# $\mu$ SR study of annealing and Ce-doping effects on the magnetism in T'- $\Pr_{1.40-x} \text{La}_{0.60} \text{Ce}_x \text{CuO}_{4+y}$ [2013A0084]

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#### 1. Introduction

It is known that the superconductivity in T'structured  $R_2$ CuO<sub>4</sub> (R: rear earth) appears after adequate oxygen reduction procedure for moderately Ce(Th)-doped system. The phase diagram of T'- $R_{2-x}$ Ce<sub>x</sub>CuO<sub>4+ $\delta$ </sub> shows the existence of robust antiferromagnetic (AF) ordered region against the electron-doping, which is commonly adjacent to the superconducting phase [1]. However, quite recently, this phase diagram has been challenged by an evidence of superconductivity in the Cefree T'- $R_2$ CuO<sub>4</sub> [2, 3]. From a careful study on thin film, Naito's group reported the importance of heat treatment to realize the stoichiometric T'structure without apical oxygen [4]. If the perfect T'-structured  $R_2CuO_4$  without chemical doping indeed shows superconductivity, the result of experimentally studied electron-hole symmetry in the physical properties should be reconsidered. Furthermore, to understand the role of magnetism in the mechanism of superconductivity, the true magnetic ground state in the parent compound and the genuine phase diagram in T'- $R_{2-x}Ce_xCuO_4$  are required to be clarified. In order to shade more light on above issue, we studied the effect of oxygen reduction and Ce substitution on magnetism in T'-Pr<sub>1.40-x-y</sub>La<sub>y</sub>Ce<sub>x</sub>CuO<sub>4+ $\delta$ </sub> (PLCCO) by zerofield muon spin rotation/relazation ( $\mu$ SR) measurements.

### 2. Experiment

For the measurement, we prepared three powder samples of  $\Pr_{1.40-x-y} \text{La}_y \text{Ce}_x \text{CuO}_{4+\delta}$ , that is, as-grown and annealed  $x{=}0$ , and as-grown  $x{=}0.16$ . From a weight-loss of the sample after the annealing, the relative change in the amount of reduced oxygen was determined to be  ${\sim}0.07$  in the x=0 sample. For each sample, the phase purity was checked by X-ray diffraction measurement. Zerofield  ${\mu}\text{SR}$  measurement was performed on D2 instrument installed at J-PARC. We measured the  ${\mu}\text{SR}$  time spectrum at the temperatures of 5, 50, 75, 100, 125, 150, 175, 200, 250 and 300 K.

## 3. Results and discussion

In Fig. 1, the  $\mu SR$  time-dependence of normal-

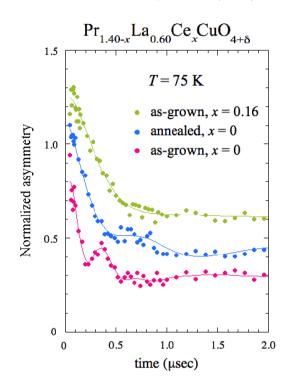


Fig. 1:  $\mu$ SR time spectrum of  $\Pr_{1.40-x-y}\text{La}_y\text{Ce}_x\text{CuO}_{4+\delta}$  at 75 K. Pink, blue and green circles represent the result for the as-grown x=0, annealed x=0 and as-grown x=0.16 samples, respectively. The spectrum for the later two samples is shifted along the vertical direction for the clear visualization.

ized asymmetry at 75K is shown for (a) as-grown x=0, (b) annealed x=0, and (c) as-grown x=0.16. The spectrum for the later two samples is lifted along the vertical direction. The spectrum in the all samples exhibits the rotation or fast relaxation components. Therefore, the magnetic order is realized in these samples at low temperature. To evaluate the magnetic ordering (Néel) temperature  $T_{\rm N}$ , we quantitatively analyzed the  $\mu {\rm SR}$  time spectrum measured at various temperatures. In the analysis, we assumed the existence of magnetic order with different internal magnetic fields at the stopped muon sites.

Figure 2 shows the relaxation rate  $\lambda$  of rotation components as a function of temperature. In the all samples, upon cooling,  $\lambda$  increases due to the devel-

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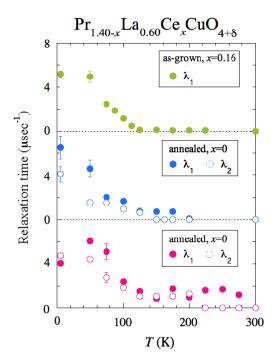


Fig. 2: Temperature dependence of relaxation rate  $\lambda$  for  $\Pr_{1.40-x-y} \operatorname{La}_y \operatorname{Ce}_x \operatorname{CuO}_{4+\delta}$ , as-grown x=0 (lower figure), annealed x=0 (middle figure) and as-grown x=0.16 (upper figure).

opment of magnetic order at low temperature. In the as-grown x=0 sample, the  $\lambda_1$  has finite value even at 250 K, indicating a relatively high  $T_{\rm N}$  in this sample. It should be noted that the two components are required to reproduce the time spectrum in the as-grown and annealed x = 0 samples, while the spectrum can be well fitted by a single rotation term for the as-grown x = 0.16 sample. Thus, the oxygen reduction and the Ce-doping give a different impact on the magnetism in the as-grown parent sample, although  $T_{\rm N}$  is reduced by both manners. We also note that there are possible two muon stopping sites, and the internal magnetic field at these sites calculated from the magnetic structure of Pr<sub>2</sub>CuO<sub>4</sub> is different. This is consistent with the observation of two rotation components in the as-grown parent sample.

From the results shown in Fig. 2, we evaluated  $T_{\rm N}$ , which is defined as the onset temperature for the appearance of relaxation components.  $T_{\rm N}$  for the as-grown and annealed x=0 and the as-grown x=0.16 is 285(15) K, 175(25) K and 125(10) K, respectively. These evaluated  $T_{\rm N}$ 's are plotted in Fig. 3 as a function of Ce concentration. The previously reported results for  ${\rm Pr}_{1-x}{\rm LaCe}_x{\rm CuO}_{4+\delta}$  is also plotted in the figure. This figure clearly indicates that the magnetic order is suppressed in a wide Ce concentration range by annealing. The ap-

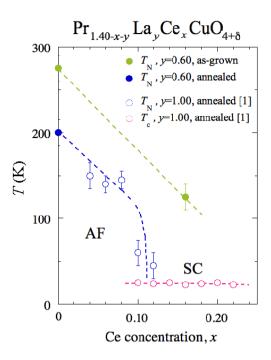


Fig. 3: Ce concentration dependence of magnetic ordering (Néel) temperature in the as-grown and the annealed  $Pr_{1.40-x-y}La_yCe_xCuO_{4+\delta}$ . Open circles are previously reported results for the annealed y=1 system [1].

pearance of superconductivity in the Ce-doped sample (x=0.16) after the full suppression of magnetic order by annealing suggests the competitive relation between magnetic order and the superconductivity. As mentioned above, we clarified the different local magnetism in the oxygen reduced and Ce-doped samples. This difference would be connected with the necessary condition for the emergence of superconductivity in the T'- $R_2$ CuO<sub>4+ $\delta$ </sub> Further systematic study on as-grown and annealed sample is important for the full understanding of mechanism of superconductivity in the T'-system.

A part of results shown in the report will be published in Key Engineering Materials ((Trans Tech Publication) in 2014.

#### References

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