(Ir)Relevance of Disorder for Superconductivity in Cuprates

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In cuprates that exhibit high-temperature superconductivity (HTS), as the doping level p is increased beyond the optimal value, the superfluid density n_{s0} decreases and eventually vanishes, closely tracking the critical temperature T_c. This has been interpreted using a dirty-d-wave extension of the Bardeen-Cooper-Schrieffer theory, assuming that T_c and n_{s0} decrease with increasing disorder and pair breaking. To test this hypothesis, we tuned the doping level in overdoped $La_{2-x}Sr_xCuO_{4-\delta}$ films by electrolyte gating, measured several parameters used to quantify the level of disorder (the residual resistivity ratio RRR, the mean free path l_0, and the electron mobility μ), and studied how T_c and n_{s0} scale with these. The experimentally measured dependence turned out to be the opposite of the expected—T_c and n_{s0} increased with increasing disorder. This brings the question of the true origin of the demise of T_c and n_{s0} with overdoping back to the center stage of the quest to decipher the HTS enigma.

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Understanding high-temperature superconductivity (HTS) in cuprates is still one of the central open problems in condensed matter physics. The unconventional and anomalous nature of both the superconducting and normal states has been unearthed through inventive and meticulous experimentation, for which a unified and comprehensive theoretical explanation still needs to be provided. One striking observation was that n_{s0}, the superfluid density extrapolated to $T \to 0$, decreases and vanishes as the doping level (the mobile carrier density per Cu atom) p is increased, closely tracking the critical temperature, T_c [1–6]. Previously, the prevailing view was that on the overdoped side of the superconducting dome, T_c decreases because of the weakening of the pairing interaction. In this case, the superconducting state should be well described by the weak-coupling Bardeen-Cooper-Schrieffer (BCS) theory. If so, as asserted by Legget's Theorem [7,8], at $T \to 0$, all the electrons should be in the superfluid. Hence, n_{s0} should continue to increase as the doping level p increases. However, the experiments showed the opposite [1–6].

To resolve this discrepancy, several theoretical models were proposed [9–14]. While they differ in detail, the common assumption is that, with increased dopant ion concentration, disorder increases, which causes pair breaking and depletes n_{s0}. In the present Letter, we put that hypothesis to a direct experimental test. In a representative HTS cuprate, we varied the doping level in fine steps by electrolyte gating. We studied the dependence of T_c and n_{s0} on several measurable quantities that can be used to quantify the effects of disorder on electron transport—the electron mobility μ, the mean-free path l_0, and the residual resistivity ratio RRR. We found that the observed dependence is precisely the opposite of what is expected from the dirty-d-wave-BCS scenario.

The main technical difficulty with such experiments is that different cuprate samples with nominally identical stoichiometry and chemical doping levels may have very different values of μ, l_0, and RRR. The underlying causes are variations in chemical composition (impurities, vacancies, or deviations from stoichiometry) and crystal-structure defects (intersite substitutions, interstitials, dislocations, etc.). In particular, p is strongly affected by the oxygen content. In HTS cuprates, oxygen is volatile at elevated temperatures (i.e., at typical synthesis conditions), and it can enter or exit the sample easily and diffusely. Depending on the external oxygen partial pressure, oxygen vacancies can be abundant, or extra oxygen ions can be intercalated into interstitial positions, affecting the total mobile hole count [15]. This disorder is hard to control, particularly in slowly cooled bulk crystals, which inevitably leads to a frozen-in gradient of oxygen composition from the surface inward. This complicates a systematic study of the interplay between T_c, n_{s0}, and disorder.

To alleviate this problem, in the present study, we used ultrathin (5-unit-cell-thick) films of the prototypical cuprate, $La_{2-x}Sr_xCuO_{4-\delta}$ (LSCO). The films were synthesized by atomic-layer-by-layer molecular beam epitaxy (ALL MBE) [6]. This technique has been amply proven to produce atomically smooth films of high crystallinity, with

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RRR exceeding 200 in some cases [16]—much higher than in bulk crystals. Here, we focused on the overdoped LSCO. To modify the doping level, we used electrolyte gating [17,18]. We varied T_c and n_{s0} in a *single* LSCO sample just by changing p. At the same time, the measured μ, l_0, and RRR varied because the defect screening improves with increased p. The samples were kept at relatively low temperatures, i.e., cycled between 300 K and 4 K, so one does not expect significant changes in the Sr^{2+} dopant concentration or the film crystallinity. At low-to-moderate gate voltages ($V_{\rm gate}$), the oxygen content is not changed, either. This greatly reduces the uncertainties due to uncontrolled sample-to-sample variations in oxygen vacancy concentration and distribution, crystalline defects, etc., allowing for unambiguous conclusions.

