PHYSICAL REVIEW B **76**, 174522 (2007)

Saddle-point van Hove singularity and the phase diagram of high-T_c cuprates

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(Received 13 July 2007; revised manuscript received 7 September 2007; published 28 November 2007)

We examine the generic phase behavior of high-T_c cuprate superconductors in terms of a universal van Hove singularity in the strongly overdoped region. Using a dispersion derived from angle-resolved photoemission spectroscopy (ARPES), we solve the BCS gap equation and show that the pairing interaction or pairing energy cutoff must be a rapidly declining function of doping. This result is prejudicial to a phonon-based pairing interaction and more consistent with a magnetic or magnetically enhanced interaction.

DOI: 10.1103/PhysRevB.76.174522 PACS number(s): 74.25.Dw, 74.25.Jb, 74.62.Dh, 74.72.−h

The high-T_c cuprates are remarkable in that, despite their varied atomic structures and consequent variation in bare electronic band structure,[1] they exhibit universal phase behavior. For example, with the exception of $La_{2-x}Sr_xCuO_4$ which seems more prone to stripe instabilities, the thermoelectric power is a universal function of hole concentration p.[2] Moreover, the evolution with doping of both the thermodynamic properties and the spin susceptibility is likewise universal.[3] Most importantly, the overall temperature-doping phase diagram seems to be universal.[4] The common features include the onset of superconductivity at $p \approx 0.05$, the location of the so-called $(1/8)$th anomalies at $p \approx 0.12$, optimal doping at $p \approx 0.16$, critical doping where the pseudogap closes at $p \approx 0.19$, the overall trend of the pseudogap line $T^*(p)$, and possibly the superconductor-metal transition at $p \approx 0.27$. To these, we now wish to add another apparently common feature, namely, the presence of a van Hove singularity (vHs) in the heavily overdoped region.[5]

The $\epsilon(\mathbf{k})$ dispersion for the hole-doped cuprates exhibits a saddle-point singularity, the so-called van Hove singularity, sited at $(\pi,0)$ on the Brillouin zone boundary. Increased hole doping moves the Fermi energy E_F down toward the vHs which it eventually crosses[5] and where the density of states (DOS), for a strictly two dimensional dispersion, diverges. Within a BCS picture, the transition temperature is given by[6]

$$k_B T_c = 1.14\hbar\omega_p \exp\left(-\frac{1}{N(E_F)V}\right), \tag{1}$$

so that if the vHs crossing occurs within the superconducting domain, the exponential dependence on the DOS should result in a local peak in T_c, precisely at the vHs. Of course, the cuprates do exhibit a superconducting phase curve that passes through a peak at optimal doping[4] and there have been many attempts to explain this phase behavior in terms of a vHs crossing there.[7–9] Further, the rise in isotope effect exponent with underdoping was also explored as a consequence of a proximate vHs.[10] Finally, underdoped cuprates exhibit a pseudogap in the normal-state DOS that, amongst other things, causes a strong suppression of the spin susceptibility χ_s at low temperature.[3,11] Some groups sought to explain this suppression in terms of a nearby vHs[12] where the Fermi window, for $T>0$, extends to the far side of the vHs thereby reducing χ_s.

These ideas all failed for various reasons. The value of χ_s near a vHs never falls more than 10% at low temperature, and so the model could never account for the nearly full suppression of χ_s as $T \to 0$.[3,11] Further, angle-resolved photoemission spectroscopy (ARPES) revealed that the vHs at $(\pi,0)$ lay at least 60 meV below E_F at optimal doping and thus could not account directly for the location of the maximum in T_c.

