Penetration Depth Measurements of 3D *XY* Critical Behavior in $YBa_2Cu_3O_{6.95}$ Crystals

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We report measurements of the electromagnetic penetration depth $\lambda(T)$ in nominally pure crystals of $YBa_2Cu_3O_{6.95}$, for temperatures close to the critical temperature T_c. Over the range $0.001 < (T_c - T)/T_c < 0.1$, we find that $\lambda(T) \propto (1 - T/T_c)^{-y}$ with $y \approx 0.33$, consistent with the critical behavior of the three dimensional *XY* model. The measured critical behavior is not affected by the presence of small amounts of Zn impurities, in agreement with the Harris criterion.

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Rather than exhibiting critical behavior, the physical properties of bulk, classic superconductors near the superconducting transition temperature T_c are generally well described by Ginzburg-Landau theory. Estimates of the Ginzburg temperature T_G, at which Ginzburg-Landau theory is expected to break down, indicate that critical behavior in classic superconductors is restricted to a temperature range that is inaccessibly close to T_c [1,2]. However, it has been recognized for some time that the small coherence length in the high-temperature superconductors leads to the possibility of observing fluctuations and perhaps critical scaling near the superconducting transition [1–3]. For a strongly type II superconductor, such as $YBa_2Cu_3O_{6.95}$, theory predicts [3] that the effective charge in the Ginzburg-Landau theory is negligible outside an inaccessibly small asymptotic regime close to T_c. Thus, for the purpose of understanding strong fluctuations in experimentally relevant temperature ranges, the order parameter may be regarded as neutral, and in the simplest case of a single complex order parameter would give rise to fluctuations in the universality class of the three dimensional (3D) *XY* model.

There has been considerable recent interest in the possibility of unconventional pairing states in the high-T_c cuprate materials [4]. Some of the possibilities under consideration involve order parameters corresponding to mixtures of irreducible representations of the symmetry group of the normal state, notably $s + id$ and $d + id'$, where s and d refer to order parameters with pure s- or d-like symmetry. Such states will either give rise to fluctuations in a universality class other than 3D *XY* near T_c if the two representations are accidentally degenerate, or produce a second transition at a lower temperature. Since no second transition has been observed, observation of 3D *XY* critical behavior would suggest a single, complex order parameter.

Early measurements of transport and thermodynamic response in zero external magnetic field were interpreted in terms of Gaussian fluctuations about a mean field background [5], although later analysis suggested the possibility of critical scaling in the universality class of the three dimensional (3D) *XY* model [6]. More recent measurements in the presence of a magnetic field have also been interpreted as evidence for 3D *XY* critical scaling [7–10]; in these measurements, the presence of magnetic field effectively reduces the dimensionality of the system, with the result that the critical region is enlarged. In all of these works, the analysis of the data is invariably complicated by the necessity to perform a background subtraction, which is difficult to quantify reliably, in order to extract the fluctuation contribution.

In this Letter, we report microwave measurements of the a-b plane electromagnetic penetration depth $\lambda(T)$ near the critical temperature of crystals of $YBa_2Cu_3O_{6.95}$. The penetration depth is a direct measure of the superfluid density $\rho_s \propto \lambda^{-2}$, which tends to zero as $T \to T_c^-$ in a manner determined by critical fluctuations. Thus, in contrast to the previous thermodynamic and transport studies [7–10], no background subtraction is required to discern any fluctuation contributions to $\lambda(T)$.

The majority of microwave studies of surface resistance and $\lambda(T)$ have been performed on thin films [11], but the question of the role of defects in the electromagnetic properties of $YBa_2Cu_3O_{7-\delta}$ led us earlier to study high quality crystals [12]. Initial measurements were focused on the linear temperature dependence of $\lambda(T)$ found at low temperatures in nominally pure crystals, but it was also apparent that $\rho_s = \lambda^2(0)/\lambda^2(T)$ rises unusually rapidly below T_c [12]. In fact, we show in the present study that the measurements provide unambiguous evidence for critical fluctuations near T_c, consistent with the 3D *XY* universality class.

In our earlier studies, the influence of disorder on the low temperature behavior of $\lambda(T)$ was examined by doping with Ni and Zn [13] in order to test a prediction based upon the hypothesis that the pairing state was d wave [14]. Here we report that disorder is found to have little influence near T_c, consistent with the Harris criterion [1,15], thus further strengthening the claim for critical behavior. We emphasize that in this Letter we only refer to the zero field scaling regime intermediate between mean field theory and the asymptotic scaling regime inaccessibly close to T_c, where fluctuations of the electromagnetic field must also be taken into account [16].

