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LETTER TO THE EDITOR

Relevance of electron–lattice coupling in cuprate superconductors

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Abstract
We study the oxygen isotope (^{16}O, ^{18}O) and finite size effects in $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ by in-plane penetration depth (λ_{ab}) measurements. A significant change of the length L_c of the superconducting domains along the c-axis and λ_{ab}^2 is deduced, yielding the relative isotope shift $\Delta L_c/L_c \approx \Delta\lambda_{ab}^2/\lambda_{ab}^2 \approx 0.14$ for $x = 0$, 0.2 and 0.3. This uncovers the existence and relevance of the coupling between the superfluid, lattice distortions and anharmonic phonons which involve the oxygen lattice degrees of freedom.

Since the discovery of superconductivity in cuprates by Bednorz and Müller [1] a tremendous amount of work has been devoted to their characterization. The issue of inhomogeneity and its characterization is essential for several reasons, including the following. First, if inhomogeneity is an intrinsic property, a re-interpretation of experiments, measuring an average of the electronic properties, is unavoidable. Second, inhomogeneity may point to a microscopic phase separation, i.e. superconducting domains, embedded in a nonsuperconducting matrix. Third, there is neutron spectroscopic evidence for nanoscale cluster formation and percolative superconductivity in various cuprates [2, 3]. Fourth, nanoscale spatial variations in the electronic characteristics have been observed in underdoped $Bi_2Sr_2CaCu_2O_{8+\delta}$ with scanning tunnelling microscopy (STM) [4–7]. They reveal a spatial segregation of the electronic structure into 3 nm diameter superconducting domains in an electronically distinct background. In contrast, a large degree of homogeneity has been observed in STM studies of $Bi_2Sr_2CaCu_2O_{8+\delta}$ [8] and NMR in $YBa_2Cu_3O_{7-\delta}$ [9]. In any case the relevance of these observations for thermodynamic properties remains to be clarified. Fifth, in $YBa_2Cu_3O_{7-\delta}$, MgB_2, 2H-$NbSe_2$ and $Nb_{77}Zr_{23}$ considerably larger domains have been established. The magnetic field induced finite size effect revealed lower bounds ranging

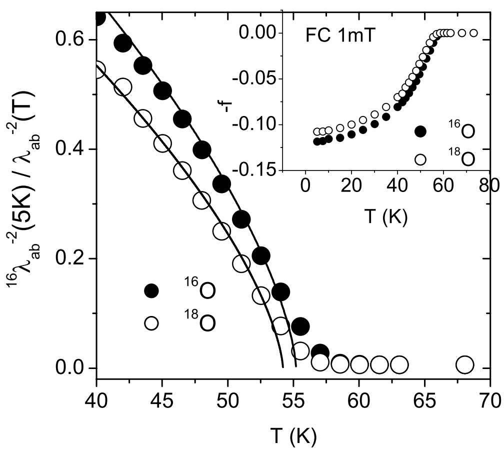

Figure 1. $(^{16}\lambda_{ab}(0)/\lambda_{ab}(T))^2$ versus T for the ^{16}O and ^{18}O samples of $Y_{0.7}Pr_{0.3}Ba_2Cu_3O_{7-\delta}$. The solid curves indicate the leading critical behaviour of the homogeneous systems as explained in the text. Inset: Meissner fraction f versus temperature (FC 1 mT). The error bars are smaller than the size of the data points.

from $L = 182$ to 814 Å [10, 11]. Sixth, since the change of the lattice parameters upon oxygen isotope exchange is negligibly small [12, 13], the occurrence of a significant modification of the domains' spatial extent will provide clear evidence for superconductivity mediated by local lattice distortions.

This letter concentrates on $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ with ^{16}O and ^{18}O. It addresses these issues by providing clear evidence for a finite size effect on the in-plane London penetration depth, revealing the existence of superconducting domains with spatial nanoscale extent and its significant change upon oxygen isotope exchange.

