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Insensitivity of T_c to the residual resistivity in high-T_c cuprates and the tale of two domes

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One of the few undisputed facts about hole-doped high-T_c cuprates is that their superconducting gap Δ has d-wave symmetry. According to 'dirty' d-wave BCS theory, even structural (non-magnetic) disorder can suppress Δ, the transition temperature T_c and the superfluid density ρ_s. The degree to which the latter is affected by disorder depends on the nature of the scattering. By contrast, T_c is only sensitive to the total elastic scattering rate (as estimated from the residual resistivity ρ_0) and should follow the Abrikosov-Gor'kov pair-breaking formula. Here, we report a remarkable robustness of T_c in a set of Bi2201 single crystals to large variations in ρ_0. We also survey an extended body of data, both recent and historical, on the LSCO family which challenge key predictions from dirty d-wave theory. We discuss the possible causes of these discrepancies, and argue that either we do not understand the nature of disorder in cuprates, or that the dirty d-wave scenario is not an appropriate framework. Finally, we present an alternative (non-BCS) scenario that may account for the fact that the superconducting dome in Tl2201 extends beyond that seen in Bi2201 and LSCO and suggest ways to test the validity of such a scenario.

KEYWORDS

superconductivity, cuprates, charge transport, dirty d-wave theory, disorder

1 Introduction

Over the past decade or so, overdoped (OD) cuprates, i.e., those with a carrier density beyond optimal doping, have become the central focus of efforts to elucidate the origin of high-T_c superconductivity. This shift of focus has emerged from two seemingly contradictory standpoints. The first is the perceived simplicity of the nature of the OD regime; the normal state pseudogap (on the hole-doped side) having been suppressed and with it, many of the associated ordering tendencies [1]. The second is the realization that this region of the cuprate phase diagram also hosts its own highly anomalous properties, both in the normal and superconducting (SC) states [2–14]. Chief among these is the report of a robust linear-in-T dependence of the superfluid stiffness ρ_s as $T_c \to 0$ on the overdoped side [5]. Prior to this discovery, the observed reduction in T_c and ρ_s with overdoping had been attributed to a combination of a diminishing pairing interaction and the pair-breaking effects of impurities treated within a 'dirty d-wave' extension of BCS theory. The robustness of the T-linear form of $\rho_s(T)$, a hallmark of clean d-wave superconductivity, was inconsistent with theoretical predictions and thus presented a challenge to the pre-existing consensus of what drives the reduction of T_c and ρ_s with overdoping.

In response to this challenge, a thorough examination of the viability of the dirty d-wave scenario was carried out on two very different OD cuprates–$La_{2-x}Sr_xCuO_4$ (LSCO) and

$Tl_2Ba_2CuO_{6+\delta}$ (Tl2201)–using realistic parameterisations of their respective electronic structures and treating the scattering potentials generated by out-of-plane defects with *ab initio* DFT calculations [15–19]. The conclusions of this work were that many facets of the SC state in both families, including the dependence of ρ_s on T and p [5], the THz optical conductivity (in LSCO) [7], the residual specific heat [20, 21] and the residual thermal conductivity [22–25] could be successfully captured within the existing framework. In order to account for the robustness of the T-linearity of $\rho_s(T)$ down to low-T, the total scattering potential was argued to consist almost exclusively of weak (Born) scatterers–due to the out-of-plane defects–combined with a small amount of strong (unitarity-limit) scatterers located within the CuO_2 plane (see also Ref. [26]).

Within the same picture, the suppression of T_c from its disorder-free value T_{c0} does not depend on the nature of the scatterer, only on the absolute magnitude of the normal-state scattering rate Γ_n, as described by the Abrikosov-Gorkov pair-breaking formula [27]. In the work of Broun, Hirschfeld and co-workers [15–19], estimates for Γ_n were deduced from the residual resistivity ρ_0 (essentially an extrapolation of the normal-state in-plane resistivity $\rho_{ab}(T)$ down to zero temperature). These estimates for Γ_n (≈20 K in OD Tl2201 and ≈55 K in OD LSCO) were then found to generate $T_c(p)$ domes with realistic values for the maximum T_c ($T_c^{\max}$) as well as the doping level (p_{sc}) at which superconductivity vanishes in both families. Initially, these $T_c(p)$ domes were derived from a single $T_{c0}(p)$ dome [17]. The later, more refined *ab initio* treatment required two $T_{c0}(p)$ domes to reproduce the experimental results though the difference between them was only slight [18].

One of the most consequential aspects of dirty d-wave theory, largely overlooked until now, is the strong dependence of T_c on Γ_n, irrespective of the nature of the scattering potential. As a rough guide, an increase in ρ_0 by 10 $\mu\Omega$cm corresponds to a decrease in both Γ_n and T_c of order 10 K. (More details of this correspondence will be presented later). Such modest variations in ρ_0 are not uncommon in samples from different growth batches or in samples synthesized in different laboratories and thus one might expect a notable variation in reported T_c values. Yet, throughout almost 4 decades of cuprate research, the SC domes reported in the literature for a particular cuprate family have been, to all intents and purposes, identical, both in terms of their $T_c^{\max}$ values and the doping extent of the dome itself.

The aim of this article is to highlight this insensitivity of T_c to changes in ρ_0 in different OD cuprates through a combination of new measurements and analysis of existing data. The article itself is divided into three parts. The first is an in-house transport study of one of the most inhomogeneous cuprate families–Pb/La-doped $Bi_2Sr_2CuO_{6+\delta}$ (Bi2201)–that exhibits a remarkable robustness of T_c to marked changes in ρ_0. The second is a survey of recent transport data on LSCO crystals and films which, when combined with multiple reports of the $T_c(x)$ dome in LSCO spanning several decades, represent a notable challenge to the applicability of dirty d-wave BCS theory to OD cuprates. In the final section, we present a simple (two-fluid) scenario for OD cuprates which offers an alternative explanation as to why p_{sc} (Tl2201) > p_{sc}(Bi2201/LSCO). While incorporating disorder-induced pair-breaking in some capacity, this scenario considers the strange metallic nature of OD cuprates [28] as its defining feature. The corollary of this study is that either we do not understand the nature of disorder in cuprates, or that the dirty d-wave scenario, at least in its present guise, is not an appropriate framework to describe the suppression of superconductivity in OD cuprates as $p \to p_{sc}$.