However, we now need to revisit these ideas in view of recent findings. In the case of $La_{2-x}Sr_xCuO_4$, the vHs is known to be crossed in the strongly overdoped region at $p = x = 0.20–0.22$.[13,14] A similar situation occurs with $Bi_2Sr_2CuO_6$,[15] and deeply overdoped $Tl_2Ba_2CuO_6$ also lies close to the saddle-point vHs.[16] We will return to these systems later. They are all single CuO_2 layer compounds. Where there are two CuO_2 layers per unit cell, the weak electronic coupling between the layers lifts the degeneracy of the electronic states in the layers causing split antibonding and bonding bands. The splitting is maximal near $(\pi,0)$ and is about 100 meV there. Surprisingly, it has recently been shown by ARPES measurements that for $Bi_2Sr_2CaCu_2O_8$, the antibonding vHs is crossed in the deeply overdoped region around $p \sim 0.225$.[5] One has therefore to consider the hypothesis that the location of a vHs around $p \sim 0.23$ is general and possibly plays a central role in defining the generic phase curve $T_c(p)$ (along with the pseudogap at p_{crit}). This is given all the more weight by the fact that both single- and double-layer cuprates cross the singularity in the same overdoped region of the phase diagram in spite of the split band in the latter case.

We have explored this hypothesis using an ARPES-derived $\epsilon(\mathbf{k})$ dispersion for $Bi_2Sr_2CaCu_2O_8$ expressed in terms of a six-parameter tight-binding fit[5] listed in Table I. The doping dependence of the resultant density of states at the Fermi level, $N(E_F)$, is shown in Fig. 1(a). Elsewhere, we showed that the entropy and superfluid density calculated from this dispersion agree with the observed magnitude, temperature, and doping dependence of these parameters.[17] Here, we ignore the pseudogap in underdoped samples, and this is a feature that would need to be added in a fuller treatment (though the general conclusions would remain unchanged).

In the superconducting state, we employ a d-wave gap of the form $\Delta_{\mathbf{k}} = \frac{1}{2}\Delta_0 g_{\mathbf{k}}$, where $g_{\mathbf{k}} = \cos k_x - \cos k_y$. The disper-

TABLE I. Bi-2212 bilayer dispersion tight-binding basis functions and coefficients where $\epsilon(\mathbf{k})=\Sigma c_i \eta_i(\mathbf{k})$.

c_i	$\eta_i(\mathbf{k})$
0.183	1
−0.654	$\frac{1}{2}(\cos k_x+\cos k_y)$
0.1236	$\cos k_x \cos k_y$
−0.1317	$\frac{1}{2}(\cos 2k_x+\cos 2k_y)$
0.0083	$\frac{1}{2}(\cos 2k_x \cos k_y+\cos k_x \cos 2k_y)$
0.0212	$\cos 2k_x \cos 2k_y$
0.053	$\pm\frac{1}{4}(\cos k_x-\cos k_y)^2$

sion in the presence of the superconducting gap is given by $E_{\mathbf{k}}=\sqrt{\epsilon_{\mathbf{k}}^2+\Delta_{\mathbf{k}}^2}$ and $\Delta_0(T)$ is determined from the self-consistent weak-coupling BCS gap equation[18]

$$1=\frac{V}{2}\sum_{\mathbf{k}}\frac{|g_{\mathbf{k}}|^2}{E_{\mathbf{k}}}\tanh\left(\frac{E_{\mathbf{k}}}{2k_BT}\right). \quad (2)$$

For a justification of this Fermi liquid approach, see Ref. 17. We adopt a pairing potential of the form $V_{\mathbf{k}\mathbf{k}'}=Vg_{\mathbf{k}}g_{\mathbf{k}'}$. The summation in Eq. (2) extends over all states in both bands up to a constant energy cutoff, chosen initially to be $\omega_c=70$ meV, and Δ is assumed to be the same for both bands.[19] We consider two cases: (i) the pairing amplitude V is constant, chosen such that $T_{c,max}$ takes the observed value, and (ii) for each p value, V is selected such that $T_c(p)$ follows the experimentally observed, approximately parabolic phase curve. Figure 1(a) shows $T_c(p)$ plotted as a function of hole concentration for the two cases.

