Phase Fluctuations and the Pseudogap in $YBa_2Cu_3O_x$

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The thermodynamics of the superconducting transition is studied as a function of doping using high-resolution expansivity data of $YBa_2Cu_3O_x$ single crystals and Monte Carlo simulations of the anisotropic 3D-*XY* model. We directly show that T_c of underdoped $YBa_2Cu_3O_x$ is strongly suppressed from its mean-field value (T_c^{MF}) by phase fluctuations of the superconducting order parameter. For overdoped $YBa_2Cu_3O_x$ fluctuation effects are greatly reduced and $T_c \approx T_c^{\mathrm{MF}}$. We find that T_c^{MF} exhibits a similar doping dependence as the pseudogap energy, naturally suggesting that the pseudogap arises from phase-incoherent Cooper pairing.

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The properties of high-temperature superconductors (HTSC) vary strongly with hole doping. Especially the underdoped region of the phase diagram, where the critical temperature (T_c) increases with doping, has attracted considerable attention due to the gradual development of a pseudogap in the low energy electronic excitations at a temperature T^*, which lies significantly above T_c [1–9]. Although the pseudogap phenomenon has been known for some time, the exact nature of this state and its relation to superconductivity is still a subject of great controversy [1–10]. Basically there are two competing scenarios. In the first, the opening of the pseudogap is attributed to phase-incoherent precursor pairing, and T_c is much smaller than T^* because of strong phase fluctuations [2,8,10]. In the second scenario, the pseudogap is a normal state gap, which is independent of and competing with superconductivity [3,9]. The origin of the pseudogap is clearly of great importance for understanding the general phase diagram of HTSC.

In this Letter, we study the doping dependent thermodynamic response at T_c of $YBa_2Cu_3O_x$ crystals using high-resolution dilatometry. Thermal expansion, which is closely related to the specific heat through the thermodynamic Ehrenfest or Pippard relations [11], was previously shown to be a very sensitive probe of the superconducting transition due to the large signal to background ratio and provided direct evidence of strong 3D-*XY* critical fluctuations at optimal doping [12]. Here we show that fluctuation effects increase dramatically and become more two dimensional in the underdoped region, resulting in a large depression of T_c from its mean-field value (T_c^{MF}). We find excellent scaling of these fluctuations with Monte Carlo specific-heat simulations of the anisotropic 3D-*XY* model, which clearly demonstrates the "superconducting" origin of the fluctuations. T_c^{MF} is shown to exhibit a similar doping dependence as T^* naturally suggesting that the pseudogap arises from phase-incoherent Cooper pairing. Our results, thus, confirm the phase diagram based on the phase-fluctuation scenario proposed by Emery and Kivelson [10].

An untwinned $YBa_2Cu_3O_x$ single crystal [12,13] was used, whose oxygen content was varied between $x \approx 6.77$ and $x \approx 7.0$ by annealing in pure O_2 gas at pressures between 5 mbar (450 °C) and 376 bars (400 °C), resulting in slightly underdoped to slightly overdoped states [14]. The thermal expansivity was measured with a high-resolution capacitance dilatometer [15]. The Monte Carlo simulations, in which anisotropy is introduced by reducing the coupling coefficient J_z between spins along the z direction, were performed using the Wolff cluster update method on systems of sizes $64 \times 64 \times 64$ ($L_x \times L_y \times L_z$) for the isotropic case ($J_z/J_{xy} = 1$), $128 \times 128 \times 16$ for $J_z/J_{xy} = 0.02$, and $128 \times 128 \times 8$ for $J_z/J_{xy} = 0.004$. For the anisotropic systems the cell sizes were chosen such that $L_z/L_{xy} \approx \xi_z/\xi_{xy}$, where ξ_z and ξ_{xy} are the correlation lengths in the different directions and $\xi_z/\xi_{xy} = \sqrt{J_z/J_{xy}}$. Each specific heat data point is from simulations with the creation and flipping of a few million clusters.

