

NanoEngineering of Oxide Heterostructures: The Field Effect & Interface Superconductivity



The Field Effect and (Interface) Superconductivity in High-T_c and Related Films

Davor Pavuna*

Physics of Complex Matter - FSB - <u>http://cream.epfl.ch</u> - station 3 Swiss Federal Institute of Technology at Lausanne (EPFL) CH-1015 Lausanne - EPFL, Switzerland

* in collaboration with Guy Dubuis (EPFL & BNL) & Ivan Bozovic et al. (BNL-USA)

Abstract

In the past decade we have systematically probed low energy electronic structure and measured properties of high-T_c films under different degree of epitaxial (compressive vs tensile) strain [1-5]. In overdoped in-plane compressed LSCO-214 films we doubled T₅, from 20K to 40K, yet Fermi surface (FS) remained essentially two-dimensional (2D) [1]. In contrast, tensile strained films exhibit 3D dispersion; T_e is drastically reduced [2]. It seems that the in-plane compressive strain pushes the apical oxygen away from the CuO₂ plane [1-3], enhances the 2D character of the dispersion and enhances T_{ex} while the tensile strain seems to act in the opposite direction and the resulting dispersion is 3D [2-5]. Evidently, such studies are directly relevant for the mechanism of high-T_c, as we have also obtained the FS topology for both cases. As the actual lattice of cuprates consists of rigid CuO, planes that alternate with softer 'reservoir' (that strains distort differently), our results tend to rule out 2D rigid lattice mean field models [3]. We have also mapped the 3D FS topology from the observed wavevector quantization. in cuprate films thinner than 18 units cells [5]. Moreover, we have successfully grown doped Bi-2201 thin films and performed XRD, transport and preliminary in-situ ARPES studies [6]. In collaboration with Ivan Bozovic and his colleagues at BNL, we have studied the field effect in new heteroepitaxial cuprate SuFET structures. Very large fields and induced changes in surface carrier density enable large shifts in the critical temperature (T_c). We were able to substantially vary T_c with the field and observed striking superconductor-insulator quantum phase transition in La_{2,4}Sr₂CuO₄ films that poses profound. challenge to contemporary theory [7]. We briefly cricitally discuss the significance of our results in the light of the emerging understanding of High-T_c and related phenomena.

References

- 1. M. Abrecht et al. PRL 91, 057002 (2003); EPFL Thesis no. 2792 (2003)
- D. Cloetta et al., Phys. Rev. B74, 014519 (2006); EPFL Thesis no. 3333 (2005)
- 3. D. Pavuna et al., Journal of Physics 108, 012040 (2008) and references therein
- C. Cancellieri et al., Phys. Rev. B76, 174520 (2007); EPFL Thesis no. 4120 (2008)
- 5. D. Ariosa et al. Appl. Phys. Lett. 92, 092506 (2008) and references therein
- D. Oezer, M.Sc. Thesis, EPFL (2009); N. Wooding et al., unpublished (2012)
- A.T. Bollinger et al. Nature 472, 458 (2011); G. Dubuis et. al. in print (2012).

Keywords : Superconductivity, Cuprates, Heteroepitaxy, ARPES, Transport, Field effect.

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Outline

Historic Introduction
The Field Effect (Disorder) ...
Interface Superconductivity
New Superconductors

Dedicated to the memory of Zlatko Tesanovic and Sasha Aleksandrov

Cuprates Electronic Phase Diagram:



UNDERSTAND = Superconductivity Textbook

INTRODUCTION TO SUPERCONDUCTIVITY AND HIGH-T_c MATERIALS

World Scientific

About the Book

"... an introductory text, with a unified, balanced point of view, is of considerable value. This is what Cyrot and Pavuna have produced. Their book still requires a significant effort for a genuine beginner, but it can be studied step by step. It sets up delicate compromises between the opposite dangers of dogmatism and oversimplification."

from the foreword by **P G de Gennes**

What sets this book apart from others on the introduction to superconductivity and high- T_c materials is its simple and pragmatic approach. The authors describe all relevant superconducting phenomena and rely on the macroscopic Ginzburg-Landau theory to derive the most important results. Examples are chosen from selected conventional superconductors like Nb-Ti and compared to those of high- T_c materials. The text should be of interest to students and researchers in all branches of science and engineering, with the possible exception of theoretical physicists, who may require a more mathematical approach.

SUPERCONDUCTIVITY AND HIGH T, MATERIALS **NTRODUCTION TO**

Cyrot Pavuna

Michel Cvrot

Davor Pavuna

Helmuth Berger @ EPFL: Growth of Single Crystals >30'000 samples' !!!









