

Effects of the Cu-O chains on the anisotropic penetration depth of $\text{YBa}_2\text{Cu}_4\text{O}_8$

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The temperature, T , dependence of the magnetic penetration depth, λ , of grain-aligned $\text{YBa}_2\text{Cu}_4\text{O}_8$ has been measured along the ab plane and c axis. Both $\lambda_{ab}(T)$ and $\lambda_c(T)$ vary as \sqrt{T} up to $\sim 0.4T_c$ implying a square-root density of states at low energy. The results are discussed in terms of a proximity model of alternating stacked superconducting and normal layers. [S0163-1829(99)51310-6]

The anisotropic magnetic penetration depth has been used to study the systematic behavior of the order parameter, superfluid density, ρ^s , and c -axis coupling of high- T_c superconductors (HTS's).^{1,2} In the case of $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (Y-123) and related materials the metallic Cu-O chains provide an additional electronic system which is responsible for a number of its enhanced features. Although infrared³ and muon spin-relaxation (μSR) (Ref. 4) studies show a strongly anisotropic penetration depth at $T=0$ associated with additional superfluid density on the chains, the temperature dependence of the superfluid density remains unclear. Studies on $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ single crystals⁵ and magnetically aligned powders^{6,7} showed a linear T dependence at low temperature. However, we emphasize that these samples possess significant, albeit small vacancy and interstitial [O(5) site] disorder on the chains, potentially causing scattering and pair breaking in this secondary system. Pair breaking is well known to alter the behavior of the density of states (DOS) and temperature dependence. An alternative, and in fact ideal, system to obtain information on chain superconductivity is $\text{YBa}_2\text{Cu}_4\text{O}_8$ (Y-124) which has two fully oxygenated chains per unit cell which can be remarkably free of disorder.⁸

With this aim we performed anisotropic magnetic penetration-depth measurements using the ac-susceptibility technique on magnetically aligned powders of Y-124. We report the values and temperature dependence of the in-plane, λ_{ab} , and out-of-plane, λ_c , penetration depths. The estimated anisotropy ratio $\lambda_c(0)/\lambda_{ab}(0)=5$ is in good agreement with earlier infrared⁵ and recent resistivity⁸ measurements. Both λ_{ab} and λ_c show a unique \sqrt{T} temperature dependence up to $\sim 0.4T_c$ indicating a square-root density of states at low energy. The results are discussed in the context of a multicomponent superfluid response associated with the double chains in Y-124.

Polycrystalline samples of $\text{YBa}_2\text{Cu}_4\text{O}_8$ were prepared at 935 °C in oxygen at 6 MPa pressure by stoichiometric solid-state reaction of high-purity dried powders of CuO , Y_2O_3 , and $\text{Ba}(\text{NO}_3)_2$, the nitrate being first decomposed by slow heating of the powders to 710 °C in air. The samples were

reacted as die-pressed pellets for a total of 96 h with intermediate milling and pelletizing every 16 h. X-ray diffraction showed the samples to be single phase and scanning electron microscope backscattered images showed the only impurity, CuO , to be present at less than 0.5%. The measured critical temperature T_c , that is the value where the onset of superconductivity occurs in the ac-susceptibility data for a measuring ac field $H_{ac}=3$ G rms and frequency $f=333$ Hz, is 81.5 K. A bulk piece was lightly ground for 45 min and sieved in an argon glove box. The powder was magnetically aligned in epoxy as described earlier.^{1,6,7,9} The average grain diameter corresponding to the 50% cumulative volume point is 2 μm . The fraction of unoriented powder in all grain-aligned samples was estimated to be $\sim 8\%$. Rocking curve analysis of the aligned samples gave a full width at half maximum of $\sim 2^\circ$. Low-field ac-susceptibility, χ , measurements were performed on four samples, aligned separately but under the same conditions, for $H_{ac}=1-4$ G rms at $f=333$ Hz with the ac field applied either in the ab plane or along the c axis. The separation of the grains and the absence of weak links was confirmed by checking the linearity of the signal for $H_{ac}=1-8$ G rms and $f=33-667$ Hz. Details of the experimental technique and the application of London's model for deriving λ from the measured χ in cuprate superconductors have been extensively discussed in earlier publications.^{1,6,7,9,10}

