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Direct angle resolved photoemission spectroscopy and superconductivity of strained high-T_c films

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Abstract. Since 1997 we systematically perform direct angle resolved photoemission spectroscopy (ARPES) on *in-situ* grown thin (<30 nm) cuprate films. Specifically, we probe low-energy electronic structure and properties of high-T_c superconductors (HTSC) under different degrees of epitaxial (compressive vs. tensile) strain. In overdoped and underdoped in-plane compressed (the strain is induced by the choice of substrate) ≈15 nm thin $La_{2-x}Sr_xCuO_4$ (LSCO) films we almost double T_c to 40 K, from 20 K and 24 K, respectively. Yet the Fermi surface (FS) remains essentially two-dimensional. In contrast, ARPES data under tensile strain exhibit the dispersion that is three-dimensional, yet T_c drastically decreases. It seems that the in-plane compressive strain tends to push the apical oxygen far away from the CuO_2 plane, enhances the two-dimensional character of the dispersion and increases T_c, while the tensile strain acts in the opposite direction and the resulting dispersion is three-dimensional. We have established the shape of the FS for both cases, and all our data are consistent with other ongoing studies, like EXAFS. As the actual lattice of cuprates is like a 'Napoleon-cake', i.e. rigid CuO_2 planes alternating with softer 'reservoir', that distort differently under strain, our data rule out all oversimplified two-dimensional (rigid lattice) mean field models. The work is still in progress on optimized La-doped Bi-2201 films with enhanced T_c.

Keywords. Condensed matter physics; high-T_c superconductivity; electronic properties; photoemission spectroscopy; angle resolved photoemission spectroscopy; cuprates; films; strain; pulsed laser deposition.

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1. Introduction: High-T_c superconductivity after 20 years

It is generally recognized that even after 20 years of research and more than 70,000 publications, the mechanism of high-T_c superconductivity in cuprates remains a highly controversial topic [1–4], partly due to the fact that most groups do not fabricate and/or fully control and analyse their (non-trivial) perovskite samples

[1–9]. ARPES measurements on high quality samples across the phase diagram of representative compounds in the form of single crystals (mainly on cleaved Bi-cuprates) and thin films (very few reports; none on strained films, except our ongoing work) continue to be very important [2]. It is precisely in that context that we focus our systematic research activities as we fabricate state-of-the-art thin films (with and without growth induced strain) [9] and in most cases the corresponding single crystal samples (grown by H Berger, EPFL). Moreover, we do all the measurements within our group with core collaborations within EPFL (Forro *et al*, Grioni *et al*). In this paper we summarize our main results on studies of electronic properties and direct ARPES on strained films of LSCO-214 and briefly outline the ongoing work on La-doped Bi-2201 and other related compounds.

2. High-T_c cuprate films: Doping, strain and superconductivity

Following several ARPES studies on our cleaved Bi-2212 single crystals [10,11], we have by now completed more than 10 years of systematic studies on films at the Wisconsin synchrotron. After numerous experiments [12], in the year 2002, we have finally succeeded in performing a direct photoemission spectroscopy of thin (<30 nm) superconducting LSCO cuprate films (without cleaving the film) [13] with a special emphasis on the role of strain and its influence on electronic properties [11]. In-plane compressive strain is known to increase the critical temperature (T_c) of high temperature superconductors (HTSC) [6], and is obviously related to the mechanism of high-T_c superconductivity [1,3,6]. The phenomenon is quite dramatic in LSCO thin epitaxial films: for $x = 0.1$, T_c doubles with respect to relaxed LSCO and increases by a factor of five with respect to films with in-plane tensile strain [6,13]. The published theoretical studies predicted the in-plane compressive strain to flatten the bands: this could provide a simple explanation for the dramatic T_c-increase, since band flattening implies an enhanced density of states (DOS) near the Fermi energy. However, our direct angle-resolved photoemission spectroscopy (ARPES) study on compressively strained LSCO-214 films flatly contradicts [13] that picture by revealing a dispersing band that crosses Fermi level.

Moreover, our ARPES measurements on films with huge tensile strain (c-axis of $\sim$13.10 Å, corresponding to the c-axis compression of 1%) show the evidence for a three-dimensional dispersion, in clear contrast with the strictly two-dimensional dispersion observed in the aforementioned compressively strained films (see figure 1; for details see [14]). Although these results are again striking and somewhat unexpected, they make sense. Namely, already the conduction band of relaxed LSCO is atypical. It has considerable apical-oxygen p_z and Cu $3d_{z^2-r^2}$ out-of-plane character, while, in other cuprate phases, those orbitals hybridize far less with the conduction band. Hence, we can relate the observed z-axis dispersion with the significant displacement of the apical-oxygen towards the CuO_2 plane, induced by the epitaxial strain. Under extreme tensile strain, resistivity measurements show an insulating behavior of films and no T_c. Films with weaker tensile strain still exhibit superconductivity, but diminished as compared to the relaxed films. In summary, while the in-plane compressive strain tends to push the apical oxygen far away from the CuO_2 plane, enhances the two-dimensional character of the dispersion

