Finite-size and pressure effects in YBa$_2$Cu$_4$O$_8$ probed by magnetic-field penetration-depth measurements

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We explore the combined pressure and finite-size effects on the in-plane penetration depth $\lambda_{ab}$ in YBa$_2$Cu$_4$O$_8$. Even though this cuprate is stoichiometric the finite-size scaling analysis of $1/\lambda_{ab}^2(T)$ uncovers the granular nature and reveals domains with nanoscale size $L_c$ along the c-axis. $L_c$ ranges from 50.8 (1.0) Å to 39.3 (8) Å at pressures from 0.0 to 10.2 kbar. These observations raise serious doubts on the existence of a phase coherent macroscopic superconducting state in cuprate superconductors.

I. INTRODUCTION

There is increasing evidence that inhomogeneities are an intrinsic property of cuprates (see, e.g., Ref. 1 and references therein). Studies of different cuprate families revealed the segregation of the material in superconducting and nonsuperconducting regions. In particular, neutron scattering, electron paramagnetic resonance (EPR), scanning tunneling microscopy (STM), and x-ray diffraction provide evidence for granular superconductivity in the cuprates. Although crystals of the cuprates are not granular in a structural sense, it occurs when microscopic superconducting domains are separated by nonsuperconducting regions through which they communicate for instance by Josephson tunneling to establish the macroscopic superconducting state.

On the other hand, there is evidence for nearly isolated, homogeneous and superconducting domains of nanoscale extent embedded in a nonsuperconducting matrix. It stems from a finite-size scaling analysis of the thermal fluctuation contributions to the specific heat and magnetic field penetration depth data. In an isolated domain there is a finite-size effect because the correlation length cannot grow beyond the length of the superconducting domain in direction $i$. Accordingly there is no sharp phase transition and the specific heat coefficient will exhibit a blurred peak with a maximum at $T_p < T_c$, where $T_p$ is the transition temperature of the homogeneous bulk system. At $T_p$ the correlation length $\xi(T)$ reaches the limiting length $L$ of the domain. Similarly, $1/\lambda^2$, where $\lambda$ is the magnetic field penetration depth, does not vanish at $T_c$, but exhibits a tail with an inflection point at $T_p$. This raises serious doubts on the existence of macroscopic phase coherent superconductivity, suggesting that bulk superconductivity is achieved by a percolative process. Therefore, superconducting properties and the spatial extent of the domains can be probed by thermal fluctuations and the finite-size effects. This includes the effects of oxygen isotope exchange and pressure on the domain size. Recently a significant change of spatial extent of the superconducting domains upon oxygen isotope exchange has been demonstrated in Y$_{1-x}$Pr$_x$Ba$_2$Cu$_3$O$_{7-x}$.$^7$ It revealed the relevance of local lattice distortions in the occurrence of superconductivity.

In this paper we address the pressure studies of the finite-size effect in YBa$_2$Cu$_4$O$_8$, which exhibits a rather large and positive pressure effect (PE) on $T_c$, with $dT_c/dp = 0.50(1)$ K/kbar.$^{9-11}$ Even though this cuprate is stoichiometric the finite-size scaling analysis uncovers the existence of nanoscale domains with a spatial extent $L_c$, along the crystallographic c-axis. The value of $L_c$ decreases from 50.8 (1.0) Å to 39.3 (8) Å with increasing pressure from 0.0 to 10.2 kbar. Accordingly, $T_c$ increases with reduced thickness $L_c$ of the domains.

The paper is organized as follows. In Sec. II we sketch the finite-size scaling theory adapted for the analysis of penetration depth data. In Sec. III we describe the sample preparation procedure and the experimental technique, adopted to deduce the in-plane magnetic field penetration depth $\lambda_{ab}$ from the Meissner fraction measurements. In Sec. IV we perform the finite-size analysis of the data for the in-plane magnetic penetration depth $\lambda_{ab}(T)$ taken at different pressures. Section V comprises the analysis of the pressure dependence of the domain lengths $L_c$ along the crystallographic c-axis.

