Critical fluctuations in high-T_c superconductors

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(Received 28 May 1987)

The Ginzburg criterion suggests that the Ginzburg-Landau theory will break down within ~ 0.1 K of the transition temperature of the new high-T_c superconductors. Theoretical consequences of this include a weak divergence in the specific heat at T_c, and a correlation length which diverges as $(T-T_c)^{-\nu}$ with $\nu \cong 0.67$. Conventional Ginzburg-Landau formulas for critical fields and penetration depths, and mean-field predictions for fluctuation-enhanced quantities, must also be modified close to T_c.

The Ginzburg-Landau (GL) theory is enormously successful in explaining the properties of conventional superconductors,[1] but fails in explaining many other second-order phase transitions.[2] This is because, strictly speaking, the GL theory neglects fluctuations—an approximation which is adequate for conventional bulk superconductors. Even when fluctuations are added on to the GL theory,[3] as is done for treating corrections to the conductivity and diamagnetism near the transition temperature, the fluctuations are assumed small, and are thus treated approximately.

The GL theory assumes that the free-energy density may be expanded in terms of the order parameter ψ by[1]

$$f=\alpha_0\left(\frac{T-T_{c0}}{T_{c0}}\right)|\psi|^2+\tfrac{1}{2}\beta|\psi|^4+\frac{1}{2m^*}\left|\frac{\hbar}{i}\nabla\psi\right|^2, \tag{1}$$

where the magnetic field is taken to be zero, the free energy is measured relative to the normal-state free energy, and T_{c0} is the mean-field transition temperature.

As mentioned above, fluctuations can be treated approximately in the GL theory, as long as they are small. The Ginzburg criterion is derived by asking when the GL predictions for fluctuations in ψ become of the same order as ψ itself. This yields a temperature range close to T_c in which the GL theory is not expected to be valid:[4]

$$\frac{|T-T_{c0}|}{T_{c0}}<\frac{1}{32\pi^2}\frac{\beta^2}{\alpha_0}\left(\frac{2m^*}{\hbar^2}\right)^3(kT_{c0})^2 . \tag{2}$$

[Equation (2), and all subsequent equations, are for three-dimensional systems.] Outside of the temperature interval defined by (2), the GL theory will hold (provided, of course, that $|T-T_{c0}|/T_{c0}$ is still small). Inside of this temperature interval (which is referred to as the critical region), the GL theory breaks down.[5]

This equation can be evaluated for superconductors by using the GL results $\alpha_0^2/2\beta=H_c^2(0)/8\pi$ and $\hbar^2/2m^*\alpha_0=\xi^2(0)$, where

$$H_c(T)\equiv H_c(0)[(T_{c0}-T)/T_{c0}] ,$$

and

$$\xi(T)\equiv\xi(0)[(T_{c0}-T)/T_{c0}]^{-1/2} ,$$

near T_{c0}. Substituting these results into (2) gives

$$\frac{|T-T_{c0}|}{T_{c0}}<\frac{1}{2}\left(\frac{kT_{c0}}{H_c^2(0)\xi^3(0)}\right)^2 . \tag{3}$$

In a type-II superconductor, the thermodynamic critical field H_c can be expressed in terms of the measurable upper critical field H_{c2} by $H_c(T)=H_{c2}(T)/\sqrt{2}\kappa$, where κ is the ratio of the penetration depth λ to the coherence length ξ. This result, and the GL result $H_{c2}(T)=\Phi_0/2\pi\xi^2(T)$, where $\Phi_0=hc/2e$ is the superconducting flux quantum, combine with (3) to give

$$\begin{aligned}|T-T_{c0}| &< \frac{1}{2}\frac{32\pi^3\kappa^4}{\Phi_0^3}\frac{(kT_{c0})^2}{H_{c2}(0)}T_{c0}\\ &< 1.07\times10^{-9}\frac{\kappa^4T_{c0}^3}{H_{c2}(0)} .\end{aligned} \tag{4}$$

In the second part of (4), T_{c0} is measured in kelvin and $H_{c2}(0)$ is measured in G. $H_{c2}(0)$ is not the experimental upper critical field at $T=0$, but the field obtained by extrapolating the linear part of $H_{c2}(T)$ near $T=T_{c0}$ [but *outside* the range defined by (4)] down to $T=0$. If a superconductor is anisotropic, κ and $H_{c2}(0)$ are replaced by their geometric means in (4).

