Thermodynamic fluctuations in the high-T_c perovskite superconductors

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The role of fluctuations in the breakdown of mean-field theory in the new high-T_c perovskite superconductors is analyzed. Both the breakdown of the classical critical exponents (the Ginzburg criterion) and the quantitative breakdown of mean-field theory (the Brout criterion) are discussed. It is found that for certain sets of observed parameters for these materials, a breakdown of mean-field theory is to be expected.

For ordinary superconductors, the superconducting transition is extremely well described by the Ginzburg-Landau theory (GL),[1] which was proven by Gor'kov[2] to be equivalent to the BCS theory[3] in the limit $T \to T_c$. This is a direct manifestation of "mean-field" behavior in its strongest form, i.e., where both the order parameter and the coefficients of the GL theory can be calculated from the microscopic mean-field theory.

However, mean-field theory does not account for most real second-order phase transitions (e.g., superfluid helium, paramagnetic-to-ferromagnetic transitions, liquid crystals, etc.) or model systems (e.g., Ising model, Heisenberg model, etc). These departures from "classical" behavior are generally attributed to thermal fluctuations, which are neglected in the mean-field approach. Standard estimates of the critical region show that in pure superconductors the range of temperatures around T_c within which fluctuations are important is as small as $|T-t_c| < 10^{-14}T_c$, whereas in alloys it can be $|T-T_c| < 10^{-7}T_c$. Both are very small intervals around T_c that are not in general experimentally accessible.[4]

Less widely appreciated is that the breakdown of mean-field theory is progressive.[4] As one approaches the critical region, the ability of mean-field theory to calculate nonuniversal quantities (prefactors, parameters of the GL theory, and T_c) breaks down before the universal critical exponents and certain ratios of amplitudes become modified. To estimate the latter, one customarily uses the Ginzburg criterion,[5] whereas to estimate the former it is necessary to use the less well-known Brout criterion.[6,7]

In this Rapid Communication we review these two criteria and analyze current experiments on the new high-T_c perovskite superconductors to estimate the extent of these two "critical" regions. We further show that for some sets of measured parameters, critical phenomenon should be expected. We emphasize the difference between an approach that is based on strict three-dimensional fluctuations[4] and one that is based on a layered model.[8]

Two other discussions of possible critical phenomena in the oxide superconductors are known to us.[9] Both are less extensive than the discussion here. Also, they do not mention the importance of the Brout critrion.

The macroscopic wave function Ψ is allowed to fluctuate from region to region, but any fluctuations of wave length shorter than L are absorbed into the definition of Ψ. Here L is the temperature-dependent coherence length in the case of the breakdown of the macroscopic mean-field behavior[5] and the zero-temperature coherence length in the case of the breakdown of the microscopic theory.

Following the standard development, fluctuations of Ψ can be calculated from the GL theory by considering the contribution from wavelengths larger than L. The GL free energy is

$$F\{\Psi(r),T\} = F_n(T) + \alpha(T)|\Psi|^2 + \tfrac{1}{2}\beta(T)|\Psi|^4 + \gamma(T)|\nabla\Psi|^2 , \qquad (1)$$

where $\beta(T)$ and $\gamma(T)$ are taken to be regular at T_c. Minimization of F with respect to Ψ gives the result

$$\langle|\Psi|\rangle^2 = -\alpha(T)/\beta(T) \equiv -\alpha' t/\beta(T) ,$$

where $t = |T-T_c|/T_c$ is the reduced temperature. By Fourier transforming the difference in free energies, one obtains the Orenstein-Zernike[10] correlation function:

$$G(q) = \frac{k_B T_c/\gamma(T_c)}{q^2+\xi^{-2}} . \qquad (2)$$

It is useful to note that the jump in the specific heat (per unit volume) $\delta c(T_c)$ at T_c and the slope of the thermodynamic critical field $|dH_c(T)/dT|_{T_c}$ are related to the GL coeffecients by

$$\frac{\alpha'^2}{\beta(T_c)} = T_c\delta c(T_c) = \frac{1}{4\pi}\left(T\frac{\partial H_c}{\partial T}\right)^2_{T_c} \approx 2\delta F_c(0) , \qquad (3)$$

Where $\delta F_c(0) = H_c(0)^2/8\pi$ is the condensation energy at zero temperature. The coherence length is given in terms of the Ginzburg-Landau coefficients by

$$\xi(T)^2 = \frac{\gamma(T)}{|\alpha(T)|} \equiv \xi(0)^2 t^{-1} . \qquad (4)$$

The critical region, within which $\langle(\delta\Psi)^2\rangle \geq \langle|\Psi|\rangle^2$, can now be estimated from the summation of the fluctuations over all wavelengths [namely, integrating Eq. (2)] and then dividing by the average order parameter:[5]

$$\frac{\langle(\delta\Psi)^2\rangle}{\langle|\Psi|\rangle^2}=\frac{k_B}{\delta c(T_c)t\xi(0)^2(2\pi)^3}\int_0^{L^{-1}}\frac{d^dq}{q^2+\xi^{-2}}\geq 1\,. \tag{5}$$