II. THEORETICAL BACKGROUND

The detailed description of the finite-size scaling theory adopted for the analysis of the magnetic field penetration depth $\lambda$ can be found in Refs. 1, 6, and 7. Briefly, the main signatures for the existence of finite-size behavior appearing in the temperature dependence of the in-plane magnetic penetration depth $\lambda_{ab}$ can be summarized as follows:$^1$

(i) Denoting the spatial extent of the domains along the crystallographic a-, b- and c-axes with $L_a$, $L_b$, and $L_c$, the transverse in-plane correlation lengths $\xi_{ab}(T)$ cannot diverge at $T=T_c$ but are limited by

$$\xi_{ab}(T) \leq L_c.$$  

(1)

Consequently, for finite superconducting domains, the penetration depth is a smooth function of temperature. As a rem-
nent of the singularity at $T_c$, $\lambda_{ab}^{-2}$ exhibits a so called finite-
size effect,\(^1\) namely an inflection point at $T_{p_c}$ so that
\[
\left.\frac{d\lambda_{ab}^{-2}(T)}{dT}\right|_{T=T_{p_c}} = \text{extremum}.
\] (2)

(ii) The absolute value of the superconducting domain size along the $c$-axis ($L_c$) can be estimated knowing $\lambda_{ab}$ at $T=T_{p_c}$ as\(^1,6,7\)
\[
L_c = \frac{16\pi^3 k_B T_{p_c} \lambda_{ab}^2(T_{p_c})}{\Phi_0^2},
\] (3)
where $\Phi_0$ is the flux quantum, and $k_B$ is the Boltzmann’s constant.

(iii) Apart from the inflection point, an essential characteristic of a finite size effect is the finite-size scaling function.\(^12\) According to Ref. 1 it is defined in terms of
\[
g(y) = \frac{\lambda_{ab}^2}{\lambda_{ab}'^2} |t|^{-\nu},
\] (4)
where $y$ is equal to
\[
y = \text{sign}(t) \left| \frac{T}{T_{p_c}} \right|.
\]
t=$T/T_c$−1, $T_{p_c}=T_p/L_c$−1, $\nu$ is the critical exponent, and $\lambda_{ab}'$ is the critical amplitude.\(^1\)

For the homogenous system ($L_c \to \infty$) and $t \neq 0$ ($y \neq 0$), $g(y)$ corresponds to the stepwise function
\[
g_+(y<0) = 1, \quad g_-(y>0) = 0,
\] (5)
while for the system confined by the finite geometry ($L_c \neq 0$) the scaling function $g(y)$ diverges at $t \to 0$ ($y \to 0$) as
\[
g(y \to 0) = g_0 \nu^{-\nu} = g_0 \left( \frac{T}{T_{p_c}} \right)^{-\nu}.
\] (6)

III. EXPERIMENTAL DETAILS

A. Sample preparation and characterization

The polycrystalline YBa$_2$Cu$_4$O$_8$ samples were synthesized by solid-state reactions using high-purity Y$_2$O$_3$, BaCO$_3$ and CuO. The samples were calcinated at $880$–$935^\circ$C in air for 110 hours with several intermediate grindings. The phase-purity of the material was examined using a powder x-ray diffractometer. Only YBa$_2$Cu$_4$O$_{7-\delta}$ and CuO phases were revealed. The synthesis was continued at a high oxygen pressure of 500 bar, at $1000^\circ$C during 30 hours. The x-ray diffraction measurements performed after the final stage of the synthesis revealed 95% of the YBa$_2$Cu$_4$O$_8$ phase. The sample was then reground in mortar for about 60 min and passed through the 10 $\mu$m sieve in order to obtain sufficiently small grains, as required for the determination of $\lambda$ from Meissner fraction measurements. As it was shown in Ref. 13 the grounding procedure does not introduce any additional defects. The grain size distribution of the powder was then determined by analyzing SEM (scanning electron microscope) photographs. The measured particle radius distribution $N(R)R^2$ of YBa$_2$Cu$_4$O$_8$ sieved powder derived from the SEM photographs. The dashed line is the stepwise $G(R)$ function used for the $\lambda(T)$ determination by means of Eq. (8). The inset shows the grain size distribution $N(R)$. Errors are statistical.

