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I. Bozovic

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Is there a path from cuprates towards room-temperature superconductivity?

I. Božović^{1,2}, J. Wu¹, X. He^{1,2} and A. T. Bollinger¹

¹*Brookhaven National Laboratory*

²*Applied Physics Department, Yale University*

Abstract

A brief account is presented of an extensive experiment performed at Brookhaven National Laboratory, aimed at understanding the nature of high-temperature superconductivity in cuprates. Over the course of the last twelve years, over 2,000 films of the prototypical high- T_c superconductor, $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, have been synthesized using atomic-layer-by-layer molecular beam epitaxy (ALL-MBE), characterized by a range of techniques, and patterned into devices. These were used to measure accurately the key physical parameters in both the superconducting and the normal states, and establish their precise dependence on doping, temperature, and external fields. The results bring in some great surprises, challenge the commonly held beliefs, rule out many theoretical models, and point to a new path for raising T_c even further.

Keywords: high-temperature superconductivity, cuprates, molecular beam epitaxy, superfluid density, electronic nematicity

1. Background and motivation

Ever since Bardeen, Cooper and Schrieffer (BCS) presented their renowned theory of superconductivity¹, it has been the standard explanation of the phenomenon, in all known superconductors. This picture has been challenged by the discoveries of high-temperature superconductivity (HTS) in various cuprate compounds, by G. Bednorz and K. A. Mueller, Ching-Wu (Paul) Chu, and others, with the record critical temperature rising within just a few years by more than six-fold to reach 135 K (and 164 K under pressure).²⁻⁴ Right away, Phillip W. Anderson asserted that the superconducting state in cuprates is not just unusual, but in fact unprecedented: a Bose-Einstein condensate (BEC) of charged bosons ('holons').⁵ Several other prominent theorists – Nevil Mott, T. D. Lee, A. F. Andreev, Pierre-Gilles De Gennes and Guy Deutscher, among others – also took this stand, as well as K. A. Mueller himself.⁶⁻¹⁰ The plausibility of this picture has been reinforced subsequently, because experiments with cold atoms have shown that a cross-over from BCS to BEC can be tuned by increasing the pairing interaction strength, and/or by making the cloud thinner and crossing over from the three-dimensional (3D) to quasi-two-

dimensional (2D) behavior. In either case, T_c increases and gets much higher on the BEC side.¹¹⁻¹³ Thus, both the high T_c and the 2D nature of cuprates point to BEC rather than to BCS.

Nevertheless, this view has not been universally accepted, largely because angle-resolved photo-emission (ARPES) experiments have shown the presence of a Fermi Surface (FS) in metallic cuprates.¹⁴⁻¹⁶ This was confirmed by spectroscopic-imaging scanning-tunneling microscopy (SI-STM), angle-resolved magnetoresistance oscillations (MRO), and most spectacularly, by quantum (de Haas-van Alfen and Shubnikov-de Haas) oscillations.¹⁷⁻²¹ These findings have pointed to the more-or-less standard picture in which a BCS-like superconducting state evolves upon cooling from a Fermi liquid (FL) state.^{22,23} It seems that the majority of the researchers in the field favor this scenario — albeit with many variants insofar as the microscopic nature of the pairing interaction(s), or ‘glue’, is concerned — at least, on the overdoped side of the phase diagram.

This dichotomy is quite important because the physics in the two limits, BCS vs. BEC, is different both in terms of what controls T_c and in what type of excitations, quasi-particle like or collective, deplete the condensate upon thermal agitation. Still, the debate about the nature of superconducting state in cuprates has remained an open question.

2. The BNL high- T_c experiment

To address this important question, one of us (I.B.) proposed in 2004 to the US Department of Energy a project along the following lines.