To demonstrate our estimates, we discuss first the results for an isotropic material. Carrying out the integral in Eq. (5) with the range of integration $L=\xi(T)$, we obtain the Ginzburg criterion[5] for the critical region $\zeta^G\equiv|T_f-T_c|/T_c$ within which mean-field exponents cease to be valid:

$$\zeta^G=A\left[\frac{1}{\xi(0)^3}\Big/\frac{k_B}{\delta c}\right]^2. \tag{6}$$

Here A is a constant number typically $A\sim10^{-4}\div10^{-3}$. The criterion for the "breakdown" of the microscopic mean-field theory (i.e., the Brout criterion) is realized when $\xi(0)$ instead of $\xi(T)$ is substituted for L in Eq. (5). In microscopic terms, the Brout criterion is equivalent to calculating the fluctuations within the range of the kernel in the Gor'kov equations, hence renormalizing the microscopic coefficients. If we insert now $\xi(0)$ for L in Eq. (5), we get the general result that the Brout critical region $\zeta^B\propto(\zeta^G)^{1/2}$. If Gor'kov-BCS theory is still valid, further relations exist between microscopic parameters and thermodynamic quantities, e.g.,

$$k_B/\delta c(T_c)=0.17k_BT_c/\delta F_c(0)$$

is a coherent volume, given in terms of the single-particle density of states at the Fermi level $g(\varepsilon_F)$ and the zero-temperature gap Δ by

$$k_B/\delta c(T_c)=k_BT_c[3g(\varepsilon_F)\Delta^2]\,.$$

In the isotropic case, this volume will be $\pi^3\xi_0/k_F^2$, where $\xi_0=hv_F/\pi\Delta$ is BCS coherence length. Hence one gets

$$\zeta^G=A'\left[\frac{\xi_0}{k_F^2\xi(0)^3}\right]^2. \tag{7}$$

where again A' is a constant of order 0.4. In the clean limit where $\xi(0)\approx\xi_0$ we get

$$\zeta^G_{\text{clean}}\approx\left[\frac{1}{k_F^2\xi_0^2}\right]^2\approx\left[\frac{\Delta}{\varepsilon_F}\right]^4. \tag{8}$$

In conventional superconductors $\Delta/\varepsilon_F\cong10^{-4}$, resulting in an extremely narrow critical region, $\zeta^G<10^{-14}$ [if we include the constant in Eq. (8)].

In the dirty limit, $\xi(0)^2=\xi_0\cdot l$ and

$$\zeta^G_{\text{dirty}}\approx A''\left[\frac{\Delta}{\varepsilon_F}\right]\frac{1}{(k_Fl)^3}\,, \tag{9}$$

where $A''\approx0.01$. The above calculation is of course valid only when the Anderson theorem[8] applies, namely, $k_Fl\gg1$. For $k_Fl\cong10$, $\zeta^G<10^{-11}$ and the critical region is very narrow. When $k_Fl<1$, it was shown by Kapitulnik and Kotliar[11] that the critical region broadens and can reach unity.

In the clean limit, the Gor'kov-BCS relations for the mean-field coefficients will no longer be valid in the region $\zeta^B\approx(\Delta/\varepsilon_F)^2\approx10^{-8}$ for conventional superconductor. Of course this region becomes larger as either the material becomes "dirtier" or Δ approaches ε_F. Still it is a very small number.

Let us now turn to the calculation of ζ^G and ζ^B for multilayered materials, which is of obvious potential relevance for the perovskite superconductors. The first, most naive approach is to use anisotropic GL theory. If we denote the in-plane coherence length by $\xi_\parallel$ and the perpendicular coherence length by $\xi_\perp$, one gets

$$\zeta^G=A\left[\frac{k_Bl}{\delta c\xi(0)_\perp\xi(0)_\parallel^2}\right]^2. \tag{10}$$

A more realistic model of a layered superconductor is that of a series array of Josephson junctions[8,12–14] in which the superconducting layers (or triplets in the case of $Y_1Ba_2Cu_3O_7$) are of atomic thickness arranged in parallel. Although quantum fluctuations in the phase of the order parameter will destroy superconductivity in a pure two-dimensional (2D) system, it was shown by Dzyaloskinskii and Kats[15] that very weak overlap of the electron wave functions of neighboring layers suppresses the phase fluctuations that disturb the long-range order in the pure 2D case. For materials exhibiting strong quasi-two-dimensional behavior, the coupling between planes will be characterized by an effective coherence length such that $[\xi(0)_\perp/s]<1$. Note that $\xi(0)_\perp$ is an exponentially decreasing function of the layer separation s. The integration of Eq. (5), which now is over two dimensions in reciprocal space, yields

$$\zeta^G\approx\left[\frac{k_B}{\delta cs\xi(0)_\parallel}\right]^2\left[\frac{s}{\xi(0)_\perp}\right]^2$$
$$\approx\left[\frac{\Delta}{\varepsilon_F}\right]^2\left[\frac{\Delta}{k_BT_c}\right]\left[\frac{s}{\xi(0)_\perp}\right]^2. \tag{11}$$

