BNL-112450-2016-JA

**On Local Pairs vs. BCS: Quo
Vadis High-Tc
Superconductivity?**

D. Pavuna

Submitted to Journal of Superconductivity and Novel Magnetism.

August 2016

Condensed Matter Physics and Material Science Department

Brookhaven National Laboratory

**U.S. Department of Energy USDOE
Office of Science (SC),
Basic Energy Sciences (BES) (SC-22)**

Notice: This manuscript has been authored by employees of Brookhaven Science Associates, LLC under Contract No. DE-SC0012704 with the U.S. Department of Energy. The publisher by accepting the manuscript for publication acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes.

DISCLAIMER

This report was prepared as an account of work sponsored by an agency of the United States Government. Neither the United States Government nor any agency thereof, nor any of their employees, nor any of their contractors, subcontractors, or their employees, makes any warranty, express or implied, or assumes any legal liability or responsibility for the accuracy, completeness, or any third party's use or the results of such use of any information, apparatus, product, or process disclosed, or represents that its use would not infringe privately owned rights. Reference herein to any specific commercial product, process, or service by trade name, trademark, manufacturer, or otherwise, does not necessarily constitute or imply its endorsement, recommendation, or favoring by the United States Government or any agency thereof or its contractors or subcontractors. The views and opinions of authors expressed herein do not necessarily state or reflect those of the United States Government or any agency thereof.

On Local Pairs vs. BCS: Quo Vadis High- T_c Superconductivity?

D. Pavuna¹, G. Dubuis^{1,2}, A.T. Bollinger³, J. Wu³, X. He⁴ and I. Božović^{3,4}

¹Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland

²MacDiarmid Institute for Advanced Materials & Nanotechnology, Robinson Research Institute, Victoria University of Wellington, Lower Hutt 5046, New Zealand

³Brookhaven National Laboratory, Upton, NY 11973, USA;

⁴Yale University, New Haven, CT 06520, USA

Corresponding Author:

Ivan Božović

Phone: (631) 344-4973

Fax: (631) 344-4071

E-mail:: bozovic@bnl.gov

Keywords (4-6)

High temperature superconductivity, ionic liquid, quantum critical point, cuprate, thin films,

Abstract

Since the discovery of high-temperature superconductivity in cuprates, proposals have been made that pairing may be ‘local’, in particular in underdoped samples. Here we briefly review evidence for local pairs from our experiments on thin films of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$, synthesized by atomic-layer-by-layer molecular beam epitaxy (ALL-MBE).

In this short letter we briefly discuss the current situation in and future prospects for high- T_c superconductivity (HTS). Over a century after the discovery of superconductivity and 30 years after the discovery of HTS in cuprates, our understanding is still incomplete and patchy. Bardeen-Cooper-Schrieffer (BCS) theory has worked well for the low- T_c ($< 30\text{-}40\text{ K}$) superconductors^{1,2}, but it is generally considered inadequate to describe HTS in cuprates³⁻¹¹. While it is still under intense debate, the physics of cuprates is widely believed to be ‘exotic’. The same is true of another, more recent HTS family of iron-based superconductors (pnictides, etc.) although in this case ‘exotic’ may have a different meaning. To make the matters more perplexing, the most recent, and record-holding ($T_c > 200\text{ K}$), entry to the HTS realm, sulfur hydride, has been claimed by numerous authors to be a conventional BCS superconductors. This makes for a very weird HTS road map¹², Figure 1. This situation is far from satisfactory — one would rather hope that the phenomenon of superconductivity could be described by a single, unified theoretical framework.