Our main finding is that as the doping decreases, μ, l_0, and RRR decrease, while T_c and n_{s0} increase. In other words, T_c and n_{s0} increase with increased disorder, so the cause of the demise of T_c and n_{s0} with overdoping must be something else. Unraveling the true origin of this phenomenon remains a central open question; answering it may help crack the HTS enigma.

In Fig. 1(a), we show the temperature dependence of the resistance R measured at different values of the applied gate voltage, $V_{\rm gate}$, in an LSCO film with Sr doping $x = 0.28$. For negative $V_{\rm gate}$, the gating effect is minimal. In contrast, a substantial increase in T_c (by more than a factor of 2) and R is observed for positive $V_{\rm gate}$. (This asymmetry is expected from our understanding of the effects of electrolyte gating [17,18].) These $R(T)$ curves are reproducible and reversible; there is no observable hysteresis if the gate voltage is kept within the range $-2.25\ \mathrm{V} \gtrsim V_{\rm gate} \gtrsim +1.25$ V. To quantify the effect of disorder on transport, we use the residual resistance ratio, $RRR = R_{\rm RT}/R_0$, where $R_{\rm RT} = R(T = 295\ \mathrm{K})$ is the resistance at room temperature and $R_0 = R(T \to 0)$ is the resistance extrapolated to zero temperature by fitting the $R(T)$ curves above the superconducting transition to a standard parabolic form $R(T) = R_0 + \mathrm{A}T + \mathrm{B}T^2$. (Very similar RRR values are obtained if we fit by a straight line, or by the parallel conductance model.) The RRR is widely used in condensed matter physics as a figure of merit to compare and quantify the quality of metallic samples in terms of order and homogeneity; a larger RRR usually indicates a sample with fewer impurities and less disorder [19]. In Fig. 1(b), we show the relation between T_c and RRR in the same LSCO$(x = 0.28)$ film. The superconducting transitions in LSCO and other cuprates are relatively wide because of some extrinsic disorder and/or the intrinsic broadening due to superconducting fluctuations, and hence T_c is not sharply defined. Various proxies for T_c were used in the literature, e.g., the temperature at which $R(T)$ drops to 90%, 50%, or 10% of the normal-state value R_n just above the transition, but the choice of R_n is subjective. Here, we determine T_c as the temperature at which $\mathrm{d}R(T)/\mathrm{d}T$ reaches the maximum; this definition is at least objective. Nevertheless, any other choice leads to the same conclusion: as T_c decreases, RRR increases. The bars in Fig. 1(b) are not an indication of the measurement errors—the latter are smaller than 0.1 K. Rather, these are FWHMs of the peaks in $\mathrm{d}R(T)/\mathrm{d}T$, which provide a measure of the transition widths. The same convention is used in other figures.

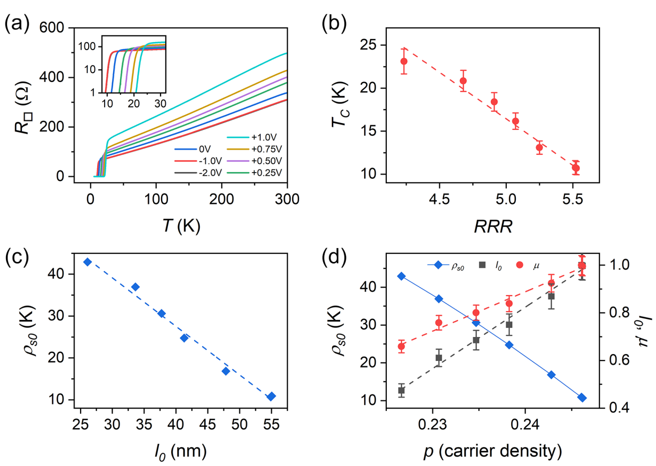

FIG. 1. Tuning the critical temperature, T_c, and the superfluid phase stiffness, ρ_{s0}, in overdoped LSCO$(x = 0.28)$ by electrolyte gating. (a) Main panel. $R(T)$ dependence measured for different values (color coded) of the applied gate voltage $V_{\rm gate}$. Inset: the same data, on log scale and enlarged on the superconducting transition to highlight a substantial change in T_c. (b) The dependence of T_c on the residual resistance ratio, $RRR = R(T = 295\ \mathrm{K})/R_0$. (c) The gating-induced changes of ρ_{s0} as a function of the mean-free path l_0. (d) The dependence of ρ_{s0} (blue diamonds), l_0 (black squares), and the hole mobility μ (red dots) on the doping level p. In all panels, T_c and ρ_{s0} increase in the direction of increasing disorder (smaller RRR, l_0, and μ).

