

Anomalous behavior of T_c and pseudogap in the superconductor $\text{Ca}_x\text{La}_{1-x}\text{Ba}_{1.75-x}\text{La}_{0.25+x}\text{Cu}_3\text{O}_y$ with respect to doping and ion size

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The cuprate superconductor $\text{Ca}_x\text{La}_{1-x}\text{Ba}_{1.75-x}\text{La}_{0.25+x}\text{Cu}_3\text{O}_y$ has been proposed as a model system which allows systematic variation in the optimum transition temperature, $T_{c,\text{max}}$, with doping and ion size. However, we show that the $T_{c,\text{max}}$ values are anomalous in this system. We determine the doping accurately and show that the phase curve, as a function of doping, is anomalously truncated so that values of $T_{c,\text{max}}$ appear to decrease with decreasing x , concealing the generic increase expected as ion size increases. We also thereby resolve its reported anomalous pseudogap behavior from NMR and anomalous superconducting gap from angle-resolved photoemission spectroscopy.

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The mechanism for electron pairing in cuprate high- T_c superconductors (HTS) remains to be conclusively identified though it is widely believed to be of magnetic origin [1]. For hole-doped cuprates, T_c follows a dome-like dependence on hole concentration, p (in units of holes per planar Cu) with T_c rising to a maximum, $T_{c,\text{max}}$, at optimal doping, $p \approx 0.16$ [2]. Because of its two-dimensional character the electronic density of states (DOS) diverges at a van Hove singularity (vHs) which usually sits in the overdoped region of the phase diagram [3] while in the optimal and underdoped regions a pseudogap opens a partial gap in the DOS that is probably due to Fermi surface reconstruction [4]. Near $p \approx 0.125$ the $T_c(p)$ phase curve is typically reduced below the idealized parabolic dome due to a competing charge density wave (CDW) [5] giving the so-called “60 K plateau” in $\text{YBa}_2\text{Cu}_3\text{O}_y$ [6,7].

The value of $T_{c,\text{max}}$ varies considerably from one cuprate to another; however, $T_{c,\text{max}}$ is found to correlate remarkably well with a composite bond valence sum (BVS) parameter, $V_+ = 6 - V_{\text{Cu}} - V_{\text{O}(2)} - V_{\text{O}(3)}$ (see Fig. 1, omitting the black circles). V_+ was established as a measure of the charge distribution between Cu and O orbitals in the CuO_2 basal plane [8] but it also is a measure of the in-plane stress, as annotated at the top of the figure. Here O(2) and O(3) are the two planar oxygen sites.

The manifest success of this correlation indicates that it captures some relatively simple underlying feature that generally accounts for the trends in $T_{c,\text{max}}$. But how do we isolate that feature in order to narrow the search for a pairing mechanism? What is needed is a means to methodically vary the interaction strength and we have advanced the idea that variations in ion size, combined with changes in external pressure, provide just such a means [9]. The effect of ion size is convolved into our parameter V_+ because as ion size is increased the CuO_2 plane is stretched and V_+ increases. Then all that is needed is a model system (or several) in which these effects can be systematically explored without substantial alteration of other structural features or the introduction of disorder.

To this end we have studied the system $R(\text{Ba},\text{Sr})_2\text{Cu}_3\text{O}_y$ where the ion size of the lanthanide, R , can be varied as can the ratio of Ba to Sr [9]. In Fig. 1 the effect of changing R is plotted with the red “+” symbols and the effect of progressively replacing Ba by Sr is plotted by the blue “+” symbols. The correlation between $T_{c,\text{max}}$ and V_+ remains well preserved.

Another extensively studied system is $\text{Ca}_x\text{La}_{1-x}\text{Ba}_{1.75-x}\text{La}_{0.25+x}\text{Cu}_3\text{O}_y$ (CLBLCO), which is amenable to progressive reduction in ion size as x increases [10]. It is isoelectronic, it apparently can be substantially overdoped, it remains tetragonal for all doping levels, and $T_{c,\text{max}}$ ranges from 45 to 80 K as x runs from 0 to 0.4. Tellingly, it was shown that $T_{c,\text{max}}$ scales with the nearest-neighbor exchange energy, J , measured by Raman scattering, suggesting a magnetic pairing scenario [11,12]. Indeed the entire phase diagram could be collapsed to a single universal phase behavior by scaling temperatures by J . This then seems an ideal model system for systematic ion-size studies.