Within the BCS theory, one expects that as the pairing interaction gets stronger and the superconducting gap and T_c increase, the coherence length and the Cooper pair size should get smaller. Eventually, when the pair size becomes comparable to, or smaller than, the distance between pairs, so that they do not overlap anymore, the BCS picture becomes inadequate and the situation may be more resembling of Bose-Einstein condensation (BEC) of ‘local’ pairs. Indeed, it has been demonstrated in experiments with cold-atom gases¹³⁻¹⁵ that the transition from BCS to BEC can be triggered by either increasing the strength of the pairing interaction, or by making the atom clouds thinner, i.e., by reducing the dimensionality from 3D to 2D. The transition from BCS to BEC seems to be smooth and gradual, as predicted in the theories of BCS-BEC crossover that were put forward by A. Leggett, P. Nozieres, C. Levin, M. Randeria, and others.

In view of this, it is not surprising that numerous physicists argued that pairing is in fact local in cuprates. While their proposed explanations vary in many other (and sometime quite important) details, this general idea of local pairs has been espoused by K. A. Mueller, P. W. Anderson, N. Mott, T. D. Lee, P.-G. De Gennes, G. Deutscher, A. F. Andreev, J. Ranninger, Y. J. Uemura, D. Mihailovic, T. Egami, A. Alexandrov, D. Emin, T. Schneider, A. Bussman-Holder, H. Keller, P. P. Edwards, J. A. Wilson, A. Auerbach, Wei Ku, and others — to many to cite here.

Here, we review briefly our own experimental results that point to local pairs in cuprates. One of this was a study of ultrathin films of $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO), synthesized by atomic-layer-by-layer molecular beam epitaxy (ALL-MBE) at Brookhaven, and exposed to electrolyte gating¹⁶. We are able to traverse the superconductor-to-insulator transition (SIT) at the critical doping $x_c \approx 0.06$, in very fine steps, by tuning the gate voltage V_G , see Figure 2. All the $R(T, V_G)$ curves can be scaled perfectly to a single (albeit double-valued) function of the compound coordinate $u \equiv |x - x_c| T^{2/3}$, as predicted for quantum critical scaling in 2D. However, the most important finding was that SIT occurred at the critical sheet resistance quite close to the quantum pair resistance $R_Q = h/(2e)^2 = 6.5 \text{ k}\Omega$, where h is the Planck constant and e the electron charge. This indicates that pairs exist on both side of SIT; they are mobile, coherent, and condensed in the superconductor while they are local(ized) on the insulator side. The same finding was reported¹⁷ almost simultaneously and using the same electrolyte-gating technique, by the group of Allen Goldman, notably in a different cuprate ($\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$). In fact, SIT occurs at the same critical sheet resistance close to $6.5 \text{ k}\Omega$ even in bulk LSCO and $\text{Bi}_2\text{Sr}_2\text{Y}_x\text{Ca}_{1-x}\text{Cu}_2\text{O}_8$ crystals^{18,19}.

The next result we wish to recollect here is that THz spectroscopy experiments on ALL-MBE grown films of $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_8$ and LSCO clearly revealed superconducting (phase) fluctuations up to about 20-25 K above T_c , (see Figures 2a and 2b, respectively), everywhere in the phase diagram^{20,21}. This indeed implies that pairs are ‘preformed’ well above T_c . Several other groups, using an array of different techniques and studying different cuprate compounds, arrived at the same conclusion²²⁻³⁰.

Finally, quite recently the results have been released³¹ of very comprehensive and accurate measurements of the magnetic penetration depth λ in high-quality LSCO films synthesized by ALL-MBE (see the accompanying article by Bozovic *et al.* in this volume). The key result is that T_c scales with, and is controlled by, the zero-temperature phase stiffness ρ_{s0} (or, equivalently, the superfluid density). This is incompatible with the conventional BCS picture, and again points to local pairing.

Getting back to our initial question, while it seems that HTS in cuprates indeed involves local pairs, one wonders what happens with other HTS families – pnictides, FeSe/STO , and H_3S . This calls for some systematic and incisive experiments designed to directly address this question.