(The second equality assumes the clean limit in the plane with BCS parameters.) We see that ζ^G is now roughly the square root of what is was in the three-dimensional case, possibly even higher.

Since $\zeta^B\propto(\zeta^G)^{1/2}$, the Brout criterion is given by

$$\zeta^B\approx\left[\frac{k_B}{\delta cs\xi(0)_\parallel}\right]\left[\frac{s}{\xi(0)_\perp}\right]\approx\left[\frac{\Delta}{\varepsilon_F}\right]\left[\frac{s}{\xi(0)_\perp}\right], \tag{12}$$

where again the second equality is an in-plane clean limit BCS expression. Note that even in the case of $\Delta\ll\varepsilon_F$, it implies a wide Brout critical region.

We turn now to a discussion of the above results in view of the experimental data available on the new high-T_c superconductor Y-Ba-Cu-O. (A similar analysis can be made for La-Sr-Cu-O but less data exists.) In order to estimate the critical regions, we proceed in two ways. In the first, we calculate the critical regions on the basis of thermodynamic quantities such as $\delta c(T_c)$ or dH_c/dt at T_c using Eq. (10). In the second, we use BCS expressions and Eqs. (11) and (12). The results are summarized in Table I.

Measurements of the coherence lengths are essential to estimate the size of the critical regions. Several groups have estimated the coherence lengths from critical field and fluctuation conductivity (and diamagnetism) data.[16–24] The largest values ($\xi_{\parallel} \cong 37$ Å and $\xi_{\perp} \cong 7$ Å) were measured by Worthington *et al.*[16] The smallest values ($\xi_{\parallel} \simeq 16$ Å and $\xi_{\perp} \cong 3.6$ Å) were obtained by the Stanford group.[21] They are about a factor 2 smaller. Combining these values with the measured $\delta c(T_c)$ (Refs. 25–27) and $|dH_c/dT|_{T_c}$,[28] we obtain a range of estimates for ζ^B and ζ^G from thermodynamic data. They are the estimates in the upper part of Table I. We also note that if the smaller value of $\xi_{\perp}$ is correct, $YBa_2Cu_3O_7$ may be quasi-two-dimensional as $\xi_{\perp} < s$, where s is the distances between the centers of the triplets of Cu-O layers.

The available data on the high-T_c superconductors, based on analyzing transport in the different directions,[29] suggest that the clean limit applies. Thus using the BCS clean limit results, taking $(s/\xi_{\perp}) = 1$ and $\Delta/k_B T_c \sim 2.5$, which is the lowest value (i.e., most conservative) inferred from recent experiments,[30] we find $\zeta^G \approx (\Delta/\varepsilon_F)^2$ and $\zeta^B \approx (\Delta/\varepsilon_F)$. Thus estimation of the critical region depends largely on the ratio Δ/ε_F. Several estimates have been put forth.

Sound velocity measurements[31] and positron anihilation experiments[32] suggest that all the electrons at the Fermi surface participate in the condensate. If these conjectures are correct, $\Delta \approx \varepsilon_F$ and both ζ^G and ζ^B will be of order unity. Also as Laughlin and Hanna[33] have emphasized, for $La_{2-x}Sr_xCuO_4$ with $x = 0.15$, a carrier density of one carrier per dopant ($n = 1.84 \times 10^{21}$) one gets $\varepsilon_F = 0.5$ eV for a free-electron approximation in 3D. If we ignore the interlayer coupling and calculate the Fermi energy for a 2D case, $\varepsilon_F = 0.25$ eV.[34] The gap as measured by tunneling or infrared experiments is found to be in the range of $\Delta = 8 \div 17$ mV.[35] We thus get Δ/ε_F ranging between 0.016 and 0.068. This is very marginal for the Ginzburg criterion, but could lead to a Brout critical region $\zeta^B \approx 0.1$, or 10 K. If one assumes a polaron of some kind with an effective mass of $6m_0$, one gets that Δ/ε_F ranges from 0.2 to 0.8. In this case there clearly would be a complete breakdown of mean-field theory in an observable region near T_c. This result suggests that even in La-Sr-Cu-O some caution may be in order in using BCS and/or GL relationships to analyze data near T_c.