The field-cooled (FC) dc-magnetization ($M$) measurements were performed with a Quantum Design SQUID magnetometer in a field of 0.5 mT for temperatures ranging from 5 K to 100 K. The absence of weak links between grains has been confirmed by the linear magnetic field dependence of the FC magnetization, measured at 0.25 mT, 0.5 mT, 0.75 mT and 1.0 mT for each pressure at $T=8$ K (see the inset in Fig. 2).

The hydrostatic pressure was generated in a copper-beryllium piston cylinder clamp that was especially designed...
for magnetization measurements under pressure. The sample was mixed with Fluorient FC77 (pressure transmitting medium) with a sample to liquid volume ratio of approximately 1/6. With this cell hydrostatic pressures up to 12 kbar can be achieved. The pressure was measured in situ by monitoring the $T_c$ shift of the small piece of In [$T_c(p=0)=3.4$ K] included in the pressure cell.

### B. Determination of the temperature dependence of $\lambda$

from Meissner fraction measurements

The temperature dependence of $\lambda^{-2}$ was extracted from the Meissner fraction $f$ deduced from low-field (0.5 mT FC) magnetization data using the relation

$$f(T) = \left(\frac{H}{M(T)} - N\right)^{-1},$$

where $H$ denotes the external magnetic field and $N$ is the demagnetization factor. $N=1/3$ was taken assuming that the sample grains are spherical. Figure 2 shows the temperature dependence of the Meissner fraction close to $T_c$ for different pressures. Three important features emerge: (i) The transition temperature $T_c$ increases with increasing pressure. The value of $dT_c/dp=0.50(1)$ K/kbar is found, which is in good agreement with the literature data. (ii) The value of $f$ (Fig. 2) is much smaller than 1, confirming that the average grain size of the sample is compatible with $\lambda$. The reduction of $f$ is caused by the field penetration at the surface of each individual grain for distances of the order of $\lambda$. (iii) The absolute value of the Meissner fraction increases with pressure. Since the average grain size does not change under pressure and the grains are decoupled from each other, the rise of $f$ must be attributed to a decrease of the magnetic penetration depth $\lambda$.

The temperature dependence of the penetration depth $\lambda$ was analyzed from the measured $f(T)$ by using the Shoenberg formula\(^{17}\) modified for the known grain size distribution:\(^{18}\)

$$f(T) = \int_0^\infty \frac{3\lambda(T)}{R} \coth \left( \frac{R}{\lambda(T)} \right) + \frac{3\lambda^2(T)}{R^2}$$

$$\times G(R)dR / \int_0^\infty G(R)dR.$$

Here $G(R)=N(R)R^3$, and $N(R)$ is the grain size distribution (see Fig. 1). By solving this nonlinear equation, $\lambda$ for each value of $f$ was extracted, and then the set of $\lambda(T,p)$’s was reconstructed. Since the sample consists of an anisotropic nonoriented powder, the extracted $\lambda$ is the so called effective penetration depth $\lambda_{\text{eff}}$ (powder average). However, for sufficiently anisotropic extreme type II superconductors, including YBa$_2$Cu$_4$O$_8$, $\lambda_{\text{eff}}$ is proportional to the in-plane penetration depth in terms of $\lambda_{\text{eff}}=1.31\lambda_{ab}$.\(^{19}\) The resulting temperature dependence $\lambda_{ab}^{-2}(T,0)$ at ambient pressure is shown in Fig. 3. Note, that $\lambda_{ab}^{-2}(T)$ curves of the present work (see Fig. 3) are very similar to those reported in Ref. 20, obtained for YBa$_2$Cu$_4$O$_8$ using the same experimental technique. The fast increase of $\lambda_{ab}^{-2}$ close to $T_c$ and change of the linear slope at low temperatures in Ref. 20 was attributed to the contribution of the double chains to $\lambda_{ab}^{-2}$.

