

## Evidence for charged critical fluctuations in underdoped Y Ba<sub>2</sub>Cu<sub>3</sub>O<sub>7-δ</sub>

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## LETTER TO THE EDITOR

**Evidence for charged critical fluctuations in underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$** **T Schneider<sup>1,5</sup>, R Khasanov<sup>1,2,3</sup>, K Conder<sup>3</sup>, E Pomjakushina<sup>3</sup>, R Bruetsch<sup>4</sup> and H Keller<sup>1</sup>**<sup>1</sup> Physik-Institut der Universität Zürich, Winterthurerstrasse 190, CH-8057, Switzerland<sup>2</sup> DPMC, Université de Genève, 24 Quai Ernest-Ansermet, 1211 Genève 4, Switzerland<sup>3</sup> Laboratory for Neutron Scattering, ETH Zürich and PSI Villigen, CH-5232 Villigen PSI, Switzerland<sup>4</sup> Laboratory for Material Behavior, PSI Villigen, CH-5232 Villigen PSI, Switzerland

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**Abstract**

We report and analyse in-plane penetration depth measurements in  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  taken close to the critical temperature  $T_c$ . In underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_{6.59}$  we find consistent evidence for charged critical behaviour. Noting that the effective dimensionless charge  $\tilde{e} = \xi/\lambda = 1/\kappa$  scales as  $T_c^{-1/2}$ , this new critical behaviour should be generically observable in suitably underdoped cuprates.

(Some figures in this article are in colour only in the electronic version)

Close to the critical temperature  $T_c$  of the normal–superconductor transition, in a regime roughly given by the Ginzburg criterion [1–4], order parameter fluctuations dominate critical properties. In recent years, the effect of the charge of the superconducting order parameter in this regime in three dimensions has been studied extensively [5–15]. While for strong type I materials the coupling of the order parameter to transverse gauge field fluctuations is expected to render the transition first order [6], it is well established that strong type II materials should exhibit a continuous phase transition, and that sufficiently close to  $T_c$  the charge of the order parameter is relevant [8–15]. However, in cuprate superconductors within the fluctuation dominated regime, the region close to  $T_c$ , where the system crosses over to the regime of charged fluctuations, turns out to be too narrow to access. For instance, optimally doped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ , while possessing an extended regime of critical fluctuations, is too strongly type II to observe charged critical fluctuations [1–4, 16]. Indeed, the effective dimensionless

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charge  $\tilde{e} = \xi/\lambda = 1/\kappa$  is in strongly type II superconductors ( $\kappa \gg 1$ ) small. The crossover upon approaching  $T_c$  is thus initially to the critical regime of a weakly charged superfluid where the fluctuations of the order parameter are essentially those of an uncharged superfluid or  $XY$  model [1]. Furthermore, there is the inhomogeneity induced finite size effect which renders the asymptotic critical regime unattainable [17, 18]. However, underdoped cuprates could open a window onto this new regime because  $\kappa$  is expected to become rather small. Here the cuprates undergo a quantum superconductor to insulator transition in the underdoped limit [4, 19–22] and correspond to a 2D disordered bosonic system with long-range Coulomb interactions. Close to this quantum transition  $T_c$ ,  $\lambda_{ab}$  and  $\xi_{ab}$  scale as  $T_c \propto \lambda_{ab}^{-2} \propto \xi^{-z}$  [4, 19–22], yielding with the dynamic critical exponent  $z = 1$  [4, 22–25],  $\kappa_{ab} \propto \sqrt{T_c}$ . Noting that  $T_c$  decreases by approaching the underdoped limit, sufficiently homogeneous and underdoped cuprates appear to be potential candidates to observe charged critical behaviour.

