Fluctuations and critical temperature reduction in cuprate superconductors

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We show theoretically that phase and amplitude fluctuations set in simultaneously in the cuprates. We then analyze fluctuations about T_c in the specific heat of (Y,Ca)Ba₂Cu₃O_{7- δ}, YBa₂Cu₄O₈, and Bi₂Sr₂CaCu₂O_{8+ δ}. The mean-field transition temperature, T_c^{mf} , in the absence of fluctuations lies well above T_c and at low doping reaches as high as 150 K. T_c^{mf} is found to be unrelated to the pseudogap temperature T^* , but scales with the gap Δ_0 , such that $2\Delta_0/k_B T_c^{\text{mf}}$ is comparable to the BCS weak-coupling value 4.3, for *d*-wave superconductivity.

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Many authors have suggested that pairing in the cuprates begins well above T_c . Emery and Kivelsen argued that the low superfluid density n_s in the cuprates leads to phase fluctuations below the mean-field (MF) transition temperature, $T_c^{\rm mf}$, resulting in a phase-incoherent state with a finite pairing amplitude.¹ Phase coherence is then established at a lower temperature, the observed T_c . Support for this picture may be found in the high-frequency optical studies by Corson *et al.*² 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 which seems to close abruptly at $p_{crit} = 0.19$ holes/Cu.³ It is often proposed that the pseudogap corresponds to the phase-incoherent pairing state between T_c and $T_c^{\text{mf.}4,5}$ The pseudogap T^* line (below which pseudogap effects are observed) would then correspond to $T_c^{\rm mf}(p)$.

Here we analyze the superconducting (SC) fluctuations in the electronic specific heat for several cuprates, from which we calculate T_c^{mf} and the MF jump $\Delta \gamma^{\text{mf}}$ in the electronic specific heat coefficient $\gamma = C_P/T$. We find that at all doping levels T_c^{mf} lies well above the observed T_c , reaching as high as 113 K for Y, Ca-123 and 150 K for Bi-2212. Our approach is similar to that of Meingast *et al.*⁵ using thermal expansion data. But, where they identified the pair-fluctuating state with the pseudogap, we show they are distinct.

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 density⁶ 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 .

We use a high-resolution differential technique⁷ to directly measure the difference in the specific heats of each SC sample and a closely related but non-SC reference sample. Typically the reference is a Zn-substituted sample. The bulk of the large phonon term is thereby backed off leaving a residual phonon term that exhibits a broad peak at 39 K in (Y,Ca)Ba₂Cu₃O_{7- δ} (Y,Ca-123)⁷ and 17 K in Bi₂Sr₂CaCu₂O_{8+ δ} (Bi-2212)⁸ which scales in magnitude with oxygen content. This is the only significant *T*-dependent correction required to determine the absolute electronic specific heat. The sample is successively annealed and quenched to alter the oxygen content and, for each doping state, the differential specific heat is then differenced against the lowest doping state and scaled by the change in oxygen content measured by mass change. This allows the residual phonon term to be determined with an accuracy better than 0.2 mJ/g.at.K^2 at the peak. Differences in absolute values between doping states are certainly less than this. And, because the residual is peaked at low temperature, and broad, uncertainties in this term have little effect on the following analysis of the SC fluctuations which peak sharply at T_c .

We start by arguing that phase and amplitude fluctuations set in simultaneously. Emery and Kivelsen¹ deduced that phase fluctuations become important when $T > T_{\theta}$ where $k_B T_{\theta} \sim AV_0, A \sim 1$, and V_0 is the phase stiffness, $V_0 = a\hbar^2 n_s(0)/4m^*$. The length scale *a* was defined as $a = \sqrt{\pi\xi}$ for isotropic three-dimensional (3D) behavior and $a = \max(d, \sqrt{\pi\xi_{\perp}})$ for two-dimensional (2D) where *d* is the mean interlayer spacing. V_0 is related to the penetration depth λ_{ab} , viz.

