Superconductor-Insulator Transition of Cuprate Thin Films Driven by Tuning of In-Plane Epitaxial Strain with a Buffer Layer

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Abstract. In order to finely tune the epitaxial strain we propose to insert a buffer layer of La$_{x}$Sr$_{1-x}$Al$_{0.5}$Ga$_{0.5}$O$_{4}$ between a superconducting cuprate thin-film and LaSrAlO$_{4}$ substrate. Thin films of La$_{1.875}$Ba$_{0.125}$CuO$_{4}$/La$_{x}$Sr$_{1-x}$Al$_{0.5}$Ga$_{0.5}$O$_{4}$ (LBCO/LSAGO) grown by pulsed laser deposition have been characterized by x-ray diffraction and transport measurements. The $c$-axis length of LBCO on the buffer layer shows a systematic change with $z$ indicating that the epitaxial strain is reasonably controlled. Superconducting transition temperature of LBCO/LSAGO films smoothly decreases with increasing $z$, and finally diminishes. Besides, LBCO films show systematic increase of normal-state resistivities with increasing $z$. However, comparing with single crystals of La$_{2-x}$Sr$_{x}$CuO$_{4}$, Hall coefficients of insulating film is much lower than expected. Therefore, the phenomena cannot be explained by nominal change of carrier and indicates that the epitaxial strain causes certain modification on the shape of Fermi surface.

1. Introduction
La$_{2-x}$Sr$_{x}$CuO$_{4}$ (LSCO) and La$_{2-x}$Ba$_{x}$CuO$_{4}$ (LBCO), the mostly studied high temperature superconductor (HTSC) prototypes, are well known to demonstrate an intimate relationship between structure and superconductivity. $T_c$ is easily modified by adding isotropic/anisotropic pressure to the crystal [1, 2]. And even more drastic change in $T_c$ occurs on epitaxial grown thin films of LSCO and LBCO [5]. They indicate greatly increased $c$-axis length, higher $T_c$ than the bulk, and vanishing of the “1/8-anomaly” [3, 4] under compressive strain along the CuO$_2$ plane [6-9]. The compounds are also known to show a decrease of $T_c$ upon increasing the in-plane cell constant of single-crystal substrate. However, detailed mechanism of the suppression of superconductivity has not been studied because we can access the limited number of single-crystal substrates. One can control epitaxial strain by employing a buffer layer between a substrate and a LSCO (or LBCO) and changing its thickness [10-12]. But the method supposed not to be suitable for quantitative discussion as the epitaxial strain is not proportional to the “thickness” of a buffer layer. Therefore, substrates with variable unit-cell parameters are indispensable for the quantitative discussion on the effect of epitaxial strain. In this paper, we tune the in-plane strain of La$_{1.875}$Ba$_{0.125}$CuO$_{4}$ (LBCO $x=0.125$) thin films by inserting a buffer layer of a solid-solution compound and characterize them by transport measurements.
2. Experimental details

We chose LaSrAl$_{1-z}$Ga$_z$O$_4$ (LSAGO) as a buffer layer for the following reasons: (1) Both end compounds—LaSrAlO$_4$ (LSAO) [13] and LaSrGaO$_4$ (LSGO) [14]—have the same K$_2$NiF$_4$-type crystal structures, and (2) the $a$-axis length of the bulk LBCO crystal is located between that of LSAO ($a=3.7564$ Å) and LSGO ($a=3.8437$ Å). Further information is available on our previous report [15].

2.1. Growth of LSAGO buffer layer

LSAGO polycrystals have been prepared by conventional solid state reaction. Stoichiometric mixtures of La$_2$O$_3$, SrCO$_3$, Al$_2$O$_3$, and Ga$_2$O$_3$ were grounded thoroughly in an agate mortar, pressed into pellets, placed on a platinum sheet, and heated first at 1300$^\circ$C for 20 h and then at 1500$^\circ$C for 50 h in air. According to the powder x-ray diffraction analysis, all products contained the LSAGO phase with no impurities. All peaks are indexed to a K$_2$NiF$_4$ structure with space group I$4/mmm$ (No. 139). The cell constants for the LSAGO compounds obtained from powder XRD data was calculated by using the program RIETAN-2000 [16]. Both of the constants ($a$ and $c$) increase with increasing $z$, linearly interpolating the values of LSAO and LSGO [see Fig. 1]. It suggests that the system forms solid solution in the entire range of $0 \leq z \leq 1$. All LSAGO buffer layers were grown by pulsed laser deposition (PLD) on the substrate of LSAO (001) at $T=850^\circ$C. The target was ablated by a KrF excimer laser (wavelength: 248 nm) under 10 mPa of pure ozone. A typical thickness of the LSAGO buffer layer is 40 nm.