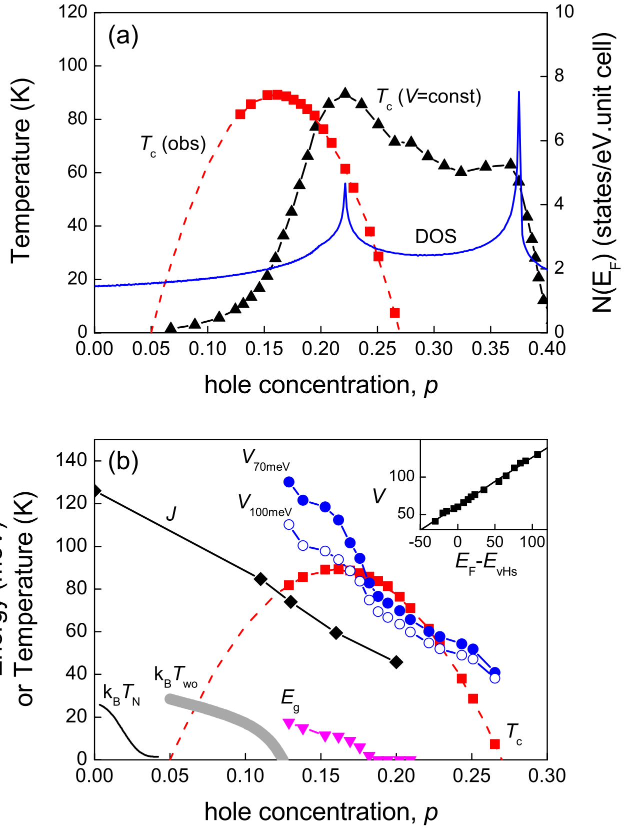

FIG. 1. (Color online) (a) The doping dependence of the DOS $N(E_F)$ and of T_c, as observed (squares) and as calculated (triangles) assuming a constant pairing interaction. (b) The doping dependence of the pairing amplitude V and of the exchange energy J, the pseudogap energy E_g, the NMR wipeout line k_BT_{wo}, and Néel line k_BT_N.

Turning first to the $T_c(p)$ curve for a constant V (up triangles), it is evident that if the pairing interaction or cutoff energy is fixed, then the phase curve (a) is more narrower than that observed, (b) maximizes at the location of the vHs in the heavily overdoped region (not at optimal doping), and (c) exhibits a second peak at the bonding-band vHs. All three difficulties are averted, and the peak broadened and shifted back to the observed optimal doping, only if either the pairing amplitude or the cutoff energy decreases rapidly with doping. This is a robust result independent of the particular details that follow. We illustrate this by the second case explored as follows.

The second $T_c(p)$ phase curve (squares) follows the empirical, approximately parabolic phase curve,[4]

$$T_c=T_{c,max}[1-82.6(p-0.16)^2]. \quad (3)$$

Using the gap equation, we have calculated values of V that reproduce these T_c values. The resultant values of V are shown by the circles plotted as a function of p in Fig. 1(b). They descend rapidly toward zero with increasing doping. If, alternatively, V is held constant and the energy cutoff ω_c is varied,[17] essentially the same result is obtained—a rapidly descending value that vanishes near $p\approx 0.3$. We thus conclude that either the pairing interaction or the energy cutoff falls rapidly with doping. However, the doping independence of the renormalization (or "kink") energy in the dispersion[20] points to the former scenario (constant ω_c, falling V) as being the more likely. In addition, the high energy slope of the dispersion is observed to decrease with doping.[20,21] It is notable that the ratio of the renormalized velocity at E_F to the unrenormalized value well below E_F shows a similar rapid decline with doping.[21] This ratio is a measure of (though overestimates) the electron-boson coupling strength $\lambda=N(E_F)V$. It seems clear therefore from our calculations and ARPES results that the electron-boson coupling strength indeed falls rapidly with doping across the phase diagram.

Such a strongly dependent interaction would not usually be associated with the electron-phonon interaction where the phonon energy scale is only a weak function of doping. These results are, rather, indicative of a magnetic, or magnetically enhanced, mechanism.