$$\lambda_{\rm ab}^{-2} = \mu_0 e^2 [n_s(0)/m^*] = \left(\frac{4\mu_0 e^2}{a\hbar^2}\right) V_0. \tag{1}$$

The condensation energy U_0 is given by

$$U_0 = \frac{1}{2}\mu_0 H_c^2 = \frac{1}{4}\mu_0 \left(\frac{1}{2\pi\mu_0}\right)^2 \left(\frac{\phi_0}{\lambda_{ab}\xi_{ab}}\right)^2, \qquad (2)$$

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

$$k_B T_\theta \sim A V_0 = (4A/\pi) U_0 \Omega(0), \qquad (3)$$

where $\Omega(0) = \pi \xi_{ab}^2 a$ is the coherence volume for Cooper pairs. Following Bulaevskii⁹ we adopt the criterion for amplitude fluctuations as

$$k_B T_{\rm amp} \sim U_0 \Omega(0), \tag{4}$$

which leads immediately from Eq. (3) to the relation

$$k_B T_{\theta} \sim A V_0 = (4A/\pi) U_0 \Omega(0) \sim (4A/\pi) k_B T_{\text{amp}}.$$
 (5)

As $A \approx 0.9$ for $2D^1$ 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 Kivelsen¹ and its implementation by Corson *et al.*² If T_{θ} and T_{amp} greatly exceed T_c^{mf} then the transition occurs essentially

at T_c^{mf} . But, if T_{θ} and T_{amp} are comparable to or less than T_c^{mf} (as is the case) then T_c will be suppressed below T_c^{mf} . Between T_c and T_c^{mf} both amplitude and phase will fluctuate. It is our aim to determine how large this T_c suppression is.

The fluctuations in C_P have been analyzed^{10,11} by separating C_P into a fluctuation term C_P^{fl} , which is symmetric about T_c and an asymmetric MF term $C_P^{\text{mf},0}$. In the 3D-XY model C_P near T_c may be approximated by

$$\delta C_P = \begin{cases} A^{-} \ln |t| + \Delta C_P^{\text{mf},0}, & [t \equiv (T/T_c - 1) < 0], \\ A^{+} \ln |t|, & (t > 0). \end{cases}$$
(6)

 $A^- \approx A^+ = 4k_B/[9\pi^2\Omega(0)]$ (Ref. 12) and $\triangle C_P^{\mathrm{mf},0}$ is the MF step at T_c . 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.¹³ For example, it accurately represents the critical behavior of He⁴ at the superfluid transition.¹⁰ A plot of C_P versus $\ln |t|$ gives two parallel lines offset by $\triangle C_P^{\mathrm{mf},0}$. 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 modeled¹⁴ by replacing t by $t^* = (t^2 + \triangle t^2)^{1/2}$ in Eq. (6).

For Bi-2212, $\Delta C_p^{\text{mf},0}$ 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 (Ref. 11). 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 anomaly below.

Figure 1(a) shows $\gamma(T)$ reported by Loram *et al.*¹⁵ for $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$ with $\delta = 0.25$ and p = 0.186. The dashed line is $\gamma_n(T)$, the NS $\gamma(T)$. Because the electronic 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 γ_n below T_c . In this case there is no pseudogap and $\gamma_n(T)$ is constant. For lower doping where the pseudogap is present we use a triangular gap which fills with increasing temperature:³

$$\gamma_n(T) = \gamma_n(\infty)[1 - \vartheta^{-1}\tanh(\vartheta)\ln(\cosh(\vartheta))], \qquad (7)$$

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

$$S^{\rm fl} = \int_0^\infty \gamma^{\rm fl} dT = \int_{T_c}^{T_c^{\rm mf}} \left(\gamma_s^{\rm mf} - \gamma_n \right) dT. \tag{8}$$