I. Vobornik et. A (1999)l

ARPES on Cleaved Bi-2212 (I. Vobornik)



Disorder in $Bi_2Sr_2CaCu_2O_{8+x}$ SC-state:



Defect-induced loss of coherence

I. Vobornik *et al*., PRL **82**, 3128 (1999)

ARPES Pseudogap & Disorder



Doping vs. deliberate disorder (L.Forro):



Disorder, doping, strains & high Tc

Verify the bulk properties as discussed by Alloul et al.!

Critical temperature of superconductors <1986



Conventional Superconductors Barisic-Labbé-Friedel theory of superconductivity (1967)



Hangzhou Workshop on Quantum Matter

Two key words: Emergence and Control

How to understand and control the remarkable emergent properties of correlated quantum matter?

Yu Lu

Ivan Bozovic & Davor Pavuna Control

Correlated Oxide Materials:

Physics & Nano-Engineering I – VI (SPIE,USA)

2. ARPES on In-Situ Grown FILMS:

Physics <-> HeteroEpitaxy

Control E(k) ... (LATTICE) STRAINS

Control 'building blocks' + **Functionality**

In-Situ ARPES on UNCLEAVED Films

Top Surface is Crucial&INT.

& INTERFACE Rules !





PLD Cuprates HeteroEpitaxy

DARPES on As-made UNCLEAVED thin films

DARPES on As-Grown Film Surfaces

Grioni's ARPES ← Laser ablation



FP**F**[

ÉCOLE POLYTECHNIQUE

FÉDÉRALE DE LAUSANNE



Pavuna et al. J. Phys. (GB) 108, 012040 (2008)



Connection

DARPES

Direct ARPES on As-made La-doped Bi-2201 Film: NOT Cleaved



In-Situ ARPES @ Synchrotron in Wisconsin





DARPES on Thin Oxide Films

La_{2-x}Sr_xCuO₄ Films Under Strains



In-plane tensile strain (SrLaGaO₄, SrTiO₃)

a)

b)

for an unstrained entirely dened single envetal (right)



Direct ARPES on Strained HTSC Films





An illustration of reconstructed FS for overdoped (x=0.2) LSCO-214 films under

compressive strain Tc is enhanced PRL, 2003

and

tensile strain .

Tc is diminished

PRB, 2006

Ultra-Thin Films: 2D-3D Crossover

Discrete generic 3D -tight-binding model

Parameter	Hopping term [mW]
t	in-plane nearest neighbor (NN)
t'	in-plane next nearest neighbor (NNN
t''	out-of-plane NN
$t^{\prime\prime\prime}$	out-of-plane NNN







r indicates predominant dimensionality of the electronic band dispersion.

The quantization of the electron wavevector









Appl. Phys. Lett. 92, 092506 (2008)



Ming Shi & Joel Mesot, PSI Fermi surface of ortho-II YBCO(123)





* In-Situ ARPES on As-grown Films Works !

* < 18UCs LSCO-214 Strained Films Show Different Dispersions (3D FS)

*

T_c scales with c-axis (apical oxygen)

DARPES on Thin Oxide Films



Increase in T_c^{max} with Lanthanum-Barium co-doped Sample

T_c vs. S(300) In doped LXCO-214

Thierry Schnyder, Jeff Tallon et al. (2010)



Figure: T_c vs. S(300)

T_c scales with c-axis (apical oxygen)

NanoEngineering & Electronic Diagram



See also Cloetta et al. PRB 2006

MBE Grown Compressively Strained LSCO Film (BNL):

Resistivity of an LSCO film on LSAO substrate.





Bozovic et al. PRL 89, 107001 (2002)

High-Tc Materials COMPLEXITY: Unwanted phases OUR MAIN PROBLEM



Phase formation in the Bi-Sr-Cu-O system (@ 800°C, in air)

Courtesy I. Bozovic



3. Electric Field Effect in LSCO Films Guy Dubuis @BNL with Ivan Bozovic et al.

• Use Electric Field to tune T_c

• Adjust carrier concentration • without changing disorder

* Intrinsic nature of quantum critical points ?





Field Effect applied to high-Tc Superconductivity (1991)

Electric field effect on superconducting $YBa_2Cu_3O_{7-\delta}$ films

J. Mannhart, J.G. Bednorz, K.A. Müller, and D.G. Schlom

Z. Phys. B - Condensed Matter 83, 307-311 (1991)

 «... density of free carriers in the YBa₂Cu₃O_{7-δ} films can be modified by 1-2% with gate voltages smaller than 50 V. »



To induce large changes in the carrier concentration, i.e. $\sim 10^{21}$ cm⁻³, the breakdown field of the insulator is required to be at least 10⁸ V/cm and such an insulator does not exist.