The values of $\lambda_{ab}(0)$ and $\lambda_c(0)$ derived from our data are 127 ± 17 nm and 615 ± 90 nm, respectively. The results shown here are characteristic of a set of data obtained for several grain-aligned Y-124 pieces as cut from four different batches of grain-aligned samples all prepared under the same conditions. Although there was a small variation in the quantity ($\pm 4\%$) and quality ($\pm 0.4^\circ$) of alignment between the four batches the results were essentially the same. From the various samples measured we have found that the temperature dependence of λ is not affected more than 3% by the uncertainties in $\lambda(0)$.

Figure 1(a) shows a plot of the temperature dependence of $\lambda_{ab}^{-2}(T)$. The inset of this figure shows a normalized plot of

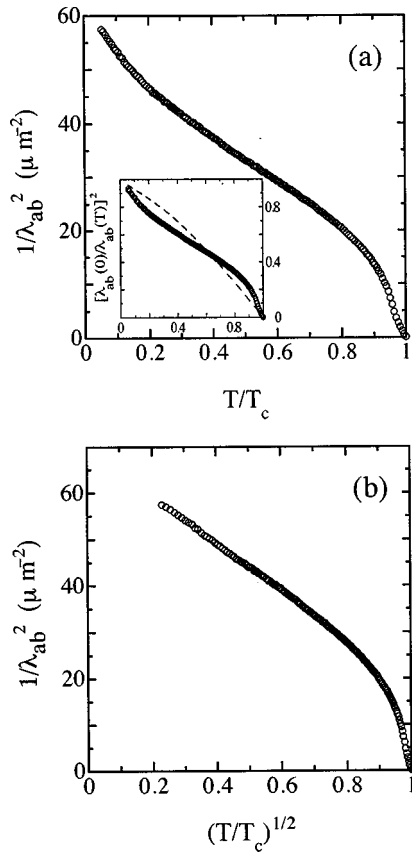


FIG. 1. (a) Temperature dependence of the measured $\lambda_{ab}^{-2}(T)$ for grain-aligned $\text{YBa}_2\text{Cu}_4\text{O}_8$. The inset shows a normalized plot of $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ compared with the weak-coupling theory (broken line) for a d -wave superconductor. (b) The data of (a) replotted as a function of $(T/T_c)^{1/2}$.

$[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ compared with the weak-coupling theory for a d -wave superconductor. Evidently, the T dependence of the superfluid density exhibits two clear departures from the weak-coupling d -wave behavior: just below T_c where the initial development of superfluid density is significantly more rapid than the model behavior and below 15 K where there appears to be a further enhancement in superfluid density. The observed behavior is independent of the magnitude of H_{ac} and f . We stress that in all the HTS materials we have investigated both behaviors are unique. The distinguishing feature of Y-124 is of course the double chains which are noted from resistivity studies to be singularly free of defects.⁸

We note that much earlier μSR investigations of Y-124, Zn-substituted Y-124, and Y-247 at different oxygen contents, showed signatures for a distinctive rise in superfluid density at low temperature that had not been seen in other cuprates.^{4,11} However, the resolution and temperature spacing was not suitable to adequately characterize this behavior. This rise in superfluid density which is common to all high- T_c cuprates with double chains must surely be interpreted as a multicomponent superfluid response associated with the coupled planes and double chains. Furthermore, more recent evidence by microwave measurements¹² on $\text{YBa}_2\text{Cu}_3\text{O}_{6.95}$ single crystals grown in BaZrO_3 crucibles also points to a two-component superconductivity although the chain enhancements to the superfluid density are not as obvious as in

the present case. This is presumably due to the high degree of order on the Y-124 double chains.