Here we report and analyse in-plane penetration depth measurements of underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  to explore the evidence for this new critical behaviour.  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  samples were synthesized by solid-state reactions using high-purity  $\text{Y}_2\text{O}_3$ ,  $\text{BaCO}_3$  and  $\text{CuO}$ . The samples were calcinated at 880–935 °C in air for 100 h with several intermediate grindings. The phase purity of the material was examined by powder x-ray diffraction. As synthesized, the samples have oxygen contents in the range 6.975–6.985. Starting with a material of maximal oxygen content a series of reduced samples has been produced. Our characterization revealed that the equilibration of the samples in closed ampoules with an appropriate amount of copper powder reacting with oxygen leads to the best results. Thus, for each sample in the series, an alumina crucible with Y123 powder of exactly known weight and oxygen content was placed in a quartz ampoule together with an exactly weighed copper powder in another crucible. To ensure a homogenous oxygen distribution the ampoule was sealed under vacuum and heated up to 700 °C (heating rate 10 °C h<sup>-1</sup>), kept at this temperature for 10 h and finally slowly cooled (5 °C h<sup>-1</sup>). Field-cooled (FC) magnetization measurements were performed with a Quantum Design SQUID magnetometer in a field of 0.5 mT for temperatures ranging from 5 to 100 K. The Meissner fraction  $f$  was deduced from the mass of the samples and the x-ray density, and assuming spherical grains. To calculate the temperature dependence of the effective penetration depth  $\lambda_{\text{eff}}(T)$  we used the Shoenberg formula [26, 27] assuming spherical grains of radius  $R$ , particle size distribution  $N(R)$  and volume fraction distribution  $g(R) = N(R)R^3$ ,

$$f(T) = \int_0^\infty \left( 1 - \frac{3\lambda(T)}{R} \cot\left(\frac{R}{\lambda(T)}\right) + \frac{3\lambda^3(T)}{R^2} \right) / \int_0^\infty g(R) dR. \quad (1)$$

We extracted the grain size distribution  $N(R)$  from an analysis of SEM (scanning electron microscope) photographs. Solving this nonlinear equation for given  $f(T)$  and  $g(R)$  we obtained the effective penetration depth  $\lambda_{\text{eff}}(T)$ . For sufficiently anisotropic superconductors ( $\lambda_c/\lambda_{ab} > 5$ ), including  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ ,  $\lambda_{\text{eff}}$  is proportional to the in-plane penetration depth, so that  $\lambda_{\text{eff}} = 1.31\lambda_{ab}$  [28]. The resulting data for  $\lambda_{ab}(T)$  agree well with the transverse-field  $\mu\text{SR}$  measurements performed on similar samples [29].

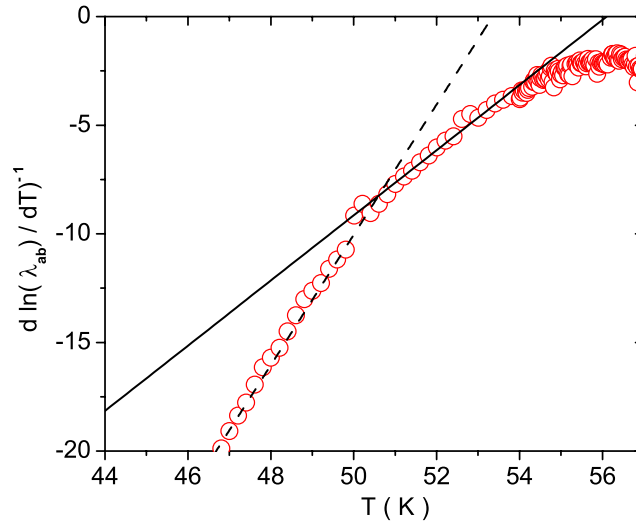
When charged fluctuations dominate, the in-plane penetration depth and the correlation length are related by [10–12, 14, 15]

$$\lambda_{ab} = \kappa_{ab}\xi_{ab}, \quad \lambda_{ab} = \lambda_{0ab}|t|^{-\nu}, \quad \nu \simeq 2/3, \quad (2)$$

in contrast to the uncharged case, where  $\lambda \propto \sqrt{\xi}$  and with that

$$\lambda_{ab} = \lambda_{0ab}|t|^{-\nu/2}, \quad (3)$$

where  $t = T/T_c - 1$ . In a plot  $(d \ln \lambda_{ab}/dT)^{-1}$  versus  $T$  charged critical behaviour is then uncovered in terms of a temperature range where the data fall on a line with slope  $1/\nu \simeq 3/2$ ,



**Figure 1.**  $(d \ln \lambda_{ab}/dT)^{-1}$  with  $\lambda_{ab}$  in  $\mu\text{m}$  versus  $T$  for  $\text{YBa}_2\text{Cu}_3\text{O}_{6.59}$ . The straight line with slope  $1/\nu \simeq 3/2$  corresponds according to equation (2) to charged criticality with  $T_c = 56.1$  K, while the dashed line indicates the intermediate 3D  $XY$  critical behaviour with slope  $2/\nu \simeq 3$ .

