Vol. **111** (2007) *ACTA PHYSICA POLONICA A* No. 1

Proceedings of the Symposium K: "Complex Oxide Materials for New Technologies"
of E-MRS Fall Meeting 2006, Warsaw, September 4–8, 2006

Strain Relaxation in Thin Films of $La_{1.85}Sr_{0.15}CuO_4$ Grown by Pulsed Laser Deposition

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X-ray diffraction, resistivity, and susceptibility measurements are used to examine the effects of film thickness d (from 17 to 250 nm) on the structural and superconducting properties of $La_{1.85}Sr_{0.15}CuO_4$ films grown by pulsed laser deposition on $SrLaAlO_4$ substrates. For each d the film sgrow with a variable strain, ranging from a large compressive strain in the thinnest films to a negligible or tensile strain in thick films. Our results indicate that the tensile strain is not caused by the off-stoichiometric layer at the substrate–film interface. Instead, it may be caused by the extreme oxygen deficiency in some of the films.

PACS numbers: 68.55.–a, 74.62.–c, 74.72.Dn, 74.78.Bz, 74.25.Fy

1. Introduction

The reproducible technology of excellent quality superconducting films is prerequisite for their potential applications in electronic devices. If the films are grown heteroepitaxially on the substrates with the mismatched lattice parameters, their quality may be compromised as a result of strain relaxation and accumulation of various growth-induced defects. The growth of thin $La_{1.85}Sr_{0.15}CuO_4$ (LSCO) films on a substrate of $SrLaAlO_4$ (SLAO) is known to produce films with compressive in-plane strain which enhances superconducting transition temperature, T_c [1–4]. However, the thick LSCO films grown on SLAO suffer film-to-film variations of T_c and other transport parameters [5–7]. We have shown recently that these films show a variable built-in strain, ranging from compressive to tensile. While the compressive strain is induced by the mismatch to the substrate, which has an in-plane lattice parameter smaller by about 0.6%, the origin of tensile strain is unclear. We have suggested that it may be caused by the thin layer of the oxygen or strontium-deficient material which grows in the initial stages of the deposition [8]. To check this hypothesis in the present study we investigate the properties of several groups of LSCO films of thicknesses d, ranging from 17 to 250 nm.

2. Experimental details and results

Epitaxial LSCO films were deposited from stoichiometric ceramic target by pulsed laser deposition (PLD) using excimer KrF laser ($\lambda = 248$ nm) with pulse duration of 28 ns, repetition rate of 1 Hz, and energy density of 1.3 J/cm^2 at the target surface. During deposition the SLAO substrates were held at temperature of 760°C in the oxygen atmosphere of 600 mTorr. After deposition, the O_2 pressure in the chamber was increased to 500 Torr, and the films were slowly cooled down to room temperature with a rate of 3 K per minute. The films were studied using X-ray diffraction with 3 different diffractometers, Siemens D-5000, Bragg-Brentano Philips XPert Pro Alpha1 MPD, and high-resolution Philips XPert MRD. The thickness of the samples was estimated from X-ray reflectivity data. The T_c was obtained from the resistivity and susceptibility measurements.

Figure 1a shows the correlation between a-axis (in-plane) and c-axis (out-of-plane) lattice parameters for several films with different d. We see that the compression of a with respect to the bulk is accompanied by the expansion of c. This type of behavior is expected in the case of strain induced by the lattice mismatch.

Figure 1c shows the dependence of c on the film thickness. For each d we observe films with a large range of c-values. The films with the largest compressive in-plane strain for each thickness d are those with the largest c. The data for these films lay along dashed line drawn in Fig. 1c. The dashed line represents the limit of the compressive strain which may be built into the film for each thickness d. We see that this limit is the largest for the thinnest films. The compressive strain is partially relieved as d grows, until c becomes comparable to the bulk value when the film thickness reaches about 250 nm. The Θ–2Θ scans for several of the films with different d and with the compressive strain close to the limiting value are shown in Fig. 1b. The peaks shift towards a high angle with the increase in d indicating a gradual decrease in c.

In addition to the films with the largest compressive strain, in each group of films with different d there are also films with the smaller c-values. In particular, this leads to the decrease in c-axis parameter below the bulk value when $d > 200$ nm, so that the tensile in-plane strain is built into the film. Indeed, the correlation shown in Fig. 1a indicates that the decrease in c is accompanied by the expansion of a. However, since this effect is similar for the films of different thicknesses, it does not seem to be caused by a substrate-related strain. Neither it cannot be attributed to a thin layer of strontium or oxygen-deficient material at the substrate–film interface since this would lead to the strongest tensile strain in the thinnest films, which is in contrary to our observations.