2 Results and discussion

2.1 Robustness of T_c to variations of ρ_0 in Bi2201

Large single crystals of Pb/La-doped Bi2201 were taken from boules grown independently at two sites via the floating-zone technique. The doping level of the crystals used in this study (p = 0.215 ± 0.005) was estimated from the measured T_c using the Presland relation $1 - T_c/T_c^{\max} = 82.6\,(p - 0.16)^2$ [29], with $T_c^{\max}$ = 36 K. Recently, it was shown that when using this relation, normalized $\rho_{ab}(T)$ curves for Bi2201 and LSCO single crystals of the same p (or T_c) value collapse on top of one another [30]. The crystals were cleaved along the ab-plane and cut into shape along the c-axis using a wire saw which leaves no burring of the edges. Typical sample dimensions were approximately 1,000 × 200 × 8–40 μm^3 (the thicknesses having been determined by a scanning electron microscope).

For the resistivity measurements, electrical contacts were made using 25 μm Au wire and fixed using Dupont 6838 paint before being annealed in flowing oxygen for 10 min at 450°C. Typical contact resistances were between 1 and 10 Ω. All samples were cooled using a ^{4}He flow cryostat and their in-plane resistivity measured using a standard four-point ac lock-in detection technique. While the dimensions of the samples could be determined to a high degree of accuracy, the absolute magnitudes of $\rho_{ab}(T)$ were subject to an uncertainty of ±25% due to the uncertainty in estimating the distance between the voltage electrodes. Complementary magnetization measurements were carried out in a commercial SQUID magnetometer.

Figure 1 shows $\rho_{ab}(T)$ curves for 6 Pb/La-doped Bi2201 crystals grown in Amsterdam (top panel) and Sendai (bottom panel). For both sets of crystals $T_c \simeq 27$ K and $p \simeq 0.20$. According to a recent combined transport and angle-resolved photoemission spectroscopy (ARPES) study [30], this doping level lies very close to the doping level p^* at which the pseudogap regime terminates in Bi2201. Correspondingly, all $\rho_{ab}(T)$ curves display a quasi-T-linear dependence from room temperature down to T_c albeit shifted with respect to each other due to the difference in their respective ρ_0 values.

In certain crystals (labelled ♯1B and ♯1C in Figure 1A), $\rho_{ab}(T)$ displays a small upward deviation from T-linearity below around 75 K. One possible origin for this upward deviation is contamination of the signal from c-axis mixing, whereby a proportion of the current flows between the CuO_2 planes. The anisotropy in the resistivity ρ_c/ρ_{ab} is extremely large in Bi2201 (≈10^5–10^6) [31] and $\rho_c(T)$ is known to exhibit only weakly metallic behaviour in the OD regime. Hence, any c-axis mixing would lead to a marked increase in the absolute value of the as-measured resistivity relative to the intrinsic ab-plane response as well as a different T-dependence. In order to minimise this possibility, each sample was mounted in a 'floating' configuration, i.e., elevated above the substrate with the silver paint fully extending across the sample thickness, in order to isolate the in-plane current response and avoid any contamination from current along the c-axis. Using only samples in which the resistivities on opposite sides of the crystal were

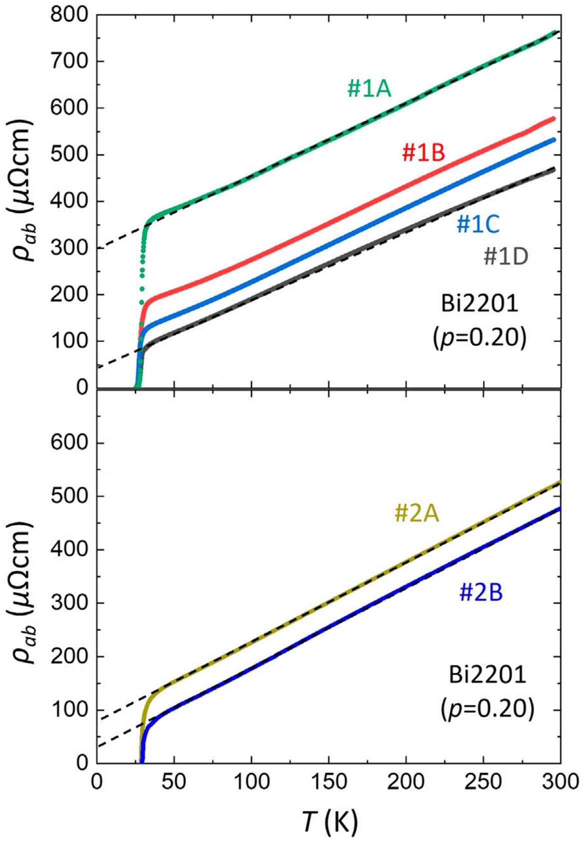

FIGURE 1
In-plane resistivity *versus* temperature $\rho_{ab}(T)$ for several Bi2201 single crystals with a doping level $p \approx 0.20$. The crystals in the two panels were grown at two distinct sites: those labeled ♯1 (top panel) were grown in Amsterdam, while those labeled ♯2 (bottom panel) were grown in Sendai. The dashed lines are extrapolations of a fit to $\rho_{ab}(T)$ between 50 and 150 K to allow a better estimate of the residual resistivity (ρ_0) values for each crystal. Note that the $\rho_{ab}(T)$ curves have been normalised such that their high-T T-linear slopes are equivalent. The adjustments required to normalise these slopes were of the order of the geometrical uncertainty (±25%) in each panel.

identical, we obtained a series of $\rho_{ab}(T)$ curves that exhibit the same T-linear slope between 75 K and 300 K (to within our geometrical uncertainty), suggesting that c-axis mixing is indeed negligible in these crystals.