Field effect & correlated oxydes



Ahn et al., Nature 424, 1015-1018(2003)

High-Density Carrier Accumulation in ZnO Field-Effect Transistors Gated by Electric Double Layers of Ionic Liquids

By Hongtao Yuan,* Hidekazu Shimotani, Atsushi Tsukazaki, Akira Ohtomo, Masashi Kawasaki, and Yoshihiro Iwasa

Adv. Funct. Mater. 2009, 19, 1046–1053



N,N-Diethyl-N-methyl-N-(2-methoxyethyl)ammonium

bis(trifluormethane sulfonyl)imide

Ivan Bozovic's ALL-MBE @BNL



The Sample



(i) 1 unit cell (UC) thick $La_{1.56}Sr_{0.44}CuO_4$ buffer layer (ii) 5 UC of insulating La_2CuO_4 (LCO) (iii) 1, 1.5, or 2 UC of $La_{2-x}Sr_xCuO_4$ (LSCO); x = 0.10-0.20Nature 472, 458 (2011)

Electric field causes large changes in carrier density and T_c





Diamagnetic response measured by 2-coil mutual inductance technique confirms the results of transport measurements

Field-induced shifts: $\Delta x = \pm 0.04, \Delta T_c = 30 \text{ K}.$

[Nature 2011]

S-I quantum phase transition induced by field effect in 1 UC thin LSCO layer



Note the T-independent separatrix at $R_0 = R_0 = 6.45 \ k\Omega$; this corresponds to $\rho \approx 400 \ \mu\Omega$ cm

$$R_Q \equiv h/(2e)^2 = 6.45 \ k\Omega$$

[Nature 2011]

Scaling Analysis (QPT)



Markovic et al., Phys. Rev. B. 60, 4320 (1999).

The same data replot as a set of R(x) curves for finely scaled values of T



Quantum critical point at $x_c = 0.06$

Good scaling to 2D S-I QPT → ultrathin active layer → field effect rather than chemistry The same data scaled against a single variable $u = |x-x_c| / T^{2/3}$



The critical resistance equal to $R_Q = h/(2e)^2 = 6.45 \ k\Omega$. Existence of electron pairs on both sides of the quantum phase transition \Rightarrow "Bosonic scenario": Cooper pair mobile and Bose-condensed, vortices localized and immobile on the S side; dual (reverse) on the I side \Rightarrow 'Elusive Bose metal' at $x = x_c$

[Nature 2011]

• The critical sheet resistance is equal to $R_Q = h/(2e)^2 = 6.45 \ k\Omega$.

Existence of electron pairs on both sides of the quantum phase transition.

→ "Bosonic scenario": on the S side Cooper pairs are mobile and Bose-condensed, while vortices are bound; dual (reverse) on the I side.

Exact self-duality > similar symmetry, interactions >
on-site Coulomb repulsion not sufficient?

Elusive Bose metal' at $x = x_c = 0.0605$

+ several experiments in progress ...

Discovery of superconductivity in KTaO₃ by electrostatic carrier doping

K. Ueno^{1,2}, S. Nakamura^{3,4}, H. Shimotani⁵, H. T. Yuan⁵, N. Kimura^{4,6}, T. Nojima^{3,4}, H. Aoki^{4,6}, Y. Iwasa^{5,7} and M. Kawasaki^{1,5,7}*



Figure 1 | Electric double-layer (EDL) transistor. a,**b**, Schematic diagrams (**a**) and photograph (**b**) of the EDL transistor with an ionic liquid electrolyte, DEME-BF₄. DEME⁺ ions comprise the cations and BF_4^- ions ore the anions. The device was fabricated on a KTaO₃ single crystal. Source, drain and gate electrodes were fabricated on the crystal (black area in the photograph), and the entire surface of the crystal, except for the channel area and electrodes, was covered by separator layer (yellow area in the photograph). A small amount of the ionic liquid was dropped on the crystal so that it covered the channel region (KTaO₃ surface) and the gate electrode. **c**, Molecular and crystal structures for the anion, cation and KTaO₃.