while in the neutral (3D  $XY$ ) case it collapses on a line with slope  $2/\nu \simeq 3$ . Clearly, in an inhomogeneous system the phase transition is rounded and  $(d \ln \lambda_{ab}/dT)^{-1}$  does not vanish at  $T_c$ . In figure 1 we display  $(d \ln \lambda_{ab}/dT)^{-1}$  versus  $T$  for  $\text{YBa}_2\text{Cu}_3\text{O}_{6.59}$ , derived from the measured in-plane penetration depth data. The data uncover a crossover from uncharged critical behaviour (dashed line) to charged criticality (solid line) with  $T_c \simeq 56.1$  K, limited by a finite size effect due to the finite extent of the grains and/or inhomogeneities within the grains. Although charged criticality is attained there is no sharp transition. Indeed,  $\lambda_{ab}$  does not diverge at  $T_c$  because the correlation length  $\xi_{ab} = \lambda_{ab}/\kappa_{ab}$  cannot grow beyond the limiting length  $L_{ab}$  in the  $ab$ -plane.

To explore the evidence for charged critical behaviour and the nature of the finite size effect further, we display in figure 2  $1/\lambda_{ab}$  and  $d(1/\lambda_{ab})/dT$  versus  $T$ . The solid curve is  $\lambda_{ab} = \lambda_{0ab}|t|^{-\nu}$  with  $\lambda_{0ab} = 0.089 \mu\text{m}$ ,  $\nu = 2/3$  and  $T_c = 56.1$  K, appropriate for charged criticality, and the dashed one its derivative. Approaching  $T_c$  of the fictitious homogeneous system the data again reveal a crossover from uncharged to charged critical behaviour, while the tail in  $\lambda_{ab}$  versus  $T$  above  $d\lambda_{ab}/dT$  points to a finite size effect. Indeed,  $d\lambda_{ab}/dT$  does not diverge at  $T_c$  but exhibits an extremum at  $T_p \simeq 54.27$  K, giving rise to an inflection point in  $\lambda_{ab}(T)$  at  $T_p$ . Here the correlation length attains the limiting length.

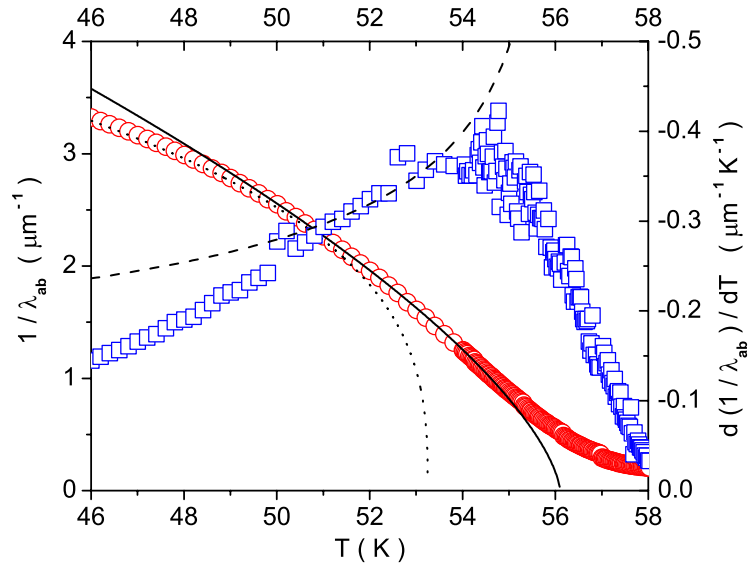
In this case the penetration depth adopts the finite size scaling form [30, 31]

$$\lambda_{ab}(T) = \lambda_{0ab}|t|^{-\nu}g(y), \quad y = \text{sgn}(t) \left| \frac{t}{t_p} \right| \quad (4)$$