Polycrystalline samples of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ ($x = 0, 0.2, 0.3$) were synthesized by solid-state reactions [14] and their phase-purity was examined using powder x-ray diffraction. For each doping concentration the samples were reground in a mortar for about 60 min. Powder samples with a grain size of $<10\ \mu$m were obtained by using a system of 20/15/10 μm sieves. Oxygen isotope exchange was performed by heating the powder in $^{18}O_2$ gas. In order to ensure the same thermal history of the substituted (^{18}O) and nonsubstituted (^{16}O) samples, two experiments (in $^{16}O_2$ and $^{18}O_2$) were always performed simultaneously [14]. To achieve complete oxidation the exchange processes were carried out at 550 °C during 30 h, followed by slow cooling (20 °C h^{-1}). The ^{18}O content, determined from a change of the sample weight after the isotope exchange, was found to be 89(2)% for all samples. Field-cooled (FC) magnetization measurements were performed with a Quantum Design SQUID magnetometer in field range 0.5–10 mT and a temperature range 5–100 K. The powder samples ($\sim$100 mg) were put in a quartz ampoule. To guarantee the same experimental conditions (sample geometry and the background signal from the sample holder), the same ampoule was used. The absence of weak links between grains was confirmed by the linear scaling of the FC magnetization measured at 5 K in 0.5, 1 and 1.5 mT. The Meissner fraction f was calculated from the mass and the x-ray density, assuming spherical grains. An example of f versus T is displayed in the inset of figure 1. Assuming spherical grains of radius R, the data were analysed on the basis of the Shoenberg formula [15], allowing us to calculate the temperature dependence of the effective penetration depth $\lambda_{eff}(T)/\lambda_{eff}(0)$. For sufficiently anisotropic extreme type II superconductors, including $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$, λ_{eff} is proportional to the in-plane penetration depth, so that $\lambda_{\rm eff} = 1.31\lambda_{ab}$ [16, 17]. Since it is not possible to extract the absolute value of λ_{ab} from our measurements, we take $\lambda_{ab}(0\ {\rm K})$ from μSR measurements [18] to normalize the data. In the main panel of figure 1 we displayed the resulting temperature dependence of $^{16}\lambda_{ab}^2(0)/\lambda_{ab}^2(T)$ for the ^{16}O and ^{18}O samples of $Y_{0.7}Pr_{0.3}Ba_2Cu_3O_{7-\delta}$. For comparison we included $^{16}\lambda_{ab}^2(0)/\lambda_{ab}^2(T) = {^{16}\lambda_{ab}^2(0)}/\lambda_{0,ab}^2|t|^{-\nu}$, where $t = (T/T_{\rm c} - 1)$, with the critical exponent $\nu = 2/3$, $^{16}T_{\rm c} = 54.9$ K and $^{18}T_{\rm c} = 53.7$ K to indicate the asymptotic critical behaviour for an infinite superconducting domain. Apparently, the data are inconsistent with such a sharp transition. It clearly uncovers a rounded phase transition which occurs smoothly and with that a finite size effect at work. In this context it is important to emphasize that this finite size effect is not an artefact of $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ or the particular technique used to evaluate $1/\lambda_{ab}^2(T)$. Indeed, this rounding is also seen in the data for $YBa_2Cu_3O_7$ [19], $La_{2-x}Sr_xCuO_4$ with $x = 0.1, 0.15$ and 0.2 [20], and $Bi_2Sr_2CaCu_2O_{8+\delta}$ single crystals [21, 22]. Moreover, independent evidence for superconducting domains of finite extent stems from the analysis of specific heat data [11]. Given the mounting evidence for these domains, their behaviour upon oxygen isotope exchange is expected to offer valuable clues on the relevance of local lattice distortions in the mechanism mediating superconductivity.