The strong dependence of (4) on T_{c0} suggests that high-temperature superconductors should have wider critical regions than conventional superconductors. Typical parameters for conventional superconductors are $\kappa\approx10$, $T_{c0}\approx10$ K, and $H_{c2}(0)\approx10^4$ G, which gives $|T-T_{c0}|<10^{-6}$ K in order to observe non-GL behavior. By contrast, transition temperatures in the 90–100 K range are reported for the new materials.[6] Extrapolated $H_{c2}(0)$ values vary,[7] but are typically between 500 and 1000 kG. Values of $\xi(T)$ can be obtained from $H_{c2}(T)$; combined with values for $\lambda(T)$, which can be inferred from $H_{c1}(T)$, these give an experimental value for κ. A value of $\kappa=100$ is plausible, but lower values (~50) and higher values (~200) have been reported, or can be inferred, from measurements on the 40-K superconductors and the 90-K superconductors.[7,8] Using $\kappa=100$, $H_{c2}(0)=750$ kG, $T_{c0}=95$ K gives $|T-T_{c0}|<0.12$ K. If $\kappa=200$, $|T-T_{c0}|<1.96$ K; a smaller value of $H_{c2}(0)$ is not inconceivable and could increase the extent of the critical region by a factor of 2 more. (If $\kappa=50$, the critical region shrinks to approximately 8 mK.[9]) It is, of course, possible to raise κ by shortening the electronic mean free path.

Inside the critical region, the behavior of a superconductor is quite different from the behavior outside. (A number of important differences are summarized in Table I.) The phase transition will occur at a temperature T_c which will be, in general, different from the GL transition temperature T_{c0}. The thermodynamic properties of a superconductor within the critical region are the same as a three-dimensional *XY* model, and can be seen experimentally[2] in the superfluid transition in ^{4}He. The specific heat should have a weak power-law divergence at T_c, in contrast to the discontinuity of the GL theory. This divergence can be written as $C \sim |T-T_c|^{-\alpha}$ with $\alpha \cong 0$. Since the specific heat is proportional to the second derivative of the free energy with respect to temperature, this says that $f \sim |T-T_c|^{2-\alpha}$. An immediate consequence of this last result is that the thermodynamic critical field, which is proportional to $f^{1/2}$, varies as $H_c(T) \sim (T_c - T)$ below T_c, just as in the GL theory, because α is small.

A second important difference between critical behavior and GL behavior is that the correlation length $\xi(T) \sim (T-T_c)^{-\nu}$, with $\nu \cong 0.67$. [In the GL theory, the correlation length diverges as $(T-T_c)^{-1/2}$.] Energetic arguments imply that the upper critical field should vary as Φ_0/ξ^2, independent of whether the GL theory holds, which implies that $H_{c2}(T) \sim (T_c - T)^{1.34}$, in contrast to the linear GL temperature dependence. It is interesting to note that measured upper critical fields[7] have an upward curvature close to T_c, crossing over to linear behavior at lower temperatures, although this may be due to sample inhomogeneity.

The order parameter ψ, which is proportional to the square root of the superfluid density n_s, varies as $\psi(T) \sim (T_c - T)^{\beta}$ close to T_c, with $\beta \cong 0.33$. (The GL result is, again, a square-root dependence.) As long as the superconductor is in the local limit, this suggests that $\lambda(T) \propto 1/n_s^{1/2} \sim (T_c - T)^{-\beta}$.

Unlike the GL case, where λ and ξ have the same temperature dependence near T_c, in the critical region the correlation length diverges faster than the penetration depth. The parameter $\kappa = \lambda/\xi \sim (T_c - T)^{0.34}$ is no longer temperature independent, but goes to zero as T approaches T_c. Close enough to T_c, superconductors will become type I. This suggests that magnetization curves close to T_c can be used to observe the crossover to critical behavior. In the GL regime, H_{c1} and H_{c2} have the same linear dependence on $(T_{c0} - T)$. Closer to T_c, $H_{c1}(T) \sim (T_c - T)^{0.66}$ while $H_{c2}(T) \sim (T_c - T)^{1.34}$; the deviations from GL behavior are in opposite directions.

The analogy to ^{4}He, plus dynamic scaling arguments,[10] can be used to make estimates of other properties. In the absence of a more complete theory, dimensional analysis and physical arguments will be used to obtain formulas for fluctuation diamagnetism and fluctuation-enhanced conductivity above T_c.

Fluctuation diamagnetism is simpler to deal with because it does not require knowledge of the time dependence of fluctuations. In an argument first suggested by Schmid,[1,3,11] a three-dimensional superconductor above T_c can be viewed as a collection of small droplets, of size $\xi(T)$, which fluctuate independently. Neglecting factors of order unity, this leads to a susceptibility χ',

$$\chi' = -(kT/\Phi_0^2)\xi(T) \; , \tag{5}$$

which will diverge as $(T-T_c)^{-0.67}$. The GL result $\chi' \sim (T-T_{c0})^{-1/2}$, which should be distinguishable from (5), is expected to hold outside the critical region. Corrections may need to be made to (5) for the effects of finite external field, as has been done for the GL case.[12]

