Electronic inhomogeneity, fluctuations, and thermodynamic transitions in cuprate superconductors

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The apparent electronic inhomogeneity in the cuprate superconductor $Bi_2Sr_2CaCu_2O_{8+\delta}$ inferred from scanning tunneling spectroscopy should induce broadening of the thermodynamic transition. Because observed thermodynamic transition widths are actually rather narrow the question has been raised as to whether the inclusion of fluctuations might sharpen the computed transition for an inhomogeneous system. We have modeled a distributed mean-field step combined with fluctuations and show that the transition is not sharpened by fluctuations. The spread of T_c values inferred from our analysis is significantly less than that implied by pairing inhomogeneity and, along with many other observations, brings into question this interpretation.

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In recent years there has been the development of a widespread belief that the high-T_c cuprate superconductors exhibit gross electronic inhomogeneity. While this has been anticipated on theoretical grounds,[1] and the cuprates generally require extrinsic dopants to introduce carriers, it was the advent of high-resolution scanning tunneling spectroscopy (STS) studies on $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) (Refs. 2 and 3) that rather forcefully brought this issue into focus. These STS studies suggested a $\sim\pm 25\%$ variation in the peak-to-peak gap magnitude on a length scale of a superconducting (SC) coherence length, ξ_0. The locations with large spectral gaps were found to lack well-developed coherence peaks, and they correlate with the locations of oxygen interstitial dopants.[4] Based on the absence of Ni resonances there, these large-gap locations were originally suggested to be nonsuperconducting.[3]

More recently, the alternative view has been adopted, namely, that these variable gaps, including the large gaps without coherence peaks, are all SC gaps.[5] Building on this notion Hirschfeld and co-workers[6,7] modeled this behavior by adopting a spatially inhomogeneous pairing interaction. By solving the local Bogoliubov–de Gennes equations they show that this pairing inhomogeneity reproduces the inhomogeneous gap maps derived from STS. The large gaps are thus presumed to correlate with a locally large pairing interaction. And then, most recently this scenario seems apparently confirmed by STS studies above T_c which reveal a locally inhomogeneous persistence of gap features to a temperature T^* which lies well above T_c and which rises with underdoping.[8] These patches of locally persistent gaps correlate with the large-gap regions in the SC state. The combination of both of these observations makes a compelling case for inhomogeneous pairing interactions, with local pairing persisting to well above T_c in the large-gap regions.

However, despite the impressive achievements of the STS community these conclusions are at odds with many other observations and we have consistently questioned them.[9,10] Further, they are at odds with an important subset of similar STS studies which actually report spatially homogeneous spectra. Because of the fundamental importance of this issue we first assemble the contrary evidence against bulk electronic inhomogeneity near optimal doping, and we then revisit in detail the broadening of the thermodynamic transition and focus on the impact of fluctuations. We conclude that the inhomogeneity envisaged in these remarkable STS papers is probably an artifact and that the fundamental physics of HTS is not essentially linked to inhomogeneity but, rather, occurs within an intrinsically homogeneous electronic environment on the timescale of STS, angle resolved photoemission spectroscopy (ARPES), or NMR studies.

First, we consider ARPES data. It is well known that in the pseudogap regime in underdoped cuprates the quasiparticle peak near $(\pi,0)$ is heavily broadened relative to that near the zone diagonal.[11] This appears to be consistent with the STS data which are spatially inhomogeneous only near the maximum in the *d*-wave gap and not at lower bias near the node. However, conventional ARPES is dominated by the outermost CuO_2 layer[11] while the more recently developed laser ARPES (Ref. 12) probes deeper and, here, the EDS background is much reduced and the quasiparticle peaks are much sharper. Figure 2d of the paper by Koralek *et al.*[12] shows a sharp quasiparticle peak near $(\pi,0)$ recorded at 25 K (below T_c) with a full width at half maximum (FWHM) breadth of 13 meV. Such a narrow peak seems inconsistent with the gap distributions inferred from STS studies with FWHM widths exceeding 20 meV (Ref. 5) around the same doping state. Improvements to the laser ARPES technique are likely to see further narrowing of the quasiparticle peaks.

Turning to other STS studies we note that measurements directly on an exposed CuO_2 plane (i.e., not through the BiO and SrO layers) revealed very homogeneous spectra.[13] This is consistent with the apparent inhomogeneity deriving in some way from the surface BiO/SrO layers containing the dopant atoms and not from the intrinsic electronic state of the CuO_2 planes. Very recent studies by Boyer *et al.*[14] show that there appears to be an inhomogeneous background gap which conceals the SC gap. By dividing out this variable gap obtained at any one location above T_c from the spectra below T_c at the same location, they obtain a highly homogeneous SC gap across the entire field of view, complete with coherence peaks.