The results in Figs. 1(a) and 1(b) are presented in terms of the variables that are directly measured or extracted by fitting the raw data. However, to make our findings more accessible, we can resort to two other inferred parameters that are commonly used to characterize metals and semiconductors. One of these is the electron mobility $\mu = 1/ne\rho$, where n is the normal state carrier density, e is the electron charge, and ρ is the resistivity. The other is the electron mean-free path $l_0 = m^* v_F / ne^2 \rho_0$, where v_F is the Fermi velocity and m^* is the quasiparticle effective mass [20]. Both μ and l_0 are expected to decrease with increasing disorder.

We also measured the magnetic penetration depth λ *in situ* upon electrolyte gating using the mutual inductance technique [6,21] and determined the two-dimensional (2D) superfluid phase stiffness, $\rho_{s0} = A/\lambda^2$, where $A = \hbar^2 d / 4\mu_0 \mathrm{k_B} e^2 = 4 \times 10^{-12}\ \mathrm{m^2\,K}$, $\mathrm{k_B}$ is the Boltzmann constant, $\hbar$ is the reduced Planck constant, $\mu_0 = 4\pi \times 10^{-7}\ \mathrm{N/A^2}$ is the vacuum permeability, d is the layer thickness. This phase stiffness is proportional to the 2D

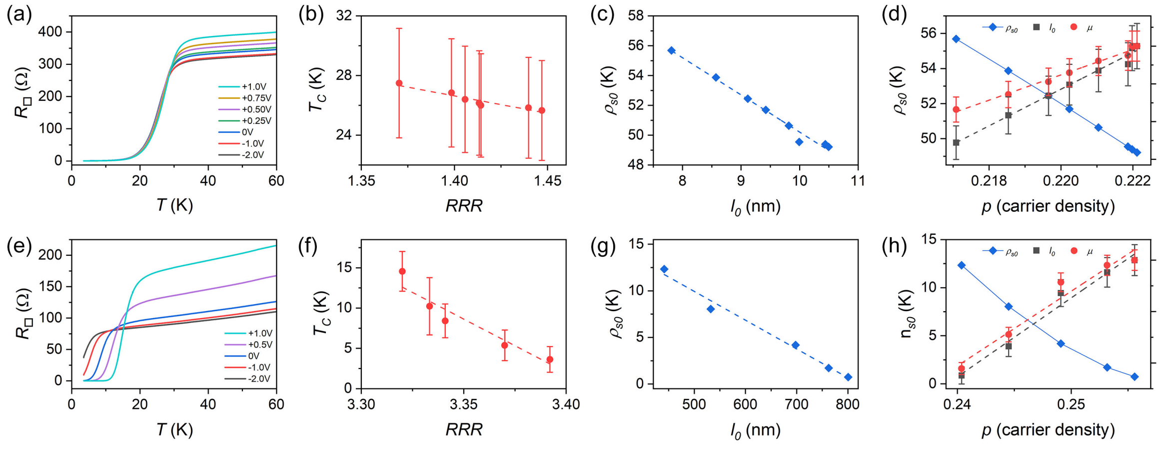

FIG. 2. Electrolyte gating of two other overdoped LSCO films, $x = 0.22$ and $x = 0.31$. (a) $R(T)$ dependence measured in LSCO($x = 0.22$) for different values (colorcoded) of the applied gate voltage V_{gate}. (b) The dependence of T_c on $RRR = R(T = 295\ \mathrm{K})/R_0$ in LSCO($x = 0.22$). (c) The dependence of ρ_{s0} on the mean-free path l_0 in LSCO($x = 0.22$). (d) The gating-induced changes of ρ_{s0} (blue diamonds), l_0 (black squares), and the hole mobility μ (red dots) as a function of the doping level p in LSCO($x = 0.22$). (e) $R(T)$ measured in LSCO($x = 0.31$) for different values of V_{gate}. (f) The dependence of T_c on RRR in LSCO($x = 0.31$). (g) The dependence of the ρ_{s0} on l_0 in LSCO($x = 0.31$). (h) The gating-induced changes of ρ_{s0} (blue diamonds), l_0 (black squares), and the hole mobility μ (red dots) as a function of p in LSCO($x = 0.31$).

superfluid density $n_s^{2\mathrm{D}} = \rho_s(4k_{\mathrm{B}}m^*/\hbar^2)$. Consistent with previous results [6], we found that ρ_{s0} decreased and tracked T_c as doping increased. In Fig. 1(c), we show the gating-induced changes in ρ_{s0} as a function of l_0. In Fig. 1(d), we show ρ_{s0}, l_0, and μ, as functions of the doping level p. To plot l_0 and μ on the same scale, we normalized both to their values at $V_{\mathrm{gate}} = 0$. As we decrease the doping, ρ_{s0} increases, as expected; however, both l_0 and μ decrease, indicating that disorder increases. Hence, both T_c and ρ_{s0} decreased with *decreasing* disorder.