The $YBa_2Cu_3O_{6.95}$ crystals used in this study were prepared by a flux-growth technique described in detail elsewhere [17]. The sharpness of the specific heat jump at T_c, which provides a measure of the *bulk* homogeneity of a crystal, is an indicator of sample quality that has particular significance in studies of critical behavior. A crystal produced by the technique used here exhibited a jump that was only 0.25 K wide (defined by the 10% to 90% points in the jump in C_p/T) [17], comparable to the narrowest transitions reported by Regan *et al.* for a crystal of $YBa_2Cu_3O_{6.95}$ [18] and by Buan *et al.* for a twin-free crystal of $LuBa_2Cu_3O_{7-\delta}$ [19].

The experimental technique, a microwave technique discussed in detail elsewhere [12,13], measures changes in the penetration depth with better than 1 Å resolution. The measurements involve cavity perturbation of a 900 MHz superconducting split-ring resonator with a thin platelike crystal aligned so that $\vec{H}_{\rm rf} \perp \hat{c}$. This geometry primarily measures the *a-b* plane penetration depth, $\lambda_\perp$, provided that the crystal is very thin. Placing a sample into a microwave cavity leads to a perturbation $\Delta\omega$ of the resonant angular frequency ω that can be expressed as

$$\frac{\Delta\omega}{\omega} = \frac{\Delta f}{f} - i\frac{1}{2}\Delta\left(\frac{1}{Q}\right), \tag{1}$$

where Δf is the shift in the resonant frequency f and $\Delta(1/Q)$ is the change in the inverse of the quality factor of the cavity. For a slab of thickness $2c$ and infinite area, the perturbation in our measurement geometry is [12]

$$\frac{\Delta\omega}{\omega} = \frac{1}{2}\frac{V_s}{V_r}\left[1 - \frac{\tanh kc}{kc}\right], \tag{2}$$

where k, the propagation constant, is related to the complex conductivity, $\sigma = \sigma_1 + i\sigma_2$, by

$$k = (1-i)\sqrt{\frac{\omega\mu_0\sigma}{2}}. \tag{3}$$

V_s is the volume of the sample and V_r, the effective volume of the resonator, is determined by measuring a Pb:Sn alloy sample in the normal and superconducting states [13].

The first term in Eq. (2), $V_s/2V_r$, is the dominant geometric frequency shift associated with the displacement of microwave fields by the entire sample volume. In order to measure the temperature dependence of $\lambda(T)$ very precisely, we mount the sample rigidly inside the resonator and measure the temperature dependence of the perturbation relative to that for the sample at 1.3 K. This helps eliminate the problem of spurious frequency shifts due to sample motion altering the $V_s/2V_r$ term. In this case, the relevant expression for the perturbation is

$$\frac{\delta\omega}{\omega} = -\frac{1}{2}\frac{V_s}{V_r}\frac{\tanh kc}{kc}, \tag{4}$$

where we use $\delta\omega$ rather than $\Delta\omega$ to denote the shift relative to that of a sample that perfectly screens out the microwave fields. In terms of our measured quantities, this perturbation is given by

$$\frac{\delta\omega}{\omega} = \left(\frac{\delta f}{f} - \frac{A\lambda(1.3\ \mathrm{K})}{V_r}\right) - i\frac{1}{2}\delta\left(\frac{1}{Q}\right), \tag{5}$$

where δf and $\delta(1/Q)$ are the measured shifts relative to those at $T = 1.3$ K. The term $A\lambda(1.3\ \mathrm{K})/V_r$ is a correction to account for the fact that the fields are not screened perfectly at 1.3 K; they penetrate a distance $\lambda(1.3\ \mathrm{K})$, close to the low temperature limit of the London penetration depth. There is no need to correct the Q measurement for the sample's microwave losses at 1.3 K because at low temperature they are much too small at 900 MHz to be measurable in our cavity. Combining Eqs. (4) and (5) yields

$$\left(\frac{\delta f}{f} - \frac{A\lambda(1.3\ \mathrm{K})}{V_r}\right) - i\frac{1}{2}\delta\left(\frac{1}{Q}\right) = -\frac{1}{2}\frac{V_s}{V_r}\left(\frac{\tanh kc}{kc}\right). \tag{6}$$

Using the measured values of $\delta f/f$ and $\delta(1/Q)$ and an assumed value for $\lambda(1.3\ \mathrm{K})$, this equation can be solved numerically for k from which σ_1 and σ_2 can be easily extracted. For all of the data in the superconducting state shown here, $\sigma_2 \gg \sigma_1$, and the penetration depth is determined by $\lambda = (\mu_0\omega\sigma_2)^{-1/2}$. This relationship between σ_2 and λ is valid provided that measurements are made at a frequency low enough that $\omega\tau \ll 1$, where τ is the relaxation time of thermally excited quasiparticles in the superconducting state. Near T_c this lifetime is roughly $\tau \sim \hbar/2k_BT_c$, ensuring that $\omega\tau \ll 1$ at microwave frequencies. However, in the far infrared this is not the case and strong temperature and frequency dependence of the relaxation time can influence the apparent behavior of λ at high frequency [20].

