Fluctuations and $T_c$ reduction in cuprate superconductors

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We analyse fluctuations about $T_c$ in the specific heat of (Y,Ca)Ba$_2$Cu$_3$O$_{7-δ}$, YBa$_2$Cu$_4$O$_8$ and Bi$_2$Sr$_2$CaCu$_2$O$_{8+δ}$. The mean-field transition temperature, $T^{mf}_c$, in the absence of fluctuations lies well above $T_c$, especially at low doping where it reaches as high as 150K. We show that phase and amplitude fluctuations set in simultaneously and $T^{mf}_c$ scales with the gap, $2\Delta_0$, such that $2\Delta_0/k_B T^{mf}_c$ is comparable to the BCS weak-coupling value, 4.3, for $d$-wave superconductivity. We also show that $T^{mf}_c$ is unrelated to the pseudogap temperature, $T^*$. 

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Many authors have suggested that pairing in the cuprates begins well above $T_c$. Emery and Kivelson argued that the low superfluid density, $n_s$, in the cuprates leads to phase fluctuations below the mean-field (MF) transition temperature, $T^{mf}_c$, resulting in a phase-incoherent state with a finite pairing amplitude. In this view phase coherence is not established until a lower temperature, the observed $T_c$. Experimental support for this picture may be found in the high-frequency optical studies by Corson et al. Additionally, the underdoped normal state (NS) exhibits a depletion of the density of states (DOS) near the Brillouin zone boundary due to the presence of a pseudogap. The pseudogap seems to close abruptly at $p_{crit}$ = 0.19 holes/Cu. Some authors have drawn these two strands together proposing that the pseudogap corresponds to the phase-incoherent pairing state between $T_c$ and $T^{mf}_c$. The pseudogap $T^*$ line (below which pseudogap effects are observed) would then correspond to the doping-dependent $T^{mf}_c$.

Here we analyze the fluctuations in specific heat, $C_p$, to calculate $T^{mf}_c$ and the mean-field jump, $\Delta^{mf}$, in specific heat coefficient, $\gamma$. We find that at all doping levels $T^{mf}_c$ lies well above the observed $T_c$, reaching as high as 113K for (Y,Ca)Ba$_2$Cu$_3$O$_{7-δ}$ (Y,Ca-123) and 150K for Bi$_2$Sr$_2$CaCu$_2$O$_{8+δ}$ (Bi-2212). Our approach is similar to that of Meingast et al. using thermal expansion data. But, where they identified the pair-fluctuating state with the pseudogap, we show they are distinct. Further, we find that $T^{mf}_c$ and $T^*$ are very different in magnitude and doping dependence, confirming that they are unrelated. Over a wide doping range $T^{mf}_c$ scales with the superconducting gap such that $2\Delta_0/k_BT_c$ remains comparable to the weak-coupling BCS $d$-wave value of 4.3.

Even without such an analysis the idea that the pseudogap is a phase incoherent pairing state faces an insurmountable obstacle. If the pseudogap arises merely from thermal phase fluctuations then at $T$=0 there should be no remnant pseudogap effects. But, even at $T$=0 the pseudogap weakens the SC ground state, abruptly reducing the condensation energy and superfluid density, as doping is reduced below $p_{crit}$. The pseudogap thus coexists with SC at $T$=0 and must be distinct from fluctuation effects above $T_c$.