To elucidate this issue we perform a finite size scaling analysis of the in-plane penetration depth data for $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ with ^{16}O and ^{18}O. Supposing that cuprate superconductors are granular, consisting of spatial superconducting domains, embedded in a nonsuperconducting matrix and with spatial extent L_a, L_b and L_c along the crystallographic a-, b- and c-axes, the correlation length ξ_i in direction i, increasing strongly when $T_{\rm c}$ is approached, cannot grow beyond L_i. Consequently, for finite superconducting domains, the thermodynamic quantities such as the specific heat and penetration depth are smooth functions of temperature. As a remnant of the singularity at $T_{\rm c}$ these quantities exhibit a so called finite size effect [23], namely a maximum or an inflection point at $T_{{\rm p}_i}$, where $\xi_i(T_{{\rm p}_i}) = L_i$. There is mounting experimental evidence that for the accessible temperature ranges, the effective finite temperature critical behaviour of the cuprates is controlled by the critical point of uncharged superfluids (3D-XY) [10, 22]. In this case there is the universal relationship

$$\frac{1}{\lambda_i^2(T)} = \frac{16\pi^3 k_{\rm B} T}{\Phi_0^2 \xi_i^{\rm t}(T)}, \tag{1}$$

between the London penetration depth λ_i and the transverse correlation length $\xi_i^{\rm t}$ in direction i [10, 24]. As aforementioned, when the superconductor is inhomogeneous, consisting of superconducting domains with length scales L_i, embedded in a nonsuperconducting matrix, $\xi_i^{\rm t} = \xi_{0i}^{\rm t}|t|^{-\nu}$ does not diverge but is bounded by

$$\xi_i^{\rm t}\xi_j^{\rm t} \leqslant L_k^2, \qquad i \neq j \neq k. \tag{2}$$

A characteristic feature of the resulting finite size effect is the occurrence of an inflection point at $T_{{\rm p}_k}$ below $T_{\rm c}$, the transition temperature of the homogeneous system. Here

$$\xi_i^{\rm t}(T_{{\rm p}_k})\xi_j^{\rm t}(T_{{\rm p}_k}) = L_k^2, \qquad i \neq j \neq k, \tag{3}$$

and equation (1) reduces to

$$\left.\frac{1}{\lambda_i(T)\lambda_j(T)}\right|_{T=T_{{\rm p}_k}} = \frac{16\pi^3 k_{\rm B} T_{{\rm p}_k}}{\Phi_0^2}\frac{1}{L_k}. \tag{4}$$

In the homogeneous case $1/\lambda_i(T)/\lambda_j(T)$ decreases continuously with increasing temperature and vanishes at $T_{\rm c}$, while for superconducting domains, embedded in a nonsuperconducting matrix, it does not vanish and exhibits an inflection point at $T_{{\rm p}_k} < T_{\rm c}$, so that

$$\left.{\rm d}\left(\frac{1}{\lambda_i(T)\lambda_j(T)}\right)\middle/{\rm d}T\right|_{T=T_{{\rm p}_k}} = \text{extremum}. \tag{5}$$

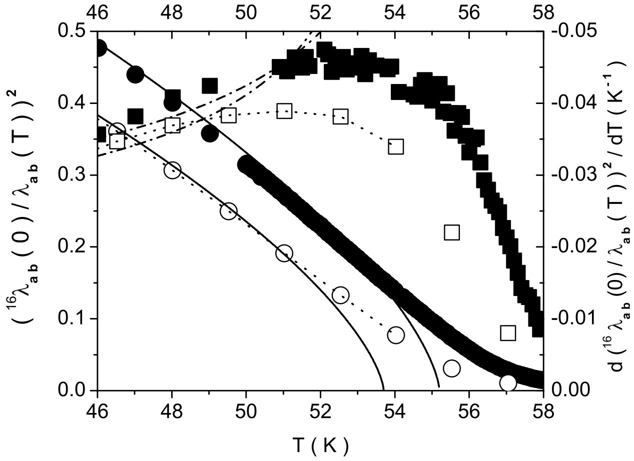

Figure 2. ${}^{16}\lambda^2_{ab}(T = 0)/\lambda^2_{ab}(T)$ (○, ^{16}O; ●, ^{18}O) and $\mathrm{d}(\lambda^2_{ab}(T = 0)/\lambda^2_{ab}(T))/\,\mathrm{d}T$ (□, ^{16}O; ■, ^{18}O) versus T for $Y_{0.7}Pr_{0.3}Ba_2Cu_3O_7$ with ^{16}O and ^{18}O. The solid and dash–dot curves indicate the leading critical behaviour of the homogeneous system as explained in the text.