Finally, we must choose a value for $\lambda(1.3\ \mathrm{K})$ in order to generate $\lambda(T)$ from our measurements. Both far-infrared [20] and muon spin rotation [21] measurements on crystals similar to the ones used here place the low temperature penetration depth between 1350 and 1450 Å. We will adopt a value of 1400 Å in the following presentation of the microwave measurements; none of our results depend sensitively on the choice of $\lambda(1.3\ \mathrm{K})$.

Now we turn to a discussion of the data. In the inset of Fig. 1 is plotted the superfluid density $[\lambda(0)/\lambda(T)]^2$ as a function of temperature T for nominally pure $YBa_2Cu_3O_{6.95}$. The plot exhibits noticeable curvature near T_c, whereas linear behavior would have been expected if the penetration depth were to follow the

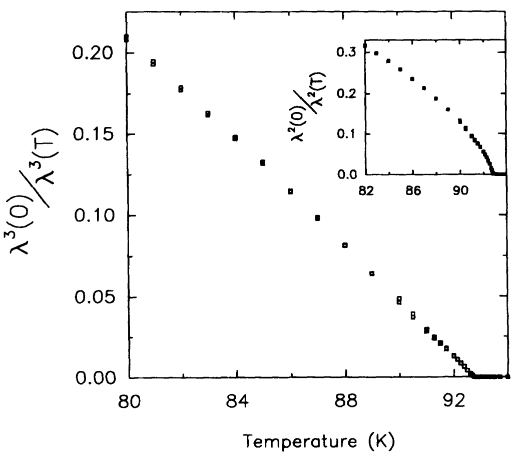

FIG. 1. Inset: $[\lambda(0)/\lambda(T)]^2$ versus T for nominally pure $YBa_2Cu_3O_{6.95}$. Main figure: The same data, but $[\lambda(0)/\lambda(T)]^3$ versus T. The value of $\lambda(0)$ was taken to be 1400 Å.

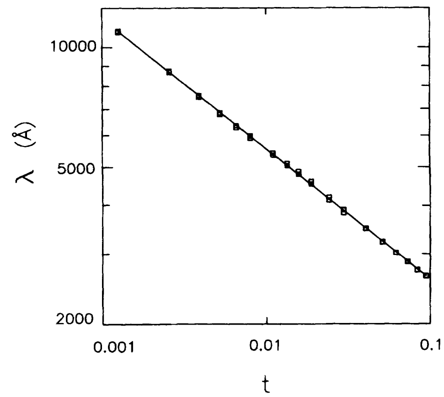

FIG. 2. A log-log plot of $\lambda^{-1}(T)$ versus the reduced temperature, $t \equiv (T_c - T)/T_c$. With the critical temperature of 92.74 K used in this figure, power-law behavior $\lambda(T) = \lambda_{\perp 0} t^{-y}$ is observed over two decades in reduced temperature. The solid line is a fit to the data, yielding $\lambda_{\perp 0} = 1186$ Å and an exponent $y = 0.33 \pm 0.01$ that is consistent with the critical behavior expected for the 3D XY model.

Ginzburg-Landau behavior $\lambda(T)/\lambda(0) \propto t^{-1/2}$, with the reduced temperature being $t \equiv (T_c - T)/T_c$. The main part of Fig. 1 shows the same date replotted with the vertical axis $[\lambda(0)/\lambda(T)]^3$. The linear behavior for about 9° below T_c indicates that $\lambda(T)/\lambda(0) \propto t^{-y}$ with $y \approx 1/3$.

Figure 2 shows $\lambda(t)$ versus t on a log-log plot, using $T_c = 92.74$ K, the value of the critical temperature obtained by extrapolating the $t^{-1/3}$ behavior shown in Fig. 1. Because of the slight breadth of the superconducting transition there is a small uncertainty, about ± 0.03 K, in the appropriate choice of T_c. This uncertainty only has a serious influence on the point near $t = 0.001$ in the log-log plot, leaving nearly two decades in t over which power-law behavior is unambiguously observed. A fit to the data shown in Fig. 2 yields power-law behavior, $\lambda(t) = \lambda_{\perp 0} t^{-y}$ with $y = 0.33 \pm 0.01$. The uncertainty in the exponent represents the range of values obtained for the range of choices of T_c. This value of the exponent is consistent with the notion that the data are well described by a 3D XY critical regime over a temperature range of order 10 K. This range agrees well with the range over which the electronic specific heat has been claimed to vary logarithmically with temperature [6,18].

