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Thermodynamic transitions in inhomogeneous *d*-wave superconductors

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We study the spectral and thermodynamic properties of inhomogeneous *d*-wave superconductors within a model where the inhomogeneity originates from atomic-scale pair disorder. This assumption has been shown to be consistent with the small charge and large gap modulations observed by scanning tunneling spectroscopy on $Bi_2Sr_2CaCu_2O_{8+x}$. Here we calculate the specific heat within the same model, and show that it gives a transition width comparable to those found in experiments on this material. This suggests that nanoscale inhomogeneity is a bulk property of the $Bi_2Sr_2CaCu_2O_{8+x}$ materials.

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There is accumulating evidence from scanning tunneling spectroscopy (STS) that at least some of the families of superconducting cuprate materials are "intrinsically" inhomogeneous at the nanoscale,[1–4] in the sense that all samples of the given material exhibit electronic disorder over length scales of ~25 Å which cannot be removed by any standard annealing procedure. In particular, differential conductance maps of $Bi_2Sr_2CaCu_2O_{8+x}$ (BSCCO) reveal the existence of spectral peaks reminiscent of the coherence peaks of a homogeneous *d*-wave superconductor whose energy varies by a factor of 2–3 over the sample. There is currently widespread interest in this phenomenon, but no consensus as to its origin, nor even as to whether the gaps between these peaks may be taken to correspond directly to the local superconducting order parameter in the sample, or whether some are related to a second, competing order.[5] While these experiments have been proposed as evidence of a general nanoscale inhomogeneity in cuprates, there have been several criticisms. One class of objections points out that other materials, particularly $YBa_2Cu_3O_{7-\delta}$ (YBCO) exhibit much narrower NMR linewidths, suggesting that in this material at least, the electronic inhomogeneity cannot be a pronounced property of the bulk.[6] A stronger critique has been offered by Loram *et al.*[7] even with regard to the BSCCO material. These authors point out that if one associates with each "gap patch" a local doping corresponding to the hole density required to produce a gap of that size in the average phase diagram, then the distribution of the doping is of order $\Delta p \sim 0.1$. Such a large range of local doping levels would in turn produce a large inhomogeneous distribution of regions with different T_c's, sufficient to yield a transition width ΔT_c of order the average T_c itself. Since this is contrary to specific heat measurements,[8,9] these authors conclude that the inhomogeneity is either a surface phenomenon, or represents a distribution of antinodal scattering lifetimes rather than of order parameters. Clearly it is very important to determine whether results from surface-sensitive probes reflect the bulk properties of these materials.

Recently, localized resonances were imaged by STS at a bias of −960 meV and identified with the O interstitials partially responsible for doping the BSCCO material.[10] This work also found a surprising positive correlation between the positions of these dopants and the local spectral gap, and furthermore argued that the charge inhomogeneity in the sample was considerably smaller than anticipated on the basis of previous scanning tunneling topographs integrated to smaller bias voltages. Nunner *et al.*[11] then argued that these and a number of other experimental observations[12] could be explained by the simple hypothesis that the disorder caused by the dopant atoms was primarily in the Cooper rather than density channel, i.e., that each dopant modulated the BCS pair interaction on an atomic scale.

In this paper we argue that the pair disorder model of Ref. 11 also allows one to avoid the argument of Loram *et al.*[7] regarding the transition width. The model, with parameters chosen to reproduce the gap maps and other correlations of the STS data on BSCCO, is shown to yield results within mean-field theory that are consistent with a relative specific heat transition width $\Delta T_c/T_c$ of ~20%, despite the fact that at $T=0$ gap modulations of order 100% are observed. The observed widths are therefore mainly attributable to nanoscale pair disorder since the samples are homogeneous at the *mesoscale* (as determined, e.g., by optical and scanning electron microscopy). Predictions of the theory for the formation and persistence of superconducting islands near the transition should be verifiable by high-temperature STS measurements.

