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The demise of superfluid density in overdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ films grown by molecular beam epitaxy

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Abstract

We have synthesized $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ thin films using atomic-layer-by-layer molecular beam epitaxy (ALL-MBE). The films are high-quality — single crystal, atomically smooth, and very homogeneous. The critical temperature (T_c) shows very little (< 1 K) variation within a film of $10 \times 10 \text{ mm}^2$ area. The large statistics (over 2,000 films) is crucial to discern intrinsic properties. In these films, we have measured the absolute value of the magnetic penetration depth λ with the accuracy better than 1%, and mapped densely the entire overdoped side of the $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ phase diagram. A new scaling law is established accurately for the dependence of T_c on the superfluid density. The scaling we observe is incompatible with the standard Bardeen-Cooper-Schrieffer picture, and points to local pairing.

Introduction

Underdoped cuprate superconductors have been studied in great detail.¹⁻³ The overdoped side has received much less attention. It seems to be almost generally believed that with overdoping the physics evolves towards the conventional one, where the normal state can be well described by the Landau Fermi liquid theory and the superconducting state by the Bardeen-Cooper-Schrieffer (BCS) theory. In variance with this simple picture, several early studies⁴⁻⁹ on a few overdoped samples showed that the measured superfluid density was much smaller than what one would expect in a clean BCS superconductor. However, this discrepancy was attributed to extrinsic factors such as phase separation⁴ or pair-breaking⁹, and largely brushed away.

Experimental methods

We have studied this question in great detail by measuring the dependence of the magnetic penetration depth λ on temperature and doping in overdoped $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) films. For film synthesis, we used a unique atomic-layer-by-layer molecular beam epitaxy (ALL-MBE) system at Brookhaven National Laboratory that provides single-crystal thin films free of usual defects such as secondary-phase precipitates, grain boundaries, pinholes, etc¹⁰⁻¹². The total number of LSCO films we have studied, in ten years, is well over 2,000, and this large statistics is critically important since it allows us to clearly identify the trends and systematics, and thus to differentiate between intrinsic behavior and the effects of various extrinsic factors. The sample homogeneity can be judged by the sharpness of the superconducting transition indicated by the onset of the Meissner effect. In our best films, T_c is homogeneous on the scale of ~ 0.1 K, over the film area of 10×10 mm².

We have measured $\lambda(T, x)$ using the two-coil mutual inductance technique¹³⁻¹⁹. To improve the accuracy, we have made a number of technical improvements in the apparatus and the procedure²⁰, ensuring great reproducibility and precision (better than 0.3 %) in the raw data. To extract from these the absolute values of λ , a critical requirement is that the thickness of the superconducting layer must be accurately known. To meet this goal, we have used ALL-MBE to engineer the samples at the atomic-layer level (including δ -doping with Zn), synthesizing multilayer structures in which the active (superconducting) layer thickness is sharply defined. Altogether, we measure the absolute value of λ with the accuracy of $\pm 1\%$.

Experimental results

In Fig. 1, we show the doping dependence of $\lambda(T)$ covering densely the entire overdoped side of the LSCO phase diagram. These data are extracted directly from the measured raw inductance data; the only sample-dependent input parameter is the film thickness, which we know accurately by virtue of our atomic-layer-by-layer (‘digital’) synthesis technique.

For few most heavily overdoped films, in Fig. 2 we show the corresponding 2D superfluid density N_s , expressed as the number of holes per one CuO_2 unit. It is determined as $N_s \equiv A/\lambda^2$, where $A \equiv m^* a_0^2 d / \mu_0 e^2$, m^* is the effective mass, $a_0 = 0.38$ nm is the in-plane lattice constant, $d = 0.662$ nm is the distance between two neighbor CuO_2 planes, $\mu_0 = 4\pi \times 10^{-7}$ N/A² is the vacuum permeability, and e is the electron charge. In this plot, we have assumed for simplicity that $m^* = 2m_e$, independent on doping; this is the largest value compatible with our data at optimal doping, and it gives $A = 5.14 \times 10^{-15}$ m². Note that this choice is made just to facilitate communication and comparison with the sample doping level, which is usually also expressed as a number of holes per Cu. We can equally well formulate all of our conclusions²⁰ without any assumptions, in terms of the superfluid phase stiffness, defined as $\rho_s \equiv (4.15 \times 10^{-12} \text{ K-m}^2) / \lambda^2$.