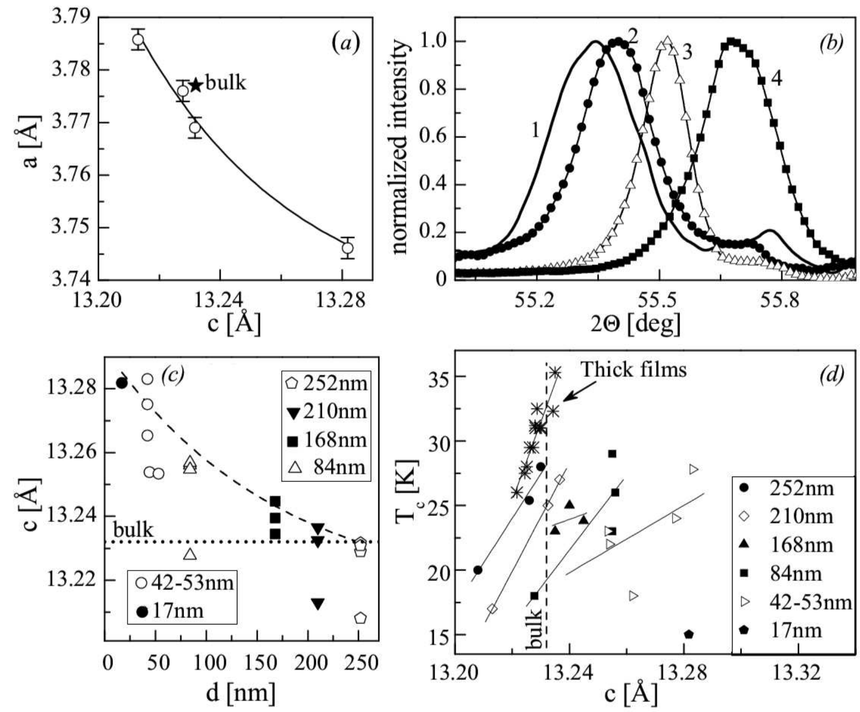

Fig. 1. (a) Correlation between a and c lattice parameters. (b) $\Theta{-}2\Theta$ scans in the vicinity of the (008) peak for film with $d = 42$ nm (1), $d = 84$ nm (2), $d = 168$ nm (3), $d = 252$ nm (4). (c) c as a function of d. (d) The dependence of T_c on c for films with different d. Data for thick films are from Ref. [8].

Figure 1d shows the dependence of T_c on c for a series of films with various thicknesses d. There is a considerable scatter of the data, particularly for the thinnest films. Nevertheless, we can say that for each d the T_c is suppressed as the c-axis parameter decreases. This suppression is similar to the one observed by us previously for the thick films (data shown by stars), and attributed to the presence of the tensile strain. This similarity suggests that the T_c suppression may be caused by similar effects in the thin and in the thick samples. Since the tensile strain is absent in the thinnest films, the presence of the tensile strain cannot be the primary cause of the T_c suppression. Rather, we suspect that the decrease in the c parameter may be related to oxygen vacancies which may be built-in independent of the film thickness. The oxygen deficiency in the LSCO films leads to the decrease in c and increase in a parameter, producing effects which are in direct competition with the substrate-induced compressive strain [4]. Therefore, the tensile strain and related to it columnar growth reported in Ref. [8] for thick films may be caused by the extreme oxygen deficiency. Further experiments on the O_3-assisted film growth are planned to verify this conclusion.

3. Conclusions

The study of LSCO films grown on SLAO substrates, with thickness ranging from 17 to 250 nm, indicate that there is a limit of the compressive strain which may be built into the film. This limit is the largest in the thinnest films and decreases with the increase in d, which excludes the possibility that the oxygen- or strontium-deficient layer is formed in the first stage of the deposition. For each d some of the films grow with smaller c, leading to a tensile strain in case of thick films. The tensile strain does not seem to be the cause of the T_c suppression. Rather, it is the buildup of some defects, most likely oxygen vacancies, which contributes both to the tensile strain, and to the suppression of T_c.

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