But we show here that the $T_{c,\text{max}}$ values in this system are anomalous and that, when $x < 0.4$, the doping is significantly lower than previously inferred. Further, this system is reported to display an anomalous pseudogap behavior [13] and anomalous angle-resolved photoemission spectroscopy (ARPES) measurements of superconducting gaps [14]. Our doping analysis resolves both these issues. Taken together, we conclude that this is not the ideal model system that had been hoped.

The first indication that there is something unusual about the title system may be seen in the relation between $T_{c,\text{max}}$ and the BVS parameter, V_+ , which is shown in Fig. 1 by the solid black circles.

V_{Cu} is calculated as before [8,9] using $V_{\text{Cu}} = (3V_2 - 2V_3)/(V_2 + 1 - V_3)$, when $V_2 > 2$. For Cu, $V_n = \sum_i \exp[(R_n - R_i)/0.37]$, $n = 2, 3$, where the sum is over the oxygens neighboring the in-plane Cu. $R_2 = 1.679 \text{ \AA}$, while $R_3 = 1.73 \text{ \AA}$, as first proposed by Brown [15]. For bond lengths in CLBLCO we use the structural refinements of Agrestini *et al.* [16] for the special case $y = 7.1$ which corresponds nearly to the maximum in the phase curve for each x value. The calculated values of V_{Cu} and V_+ are listed in Table I, and as noted $T_{c,\text{max}}$ is plotted versus V_+ in Fig. 1.

Strikingly, the data in Fig. 1 do not fit the generic correlation but are actually orthogonal to it. This immediately signals that the T_c values may be anomalous, that is, all except for the $x = 0.4$ family which sits nicely on the correlation line. We also list in Table I the values of $T_{c,\text{max}}$ expected from the V_+

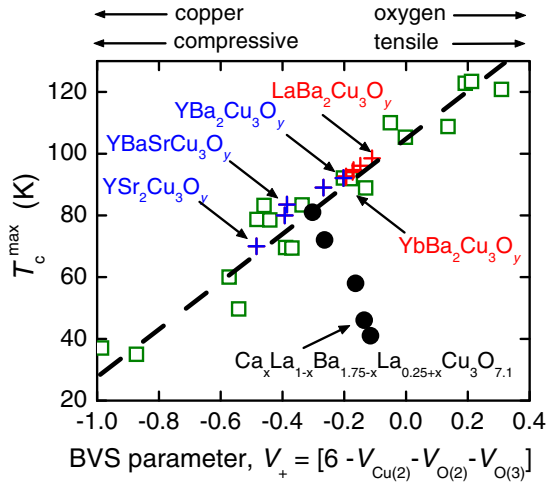


FIG. 1. (Color online) $T_{c,max}$ plotted versus the bond valence sum parameter V_+ for many HTS cuprates, as reproduced from [9]. To this is added the data for $\text{Ca}_x\text{La}_{1-x}\text{Ba}_{1.75-x}\text{La}_{0.25+x}\text{Cu}_3\text{O}_y$ with $x = 0.4, 0.3, 0.2, 0.1,$ and 0 where V_+ is calculated from the crystallographic data reported in [16]. Only the $x = 0.4$ data point fits the correlation and the trend with decreasing x is orthogonal to all other HTS cuprate data.

values if the correlation were to be satisfied, i.e., progressing up the dashed line in Fig. 1. These rise as high as $T_{c,max} = 96$ K for $x = 0$, i.e., for $\text{LaBa}_{1.75}\text{La}_{0.25}\text{Cu}_3\text{O}_y$. This is not unreasonable since $T_{c,max}$ for $\text{LaBa}_2\text{Cu}_3\text{O}_y$ is at least 98.5 K when properly optimized [9]. Clearly then there is something anomalous about the title system.

Recently Wulferding *et al.* [11] reported Raman two-magnon scattering in CLBLCO to determine the magnitude of J . As noted, they find that $T_{c,max}$ correlates with J in contrast to our result where $T_{c,max}$ was found to anticorrelate with J for $R(\text{Ba,Sr})_2\text{Cu}_3\text{O}_y$ [9]. Aware of this contradiction Wulferding *et al.* suggest that it is the J values which are inverted in the latter case due to inadequate deoxygenation. But in fact the trend in J with ion size is rather similar in both cases. It is the $T_{c,max}$ values which are inverted for CLBLCO, as indicated by Fig. 1. We now examine this more closely.