In cuprates, the phase diagram on the underdoped side is quite complicated and controversial, with various types of order (charge density waves, spin density waves, etc.) observed, and many more hypothesized. Pretty much every detail, including the relevance for the HTS phenomenon of these other orders, competing or intertwined, has been hotly debated. It seemed that the only detail that almost everyone agrees upon is that at the extreme overdoped side of the phase diagram the superconducting state is of the standard BCS nature (except for the unusual d-wave symmetry of the order parameter), and that it emerges from a standard FL state. If this is the case, one could start from this well-understood fixed point and move adiabatically towards the optimum doping, measuring on the way the most relevant physical properties of the superconducting state (T_c , the magnetic penetration depth, the coherence length) and of the normal state (the resistivity, the Hall constant, the magnetoresistance). Of course, one needs to make sure that the measured properties are intrinsic, which means that the samples must be of the highest quality, the measurements accurate, and the results robust and reproducible. Then one of two things could happen. All the physical properties may turn to evolve smoothly, in which case we would infer that the superconducting state at optimal doping is at least qualitatively similar to the BCS state. Alternatively, we may encounter some jumps and singularities, in which case all bets are off — the superconducting state at optimal doping may be of an entirely different nature, like the resonant valence bond (RVB) state of P. W. Anderson, anyons, loop currents, etc.

The project was approved and funded by U.S. Department of Energy, and the work at Brookhaven National Laboratory (BNL) started in earnest in 2005. Conceptually simple, this experiment turned out to require much technical development and innovation, and took a dozen years to complete.

3. Technical advances

‘Tuning the doping adiabatically’ translates into a need to produce many samples, with exquisite control of the chemical composition. For this, we have developed a special tool, an atomic-layer-by-layer molecular beam epitaxy (ALL-MBE) system.²⁴ It is equipped with advanced surface-science tools that enable detailed studies of the film growth. We designed the system to be completely modular, thus achieving a very high up-time and productivity; we perform over 200 synthesis experiments every year. Thus we were able to establish the ‘recipes’ for synthesis of the highest-quality, single-crystal thin films of HTS cuprates and other complex oxides, with atomically smooth surfaces and interfaces. In this study, we have focused on the prototypical high- T_c superconductor, $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO), because it is the simplest and because by varying the Sr doping level x we can cover the entire phase diagram, from insulator to superconductor to a non-superconducting metal. The best LSCO films show T_c uniform to the ± 0.1 K level, across a relatively large area ($10 \times 10 \text{ mm}^2$). Moreover, by virtue of our ‘digital’ synthesis, we can engineer the samples at an atomic-layer level, optimizing them according to the needs of a particular experiment²⁵⁻³⁵, which also turned out to be crucial for the task at hand.

In parallel, we have been developing and improving various techniques for measurements of the relevant physical properties. Important examples are the advances we made in the mutual inductance technique for measuring the magnetic penetration depth λ , a key parameter of the superconducting state. We have invented a device to suppress the thermal noise in a cryocooler to below 1 mK, an improvement by one-two orders of magnitude over the performance of commercially available units.³⁶ Tightening the electronic, mechanical and thermal stability of the mutual-inductance apparatus, we have achieved remarkable reproducibility; we have demonstrated that measurements on the same sample reproduced to within 0.1 % after one year.³⁷ We have also expanded the range of temperatures (down to 300 mK), frequencies (up to 50 MHz), and fields (up to 9 T). At this point, the accuracy of our measurements of $\lambda(T)$ is not limited by the hardware, but rather by how well one knows the film thickness.

The later also turned out to be nontrivial; it took a decade of experimentation and study. By virtue of our layer-by-layer synthesis technique we know the total film thickness to the crystallographic accuracy, but the problem is in the formation of the dead layer, of unknown thickness and composition, next to the substrate, and another one next to the surface, because of the interaction with the atmosphere. To solve this problem, we protect the film by a metallic buffer layer and an overlayer. This, however, introduces another problem: the chemical potential in the active HTS material is in general different from that in the compound used for the protective layers, so there is some charge redistribution across each interface. To minimize this, we use graded doping near the two interfaces, a technique developed for growth of III-V heterostructures. Our final touch is Zn δ -doping in the transition layers (see Figure 1), a technique we invented and used to demonstrate that the HTS interface superconductivity in LSCO/LCO heterostructures is confined to a single CuO_2 plane.²⁷ This removes the interface superconductivity in the transition layers, and we are left with a precisely known number of HTS CuO_2 planes.³⁴ In this way, we can measure the absolute value of the penetration depth to better than $\pm 1\%$ absolute accuracy.³⁷ One could call this metrology; this is as quantitatively accurate as can be done today.