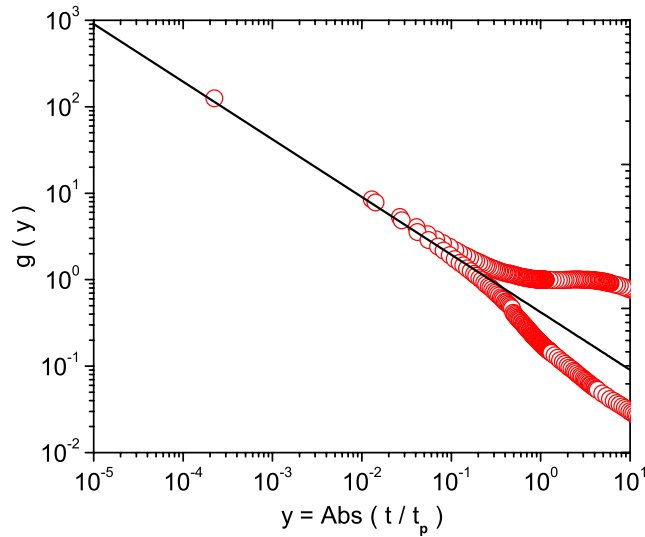
where  $\xi_{ab}(T_p) = \xi_{0ab}|t_p|^{-\nu} = L_{ab}$ . For  $t$  small and  $L_{ab} \rightarrow \infty$  the scaling variable tends to  $y \rightarrow \pm\infty$  where  $g(y \rightarrow -\infty) = 1$  and  $g(y \rightarrow +\infty) = 0$  while for  $t = 0$  and  $L_{ab} \neq 0$ ,  $g(y \rightarrow 0) = g_0|y|^\nu = g_0|t/t_p|^\nu$ . In this limit we obtain

$$\frac{\lambda_{ab}(T_c, L_{ab})}{\lambda_{0ab}} = g_0 \frac{L_{ab}}{\xi_{0ab}}. \quad (5)$$

Another essential property of the finite size scaling function stems from the existence of an inflection point in  $\lambda_{ab}(T)$ . It implies an extremum in  $d\lambda_{ab}/dT$  at  $T_p > T_c$  and with that

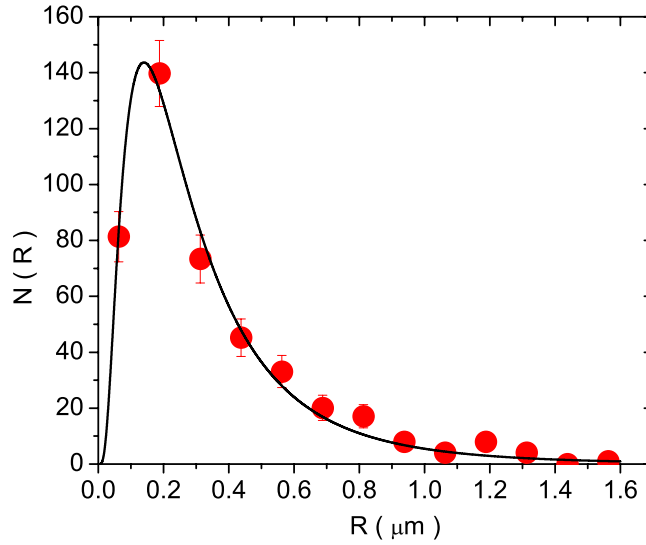


**Figure 2.**  $1/\lambda_{ab}$  and  $d(1/\lambda_{ab})/dT$  versus  $T$  for  $\text{YBa}_2\text{Cu}_3\text{O}_{6.59}$ . The solid curve is  $\lambda_{ab} = \lambda_{0ab}|t|^{-\nu}$  with  $\lambda_{0ab} = 0.089 \mu\text{m}$ ,  $\nu = 2/3$  and  $T_c = 56.1 \text{ K}$ , appropriate for charged criticality, and the dashed one its derivative. The dotted curve indicates uncharged critical behaviour.



**Figure 3.** Finite size scaling function  $g(y)$  deduced from the measured data with  $\lambda_{0ab} = 0.089 \mu\text{m}$ ,  $\nu = 2/3$ ,  $T_c = 56.1 \text{ K}$  and  $T_p \simeq 54.27 \text{ K}$ . The solid line indicates the asymptotic behaviour  $g(y \rightarrow 0) = g_0|y|^\nu$  with  $g_0 = 0.42$ .