Electrostatic Control of the Evolution from a Superconducting Phase to an Insulating Phase in Ultrathin YBa₂Cu₃O_{7-x} Films

Xiang Leng,¹ Javier Garcia-Barriocanal,¹ Shameek Bose,² Yeonbae Lee,¹ and A. M. Goldman¹



Superconductor-to-Insulator Transition and Transport Properties of Underdoped YBa₂Cu₃O_y Crystals

Kouichi Semba and Azusa Matsuda

NTT Basic Research Laboratories, 3-1, Morinosato Wakamiya, Atsugi-shi Kanagawa 243-0198, Japan (Received 30 November 1998; revised manuscript received 5 July 2000)

The carrier-concentration-driven superconductor-to-insulator (SI) transition as well as transport properties in underdoped YBa₂Cu₃O_y twinned crystals is studied. The SI transition takes place at $y \approx$ 6.3, carrier concentration $n_H^{SI} \approx 3 \times 10^{20}$ cm⁻³, anisotropy $\rho_c/\rho_{ab} \approx 10^3$, and the threshold resistivity $\rho_{ab}^{SI} \sim 0.8 \text{ m}\Omega$ cm which corresponds to a critical sheet resistance $\frac{h}{4e^2} \approx 6.5 \text{ k}\Omega$ per CuO₂ bilayer. The evolution of a carrier, $n_H \propto y - 6.2$, is clearly observed in the underdoped region. The resistivity and Hall coefficient abruptly acquire strong temperature dependence at $y \approx 6.5$ indicating a radical change in the electronic state.



Again Rc ≈ h/(2e)² (Arrow)

Two-dimensional electron localization in bulk single crystals of $Bi_2Sr_2Y_xCa_{1-x}Cu_2O_8$

D. Mandrus, L. Forro,* C. Kendziora, and L. Mihaly

Department of Physics, State University of New York at Stony Brook, Stony Brook, New York 11794-3800 (Received 25 February 1991; revised manuscript received 16 May 1991)

The temperature dependence of the *ab*-plane electrical transport was investigated on a series of single-crystal $Bi_2Sr_2Y_xCa_{1-x}Cu_2O_8$ samples with a wide range of yttrium content. The observed metal-insulator transition is discussed in terms of two-dimensional localization. The Hall-effect and thermopower results indicate that the driving force of the localization is the random impurity potential.



One of the first observations of $Rc \approx h/(2e)^2$ (Arrow)

Interface Superconductivity & New Superconductors



Ivan Bozovic (BNL)

Interface superconductivity: a modern classic

D. Saint-James and P. G. De Gennes	Phys. Lett. 7 306 (1963)
V. L. Ginzburg	Phys. Lett. 13 , 101 (1964)
M. Strongin	Phys. Rev. Lett. 10, 442 (1964)

Web of Science: "Interface superconductivity"			
1973-2012	1006 papers		
1973-1976	3 papers	(Overton '73, Tsuei '74, Ickson '75)	
2009-2012	292 papers (<i>Nature</i> 12, <i>NM</i> 8, <i>N</i>	<i>IP</i> 4, <i>NC</i> 2, <i>Science</i> 6, PRL 40, PRB 188; Σ = 260)	

Why is *IS* difficult?

Conventional metals - BCS superconductors: $n \sim 1023 \text{ cm}-3 \implies I_{TF} \sim 2-3 \text{ Å}$ $\xi \sim 100-1,000 \text{ Å}$ (870 Å in lead, 16,000 Å in Al,...) \Rightarrow the modified (interface) layer is too thin for SC to occur! Plus, reduction in carrier density generally *decreases* T_c.

Cuprates: $n \sim 1.5^*10^{21} \text{ cm}^{-3} \Rightarrow I_{TF} \sim 5.10 \text{ Å}$ high $T_c \Rightarrow$ short coherence lengthextreme anisotropy $\Rightarrow \xi_c \sim 1.2 \text{ Å}$ in OD cuprates, reduction in n *increases* T_c .

SrTiO₃:

$$n \sim 10^{18} \cdot 10^{19} \text{ cm}^{-3} \implies I_{TF} \sim 5 \cdot 10 \text{ nm}$$

a chance to realize interface superconductivity!

LaAIO₃ / SrTiO₃ & related

2004: Discovery of high-mobility 2DEG (Ohtomo & Hwang)

2007: Discovery of interface superconductivity (Triscone & Mannhart)

2008: Electric field control (Triscone & Mannhart)

2009: Thickness of 2DEG measured by cross-section AFM (Basletic)

2009: Quantum oscillations (Hwang)

2009-11: Ferromagnetism (Hilgenkamp, Ashoori, Moller, Mannhart, Eom, Schuller)

2010: Tunable Rashba Spin-Orbit coupling (Triscone, Dagan, Hwang)

2010: Interface superconductivity in LaTiO₃ / SrTiO₃ (Lesueur)

2011: Enhancement of T_c in H_I (Schlom)

Devices used to produce 2D or interface superconductivity



Hwang, Iwasa, Kawasaki, Keimer, Nagaosa & Tokura, Nature Matter. 11, 108 (2012)

INTERFACE SUPERCONDUCTIVITY IN CUPRATES





R(T) for various bilayers.