To estimate the error in the absolute value of $\lambda(0)$ introduced by the uncertainty in the grain-size distribution we performed reconstructions with two “extreme” conditions. For the first we took the grain-size distribution in the form $N^*(R)=N(R)-\sqrt{N(R)}$; for the second we took $N^*(R)=N(R)+\sqrt{N(R)}$. Appropriate $\lambda_{ab}^{-2}(T)$ dependencies for $N^*(R)$, $N(R)$ and $N^*(R)$ are shown in the inset of Fig. 3. The linear fit of the low-temperature part of $\lambda_{ab}^{-2}$ yields 142 nm for the lowest and 170 nm for the highest values of $\lambda(0)$. From these the absolute value of $\lambda_{ab}$ can be estimated as $\lambda_{ab}(0)=156(14)$ nm that is in good agreement with the $\lambda_{ab}(0)$ obtained by means of $\mu SR$.\(^{21-23}\)

Now we are turning to analyze the pressure dependence of $\lambda_{ab}$. As it was already mentioned, the procedure of the $\lambda^{-2}(T)$ reconstruction is sensitive to the grain-size distribution. The good thing here is that we study relative effects measured with the same sample in the same pressure cell, where most of the systematic errors are eliminated. The main systematic error for such measurements comes from misalignments of the experimental setup after the cell was removed from the SQUID magnetometer and put back again. We checked this procedure with a set of measurements at constant pressure. The systematic scattering of the magnetization data is of about 0.5%, giving a relative error in $\lambda^{-2}(T)$ of about 2%. Temperature dependencies of $\lambda_{ab}^{-2}$ evaluated at different pressures are depicted in Fig. 4. The solid lines indicate the leading critical behavior of $\lambda_{ab}^{-2}(T)$ for homogenous and infinite domains.\(^{1,24}\)

$$\lambda_{ab}^{-2}(T) = \lambda_{ab}^{-2}(T)^\nu,$$

with $\nu=2/3$ in the 3D-XY universality class,\(^{1,7}\) and the critical amplitude
The values of $\lambda_{ab}$ and $T_c$ obtained from the fit of Eq. (9) to the experimental data presented in Fig. 4 are summarized in Table I. Since Eq. (9) is valid in the vicinity of $T_c$ only, we restricted the fit to the interval from $T_c$ to $T_c - 10$ K.

IV. THE FINITE-SIZE ANALYSIS

The essential characteristic of a homogeneous bulk cuprate superconductor is a sharp superconductor to normal state transition. A glance to the inset of Fig. 4 shows that in the samples considered here that is not the case. The transition occurs smoothly and there is a tail pointing to a finite-size effect associated with an inflection point at some characteristic temperature $T_p$. As outlined above [Eqs. (1) and (2)] at $T_p$, the transverse correlation length $\xi_{ab}^t$ attains the limiting length along the c-axis. To substantiate the occurrence of an inflection point we show in Fig. 5, $d\lambda_{ab}^{-2}(T)/dT$ versus $T$ for YBa$_2$Cu$_4$O$_8$ samples at different pressures. The extreme in the first derivative of $\lambda_{ab}^{-2}(T)$ clearly reveals the existence of an inflection point at $T_p < T_c$. The absolute values of $T_p$ obtained from a parabolic fit to experimental data around maximum point are summarized in Table I.