The most striking feature of Figure 1 is the invariance of T_c (= 27 ± 1 K), irrespective of the magnitude of ρ_0, that itself varies by over 250 $\mu\Omega$cm. (For each crystal, ρ_0 is obtained by extrapolating a fit to $\rho_{ab}(T)$ between 50 K and 150 K). Moreover, any small variations in the value of T_c do not appear to be correlated with ρ_0. As we will discuss in the following section, this level of impurity scattering should be enough to destroy superconductivity many times over. Such robustness is contrary to expectations within dirty d-wave theory in which changes in T_c and ρ_0 are strongly correlated (via Γ_n).

2.2 Comparison with dirty *d*-wave theory

Dirty d-wave theory, as applied to cuprate superconductivity, is an extension of an original treatment of paramagnetic impurities in a conventional (s-wave) BCS superconductor [27]. Due to the sign change of the SC order parameter Δ occurring at the nodes, even non-magnetic impurities can induce pair breaking in a d-wave superconductor. This in turn leads to a residual density of zero-energy states and a suppression of both T_c and the superfluid density n_s. More quantitatively, Δ closes at a T_c value that is reduced in the presence of disorder from its optimal value T_{c0} according to the Abrikosov-Gor'kov (AG) equation:

$$\ln \frac{T_{c0}}{T_c} = \psi_0\left(\frac{1}{2} + \frac{\hbar\Gamma_n}{2\pi k_B T_c}\right) - \psi_0\left(\frac{1}{2}\right), \quad (1)$$

where ψ_0 is the usual digamma function, Γ_n is the normal-state impurity scattering rate, and $\hbar$ and k_B are the reduced Planck constant and Boltzmann constant, respectively.

As mentioned in the Introduction, a recent series of studies based on the self-consistent T-matrix approximation (SCTMA) have indicated that a number of experimental observations in LSCO and Tl2201 can be successfully accounted for by carefully treating the scattering potentials arising from different types of out-of-plane disorder within the dirty d-wave formalism [15–19]. Within their picture, the momentum relaxation rate is assumed to be the same as the single-particle scattering rate, as deduced, for example, from ARPES. Accordingly, Γ_n can be estimated from the Drude expression for the (residual) dc resistivity:

$$\rho_0 = \frac{m^*}{ne^2}\Gamma_n. \quad (2)$$

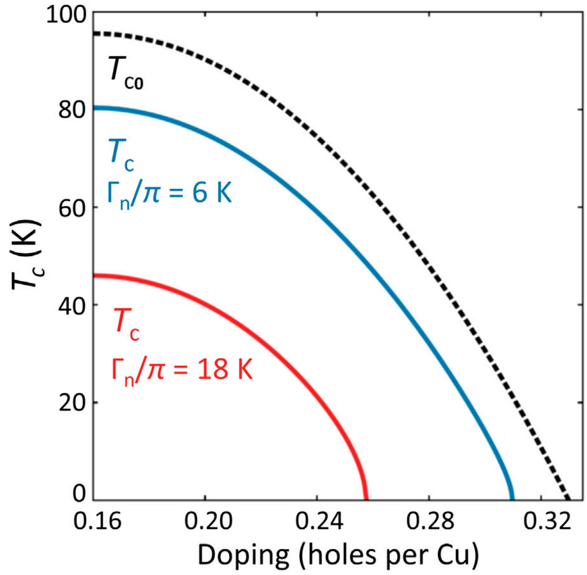

FIGURE 2
Predictions of dirty *d*-wave theory. For a single parabolic doping dependence of the underlying $T_{c0}(p)$, different choices of Γ_n result in superconducting domes $T_c(p)$ reminiscent of Tl2201 and LSCO. The quoted values of $\Gamma_n = 6\pi$ (18π) K correspond to ρ_0 values of 6 (20) $\mu\Omega$cm for Tl2201 (LSCO), respectively. For Bi2201, ρ_0 = 50 $\mu\Omega$cm corresponds to $\Gamma_n \sim$ 90 K or 30π K. Reproduced with kind permission from Ref. [17].

Here m^* is the effective mass, n is the carrier density and e is the electronic charge. Within the SCTMA, Born and unitarity scattering contribute additively to Γ_n and thus, from the perspective of Eq. 1, the suppression of T_c is largely independent of the impurity phase shift. Note too that the above expression does not take into account the effects of small-angle scattering, which can cause the momentum relaxation rate to be substantially smaller than the single-particle scattering rate. Hence, the estimate of Γ_n from Eq. 2 is in fact a lower bound for input into Eq. 1.

Figure 2 reproduced from Ref. [17]–shows how different $T_c(p)$ domes for LSCO (red line) and Tl2201 (blue line) can be derived from a singular form of $T_{c0}(p)$ using estimates for Γ_n that are consistent with experimental observations. The quoted values of $\Gamma_n = 6\pi$ (18π) K correspond to ρ_0 = 6 (20) $\mu\Omega$cm for Tl2201 (LSCO), respectively. (As mentioned above, in a subsequent study [18], slightly modified $T_{c0}(p)$ domes for Tl2201 and LSCO were incorporated into the model, though the differences were only minor.) The essential feature of Figure 2 is that the reduction in T_c of ~ 20 K (60 K) from its inferred T_{c0} value in optimally doped Tl2201 (LSCO) is attributed to a normal state scattering rate of approximately the same magnitude. At the same time, the extent of the SC dome in LSCO (on the overdoped side) is reduced by $\Delta p \approx 0.06$ for the same level of impurity scattering.

In order to link these estimates for Γ_n to ρ_0, we must also derive estimates for m^* and $n = (1 + p)/V_{\text{cell}}$ into Eq. 1, where V_{cell} is the volume of the unit cell. First, let us consider LSCO. For $p = x = 0.20$, we obtain $m^* \sim 10\ m_e$ (the bare electron mass) from the electronic specific heat [32] and $n = 1.3 \times 10^{28}$ m^{-3} (using $1 + p$ instead of p). Hence, Γ_n = 55 K corresponds to $\rho_0 \sim$ 20 $\mu\Omega$cm, as quoted above. Similarly for Tl2201 ($p = 0.27$), $m^* \sim 5\ m_e$ [33, 34], $n = 7.4 \times 10^{27}$ m^{-3} and Γ_n = 18 K, giving $\rho_0 \sim$ 6 $\mu\Omega$cm.