Moreover, as noted, an important subset of conventional STS studies (on surfaces capped by BiO layers) actually report spatially homogeneous spectra. The recent review of STS by the Geneva group[15] shows spatially homogeneous spectra for Bi-2212, Bi-2223, and Y123 over a similar scan-

ning distance to that reported by the Davis group.[2,3] (See their figures 17(b)–17(d)). Further, Liu *et al.*[16] showed that even the pseudogap state above T_c can be homogeneous. In a further paper presented at the International Conference on Spectroscopy of Novel Superconductors (Sendai, 20–24 Aug 2007) these authors showed that, even in underdoped Bi-2212, homogeneous spectra could be obtained in both the SC state and the normal state pseudogap region above T_c.[17] Thus some groups report spatially homogeneous spectra while others report spatially inhomogeneous spectra. The conclusion to be drawn from these results is surely that the homogeneous spectra are intrinsic while the inhomogeneous spectra are of extrinsic origin. The reverse scenario is hardly likely. And indeed, as we have noted the large-gap regions in the inhomogeneous spectra correlate with oxygen interstitial dopants.[4]

In this regard intrinsic tunneling would appear to be regrettably overlooked. These studies reveal very sharp coherence peaks.[18,19] Typically involving up to 20 bilayer stacks (40 CuO_2 planes) they are much more of a bulk tunneling technique than is STS. Gross inhomogeneity, on the scale inferred from STS studies, would severely broaden these peaks, and their observed sharpness is indicative of a highly homogeneous state. These features can no longer be dismissed as overheating effects.[20]

Thus whether it is ARPES or STS the deeper is the probe the sharper and more homogeneous are the spectra. A possible resolution of these conflicting results has been proposed by Cooper[21] who suggested that, in defect free regions, the cone of the tunneling current is so narrow that only states around the nodes are probed. Due to Coulomb scattering, tunneling near an oxygen interstitial broadens the cone and thus probes nearer to the antinodes where the gap is larger. Moreover, recent studies in optimal or underdoped samples confirm the presence of two energy scales.[22–24] The SC gap is seen in the nodal region near the zone diagonal, while the pseudogap, which in underdoped samples is much larger, resides at the antinodes. The model of Cooper would suggest then that tunneling near an oxygen interstitial would pick up the large (nonstates-conserving) pseudogap, consistent with the observed absence of coherence peaks for the large gap. A spatially uniform gap structure would thus present an inhomogeneous field of STS spectra that correlates with the locations of oxygen interstitials, as observed.[4] At subgap bias voltages a uniform local density of states (LDOS) would be, and is, observed.[3] Details have yet to be refined (and agreed), but the seeds of a resolution appear to be planted.

To these observations one may add that both NMR (Refs. 9 and 25) and specific-heat[9,26] data strongly support a general absence of gross electronic inhomogeneity in the best samples. We must stress that this is only for doping states above $p=0.125$. Below this doping level there is ample evidence for spin and charge inhomogeneity.[9,10] Above this doping, our analysis shows that the specific-heat transition anomaly in Bi-2212 is sufficiently sharp as to require a quantifiably high degree of homogeneity.[9]

A direct comparison between the STS data and the specific-heat anomaly is available thanks to the calculations of Andersen *et al.*[7] These authors modeled the spectroscopic inhomogeneity by adopting a spatially inhomogeneous pairing interaction in a *d*-wave BCS pairing model. They solved self-consistently for the local gap magnitude which closely mimics the observed STS gap maps. They then computed the specific heat which is, naturally, broadened relative to the standard mean-field (MF) transition. Much can be learned from a detailed comparison of the computed and observed specific heat.

Quite generally,[27] the specific heat near T_c comprises an asymmetric MF step and a nearly symmetric fluctuation term which for Bi-2212 extends some 30 K above and below T_c. Such a broad fluctuation term could be naively interpreted as transition broadening, and indeed Andersen *et al.*,[7] who omitted the fluctuation contribution, mad this association. Well away from T_c their broadened MF transition matches the observed fluctuation specific heat. However, near T_c the observed anomaly is very sharp while the calculated anomaly remains broadened.[26] Aware of this disparity they make the interesting suggestion that the introduction of fluctuations may possibly lead to a sharpening of a broadened MF step. It is easy to see how such a perception may arise. The inclusion of fluctuations increases the apparent step height of the specific heat which can give the illusion of a sharper anomaly. But as we now show it does not narrow the transition.