Firstly, we show that phase and amplitude fluctuations set in simultaneously. Emery and Kivelson deduce that phase fluctuations become important above a temperature $T_0$ given by $k_BT_0 \sim AV_0$ where $A \approx 1$ and $V_0$ is the phase stiffness, $V_0 = \hbar^2 n_s(0)/4m^*$. The length scale, $a$, was defined as $a = \sqrt{\pi \xi}$ for isotropic 3D behavior and $a = \max(d, \sqrt{\pi \xi_{ab}})$ for 2D where $d$ is the mean interlayer spacing. $V_0$ is related to the penetration depth, $\lambda_{ab}$, via:

$$\lambda_{ab}^{-2} = \mu_0 c^2(n_s(0)/m^*) = \left(\frac{4\mu_0 c^2}{\hbar^2}\right) V_0. \quad (1)$$

The condensation energy, $U_0$, is given by

$$U_0 = \frac{1}{2} k_B T_c^2 = \frac{1}{4} \mu_0 \left( \frac{1}{2\pi m^*} \right)^2 \left( \frac{\phi_0}{\lambda_{ab} c_{ab}} \right)^2, \quad (2)$$

where the last equality comes from Ginzburg-Landau theory. Combining these, we find

$$k_BT_0 \sim AV_0 = (4A/\pi) U_0 \Omega(0), \quad (3)$$

where $\Omega(0) = \pi \xi_{ab}^2$ is the coherence volume for Cooper pairs. Following Bulaevskii we adopt the criterion for amplitude fluctuations as

$$k_BT_{amp} \sim U_0 \Omega(0), \quad (4)$$

which leads immediately from Eq. 3 to the relation

$$k_BT_0 \sim AV_0 = (4A/\pi) U_0 \Omega(0) \sim (4A/\pi) k_BT_{amp}. \quad (5)$$

As $A \approx 0.9$ for 2D then the conditions for phase and amplitude fluctuations are equally restrictive. For a homogeneous system they both must set in simultaneously. We thus question the widely-accepted phase-fluctuation model of Emery and Kivelson and its implementation by Corson et al. If $T_0$ and $T_{amp}$ greatly exceed $T^{mf}_c$ then the transition occurs essentially at $T^{mf}_c$. But, if $T_0$ and $T_{amp}$ are comparable to or less than $T^{mf}_c$ (as is the
case) then $T_c$ will be suppressed below $T_{mf}^c$. Between $T_c$ and $T_{mf}^c$ both amplitude and phase will fluctuate, not just the phase. It is our aim to determine how large this $T_c$ suppression is.

The fluctuations in $C_P(T)$ have been analyzed by separating $C_P$ into a symmetric fluctuation term, $C_{P}^{\gamma f}$, and an asymmetric MF term, $C_{P}^{mf,0}$. In the 3D-XY model $C_P$ near $T_c$ may be approximated by

$$\Delta C_{P} = \begin{cases} 
A^{-} \ln |t| + \Delta C_{P}^{mf,0} & (t \equiv (T/T_{c} - 1) < 0) \\
A^{+} \ln |t| & (t > 0).
\end{cases}$$

(6)

$\Delta C_{P}^{mf,0}$ is the MF step at $T_c$ and $A^{-} \approx A^{+} = 4k_B/(\pi n^2 \Omega(0))$ [11]. While Eq. (6) is not strictly correct deep in the critical region it captures all the main physical features of the more complex crossover from critical to MF behavior [12] and e.g. accurately represents the critical behavior of He$^4$ at the superfluid transition [9]. A plot of $\Delta C_{P}$ versus $\ln |t|$ gives two parallel lines offset by $\Delta C_{P}^{mf,0}$. In practice this plot exhibits negative curvature for sufficiently small $|t|$ due to minor transition broadening. The effect of the resulting spread in $T_c$ was modeled by replacing $t$ by $t^* = (t^2 + \Delta t^2)^{1/2}$ in the above expressions for $\Delta C_{P}$ [13].

For Bi-2212 $\Delta C_{P}^{mf,0}$ was found to collapse rapidly with the opening of the pseudogap at $p_{crit}$ falling to zero near optimal doping $p = 0.16$ holes/Cu [10]. Below this, $C_P(T)$ is dominated by fluctuations alone and is symmetrical about $T_c$. This is puzzling because the specific heat jump should remain finite, consistent with the second-order phase transition. We resolve this below.