In Fig. 2, we show analogous data for two other LSCO films: one less overdoped ($x = 0.22$) and the other more overdoped ($x = 0.31$). Because of some sample-to-sample variability in the frozen disorder (e.g., from Sr^{2+} dopant ions and oxygen vacancies), the starting point (i.e., R_0) varies. The effects of electrolyte gating on T_c vary, as well; they are smaller closer to the optimal doping because the $T_c(p)$ dome is presumably a parabola, i.e., flat near the top. On the other hand, at very high doping, screening improves and the relative change in p is smaller, so T_c varies less for the same V_g. Nevertheless, the trends are always the same—T_c and ρ_{s0} increase in the direction of increasing disorder.

More information can be inferred by cycling the sample repeatedly between positive and negative gate voltages. We illustrate this in Fig. 3 for the LSCO($x = 0.28$) film. For $-2.25\ \mathrm{V} < V_{\mathrm{gate}} < +1.25\ \mathrm{V}$, the same $R(T)$ curves are reproduced; there is no hysteresis. However, for $V_{\mathrm{gate}} \gtrsim +1.25\ \mathrm{V}$, the sample enters a different regime in which the resistance increases abruptly across the entire temperature range. At the same time, the superconducting transition broadens and remains incomplete down to 4 K

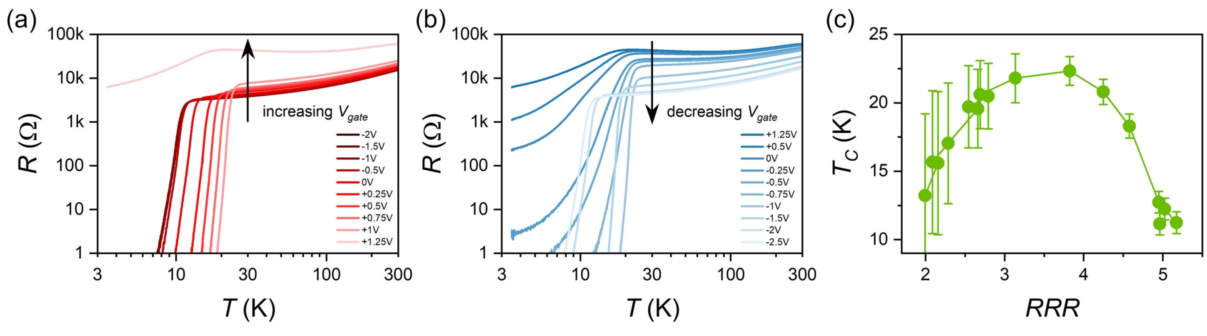

FIG. 3. Cycling V_{gate} in LSCO($x = 0.28$). (a) When V_{gate} reaches $+1.25$ V, $R(T)$ increases abruptly in the entire temperature range, while the T_c onset becomes lower and the superconducting transition much broader. (b) When ramping the gate voltage down to -2.5 V, the set of $R(T)$ curves differs from that in panel a; here, while $R(T)$ decreases, T_c increases. (c) T_c vs RRR for the $R(T)$ curves in panels a and b. Here, T_c is no longer a single-valued function of the disorder. The vertical bars indicate the transition width, which increases with disorder, as expected.

[see Fig. 3(a)]. As we reduced V_{gate} back to zero, the $R(T)$ curves became substantially different from the initial ones [Fig. 3(b)], i.e., there was a pronounced hysteresis. This phenomenon is not an experimental artifact; it arises from the chemical modifications to the sample, well known to occur in electrochemistry when the characteristic threshold voltages are exceeded. Irrelevant of the microscopic details, the point here is that in this case, T_c is not a single-valued function of RRR anymore [Fig. 3(c)]. RRR can be reduced by a factor of 2.5, implying a greatly enhanced disorder, and yet T_c remains the same. This is an independent, strong indication that the demise of superconductivity with overdoping is not (primarily) controlled by disorder.