Further, the pairing potential V is clearly large and grows with underdoping toward the magnitude J of the exchange interaction. For comparison, we plot in Fig. 1(b) the magnitude of J determined from two-magnon Raman scattering[22] (where J is taken, as usual, as 1/3 the frequency of two-magnon scattering peak). The magnitude and doping dependence of V is very similar to that of J, suggesting a close relationship between these. Also plotted (open circles) is V when $\omega_c=100$ meV. A similar rapid fall is found (and is also

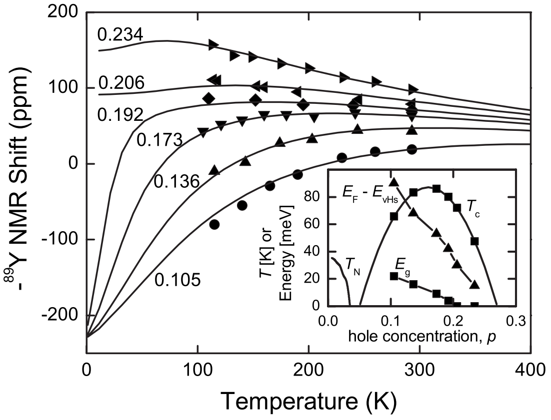

FIG. 2. The ^{89}Y Knight shift for $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$ at the indicated p values. Solid curves are the fits using a two-band dispersion, as described in the text. The inset shows the deduced values of the pseudogap energy E_g and of E_F-E_{vHs} showing that the vHs is crossed in the deeply overdoped region.

found for $\omega_c=125$ meV), thus showing that the choice of ω_c is not too critical.

This rapid fall in energy scales with doping and is also reflected in several other energy scales also shown in Fig. 1(b). These are the pseudogap energy scale E_g and line $k_B T_{wo}$, where T_{wo} is the temperature where NMR intensity wipeout effects are observed, indicating the onset of inhomogeneous spin and charge distribution.[23,24] T_{wo} is for $(Y,Ca)Ba_2Cu_3O_{7-\delta}$.[24] It is also in this region that the 4×4 checkerboard structure is observed in scanning tunneling spectroscopy.[25] These lines all expand out from the antiferromagnetic phase curve $T_N(p)$,[26] like ripples of remanent magnetic effects suggesting a common magnetic origin for these.

Here, the doping is estimated from the parabolic phase curve,[4] which we know to be approximate only. In fact, V is very linear in E_F-E_{vHs}, as shown in the inset of Fig. 1(b). This suggests that the overall phase curve $T_c(p)$ is indeed governed by the proximate vHs combined with a rapidly declining bosonic energy scale. The value of ω_c or V need not vanish at $p\approx0.27$. Eventually, the superconducting energy gap will fall below the pair-breaking scattering rate and T_c will be reduced to zero[27] even if ω_c or V is not quite zero.

If the overall phase diagram in the above-noted single-layer cuprates and in Bi-2212 is controlled by the proximate vHs, then one might expect a similar vHs crossing in strongly overdoped $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$. Here, we now present evidence for this from the T-dependent ^{89}Y Knight shift measurements reported previously by Williams *et al.*[28] and shown in Fig. 2. The Knight shift is linearly related to the spin susceptibility χ_s as follows:

$$^{89}K(T)=a\chi_s(T)+\sigma_c, \tag{4}$$

where σ_c is the T- and p-independent chemical shift. For a Fermi liquid, the spin susceptibility is

$$\chi_s=2\mu_B\int(\partial f/\partial E)N(E)dE. \tag{5}$$

Here, μ_B is the Bohr magneton and $f(E)$ is the Fermi function. We adopt our previous model which we used to model the entropy and superfluid density.[17] Using a tight-binding two-band fit (provided by the authors of Ref. 29) to the electronic dispersion for $YBa_2Cu_3O_{7-\delta}$ we have calculated χ_s and fitted to the Knight shift data shown in Fig. 2. As before,[17] we adopt a Fermi-arc model of the pseudogap and ignore all states in the integral with energy below the k-dependent gap value $E_g(\theta)$. The fits are shown by the solid curves in the figure, and the deduced values of $E_g[=E_g(\theta=0)]$ and E_F-E_{vHs} are plotted in the inset as a function of doping.