Our modeling is based on well-established results from microscopic theory: (i) both the MF and fluctuation contributions to the specific-heat anomaly experience the same broadening due to finite-size effects, inhomogeneity, or an applied field;[27,28] and (ii) in granular systems which are not strongly coupled (as here) the anomaly is dominated by the specific-heat peaks associated with each local grain, and the resultant anomaly is obtained by summing over the individual grains.[29] With these features in mind, the effects of broadening are simulated by integrating sharp specific-heat transitions over Gaussian distributions of T_c. The data are then compared with similar plots for some of our Bi-2212 data. To cover a wide range of situations we have considered (a) transitions with a simple MF step, (b) pure $\ln(t)$ fluctuations with no MF step, and (c) admixtures of the two. In all following cases C corresponds to the difference of $C - C_{\text{normal}}$.

For the simulated MF specific-heat function we adopt a form which conserves entropy at T_c,

$$C_{\text{mf}}(x) = x(3x^2 - 1) \quad \text{for} \quad x = T/T_c \leq 1,$$

$$C_{\text{mf}}(x) = 0 \quad \text{for} \quad x > 1. \tag{1}$$

For the fluctuation specific heat we assume

$$C_{\text{fluc}}(x) = \ln(1/|x-1|) \quad \text{for all } x. \tag{2}$$

This has a symmetric divergence at T_c. Equation (1) is not dissimilar to the weak-coupling *d*-wave result, and Eq. (2) is pertinent to three-dimensional (3D)-*XY* critical fluctuations, but in fact we could have used any mean-field and fluctuations terms with little change in our results. For the distribution of T_c values we assume a Gaussian function centered on T_{co} with half width $w=0.001$, 0.02, 0.05, 0.1, 0.15, and 0.2,

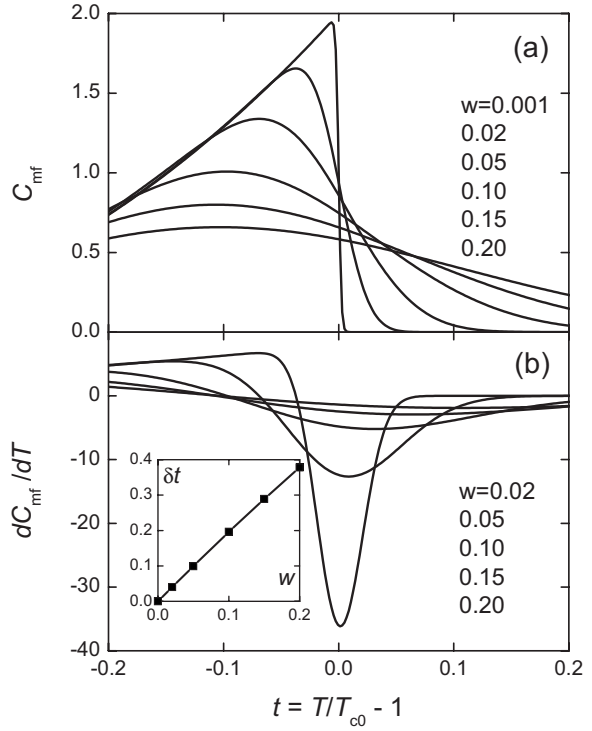

FIG. 1. (a) The MF specific-heat anomaly modeled using Eq. (1) with a normal distribution of T_c values with half-width, $w=0.001$, 0.02, 0.05, 0.10, 0.15, and 0.20. (b) The derivative dC_{mf}/dT. Inset: the separation δt of points of maximum positive and negative curvatures d^2C/dT^2 vs w.

