Superconductivity of Ln$_{1.85-x}$La$_x$Ce$_{0.15}$CuO$_4$ (Ln=Pr,Sm)

AKIRA ITO, HIDEO IWASAKI, and NORIO KOBAYASHI

Institute for Materials Research, Tohoku University, 1-1, Katahira 2-chome, Aoba-ku, Sendai, 980 Japan

ABSTRACT

Superconducting properties of the N-type superconductors Ln$_{1.85-x}$La$_x$Ce$_{0.15}$CuO$_4$ (L=Pr and Sm) have been studied. Sintered samples were prepared by solid state reaction. It is possible to synthesize the samples with single phase of the Nd$_2$CuO$_4$ type structure in wide concentration ranges 0.5x<1.2 for Pr-based system and 0.5x<0.5 for Sm-based one. The lattice parameters a and c increase monotonically with increasing x for both systems. While it is expected that the substitution of Pr or Sm atoms by La does not change the carrier concentration of these systems because the oxygen content 4-y is kept almost constant. The superconducting transition temperature $T_c$ decreases with increasing La-concentration x. The suppression of $T_c$ is more rapid for Sm-based system than for Pr-based one. The x dependence of $T_c$ for both systems could be scaled by the reduced lattice parameter $(a/a_0)/a_0$.

KEYWORDS: electron-doped superconductor, substitution effect, Pr$_{1.85}$Ce$_{0.15}$CuO$_4$, Sm$_{1.85}$Ce$_{0.15}$CuO$_4$

INTRODUCTION

Since the discovery[1] of the electron-doped system Nd$_{1-x}$Ce$_x$CuO$_{4-y}$, many investigations have been performed to find out another N-type superconductor with different crystal structure. It is, however, only known that Ln$_{1-x}$M$_x$CuO$_{4-y}$ (Ln=Pr, Nd, Sm or Eu[2] ; M=Ce or Th[2]) with Nd$_2$CuO$_4$-type (T'-type) structure exhibits the N-type superconductivity. In these compounds, trivalent rare-earth ions can be substituted by tetravalent Ce or Th in contrast with La$_{1-x}$M$_x$CuO$_{4-y}$ (M=Ca, Sr or Ba) hole-doped system with the K$_2$NiF$_4$-type (T-type) structure. Furthermore, the compounds with T'-type structure must be reduced by heat treatment in a reducing condition, in order to obtain superconductivity. By this heat treatment, a small amount of oxygen is lost and superconductivity is realized.

Recently, Oh-ishi et al.[3] and Takayama-Muromachi et al.[4] reported that Nd sites in Nd$_{1.85}$Ce$_{0.15}$CuO$_4$-y system could be substituted by La in wide concentration range and the transition temperature $T_c$ was suppressed monotonically with increasing La concentration in spite of the fact that the La ions have the same trivalence as Nd ions.

It is very interesting to know what kind of mechanism is responsible for the $T_c$-suppression in the La-doped system. In this paper, we present experimental results on lattice parameters, oxygen deficiency and $T_c$ in Ln$_{1.85-x}$La$_x$Ce$_{0.15}$CuO$_{4-y}$ (Ln=Pr and Sm) system and discuss relations among them.

EXPERIMENTAL

Samples with compositions of Ln$_{1.85-x}$La$_x$Ce$_{0.15}$CuO$_4$-y (Ln=Pr and Sm) were prepared in the concentration ranges of 0.5x<1.3 for Ln=Pr and 0.5x<0.5 for Ln=Sm by a solid state reaction. Appropriate amounts of 4N-Pr$_2$O$_3$, Sm$_2$O$_3$, La$_2$O$_3$ CeO$_2$ and CuO powders were carefully mixed to improve homogeneity and pressed into pellets. The pellets were calcined at 950°C for 12h. Reacted materials were followed by grinding, pelletizing, reheating at 1150°C for 24h and then slowly cool down. These processes were done in air. A part of each sample was reduced in pure Ar atmosphere at 900°C for Pr system or 850°C for Sm system for 40h and then cooled down in the furnace.
Powder x-ray diffraction of the samples were taken using Cu-Kα radiation. The diffraction patterns showed that any impurity phase was not observed up to x=1.2 for Ln=Pr and in the concentration range studied for Ln=Sm. Oxygen content 4−y of the Pr1.85−xLaxCe0.15CuO4−y samples was measured by the iodometric titration method with a TDA auto-titrator. The electrical resistance was measured by an usual four probe method. The temperature was measured by a carbon glass and a platinum resistance thermometers. DC susceptibility measurements were made by a SQUID magnetometer (Quantum Design).

RESULTS AND DISCUSSION

Figure 1 shows the concentration dependence of lattice parameters a and c. The lattice parameters for both Pr- and Sm-based systems increase almost linearly with increasing La concentration x. This behavior suggests that the T'-type structure is held in whole concentration range studied. On the other hand, the lattice parameters seem to become slightly longer by the annealing in Ar atmosphere. As seen in Fig. 2, the oxygen content for the Pr-based