Just like S^{el}/T, where S^{el} is the electronic entropy, the Knight shift is observed to exhibit a fanning out behavior as T is decreased, with underdoped samples decreasing (due to the pseudogap) and overdoped samples increasing (due to the proximate vHs).[3,17] In fact, it has previously been shown that $\chi_s\approx a_W S^{el}/T$ over a broad range of T and p for a number of cuprates, with a_W being equal to the Wilson ratio for nearly free electrons.[3] With increasing doping, both $\chi_s(p)$ and $S^{el}(p)/T$ should rise steadily to a peak at the vHs then fall, as has been observed in $La_{2-x}Sr_xCuO_4$.[3] The apparently classical increase in $S^{el}(p)$ of 1 k_B per added hole[3] would appear simply to arise from the increased DOS on approaching the vHs. The inset of Fig. 2 shows that for $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$, the Fermi level will reach the vHs in the deeply overdoped region, just like the other cuprates.

We therefore believe this situation to be universal in the cuprates. It raises the issue as to whether the phase diagram is truly a universal function of hole concentration p or whether it is possibly merely a universal function of the distance away from the vHs, i.e., E_F-E_{vHs}. From our point of view, this would be a significant concession as we have long argued for a universal T-p phase diagram.[4,27] However, it is not straightforward to determine doping levels except in the case of $La_{2-x}Sr_xCuO_4$ and $La_{2-x}Sr_xCaCu_2O_6$, which both follow the universal phase curve.[4] To these, we can add the fully deoxygenated material $Y_{1-x}Ca_xBa_2Cu_3O_6$, where $p=x/2$ exactly. Here, the universality of the phase diagram seems to be preserved[4] and the careful use of bond valence sums also seems to confirm this for $Y_{1-x}Ca_xBa_2Cu_3O_{7-\delta}$ when $\delta<1$.[4]

Turning to other cuprates, very precise measurements of the cation content of oxygen-stoichiometric $Tl_{0.5}Pb_{0.5}Sr_2Ca_{1-x}Y_xCu_2O_7$, $Tl_{0.5}Pb_{0.5}Sr_2Ca_2Cu_3O_9$, and $Tl_2Ba_2Ca_2Cu_3O_{10}$ also confirm the basic phase diagram with optimal doping at $p\approx0.16$.[4] For other cuprates, we have usually estimated the doping state either from the room-temperature thermopower[2] or from the parabolic phase behavior given by Eq. (3). $HgBa_2CuO_{4+\delta}$ is also found to be consistent with both of these correlations.[30] However, we have recently found that the overall doping dependence of the thermopower, $Q(T,p)$, is governed by the approach to the vHs,[31] with the change in sign of $Q(T,p)$ at low temperature occurring precisely at the vHs. Thus, $Q(T,p)$ is primarily governed by $(E_F - E_{vHs})$. The thermopower is therefore only a universal function of p if $(E_F - E_{vHs})$ is a universal function of p. This can be tested by determining the absolute doping state from the area of the Fermi surface A_{pFS}, using the relation[32]

$$p_{FS} = 2\left(\frac{A_{pFS}}{A_{BZ}}\right) - 1, \tag{6}$$

where A_{BZ} is the area of the first Brillouin zone.