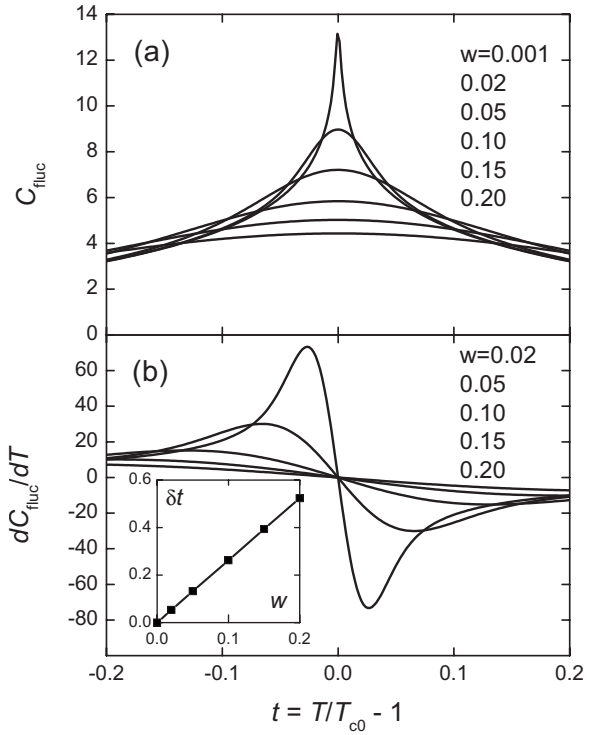

FIG. 2. (a) The fluctuation specific-heat anomaly modeled using Eq. (2) with a normal distribution of T_c values with half-width, $w=0.001$, 0.02, 0.05, 0.10, 0.15, and 0.20. (b) The derivative dC_{fluc}/dT. Inset: the separation δt of points of maximum negative and positive slopes dC/dT vs w.

$$P(y) = \frac{1}{w\sqrt{2\pi}}\exp\left[-\frac{(y-1)^2}{2w^2}\right]; \quad y = T_c/T_{co}. \tag{3}$$

The specific heat resulting from this T_c distribution is

$$C(T) = \int_0^\infty C(x)P(y)dy. \tag{4}$$

Alternatively, we note that averaging over the free energy and then differentiating twice to obtain the specific heat again gives very similar results to those discussed below.

For a range of widths w, plots of C and dC/dT versus $t=(T/T_{co})-1$ are shown in Fig. 1 for C_{mf} and in Fig. 2 for C_{fluc}. Admixtures $C_{\mathrm{mix}}=C_{\mathrm{mf}}+mC_{\mathrm{fluc}}$ with $m=0$, 0.2, 0.4, 0.6, 0.8, and 1 are plotted versus t in Figs. 3(a) and 3(b) for $w=0.02$.

We note, first, that despite the very different T dependencies of the unbroadened anomalies, the region affected by broadening is very similar for C_{mf}, C_{fluc}, and C_{mix}. This shows that the addition of fluctuations will not narrow the transition width as measured by the negative-curvature region around T_c. We also find that the relative positions of key features of the T derivatives of the broadened curves are insensitive to the detailed T dependencies assumed for C_{mf} and C_{fluc}.

(i) For the MF anomaly, shown in Fig. 1, a reliable measure of the broadening is given by the separation δt of points of maximum positive and negative curvature d^2C/dT^2 since this quantity is insensitive to the slope of the MF term below T_c. In the inset to Fig. 1(b) we show δt plotted as a function of the half width w. Over most of the range of w, $\delta t \approx 2.0w$.

(ii) For the symmetric fluctuation anomaly, shown in Fig. 2, a reliable measure of the broadening is given by the separation δt of points of maximum negative and positive slopes dC/dT, which is insensitive to the detailed T dependence of the fluctuation term. In the inset to Fig. 2(b) we show δt plotted as a function of w. Over the entire range, $\delta t \approx 2.63w$.

(iii) For the admixture of a MF anomaly and fluctuations, Fig. 3 shows plots of $C_{\mathrm{mix}}=C_{\mathrm{mf}}+mC_{\mathrm{fluc}}$ for $w=0.02$ with $m=0$, 0.2, 0.4, 0.6, 0.8, and 1. These are useful for comparison with typical cuprate specific-heat data and cover the range from asymmetric ($m=0$) to almost symmetric ($m=1$) anomalies. A value of $m \approx 0.8$ is appropriate for weakly overdoped Bi-2212. From Fig. 3(b) we obtain the positions of the points of maximum positive and negative slopes at t_- and t_+, respectively, and the difference $\delta t = t_+ - t_-$. For larger values of m, typical of our Bi-2212 samples, we find $\delta t \approx 2.7w$. For $m=\infty$ (pure fluctuations) we found, above, $\delta t \approx 2.63w$.