2.3 Application of dirty *d*-wave theory to Bi2201

For Bi2201, m* ~ 7–10 m_e for $p \sim 0.23$ (T_c = 18 K) [35] and $n = 7.0 \times 10^{27}$ m^{-3}. Hence, the spread in ρ_0 shown in Figure 1 (40–290 $\mu\Omega$cm) corresponds to 70 K $\lesssim \Gamma_n \lesssim$ 500 K. In other words, while the impurity scattering rate deduced from ρ_0 varies on the scale of 500 K, the superconducting transition temperature is found to be constant to within 1 K. Such extreme inequality is clearly at odds with expectations from dirty *d*-wave theory but is likely, at least in part, to reflect the presence of some form of defect that contributes to an enhanced ρ_0 while creating, by itself, little or no pair-breaking. Before addressing the viability of the BCS pair-breaking picture, therefore, let us first consider alternative explanations for this surprising finding. (Recall that we have already dismissed *c*-axis mixing in the current flow as a possible cause of this variation in ρ_0.)

In this present study, T_c values are quoted based on $\rho_{ab}(T)$ measurements. Resistivity is effectively a one-dimensional probe of superconductivity, in the sense that a transition to zero resistivity requires only a single, filamentary SC path to be realized. Hence, if a sliver of nominally pristine Bi2201 (i.e., with minimal disorder) permeates each crystal, the apparent robustness of T_c may be illusory. In the normal state, by contrast, the current distribution will be sensitive to all regions of the sample and indeed, if the SC filament is sufficiently thin, it will be dominated by those non-SC regions with higher ρ_0. Such a scenario may help explain why T_c is so insensitive to marked variations in ρ_0.

Simulations presented in Supplementary Appendix SA indicate that for such a scenario to be applicable, the SC region must occupy ~ 1% of the total volume of the sample; otherwise the T-dependence of $\rho_{ab}(T)$ will visibly deviate downwards from its intrinsic (T-linear) behaviour, which is not observed. In order to estimate the SC volume fraction of our crystals, we measured the dc magnetisation of two of them (with ρ_0 values of 80 $\mu\Omega cm$ (ρ(T) data not shown) and 300 $\mu\Omega cm$ (Sample #1A in Figure 1), respectively) using a SQUID magnetometer with the magnetic field applied parallel to the *ab*-plane (where the demagnetisation factor is minimised). The results are shown in Supplementary Appendix SB and reveal an estimated volume fraction in both crystals of ~ 100%. Thus, it seems unlikely that the presence of a filamentary SC path (which would have to be very similar in form in all crystals studied) can account for the observed robustness of the SC transition.

A more plausible origin of this insensitivity of T_c on ρ_0 is the presence of specific (extended) forms of microstructural defects (e.g., dislocations, columnar defects, grain boundaries, etc., …) that adversely affect ρ_0 while contributing minimally to pair breaking. An example of such extended defects having a profound effect on $\rho_{ab}(T)$ but a minimal effect on T_c can be found in the infinite-layer nickelates [36, 37]. Structural and electronic nanoscale inhomogeneity is a well-known feature of Bi-based cuprates [14, 38], but this inhomogeneity tends to be more point-like than extended and as such, should also have a similar effect on both T_c and ρ_0. Larger defects, such as microcracks, could also cause an increased resistivity though in this case, one might expect to see Arrhenius-type behaviour ($\rho(T) \propto \exp(-\Delta/k_BT)$) due to tunneling across the crack, as one finds in polycrystalline samples with multiple grain boundaries. While we noted earlier that there were small upturns observed in the $\rho_{ab}(T)$ curves of some of our Bi2201 crystals, there did not appear to be any correlation between the value of ρ_0 and the presence of an upturn.

A dedicated transmission electron microscopy (TEM) study is currently underway to look for evidence for the type of defect that might cause this dichotomy between T_c and ρ_0, the results of which will be published elsewhere. Although we cannot rule out the presence of such extended defects affecting ρ_0 without impacting T_c, in the following section we present a body of evidence on the LSCO family that provides arguably a greater challenge to the viability of the dirty *d*-wave scenario to cuprates within the strange metal regime.

2.4 Review of T_c dependence on ρ_0 in LSCO

It is well known that superconductivity in cuprates is strongly suppressed upon Zn substitution on the planar Cu site. In LSCO, for example, 4% Zn substitution can destroy superconductivity entirely while at the same time raising ρ_0 by ~ 50 $\mu\Omega$cm [39]. The origin of this suppression is not entirely clear. Despite being a non-magnetic impurity, Zn dopants appear to influence strongly the magnetic environment within the CuO_2 plane as well as act as unitarity-limit scatterers [39].

Mahmood *et al.* [40] recently reported a study on OD LSCO thin films irradiated using 1 MeV oxygen ions. Ion irradiation is believed to create narrow columnar defect tracks throughout the film. Figure 3A shows a series of $\rho_{ab}(T)$ curves obtained on an optimally doped LSCO film exposed to different fluences using a flux gradient to produce a spread in defect density. Irradiation leads to a maximal increase in ρ_0 of 36–40 $\mu\Omega$cm without the *T*-dependence or the slope of the $\rho_{ab}(T)$ curves changing. Such adherence to Matthiessen's rule, coupled with accompanying measurements of the low-frequency Drude response, suggests that the irradiation is simply creating additional elastic scattering centres. According to the calculations in Section 2.2, the magnitude of $\Delta\rho_0$ corresponds to $\Delta\Gamma_n$ > 100 K, and given that Eqs 1, 2 predict a ~ 1 K drop in T_c for every 1 K increase in Γ_n, clearly such a level of disorder should be sufficient to remove all vestiges of superconductivity in the film. Yet T_c itself is found to drop by less than 5 K.