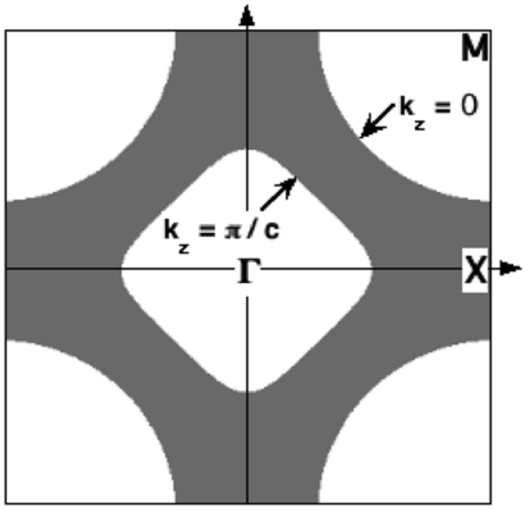

Figure 1. Projection of the reconstructed Fermi surface of an overdoped LSCO-214 film under tensile strain.

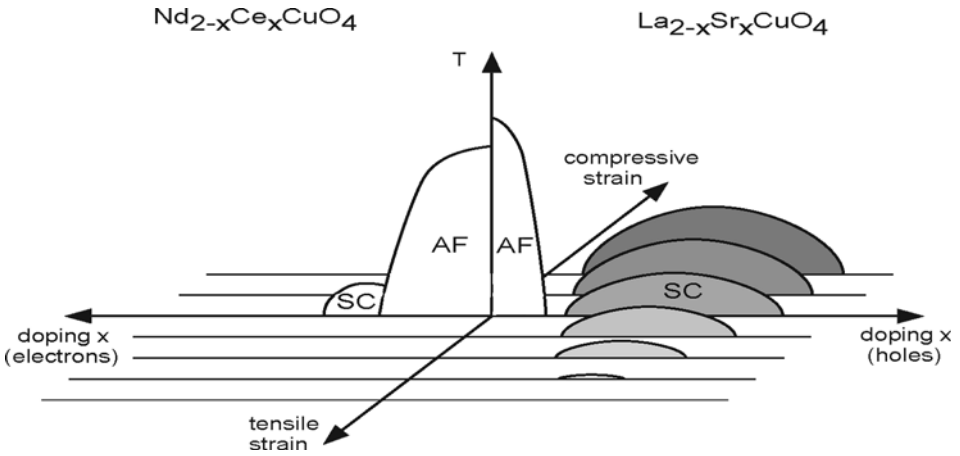

Figure 2. Schematic presentation of the electronic phase diagram of La$_{2-}$ as a function of doping, taking into consideration compressive and tensile strain for the superconducting phase (SC). For its electron-doped counterpart, $Nd_{2-x}Ce_xCuO_4$, there are still no strain data.

and increases T_c, the tensile strain seems to act exactly in the opposite direction [14] and the resulting dispersion is three-dimensional as discussed by Bansil *et al* [15].

For completeness, here is also a summary of other relevant results obtained so far in our ongoing systematic studies on direct ARPES on strained high-T_c films [12,13]:

(i) We have measured virtually identical ARPES dispersion on cleaved LSCO-214 single crystals and on as-made relaxed (i.e. no strain) films. That clearly removes any ambiguity on whether results on cleaved crystals are different from those measured on films (apart from the changes deliberately induced by the growth-induced strain as we do in our studies) [12].

(ii) Cuprate films with ‘chains’, like RBCO-123 (R=Y,Nd), tend to lose oxygen from the ‘chains’ above 100 K. So their surfaces do not show clear, stable evidence of the Fermi edge when measured using a cylindrical mirror analyser (CMA). Therefore, these films are not suitable for ARPES studies, and so we now concentrate on other compounds, notably LSCO and the BSCCO films [13,14].

(iii) Interesting new observations were made on ultra-thin films of $Bi_2Sr_2CaCu_2O_{8+x}$ (Bi-2212) grown on STO substrates. All films (even those whose thickness corresponds to 1/4 of a unit cell) show metallic-like Fermi edge in the PES spectra. However, a structural phase transition occurs at a nominal thickness of one unit cell (UC), converting the precursor $Bi_2O_{2.33}$(Bi_6O_7) highly coherent ultra-thin film into the $Bi_2Sr_2CaCuO_{8+x}$ epitaxial structure. In other words, the BSCCO-2212 phase forms for deposits above 1 UC, yet the very first UC represents metallic Bi_6O_7 that can be considered as a natural (self-organised) buffer for the growth of BSCCO-2212 films. In contrast, our preliminary results indicate that thin LSCO-214 films form by gradually connecting deposited ‘islands’ ∼60 nm in diameter [12–14].