Now we compare this model data with measured data for Bi-2212. Plots of dC/dT versus $\tau = T/T_p - 1$ are shown in Fig. 3(c) for eight values of p (ranging from well underdoped to well overdoped). Comparison with Fig. 3(b) shows that T_c is only 1%–2% above T_p, so separations of peaks in t and in τ are almost identical. When underdoped the anomaly is pure fluctuations with no MF step while the increase in MF step is evident for overdoping from the increasingly different relative magnitudes of the positive and negative peaks in dC/dT.

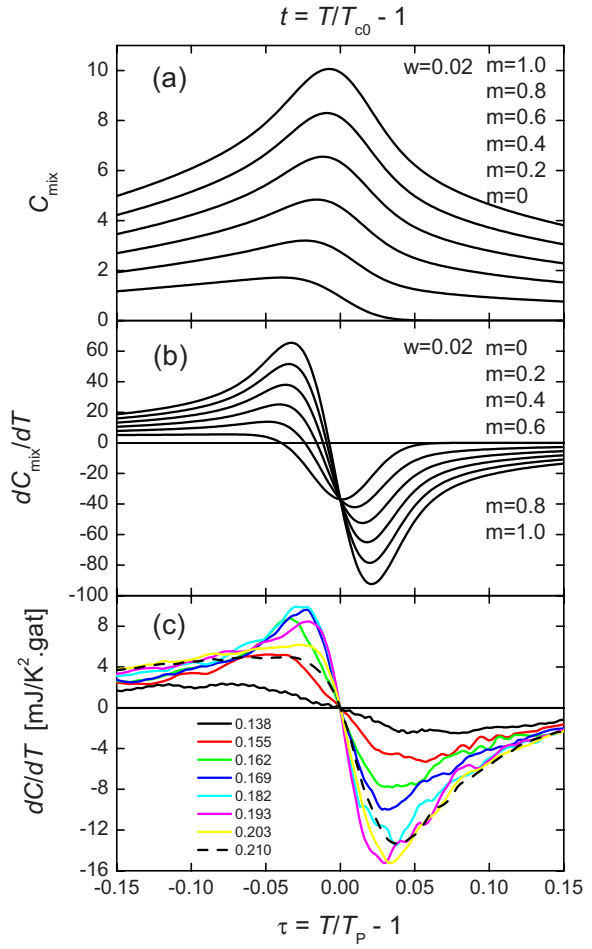

FIG. 3. (Color online) (a) The admixture specific-heat anomaly $C_{\rm mix}=C_{\rm mf}+mC_{\rm fluc}$ with half-width, $w=0.02$ and various mixing ratios, m, plotted versus $t=(T/T_{co})-1$. (b) The derivative $dC_{\rm mix}/dT$ vs t. The zero crossing occurs at T_p and the curves intersect at T_{c0}. (c) dC/dT vs τ for Bi-2212 at different doping levels (annotated) showing a crossover from pure fluctuations for $p<0.162$ to an admixture with a MF step for $p>0.162$.

We estimate the transition width, w, from the separation of the positive and negative peaks in dC/dT. For all samples with $p\geq 0.169$ the separation is $\delta t\approx 0.05-0.06$. Taking $\delta t\approx 2.7w$ for $m\approx 1$ [see Fig. 3(b)] gives $w\approx 0.019-0.021$ for the half width of the distribution of T_c values. For $p=0.162$ we have $\delta t\approx 0.073$ or $w\approx 0.028$; and for the most underdoped, $p=0.138$, we have $\delta t\approx 0.14$ or $w\approx 0.056$. All these values are in good agreement with the values of the half width Δt shown in Fig. 2 of our previous work.[9]

As a check on our previous method[9] for estimating these half widths, Δt, we show in Fig. 4 plots of $C_{\rm mix}$ versus $\log_{10}(t)$ and $\log_{10}(t^*)$, respectively, for $w=0.02$ and $m=0$ to 1, where $t^*=\sqrt{t^2+\Delta t^2}$. Our normal procedure is to choose a value of Δt that just averts the negative curvature close to T_c seen in plots of $C_{\rm mix}$ versus $\log_{10}(t)$, and it is evident from the solid curves that the choice $\Delta t=w$ achieves this result. This confirms that estimates of the broadening Δt from plots of ΔC versus $\log_{10}(t^*)$ give reliable values comparable to the true half width w. The plots in Fig. 4 also show that this procedure provides a reliable estimate of the MF step $\Delta C_{\rm mf}$