Consider a few examples. (i) Firstly, it has already been shown that $La_{2-x}Sr_xCuO_4$ appears to follow the Luttinger theorem and $p_{FS} \approx x$ across the phase diagram;[13] (ii) ARPES measurements on fully oxygenated $YBa_2Cu_3O_{6.993}$, which we have previously identified as having a doping state of $p = 0.19$,[27] reveal that it has a Fermi surface area of 60% of the Brillouin zone corresponding to $p = 0.20$; and (iii) the Fermi surface of Tl-2201 has been measured by angular magnetoresistance oscillations,[33] and for a sample with $T_c = 30$ K, p_{FS} was found to be 0.24 ± 0.02 hole/Cu. Using Eq. (3), its T_c value implies $p = 0.25$. This success looks very promising. However, in the case of Bi-2212, the doping state is a little higher[34] than estimated from thermopower or Eq. (3), and for Bi-2201, it is seriously higher.[15] It is possible, therefore, that the phase diagram is a universal function of $(E_F - E_{vHs})$ rather than of p (though it is often also a universal function of p).

Turning from doping state to phase diagram, we now apply the above results to compare T_c values for several different compounds. In their paper, Kondo *et al.*[32] list the tight-binding parameters for the Bi-2201 energy-momentum dispersion as well as $E_F - E_{vHs}$ and T_c values for several doping levels. From these data, V was calculated assuming a rigid band and $\omega_c = 70$ meV. Having established V as a function of $E_F - E_{vHs}$, we now assume this to be a universal function for the other cuprates and calculate T_c curves using the bilayer Bi-2212 dispersion (Table I) and an ARPES-derived Tl-2201 dispersion.[16] These curves are plotted in Fig. 3 together with the Bi-2201 T_c curve as a function of p_{FS}/p_{opt}. For Bi-2212, the hole concentration was calculated from the average of the bonding and antibonding band Fermi surface areas. Bi-2201, Tl-2201, and Bi2212 have experimentally observed $T_{c,max}$ values of 35 K,[32] 90 K,[35] and 96 K,[36] respectively. Remarkably, the calculated T_c curves display maximal values roughly comparable to these values. The t'/t values are 0.208, 0.417, and 0.189, respectively. Thus, the overall variation is not due principally to trends in t'/t as suggested by Pavarini *et al.*[1] Comparison of the calculated densities of states shown in Fig. 3(b) shows rather that the T_c values correlate with the density of states. In Tl-2201, the dominant increase in DOS appears on the flanks of the vHs, while in Bi-2212, the bilayer splitting leads to an overall increase. $La_{2-x}Sr_xCuO_4$ should provide an exacting test of this idea. Incidentally, this compound with an apparent vHs crossing at $x = 0.20$ (Refs. 13 and 14) looks to be an outlier. However, the Fermi surface area indicates that the $x = 0.22$ sample actually has a higher doping level (Fig. 6 of Ref. 13), and specific heat measurements on ceramic samples suggest the vHs crossing occurs at about $x = 0.23 - 0.24$.[3]

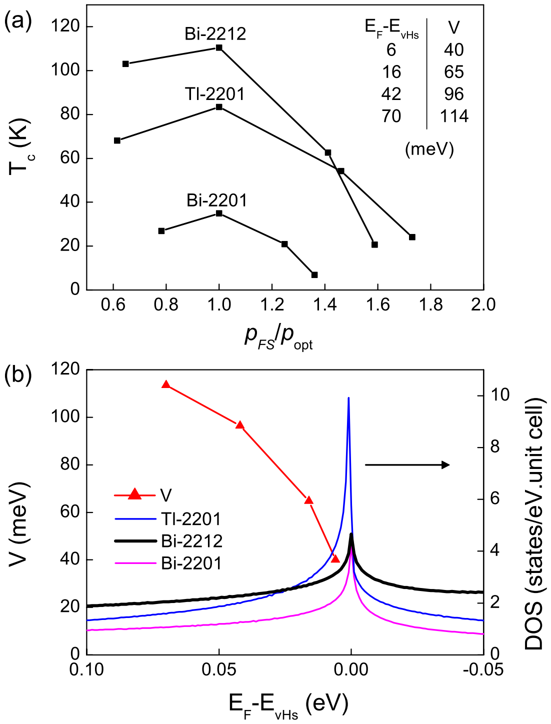

FIG. 3. (Color online) (a) Transition temperatures of different cuprates calculated from their respective ARPES dispersions assuming an identical pairing potential as a function of $E_F - E_{vHs}$. (b) Comparison of the densities of states and a plot of the pairing potential used in (a).