There are many studies of inhomogeneous superconductivity,[13] only a few of which are directly relevant to the questions addressed below. If one adds nonmagnetic disorder to an ordinary *s*-wave superconductor, one expects only small changes in superconducting one-particle properties such as the gap and T_c due to Anderson's argument.[14] Nevertheless, as disorder increases and the mean free path ℓ becomes comparable to the Fermi wavelength λ_F, localization effects can destroy superconductivity. Recently, local aspects of this transition were studied by Ghosal *et al.*,[15] who showed using numerical solutions of the Bogoliubov–de Gennes (BdG) equations including nonmagnetic disorder that near the transition the highly disordered system separates into islands with finite order parameter surrounded by an insulating sea. In the *d*-wave case, where ordinary disorder is pair breaking and the transition occurs when $\ell \sim \xi_0$, where ξ_0 is the coherence length, several numerical studies have investigated local properties of disordered systems.[16–19] Only Ref. 16 considered the finite-temperature transition, however, pointing out that the inhomogeneity induced by ordinary disorder (in this case the suppression of the order

parameter around strong scattering centers) strongly affects the transition, leading to a T_c onset (defined by the formation of superconducting islands as temperature T is reduced from above) considerably higher than the T_c predicted by the usual disorder-averaged theory of T_c suppression, analogous to the theory of Abrikosov and Gorkov.[20] The width of the transition was not discussed explicitly in this work, nor was the effect of pairing disorder considered.

More recently, it has been proposed that T_c can even be increased by local inhomogeneities compared to its value for the homogeneous system.[21,22] Within a weak-coupling BCS framework it was shown that periodic modulations of the pairing and/or electron density can lead to a substantial enhancement of T_c when the characteristic modulation length scale is of order ξ_0.[21] This agrees with recent numerical calculations for the attractive Hubbard model with various disorder distributions of the interaction, verifying that T_c enhancement by inhomogeneity is a general result arising from the proximity effect.[22] These calculations did not investigate the details of the transition below T_c where the local order parameter Δ first nucleates.

In the following model calculation we use the standard d-wave BCS Hamiltonian

$$\hat{H} = \sum_{\langle ij\rangle\sigma} t_{ij}\hat{c}^{\dagger}_{i\sigma}\hat{c}_{j\sigma} + \sum_{i\sigma}(V_i - \mu)\hat{c}^{\dagger}_{i\sigma}\hat{c}_{i\sigma} + \sum_{\langle ij\rangle}(\Delta_{ij}\hat{c}^{\dagger}_{i\uparrow}\hat{c}^{\dagger}_{j\downarrow} + \text{H.c.}), \tag{1}$$

where in the first term we include nearest t and next-nearest $t'=-0.3t$ neighbor hopping. For the chemical potential μ, we set $\mu=-t$ in order to model the Fermi surface of BSCCO near optimal doping $\sim$16%. $\Sigma_{\langle ij\rangle}$ denotes summation over neighboring lattice sites i and j. Disorder is included by the impurity potential $V_i=V_0 f_i$ where $f_i=\Sigma_s \exp(-r_{is}/\lambda)/r_{is}$, r_{is} being the distance from a defect s to the lattice site i in the plane. Distances are measured in units of $\sqrt{2}a$, where a is the Cu-Cu distance. Note that the particular Yukawa form of f_i is merely a convenient way to vary the smoothness of the potential landscape through the parameter λ. The d-wave order parameter $\Delta_{ij}=g_{ij}\langle\hat{c}_{i\uparrow}\hat{c}_{j\downarrow}-\hat{c}_{j\downarrow}\hat{c}_{i\uparrow}\rangle/2$ is determined self-consistently by iterations of

$$\Delta_{ij} = \frac{g_{ij}}{2}\sum_n [u_n(i)v_n(j) + v_n(i)u_n(j)]\tanh\left(\frac{E_n}{2T}\right), \tag{2}$$

until convergence is achieved. Here, $\{E_n, u_n, v_n\}$ is the eigensystem resulting from diagonalization of the BdG equations associated with Eq. (1). The pairing interaction g_{ij} varies in space relative to its homogeneous value $g_0=1.16t$ as $g_{ij}=g_0+\delta g(f_i+f_j)/2$, where δg is the modulation amplitude and i,j are nearest neighbors. In the following, when including potential (τ_3 channel, $V_0\neq 0$) or pair (τ_1 channel, $\delta g\neq 0$) disorder, we make sure to adjust μ and g_0, respectively, in order to maintain the same doping and average gap as in the corresponding homogeneous system. Note that in this approach the inclusion of spatial pair potential variations is purely phenomenological. However, a likely candidate for the pairing modulations is the oxygen dopant disorder. Various possible microscopic origins for this unusual disordered state have been discussed in Refs. 11 and 12. There it was also argued that τ_3 disorder alone cannot account for the scanning tunneling microscopy data.