We believe that the enhancements at low and at high temperature both must be associated with the chains. The jump $\Delta C_p/T_c$ in heat capacity at T_c for Y-124 is 1.1 mJ/g at K,¹³ while for oxygen-deficient Y-123 with $T_c = 81$ K the jump is only 0.55 mJ/g at K.¹⁴ As the jump in heat capacity is a direct measure of the pair density, the enhancement for Y-124 is probably due to proximity-induced condensation of carriers on the “double chains” occurring at T_c . To underscore the fact that this enhancement in $\Delta C_p/T_c$ is significant we note that many other properties are identical for Y-124 and oxygen-deficient Y-123 with the same T_c . These include the temperature-dependent Knight shift¹⁵ which mirrors the entropy, S/T ,¹⁶ and the rate of depression of T_c with Zn substitution^{4,6} which indicates that the density of states at T_c is the same for the two compounds.

In Fig. 1(b) we plot $\lambda_{ab}^{-2}(T)$ as a function of $(T/T_c)^{1/2}$. A striking \sqrt{T} dependence of $\lambda_{ab}^{-2}(T)$ is observed all the way to $\sim 0.4T_c$. To the best of our knowledge this is the first observation of a \sqrt{T} power law for $\lambda_{ab}(T)$, for a cuprate superconductor. A \sqrt{T} behavior directly implies that the DOS of quasiparticles has a square-root energy dependence at low energy¹⁷ which differs from the pure two-dimensional d -wave superconducting system where the low-energy density of states is linear.

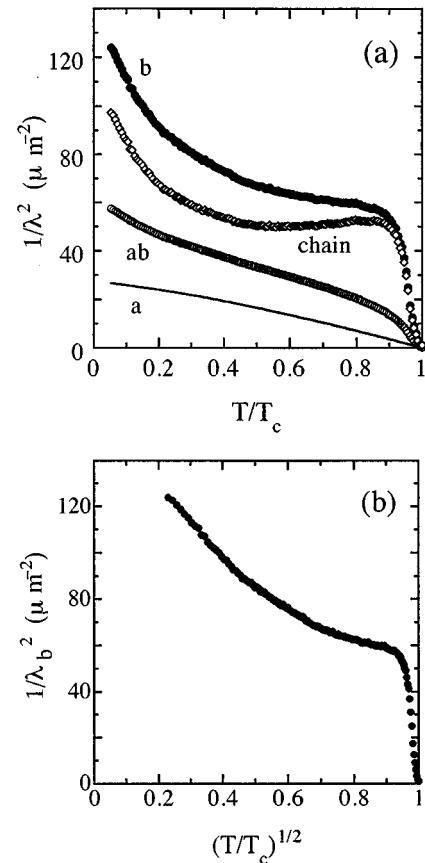


FIG. 2. (a) λ_{ab}^{-2} (open circles), λ_a^{-2} (solid line), and λ_b^{-2} (closed circles), and $\lambda_{\text{chain}}^{-2}$ (open diamonds) as functions of T/T_c for grain-aligned $\text{YBa}_2\text{Cu}_4\text{O}_8$. (b) Plot of the extracted λ_b^{-2} as a function of $(T/T_c)^{1/2}$.

Due to the powder nature of our samples we are not able to measure separately the a and b components. However, we can estimate the T dependence of the in-plane anisotropy as follows. μ SR studies show the 90 K optimally doped cuprates without chains to have a $T=0$ muon spin depolarization rate of $3.0 \mu\text{s}^{-1}$. This corresponds to $\lambda_a(0)=154 \text{ nm}$ [$=\lambda_{ab}(0)$ for the nonchain cuprates]. Underdoped cuprates such as Y-124 have a superfluid density which is diminished by the presence of the normal-state pseudogap. Using the theory of Williams, Tallon, and Haines¹⁸ a pseudogap energy sufficient to reduce T_c from 93.5 K to 81 K reduces the superfluid density and increases $\lambda_a(0)$ from 154 nm to 188 nm. This is in excellent agreement with the value 190 nm obtained by Basov *et al.*³ from infrared studies. Our experimental value for Y-124 of $\lambda_{ab}(0)=127 \text{ nm}$ allows λ_b to be calculated from $\lambda_{ab}^2=\lambda_a\lambda_b$ (Ref. 19), giving $\lambda_b(0)=86 \text{ nm}$, again in excellent agreement with the infrared studies of Basov *et al.*³ This concurrence gives us confidence in applying the same analysis to the temperature-dependent data.