One expects that an optimally doped film will possess a more robust SC state than those at a higher doping level with a lower T_c. In reality, Mahmood *et al.* observed the opposite trend. As shown in Figure 3B, for a pristine film with T_c ~ 10 K, there was no discernible change in T_c for the same level of fluence that induced a ~ 5 K reduction in T_c in the optimally-doped film. It seems that the more overdoped the pristine film is and the lower its initial superfluid density, the more robust is the superconductivity to similar levels of irradiation.

Two further examples of the insensitivity of T_c to changes in ρ_0 in OD LSCO are shown in Panels C and D of Figure 3. Panel C shows $\rho_{ab}(T)$ curves for two single crystals (x = 0.24) whose ρ_0 values differ by ~ 35 $\mu\Omega$cm, corresponding to $\Delta\Gamma_n$ ~ 100 K [41], yet the difference in T_c in the two crystals is less than 2 K. Panel D shows $\rho_{ab}(T)$ for a single crystal [2] and thin film [42] with x = 0.23. Note that the form of $\rho_{ab}(T)$, as well as their derivatives [43], are the same, implying that their doping levels are essentially equivalent. The difference in ρ_0 of the two samples (after normalising their slopes [43]) is such that $\Delta\Gamma_n$ ~ 85 K, but yet again, their T_c values are almost indistinguishable.

In the previous section, we discussed the possibility that in Bi2201, the insensitivity of T_c on ρ_0 reflects the fact that elastic scattering is dominated by extended defects that contribute largely to ρ_0 but do not, by themselves, break pairs and thereby cause a suppression in T_c. In the study by Mahmood *et al.*, extended (columnar) defects were found to cause an increase in scattering, as deduced from the width of the Drude conductivity peak. We note too that the superfluid density also decreased, implying that such defects do indeed cause pair breaking, yet T_c itself remained remarkably robust.

According to Figure 2 and the corresponding relation between Γ_n and ρ_0, dirty *d*-wave theory predicts that for every 1 $\mu\Omega$cm increase in ρ_0, T_c in optimally doped LSCO should decrease by around 2.5 K [17], irrespective of the phase shift of the dominant scattering process. At the same time, the full extent of the SC dome diminishes by Δp ~ 0.0075 per 1 $\mu\Omega$cm increase in ρ_0. This extreme sensitivity of the $T_c(p)$ dome to small changes in ρ_0 represents arguably the greatest challenge to the theory's applicability. Indeed, one of the most striking and largely overlooked features of cuprate research is the immutability of the $T_c(p)$ dome.

The six panels in Figure 4 reproduce full $T_c(p)$ domes for bulk LSCO reported over a period of 2 decades (1989–2009) [44–49]. The samples in question are both poly- and single crystalline and were prepared by various techniques, including flux and travelling-solvent floating-zone growth of single crystals and spray-drying or powder-mixing procedures for the ceramic powders. Note that all T_c values, bar those in panel (E), were determined by magnetisation or susceptibility measurements. The dashed green line in each panel represents the Presland formula [29] with $T_c^{\max}$ = 38 K and assuming

