Growth of Single Unit-Cell Superconducting 

\( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) Films

A. Rüfenacht\textsuperscript{a,b,*}, P. Chappatte\textsuperscript{a}, S. Gariglio\textsuperscript{c,b}, C. Leemann\textsuperscript{a},

J. Fompeyrine\textsuperscript{b}, J.-P. Locquet\textsuperscript{b}, and P. Martinoli\textsuperscript{a}

\textsuperscript{a}Institut de Physique, Université de Neuchâtel, CH-2000 Neuchâtel

\textsuperscript{b}IBM Research Division, Zurich Research Laboratory, CH-8803 Rüschlikon

\textsuperscript{c}DPMC, Université de Genève, CH-1211 Genève 4

Abstract

We have developed an approach to grow high quality ultrathin films of \( \text{La}_{2-x}\text{Sr}_x\text{CuO}_4 \) with molecular beam epitaxy, by adding a homoepitaxial buffer layer in order to minimize the degradation of the film structure at the interface. The advantage of this method is to enable a further reduction of the minimal thickness of a superconducting \( \text{La}_{1.9}\text{Sr}_{0.1}\text{CuO}_4 \) film. The main result of our work is that a single unit cell (only two copper oxide planes) grown on a \( \text{SrLaAlO}_4 \) substrate exhibits a superconducting transition at 12.5 K (zero resistance) and an in-plane magnetic penetration depth \( \lambda_{ab}(0) = 535 \text{ nm} \).

Key words: LSCO, Superconductivity, Homoepitaxial growth, Single unit cell

PACS:

* Corresponding author : alain.rufenacht@unine.ch - FAX : +41 32 718 29 01
1 Introduction

Growing very thin superconducting films with well-characterized and interesting physical properties is a difficult challenge for the material scientist. In an attempt to reduce the smallest superconducting thickness, we have developed a homoepitaxial technique for the growth of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO), using a normal metallic buffer layer with a strontium (Sr) concentration $x=0.4$. On this metallic buffer, we grow the physically relevant structure, i.e. a few unit cells of $\text{La}_{1.9}\text{Sr}_{0.1}\text{CuO}_4$. On top of this film an additional two unit cell thick $\text{La}_{1.6}\text{Sr}_{0.4}\text{CuO}_4$ metallic cap ($x=0.4$) is deposited for protection (Fig. 1 a).

So far the thinnest superconducting optimally doped LSCO film directly grown on a SrLaAlO$_4$ substrate was 2 unit cell$^1$ (UC) thick with a superconducting critical temperature of 10 K and a superconductivity onset around 35 K [1]. We have reproduced this result, with the same thickness on the same substrate, resulting in a $T_c(R=0)$ of 10 K and a superconductivity onset near 35 K (SLAO 0/2/0).

The use of our buffer technique eliminates the hazardous effects of the film-substrate interface (due to mismatch and chemical interactions) on the superconducting properties of these extremely thin films. Our method allows the experimentalist to grow quasi two-dimensional superconducting films, an interesting tool for physical investigations and device applications.

$^1$ Notice that the crystallographic unit cell of LSCO contains two copper oxide planes.
2 Experimental and fabrication details

The samples studied in this report were thin films grown in a molecular beam epitaxy (MBE) system. The sample fabrication details were reported elsewhere [2]. The method used to grow epitaxial thin films consists of a block-by-block deposition [3]. The oxygenation of the films during the deposition and the cool down is provided by a RF plasma source of atomic oxygen. A reflection high-energy electron diffraction (RHEED) system allows us to monitor the growth process, in particular the quality of the in-plane growth.

The choice of the substrate and the resulting strain effects play an important role in the structural quality [4] and physical properties [5] of the film. In this work, ultra-thin films were deposited on (100) SrTiO$_3$ (STO) and (001) SrLaAlO$_4$ (SLAO) substrates. The thickness of the buffer layers was adapted to the two substrates. Due to a larger mismatch, the buffer layer on a STO substrate is thicker (10 UC) than on a SLAO substrate (4 UC), allowing a better film-substrate interface decoupling$^2$ (Fig. 1 a).

In order to understand whether the Sr interlayer diffusion across the interface increases the effective number of UC necessary for superconductivity, we consider the case where the interlayer diffusion is limited to the single unit cell film with nominal concentration $x$=0.1 and the adjacent bottom and top layers, half-UC thick, with nominal concentration $x$=0.4. The rest of the layers are assumed to act as reservoirs with an almost constant concentration $x$=0.4. The interaction of the upper superconducting half-UC with the first cap half-UC

$^2$ SLAO (compressive) $a_{\text{axis}} = 3.754$ Å, STO (tensile) $a_{\text{axis}} = 3.905$ Å, LSCO with $x = 0.1$, $a_{\text{axis}} = 3.784$ Å.
will give for each half-UC an average concentration \( x = (0.1 + 0.4)/2 = 0.25 \) (Fig. 1 b). The scenario for the lower superconducting half-UC is identical. The final result would be a film with 4 half UC with \( x=0.25 \). LSCO compounds with this Sr concentration are metallic and at the upper overdoped limit of the superconducting phase boundary. With this assumption, a lower amount of Sr diffusion will not increase the number of superconducting layers. However, we can not exclude a rise of the effective doping level in the superconducting layer(s).