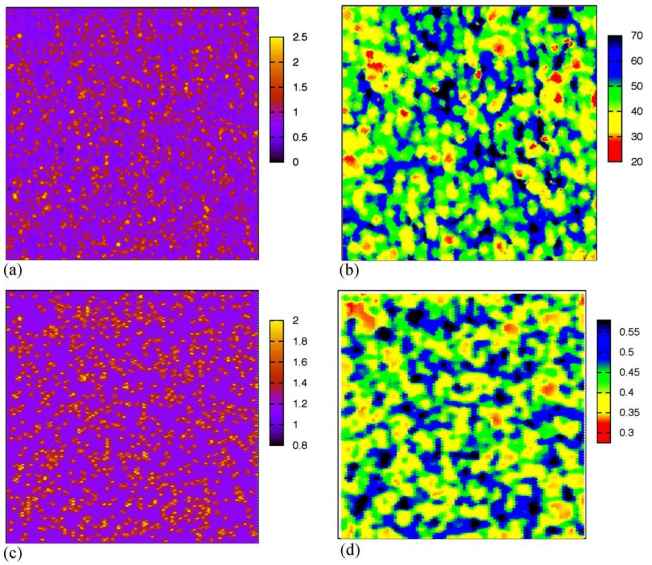

FIG. 1. (Color online) (a) Experimental dI/dV map (arb. units) at −960 meV of an optimally doped BSCCO sample from Ref. 10. (b) Experimental gap map (meV) in the same region as (a) at $T=4$ K. (c) The theoretical impurity potential extracted from (a) assuming a distance from the CuO_2 plane $r_z=0.5$ and $\lambda=0.5$. (d) The gap map resulting from using (c) as the "pairing potential" in the BdG equations with $\delta g=1.5t$ and $T=0$. Both (c) and (d) are shown in units of t.

Figure 1 shows, in a field of view (FOV) of 49×49 nm^2, the experimental local density of states (LDOS) at −960 meV (a), and the corresponding gap map (b) obtained by McElroy *et al.*[10] The bright resonances in Fig. 1(a) reveal the location of the oxygen dopants. In Fig. 1(c) we show the impurity potential generated by using each of these dopants (833 in the above FOV) as a defect located out of the CuO_2 plane. This map resembles Fig. 1(a) as it should. The experimental FOV is modeled as a 90×90 lattice system rotated 45° with respect to the 3.83-Å-long Cu-Cu bonds, i.e., it includes $2\times 90\times 90$ sites and is aligned with the experimental FOV.[23] The theoretical gap map (as extracted from the LDOS) using Fig. 1(c) as a τ_1 potential with $\delta g=1.5t$ at $T=0$ is shown in Fig. 1(d). We find that gap maps consistent with experiment are found for $1.0<\delta g/t<2.0$. The correlation coefficient[24] between the gap maps Figs. 1(b) and 1(d) is 0.17,[25] a reasonably large value given the simplicity of the pure τ_1 calculation and the fact that the correlation between the dopant positions and the experimental gap map is 0.3.[10]

Now we turn to the discussion of the results for $T\neq 0$. Below, we set $N=50$ (corresponding to the upper left 5/9 square of each image in Fig. 1) and study the resulting order parameter (OP) maps, entropy $S(T)$, and specific heat $C(T)$ with focus on the temperature region near T_c. The number of iterations necessary for converged results increases dramatically near T_c, whereas just a few degrees away from T_c we typically find that 25–50 iterations suffice. In Fig. 2, we show the OP map $\Delta_i=(\Delta_{i,i+\hat{x}}+\Delta_{i,i-\hat{x}}-\Delta_{i,i+\hat{y}}-\Delta_{i,i-\hat{y}})/4$, for temperatures near T_c. The bulk transition temperature for the homogeneous system is $T_{c0}=0.182t$. Here, one clearly sees how separate islands of finite Δ_i start forming above T_{c0} when cooling down and eventually overlap at lower T. In