Figure 3 shows the effect of small concentrations of Zn impurities. The value of the exponent seems to be unchanged from that of the nominally pure sample and the temperature range over which the critical behavior is observed changes little with impurity content. The observation that the exponent is not affected by the addition of low levels of impurities is consistent with the identification of this behavior as a 3D XY critical regime; there, the heat capacity exponent is slightly negative, and thus the Harris criterion predicts that the critical behavior is not affected by weak disorder [1,15].

Finally, we mention two possible complications to the simple interpretation given here to the data. The first is the possibility of a crossover to two dimensional behavior. A rough criterion for the observation of three dimensional, rather than two dimensional, behavior is that the correlation length ξ_z in the c direction, as defined in [3], be greater than half of the interlayer spacing. In the 3D XY critical regime the a-b plane correlation length and penetration depth for $T \to T_c^-$ are given by $\xi_\perp(t) = \xi_{\perp 0} t^{-\nu}$ and $\lambda_\perp(t) = \lambda_{\perp 0} t^{-\nu/2}$, respectively, with $\nu \approx 2/3$. The coefficients are related by a universal amplitude ratio, which follows from two-scale-factor universality [3,22]:

$$\frac{\lambda_{\perp 0}^2}{\xi_{\perp 0}} = \frac{\gamma \Lambda_{T_c}}{\pi c_s}, \tag{7}$$

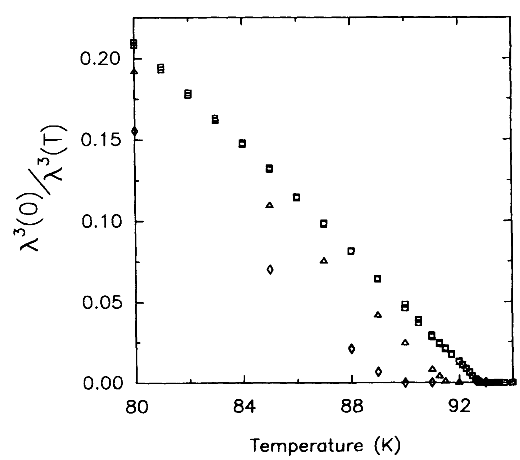

FIG. 3. $[\lambda(0)/\lambda(T)]^3$ versus T for $YBa_2(Cu_{1-x}Zn_x)_3O_{6.95}$ with $x = 0$ (squares), 0.0015 (triangles), and 0.0031 (diamonds). The addition of Zn impurities does not appreciably affect the power-law behavior below T_c.

where Λ_{T_c} is 2×10^8 Å K/T_c and c_s is a universal constant that has been estimated to be 0.5 [22]. ξ_z is related to $\xi_\perp$ by $\xi_z = \gamma \xi_\perp$ with an anisotropy parameter $\gamma \approx 0.2$ for $YBa_2Cu_3O_{6.95}$. Thus, in the 3D *XY* critical regime $\xi_z(t) \simeq c_s \pi \lambda_\perp^2(t)/\Lambda_{T_c}$, which gives $\xi_z = 4.8$ Å at $t = 0.1$ and 103 Å at $t = 0.001$. If one assumes that the CuO_2 bilayers are well coupled, then half the interlayer spacing is 5.9 Å and the above estimate of ξ_z implies that the condition for three dimensional critical fluctuations is satisfied over the range from $t = 0.001$ to 0.1. There is in principle the possibility of a gradual crossover to 2D fluctuations at lower temperatures, although the fluctuation dominated regime may not extend that far.

The second possible complication is that of strong-coupling effects. Although strong coupling can increase the slope of $\lambda^2(0)/\lambda^2(T)$ near T_c [23], the qualitative mean field behavior is retained; that is, $\lambda^2(0)/\lambda^2(T)$ remains linear in t. The curvature of the data near T_c shown in the inset of Fig. 1 is a qualitative deviation from both the weak-coupling BCS and strong-coupling mean field results.

In conclusion, we have presented unambiguous evidence for 3D *XY* critical scaling behavior in a temperature interval of order 10 K below T_c. This is critical behavior expected for a transition to a superconducting state with a single, complex order parameter such as pure s wave or $d_{x^2-y^2}$. The implications of our findings for the fluctuation contribution to the dc conductivity [24] will be presented elsewhere [25].

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