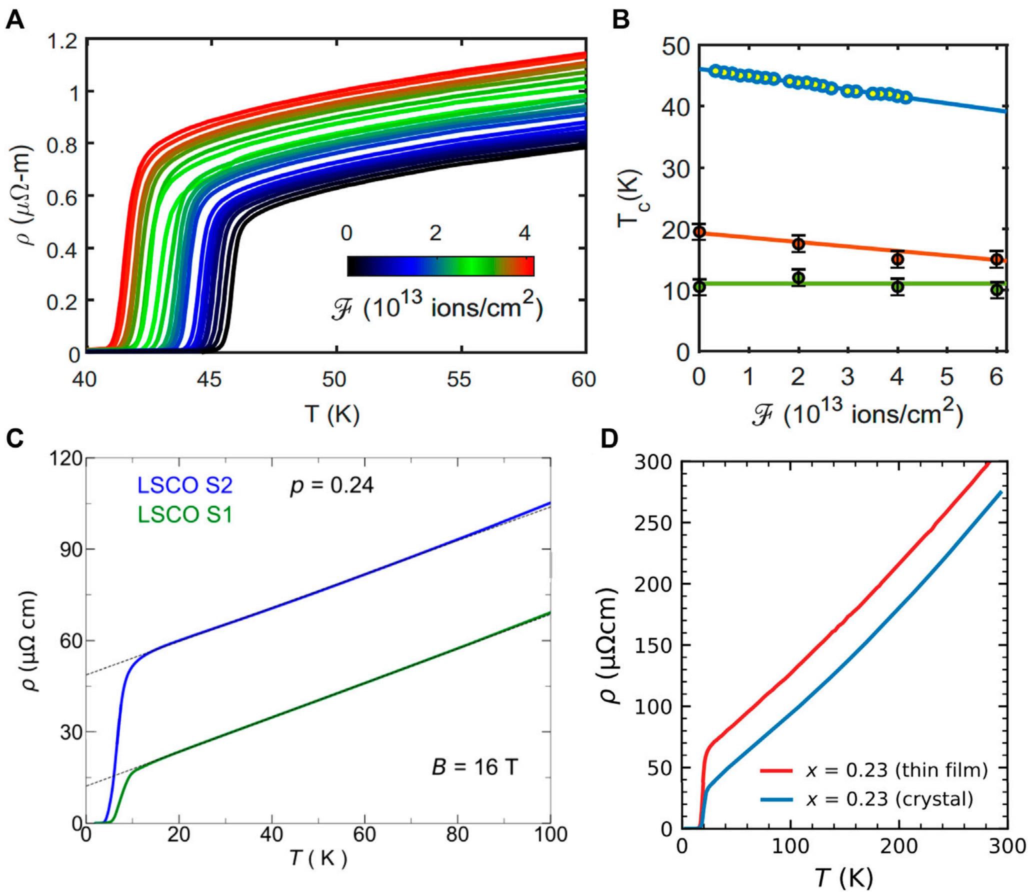

FIGURE 3
Examples of the robustness of T_c to changes in ρ_0 in LSCO. **(A)** $\rho_{ab}(T)$ of an optimally doped LSCO thin film (x = 0.16) irradiated with 1 MeV oxygen ions, but with a total fluence that varies across the devices between 0 and 4 × 10^{13} ions/cm^2, as indicated by the inset color bar. Reproduced with kind permission from Ref. [40]. **(B)** T_c as a function of irradiation fluence $\mathcal{F}$ for an optimally doped film (blue dots) and two overdoped LSCO films with different initial T_c values (red and green dots). Solid lines are guides to the eye. Reproduced with kind permission from Ref. [40]. **(C)** $\rho_{ab}(T)$ of two different crystals of LSCO (x = 0.24) with ρ_0 values differing by 36 $\mu\Omega$cm, corresponding to $\Delta\Gamma_n \sim 100$ K. Despite this large value of $\Delta\Gamma_n$, the respective T_c values differ by only 1.5 K (Note that these $\rho_{ab}(T)$ curves were obtained in a magnetic field of 16 T applied perpendicular to the CuO_2 planes). Reproduced with kind permission from Ref. [41]. **(D)** Comparison of $\rho_{ab}(T)$ of a LSCO single crystal (x = 0.23, blue curve) and a LSCO thin film (x = 0.23, red curve). Reproduced from Ref. [43]. The $\rho_{ab}(T)$ data for the thin film has been divided by two in order to normalize the slopes. The corresponding ρ_0 values are 20 and 50 $\mu\Omega$cm respectively, corresponding to $\Delta\Gamma_n \sim 85$ K.

$p = x$ (the Sr content). The consistent overlap between the dashed lines and the data indicates that the $T_c(p)$ dome in bulk, as-grown LSCO is invariant, independent of the quality of the starting materials or the way in which the samples have been synthesized. Moreover, as highlighted by the single horizontal line spanning all six panels, $T_c^{\max}$ is the same for all series of samples to within 1 K. For such consistency to be accounted for within the dirty d-wave scenario, ρ_0 would have to be identical to within 0.4 $\mu\Omega$cm ($<2\%$) for every sample at every single doping level.

Such extreme levels of reproducibility are clearly beyond all reasonable expectations (requiring as it does that all LSCO samples have identical values of Γ_n to within 1 K) and highlight a key feature of the superconductivity in LSCO that remains unresolved. It appears that the $T_c(p)$ dome in bulk LSCO is not, as has been argued [15], set by the level of disorder in the material, but by some other driving mechanism, such as the strength of next-nearest hopping [50, 51]. Certainly, it would be remarkable if the drop from $T_{c0}(p)$ to $T_c(p)$ for LSCO in Figure 2 were due to a scattering rate Γ_n whose magnitude is fixed and commensurate with a ρ_0 value equivalent to 20 $\mu\Omega$cm and that all other contributions to ρ_0, e.g., flux or crucible inclusions, were extraneous and had no further pair-breaking effect. A similar fundamental limit to ρ_0 would also have to exist for Bi2201, despite the fact that Bi2201 may contain multiple elements (e.g., Bi/Pb, La/Sr) in its formula unit.

Although a number of SC properties of OD LSCO and Tl2201 have been successfully modeled by considering the differences in the impurity potential, its phase shift and its location relative to the CuO_2 plane [15–19], the relation between T_c and Γ_n is, by and large, independent of these details and as such, should be a robust test of the theory's applicability. The inability of dirty d-wave theory to account for the remarkable insensitivity of T_c to changes in ρ_0–highlighted in Figures 1, 4 – thus implies that either we do not understand the true causes of residual resistivity in cuprates (i.e., that the correspondence between Γ_n and ρ_0 is somehow lost), or that the basis of the theory is not the right

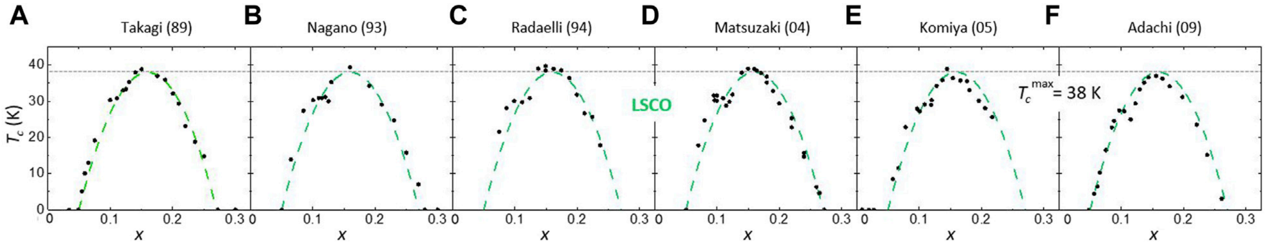

FIGURE 4
Superconducting domes in LSCO over the decades. $T_c(x)$ as measured on **(A–C)** polycrystalline pellets grown via various techniques [44–46]; **(D)** flux-grown single crystals [47]; **(E,F)** travelling-solvent floating-zone crystals [48, 49]. For all panels except (E), T_c was determined from the onset of the Meissner (diamagnetic) signal. For panel **(E)**, T_c was determined from the onset of zero resistivity. The dashed line in each panel is the Presland formula $T_c = T_c^{max}(1–82.6\ (x - 0.16)^2)$ with T_c^{max} = 38 K. All $T_c(x)$ domes appear to follow the same trajectory with the same T_c^{max} value (±1 K) and most remarkably, the same beginning (x = 0.05) and end (x = 0.27) points.

framework to describe the transition from strange metal to superconductor.

Motivated by these findings, we present below an alternative (non-BCS) scenario for the robust $T_c(p)$ domes in OD cuprates in which the non-FL nature of the cuprate strange-metal plays a central role. In the process, we offer an alternative explanation as to why the SC dome in cleaner Tl2201 extends to a higher p-value than in Bi2201 and LSCO and suggest ways to test the validity of such a scenario.

