Low-temperature structural transitions and $T_c$-suppression in $\text{La}_{2-x-y}\text{Ba}_x\text{Nd}_y\text{CuO}_4$

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ABSTRACT

Features of low-temperature structural transitions in both $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$ and $\text{La}_{1.675}\text{Ba}_{0.125}\text{Nd}_{0.2}\text{CuO}_4$ have been investigated by transmission electron microscopy. The low-temperature transitions in both oxides were found to accompany 1/2 1/2 0-type superlattice spots in electron diffraction patterns. A feature of the superlattice spot is that the spot in La-Ba-Nd-Cu-O does not appear at a transition temperature of about 110K, but around 70K below it. In addition, bright- and dark-field images suggest that high- and low-temperature phases in the transition always coexist below the transition temperature.

KEYWORDS: $\text{La}_{2-x-y}\text{Ba}_x\text{Nd}_y\text{CuO}_4$ superconducting oxide, low-temperature structural transition, $T_c$-suppression, transmission electron microscopy

INTRODUCTION

In the La-cuprates such as $\text{La}_2\text{Ba}_2\text{CuO}_4$, a suppression of $T_c$ has been found around $x=0.125$ and so far discussed in relation to the low-temperature structural transition [1-2]. Yamada et al. measured a Seebeck coefficient in $\text{La}_2\text{Ba}_2\text{CuO}_4$ and found that the coefficient becomes a negative value below about 40K[3]. This seems to indicate that the transition results in a change in an electronic state at a Fermi level. Recently both the low-temperature transition and the $T_c$-suppression were also found to occur in $\text{La}_{1.675}\text{Ba}_{0.125}\text{Nd}_{0.2}\text{CuO}_4$ [4]. A remarkable feature of the Seebeck coefficient in this oxide is that the negative value of the Seebeck coefficient was obtained only below $T_c$ of about 40K, while the low-temperature transition takes place at $T_m$ of about 100K. That is, the Nd-substitution leads to an increase in $T_m$, but $T_c$ is independent of the Nd content. This clearly indicates that the transition does not necessarily produce a change in the electronic state. It can be then said that a correlation between the $T_c$-suppression and the low-temperature transition is still an open question. In addition to an origin of the $T_c$-suppression, a change in microstructures during the transition has not been sufficiently understood. Two groups, Chen et al. [5-6] and Zhu et al. [7], have already investigated by transmission electron microscopy. The former group reported that antiphase boundaries with respect to oxygen-atom displacements for a tilt of an octahedron were observed in a low-temperature-tetragonal (LTT) phase, which is the low-temperature phase in the transition. On the other hands, the latter group pointed out that the LTT structure exists only in twin boundaries of low-temperature-orthorhombic (LTO) domains. Note that the LTO phase is the high-temperature phase in the transition. In the present work, then in order to clarify the above-mentioned discrepancies, we have reexamined features of the low-temperature transitions in both $\text{La}_{1.675}\text{Ba}_{0.125}\text{CuO}_4$ and $\text{La}_{1.675}\text{Ba}_{0.125}\text{Nd}_{0.2}\text{CuO}_4$ by transmission electron microscopy.

EXPERIMENTAL PROCEDURE

Samples of both $\text{La}_{1.675}\text{Ba}_{0.125}\text{CuO}_4$ and $\text{La}_{1.675}\text{Ba}_{0.125}\text{Nd}_{0.2}\text{CuO}_4$ examined here were prepared by a conventional
solid state reaction. Starting powders of La$_2$O$_3$, BaCO$_3$, Nd$_2$O$_3$, and CuO were mixed mechanically and then pressed into pellets. In the present work, each pellet was sintered for 24 hr at three temperatures, 1273K, 1323K, and 1373K. After the sintering, the pellets were annealed at 673K for 48 hr in flowing oxygen atmosphere. In order to elucidate features of the transitions in both oxides, then in-situ observation was performed, using an H-800 type transmission electron microscope equipped with a liquid He reservoir in a temperature range between 11K and room temperature. Because of a stability of temperature, we mainly took bright- and dark-field images as well as electron diffraction patterns during a heating process. As for an intensity of a diffraction spot, we measured it by photodensitometry. Flakes obtained by crushing the pellets were used as a specimen for the in-situ observation.

RESULTS AND DISCUSSION

Both La$_{1.875}$Ba$_{0.125}$CuO$_4$ and La$_{1.675}$Ba$_{0.125}$Nd$_{0.200}$CuO$_4$ were already found to exhibit the low-temperature structural transition. Figures 1(a) and 1(b) show electron diffraction patterns taken from La$_{1.875}$Ba$_{0.125}$CuO$_4$ at 19K and from La$_{1.675}$Ba$_{0.125}$Nd$_{0.200}$CuO$_4$ at 46K, respectively. Electron incidences of these patterns are parallel to the [001] direction. Note that both oxides have a high-temperature-tetragonal (HTT) structure at room temperature and diffraction spots in the patterns are indexed in terms of the tetragonal system. As is easily understood from these figures, features of the pattern in La-Ba-Cu-O is entirely identical to those in La-Ba-Nd-Cu-O. That is, there exist two types of diffraction spots, in addition to fundamental spots due to the HTT structure. One is the 1 0 0-type spot, which is forbidden in the HTT structure. Then the appearance of this spot means that the specimens at these temperatures have the LTT structure. The other is the 1/2 1/2 0-type superlattice spot, which does not come from the atomic displacements for the tilt of the oxygen octahedron. In order to understand a change in intensities of both spots, we measured the intensities by using an electron diffraction pattern at each temperature by photodensitometry. Figure 2 represents measured intensities of both the 1 0 0 forbidden and 1/2 1/2 0 superlattice spots at various temperatures for La$_{1.875}$Ba$_{0.125}$CuO$_4$ and La$_{1.675}$Ba$_{0.125}$Nd$_{0.200}$CuO$_4$. Note that the temperature dependence of these intensities in La-Ba-Cu-O is shown in an inserted diagram. Closed and open circles in the figure represent the intensities of the 1 0 0 forbidden and 1/2 1/2 0 superlattice spots, respectively. Although both intensities start to raise at the transition temperature T$_m$ in La-Ba-Cu-O, in a case of La-Ba-Nd-Cu-O the 1/2 1/2 0 spot appears around 70K below T$_m$. This implies that the 1/2 1/2 0-type superlattice spot does not have a direct relation to the low-temperature transition. It is worth noticing that the Seebeck coefficient in La-Ba-Nd-Cu-O exhibits a rapid decrease around 70K, where the 1/2 1/2 0-type superlattice spots appear, and then becomes the negative value at T$_c$ of about 40K. This fact strongly suggests that the change in the electronic state, which results in the T$_c$-suppression, is not due to the low-temperature transition itself but is related to the appearance of the 1/2 1/2 0-type superlattice spots. As was pointed out in our previous paper [1], the superlattice spot is presumably a response of a lattice system to the appearance of charge density waves.

The features of the diffraction patterns in the lower temperatures were sufficiently understood, as was mentioned just above. A change in microstructures during the low-temperature transition was then examined by using a different specimen of La$_{1.875}$Ba$_{0.125}$CuO$_4$ from that for the electron-diffraction-pattern observation. Note that the specimen was