Figure 2(a) shows the reported variation in T_c with oxygen content, y , for $x = 0, 0.1, 0.2, 0.3,$ and 0.4 [16]. This reveals

TABLE I. Bond valence sums, V_{Cu} and V_+ , and $T_{c,max}$ values calculated for $\text{Ca}_x\text{La}_{1-x}\text{Ba}_{1.75-x}\text{La}_{0.25+x}\text{Cu}_3\text{O}_y$ using the crystal data of Agrestini *et al.* [16] as described in the text. $T_{c,max}^{\text{obs}}$ is the observed value and $T_{c,max}^{\text{calc}}$ is the value calculated from V_+ from the dashed line in Fig. 2. Also shown is the constant offset in doping, Δp , between the doping curves shown in Fig. 3.

x	y	V_{Cu}	V_+	$T_{c,max}^{\text{obs}}$ (K)	$T_{c,max}^{\text{calc}}$ (K)	Δp (from TEP)
0	7.1	2.064	-0.1152	41	96	0.027
0.1	7.1	2.117	-0.1354	46	94.5	0.021
0.2	7.1	2.111	-0.1634	58	92.3	0.013
0.3	7.1	2.209	-0.2634	72	84.6	0.0066
0.3	6.9	2.170	-0.2480			
0.3	6.4	2.114	-0.2866			
0.4	7.1	2.252	-0.3027	81	81.6	0

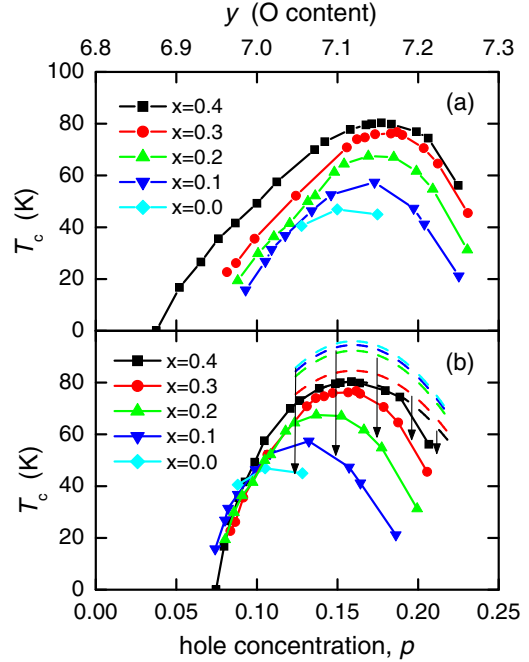


FIG. 2. (Color online) (a) T_c plotted as a function of oxygen content y in $\text{Ca}_x\text{La}_{1-x}\text{Ba}_{1.75-x}\text{La}_{0.25+x}\text{Cu}_3\text{O}_y$ for each of the families $x = 0, 0.1, 0.2, 0.3,$ and 0.4 (reproduced from [16]). (b) T_c plotted as a function of doping p determined from the TEP as described in the text. The peak moves rapidly to lower doping as x is decreased. Dashed curves show the parabolic curves $T_c = T_{c,max} [1 - 82.6(p - 0.16)^2]$ for each family [2] where $T_{c,max}$ is determined from the V_+ correlation shown in Fig. 1. Arrows show anomalous suppression of T_c .

dome-shaped curves for each x where $T_{c,max}(x)$ decreases as x decreases. The reduction is presumably not due to disorder scattering because, as expected, disorder decreases as x decreases. This is evidenced by high-resolution x-ray diffraction [16] and NMR linewidths [17].

This data has been interpreted under the assumption that $T_{c,max}$ for each x occurs at optimal doping, generally taken to be $p \approx 0.16$ holes/Cu [2]. Moreover, by scaling the doping state on the abscissa with respect to a stretching parameter, $K(x)$, and by scaling $T_c, T_{\text{Néel}},$ or J on the ordinate with respect to $T_{c,max}$, then the varied phase diagrams for each x value appear to collapse to a single universal phase diagram [11,12]. However, this result is at odds with other cuprates and the problem, as noted, seems to be with the T_c values, not the J values.