the scaling form  $g^+(y) = y^\nu(1 + f(y))$  with  $df/dy \neq 0$  and  $d^2f/dy^2 = 0$  at  $y = 1$ , e.g.  $f(y) = ay + b(1 - y)^3 + c$ . In figure 3 we displayed the finite size scaling function  $g(y)$  deduced from the measured data with  $\lambda_{0ab} = 0.089 \mu\text{m}$ ,  $\nu = 2/3$ ,  $T_c = 56.1 \text{ K}$  and  $T_p \simeq 54.27 \text{ K}$ . The solid line indicates the asymptotic behaviour  $g(y \rightarrow 0) = g_0|y|^\nu$  with  $g_0 = 0.42$ . The upper branch corresponding to  $T < T_c$  tends to  $g(y \rightarrow -\infty) = 1$ , while



**Figure 4.** Grain size distribution  $N(R)$  of our  $\text{YBa}_2\text{Cu}_3\text{O}_{6.59}$  sample derived from an analysis of SEM (scanning electron microscope) photographs. The solid curve is a fit to the log-normal distribution [34].

the lower one referring to  $T > T_c$  approaches  $g(y \rightarrow +\infty) = 0$ . Consequently, the absence of a sharp transition (see figures 1 and 2) is fully consistent with a finite size effect arising from a limiting length  $L_{ab}$  in the  $ab$ -plane, attributable to the finite extent of the grains and/or inhomogeneities within the grains.

To disentangle these options we note that  $L_{ab}/\xi_{0ab} \simeq 51$  follows from the finite size scaling analysis by invoking equation (5) with  $\lambda_{ab}(T_c, L_{ab}) \simeq 1.898 \mu\text{m}$ ,  $\lambda_{ab0}(T_c, L_{ab}) \simeq 0.089 \mu\text{m}$  and  $g_0 = 0.42$ . Noting that  $\xi_{ab0} = \gamma \xi_{c0}$ , where  $\gamma$  is the anisotropy, we obtain with  $\gamma \approx 20$  [32] and  $\xi_{c0} \approx 10 \text{ \AA}$ , appropriate for  $\text{YBa}_2\text{Cu}_3\text{O}_{6.59}$  [33], the estimate  $L_{ab} \approx 510 \text{ \AA}$  compared to  $L_{ab} \approx 572 \text{ \AA}$  found in  $\text{YBa}_2\text{Cu}_3\text{O}_{6.7}$  [18]. On the other hand, a glance at figure 4 shows that the grain size distribution of our  $\text{YBa}_2\text{Cu}_3\text{O}_{6.59}$  sample exhibits a maximum at  $2R = 2800 \text{ \AA}$  and decreases steeply for smaller grains. Hence, the smeared transition is not attributable to the finite extent of the grains but due to inhomogeneities within the grains. However, this does not point to bad sample quality but to a genuine feature of underdoped cuprates reflecting the large value of  $\xi_{ab0}$  in this doping regime.

Nevertheless, our analysis of the in-plane penetration depth data of underdoped  $\text{YBa}_2\text{Cu}_3\text{O}_{6.59}$  provides remarkable consistency for critical fluctuations, consistent with the charged universality class, limited close to  $T_c$  of the fictitious infinite and homogeneous counterpart by a inhomogeneity induced finite size effect. Since  $\kappa_{ab} \propto \sqrt{T_c}$  this will no longer hold true in the optimally doped counterparts. In this doping regime there is mounting evidence for neutral (3D  $XY$ ) critical behaviour [3, 4, 16, 22, 33]. A prominent example is  $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$  revealing in the in-plane penetration depth [16] 3D  $XY$  behaviour over three decades in reduced temperature, with no sign pointing to a crossover to charged criticality.

In summary, we have presented in-plane penetration depth data for  $\text{YBa}_2\text{Cu}_3\text{O}_{6.59}$  providing consistent evidence for the charged critical behaviour of the superconductor to normal state transition in type II superconductors ( $\kappa > 1/\sqrt{2}$ ). Since the effective dimensionless charge  $\tilde{e} = \xi/\lambda = 1/\kappa$  scales as  $T_c^{-1/2}$  this new critical behaviour should be observable and generic in suitably underdoped cuprates. In this regime the crossover upon

approaching  $T_c$  is thus to the charged critical regime, while near optimum doping it is to the critical regime of a weakly charged superfluid where the fluctuations of the order parameter are essentially those of an uncharged superfluid (3D XY). Furthermore, there is the inhomogeneity induced finite size effect which renders the asymptotic critical regime and with that the charged regime of nearly optimally doped samples difficult to attain.

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