To summarize this section, we have advanced the technique of synthesis of cuprate superconductors to a new level, both in terms of film quality and by engineering the sample at the atomic-layer level. As important, by virtue of great productivity of our ALL-MBE system, we have synthesized and studied a very large sample set of over 2,000 LSCO films, perhaps without precedence in condensed matter physics. This has been decisive for the success of this project. When

one deals with complex materials or objects, more is different; a large statistics reveals clear trends and intrinsic properties.

4. Penetration depth study: a strange superconducting state in LSCO

The result of “the Brookhaven HTS Experiment” that has been easiest to disclose, if not to interpret, has been a systematic dependence of λ on doping and temperature, covering densely the entire overdoped side of the LSCO phase diagram.³⁴ In a nutshell, the main finding is that, as we vary the doping from optimally doped all the way to non-superconducting metal, the penetration depth evolves smoothly and continuously, without any jumps. The data carry other important messages that get clearer if we plot the superfluid density per one LSCO formula unit, $N_s(T, p)$, which is proportional to $1/\lambda^2$. In Figure 2a, one can see that, surprisingly, N_s is essentially linear in temperature all the way to T_c , even in the most overdoped LSCO films. The same is true all the way up to the optimum doping.³⁴ Second, in BCS theory, at $T = 0$ the superfluid density $N_{s0} \equiv N_s(T \rightarrow 0)$ should be equal to the carrier density, so like the later N_{s0} should also increase with doping. What we see, however, is that as we increase the doping, N_{s0} decreases, all the way to zero. Third, one can express the superfluid density in the units of Kelvin to get the so-called superconducting phase stiffness. Intuitively, the later indicates the temperature scale at which thermal fluctuations of the *phase* of the superconducting order parameter get strong enough to destroy macroscopic superconductivity. We find that in cuprates this temperature is extremely small, roughly equal to T_c itself. To illustrate this point, we can compare a standard BCS superconductor such as Nb with an LSCO sample overdoped to exactly match the T_c of Nb. We find that in LSCO, the phase stiffness is three orders of magnitude smaller than in Nb — the smallest it could possibly be. We conclude that in cuprates, the physics is dominated by massive phase fluctuations, present over a very broad temperature range, as argued by Kivelson and Emery.³⁸ But, as seen from Figure 2b, a sharp Berezinskii-Kosterlitz-Thouless (BKT) phase transition is not observed, even in the films that are just a single unit cell thick. It can be observed only at very high (MHz to THz) frequencies.³⁹⁻⁴¹

From the same $\lambda(T, p)$ data set, one can extract one more precious piece of information, the scaling of T_c with the zero-temperature superfluid density $N_{s0} \equiv N_s(T \rightarrow 0)$. We have found this dependence to be smooth and essentially linear, but with a finite offset, except close to the origin where it turns to a square-root dependence.³⁰ This is incompatible with the BCS theory, which fails spectacularly both quantitatively and qualitatively. Here, we are referring not only to the standard (‘clean’) BCS description, but also to its various extensions that take into account the d-wave symmetry of the order parameter, as well as the effects of disorder, impurities, pair breaking, etc. While there is no space here to present the detailed arguments and evidence, let us just remark that no BCS-based approach that we have tried or seen so far is compatible with our entire data set, and that a dirty-BCS picture can also be ruled out conclusively.³⁴

To summarize this section, we have established that the superconducting state in cuprates is very strange, and non-BCS like. In BCS theory, T_c is essentially controlled by the phonon frequency, electron density of states near the Fermi level, and the strength of electron-phonon interaction. None of these matter here; T_c is determined by the kinematics (the super-electron density) rather than dynamics. This, of course, resembles more what one would expect from local pairing and the BEC scenario. In fact, it has been shown by Z. Radovic and M. Vanevic (unpublished) that both the temperature and the doping dependence of N_s can be reproduced quantitatively using a simple quasi-2D Bose gas model.