Instead of using a metallic buffer, it is also possible to choose an insulating material, by reducing the Sr content of the LSCO buffer below \( x=0.05 \). In this way, the Sr in-diffusion is excluded. However, such LCO and low doped LSCO can be driven superconducting by adding interstitial oxygen [6]. This process occurs preferentially at low Sr concentration. Note that the growing LSCO or LCO films doped with interstitial oxygen is more difficult to master than by varying the Sr concentration.

3 Structural properties

The x-ray diffraction measurements confirm the \( c \)-axis growth of the films deposited on SLAO and STO. From conventional \( \theta - 2\theta \) scans, a linear regression of the (00\( \ell \)) peaks positions gives an average lattice parameter\(^3\) which for the film 4/1/2 on SLAO substrate is \( c = (13.285 \pm 0.010) \text{ Å} \) (Fig. 2). Finite-size effect oscillations were observed at low angle and around the (00\( \ell \)) peaks (\( \ell = 2, 4, 6, 8 \)), allowing a good estimate of the total film thickness. For the 4/1/2

\(^3\) to be compared with the bulk value (\( x=0.4 \)) : \( c = (13.26 \pm 0.01) \text{ Å} \) [7]
film grown on SLAO, finite-size effect oscillation observed around the (004) peak give a thickness of 89.4 Å, in agreement with the presence of 7 UC of 13.285 Å. Pole figure of the (103) plane of the film, measured on the same sample, confirms the epitaxial growth.

4 Superconducting properties

4.1 Resistivity measurements

Resistivity measurements were performed in a cryoprobe (range 300K → 4.2 K), using a standard DC four-point method. Contacts were made with four indium wires pressed onto the top of the film structures. The measurements (Fig. 3) performed on different samples provide evidence of superconductivity in samples as thin as 1 UC on SLAO and 2 UC on STO. No superconductivity was observed in the film with a similar nominal Sr concentration but with only one copper oxide plane. These results demonstrate the validity of the homoepitaxial buffer layer technique and tell us that the Sr interdiffusion is small enough not to depress the superconducting behavior. The thickness has a major influence on the superconducting properties. The superconducting critical temperatures $T_{c0}$ (as defined by the bottom of the resistivity transition) for the layers grown on SLAO are respectively 12.5 K and 25 K for the 1 UC and 2 UC thin films. On STO substrates, the $T_{c0}$ are 5.7 K for a 2 UC and 11.3 K for a 4 UC thin film. The fact that $T_{c0}$ is found to be proportional to the film thickness is consistent with two-dimensional behavior [8]. At identical thickness, the critical temperature of layers grown on STO is much lower than the one obtained for a film grown on the SLAO. The overall resistance of the
whole three-layers structure results from the contributions of the individual layers, which have not been investigated in detail. Nevertheless, the temperature dependence in the normal state is consistent with the value obtained for $x=0.4$ doped LSCO films [9].

4.2 Magnetic penetration depth measurements

The measurements of the magnetic penetration depth were performed using a two-coil technique [10]. Briefly, this method measures, contact-less and with an astatically wound coil, the response (due to screening currents) of a sample excited by an external AC electromagnetic field. The complex sheet impedance of the film is extracted from the signal measured with a lock-in amplifier. The inductive part of the sheet impedance of the superconducting film (the so-called kinetic inductance $L_k$) is related to the bulk in-plane magnetic penetration depth $\lambda_{ab}$ by the following relation: $L_k(T) = \frac{\mu_0 \lambda_{ab}^2(T)}{d}$, where $d$ is the thickness of the film.

All inductive measurements were performed in a $^3$He cryostat. Notice that the inductive method probes the onset of global superconducting phase coherence. For this reason the $T_c$ deduced from the inductive measurements is lower than that extracted from the resistive transitions [11].

As shown in figure 4, the zero-temperature value $\lambda_{ab}^{-2}(0)$ for the 1 UC film grown on SLAO was deduced by fitting the kinetic inductance data to the parabolic expression $\lambda_{ab}^{-2}(T) = \lambda_{ab}^{-2}(0) \cdot (1 - \left(\frac{T}{T_c}\right)^2)^2$ [12]. A similar procedure was used to fit the low-temperature data of the 4 UC film grown on STO. Although both films exhibit almost identical critical temperatures ($\sim$8.5 K), the
1 UC film has a smaller value of the magnetic penetration depth ($\lambda_{ab}(0)=535$ nm) than the 4 UC film ($\lambda_{ab}(0)=760$ nm). This shows the superior structural quality of the film grown on SLAO, in agreement with previous observations [5]. We also notice that the values of $\lambda_{ab}(0)$ obtained using the buffer technique are much smaller than that ($\lambda_{ab}(0)=2.3 \mu$m) for a 2 UC film grown directly on SLAO (see Fig. 4), thereby demonstrating the significant advantage of our method. An other remarkable feature emerges if one compares the penetration depth of the 1 UC film grown on SLAO ($\lambda_{ab}(0)=535$ nm) with that of optimally doped LSCO (bulk) single crystals ($T_c=35$ K and $\lambda_{ab}(0)=300$ nm [13]). Assuming two-dimensional behavior, for which one expects $T_c \propto \lambda_{ab}^{-2}$, one deduces that the structural quality of the ultrathin films grown on SLAO with our homoepitaxial buffer technique is comparable to that of LSCO single crystals.