Fig. 1(a) shows $\gamma(T)$ reported by Loram et al. [6] for $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$ with $\delta = 0.25$ and $p = 0.186$. The dashed line is $\gamma_n(T)$. Because entropy $S = \int \gamma dT$ there are two entropy balance conditions: (i) the area abc equals the area cde. This helps to establish the T-dependence of $\gamma_n$ below $T_c$. In this case there is no pseudogap and $\gamma_n(T)$ is constant across the entire $T$-range. For lower doping where the pseudogap is present we use a triangular gap with $\gamma_n$ increasing with temperature [3]:

$$\gamma_n(T) = \gamma_n(\infty) \left[1 - \vartheta^{-1} \tanh(\vartheta) \ln(\cosh(\vartheta))\right],$$

(7)

where $\vartheta = E_g/2k_BT$. The second entropy balance condition concerns the fluctuation term which reduces $T_c$ below $T_{mf}^c$. Thus, (ii) the entropy equal to the forward cross-hatched area between $T_c$ and $T_{mf}^c$ equals the fluctuation entropy given by the backward cross-hatched area under the fluctuation term, $\gamma_f$, which includes both critical and Gaussian fluctuations. That is

$$S^{\gamma f} = \int_{0}^{\infty} \gamma_f dT = \int_{T_c}^{T_{mf}^c} (\gamma_{mf} - \gamma_n) dT,$$

(8)

This construction enables $T_{mf}^c$ to be estimated. Furthermore, the apparent mean-field step, $\Delta \gamma_{mf,0}$, at $T_c$ is also smaller than the (hypothetical) mean-field step, $\Delta \gamma_{mf}$, that would occur at $T_{mf}^c$. These jumps are defined in the figure. (The same superscript notation is used for the jumps in specific heat $\Delta C_{P}^{tot}$, $\Delta C_{P}^{mf,0}$ and $\Delta C_{P}^{mf}$).

We proceed as follows. We combine the first entropy condition with Eq. (7) to establish $\gamma_n(T)$. We then plot $C_P$ above and below $T_c$ using Eq. (6) to determine $\Delta C_{P}^{mf,0}$ and hence $\Delta \gamma_{mf,0}$ [10]. We then construct a power-law fit to $\gamma_n(T)$ at low $T$ that reproduces this value of $\Delta \gamma_{mf,0}$. This is $\gamma_{mf}(T)$ which is slightly superlinear consistent with the predominant d-wave gap structure. Lastly, we impose the second entropy condition (Eq. (8) to deduce $T_{mf}^c$ and $\Delta \gamma_{mf}$. Naturally there are errors inherent in each a construction. But while they grow with underdoping they do not impact on any of our conclusions.

In the example shown in Fig. 1(a) $T_c$ is reduced by (14.7 ± 1.5) K below $T_{mf}^c$. At the same time $\Delta \gamma_{mf}(2.31 ± 0.08)$ mJ/g.at.K$^2$, significantly more than $\Delta \gamma_{mf,0}=1.51$ mJ/g.at.K$^2$. The analysis was carried out for many sam-
for Bi-2212, reflecting the larger anisotropy in the latter doping levels. Underdoped samples show a reduction in \( \Delta_0 \) (red open triangles), and pseudogap \( E_g \) (blue crosses) from ref. [6] are also plotted, scaled by the factor \((1/2.5k_B)\). (c) also shows \( \Delta_0 \) values for Bi-2212 from \( B_{1g} \) Raman (red open triangles). (e) shows \( T_c, T^*_{c,mf} \) and \( T^* \). [21].

