Tc, both electrical resistivity and magnetic susceptibility were measured. Zero-field (ZF) μSR measurements were performed at temperatures down to 2 K at the RIKEN-RAL Muon Facility at the Rutherford-Appleton Laboratory in UK.

3. Results and discussion

The obtained μSR time spectra of impurity-free Pr0.86LaCe0.14CuO4+δ are roughly grouped into 4 classes with different δ values: as-grown (δ = 0), very small δ (δ < 0.01), small δ (0.01 ≤ δ < 0.04), and large δ (0.04 ≤ δ ≤ 0.09). Samples with the small and large δ show superconductivity with Tc ranging from 15 K to 17 K (average Tc ~ 16 K) and from 18 K to 22 K (average Tc ~ 20 K), respectively, while the as-grown sample and the sample with very small δ are not superconducting above 4.2 K.

Figure 1 shows typical ZF-μSR time spectra of impurity-free Pr0.86LaCe0.14CuO4+α−δ. For the as-grown sample, a muon-spin precession is observed even at high temperature of 100 K due to long-range antiferromagnetic order. For the samples with very small δ, small δ and large δ, a Gaussian-like behavior is observed at high temperatures above ~100 K due to randomly oriented nuclear spins, and an exponential-like depolarization of muon spins is observed at low temperatures below ~50 K. For the sample with very small δ, a muon-spin precession is

![FIGURE 1. Typical ZF-μSR time spectra of Pr0.86LaCe0.14CuO4+α−δ with different δ values at various temperatures. Solid lines for the small and large δ are the best-fit results using a two-component function: A(t) = A_G exp[-σ^2 t^2] + A_S exp[-(λt)^β].]
observed at low temperatures below ~ 5 K. For the samples with small and large $\delta$, on the other hand, no muon-spin precession is observed, indicating the absence of any long-range magnetic order above 2 K. The temperature-dependent change of the spectra above 20 K is regarded as being due to the static random magnetism of small Pr$^{3+}$ moments [8]. Concerning the samples with small and large $\delta$ ($0.01 \leq \delta \leq 0.09$), it is found that the asymmetry $A(t)$, namely, the $\mu$SR time spectrum in the long-time region around 10 $\mu$sec, increases with decreasing temperature at low temperatures, suggesting possible slowing-down of the Cu-spin fluctuations.

The ZF-$\mu$SR time spectra are analyzed with the following two-component function: $A(t) = A_G \exp\left[-\sigma t^2\right] + A_s \exp\left[-(\lambda t)^\beta\right]$. The first term is a static Gaussian component in the region where the relaxation due to nuclear spins and small Pr$^{3+}$ moments [8] is dominant. The second term is a dynamical stretched-exponential component in the region where the Cu-spin fluctuations exhibit slowing down. The increase of $A(t)$ in the long-time region at low temperatures is reflected by the increase in $A_s$. The time spectra are well fitted with this function as shown in Fig. 1.

Figure 2 shows the temperature dependence of the fitted parameter $A_s$ for samples with various $y$ and $\delta$ values in the range $0.01 \leq \delta \leq 0.09$. For the impurity-free sample of $y = 0$ with large $\delta$, for example, it is

![Figure 2](image_url)

**FIGURE 2.** Temperature dependence of $A_s$ for Pr$_{0.86}$LaCe$_{0.14}$Cu$_{1-y}(Zn,Ni)_y$O$_{4+\alpha-\delta}$ with $y = 0, 0.01, 0.02, 0.05$ and small and large $\delta$ values at temperatures down to 2 K. Arrows indicate the temperature where $A_s$ exhibits the minimum.
found that $A_s$ decreases with decreasing temperature down to 30 K, which is due to the growing effect of the Pr$^{3+}$ moments. Below ~ 30 K, $A_s$ increases with decreasing temperature, indicating the slowing down of the Cu-spin fluctuations. For the impurity-free sample with small $\delta$, on the other hand, $A_s$ increases with decreasing temperature below 45 K. The difference of the temperature where $A_s$ shows the minimum may be due to the residual effect of a small amount of antiferromagnetically ordered Cu-spins in the impurity-free sample with small $\delta$. In any case, it attracts interest that the slowing down of the Cu-spin fluctuations is observed even in the impurity-free sample. This may be due to possible enhancement of the Cu-spin correlation assisted by the Pr$^{3+}$ moments.

The increase in $A_s$ at low temperatures is still observed for both Zn- and Ni-substituted samples up to $y = 0.05$. However, no significant difference in the temperature dependence of $A_s$ between Zn- and Ni-substituted samples is observed. It appears that the effect of the Pr$^{3+}$ moments is stronger than that of a small amount of Zn and Ni impurities. These behaviors are very different from those observed in the hole-doped high-T$_c$ cuprates [2,3].

### 4. Summary

We have investigated the Zn- and Ni- substitution effects on the Cu-spin dynamics from ZF-$\mu$SR measurements in the electron-doped cuprate Pr$_{0.88}$LaCu$_{0.14}$Cu$_{1-y}$(Zn,Ni)$_y$O$_{2-\delta}$ with $y = 0, 0.01, 0.02, 0.05$ and various $\delta$ values at temperatures down to 2 K. It has been found that the Cu-spin fluctuations exhibit slowing down at low temperatures in both impurity-free and impurity-substituted samples, regardless of the $y$ value for moderately oxygen-reduced samples ($0.01 \leq \delta \leq 0.09$). A possible origin of the slowing down observed even in the impurity-free sample is enhancement of the Cu-spin correlation assisted by the Pr$^{3+}$ moments. No significant difference in the temperature dependence of $A_s$ between Zn- and Ni-substituted samples may be due to the stronger effect of the Pr$^{3+}$ moments than that of a small amount of Zn and Ni impurities.

### Acknowledgments

This work was supported by Joint Programs of the Japan Society for the Promotion of Science, TORAY Science and Technology Grant and also Grant-in-Aid for Scientific Research from the Ministry of Education, Culture, Sports, Science and Technology, Japan.

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