2.5 A tale of two domes

In the previous section, we highlighted various types of extended defects that could, in principle, enhance ρ_0 without necessarily inducing substantial pair breaking within the CuO_2 plane. In order to investigate whether such defects are indeed the root cause of this behaviour, more detailed microstructural studies of each of the relevant cuprate families are strongly advocated. Certainly, it is something that has been largely overlooked by the community. Until such time, however, it is worthwhile to at least consider alternative explanations for the demise of superconductivity on the overdoped side.

Franz et al. [52] have argued that the predicted drop in T_c with disorder is higher than observed experimentally due to the fact that within AG theory, the order parameter is spatially averaged, an assumption that may not be applicable to high-T_c cuprates by virtue of their short coherence lengths. Allowing for the spatial variation of the order parameter within a Bogoliubov-de Gennes formalism leads to a suppression of T_c that is indeed weaker than that predicted by the AG theory, but only by a factor of 2. Moreover, naively, one would expect the coherence length to diverge as $p \rightarrow p_{sc}$, yet according to the study of Mahmood et al., T_c becomes even more robust at higher doping levels [40].

The SCTMA used by Lee-Hone et al. [15] treats disorder using an effective medium theory in which the SC state is also assumed to be homogeneous. Other treatments, however, have considered inhomogeneity or granularity in the SC state [53, 54]. When the Cooper pair coherence length becomes comparable to the correlation length of the disorder potential, the order parameter is found to vary spatially while the superconductor segregates into regions of high and low superfluid density, that in turn enhances the propensity for SC phase fluctuations. Indeed, evidence has emerged for both granular superconductivity [14, 55] and enhanced SC phase fluctuations [56] in OD cuprates.

While the granular model described in Ref. [54] captures a number of key observations, it cannot be the complete picture. What this model–and indeed the majority of disorder models that consider this problem–assumes it that the OD cuprates are essentially Fermi-liquids that transition into a BCS superconductor (homogeneous or otherwise) below T_c. Yet, as stressed elsewhere, there is now mounting evidence that OD cuprates are in fact strange metals, with a dominant non-FL T-linear resistivity extending over the entire doping region [2]. Moreover, this strange metal is claimed to exhibit dual character [9, 10] with coexisting but spatially separated regions with FL and non-FL character, respectively [57].

The natural question that arises is whether the non-FL sector possesses the necessary qualities to preserve the size of the pairing amplitude in the presence of disorder. While there is currently no microscopic picture that addresses this, we consider here a simple 'patchwork' model for OD cuprates in which intrinsic superconductivity emerges uniquely from the non-FL sector and is resilient to large changes in Γ_n. Figure 5A shows a schematic of such a patchwork cuprate comprising distinct regions of non-FL (in blue) and FL (in red). In a related article [59], we applied both effective medium theory and random resistor networks to a binary mixture of FL and non-FL patches to capture the evolution of the low-T resistivity from purely T^2 at high dopings to T-linear near p^* (≈0.19) with an increasing fraction f of non-FL sector. The same model also explains the correlation between the T-linear resistivity coefficient and the slope of the H-linear magnetoresistance [59]. In Figure 5A, the doping level is set such that there is no percolation path available for the non-FL component. Supercurrent could, in principle, flow through the FL regions via the proximity effect and thus maintain the SC state. In inhomogeneous systems like LSCO and Bi2201, however, the FL sector will be susceptible to strong pair-breaking effects due to scattering off such inhomogeneities, thereby inhibiting the flow of supercurrent between the SC patches. Hence, as soon as the percolation limit is exceeded, the zero-resistance state is lost. For LSCO and Bi2201, this percolation limit is assumed to coincide with the end of the SC dome at p_{sc} = 0.27.

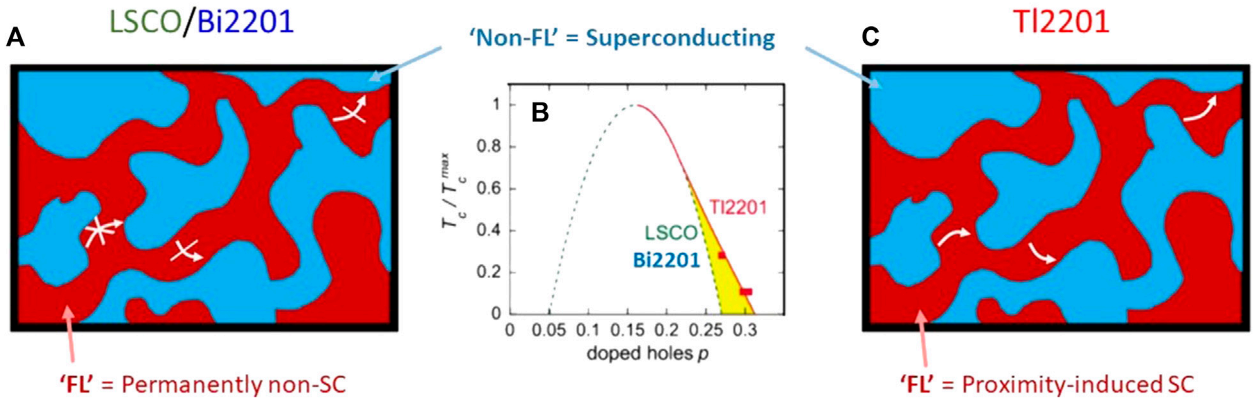

FIGURE 5
A tale of two superconducting domes. **(A)** Patchwork model for a hole-doped cuprate comprising distinct regions of non-FL (in blue) and FL (in red). Here, the doping level is such that there is no percolation path available for the non-FL sector. In LSCO and Bi2201, supercurrent could, in principle, flow through the FL sectors via the proximity effect (and thus maintain superconductivity. Due to the presence of strong inhomogeneities, however, pair-breaking effects inhibit the formation of proximity-induced superconductivity within the FL sectors. **(B)** Normalized $T_c(p)$ domes for LSCO and Bi2201 (dashed line) and Tl2201 (solid line and squares). Reproduced from Ref. [58]. The yellow shaded area represents the proposed region of suppressed superconductivity in OD LSCO and Bi2201. **(C)** Patchwork model for Tl2201 with the same concentration (and distribution) of non-FL sectors. Due to its lower levels of disorder, superconductivity can survive beyond the percolation limit because the supercurrent is now able flow through the FL sectors via the proximity effect. Once the non-FL sector vanishes at p = 0.31, however, then all traces of superconductivity are lost.