To substantiate the finite-size scenario further, we explore the scaling properties of the data with respect to the consistency with the finite-size scaling function. Noting that $\nu = 2/3$ in the 3D-XY universality class, Eq. (4) can be rewritten as

$$\frac{\lambda_{ab}^{-2}(T)}{\lambda_{ab}^{-2}} |_{\nu = 2/3} = g\left(\frac{T}{T_p}\right).$$

Figure 6(a) shows the resulting scaling function. For a comparison, we included the limiting behavior of the finite-size scaling function $g_\infty$ [see Eq. (5)] for a homogeneous ($L_c \rightarrow \infty$) system [the solid line in Fig. 6(a)]. As it is seen, there is quite a good agreement between the experimental data and $g_\infty$ function for $t/t_{p_c}$ far from 0 ($T$ far from $T_c$), whereas in the vicinity of 0 ($T \sim T_c$) the data are completely inconsistent with such a stepwise behavior. The experimental data have to diverge at $t/t_{p_c} \sim 0$. As outlined in Sec. II divergence of the scaling function $g(t/t_{p_c})$ at $t/t_{p_c} \rightarrow 0$ implies that the system is confined by a finite geometry. To strengthen this point we

<table>
<thead>
<tr>
<th>$T_c$ (K)</th>
<th>$T_p$ (K)</th>
<th>$\lambda_{ab}^{-2}$ (um$^{-2}$)</th>
<th>$\lambda_{ab}^{-2}(T_p)$ (um$^{-2}$)</th>
<th>$L_c$ (Å)</th>
<th>$\Delta T_c/T_c$ (%)</th>
<th>$\Delta T_{p}/T_{p_c}$ (%)</th>
<th>$\Delta \lambda_{ab}^{-2}(T_p)/\lambda_{ab}^{-2}(T_p)$ (%)</th>
<th>$\Delta L_c/L_c$ (%)</th>
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</thead>
<tbody>
<tr>
<td>0.0 kbar</td>
<td>79.07(1)</td>
<td>77.5(1)</td>
<td>34.3(7)</td>
<td>2.44(5)</td>
<td>50.8(1.0)</td>
<td>-</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>4.29 kbar</td>
<td>81.40(1)</td>
<td>79.7(1)</td>
<td>39.2(8)</td>
<td>2.92(6)</td>
<td>43.7(9)</td>
<td>2.95(2)</td>
<td>2.84(5)</td>
<td>19.7(2.8)</td>
</tr>
<tr>
<td>7.52 kbar</td>
<td>82.73(3)</td>
<td>81.1(1)</td>
<td>44.1(9)</td>
<td>3.29(7)</td>
<td>39.4(8)</td>
<td>4.63(4)</td>
<td>4.65(5)</td>
<td>34.8(2.8)</td>
</tr>
<tr>
<td>10.2 kbar</td>
<td>84.22(3)</td>
<td>82.5(1)</td>
<td>45.0(9)</td>
<td>3.36(7)</td>
<td>39.3(8)</td>
<td>6.51(4)</td>
<td>6.45(5)</td>
<td>37.7(2.8)</td>
</tr>
</tbody>
</table>

TABLE I. Finite size estimates for $T_{p_c}$, $\lambda_{ab}^{-2}$, $\lambda_{ab}^{-2}(T_p)$ and the resulting relative shifts $\Delta T_p/T_c$ and $\Delta \lambda_{ab}^{-2}(T_p)/\lambda_{ab}^{-2}(T_p)$ for different pressures. $L_c$ and $\Delta L_c/L_c$ are deduced from Eq. (3).
YBa$_2$Cu$_3$O$_{6.7}$ oriented powder and for Bi$_2$Sr$_2$CaCu$_2$O$_{8+\delta}$

films. Much larger values, consistent with all three characteristics of a finite-size effect, are presented in Table I. We established the consistency of our estimates we plot in Fig. 8 the relative shifts of $T_{P_c}$, $\lambda_{ab}^{-2}(T_{P_c})$, and $\Delta T_{P_c}$ versus the relative shift of $T_c$. According to Eq. (3) these shifts are not independent but related by

\[ \Delta T_c/T_c = \Delta T_{P_c}/T_{P_c} \cdot \lambda_{ab}^{-2}(T_{P_c})/\lambda_{ab}^{-2}(T_c) \]