2.2. Growth and characterization of LBCO thin film

La$_{1.875}$Ba$_{0.125}$CuO$_4$ films were grown under almost the same conditions as for the LSAGO buffer layers, except for the deposition temperature ($820^\circ$C) and the thickness of the films (100 nm). Figure 2 shows the $c$-axis length of the La$_{1.875}$Ba$_{0.125}$CuO$_4$ film as a function of $z$ of the LSAGO buffer layer. The data reveal that $c$-axis length decreases with increasing $z$, and the result agrees with our exception. Resistivity and Hall coefficient measurements were carried out in a Quantum Design PPMS system.

3. Results and discussion

The transport properties of the La$_{1.875}$Ba$_{0.125}$CuO$_4$ film on the LSAGO/LSAO substrate demonstrates an intimate relationship between physical properties and epitaxial strain. The resistivity data, displayed in Fig. 3, clearly show the superconductor-insulator transition driven by the in-plane epitaxial strain. The room-temperature in-plane resistivity of the La$_{1.875}$Ba$_{0.125}$CuO$_4$ film on LSAO ($z=0$) is $3.8 \times 10^{-4}$ $\Omega$cm. A systematic increase of the resistivity with increasing $z$, i.e. the $a$-axis length of the buffer layer, is observed in the entire
Figure 2. The \( c \)-axis length of the 100 nm \( \text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4 \) film on the 40 nm LSAGO/LSAO substrate (there is no buffer layer for \( z = 0 \)). The dashed line is a linear fit in the region of \( 0 \leq z \leq 0.9 \).

Figure 3. Resistivity vs. temperature curves for \( \text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4 \) films on the LSAGO/LSAO substrates. For \( z = 0 \), the LBCO film is deposited on LSAO without buffer layer.

Figure 4. The superconducting onset (open square) and zero-resistivity temperature (filled square) for \( \text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4 \) films on the LSAGO/LSAO substrates (there is no buffer layer for \( z = 0 \)).

temperature range, and the ratios of resistivity measured at \( T_c \) to that at room temperature \([R(T_c)/R(300\text{K})]\) are 0.216, 0.262, 0.277, and 0.280 for \( z = 0, 0.2, 0.4, \) and 0.6, respectively. The superconducting transition temperatures are also sensitive to the epitaxial strain; \( T_c \) decreases from 38 to 26 K as \( z \) increases, and the superconductivity vanishes at \( z = 0.8 \) [see Fig. 4]. This tendency—the in-plane compressive strain enhances superconductivity while tensile strain suppresses it—agrees with previous studies [6, 7].

The Hall coefficient data of \( \text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4 \) film on the buffer layer of \( z = 0.8 \), which shows insulating behavior at low temperature, indicates the unique feature of this superconductor-insulator transition. Figure 5 shows the result of preliminary measurement of the Hall coefficient for superconducting (\( z = 0 \)) and insulating (\( z = 0.8 \)) \( \text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4 \) films. The Hall coefficient shows a clear increase with increasing \( z \). Assuming that the Hall coefficient correctly reflects the number of carrier, the insulating behavior of the \( \text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4 \) film with the buffer layer of \( z = 0.8 \) seems to be realized by nominal change of carrier. However, comparing with
the LSCO single crystals, the Hall coefficient value of the insulating $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$ film ($2.0 - 3.7 \times 10^{-3} \text{cm}^3/\text{C}$) is much lower than that of superconductor-insulator boundary in LSCO single crystals ($\sim 10 \times 10^{-3} \text{cm}^3/\text{C}$) [17]. In other word, insulating phase of this film shows up in spite of the presence of enough carriers. Therefore, the phenomena strongly suggest that the in-plane tensile surppresses the superconductivity mainly through the change of electronic structure, specifically the shape of Fermi surface. Next, we have to find out which part of the Fermi surface is directly tied to the occurrence of superconductivity.

4. Summary
We have succeeded in controlling physical properties of the $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$ thin films by tuning the in-plane strain with buffer layer of $\text{LaSrAl}_{1-z}\text{Ga}_z\text{O}_4$. The $\text{La}_{1.875}\text{Ba}_{0.125}\text{CuO}_4$ films demonstrate a systematic change in $T_c$ and normal-state resistivity. The Hall coefficient of films indicates that the principal origin of insulating phase is not the insufficiency of carrier but the change of the shape of Fermi surface.

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