A previous study of $YBa_2Cu_3O_x$ at optimal doping [12] showed that it is of great advantage to examine the difference between the expansivities of the b and a axes, $\alpha_{b-a} \equiv \alpha_b - \alpha_a$, because the anomalies at T_c are of approximately equal magnitude but of opposite sign. Taking this difference, thus, doubles the size of the anomaly and, at the same time, reduces the background [12]. In Fig. 1(a) $\alpha_{b-a}(T)$ is shown for six different values of x. Large, in comparison to the background, λ-shaped anomalies are seen at T_c, which decrease in size as the oxygen content is reduced from $x = 7.0$ to $x = 6.77$. For $x = 6.77$, the anomalies in the a and b axes have the same sign and magnitude and, thus, the anomaly in α_{b-a} vanishes [16]. This is actually a blessing in disguise, since the α_{b-a} curve for $x = 6.77$ provides us with a very good approximation of the curvature of the phonon background expansivity, $\alpha_{b-a}^{\mathrm{back}}$. By simply vertically scaling the smooth

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α_{b-a} ($x = 6.77$) data until they coincide with the various data sets at high temperatures, we obtain $\alpha_{b-a}^{\text{back}}$ for the different x values (see gray lines in Fig. 1). We believe that this procedure produces a very physical background for the different x values because the background matches the curvature of the high-temperature data very well for all x values and the variation of this background with x intuitively matches what one would expect as one progressively removes O atoms from the chain sites. The origin of $\alpha_{b-a}^{\text{back}}$ is the orthorhombic structure, i.e., the chains along the b axis. The magnitude of $\alpha_{b-a}^{\text{back}}$ is thus expected to be largest when the chains are fully oxidized ($x = 7.0$) and to decrease as oxygen is removed from the chains (at $x \approx 6.35$ the material becomes tetragonal, and $\alpha_{b-a}^{\text{back}}$ must vanish). This is exactly the observed behavior, which gives us a great deal of confidence in $\alpha_{b-a}^{\text{back}}$ [17].

In Fig. 1(b) $\alpha_{b-a}^{\text{back}}$ has been subtracted from all original data sets yielding the electronic thermal expansivity $\alpha_{b-a}^{\text{elec}}$. The magnitude of $\alpha_{b-a}^{\text{elec}}$ at T_c decreases drastically with oxygen depletion. This is largely due to the fact that the difference between the uniaxial pressure coefficients $dT_c/dp_{b-a} \equiv dT_c/dp_b - dT_c/dp_a$ vanishes at $x \approx 6.77$ [16], since $\alpha_{b-a}^{\text{elec}}$ is directly proportional to dT_c/dp_{b-a} [11]. In order to compare the shapes of these anomalies, we normalize the temperature scales by T_c and then divide $\alpha_{b-a}^{\text{elec}}(x, T/T_c)$ by $\alpha_{b-a}^{\text{elec}}(x, T/T_c = 0.7)$, which results in very good scaling in the low-temperature region [$T/T_c < 0.75$; see Fig. 2(a)]. By scaling the anomalies at low temperatures, we have in effect normalized all curves by the mean-field (MF) behavior, which allows us to quantify the effects of fluctuations relative to the MF component. For the slightly overdoped case ($x = 7.0$), $\alpha_{b-a}^{\text{elec}}$ resembles classical MF behavior with small fluctuation corrections close to T_c. As x decreases, the magnitude of the singular part at T_c increases and at the same time the range of the fluctuations increases dramatically, especially above T_c. For our most underdoped sample in Fig. 2(a) ($x = 6.81$), the fluctuation contribution extends up to about $2 \times T_c$, and the integrated signal above T_c is actually larger than the one below T_c. This manifests a drastic deviation from MF behavior, where all of the "action" is below T_c and clearly demonstrates