The present calculations strongly suggest that V is a universal energy scale in the single-layer cuprates (and possibly the multilayer cuprates), with the variations in $T_{c,max}$ governed by the comparative magnitudes of the DOS close to the vHs. We can test this further. A rough extrapolation of the curve in Fig. 3(b) gives a value for V at the vHs of about 25 meV. By assuming this value, we calculate T_c for Tl-2201 to be about 7 K at the vHs. This places the vHs very close to where $T_c \rightarrow 0$ for this compound. Interestingly, the thermopower of Tl-2201 seems to confirm this. Generally, the linear part of the thermopower takes the form $S(T) = S_0 - \alpha T$, where $S_0 > 0$ if $E_F - E_{vHs} > 0$ and $S_0 < 0$ if $E_F - E_{vHs} < 0$.[37] In the thermopower data of Ref. 2, the Tl-2201 sample near the edge of the superconducting dome with $T_c < 4$ K appears to exhibit $S_0 = 0$, thus locating it right at the vHs. We therefore have overall consistency which, again, confirms this particular approach on the systematics of the phase diagram and the thermopower.

Finally, we note that close to a vHs, there should be a tendency to structural change or local symmetry reduction so as, for example, to split the vHs and thus to lower the electronic energy. In this regard, it is interesting that most, perhaps all, of the above mentioned high-temperature superconductors tend to be unstable in the heavily overdoped region. Firstly, the vHs is evidently close to the absolute limits of overdoping of Bi-2212 and Y,Ca-123. Our experience is that these systems tend to decompose near these limits, and so we typically have to oxygenate at high oxygen pressures using rather low temperatures (<350 °C) to avoid decomposition.

For a long time, La-214 has been known to phase separate in this overdoped region[38] and only by quenching from high temperature synthesis and reoxygenating (to stoichiometric O=4) at substantially lower temperatures can this phase separation be avoided.[39] These effects are most noticeable in polycrystalline samples where there is a large surface to volume ratio. Single crystals enjoy a metastable state to higher temperatures. Oxygenation of Tl-2201 into the heavily overdoped region results in precipitation of Tl_2O_3 on the surface of single crystals or in grain boundaries of polycrystalline samples. Our attempts to overdope Tl-2201 or $Tl_{0.5}Pb_{0.5}Sr_2CaCu_2O_7$ into this region by Cd substitution for Tl (generally successful in other circumstances) failed. Then, Bi-2201 undergoes a change in electronic state in this same region. These various instabilities have always been a puzzle as has the fact that so many cuprates, as prepared, reside close to optimal doping. The proximate vHs could be the common cause.

In summary, we have used an ARPES-derived dispersion for $Bi_2Sr_2CaCu_2O_{8+\delta}$ and computed the phase curve $T_c(p)$, showing that the only way in which the observed phase curve may be recovered is if the pairing interaction is a rapidly falling function of doping. We find that this closely follows the doping dependence of the exchange interaction J. We also argue that all HTS cuprates exhibit a van Hove singularity in the deeply overdoped region, and this probably underlies the universal phase diagram observed in these systems. Using a universal value for the pairing potential $V = V(E_F - E_{vHs})$, we calculate T_c phase curves for Bi-2201, Tl-2201, and Bi-2212 and find very good agreement with the observed maximum T_c values. This suggests that the differences in $T_{c,max}$ values are governed by the density of states rather than the ratio t'/t, as has been suggested by other authors.[1]

We acknowledge funding from the New Zealand Marsden Fund. We thank A. Kaminski for the tight-binding fits to the electronic dispersion of Bi-2212, and J. W. Loram for helpful comments on the manuscript.

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