According to Pelc et al., percolation emerges when the fraction of SC patches reaches a critical value of 0.3 (assuming SC and non-SC patches of equivalent size) [60]. Related to this, α_1–the coefficient of the low-T T-linear resistivity in OD cuprates–is found to grow linearly from p = 0.31 up to its maximum value $\alpha_1^{\max}$ at $p^* \sim$ 0.20 where the pseudogap opens. Thus, at p = 0.27, $\alpha_1/\alpha_1^{\max} \sim 0.35$. In a recent high-field transport study [59], we argued that $\alpha_1/\alpha_1^{\max}$ is a measure of the fraction of Planckian carriers that are present at a particular doping. If these carriers, and only these carriers, form the superfluid condensate in LSCO and Bi2201, then the reason for the onset of superconductivity at $p = p_{sc}$ becomes self-evident–it is the point at which the supercurrent can travel percolatively between adjacent patches.

Figure 5B compares the normalised T_c dome for LSCO and Bi2201 (dashed line) with that of Tl2201 (solid line and squares). The yellow shaded area in Figure 5B represents the region of the phase diagram where the superconductivity in Tl2201 is enhanced relative to that seen in LSCO and Bi2201. (This extended region of superconductivity is essentially the same as that shown in Figure 2.) Tl2201 is known to be more homogeneous than LSCO and Bi2201 and possess longer mean-free-paths (as manifest in lower ρ_0 values) [34]. According to the above picture, the level of disorder scattering in Tl2201 is low enough to allow supercurrent to traverse the FL sectors via the proximity effect (see Figure 5C) and for superconductivity to persist beyond the percolation limit. Once the non-FL sector vanishes at p = 0.31, however, then all traces of superconductivity are lost.

This picture represents a marked departure from the extended BCS description for a disordered d-wave superconductor, yet is clearly nothing more than a toy model at present. Before closing, therefore, let us consider some of the consequences of the proposed picture and how it might be tested experimentally. One such consequence may in fact have been tested already. In an earlier electron irradiation study on OD Tl2201 (T_c = 31 K) [61], an increase in ρ_0 by 70 $\mu\Omega$cm was found to cause a reduction of 20 K in T_c. Using the values quoted in Section 2.2, we find that $\Delta\Gamma_n \sim 210\ \mathrm{K} > 10\ \Delta T_c$. Despite this level of T_c reduction being far smaller than expected by modified AG theory ($\Delta T_c = -(\pi/4)\Gamma_n$ [61]), it is still more than is seen in LSCO or in Bi2201. (In Ref. [61], the authors used p, rather than $1 + p$, for the carrier density, making the agreement with their expectations from AG theory appear reasonable.). If superconductivity within the FL sector is susceptible to disorder (in accordance with dirty d-wave theory) but is resilient within the non-FL sector, electron radiation might induce pair-breaking predominantly or uniquely within the FL sector. Moreover, if the doping level sits close to the percolation threshold, superconductivity will be appreciably suppressed. Further irradiation, on the other hand, would not cause a further deterioration in T_c due to the resilience of the superfluid residing the non-FL sector. A more dedicated irradiation study, over a range of dopings and to higher fluences, could thus serve as a robust test of the validity of this proposal.

The other corollary of this picture is the presence of SC droplets beyond p_{sc} = 0.27. According to Ref. [59], signatures of superconductivity are intimately tied to the existence of the T-linear component in $\rho(T)$ that itself indicates the fraction of carriers that are not standard Landau quasiparticles. Hence, for $0.27 \leq p \leq 0.31$, one expects SC patches to survive, fluctuating or otherwise. A recent STM study [14] observed gap features persisting in nominally non-SC Bi2201, albeit with a low filling fraction, consistent with this picture. In order to test this idea more rigorously, however, one would need to track the evolution of these features in combination with $\rho(T)$ measurements up to p = 0.31 and beyond.

3 Conclusion

The measurements, reproductions and analysis presented in this article serve to highlight serious shortcomings in our understanding of the normal and SC properties of overdoped cuprates. The fundamental problem can be expressed as follows; either we do not understand the relation between Γ_n and ρ_0 and the origins of residual resistivity in OD cuprates, or dirty d-wave theory, at least in its present guise, is not the appropriate framework to describe OD cuprates. The reality is probably a combination of the two. The robustness of the $T_c(p)$ domes in LSCO is particularly challenging for scenarios based on standard pair-breaking effects in d-wave superconductors. At the same time, there clearly needs to be a more concerted effort to understand the nature of defects and their contribution to pair-breaking and to ρ_0.

In the absence of a consistent picture, we have introduced here an alternative explanation for the robustness of T_c in different OD cuprates, based on a 'patchwork' model that recognises the dual character and non-FL nature of the strange metal regime and the importance of the latter for pair condensation. Within this model, dirty d-wave theory is still found to play some role, accounting for difference in the extent of the $T_c(p)$ domes in LSCO, Bi2201 and Tl2201. Further irradiation studies on samples located at the edge of the SC dome may allow us to differentiate between the different explanations for these striking effects.

Data availability statement

The raw data supporting the conclusion of this article will be made available by the authors, without undue reservation.

Author contributions

DJ: Conceptualization, Data curation, Formal Analysis, Investigation, Methodology, Writing–original draft, Writing–review and editing. JA: Data curation, Investigation, Validation, Writing–review and editing, Writing–original draft. RN: Data curation, Investigation, Validation, Writing–review and editing. NH: Conceptualization, Formal Analysis, Funding acquisition, Methodology, Project administration, Resources, Supervision, Writing–original draft, Writing–review and editing.

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Conflict of interest

The authors declare that the research was conducted in the absence of any commercial or financial relationships that could be construed as a potential conflict of interest.

Supplementary material

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fphy.2024.1396463/full#supplementary-material

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