Based on previous experience with the cuprates^{1,2,6,7,9} we assume that λ_a^{-2} has the weak-coupling d -wave temperature dependence. Using this temperature dependence for λ_a^{-2} we determine $\lambda_b(T)$ using $\lambda_{ab}^2(T)=\lambda_a(T)\lambda_b(T)$. Finally, assuming that for the b direction, the superfluid density on the chains is additive to that on the planes, then $\lambda_{\text{chain}}^{-2}=\lambda_b^{-2}-\lambda_a^{-2}$. The T dependence of these parameters, λ_{ab}^{-2} , λ_a^{-2} , λ_b^{-2} , and $\lambda_{\text{chain}}^{-2}$ is plotted in Fig. 2(a). It is clear that λ_b^{-2} shows a pronounced increase at low T and $\lambda_{\text{chain}}^{-2}$ even more so. Surprisingly, the enhancement actually starts from T_c . The details of the deduced T dependence are of course influenced by our assumptions in extracting λ_b^{-2} but it is clear that the strong enhancement below T_c is evident in the basic data for λ_{ab}^{-2} . Factors which might affect the T dependence here include phase fluctuations, inelastic scattering, and the strongly T -dependent anisotropy.

Following the ideas of Kresin *et al.*²⁰ and other authors²¹ on proximity-induced chain superconductivity, Xiang and Wheatley¹⁷ (XW) have investigated a model system of coupled CuO_2 planes and CuO chains exploring two scenarios: (i) a proximity model where plane and chain layers are coupled through single electron tunneling, and (ii) an interlayer pair-tunneling model where planes and chains are Josephson coupled. For the former they found, to the leading order approximation, the low-temperature limits $\rho_b^s \sim \sqrt{T}$ for the b axis and $\rho_c^s \sim T$ for the c axis, while for the latter $\rho_b^s \sim T$ and ρ_c^s was found to follow a T^2 dependence. The low-temperature data shown in Figs. 1 and 2 agree well with the proximity model although the enhancement seen near T_c is not predicted. However, we note that the XW model is valid only at low temperatures, where in fact agreement with experiment is found.

Figure 3(a) shows the T dependence of λ_c^{-2} and here also there is a clear enhancement in the superfluid just below T_c and, notably, a \sqrt{T} dependence at low T . Below $0.1T_c$ the data tends towards linear but it is not clear whether this is significant without lower temperature data. In Figs. 2(b) and 3(b) we replot $\lambda_b^{-2}(T)$ and $\lambda_c^{-2}(T)$ as functions of $(T/T_c)^{1/2}$. For both, the \sqrt{T} dependence persists to rather high values of T/T_c . An enhancement in the low temperature $\lambda_c(T)$ is a