5. Additional evidence for preformed local pairs

Various other independent experimental results also point to formation of local pairs in cuprates. Here, we will only mention two that we were involved with. One of this was electrolyte gating²⁸ of ultrathin LSCO films. By varying the gate voltage V_G , we tuned the carrier density in very fine steps across the superconductor-to-insulator transition (SIT) that occurs at the critical doping $x_c \approx 0.06$, see Figure 3. As shown in the inset, all the $R(T, V_G)$ curves can be scaled very well to a universal function of the new coordinate $u \propto |x - x_c| T^{2/3}$, indicative of 2D quantum critical behavior. Notably, the SIT occurs when the film sheet resistance reaches the quantum resistance for pairs, $R_Q = h/(2e)^2 = 6.5 \text{ k}\Omega$. (Here, h is the Planck constant and e the electron charge.) This is consistent with the existence of pairs on both sides of the SIT, mobile in the superconducting state and localized in the insulating state. Similar findings were also reported in thin films and bulk crystals of other cuprates.⁴²⁻⁴⁴

Another piece of evidence came from THz spectroscopy experiments on films of LSCO, which showed clear signs of superconducting (phase) fluctuations⁴¹ well above T_c (at least 20-25 K) at all doping levels, see Figure 4. This corroborated similar earlier findings in $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ (BSCCO) films.⁴⁰ The same conclusion was reached by a number of other researchers, who studied different cuprate compounds and used different measurement techniques.⁴⁵⁻⁵¹

6. A paradox and a possible resolution: a two-fluid picture

The problem with the above conclusions about local pairing and BEC-like behavior is that they collide with equally convincing evidence about the existence of the Fermi surface compounded by ARPES, STM, and quantum oscillations measurements. Of course, one possibility is that simply something is wrong, but at this point it is not obvious what. True, ARPES and STM are surface sensitive, but essentially the same Fermi surfaces are inferred from quantum oscillation experiments that probe the bulk. The (extremely) high magnetic fields used in quantum oscillation experiments could affect the electronic states and even generate new features that are absent in the ground state, but ARPES and STM measurements are performed in zero field. As for the demise of superfluid density with overdoping, it has been observed independently in LSCO by the same (mutual inductance) technique by the group of T. Lemberger,^{52,53} as well as by muon spin rotation measurements on heavily overdoped Tl-2201 bulk crystals, by two different groups.^{54,55} In the later case, N_{s0} can be two orders of magnitude smaller than the carrier density, while at the same time spectacularly sharp quantum oscillations show without doubt that the electron mean-free path is very long (on the scale of 1,000 Å or larger) and the crystals are in the ultra-clean limit.^{20,21} The later, by the way, leaves very little space for speculations about phase separation (electronic or otherwise), disorder, pair-breaking, etc.

One possible way out of the paradox is to assume that we are dealing with something akin to a two-fluid model, a fermion-boson mixture, where fermionic and bosonic excitations coexist, in the normal state. This separation could even occur in k -space, with preformed pairs originating from the states near the anti-nodes, while the nodal quasi-particles remain unpaired and fermionic. The boson-fermion ratio should vary with doping, with the bosonic fraction progressively decreasing. Assuming that superconductivity originates from the bosonic fraction, this could explain why both T_c and N_s decrease with doping, and at the same time, why the normal state increasingly looks like an ordinary Fermi Liquid.