A closer examination shows that T_c is progressively suppressed in this system as x is reduced. This becomes apparent when we consider the BVS, V_{Cu} , for a fixed value of $y = 7.1$ as x is varied. Table I reveals that the doping of the planes steadily increases with x even though y is fixed and the system is therefore isoelectronic. Because of the isoelectronic doping this residual charge transfer must come from the chain layer or apical oxygen and indeed is precisely what is expected from purely electrostatic considerations. Thus the inference of Wulferding *et al.* [11] and Ofer *et al.* [12] that the doping is fixed at the maximum in the phase curves is manifestly not satisfied. If we were to plot T_c as a function of p the curves for smaller x must be displaced to lower doping relative to the curves for higher x . It is our task now to make this conversion explicit.

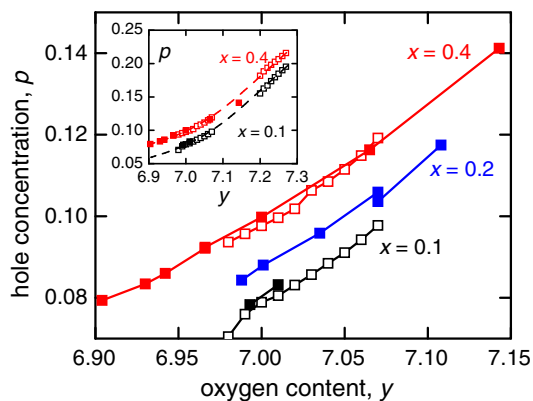


FIG. 3. (Color online) Hole concentration, p , calculated from the thermoelectric power data in Refs. [19] and [20] plotted as a function of oxygen content, y , for $x = 0.1, 0.2$, and 0.4 . The inset shows the data for $x = 0.1$ and 0.4 over a wider range. Note the parallel shifts.

Values of V_{Cu} can only be used as a guide to the trend in doping state. More reliably the thermoelectric power (TEP) can be used as a measure of doping [18]. Fortunately there is TEP data available on this system [19,20] and it is entirely consistent with the BVS. We convert TEP data from these two sources to doping, p , using the equations reported in Tallon *et al.* [2] and plot p versus y in Fig. 3. The inset shows a wider range of y values.

Two things are evident:

(i) The data for each x are simply offset from each other by a constant p value that scales with x . The inset shows this is satisfied over a very broad range of oxygen content where the two fit curves are identical except for the offset of $\Delta p = 0.021$. Thus decreasing x at constant y results in a decreasing p —as we have claimed and contrary to previous articles [11,12]. Key to the approach of these authors was the inference that the relationship between y and p involves an x -dependent “stretching parameter,” $K(x)$, but Fig. 3 shows that these doping curves are simply shifted by a constant offset.

(ii) For $y < 7.05$ the relationship between y and p is linear with slope $\Delta p / \Delta y \approx -0.21$ as was found previously for $Y_{1-x}Ca_xBa_2Cu_3O_y$ [2] but when $y > 7.05$ the slope increases markedly. The doping efficiency per added oxygen more than doubles. Presumably this is associated with the high percentage of adjacent O(1) and O(5) occupancy for every added oxygen once $y > 7$. This behavior is independent of x and only dependent on y .

Following these results we calculate the doping for each (x, y) pair using the y -dependent fit curve shown in the inset to Fig. 3 for $x = 0.4$. For other x values we subtract the constant offset, Δp , listed in the last column of Table I. Using these p values we replot T_c now as a function of p in Fig. 2(b). For comparison, the dashed curves in Fig. 2(b) show the *nominally* universal phase curve [2]:

$$T_c = T_{c,\max}[1 - 82.6(p - 0.16)^2], \quad (1)$$

for each family where $T_{c,\max}$ is found for each x from the V_+ correlation shown in Fig. 2 and is listed in Table I. These curves are only plotted above $p \approx 0.125$ as T_c often dips there

due to CDW formation [5,7]. Several features now emerge that show why this system is anomalous.

First and encouragingly, in contrast to Fig. 2(a), there is now a common onset for superconductivity at low doping. This agrees with an early conclusion made by Goldschmidt *et al.* [20] for this system. In this region we infer that the behavior for all x is “conventional.” Secondly, the peak at $T_{c,\max}$ is seen to be displaced progressively to lower doping (by as much as $\Delta p = 0.055$) as x is reduced. This is fundamentally unlike Ca and La doping in $YBa_2Cu_3O_y$ [6]. For these lower x values, T_c is already reduced well below the dashed curves before reaching the conventional “optimum doping” at $p = 0.16$ (see arrows). While conventional behavior extends to optimal doping and beyond for $x = 0.4$, for smaller values of x superconductivity seems to be pathologically suppressed at progressively lower doping as x decreases. It is these systematics that lie at the heart of the anomalous behavior in this system.