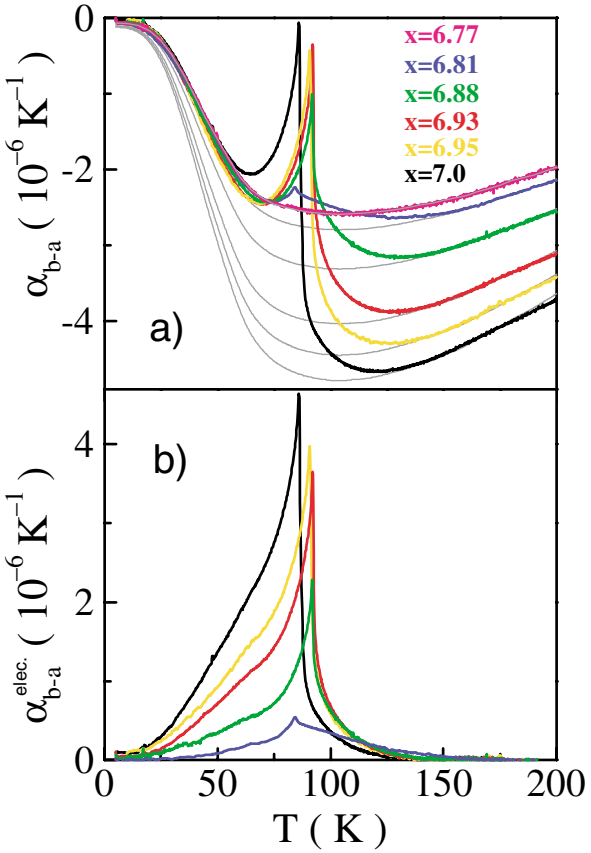

FIG. 1 (color). (a) Difference between the expansivity along the b and a axes $\alpha_{b-a}(T)$ of $YBa_2Cu_3O_x$ for $x = 6.77-7.0$. The gray lines represent the backgrounds, which were obtained by scaling the smooth $x = 6.77$ data (see text). (b) Electronic expansivity $\alpha_{b-a}^{\text{elec}}$ obtained after subtracting the backgrounds from the original data sets.

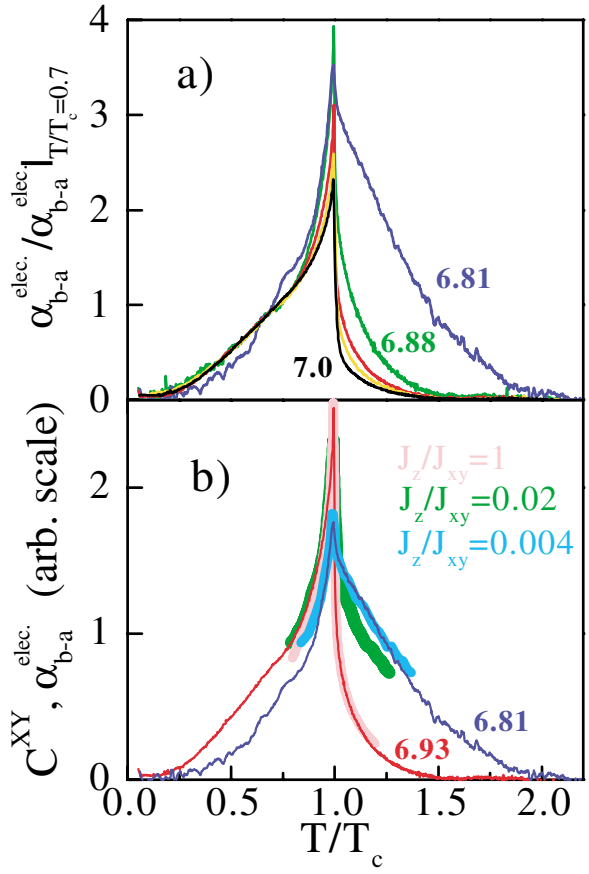

FIG. 2 (color). (a) Low-temperature scaled $\alpha_{b-a}^{\text{elec}}$ showing a dramatic increase of the fluctuation signal above T_c in the underdoped samples (see text). (b) Specific heat of the 3D-XY model (C^{XY}) for three different anisotropies (thick light-colored lines). Excellent scaling of $\alpha_{b-a}^{\text{elec}}$ and C^{XY} is observed over an extended temperature range for $x = 6.93$ (thin red line) and $x = 6.81$ (thin blue line).

the unusual character of the superconducting transition in underdoped $YBa_2Cu_3O_x$.