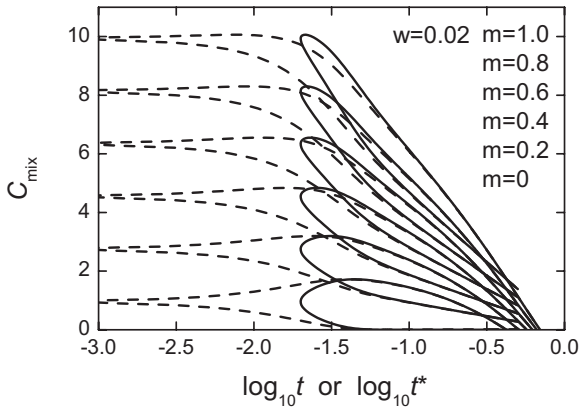

FIG. 4. The admixture specific-heat anomaly $C_{\rm mix}=C_{\rm mf}+mC_{\rm fluc}$ with half-width, $w=0.02$, plotted against $\log_{10}(t)$ (dashed curves) and against $\log_{10}(t^*)$ (solid curves) with $t^*=\sqrt{t^2+\Delta t^2}$ and setting $\Delta t=w$.

≈ 1.5 even in the presence of large fluctuations.

Turning to the MF calculation of the specific-heat anomaly by Andersen *et al.*,[7] and recalling that their model does not include fluctuations, we compare their results [shown in Fig. 2b of their paper] with our plots for a fluctuation-free broadened MF step, shown here in Fig. 1. From the location of the temperatures T_- and T_+ of their maximum negative and positive curvatures, we obtain $\delta t=(T_+-T_-)/T_{av}\approx 0.146$, 0.204, and 0.232 for their curves for $\delta g/t=1.0$, 1.5, and 2.0, respectively. The last of these gave the closest match of the calculated and experimental specific heat well away from T_c. Taking $\delta t=2w$ from Fig. 1(b) we find $w\approx 0.073$, 0.102, and 0.116, respectively, with the last of these being favored, as noted. Consistent with this, we observe that the spread of gaps deduced from STS gapmaps exhibits a constant relative half width $w\approx 0.12$ across the entire under- and overdoped regions.[5] In contrast, we found from our Bi-2212 data $w\approx 0.02$ over most of the doping range, a factor of 6 lower than both estimates. We conclude that the spectroscopic and thermodynamic data are not consistent. The specific heat for Bi-2212 implies a significantly more homogeneous electronic state than is inferred from the STS gap maps. In the case of $YBa_2Cu_3O_{7-\delta}$ the thermodynamic transitions are very much sharper still, implying an even more homogeneous electronic state.

The source of this inconsistency between the spectroscopic and thermodynamic data is undoubtedly to be found in the fact that the specific heat is a bulk property while STS is a surface property dominated by the outermost CuO_2 plane and strongly modified by scattering effects of extrinsic origin. The observation of gaps persisting to well above T_c in local patches[8] does not negate this. These exhibit features characteristic of the pseudogap (no coherence peaks and the gap fills rather than closes with increasing T). Within the model of Cooper[21] the pseudogap, located as it is around $(\pi,0)$, would appear patchy. Persistence of these large gaps below T_c shows that the pseudogap coexists with SC, and this fact helps convey the false impression that the SC gap evolves smoothly into the pseudogap above T_c, as widely reported.[30]

As noted, more complex ways of distributing T_c values in our model would give similar results. Andersen *et al.*[7] used a distributed pairing interaction, and we have correctly identified the initial spread of gaps used in their calculations for the pure mean-field case. Inclusion of fluctuations will not alter this. This case is treated by Ebner and Stroud[29] using Monte Carlo calculations for Josephson-coupled grains with a random distribution of T_c values. For moderately or weakly linked grains the computed specific-heat peak is strongly broadened (as shown by the separation of inflexion points) despite the presence of fluctuations. For the case of uniform T_c across all grains, rounded anomalies are still observed for weak to moderate couplings due to the finite-size effects.[29] The two cases illustrate the separate effects of distributed T_c values and finite-size effects, and in neither cases are sharp anomalies observed due to the presence of fluctuations. We believe therefore that, quite generally, fluctuations will not sharpen the specific-heat anomaly for an inhomogeneous system that is not strongly coupled.

In summary, by modeling a distributed mean-field step combined with fluctuations we have shown that the transition cannot be sharpened by fluctuations. The spread of T_c values inferred from our analysis of the thermodynamic data is significantly less than that implied by pairing inhomogeneity. This seems to concur with the wider thermodynamic,[9,26] NMR,[25] ARPES,[12] STS,[13–17] and intrinsic tunneling[18–20] data.

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