The situation with Y-Ba-Cu-O is similar as can be seen from Table I. The data presented there have been collected from Refs. 16–36 depending on the particular quantity. Since the carrier density is twice as large,[35,36] ε_F will be approximately twice its value in the La-Sr-Cu-O (in the 2D case) but Δ is also larger. Thus for $m/m_0 \approx 1$, the critical region will be $0.06 \div 1.6$ K. For heavy masses the GL critical region will be $2 \div 34$ K and the Brout critical region is much larger.

We note some of the consequences of a breakdown of mean-field theory. For a breakdown of only the microscopic mean field, we expect the same critical behavior as in BCS theory but with different coefficients. In particular, in the range where $\zeta^B < 1$, T_c would be shifted[37] such that $2\Delta/k_B T_c \cong 3.52(1+\zeta^B)$. This is an intriguing point since it shows that $2\Delta/k_B T_c$ can be large even without invoking strong coupling superconductivity. Consequently the jump of the specific heat at T_c will also increase. The experimental data on the specific heat are too broad to decide whether a regular BCS shape is being measured with the correct BCS amplitude.

In the case of the complete breakdown of the mean-field theory, the critical behavior (near T_c) should also be modified. In particular, the upper critical field should behave now as

$$H_{c2} = \frac{\Phi_0}{2\pi\xi(0)^2}\left(\frac{T_c - T}{T_c}\right)^{2\nu}, \tag{13}$$

with $2\nu > 1$. If the transition is a 3D x-y transition (as in liquid helium) $2\nu \approx 1.3$. Curvature in $H_{c2}(T)$ is common in these materials (and in particular near zero resistance where fluctuations are expected), and was found with the predicted exponent even for high-quality materials[17,21] but again the role of homogeneity must be considered. Specifically we have to remember that granularity due to weak links can lead to an exponent artificially greater than one.

The penetration depth λ diverges near T_c with the superfluid density exponent[11,38] $\zeta/2 \approx \beta$, (β is the order parameter exponent). Thus the Ginzburg-Landau param-

TABLE I. Calculated critical regions based on experimental data for $YBa_2Cu_3O_7$.

Experiment	ζ^G_{min}	ζ^G_{max}	ζ^B_{min}	ζ^B_{max}
Specific-heat jump at T_c[a] $\xi(0)_{\perp} = 3.6$ Å, $\xi(0)_{\parallel} = 16$ Å[b]	0.35	0.7	0.6	0.85
Specific-heat jump at T_c[a] $\xi(0)_{\perp} = 7$ Å, $\xi(0)_{\parallel} = 37$ Å[c]	0.01	0.03	0.1	0.2
Thermodynamic critical field[d] $\xi(0)_{\perp} = 3.6$ Å, $\xi(0)_{\parallel} = 16$ Å[b]	0.006	0.01	0.08	0.1
Thermodynamic critical field[e] $\xi(0)_{\perp} = 7$ Å, $\xi(0)_{\parallel} = 37$ Å[c]	2×10^{-4}	5×10^{-4}	0.015	0.025
BCS clean limit, using Δ, ε_F, and 2D electron gas, $m = m_0$[f]	4×10^{-4}	0.01	0.02	0.1
BCS clean limit, using Δ, ε_F, and 2D electron gas, $m = 6m_0$[f]	0.04	0.65	0.2	0.8

[a]References 25–27.
[b]Reference 21.
[c]Reference 16.
[d]Reference 28.
[e]Reference 24.
[f]References 30–36.

eter κ becomes

$$\kappa(T)=\frac{\lambda(T)}{\xi(T)}=\kappa_0\left[\frac{T_c-T}{T_c}\right]^{\nu-\beta}. \quad (14)$$

κ_0 is approximately the mean-field Ginzburg-Landau parameter. It was measured to be $\kappa_0 \approx 100$ in these new high-T_c materials. If we take the superfluid He exponents $\nu-\beta\approx 0.33$, the calculation of κ indicates that a crossover from a type-II to a type-I superconductor will hardly be observable in these materials (the temperature range will be $=10^{-6}T_c$). We can therefore conclude that with the assumption of a simple 3D x-y transition, no first-order transition along the H_{c2} line will be observable. Consequently, fluctuations of the electromagnetic field will not affect the nature of the transition.[39,40]

In summary, we have discussed in this paper the various fluctuation effects on the new high-T_c superconductors. We suggest that it is very plausible that mean-field critical behavior is not adequate to describe the superconducting transition. A possible theory is that of liquid ^{4}He. More generally the applicability of the microscopic coefficients in the analysis of experiments on these superconductors near T_c has been drawn into question. It is shown that large corrections may be needed to incorporate the strong fluctuations within the range of the Ginzburg-Landau kernel. The problem of paraconductivity and that of diamagnetic fluctuations above the superconducting transition are also of chief importance in understanding the nature of the superconducting state in these new materials.

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