In addition, various *ex-situ* measurements were performed by collaborating with others: EXAFS (S Conradson, LANL), microwave studies (A Dulcic, PMF, Zagreb), and paraconductivity (F Vidal *et al*, Santiago) and the work is in progress on oxygen-treated LBCO films grown by MBE (I Bozovic *et al*, BNL). All these measurements allow us to study the normal and the superconducting state, and the overall property changes for each film sample. However, more studies are necessary to arrive at a more quantitative picture and discuss subtle consequences for the microscopic mechanism of high-T_c superconductivity. Still, it is evident that our results on strained films cannot simply be explained within any simple two-dimensional rigid lattice model. This is mainly due to the fact that the lattice is like a ‘Napoleon-cake’, i.e. CuO_2 planes are rigid, while the out-of-plane ‘reservoir’ is softer, and so the strain differently distorts these respective sublayers of the lattice; to determine the exact changes due to strain of all various coordinates is at present too demanding a task. Nevertheless, considering the strong influence of strain on T_c, the electronic phase diagram for LSCO (and cuprates in general) has to be extended by a third strain axis. The resulting, illustrative phase diagram most likely resembles the one presented in figure 2, and shows that our experimental work is rather promising to eventually understand the mechanism of high-T_c superconductivity. After we demonstrate the direct ARPES on thin cuprate films [12–14], similar approach is now being pursued by several leading groups worldwide, Tokyo, Tsukuba, Brookhaven, Berkeley, Argonne, Stanford and Villingen, among others.

Table 1. Measured critical temperature T_c of the unstrained and compressively strained LSCO films.

Doping (x)	No strain/T_c (K)	Compressive strain/T_c (K)
0.10	20	40
0.15	38	44
0.20	24	40

3. Concluding remarks and further work

As summarised in table 1, T_c is enhanced in all our compressively strained films, yet it diminishes and ultimately disappears in tensile strained films. What is truly striking is that by using essentially the same growth and oxidation procedure in underdoped ($x = 0.1$) and overdoped ($x = 0.2$) LSCO films we essentially double T_c in both cases simply by changing the substrate of the film (thereby altering the compressive strain).

It is highly unlikely that the aforementioned, measured changes in the in-plane electronic structure (see the discussion in the previous section), are sufficient to account for the striking T_c-enhancement effects. Especially interesting is the case of overdoped films ($x = 0.2$). In that regime, where samples should behave almost as in the Fermi liquid/BCS picture, an enhancement of density of states would be a natural explanation, yet that does not seem to be the case according to our ARPES results [14]. As the compressive strain clearly alters the lattice and increases the c-axis length, one has to seriously take models that take into account the role of the lattice and of the strain (see numerous contributions by the authors cited in ref. [16]). It seems difficult to argue that some sort of 'magnetic' mechanism that completely neglects the lattice effects would give such a T_c-enhancement, especially in the overdoped samples. Yet this possibility cannot be ruled out without additional work. However, a better understanding of basic mechanisms that govern superconductivity in these complex oxide films is an immediate prerequisite and our main goal.

We now have the ARPES data on Fermi surface (FS) mapping in both, relaxed, compressively and tensile strained optimal and overdoped LSCO-214 films [14]. We also have first spectra for the Bi-2201 films. We have already made systematic improvements in the growth and property optimization of strained La-doped BSCCO-2201 epitaxial films. So, in the near future we shall perform ARPES measurements (high resolution in high symmetry directions, temperature dependence) on optimized Bi-2201, single-CuO_2-layer strained films. We want to determine the changes of the Fermi surface with strain, and determine the variation of T^* and T_c (with doping and strain) and compare the results with those made by our other complementary *ex-situ* studies. Eventually, we intend to grow and study the electron-doped compounds $Nd_{2-x}Ce_xCuO_4$; these have never been studied under strain (see also figure 2). Finally, beyond 2008 we intend to use the resonant inelastic X-ray scattering technique (RIXS), that is currently being developed by M Grioni *et al.* The RIXS should enable us to verify in-depth electronic features of these layered materials (as compared to ARPES data).

All our experiments are directly relevant for understanding the superconductivity mechanism in cuprates as our strain experiments drastically alter, not only T_c but also the electronic structure and the normal state properties. Crudely speaking, if the compressive strain (that enhances T_c, i.e. the phase coherence) enhances also the ARPES-pseudogap T^* it would mean that the pairing is also enhanced and we want to determine by how much (if at all). Moreover, the respective changes of the density of states near the (antinodal) hot spots (related to T^*) and the cold spots (more itinerant states) of the Fermi surface should give us further insight into subtle interactions that play a role in high-T_c cuprates. To the best of our knowledge such an approach is still unique in that we fabricate our own samples and perform most of the experimental measurements.

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