μSR study of the Cu-spin correlation in heavily electron-doped high-$T_c$ T’-cuprates

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In the history of research on high-$T_c$ superconductivity, numerous efforts have been made to the establishment of the phase diagram of both hole-doped and electron-doped cuprates. However, Matsumoto et al. reported that superconductivity appears even in the parent compound of $x = 0$ and in a wide range of $x$ in Nd$_{2−x}$Ce$_x$CuO$_4$ with the so-called T’ structure through the appropriate reduction of excess oxygen from the as-grown films, resulting in a completely different phase diagram from that formerly suggested.1) The superconductivity in the parent compounds of T’-cuprates has also been confirmed for polycrystalline powdered samples of La$_{1−x}$Eu$_{0.2}$CuO$_4$.2) These suggest that the superconductivity in electron-doped T’-cuprates cannot be understood in terms of the doping of carriers into Mott insulators as in the case of hole-doped cuprates.

Recently, through improved reduction annealing, we have succeeded in obtaining high-quality superconducting (SC) single crystals of underdoped T’-Pr$_{1−x}$La$_x$Ce$_{4+δ}$CuO$_{4+δ}$ with $x = 0.05 − 0.10$ whose ground states were believed to be antiferromagnetic (AF).3) Transport measurements have revealed that the strongly localized state of carriers accompanied by the pseudogap due to AF fluctuations in the as-grown crystal changes to a Kondo state without the pseudogap in the SC crystal through reduction annealing. Moreover, our recent μSR measurements of SC crystals of $x = 0.10$ have revealed fast depolarization of muon spins and recovery of asymmetry in a long time region at low temperatures, suggesting the coexistence of superconductivity accompanied by a short-range magnetic order.4) These results as well as the superconductivity in the parent compounds can be understood in terms of our proposed band picture based on the strong electron correlation.3,4)

One of the next issues is investigating how superconductivity disappears through Ce doping. Our previous μSR measurements of the SC polycrystal of T’-Pr$_{1−x}$LaCe$_x$CuO$_{4+δ}$ (PLCCO) with $x = 0.14$ revealed slowing down of the Cu-spin fluctuations at low temperatures, but no short-range magnetic order was observed.5) Therefore, we performed μSR measurements using PLCCO single crystals in the heavily overdoped regime of $x = 0.17$ and 0.20 to obtain detailed information on the Cu-spin correlation. ZF and longitudinal-field μSR measurements were carried out using a MiniCryo and a fly-past-type $^3$He cryostat at temperatures down to 0.3 K at RIKEN-RAL.

Figure 1 shows ZF μSR spectra of heavily overdoped PLCCO with $x = 0.20$ where superconductivity disappears. At high temperatures around 200 K, the depolarization of muon spins is slow, indicating that the development of the Cu-spin correlation is weak. It is found that, with decreasing temperature, the depolarization of muon spins becomes fast due to the growing effect of Pr$^{3+}$ moments. At low temperatures, it is found that the recovery of the asymmetry in a long time region corresponding to the development of the Cu-spin correlation is negligibly small. This suggests that the Cu-spin correlation is barely developed in the non-SC heavily overdoped regime of PLCCO. Combined with the results in the underdoped4) and optimally doped5) regimes, it is concluded that the Cu-spin correlation is crucial for the appearance of superconductivity in T’-cuprates.

References