Table 1. Finite size estimates for ${}^{16}T_{\mathrm{p}_c}$, ${}^{18}T_{\mathrm{p}_c}$, ${}^{16}\lambda^2_{ab}({}^{16}T_{\mathrm{p}_c})/{}^{16}\lambda^2_{ab}(0)$ and ${}^{18}\lambda^2_{ab}({}^{18}T_{\mathrm{p}_c})/{}^{16}\lambda^2_{ab}(0)$, and the resulting relative shifts $\Delta T_{\mathrm{p}_c}/T_{\mathrm{p}_c}$ and $\Delta\lambda^2_{ab}(T_{\mathrm{p}_c})/\lambda^2_{ab}(T_{\mathrm{p}_c})$ for an ^{18}O content of 89%. ${}^{16}L_{\mathrm{p}_c}$, ${}^{18}L_{\mathrm{p}_c}$ and $\Delta L_{\mathrm{p}_c}/L_{\mathrm{p}_c}$ follow from equation (4). ${}^{16}\lambda_{ab}(0)$ are μSR estimates [18].

x	0	0.2	0.3
$\Delta T_{\mathrm{p}_c}/T_{\mathrm{p}_c}$	−0.000(2)	−0.015(3)	−0.021(5)
$\Delta L_c/L_c$	0.12(5)	0.13(6)	0.16(5)
$\Delta\lambda^2_{ab}(T_{\mathrm{p}_c})/\lambda^2_{ab}(T_{\mathrm{p}_c})$	0.11(5)	0.15(6)	0.15(5)
${}^{16}\lambda^2_{ab}({}^{16}T_{\mathrm{p}_c})/{}^{16}\lambda^2_{ab}(0)$	4.4(2)	4.0(2)	4.4(2)
${}^{18}\lambda^2_{ab}({}^{18}T_{\mathrm{p}_c})/{}^{16}\lambda^2_{ab}(0)$	4.9(2)	4.6(2)	5.2(2)
${}^{16}T_{\mathrm{p}_c}$ (K)	89.0(1)	67.0(1)	52.1(2)
${}^{18}T_{\mathrm{p}_c}$ (K)	89.0(1)	66.0(2)	51.0(2)
${}^{16}L_c$ (Å)	9.7(4)	14.2(7)	19.5(8)
${}^{18}L_c$ (Å)	10.9(4)	16.0(7)	22.6(9)
${}^{16}\lambda_{ab}(0)$ (Å)	1250(10)	1820(20)	2310(30)

We are now prepared to perform the finite size scaling analysis of the penetration depth data. In figure 2 we display ${}^{16}\lambda^2_{ab}(T = 0)/\lambda^2_{ab}(T)$ and $\mathrm{d}(\lambda^2_{ab}(T = 0)/\lambda^2_{ab}(T))/\,\mathrm{d}T$ versus T for $Y_{0.7}Pr_{0.3}Ba_2Cu_3O_{7-\delta}$. The solid curves are $({}^{16}\lambda_{ab}(0)/{}^{16}\lambda_{ab}(T))^2 = 1.62(1 - T/{}^{16}T_{\mathrm{c}})^{\nu}$, $({}^{16}\lambda_{ab}(0)/{}^{18}\lambda_{ab}(T))^2 = 1.4(1 - T/{}^{18}T_{\mathrm{c}})^{\nu}$ with $\nu = 2/3$, ${}^{16}T_{\mathrm{c}} = 54.9$ K, ${}^{18}T_{\mathrm{c}} = 53.7$ K and the dash–dot curves the corresponding derivatives, indicating the leading critical behaviour of a domain, infinite in the c-direction. The extremes in the first derivative around $T \approx 52.1$ and 51 K for ^{16}O and ^{18}O respectively clearly reveal the existence of an inflection point, characterizing the occurrence of a finite size effect in $1/\lambda^2_{ab}$ (equation (5)). Using equation (4) and the estimates for T_{p_c}, ${}^{16}\lambda^2_{ab}(T_{\mathrm{p}_c})/{}^{16}\lambda^2_{ab}(0)$, ${}^{18}\lambda^2_{ab}(T_{\mathrm{p}_c})/{}^{16}\lambda^2_{ab}(0)$ and ${}^{16}\lambda_{ab}(0)$ listed in table 1, we obtain ${}^{16}L_c = 19.5(8)$ Å and ${}^{18}L_c = 22.6(9)$ Å for the spatial extents of the superconducting domains along the c-axis. Note that the rather broad peak around the inflection point reflects the small value of L_c. Indeed, in Bi2212, where the same analysis gives $L_c \approx 68$ Å, this peak was found to be considerably sharper [25]. To explore the dependence