FIG. 2: (Color online) The doping dependence of evaluated parameters. (a) and (b) show \( T_c, T^*_{c,mf} \), \( \Delta_0 \) and \( \Delta_{c,mf} \) for \( Y_{0.8}Ca_{0.2}Ba_2Cu_3O_7-\delta \). In (a) values are also shown for \( Bi_2Sr_2CaCu_2O_{8+\delta} \) (arrows). The SC gap, \( \Delta_0 \) (red open triangles), and pseudogap \( E_g \) (blue crosses) from ref.[6] are also plotted, scaled by the factor \((1/2.5k_B)\). (c) also shows \( \Delta_0 \) values for Bi-2212 from \( B_{1g} \) Raman (red open triangles). (e) shows \( T_c, T^*_{c,mf} \) and \( T^* \). [21].

Panel (d) shows \( \Delta_{p,0} \) and \( \Delta_{p,mf} \) values for (Y,Ca)-123 from infra-red conductivity[17] we obtain \( 2\Delta_0/k_BT^*_{c,mf} \approx 4.2 - 4.4 \) in even better agreement with the BCS weak-coupling value. In fact, it is probably fair to state that gap magnitudes are not known sufficiently accurately to discount precise agreement with the weak-coupling value.

Fig. 1(b) shows a similar fluctuation analysis on unpublished \( \gamma(T) \) data for \( YBa_2Cu_3Zn_4O_8 \) with 0, 2 and 4\% Zn on the planar Cu sites. This shows the rapid suppression of both \( T_c \) and the jump in specific heat due to impurity scattering. The high rate of suppression \( dT_c/dx = 13 \text{ K/\%Zn} \) is typical of underdoped cuprates and reflects the presence of the pseudogap which enhances the pairbreaking scattering rate due to the reduced DOS [18]. We use Eq. (7) to fit the pseudogapped

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**Note:** The image contains multiple figures (a) to (e) which are not transcribed here due to the complexity and visual nature of the figures. The text describes the analysis of various parameters such as the SC gap, pseudogap, and mean-field gaps in different copper-oxide materials with different oxygen contents. The figures illustrate the doping dependence of these parameters, showing how they change with doping levels. The text also discusses the implications of these gaps and their effects on the superconducting transition temperatures. 

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**Key Points:***

- The doping dependence of parameters such as \( T_c \), \( \Delta_0 \), \( E_g \), and \( \Delta_{p,0} \) is shown through various panels (a) to (e).
- The SC gap, \( \Delta_0 \), and pseudogap, \( E_g \), are plotted in Fig. 2(a) along with values of \( \Delta_{c,mf} \) and \( \Delta_{p,mf} \) for (Y,Ca)-123.
- The pseudogap was implemented as before using a finite-Fermi-arc model.
- The shift in Fermi-arc opening and the van Hove singularity and pseudogap are discussed.
- Several key conclusions are drawn, including the role of fluctuation effects on the pseudogap.

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**Footnotes:**

1. Meingast et al. 2. Previous reports 3. YBa2Cu3O6.97 4. 92.9K 5. 112.3K 6. Previously reported 7. YBa2Cu3O8 8. YBa2Cu3Zn4O8 9. Tc 10. E gamma 11. Tc,mf 12. E g 13. Tc 14. Mg 15. YBa2Cu3O6.97 16. YBa2Cu3Zn4O8
$\gamma_n(T)$ (shown by the dashed red curve) and the fit is confirmed by the 2% and 4% curves for $\gamma(T)$ in Fig. 1b) for which the NS values coincide with the dashed red curve. The upturns in $\gamma(T)$ at low $T$ are due to a small fraction of impurity and need not concern us.