Fluctuation-enhanced conductivity is more complicated because it depends on the time dependence of fluctuations. In the mean-field regime, the fluctuations cause the electrical conductivity to increase by an amount σ' which is given by $\sigma' \propto \xi(T)$, according to Aslamazov and Larkin (AL).[1,3,13] This suggests that σ' should vary as $(T-T_{c0})^{-1/2}$ in the mean-field regime, crossing over to $(T-T_c)^{-0.67}$ in the critical region defined by (4). This *differs* from the thermal conductivity of ^{4}He, which experimentally[14] diverges as $(T-T_c)^{-0.334 \pm 0.005}$. A full dynamical scaling theory[10] predicts that the thermal conductivity varies as $\xi^{1/2} \sim (T-T_c)^{-0.33}$, which agrees well with experiment. This difference occurs because the temperature dependence of relaxation times changes close to T_c. It is thus expected that

$$\sigma' \cong \frac{e^2}{h\xi(0)} \left(\frac{T_c}{T-T_c} \right)^{a(T)} , \tag{6}$$

where $a(T) \cong \frac{1}{2}$ outside the critical region, crossing over to $a(T) \cong 0.67$ in the static critical region, and finally crossing over to $a(T) \cong 0.33$, closer still to T_c, where dynamical scaling effects come into play. Corrections may need to be made to account for the Maki-Thompson term, especially for samples in the clean limit.

Freitas, Tsuei, and Plasket[15] have recently reported measurements of fluctuation diamagnetism and fluctuation-enhanced conductivity in $Y_1Ba_2Cu_3O_{9-\delta}$. The data presented in their figures, which extends from about 1 K above T_c to more than twice T_c, are well fit by

TABLE I. Exponents which characterize the temperature dependence of various physical quantities near T_c. Critical exponents characterize the transition close to T_c; farther from T_c, the GL exponents should be seen. The σ' exponent crosses over twice as T approaches T_c, from $-\frac{1}{2}$ to -0.67, and then from -0.67 to -0.33 still closer to T_c.

Quantity	Critical exponent	GL exponent
C	$-\alpha \cong 0$	(discontinuity)
ξ	$-\nu \cong -0.67$	$-\frac{1}{2}$
ψ	$\beta \cong 0.33$	$\frac{1}{2}$
λ	$-\beta \cong -0.33$	$-\frac{1}{2}$
H_c	$1-\alpha/2 \cong 1$	1
H_{c1}	$2\beta \cong 0.66$	1
H_{c2}	$2\nu \cong 1.34$	1
χ'	$-\nu \cong -0.67$	$-\frac{1}{2}$
σ'	$-\nu \cong -0.67 \rightarrow -\nu/2 \cong 0.33$	$-\frac{1}{2}$

the classical AL and Prange theories, as might be expected since most of their data are outside the critical region defined by (4). They do, however, note deviations from the Al theory for $(T-T_c)<0.5$ K, which is suggestively close to the estimates given for the critical region here. Further measurements are clearly needed. For comparing theory with experiment, it is useful to note that (5) and (6) predict different deviations from the GL-based theories. The susceptibility χ is expected to diverge faster than the Schmidt-Schmid result, while the conductivity is expected to initially diverge faster, crossing over to a slower divergence very close to T_c.

In summary, the high values of T_c and κ in the new high-temperature superconductors should cause the GL theory to break down within 0.1 K or more of the transition temperature. The behavior of these materials close to T_c should be analogous to the critical behavior of ^{4}He near the λ transition. Existing experimental results contain hints of such behavior, but samples which have a narrower distribution of T_c's (due to inhomogeneity), and measurements closer to T_c, are needed. The accessibility of the critical region creates a variety of opportunities for experiments on more exotic effects. Depending on material-dependent parameters, it may be possible to observe a predicted weakly first-order transition[16] closer still to T_c than the region described here, when the crossover to type-I behavior has occurred. If, on the other hand, type-II behavior persists close enough to T_c, an inverted-XY transition is expected.[17] Finally, in a magnetic field, a type-II superconductor is expected to have a first-order transition at H_{c2}.[18]

After this manuscript was submitted, I received a copy of unpublished results[19] describing specific-heat measurements in $YBa_2Cu_3O_{7-\delta}$. The authors of that paper estimate the Ginzburg criterion using the specific-heat jump, and obtain a predicted critical region of a few mK, close to the smallest estimate obtained above. The reason for the difference between the specific-heat estimate and the critical-field estimates presented here is probably the sixth-power dependence of the Ginzburg criterion on correlation length $\xi(0)$; more accurate measurements are needed to resolve this discrepancy.

I am extremely grateful to B. I. Halperin and G. C. Spalding for their comments and suggestions in the course of this work, and to M. Tinkham, M. G. Forrester, and S. Benz for their critical readings of the manuscript. I am also grateful to D. Rudman for providing some of the experimental parameters for high-T_c materials. This work was supported in part by the National Science Foundation through Grant No. NSF-DMR84-04489.

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[9]These estimates of the extent of the critical region are based on parameters measured in polycrystalline samples. Using the plausible assumption that measured values of the critical fields are roughly twice the critical fields in the weak directions, rather than an average of the critical fields over direction, it is found that an underestimate of the geometric mean of κ is made. To correct for this, the estimates of the critical region should be multiplied by roughly the square root of the critical-field anisotropy, which increases the predicted critical region by a factor of 3 or so.

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