As seen from Fig. 2, the $N_s(T)$ curves are essentially linear, apart from some small deviations from linearity at very low T and/or also very close to T_c . This is indeed expected theoretically²¹ and observed experimentally^{22,23} in very clean d-wave superconductors. However, the absolute values of $N_{s0} \equiv N_s(T \rightarrow 0)$ are unexpectedly extremely small, as low as 1% of the total mobile carrier density (if we assume that the later is equal to p , and even less if we equate it with $1+p$, as frequently postulated). Since N_{s0} cannot be measured directly, but only extrapolated from the values measured down to the experimental limit, there is some uncertainty due to this extrapolation. However, note that our measurements extend down to $T = 300$ mK, so this error is generally negligible. This is illustrated in the inset to Figure 2, for two samples, one very highly overdoped with a low $T_c \approx 5$ K, and the other much closer to the optimal doping, with $T_c \approx 36$ K. (The corresponding doping levels, $p = 0.255$ and $p = 0.195$, are only nominal, and used just for communication – p is not a directly measured quantity²⁰.) Both are fitted to $N_s(T) = N_{s0} - AT - BT^2$, and the fits are essentially perfect ($R = 0.99981$ and $R = 0.99999$, respectively). More to the point, the difference between the extrapolated N_{s0} and the closest measured value $N_s(T=300 \text{ mK})$ is

very small to begin with at any doping ($\Delta N_s = 0.0002$ for $p = 0.255$ and $\Delta N_s = 0.0005$ for $p = 0.195$ film, respectively). The differences between the values extrapolated using different functional forms (e.g., strictly linear or strictly quadratic) are even smaller, on the order of 1% in the worst case (the lowest T_c) and much less elsewhere.

Fig. 3 shows the dependence of T_c on N_{s0} . For the most part, the $T_c(N_{s0})$ scaling in Fig. 3 is linear but with an offset, $T_c = T_0 + \alpha N_{s0}$, with $T_0 = (7.0 \pm 0.1)$ K and $\alpha = (2.5 \pm 0.1) \times 10^2$ K. This scaling breaks down very close to the origin, i.e., for $N_{s0} < 0.02$ or so; there, the curve can be fitted well to $T_c = \gamma \sqrt{N_{s0}}$ with $\gamma = (1.1 \pm 0.1) \times 10^2$ K. Note that in this figure the error bars are smaller than the marker size.

Conclusions

This accurate scaling law imposes a very tight constraint on the theory. We have gathered and presented²⁰ ample evidence that it cannot be explained by assuming impurities and disorder within the standard dirty-d-wave-BCS picture. The precise form of this $T_c(N_{s0})$ scaling is novel and surprising, and it challenges the prevalent creeds^{24,25}. An important inference is that T_c appears to be essentially controlled by the superfluid density, i.e., by the kinematics. This is more consistent with local pairing rather than with the BCS picture, in which T_c is determined by dynamics of electrons and phonons, and their interaction. Note, however, that for any single-species system that is translation- and time reversal-invariant, the superfluid density must be equal to the total particle density, as shown by A. Leggett^{26,27}. The fact that this law is so dramatically violated in samples that are evidently very homogeneous and clean poses a serious problem to the theory. We may need to rethink some of our basic premises.

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Fig. 1. The dependence of the magnetic penetration depth (λ) on temperature (T) and doping level (p) in overdoped LSCO films synthesized by ALL-MBE. Note that p is not directly measured but rather (as it is customary in this field) inferred from the measured value of T_c assuming a phenomenological parabolic $T_c(p)$ dependence²⁰.

Fig. 2. The temperature and doping dependence of the in-plane superfluid density $N_s \equiv A/\lambda^2$, where $A = 5.4 \times 10^{-15} \text{ m}^2$, for ten heavily overdoped LSCO films grown by ALL-MBE. Note that it in the most overdoped film N_{s0} has dropped to 1% of the total mobile carrier (hole) density. The inset shows the fits to $N_s(T) = N_{s0} - AT - BT^2$, for two representative but widely different doping levels.

Fig. 3. Solid blue diamonds: the experimental data showing the dependence of T_c on zero-temperature superfluid density $N_{s0} \equiv N_s(T \rightarrow 0)$. The green dashed line: fit to $T_c = T_0 + \alpha N_{s0}$, where $T_0 = (7 \pm 0.1)$ K, and $\alpha = (2.5 \pm 0.1) \times 10^2$ K, for $N_{s0} > 0.02$. The red dashed line: fit to $T_c = \gamma \sqrt{N_{s0}}$, with $\gamma = (1.1 \pm 0.1) \times 10^2$ K, for $N_{s0} < 0.02$.