References

1. Cyrot, M.; Pavuna, D.: Introduction to superconductivity and high- T_c materials (World Scientific, Singapore, 1992).
2. Cooper, L.N.; Feldman, D., editors: BCS: 50 Years (World Scientific, Singapore, 2011).
3. Phillips, J. C.: Physics of high- T_c superconductors (Academic Press, London, 1989).
4. Wilczek, F.: Fractional statistics and anyon superconductivity (World Scientific, Singapore, 1990).
5. Plakida, N. M. High temperature superconductivity: experiment and theory (Springer, Berlin, Germany, 1995).
6. Anderson, P. W.: The theory of superconductivity in the high- T_c cuprates (Princeton University Press, Princeton, New Jersey, 1997).
7. Manske, D.: Theory of unconventional superconductors. Cooper-pairing mediated by spin excitations (Springer Tracts in Modern Phys. Vol. 202; Springer, Berlin, Germany, 2004).
8. Zaanen, J.: Superconductivity - Why the temperature is high. *Nature* **430**, 512-513 (2004).
9. Lee, P. A., Nagaosa, N. & Wen, X. G.: Doping a Mott insulator: Physics of high-temperature superconductivity. *Rev. Mod. Phys.* **78**, 17-85 (2006).
10. Zaanen, J., Chakravarty, S., Senthil, T., Anderson, P. W., Lee, P., Schmalian, J., Imada, M., Pines, D., Randeria, M., Varma, C., Vojta, M. & Rice, M.: Towards a complete theory of high T_c . *Nature Phys.* **2**, 138-143 (2006).
11. Keimer, B., Kivelson, S. A., Norman, M. R., Uchida, S. & Zaanen, J.: From quantum matter to high-temperature superconductivity in copper oxides. *Nature* **518**, 179-186 (2015).
12. Božović, I.: High-temperature superconductivity: a conventional conundrum. *Nature Physics* **12**, 22-24 (2016).
13. Greiner, M., Regal, C. A., Jin, D. S.: Emergence of a molecular Bose-Einstein condensate from a Fermi gas. *Nature* **426**, 537-540 (2003).
14. Ries, M. G., Wenz, A. N., Zürn, G., Bayha, L., Boettcher, I., Kedar, D., Murthy, P. A., Neidig, M., Lompe, T., Jochim, S.: Observation of Pair Condensation in the Quasi-2D BEC-BCS Crossover. *Phys. Rev. Lett.* **114**, 230401 (2015).
15. Sommer, A. T., Cheuk, L. W., Ku, M. J. H., Bakr, W. S., Zwierlein, M. W.: Evolution of Fermion Pairing from Three to Two Dimensions. *Phys. Rev. Lett.* **108**, 045302 (2012).
16. Bollinger, A. T. *et al.*: Superconductor-insulator transition in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ at the pair quantum resistance. *Nature* **472**, 458-460 (2011).
17. Leng, X., Garcia-Barriocanal, J., Bose, S., Lee, Y. & Goldman, A. M.: Electrostatic control of the evolution from a superconducting phase to an insulating phase in ultrathin $\text{YBa}_2\text{Cu}_3\text{O}_{7-x}$ films. *Phys. Rev. Lett.* **107**, 027001 (2011). Erratum *Phys. Rev. Lett.* **107**, 039901 (2011).
18. Mandrus, D., Forro, L., Kendziora, C., Mihaly, L.: Two-dimensional electron localization in bulk single crystals of $\text{Bi}_2\text{Sr}_2\text{Y}_x\text{Ca}_{1-x}\text{Cu}_2\text{O}_8$. *Phys. Rev. B* **44**, 2418(R) (1991).
19. Semba, K. and Matsuda, A.: Superconductor-to-Insulator Transition and Transport Properties of Underdoped $\text{YBa}_2\text{Cu}_3\text{O}_y$ Crystals. *Phys. Rev. Lett.* **86**, 496 (2001).
20. Corson, J., Mallozzi, R., Orenstein, J., Eckstein, J. N. & Bozovic, I.: Vanishing of phase coherence in underdoped $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$. *Nature* **398**, 221-223 (1999).
21. Bilbro, L. S. *et al.*: Temporal correlations of superconductivity above the transition temperature in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ probed by terahertz spectroscopy. *Nat. Phys.* **7**, 298-302 (2011).
22. Madan, I. *et al.*: Separating pairing from quantum phase coherence dynamics above the superconducting transition by femtosecond spectroscopy. *Sci. Rep.* **4**, 5656 (2014).