Needless to say, this is just a vague suggestion that must first be put on solid theoretical grounds, a task that is beyond the present paper. Here, we just wish to bring up one more piece of experimental evidence. It has been shown by two independent techniques that the doping dependence of the chemical potential μ in LSCO is very strange and unlike what would be expected from a simple band structure model. From undoped to optimally doped LSCO, μ stays almost constant, as demonstrated directly (and reproduced thousand-fold) by the absence of electron transfer and redistribution across the LSCO/LCO interface. On the overdoped side, however, μ drops fast and in a linear manner with doping, exactly as one would expect from a quasi-2D metal with a cylindrical Fermi surface, see Figure 5. Such a strange ‘broken’ $\mu(p)$ dependence is indeed indicative of formation of some more-or-less localized mid-gap states at early stages of doping, and their concomitant broadening into a wide electron band as the doping level is increased further. It is worth noting that this is exactly what has been predicted by the Dynamic Mean-Field Theory (DMFT) studies of the Mott-Hubbard model and its evolution with doping.⁵⁶

Another possibly related experimental fact is that in LSCO we have observed a spontaneous breaking of the rotational symmetry in the mobile electron fluid, by angle-resolved transverse resistivity measurements.³⁵ By other techniques, this ‘electronic nematicity’ has been observed in other cuprates as well as in Fe-based superconductors. New here is that we observe it in a very large portion of the (T, p) phase diagram, up to the room temperature for all but the highest doping levels. This, of course, is quite unusual; since our LSCO films are tetragonal their resistivity should be isotropic in-plane. Notably also, the amplitude of nematicity — the relative degree of orthorhombic electronic anisotropy in a tetragonal crystal — decreases with doping, in line with the decrease of T_c , N_{s0} , $\mu(p)$, and the Marginal Fermi Liquid features such as the T -linear component of resistivity.

7. Summary and outlook

To wrap this up, we have succeeded in identifying some surprising and unique features of both the normal-metal and superconducting states in LSCO. Hence, in this prototypical HTS cuprate, a strange, non-BCS-like superconducting state develops upon cooling from an equally strange metallic state that is not a simple Fermi Liquid but a Fermi Liquid Crystal. This calls for in-depth theoretical studies, possibly involving new models; it is likely imperative to also take into account fluctuations properly. Experimentally, our immediate next goal is to check how universal the above findings are for various HTS cuprates as well as for Fe-based superconductors. Then, armed with these important new insights, and with our arsenal of unique synthesis techniques and characterization probes, we plan to embark on an enlightened search for new HTS materials. Obviously, we will be looking for materials in which we expect formation of local pairs at a sufficiently high density and sufficiently mobile. If such a material could be found, or engineered at the atomic level — perhaps as a metastable, or an epitaxially stabilized compound — it is not obvious to us why one should not be able to increase T_c beyond the current record. Time will tell.

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$10 \times \text{La}_{1.60}\text{Sr}_{0.40}\text{CuO}_4$
$\text{La}_{1.65}\text{Sr}_{0.35}\text{Cu}_{0.97}\text{Zn}_{0.03}\text{O}_4$
$\text{La}_{1.70}\text{Sr}_{0.30}\text{Cu}_{0.97}\text{Zn}_{0.03}\text{O}_4$
$10 \times \text{La}_{1.75}\text{Sr}_{0.25}\text{CuO}_4$
$\text{La}_{1.70}\text{Sr}_{0.30}\text{Cu}_{0.97}\text{Zn}_{0.03}\text{O}_4$
$\text{La}_{1.65}\text{Sr}_{0.35}\text{Cu}_{0.97}\text{Zn}_{0.03}\text{O}_4$
$10 \times \text{La}_{1.60}\text{Sr}_{0.40}\text{CuO}_4$
LSAO SUBSTRATE

Figure 1. Schematics of atomic-layer sample engineering to isolate a HTS layer of a known thickness.³⁴

Figure 2. Left panel: Temperature and doping dependence of the superfluid density N_s as a function of temperature and doping, in heavily overdoped LSCO films.³⁴ Right panel: the doping dependence of $N_{s0} \equiv N_s(T \rightarrow 0)$. Blue diamonds: our experimental data. Red line: the dependence expected from the BCS theory.²³

Figure 3. Electrolyte gating of a one unit cell thick LSCO layer.²⁸ Inset: The same data rescaled against $u = |x - x_c| / T^{2/3}$.

Figure 4. Superconducting fluctuations in LSCO seen well above T_c in THz spectroscopy.⁴¹

Figure 5. A strange ‘broken’ doping dependence of the chemical potential in LSCO.^{25,31}