It is well known that the superconducting and normal state properties of $YBa_2Cu_3O_x$ and other HTSCs are highly anisotropic and also that this anisotropy increases strongly in the underdoped region [18,19]. We now show that the unusual behavior in the underdoped region can be understood in terms of strong superconducting phase fluctuations in a system with reduced dimensionality. For this purpose we compare $\alpha_{b-a}^{\rm elec}$ with the specific-heat simulations of the anisotropic 3D-XY model (C^{XY}), which are shown in Fig. 2(b) for three different anisotropy values ($J_z/J_{xy} = 1$, 0.02, and 0.004). This model belongs to the same universality class as superconductivity (in the limit $\kappa \gg 1$) and is expected to reproduce the critical behavior of the superconducting transition if one has strong fluctuations [18,20,21]. As anisotropy is introduced into the XY model, the shape of the C^{XY} curves changes in a very similar fashion as $\alpha_{b-a}^{\rm elec}$ curves change for decreasing x [Fig. 2(a)]; that is, more and more of the area under the transition is shifted to higher temperatures, and the "jump" component at T_c decreases, resulting in a more symmetric anomaly. Universality implies that C^{XY} and $C_p^{\rm elec}$ of $YBa_2Cu_3O_x$ follow the same scaling laws, or in other words, the anomalies should have the same shape near T_c [18]. In Fig. 2(b) we show that this is actually the case by directly scaling our $\alpha_{b-a}^{\rm elec}$, which are a direct reflection of $C_p^{\rm elec}$, with C^{XY}. C^{XY} for the isotropic case [$J_z/J_{xy} = 1$, thick light red line in Fig. 2(b)] excellently matches the $\alpha_{b-a}^{\rm elec}$ curve near optimal doping [$x = 6.93$, thin red line in Fig. 2(b)], where isotropic 3D-XY scaling has been found to be well obeyed [12,18,19]. In the underdoped region we find excellent agreement between C^{XY} of the most anisotropic simulation ($J_z/J_{xy} = 0.004$, thick light blue line) and $\alpha_{b-a}^{\rm elec}$ for $x = 6.81$ [thin blue line in Fig. 2(b)]. In both cases, scaling is observed over a very wide temperature range, implying an extremely large critical region. We note that the scaling of C^{XY} and $\alpha_{b-a}^{\rm elec}$ in Fig. 2(b) is obtained by a *single* multiplicative parameter, which can be expected only if the background subtraction in Fig. 1 is essentially correct. Also, the finite-size broadening of the transitions in both sets of data is of very similar magnitude, which greatly facilitates this comparison. In the overdoped region ($x = 7.0$) the fluctuations decrease in size and the simulations, which are based on strong critical behavior, no longer match the experimental data.

The nearly perfect scaling of C^{XY} and $\alpha_{b-a}^{\rm elec}$ around T_c shown in Fig. 2(b) implies that the physics of the superconducting transition in underdoped and optimally doped $YBa_2Cu_3O_x$ is correctly accounted for by the simple anisotropic 3D-XY model, in which only phase fluctuations of the order parameter occur, since the amplitude of the spin J is fixed. For superconductors, this corresponds to a fixed amplitude of Cooper pairs, and, thus, Fig. 2(b) provides direct evidence for strong phase fluctuations [18,20,21] in underdoped and optimally doped states. We note that the large reduction in the specific heat jump at T_c in the underdoped region, which has previously been taken as evidence of a normal-state pseudogap [3], chain superconductivity [22], or a MF-Bose-Einstein crossover [19], is a natural consequence of the crossover from anisotropic 3D-XY behavior in the underdoped region to MF-like behavior in the overdoped region [see Figs. 2(a) and 2(b)].

It is instructive to compare the values of the anisotropy of the simulations with the closely, but not trivially, related anisotropy of the superconducting coherence lengths $\Gamma = \xi_{ab}/\xi_c$. There are two limiting cases. First, if the out-of-plane "bare" coherence length $\xi_c^{\rm bare}$ at T_c is smaller than the spacing d between superconducting layers, then $\sqrt{J_{xy}/J_z} = \Gamma \cdot d/\xi_{ab}^{\rm bare}$, where $\xi_{ab}^{\rm bare}$ is the bare in-plane coherence length and bare refers to the fluctuation uncorrected, i.e., MF, values [21]. This appears to be the situation for $x = 6.81$, where $\Gamma \approx 12$ [23] and $\sqrt{J_{xy}/J_x} \approx 15$ are nearly equal, which implies that $d/\xi_{a,b} \approx 1$. Such small values of the bare coherence lengths at T_c ($\xi_c^{\rm bare} \ll 12$ Å and $\xi_{ab}^{\rm bare} \approx 12$ Å) imply very large fluctuation corrections, since usually one expects the coherence lengths to diverge at T_c. The bare coherence lengths, on the other hand, diverge at the fluctuation uncorrected $T_c^{\rm MF}$, and our result suggests that $T_c^{\rm MF}$ lies considerably higher than the real T_c. We show below that this is actually the case in the underdoped region. For the second case ($\xi_c > d$), d in the above equation should be replaced by $\xi_c^{\rm bare}$, resulting in the isotropic model ($J_z/J_{xy} = 1$) [21], which we observe near optimal doping ($x = 6.93$). Thus, although a considerable anisotropy ($\Gamma \approx 7$) still exists at optimal doping [23], the system behaves isotropically because of the strong coupling along the c axis. Our results point to a doping-induced crossover from quasi-2D behavior of weakly Josephson-type coupled planes in the underdoped region, where $\xi_c^{\rm bare} < d$, to the 3D behavior of strongly coupled planes in the optimal and overdoped regions, where $\xi_c^{\rm bare} > d$.