V. THE PRESSURE DEPENDENCE OF THE DOMAIN SIZE

Using Eq. (3) and the estimates for $T_{P_c}$ and $\lambda_{ab}^{-2}(T_{P_c})$ listed in Table I, the domain size $L_c$ along the $c$-axis is readily calculated. In Fig. 7 we display the pressure dependence of $L_c$, $T_c$, and $T_{P_c}$, and the relative shifts of $T_{P_c}$, $T_c$, and $\lambda_{ab}^{-2}(T_{P_c})$ versus the relative shift of $T_c$. The solid lines represent the linear fits with adjustable parameters $\Delta L_c/L_c$ and $\Delta T_{P_c}/T_{P_c}$.

\[ \Delta L_c/L_c = -4.0(5) \cdot \Delta T_c/T_c \]

\[ \Delta T_{P_c}/T_{P_c} = 1.00(7) \cdot \Delta T_c/T_c \]

\[ \Delta \lambda_{ab}^{-2}(T_{P_c})/\lambda_{ab}^{-2}(T_c) = 6.4(5) \cdot \Delta T_c/T_c \]
The straight lines correspond to the linear fits with \( \Delta L_c/L_c = -4.0(5) \Delta T_p/T_p \). \( \Delta T_p/T_p = 1.00(7) \cdot \Delta T_s/T_s \) and \( \Delta \lambda_{ab}^2(T_p)/\lambda_{ab}^2(T_p) = 6.4(5) \cdot \Delta T_s/T_s \), revealing that Eq. (12) is almost satisfied. Furthermore, these estimates show that the reduction of \( L_c \) with pressure reflects the fact that the pressure effect on \( \lambda_{ab}^2(T_p) \) exceeds the effect on \( T_c \) considerably. Having established the consistency of our estimates it is essential to recognize that the occurrence of nanoscale superconducting domains is not an artefact of YBa\(_2\)Cu\(_4\)O\(_8\) and our samples. Indeed the existence of nanoscale domains has been established in a variety of cuprates, including films, powders and single crystals.\(^{1,6–8}\)

VI. CONCLUSION

To summarize, we reported the observation of combined finite-size and pressure effects on the lengths \( L_c \) of the superconducting domains along the \( c \)-axis and the in-plane penetration depth \( \lambda_{ab} \) in a cuprate superconductor. The evidence for a finite-size behavior of the system stems from the tail in \( \lambda_{ab}^2(T) \) observed in the vicinity of \( T_c \). We have shown that the scaling properties of the tail are fully consistent with a finite-size effect, arising from domains with nanoscale size along the \( c \)-axis. Indeed the essential characteristics of a finite-size effect, as (i) the limiting properties of the scaling function \( g(t/T_p) \), (ii) the existence of an inflection point in \( \lambda^2(T) \) and (iii) the extremum in \( d\lambda^2(T)/dT \) at \( T_p \) have been verified. Even though YBa\(_2\)Cu\(_4\)O\(_8\) is stoichiometric we have shown that the size of the domains along the \( c \)-axis is of nanoscale only, ranging from 50.8(1.0) Å to 39.3(8) Å at pressures from 0.0 to 10.2 kbar. This raises serious doubts on the existence of macroscopic phase coherent superconductivity. Contrary-wise, this does not exclude a percolative resistive superconductor to normal state transition, when the superconducting domains percolate. Indeed, a granular superconductor is usually characterized by two parameters,\(^{26}\) the first is the domain’s size and the second the coupling between them. The coupling is accomplished randomly with a temperature dependent probability. Such a mechanism is a percolation process; at the temperature where the coupling probability is equal to the percolation threshold, an infinite cluster of coupled superconducting grains is formed. This suggests that resistive bulk superconductivity is achieved by a percolative process, while the phase coherent superconducting properties and the spatial extent of the domains can be probed by thermal fluctuations and finite-size effects.

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