This construction enables T_c^{mf} to be estimated. Furthermore, the apparent MF step $\Delta \gamma^{\text{mf},0}$ at T_c is also smaller than the "true" MF step $\Delta \gamma^{\text{mf}}$ that would occur at T_c^{mf} in the absence of fluctuations. These jumps are defined in the figure. (The same superscript notation is used for $\Delta C_P^{\text{mf},0}$, $\Delta C_P^{\text{mf},0}$, and ΔC_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 $\Delta C_P^{\text{mf},0}$ and



FIG. 1. (Color online) (a) Analysis of $\gamma(T)$ for $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{6.75}$ showing the deduced MF component γ_s^{mf} and NS value γ_n . Dashed (magenta) curve is the BCS γ_s^{mf} for $2\Delta_0/k_BT_c^{\text{mf}} = 5$. (b) A similar analysis for YBa₂Cu₄O₈ showing γ_s^{mf} (blue dash-dot curve) and the pseudogapped $\gamma_n(T)$ (red dashed curve). The $\gamma(T)$ curves for 2% Zn (cyan solid line) and 4% Zn (green solid line) coincide with $\gamma_n(T)$ and thus confirm our pseudogap model. Upturns at low T are due to impurities.

hence $\Delta \gamma^{\text{mf},0}$.¹¹ We then construct a power-law fit to $\gamma_s(T)$ at low *T* that reproduces this value of $\Delta \gamma^{\text{mf},0}$. This is $\gamma_s^{\text{mf}}(T)$ which is slightly superlinear, consistent with the predominant *d*-wave gap structure. Finally, we impose the second entropy condition [Eq. (8)] to deduce T_c^{mf} and $\Delta \gamma^{\text{mf}}$. There are errors inherent in such a construction, but while they grow with underdoping, as shown, they do not impact on any of our conclusions.

In the example shown in Fig. 1 $T_c = (82.27 \pm 0.3)$ K, $T_c^{\text{mf}} = (97.72 \pm 3.0)$ K, $\Delta \gamma^{\text{mf}} = (2.32 \pm 0.08)$ mJ/g.at.K², $\Delta \gamma^{\text{mf},0} = (1.51 \pm 0.04)$ mJ/g.at.K², and $\Delta \gamma^{\text{tot}} = (3.00 \pm 0.04)$ mJ/g.at.K². For comparison, the dashed magenta curve shows the theoretical *d* wave $\gamma_s^{\text{mf}}(T)$ for $2\Delta_0/k_B T_c^{\text{mf}} = 5$. The agreement is excellent. The analysis was carried out for ten different doping states. Values of T_c and T_c^{mf} are plotted versus *p* in Fig. 2(a) along with values of $\Delta \gamma^{\text{mf}}$ and $\Delta \gamma^{\text{mf},0}$ in Fig. 2(b). A similar analysis was done for Bi-2212 (Ref. 15). Here, instead of using Eq. (7), the full bilayer ARPES dispersion was used,¹⁶ thus incorporating the van Hove singularity and pseudogap. The pseudogap was implemented as before¹⁶ using a finite-Fermi-arc model. The results are plotted in Figs. 2(c) and (d). Several key conclusions can be made.

(i) Like Meingast *et al.*⁵ we find $T_c^{\text{mf}}(p)$ continues to rise with decreasing doping and only falls at the lowest doping levels. Underdoped samples show a reduction in T_c below T_c^{mf} as large as 35–40 K for (Y,Ca)-123 and 60 K for Bi-2212, reflecting the larger anisotropy in the latter compound. The shift is also large for pure YBa₂Cu₃O_{6.97} with $T_c = (92.9 \pm 0.05)$ K and $T_c^{\text{mf}} = (112.3 \pm 0.9)$ K [see arrowed green data points Fig. 2(a)].