23. Grbić, M. S. *et al.*: Temperature range of superconducting fluctuations above T_c in $\text{YBa}_2\text{Cu}_3\text{O}_{7.8}$ single crystals. *Phys. Rev. B* **83**, 144508 (2011).
24. Emery, V. J., Kivelson, S. A.: Importance of phase fluctuations in superconductors with small superfluid density. *Nature* **374**, 434-437 (1995).
25. Uykur, E., Tanaka, K., Masui, T., Miyasaka, S., Tajima, S.: Persistence of the superconducting condensate far above the critical temperature of $\text{YBa}_2(\text{Cu,Zn})_3\text{O}_y$ revealed by c-axis optical conductivity measurements for several Zn concentrations and carrier doping levels. *Phys. Rev. Lett.* **112**, 127003 (2014).
26. Xu, Z. A., Ong, N. P., Wang, Y., Kakeshita, T., Uchida, S.: Vortex-like excitations and the onset of superconducting phase fluctuation in underdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$. *Nature* **406**, 486-488 (2000).
27. Wang, Y. Y. *et al.*: Onset of the vortex-like Nernst signal above T_c in $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ and $\text{Bi}_2\text{Sr}_{2-y}\text{LaCuO}_6$. *Phys. Rev. B* **64**, 224519 (2001).
28. Wang, Y., Li, L., Ong, N. P.: Nernst effect in high- T_c superconductors. *Phys. Rev. B* **73**, 024510 (2006).
29. Li, L. *et al.*: Diamagnetism and Cooper pairing above T_c in cuprates. *Phys. Rev. B* **81**, 054510 (2010).
30. Kondo, T. *et al.*: Point nodes persisting far beyond T_c in Bi2212 . *Nature Commun.* **6**, 7699 (2015).
31. Božović, I., He, X., Wu, J., Bollinger, A. T.: Dependence of critical temperature in overdoped copper oxides on superfluid density. *Nature* (2016) in press.

Acknowledgements. The experimental work was done at BNL and was supported by the U.S. Department of Energy, Basic Energy Sciences, Materials Sciences and Engineering Division. X.H. is supported by the Gordon and Betty Moore Foundation's [EPiQS Initiative](#) through Grant [GBMF4410](#).

Fig. 1: A cartoon of the ‘Road-map to room-temperature superconductivity’ (after Ref. 12). This (joke) was an attempt to expose how the current picture of HTS is inconsistent and unsatisfactory.

Figure 2. (a) The dependence of the sheet resistance, normalized to the quantum pair resistance $R_Q = h/(2e)^2 = 6.5 \text{ k}\Omega$, on temperature, for various values of the gate voltage V_G , in an ultrathin LSCO film grown by ALL-MBE (after Ref. 16). (b) The same data, re-plot as a function of the compound scaling coordinate $u \equiv |x - x_c| T^{2/3}$, show perfect 2D quantum critical scaling.

Figure 3: (a) THz spectroscopy data measured on two underdoping $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$ films (with $T_c = 33$ K and $T_c = 74$ K, respectively) grown by ALL-MBE (after Ref. 20). Superconducting phase fluctuations are clearly seen at least 20 K above T_c . (b) The summary of the results of similar THz experiments on a series of LSCO grown by ALL-MBE (after Ref. 21). Again, superconducting fluctuations are clearly seen about 20 K (the broad amber line) above T_c (solid black squares) everywhere in the phase diagram. The blue triangles show the onset of diamagnetic signal in LSCO single crystals (after Ref. 29), at even higher temperature.