If there were any doubt that this x -dependent shift in doping occurs we note that Knizhnik *et al.* [19] also report the resistivity for different y values in the $x = 0.1$ sample. At maximum T_c (nominally optimal doping) the resistivity shows the canonical pseudogap-induced downturn at $T^* = 160$ K. From the T^* versus p curve [21] this indicates a doping state of $p \approx 0.145$, actually well below optimal doping ($p_{\text{opt}} = 0.16$) and numerically consistent with the TEP data.

To this we can add that Knizhnik *et al.* [19] also report the effect of impurity scattering due to substituting Ni. We have previously reported that the phase curves for increasing impurity concentration collapse asymmetrically down the pseudogap T^* line [22,23] and the last point of superconductivity lies at critical doping, $p_{\text{crit}} = 0.19$ where $T^* \rightarrow 0$. Precisely the same behavior is seen for Ni substitution in CLBLCO [19] with the phase curve collapsing down asymmetrically in the T - y plane to vanish at $y_{\text{crit}} = 7.225$. From the inset to Fig. 3 we find this corresponds to $p_{\text{crit}} = 0.193$ in excellent agreement with our previous reports. Finally, the TEP data of Goldschmidt [20] also confirms that, for a fixed y ($= 7.00$), doping increases with x . Thus, $x = 0.1 \rightarrow p = 0.0784$, $x = 0.2 \rightarrow p = 0.0880$, and $x = 0.4 \rightarrow p = 0.0997$. In summing up these authors state “this proves unambiguously that *doping takes place under isoelectronic conditions*”—just as we assert here. This seems to disagree with x-ray absorption (XAS) studies which suggest a constant doping level for fixed y , independent of x [16]. We can only note that XAS here measures the combined contribution of the CuO_2 planes and the apical oxygen.

We thus conclude that, as x decreases, there is a progressive pathological suppression of the peak in the $T_c(p)$ phase curve which, as a consequence, is progressively displaced to lower doping. As such there can be no meaningful reduced phase curve in this system. This suggests that some electronic instability or scattering process is responsible for suppressing T_c at progressively lower doping as x is reduced. (This instability is not, for example, the above-noted CDW because this is clearly pinned rigidly just to $p \approx 0.125$ [6,7]). In its absence, $T_{c,\max}$ is expected to increase with increasing ion size. It becomes evident that the apparent correlation of $T_{c,\max}$ with J in this system is merely fortuitous.

The cause of the suppression of T_c is not yet obvious. But note that, for each x value, T_c begins to decline when

$y \geq 7.1$. This also coincides with the increase in slope of p versus y already alluded to. It could also be significant that the nuclear quadrupole resonance linewidth increases abruptly above $y = 7.1$ in this system [24]. A final point of possible relevance is the unusually proximate vHs in this system. In $\text{YBa}_2\text{Cu}_3\text{O}_y$ the antibonding band vHs lies close to $p = 0.28$ in the heavily overdoped region just beyond the T_c dome [25]. The T -dependent TEP may be used to locate the vHs from the last positive excursion of the TEP at low T [26]. For CLBLCO with $x = 0.4$ the crossover in sign occurs between $y = 7.217$ and $y = 7.253$ [19], more or less corresponding to the rightmost data point (black square) in Fig. 2—in the lightly overdoped region. Electronic instability can occur near a vHs and indeed the T_c of this last data point already sits low (see arrow). Also, T_c is expected to fall sharply beyond a vHs due to the rapid fall in DOS as the Fermi surface retreats from the $(\pi, 0)$ antinodes [27]. It is significant that increasing ion size shifts the vHs closer to half filling [28]. Thus as x decreases the already low doping state of the vHs will shift to even lower doping, thus potentially accounting for the overall phenomenology described here. This remains an interesting topic for future study. T -dependent TEP studies for smaller x would be a useful starting point.