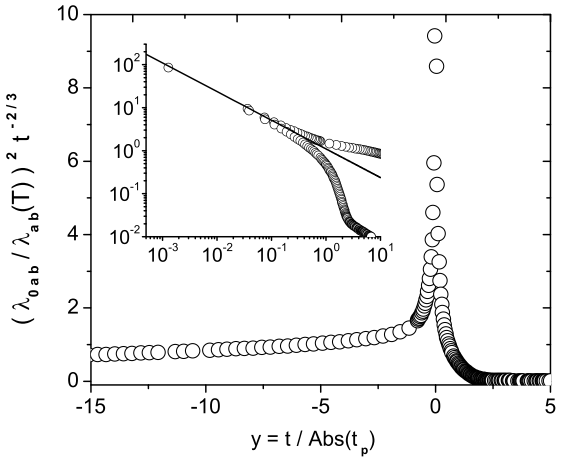

Figure 3. Finite size scaling function $g(y) = (\lambda_{0ab}/\lambda_{ab}(T))^2|t|^{-\nu}$ versus $y = t/|t_{p_c}|$ for the ^{16}O data shown in figure 2. The solid line in the inset is equation (7) with $g_0 = 1.1$.

of this change on Pr concentration we performed analogous magnetization measurements on $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ with $x = 0$ and 0.2 and extracted the quantities of interest as outlined above. The finite size estimates and the corresponding isotope shifts, for an ^{18}O content 89%, are summarized in table 1 (we define the relative oxygen isotope shift of a physical quantity X as $\Delta X/X = (^{18}X - {}^{16}X)/{}^{16}X$).

To substantiate the occurrence of a finite size effect further, we consider the scaling function [26]

$$\left(\frac{\lambda_{0ab}}{\lambda_{ab}(T)}\right)^2 |t|^{-\nu} = g(y), \qquad y = \mathrm{sign}(t)\frac{|t|}{t_{p_c}}. \tag{6}$$

Given a finite size it tends to $g(y \to \infty) = 1$ and $g(y \to -\infty) = 0$ for small t and $L_c \to \infty$, while for $t = 0$ and $L_c \neq 0$ it diverges as

$$g(y \to 0) = g_0 y^{-\nu} = g_0\left(\frac{|t|}{t_{p_c}}\right)^{-\nu}, \tag{7}$$

so that in this limit $(\lambda_{0ab}/\lambda_{ab}(T_c, L_c))^2 = g_0|t_p|^\nu = g_0\xi^t_{0ab}/L_c$. As expected, a sharp superconductor to normal state transition requires homogeneous domains of infinite extent. Moreover at t_{p_c}, $y = 1$ and $\mathrm{d}(\lambda_{0ab}/\lambda_{ab}(T, L))^2/\,\mathrm{d}t = 0$. Accordingly, there is an inflection point at t_{p_c}. Since the so-called finite size scaling function $g(y)$ depends on the type of confining geometry and on the conditions imposed at the boundaries of the domains, this applies to the amplitude g_0 as well. As an example we display in figure 3 the scaling function obtained from the ^{16}O data shown in figure 2. Apparently it is fully consistent with a finite size effect. Noting that the same behaviour was found in the six $Y_{1-x}Pr_xBa_2Cu_3O_{7-\delta}$ samples with ^{16}O and ^{18}O considered here, as well as in the in-plane penetration depth data of Panagopoulos *et al* [19], Jacobs *et al* [21] and Osborn *et al* [22], it becomes clear that the tail in the penetration depth near T_{p_c} is fully consistent with a finite size effect.