Consider the assumption that pair breaking is caused by scattering on randomly distributed Sr^{2+} dopant ions. Since the average distance L decreases with the doping level (as $L = a/\sqrt{p}$, where a is the in plane lattice constant), the pair density could decrease with increased doping in principle, but the numbers do not check. If we decrease doping by a relatively small amount, for example, from $p = 0.25$ to $p = 0.23$, the average distance L between Sr ions changes very little, $\Delta L/L = 4\%$. However, the effect on T_c and ρ_{s0} is huge, with $\Delta T_c/T_c = \Delta\rho_{s0}/\rho_{s0} = 270\%$ [6]. It is hard to see how such a large change in T_c and ρ_{s0} could arise from a tiny decrease in the defect spacing.

An alternative assumption is that intrinsic inhomogeneity arises from electronic phase separation [11,14], leading to granular superconductivity [13]. In this picture, the reduction in n_{s0} comes from a progressive decrease in the superconducting volume fraction. However, several independent experiments—mutual inductance, scanning superconducting quantum interference device, and STM measurements—all show spatially uniform superfluid density [6,22].

In dirty-BCS scenarios, T_c is expected to drop as the pairing interaction weakens, while n_{s0} decreases due to increasing atomic disorder, so these two phenomena have different causes. However, experimentally, T_c and n_{s0} stay linearly proportional over an extensive doping range and vanish at the same critical doping $p = p_{c2}$. This holds in very dirty ($l_0 < \xi_0$, where ξ_0 is the coherence length) superconductors like $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi2212), clean ($l_0 > \xi_0$) like LSCO, and ultraclean ($l_0 \gg \xi_0$) like $Tl_2Ba_2Cu_2O_{8+\delta}$ (Tl2202) [1–6], i.e., independently of the initial level of the disorder. Very telling, the $T_c(n_{s0})$ dependence is precisely the same in overdoped LSCO [6] and underdoped $YBa_2Cu_3O_{7-\delta}$ (YBCO) [22], even though the insulator-superconductor transition in the underdoped YBCO is attributed to quantum phase fluctuations rather than the disorder [23]. To account for all these experimental facts within a dirty-BCS scenario, one must postulate a set of quite unlikely coincidences.

In LSCO, the temperature dependence of the superfluid density is accurately linear, $n_s(T) = n_{s0} - \alpha T$, all the way to the edge of the superconducting dome [6]. In BCS theory, the superconducting gap is a collective property dependent on the pair density, so one would expect $n_s(T)$ to bend down as the gap closes near T_c. However, STM and angle-resolved photoemission spectroscopy experiments on LSCO and Bi2212 showed that the gap filled rather than shrinking with temperature; this is sometimes called "Dynes superconductor" physics [24,25]. Terahertz spectroscopy also revealed that a significant fraction of the carriers in overdoped LSCO do not condense even at $T \to 0$, and it increases with doping. At the same time, the pairing energy scale remains the same [26].

Finally, in BCS theory, upon cooling the sample, the superconducting state emerges from the Fermi liquid (FL) state. However, the normal state of the cuprates does not conform to the standard Landau FL theory. For example, at $p = 0.19$, the $\rho(T)$ dependence is strictly linear and extrapolates to the origin. Moreover, the magnetoresistance is linearly dependent on the magnetic field strength [16]. This strange metal behavior cannot happen in an FL. Neither can electronic nematicity, i.e., the spontaneous breaking of rotational symmetry in the electron fluid, which in LSCO occurs at all doping levels up to the dome edge [27].

In conclusion, we synthesized overdoped LSCO films by ALL MBE and performed transport and magnetic susceptibility measurements. We studied how T_c and ρ_{s0} depend on the disorder quantified by several parameters (RRR, l_0, or μ). We focused on bulk probes to avoid concerns about surface-specific effects and tuned hole doping in LSCO films via electrolyte gating to reduce uncertainties from sample-to-sample variations. In the range $-2.5\ \mathrm{V} < V_{\mathrm{gate}} < +1\ \mathrm{V}$, where electrolyte gating effects are reversible, we observed that T_c and n_{s0} decreased as disorder decreased. At higher $V_{\mathrm{gate}} > +1\ \mathrm{V}$, the changes in the film were hysteretic, and T_c was a multivalued function of RRR; therefore, the disorder does not uniquely determine T_c. Both findings contradict the hypothesis that the increasing disorder drives the demise of T_c and n_{s0} with overdoping in cuprates, at least within the framework of models of disorder proposed so far. This brings back to center stage the following questions: Why does T_c decrease and vanish with overdoping? Why does n_{s0} track T_c? Why is the $T_c(n_{s0})$ relation independent of the details of the band structure and even of whether the cuprate is underdoped or overdoped? The correct picture of HTS in cuprates should provide at least qualitative answers, and explain the other observed departures from the standard Landau FL and BCS behavior.

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Data availability—The source data files for all the figures are openly available from the authors upon request.

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