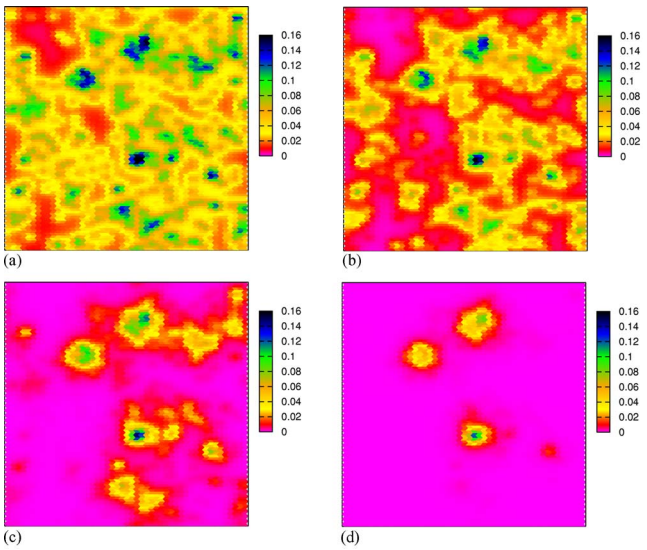

FIG. 2. (Color online) OP maps, parameters from Fig. 1(d): $T=0.18t$ (a), $0.20t$ (b), $0.22t$ (c), and $0.24t$ (d).

principle, STS measurements at T close to T_c should be able to probe these superconducting islands.

In order to address the effect of the inhomogeneity on the transition widths, we first calculate the quasiparticle entropy $S(T)$ according to the well-known expression

$$S=-2\sum_{E_n>0}[f(E_n)\ln f(E_n)+f(-E_n)\ln(f(-E_n))], \quad (3)$$

where $f(E)$ is the Fermi distribution function. From the resulting entropy $S(T)$ curve we extract the specific heat at constant volume $C(T)=T(\partial S/\partial T)$. Figure 3(a) compares the electronic specific heat for the clean and pair disordered systems described by different disorder strengths δg. In Fig. 3(b) we compare $C(T)$ for the different Nambu channels τ_1 and τ_3. We can check the BdG results by comparing to the pure "infinite" system result [black, dashed line in Fig. 3(a)] obtained by replacing $E_n \to E_{\mathbf{k}} \equiv \sqrt{\epsilon_{\mathbf{k}}^2+\Delta_{\mathbf{k}}^2}$, with $\epsilon_{\mathbf{k}}$ the normal-state dispersion with hopping integrals and chemical potential identical to the values given above, and $\Delta_{\mathbf{k}}=\Delta_0(T)(\cos k_x-\cos k_y)/2$ with $\Delta_0(T)$ obtained self-consistently from the usual gap equation. It is clear that the 50×50 BdG lattice problem is large enough to capture the sharp transition of the clean system.

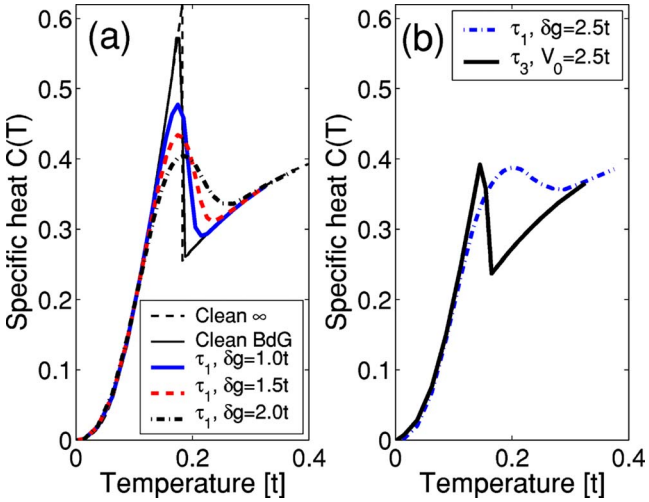

FIG. 3. (Color online) Specific heat $C(T)$ for the impurity distribution of Fig. 1(c) vs T. (a) Clean 50×50 BdG system (solid, black line), "infinite" clean system (dashed, black line), and τ_1 disordered with $\delta g=1.0t$ (thick solid, blue line); $\delta g=1.5t$ (thick dashed, red line); $\delta g=2.0t$ (thick dash-dotted, black line). (b) Comparison of $C(T)$ for the Nambu channels τ_1 and τ_3 with otherwise identical impurity parameters.

