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Temperature Dependence of Electrical Transport Properties of $La_4BaCu_{5-x}Co_xO_{13+\delta}$ Conducting Oxide Thin Films

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Abstract

The temperature dependence of the resistivity of the Co-doped $La_4BaCu_5O_{13+\delta}$ ($La_4BaCu_{5-x}Co_xO_{13+\delta}$) conducting oxide films deposited on the single crystal substrate is reported in this paper. The metallic behavior of the non-doped $La_4BaCu_5O_{13+\delta}$ film in the electric transport property changed to semiconducting behavior with increase of the Co-substitution amount. In the case of the films with low Co-substitution (x=0.13, x=0.26), the resistivity was independent of temperature. The temperature coefficient of resistance (T. C. R.) between 150 K and 350 K was 0.20×10^{-3} /K in the x=0.26 film. This low T. C. R. value would be originated in the coexistance of the metallic and semiconducting transports.

1. Introduction

Among a lot of copper oxides with various electric properties such as superconductor or insulator, $La_4BaCu_5O_{13+\delta}$ is unusual because it shows metallic behavior at extremely low temperature [1]. It has been reported that the electric resistivity is lower than 1.0 mΩcm at room temperature, and the metallic behavior is maintained up to high temperature around 600 K. Therefore, $La_4BaCu_5O_{13+\delta}$ is expected to be a substitution material for Pt which is used as the electrodes and heaters in the micro-electronic devices.

In addition, $La_4BaCu_5O_{13+\delta}$ has an electric anisotropy [2], and the electric properties change with increase of the Cu-site substitution amount by the other transition metal elements (Co, Fe) [3]. $La_4BaCu_5O_{13+\delta}$ is such an interesting and useful material. However, the Cu-site substitution amount dependence of electric properties and the electric properties especially at higher temperature than room temperature are not investigated in detail.

We fabricated Co-doped $La_4BaCu_5O_{13+\delta}$ ($La_4BaCu_{5-x}Co_xO_{13+\delta}$) epitaxial thin films with various substitution amounts on the single crystal substrates by a pulsed laser deposition (PLD) method, and investigated the electric properties under the wide temperature range.

2. Experimental Details

$La_4BaCu_{5-x}Co_xO_{13+\delta}$ thin films were deposited on single-crystal $SrTiO_3$ (100) substrates using a conventional PLD method with a fourth-harmonic Nd:YAG laser (λ=266 nm) at a repetition rate 5 Hz. The laser energy density, the distance between the substrate and targets, and the O_2 partial pressure during the deposition were 2.9 J/cm^2, 55 mm, and 140 mTorr, respectively. In order to control the Co-substitution amount x in the films, we used an alternating targets (ALT) technique [4, 5] with a $La_4BaCu_5O_{13+\delta}$ target and a $La_4BaCu_4CoO_{13+\delta}$ target. We fabricated five $La_4BaCu_{5-x}Co_xO_{13+\delta}$ thins with different x (x=0.00, 0.13, 0.26, 0.66, 1.29). All films were annealed in O_2 atmosphere at 450ºC for 5 hours.

The crystalline orientation of the films was established by X-ray diffraction (XRD) analysis. The Co-substitution amounts in each film were identified by Inductively Coupled Plasma Atomic Emission Spectroscopy (ICP-AES). The resistivity was measured by standard four-probe method under the temperature range of 70 - 1000 K.

3. Results and Discussion

Figure 1 (a) shows the XRD pattern of $2\theta/\omega$ for the $La_4BaCu_5O_{13+\delta}$ (x=0.00) film. Only the diffraction peaks corresponding to (00n) indices of the $La_4BaCu_5O_{13+\delta}$ are observed except the peaks of the $SrTiO_3$ substrate. Fig. 1 (b) shows the ϕ-scan measurement result of $La_4BaCu_5O_{13+\delta}$ film, and the peaks of $La_4BaCu_5O_{13+\delta}$ exist every 90º. These XRD analysis results suggest that the a-axis and c-axis oriented epitaxial $La_4BaCu_5O_{13+\delta}$ film has grown on the substrate. In addition, the angles of the peaks corresponding to (211) of the $La_4BaCu_5O_{13+\delta}$ and (101) of the $SrTiO_3$ are identical. Accordingly, the $La_4BaCu_5O_{13+\delta}$ film has grown on the $SrTiO_3$ substrate with 26.6º rotation in the ab-plane. All films with various Co-substitution amounts showed same orientation and crystallinity.