Phase transition temperatures are always lowered by strong fluctuations [10,18,20], and it is instructive to ask the following question: How much higher would T_c be if there were no fluctuations, or, in other words, what is $T_c^{\rm MF}$? In order to extract $T_c^{\rm MF}$ from our data, we have extrapolated the low-T MF component of $\alpha_{b-a}^{\rm elec}$ beyond the real T_c value, and $T_c^{\rm MF}$ is defined as the temperature where the area under the MF curve equals the total area under $\alpha_{b-a}^{\rm elec}$. The inset in Fig. 3 shows this construction for $x = 6.88$ yielding $T_c^{\rm MF} \approx 1.25 T_c$. This procedure, which is analogous to making an entropy conserving construction in heat capacity data, was carried out for the rest of the doping levels and the results are plotted versus the doping level [24] in Fig. 3. We find the astonishing result that $T_c^{\rm MF}$ decreases approximately linearly with increasing doping at

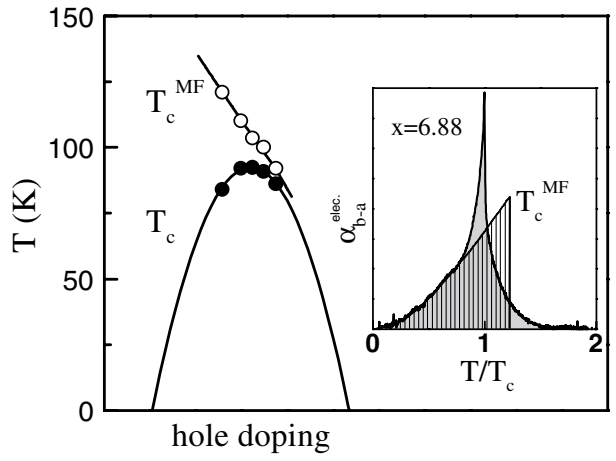

FIG. 3. Phase diagram of $YBa_2Cu_3O_x$ showing the doping dependence of both T_c and T_c^{MF}. The inset shows how T_c^{MF} is defined (see text).

the same time as the real T_c goes over a maximum. In the overdoped region the two T_c values merge because the fluctuations are greatly reduced. We note that the above estimate of T_c^{MF} depends only on the shape of $\alpha_{b-a}^{\mathrm{elec}}$ and not on the absolute scale.

The doping dependence T_c^{MF} shown in Fig. 3 presents one of our most important results, and several implications follow immediately. First, T_c^{MF} and the pseudogap temperature [5], or energy scale [3,9], exhibit a very similar doping dependence, which strongly suggests that the pseudogap is due to local Cooper pairing without phase coherence. The fact that T_c^{MF} also shows the same linear doping dependence as the single particle excitation gap at zero temperature $\Delta(0)$ seen in both scanning tunneling microscopy [6,7] and angle-resolved photoemission data [4,5] supports this conclusion, since in a MF theory $\Delta(0)$ is a direct measure of T_c^{MF}. The strong doping dependence of the ratio $2\Delta(0)/kT_c$ [4–7], in our view, is a fluctuation artifact. We also expect $2\Delta(0)/kT_c$ of $Bi_2Sr_2CaCu_2O_{8+d}$ to be larger than that of $YBa_2Cu_3O_x$ at optimal doping, because $Bi_2Sr_2CaCu_2O_{8+d}$ is much more two dimensional and fluctuations play an even more important role than in $YBa_2Cu_3O_x$ [20,25,26]. There is some evidence that this is actually the case [6,27]. Second, the maximum in T_c at optimal doping is really an artifact due to fluctuations; that is, the pairing energy, as reflected by T_c^{MF}, decreases smoothly with increasing doping with no indication of any special behavior near optimal doping. Our phase diagram is in excellent agreement with the phase-fluctuation scenario proposed by Emery *et al.* [10] and also with a recent analysis of the *t-J* model [28]. Finally, extrapolating our T_c^{MF} to even lower doping levels suggests that the strongest pairing interaction in $YBa_2Cu_3O_x$ occurs at, or near, the antiferromagnetic phase boundary. This may be an indication of a magnetic pairing mechanism, as recently proposed for several heavy-fermion compounds [29] and also suggested from magnetic neutron scattering data [30].

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