Next, the values of $\gamma_m^m f(T)$ (blue curve) are determined by fitting a power law to $\gamma_m(T)$. The complication of the upturn in the experimental data at low $T$ is averted by insisting on entropy balance such that the area between the dashed red curve and blue curve below the crossing temperature, $T_{cross} = 61.2K$, equals the area between the black and dashed red curves above $T_{cross}$. We thus obtain $T_m^m f = 91.92K$ from $T_c = 81.00K$, while $\Delta \gamma_T^{\text{tot}} = 1.11, \Delta \gamma_m^{\text{f.o}} = 0.90$ and $\gamma_m^{\text{f.o}} = 0.52$ mJ/g.at.K$^2$. The depression in $T_c$ due to fluctuations is $\Delta T_c = 10.92K$, rather less than the value $\Delta T_c = 33.5K$ obtained for Y$_{0.8}$Ca$_{0.2}$Ba$_2$Cu$_3$O$_{7-\delta}$ at the same doping state. This shortfall is probably due to the enhanced superfluid density in YBa$_2$Cu$_3$O$_6$ which, according to Eqs. 1 and 3, will suppress fluctuations. This implies that the gap magnitude is less in the latter compound, perhaps due to the proximity effect between Cu$_2$O$_2$ chains and CuO$_2$ planes which is expected to lower the SC gap magnitude.

There is little data available on the gap magnitude in YBa$_2$Cu$_3$O$_6$ but Jánošsy et al. [11] have carried out precise measurements of the $T$-dependence of the spin susceptibility below $T_c$ using Gd electron spin resonance. They find an excellent MF $d$-wave fit with maximum (antinodal) gap $\Delta_0 = 190K$, giving $2\Delta_0/k_B T_c = 4.75$. However, by referencing to $T_c^m f$ we obtain $2\Delta_0/k_B T_c^m f = 4.14$, now very close to the weak-coupling value.

Finally, Fig. 2(e) compares the various relevant temperature scales, $T_c(p)$, $T_c^m f(p)$ and the pseudogap line $T^*(p)$ for Y$_{0.8}$Ca$_{0.2}$Ba$_2$Cu$_3$O$_{7-\delta}$. A central conclusion is that, contrary to some authors [20] the pseudogap line $T^*(p)$ does not merge on the overdoped side with $T_c(p)$, still less with the more fundamental quantity $T_c^m f(p)$. This question was effectively put to rest several years ago [21] by an extensive study of the resistivity of high-quality thin films of (Y,Ca)Ba$_2$(Cu,Zn)$_3$O$_{7-\delta}$. The combination of Zn substitution and high magnetic fields allowed $T_c$ to be suppressed so as to expose the evolution of $T^*$ below $T_c$. Panel (e) reproduces these values of $T^*(p)$ (blue data points; solid=films; open=sintered). They extend below the unperturbed $T_c$ value, descending towards zero at $p \approx 0.19$. The solid blue curve is a power-law fit consistent with a terminating quantum critical point [22]. Importantly Zn substitution and moderate magnetic fields do not modify $T^*$ [21] while they do suppress $T_c$. In this way it is straightforward to distinguish between pseudogap effects and SC fluctuation effects in the transport properties. $T^*$ evidently dips well below $T_c^m f$. The temperature scales shown in Fig. 2(e) are all of comparable magnitude and it is therefore not surprising that they have been confused in the past.

In summary, we have carried out a fluctuation analysis of specific heat data to determine the mean-field transition temperature, $T_c^m f$, and the mean-field jump in specific heat coefficient, $\Delta \gamma_c^{m f}$. $T_c^m f$ rises rapidly above $T_c$ with decreasing doping, reaching values of about 110K for YBa$_2$Cu$_3$O$_{7-\delta}$ and Y$_{0.8}$Ca$_{0.2}$Ba$_2$Cu$_3$O$_{7-\delta}$, and as high as 150K for Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$. This shows the fundamental importance of fluctuations in the cuprates. $\Delta \gamma_c^{m f}$ remains non-zero across the phase diagram, as it must for a second-order phase transition. The long-standing puzzle that $2\Delta_0/k_B T_c$ grows with reducing doping is resolved by replacing $T_c$ by $T_c^m f$. Across much of the SC phase diagram $2\Delta_0/k_B T_c^m f$ remains close to the weak coupling BCS value. $T^*$ is shown to be distinct from $T_c^m f$.

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