FIG. 2. (Color online) The doping dependence of evaluated parameters. Panels (a) and (b) show T_c , T_c^{mf} , $\Delta\gamma^{\text{mf}}$, and $\Delta\gamma^{\text{mf},0}$ for $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$; (c) and (d) show the same for $Bi_2Sr_2CaCu_2O_{8+\delta}$. In (a) values are also shown for $YBa_2Cu_3O_{7-\delta}$ (arrows). The SC gap, Δ_0 (red open triangles), and pseudogap E_g (blue crosses) from Ref. 15 are also plotted, scaled by the factor $(1/2.5k_B)$. Panel (c) also shows Δ_0 values for Bi-2212 from B_{1g} Raman (red open triangles). Panel (e) shows T_c , T_c^{mf} , and T^* .²³

(ii) While $\Delta \gamma^{\text{mf},0} \approx 0$ at lower doping (and the specific heat anomaly then becomes a pure symmetric fluctuation term) $\Delta \gamma^{\text{mf}}$ remains finite in the absence of fluctuations and may only reach zero near the onset of SC at $p \approx 0.05$. This removes the puzzle of the seemingly zero MF step.

(iii) Figs. 2(a) and (c) show the pseudogap energy E_g , as previously reported.^{15,16} Coincident with the abrupt opening of the pseudogap at $p \approx 0.19$ there is an abrupt reduction in all values of $\Delta \gamma$ showing that, even after removing fluctuation effects, the pseudogap still plays a decisive role in weakening the condensate.

(iv) We compare T_c^{mf} with the SC gap, Δ_0 , at T = 0 for the two systems in Figs. 2(a) and (c). For (Y,Ca)-123 values of Δ_0 are from the specific heat¹⁵ and for Bi-2212 from the Raman B_{1g} gap.¹⁷ In both cases $2\Delta_0/k_B T_c^{\text{mf}} \approx 5$ across the entire overdoped region, little more than the *d*-wave MF BCS value of 4.3. The old puzzle that $2\Delta_0/k_BT_c$ increases steadily with decreasing doping¹⁷ is now resolved by referencing to $T_c^{\rm mf}$ rather than T_c . Eventually, at low doping $T_c^{\rm mf}$ falls below $\Delta_0/2.5k_B$ due to the pseudogap progressively removing spectral weight. If it were not for the pseudogap $T_c^{\rm mf}$ would probably track $\Delta_0/2.5k_B$ across the entire SC domain. Thus T_c is reduced both by fluctuations and by the pseudogap and is not the fundamental energy scale. It is the T = 0 *d*-wave gap Δ_0 , which is the truly fundamental quantity and in a BCS scenario $T_c^{\rm mf}$ will scale with Δ_0 until the pseudogap opens, as observed.

(v) Using Δ_0 values for (Y,Ca)-123 from infrared *c*-axis conductivity¹⁸ we obtain $2\Delta_0/k_B T_c^{\text{mf}} \approx 4.2$ -4.4 in even better agreement with the BCS ratio. In fact, gap values are probably not known sufficiently accurately to discount precise agreement with the weak-coupling value.

(vi) The presence of strong fluctuations, in both amplitude and phase, well above T_c implies the persistence of the SC gap above T_c , as has been observed.¹⁹ Indeed, the maximum temperature at which the gap is seen coincides with our deduced T_c^{mf} . It also probably explains the anomalous Nernst effect observed well above T_c (but below T_c^{mf}) in both underdoped and overdoped samples.⁴ The anomalous Nernst effect can then be seen to be unrelated to the pseudogap.