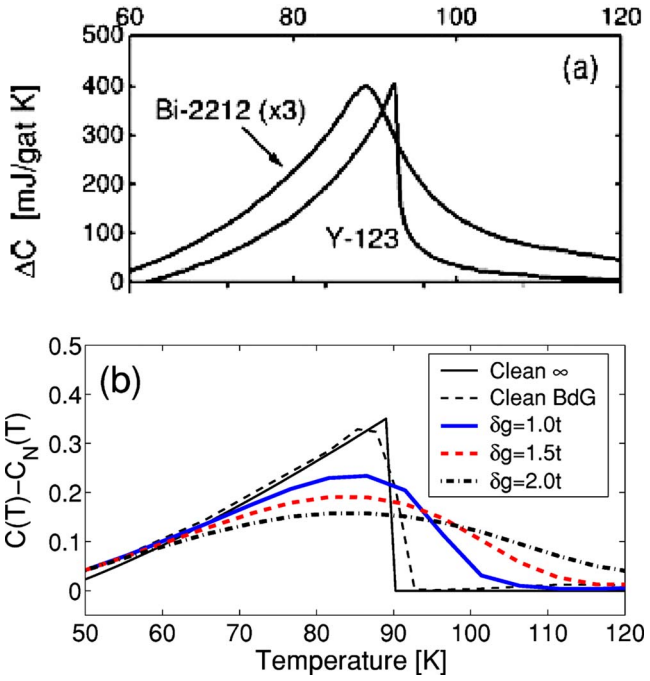

FIG. 4. (Color online) (a) Specific heat vs T for YBCO and BSCCO optimally doped powders, from Ref. 7. (b) $C(T)-C_N(T)$ (N, normal state) vs T with parameters from Fig. 3(a), assuming that $T_{c0}=0.182t$ corresponds to $T_c=90$ K.

Conventional potential disorder corresponding to $\sim 10\%$ dopant atoms causes only negligible broadening in the specific heat transition width compared to the pair disordered case. This is shown in Fig. 3(b), where both curves are produced from the impurity distribution in Fig. 1(c). The transition is sharp because potential scatterers cause local suppressions of the order parameter, whereas in interstitial regions Δ_i decreases with increasing temperature in a manner similar to the pure system. We have also studied distributions of strong scatterers at the percent level, and find similarly sharp transitions. On the other hand, potential disorder smooth on a scale of ξ_0 can lead to large modulations of μ which, even within BCS theory, will give larger transition widths. In this case, however, the associated local spectra are not consistent with STS measurements, as shown in Ref. 11.

As evident from Fig. 3(a), pairing disorder with parameters fixed to yield reasonable variations of the gap size and graininess at low temperature[11] leads to a broadened transition width similar to the experimental observations.[7] This is shown more clearly in Fig. 4, where we compare the experimental specific heat $C(T)$ for clean YBCO and optimally doped BSCCO near T_c with our results for the clean and pair disordered cases. It is clear that in the case of YBCO, the

transition is extremely narrow, and the deviations from the mean-field treatment discussed here are consistent with critical order parameter phase fluctuations in three spatial dimensions (3D *XY*) over a range of a few degrees.[26,27] On the other hand, in the case of BSCCO, the 3D *XY* description of the specific heat transition is controversial. It has in fact been discussed as closer to a Bose-Einstein condensate with specific heat exponent of $\alpha=-1$, although measured values are closer to $\alpha=-0.7$.[8] Samples of other authors have suggested that a 3D *XY* divergence in $C(T)$ cut off by bulk nanoscale inhomogeneities over a small range $\Delta T_c/T_c \sim 0.03$ could be consistent with the data.[7,28] Here we have put forward a related scenario, but our microscopic model implies that the inhomogeneities dominate over a larger range $\Delta T_c/T_c \sim 0.1-0.2$ to be consistent with STS.

Additional contributions from critical fluctuations must be added to the mean-field effects discussed here to obtain a complete description, and in a strongly anisotropic material like BSCCO such contributions may be significant. However, the nontrivial effects of strong disorder on the fluctuation contribution to $C(T)$ (Ref. 29) (and whether it leads to an overall sharpening or broadening of the mean-field result) are beyond the scope of the present paper. In addition, we note that the present theory does not allow for the presence of a residual pseudogap state, which may also complicate direct comparison with experiments even at optimal doping.

In conclusion, we have presented theoretical calculations for *d*-wave superconductors with atomic-scale pair disorder, using impurity parameters appropriate to reproduce semiquantitatively the gap maps produced by STM experiments on optimally doped BSCCO, and shown that the width of the specific heat transition can be captured by this model as well. This suggests that substantial nanoscale electronic inhomogeneity is characteristic of the bulk BSCCO system.

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[23] A small mismatch is introduced here because the experimental FOV is slightly skewed, and will be close to, but not exactly equal to, a 90×90 system.
[24] See Ref. 10 for a definition of the correlation function.
[25] This value of the correlation is sensitive to the details of the impurity potential: for instance, if one uses the raw LDOS data in Fig. 1(a) as input, it increases to 0.3.
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