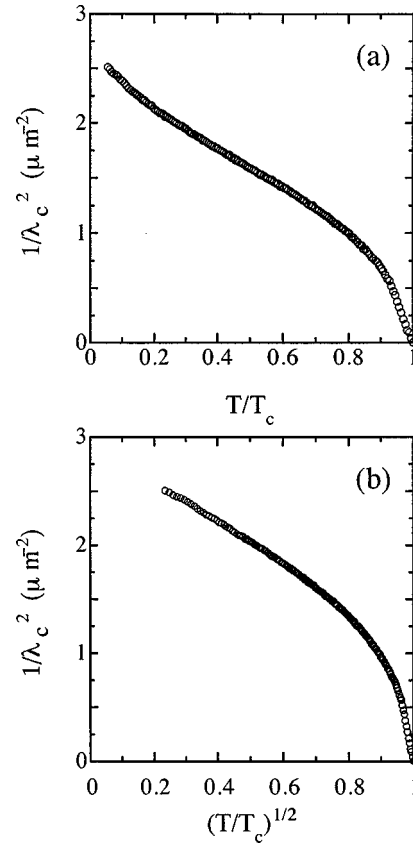


FIG. 3. Plots of the measured λ_c^{-2} as a function of T/T_c (a) and $(T/T_c)^{1/2}$ (b) for grain-aligned $\text{YBa}_2\text{Cu}_4\text{O}_8$.

natural consequence of the proximity model but XW predict this enhancement to vary linearly with T rather than as the experimentally observed \sqrt{T} dependence. However, we emphasize that the $\lambda_c(T)$ theoretical result is purely numerical. We can only assume that this feature did not emerge from the XW analysis because their approach did not incorporate the double chains of the Y-124 compound. c -axis transport necessarily involves some b -axis transport along the chains because of the $b/2$ offset of the chains relative to each other. In this case, to analyze the detailed T dependence of λ_c , a proximity (NSSN) (NSSN)' model should be considered, where S represents a CuO_2 layer, N a CuO layer, and (NSSN)' represents the unit cell which is offset by a half lattice constant along the b axis with respect to its neighboring unit cell (NSSN). On the other hand, the \sqrt{T} dependence may point to the c -axis superfluid response being governed by the DOS as, for example, in Anderson's interlayer coupling model.²²

There is no evidence for an independent pairing potential on the chains as discussed by XW that would cause a sudden rise in superfluid density over and above the plane contribution near some temperature T_c^{chain} lying below T_c^{plane} . In spite of the observation of superconductivity in the ladder compound SrCuO_2 (Ref. 23) there is no indication of intrinsic chain superconductivity in Y-124. Otherwise the substitution of Zn or Ni exclusively on the CuO_2 planes in Y-124 would effectively expose the chain superconductivity by destroying the plane superconductivity. The critical temperature would fall, then level out at a concentration of a few percent. On the contrary the suppression of T_c by such substitutions is rapid and monotonic.²⁴

We turn now to the rapid rise in the observed superfluid density, just below T_c . We find unlikely the possibility that, with the onset of superconductivity there occurs additional charge transfer from the chains to the planes giving a growth in superfluid density towards a value more in line with a higher doping state and a higher T_c . Such charge transfer would be a natural consequence of the condensation energy on the planes being higher than on the chains. Bond valence sums (BVS's) provide a means to assess doping state and charge transfer within the high- T_c cuprates. The parameter $V_- = 2 + V_{\text{Cu}(2)} - V_{\text{O}(2)} - V_{\text{O}(3)}$, where V_{Cu} is the copper BVS and $V_{\text{O}(2)}$ and $V_{\text{O}(3)}$ are the oxygen BVS's, has been found to provide a reliable estimate of the hole concentration, p , on the CuO_2 planes.²⁵ We have calculated the temperature dependence of V_- from bond lengths using the neutron struc-

tural refinements of Kaldis *et al.*²⁶ and find no systematic variation above or below T_c within ± 0.003 .

In summary, using the ac-susceptibility method on grain-aligned $\text{YBa}_2\text{Cu}_3\text{O}_8$ we have measured the anisotropic penetration depths $\lambda_{ab}(T)$ and $\lambda_c(T)$ and compared the data with a recent proximity-coupled model. We find that both λ_{ab}^{-2} and λ_c^{-2} possess a \sqrt{T} dependence at low temperature. Such a behavior is not seen in other HTS cuprates and is attributable to the effect of the double Cu-O chains on the T dependence of the superfluid density, resulting in a new type, \sqrt{T} , of low-temperature DOS.

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