I-M and *M-I* bilayers show superconductivity even though neither of the two constituents does.

In single-phase S or S' films grown under the same conditions $T_c \leq 40$ K.

Gozar et al., Nature 455, 782 (2008)

INTERFACE-ENHANCED SUPERCONDUCTIVITY



Superconductivity at ELT / YBa₂Cu₃O_{7-x} Interface



Sheet resistance vs temperature, tuned by the gate voltage.

T_c vs carrier density

R(x) isotherms. Inset: 2D QCP scaling, for $R_e = h/4e^2$; vz = 2.2.

Leng et al., PRL 107, 027001 (2011)

Field effect reveals new physics in YBa₂Cu₃O_{7-x}



Electronic phase transition in the Fermi surface near the optimal doping level.

Leng et al., PRL 108, 067004 (2012)



Metallic but non-superconducting YBCO, doped n-type by electrolyte field-effect

Nojima et al., PRB 84, 020502 (2011)

Superconductivity at ELT / ZrNCI Interface





- (a) Schematics of EDLT
- (b) The crystal structure of ZrNCI

Temperature dependence of the channel sheet resistance at different gate voltages for a ZrNCI/SrTiO₃ bilayer.

Ye et al, Nature Matter. 9, 125 (2010)

INTERFACE-ENHANCED SUPERCONDUCTIVITY IN FeSe



a) SrTiO₃ substrate, b) FeSe film, c) schematics, d) topograph showing Se termination, e) 1 UC film, f) 2UC film.



NB: In bulk FeSe, T_c = 9.4 K

Yan et al., Chin. Phys. Lett. 29, 037402 (2012)

New Superconductors

Superconductivity at ELT / KTaO₃ interface



(a) EDLT schematics; (b) EDLT photo.
(c) the structure of BF⁴⁻, DEME⁺ and KTaO₃.



- (a) Sheet resistance vs temperature, for V_G = 5 V.
- (b) $R_s vs H$, at T = 20 mK.
- (c) I-V characteristics, at T = 20 mK.

Ueno et al., Nature Nanotech. 6, 408 (2011)

Prediction of HTS in $LaMO_3/LaNiO_3$ (M = AI, Ga, Ti,...)

PRL 100, 016404 (2008)

PHYSICAL REVIEW LETTERS

week ending 11 JANUARY 2008

Orbital Order and Possible Superconductivity in LaNiO3/LaMO3 Superlattices

Jiří Chaloupka1,2 and Giniyat Khaliullin1



Hansmann et al, PRL 103, 016401 (2009)

Summary: What have we learned so far?

- Interface superconductivity has been realized by (a) atomic-layer synthesis, and (b) electric-field effect (mainly using electrolytes).
- New techniques have been developed to measure profiles of chemical composition, crystallographic structure, mobile carrier concentration, and superfluid density across the interfaces, with atomic resolution.
- In the cleanest samples, the predominant mechanism is electronic reconstruction and ionic displacements.
- quasi-2D superconductivity has been observed in SrTiO₃, LSCO, YBCO, etc., with T_c as high as in bulk samples.

5. A new superconductor, KaTaO₃ ($T_c = 0.047$ K), has been created by field effect.

 Enhanced superconductivity has been observed at LSCO/LCO (+25%), SrTiO₃/FeSe (+200%), and PbTe/PbS (+600%) interfaces.

Outlook: some open questions

A. SrTiO₃

Inhomogeneity: electronic or atomic (e.g. oxygen vacancies)? Role of ferromagnetism, Rashba spin-orbit coupling, Kondo effect? Partners other than LaAlO₃: band, Mott, FM, AF, FE, etc. Other titanates: BaTiO₃, CaTiO₃, DyTiO₃, LaTiO₃; Sr₃Ti₂O₇, ...?

The mechanism of superconductivity – conventional?

B. Cuprates

Cuprates other than LSCO and YBCO: CaCuO₂, SrCuO₂, n-type? Non-cuprate partner materials (insulators, metals)?

C. Other superconductors

Heterostructures based on other known superconductors?

Electric-field tuning of S-I and S-M quantum phase transitions?

Beyond 2012: 'Gate Every Electronic Material'

>300 K

Our (Students') Dream !

2100