Figure 2 shows the temperature dependence of resistivity (ρ-T) for the $La_4BaCu_{5-x}Co_xO_{13+\delta}$ films for the temperature range of 70 - 350 K. The non-doped $La_4BaCu_5O_{13+\delta}$ (x=0.00) film shows metallic behavior in this temperature range. The absolute values of the resistivity are lower than 1.0 mΩcm, and the values are in the same

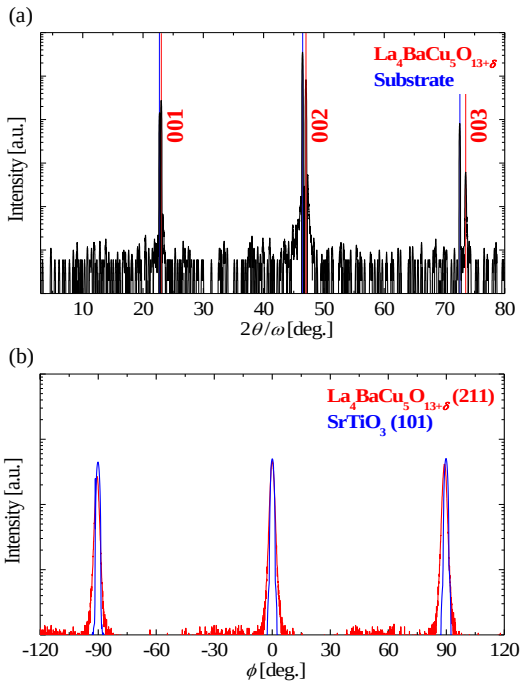

Fig. 1 XRD patterns of (a) $2\theta/\omega$ and (b) ϕ-scan for the $La_4BaCu_5O_{13+\delta}$ film on $SrTiO_3$ single crystal substrate.

range with the reported values in the bulk samples [1, 2]. The x=0.13 and x=0.26 films also show metallic behavior, and the resistivity are lower than that of the x=0.00 film. It should be noted that the temperature dependency of the resistivity for these films are extremely small. The temperature coefficient of resistance (T. C. R.) of each film are shown in Table I. We defined T. C. R. as eq. (1) using the resistivity at 150 K (ρ^{150}) and 350 K (ρ^{350}),

$$\mathrm{T.C.R.} = \frac{1}{\rho^{150}} \cdot \frac{\rho^{350} - \rho^{150}}{200} \qquad (1)$$

The resistivity of the x=0.66 and x=1.29 films increase with falling of temperature. The x=1.29 film shows the semiconducting behavior in particular. When we observe these ρ-T curves systematically along with the Co-substitution amount, the electric behavior of $La_4BaCu_{5-x}Co_xO_{13+\delta}$ transit from metallic to semiconducting with increase of Co-substitution amount. In the case of bulk, $La_4BaCu_5O_{13+\delta}$ and $La_4BaCu_4CoO_{13+\delta}$ showed metallic and semiconducting behaviors, respectively [3]. Then, it could be considered that the low T. C. R. of the x=0.13 and x=0.26 films originates in the coexistence and balance of the metallic and semiconducting transports. We will identify and compare the electric carrier of each film at various temperatures in order to reveal the transiting mechanism of the electric transport property.

4. Conclusions

We fabricated the Co-doped $La_4BaCu_5O_{13+\delta}$ films with various Co-substitution amount by conventional PLD method, and investigated the electric transport properties.

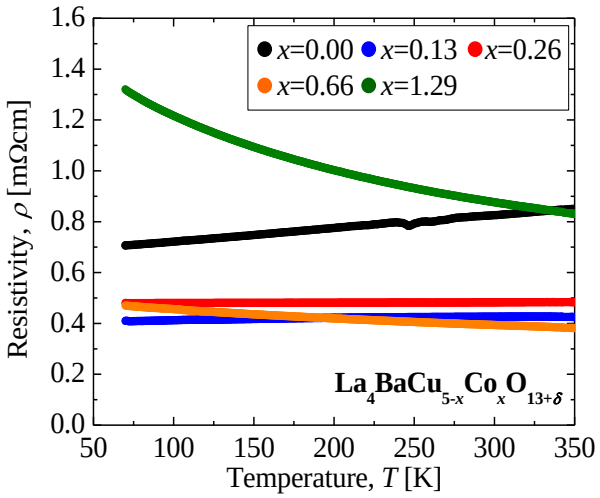

Fig. 2 Temperature dependence of resistivity for $La_4BaCu_{5-x}Co_xO_{13+\delta}$ films on $SrTiO_3$ single crystal substrates.

Table I Temperature coefficient of resistance (T. C. R.) of $La_4BaCu_{5-x}Co_xO_{13+\delta}$ films.

Film	T. C. R. [x10^{-3}/K]
x=0.00	0.65
x=0.13	0.10
x=0.26	0.02
x=0.66	-0.62

The ρ-T curve of the non-doped $La_4BaCu_5O_{13+\delta}$ film showed metallic behavior, and the resistivity was lower than 1.0 mΩcm. On the other hand, $La_4BaCu_{5-x}Co_xO_{13+\delta}$ (x=1.29) film showed semiconducting behavior. In the case of the low Co-substitution amount films such as x=0.13 and 0.26, the resistivity were almost independent of temperature. This phenomenon would be originated in the coexistance of the metallic and semiconducting transports.

We will report the other electric properties such as Seebeck coefficient and the ρ-T curves measured at higher temperature at the presentation.

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