Having corrected the doping state for this system we can now revise several other conclusions reported in the literature. Notably, a recent ^{17}O NMR study on this system concluded that the T^* line derived from fitting the T dependence of the Knight shift was not the same for each x value and extrapolated to zero at a doping state significantly beyond the overdoped superconducting phase curve [13]. Accordingly they rejected a quantum critical point (QCP) scenario for the pairing interactions. Leaving aside the point that by fitting the T dependence they are actually extracting the pseudogap energy scale, not a temperature scale, we show here that these conclusions must be reversed once the doping is corrected as above. Figure 4(a) shows T_c and T^* plotted versus y as reproduced from their work. The separate y dependence of T^* for $x = 0.1$ and $x = 0.4$ is evident. However, in Fig. 4(b) we replot the data versus p as calculated above and we also add the four data points for $x = 0.2$ and $x = 0.3$. The combined data, within the scatter, is now fully consistent with a single behavior. Further, Zaanen and Hosseinkhani [29] showed that near a QCP $T^*(p)$ has a sublinear power-law behavior $T^* = T_0(1 - p/p_{\text{crit}})^\alpha$. This is the solid purple curve with exponent $\alpha = 0.85$. While for other reasons we reject a QCP scenario, this T^* data is nonetheless now fully consistent with a single behavior for all x where $T^* \rightarrow 0$ at $p_{\text{crit}} = 0.19$, within the dome. This convergence further validates our analysis.

It now also becomes obvious why recent low- T ARPES measurements [14] on an $x = 0.4$ crystal show an energy gap

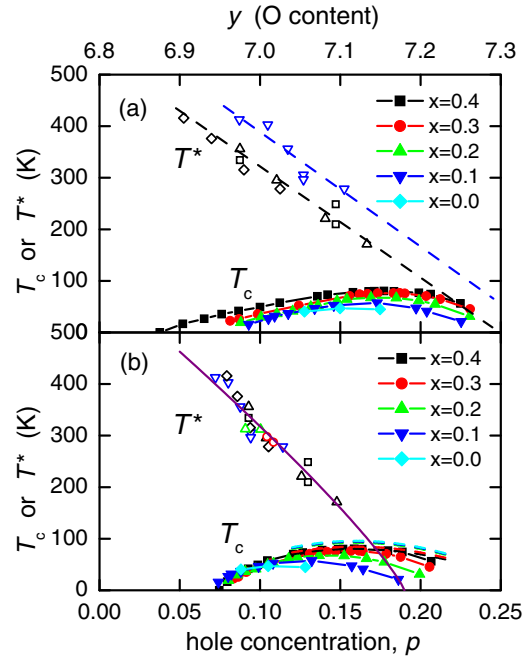


FIG. 4. (Color online) Reproduction of Fig. 1 with T^* values added from the ^{17}O and ^{43}Ca Knight shift [13] in CLBLCO. In (a) $T^*(y)$ is shown for $x = 0.1$ and 0.4 (reproduced from [13]) suggesting two distinct behaviors. But in (b) $T^*(p)$ plotted as a function of doping determined from the TEP reveals a single behavior for all x and is consistent with the required power-law behavior (solid purple curve) for a QCP scenario [29] with $p_{\text{crit}} = 0.19$. Symbol colors indicate x value.

of 40 meV, while for an $x = 0.1$ crystal there is *no gap*. From the Fermi surface area both samples had a doping of $p = 0.23 \pm 0.02$. Examination of Fig. 2(b) shows that this lies beyond the superconducting dome for $x = 0.1$, so there is no gap; but it lies within the dome for $x = 0.4$ for which there must be a gap—just as observed.

In conclusion, we have investigated the available data for the much-studied isoelectronic system $\text{Ca}_x\text{La}_{1-x}\text{Ba}_{1.75-x}\text{La}_{0.25+x}\text{Cu}_3\text{O}_y$ and conclude that T_c is anomalously suppressed once y exceeds a critical value. As a consequence, as x is reduced $T_{c,\text{max}}$ falls instead of increasing as it would in the absence of this suppression. The hole concentration at this apparent, but spurious, $T_{c,\text{max}}$ value is progressively reduced from the generic value of $p_{\text{opt}} = 0.16$ for $x = 0.4$ to $p_{\text{opt}} \approx 0.10$ for $x = 0$. As a further consequence, the previously reported correlation of $T_{c,\text{max}}$ with J must also be spurious.

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