5 Conclusions

The homoepitaxial buffer technique is an efficient tool to further reduce the superconducting minimal thickness. An X-ray characterization proved the high structural quality (c-axis orientation and epitaxy) of the films. Values obtained for the penetration depth and $T_c$ confirm the correlated role of substrate choice and homoepitaxial buffer technique when growing very high quality ultra-thin La$_{2-x}$Sr$_x$CuO$_4$ films. Finally, the work we present here forms a new starting point for investigations of physical properties in ultra-thin superconducting layers and research of device applications.
6 Acknowledgments

We thank M. Dodgson, H. Siegwart, C. Rossel, A. Guiller, J. W. Seo, E. Koller and J.-M. Triscone for helpful discussions. This work was supported by the Swiss National Science Foundation.

References

[1] H. Sato, H. Yamamoto, M. Naito, Physica C 274, 227 (1996).

[2] Y. Jaccard, A. Cretton, E. J. Willams, J.-P. Locquet, E. Mächler, C. Gerber, T. Schneider, Ø. Fischer and P. Martinoli, in Oxide Superconductor Physics and Nano-Engineering, Proc. SPIE 2158, 200 (1994).

[3] J.-P. Locquet and E. Mächler, MRS Bulletin 19, 39 (1994).

[4] J. W. Seo, J. Perret, J. Fompeyrine, G. Van Tendeloo, and J.-P. Locquet, in Superconducting and related oxides : Physics and Nanoengineering III, SPIE Proc. 3481, 300 (1998).

[5] J.-P. Locquet, J. Perret, J. Fompeyrine, E. Mächler, J.W. Seo, and G. Van Tendeloo, nature 394, 453 (1998).

[6] F. Arrouy, J.-P. Locquet, E. J. Williams, E. Mächler, R. Berger, C. Gerber, C. Monroux, J. C. Grenier, and A. Wattiaux, Phys. Rev. B 54, 7512 (1996).

[7] P. G. Radaelli, D. G. Hinks, A. W. Mitchell, B. A. Hunter, J. L. Wagner, B. Dabrowski, K. G. Vandervoort, H. K. Viswanathan, and J. D. Jorgensen, Phys. Rev. B 49, 4163 (1994).

[8] T. Schneider and J. M. Singer, Phase Transition Approach To high Temp. Supercond., Imperial College Press, London, 2000.
[9] H. Sato, A. Tsukada, M. Naito, and A. Matsuda, Phys. Rev. B 61, 12447 (2000).

[10] B. Jeanneret, J. L. Gavilano, G. A. Racine, Ch. Leemann, and P. Martinoli, Appl. Phys. Lett. 55, 2336 (1989).

[11] Ch. Leemann, Ph. Flückiger, V. Marsico, J. L. Gavilano, P. K. Srivastava, Ph. Lerch, and P. Martinoli, Phys. Rev. Lett. 64, 3082 (1990).

[12] M. Tinkham, Introduction to Superconductivity, McGrow Hill International edition, Singapore, 1996.

[13] T. Shibauchi, H. Kitano, K. Uchinokura, A. Maeda, T. Kimura, and K. Kishio, Phys. Rev. Lett. 72, 2263 (1994).
Fig. 1. Homoeptaxial buffer technique applied to La$_{2-x}$Sr$_x$CuO$_4$ compounds. (a): general scheme for the two different kinds of substrate. Notation: B/F/C corresponds, respectively, to the number of buffer layer unit cells (B), superconducting thin film (F), and metallic cap (C). (b): model for the influence of Sr interdiffusion in the SLAO 4/1/2 compound.

Fig. 2. X-ray diffraction pattern of 4/1/2 LSCO thin film on SLAO: $\theta$-2$\theta$ scan showing the LSCO (00$\ell$) reflection with their finite size effect oscillations as well as the peaks due to the substrate (S). The peak denoted by (M) comes from the material used to hold the sample in the diffractometer.
Fig. 3. Resistance measurements for different thicknesses and kinds of substrate. The curves are plotted in terms of the resistance as measured between two indium contacts (10 mm long) separated by 4 mm.
Fig. 4. Inverse kinetic inductance \((\propto \lambda_{ab}^{-2})\) vs the temperature for samples on different substrates and with different thicknesses. The inductive measurement presented here was performed with a frequency of 1kHz and a drive current of 10 \(\mu\)A. All films (LSCO 4/1/2, STO 10/4/2 and buffer-less SLAO 0/2/0) present a similar critical temperature. The dashed lines show the quadratic fit of the data in the low temperature regime.