From table 1 several observations emerge. First, L_c increases systematically with reduced T_{p_c}. Second, L_c grows with increasing x and upon isotope exchange (^{16}O, ^{18}O). Third, the relative shift of T_{p_c} is very small. This reflects the fact that the change of L_c is essentially due to the superfluid, probed in terms of λ^2_{ab}. Accordingly, $\Delta L_c/L_c \approx \Delta\lambda^2_{ab}/\lambda^2_{ab}$ for $x = 0,\ 0.2$

and 0.3. Indeed the relative shifts of T_{p_c}, $\lambda_{ab}^2(T_{p_c})$ and L_c are not independent. Equation (4) implies

$$\frac{\Delta L_c}{L_c} = \frac{\Delta T_{p_c}}{T_{p_c}} + \frac{\Delta \lambda_{ab}^2(T_{p_c})}{\lambda_{ab}^2(T_{p_c})}. \qquad (8)$$

To appreciate the implications of these estimates, we note that for fixed Pr concentration the lattice parameters remain essentially unaffected [12, 13]. Accordingly, an electronic mechanism, without coupling to local lattice distortions and anharmonic phonons, implies $\Delta L_c = 0$. In contrast, a significant change of L_{p_c} upon oxygen exchange uncovers the coupling to local lattice distortions and anharmonic phonons involving the oxygen lattice degrees of freedom. A glance at table 1 shows that the relative change of the domains along the c-axis upon oxygen isotope exchange is significant, ranging from 12 to 16%, while the relative change of the inflection point or the transition temperature is an order of magnitude smaller. For this reason the significant relative change of L_c at fixed Pr concentration is accompanied by essentially the same relative change of λ_{ab}^2, which probes the superfluid. Noting that the oxygen isotope exchange modifies the lattice degrees of freedom, this uncovers unambiguously the existence and relevance of the coupling between the superfluid, lattice distortions and anharmonic phonons involving the oxygen. Potential candidates are the Cu–O bond-stretching-type phonons showing temperature dependence, which parallels that of the superconductive order parameter [27]. Independent evidence for the shrinkage of limiting length scales upon isotope exchange stems from the behaviour close to the quantum superconductor to insulator transition where T_c vanishes [28]. Here the cuprates become essentially two dimensional and correspond to a stack of independent slabs of thickness d_s [29, 30]. It was found that the relative shift $\Delta d_s/d_s$ upon isotope exchange adopts a rather unique value, namely $\Delta d_s/d_s \approx 0.03$ [28]. Furthermore, our estimates for L_{p_c} reveal that the inhomogeneities in our samples are of nanoscale, in spite of the evidence for the absence of phase separation into hole-rich and hole-poor regions [9].

In conclusion, we have reported the first observation of the combined finite size and oxygen isotope exchange effects on the spatial extent L_c of the superconducting domains along the c-axis and the in-plane penetration depth λ_{ab}. Although the majority opinion on the mechanism of superconductivity in the cuprates is that it occurs via a purely electronic mechanism involving spin excitations, and lattice degrees of freedom are supposed to be irrelevant, we have shown that the relative isotope shift $\Delta L_c/{}^{16}L_c \approx \Delta\lambda_{ab}^2/{}^{16}\lambda_{ab}^2 \approx 0.15$ uncovers clearly the existence and relevance of the coupling between the superfluid, lattice distortions and anharmonic phonons which involve the oxygen lattice degrees of freedom.

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