Figure 1(b) shows a similar fluctuation analysis on new $\gamma(T)$ data for YBa₂(Cu,Zn)₄O₈ with 0% , 2%, and 4% Zn on the planar Cu sites. This shows the rapid suppression of both T_c and $\Delta\gamma^{\text{tot}}$ 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.²⁰ We use Eq. (7) to fit the pseudogapped $\gamma_n(T)$ (shown by the dashed red curve) and the fit is confirmed by the 2% and 4% curves for $\gamma(T)$ in Fig. 1(b) 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 $\gamma_s^{\rm mf}(T)$ (blue dash-dot curve) are determined by fitting a power law to $\gamma_s(T)$. 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 dash-dot (blue) curve below the crossing temperature $T_{\text{cross}} = 61.2$ K equals the area between the black and dashed (red) curves above T_{cross} . We thus obtain $T_c^{\text{mf}} = (91.92 \pm 0.75) \text{ K}$ from $T_c = (81.00 \pm 0.09) \text{ K}$; while $\Delta \gamma^{\text{tot}} = (1.11 \pm 0.008)$, $\Delta \gamma^{\text{mf}} = (0.90 \pm 0.029)$, and $\Delta \gamma^{\text{mf},0} = (0.52 \pm 0.005) \text{ mJ/g.at.K}^2$. The depression in T_c due to fluctuations is $\Delta T_c = (10.92 \pm 0.75)$ K, rather less than the value $\Delta T_c = 33.5$ K obtained for Y_{0.8}Ca_{0.2}Ba₂Cu₃O_{7- δ} at the same doping state. This is probably due to the large superfluid density in YBa₂Cu₄O₈ (Ref. 21) which, according to Eq. (3), will suppress fluctuations. This implies that the gap magnitude is less in the latter compound, perhaps due to the proximity effect between Cu_2O_2 chains and CuO_2 planes which will lower the SC gap magnitude.

There are little data available for Δ_0 in YBa₂Cu₄O₈ but Jánossy *et al.*²² 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 $\Delta_0 = 190$ K, giving $2\Delta_0/k_BT_c = 4.75$. By referencing to T_c^{mf} we obtain $2\Delta_0/k_BT_c^{\text{mf}} = 4.14$, now very close to the weak-coupling ratio.

Finally, Fig. 2(e) compares the various relevant temperature scales, $T_c(p)$, $T_c^{\text{mf}}(p)$, and the pseudogap line $T^*(p)$ for Y,Ca-123. Values of T^* are from an extensive study²³ of the resistivity of high-quality epitaxial thin films of $(Y,Ca)Ba_2(Cu,Zn)_3O_{7-\delta}$. 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 . Importantly, Zn substitution and moderate magnetic fields do not modify T^* (Ref. 23) 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. Figure 2(e) reproduces these values of $T^*(p)$ (blue data points; solid = films; open = sintered). They extend below the unperturbed T_c value, descending toward zero at $p \approx 0.19$. The solid (blue) curve is a power-law fit consistent with a terminating quantum critical point.²⁴ We conclude that, contrary to some authors,²⁵ the pseudogap line $T^*(p)$ does not merge on the overdoped side with $T_c(p)$, still less with the more fundamental quantity $T_c^{\text{mf}}(p)$. The temperature scales shown in Fig. 2(e) are all of comparable magnitude so it is 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 MF transition temperature T_c^{mf} and the MF jump in specific heat coefficient, $\Delta \gamma^{\text{mf}}$. T_c^{mf} rises rapidly above T_c with decreasing doping, reaching 110 K for YBa₂Cu₃O_{7- δ} and Y_{0.8}Ca_{0.2}Ba₂Cu₃O_{7- δ}, and as high as 150 K for Bi₂Sr₂CaCu₂O_{8+ δ}. This exposes the fundamental importance of fluctuations in the cuprates. $\Delta \gamma^{\text{mf}}$ remains nonzero across the phase diagram, as it must for a second-order phase transition. The long-standing puzzle that $2\Delta_0/k_BT_c$ grows with reducing doping is resolved by replacing T_c by T_c^{mf} . Across much of the SC phase diagram $2\Delta_0/k_BT_c^{\text{mf}}$ remains close to the weak coupling BCS value. T^* is shown to be distinct from T_c^{mf} .

Since submitting this work a similar entropy analysis has been reported for $Bi_2Sr_{2-x}La_xCuO_{6+\delta}$ (Ref. 26).

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