Systematic behavior of the in-plane penetration depth in *d*-wave cuprates

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We report the temperature (T) and oxygen concentration dependences of the penetration depth of grain-aligned $YBa_2Cu_3O_{7-\delta}$ with $\delta=0.0$, 0.3, and 0.43. The values of the in-plane $\lambda_{ab}(0)$ and out-of-plane $\lambda_c(0)$ penetration depths, the low-temperature linear term in $\lambda_{ab}(T)$, and the ratio $[\lambda_c(0)/\lambda_{ab}(0)]$ were found to increase with increasing δ. The systematic changes of the linear term in $\lambda_{ab}(T)$ with T_c found here and in recent work on $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$ ($n=1$ and 3) are discussed. [S0163-1829(98)04121-6]

In a recent study[1] of the c-axis coupling of d-wave high-T_c cuprates we reported the values and temperature (T) dependences of the in-plane (λ_{ab}) and out-of-plane (λ_c) penetration depths for slightly overdoped[2,3] $HgBa_2CuO_{4+\delta}$ (Hg-1201) with critical temperature $T_c=93$ K and slightly underdoped[4,5] $HgBa_2Ca_2Cu_3O_{8+\delta}$ (Hg-1223) with $T_c=135$ K. For both compounds the low-temperature dependence of λ_{ab} was found to be linear as expected for d-wave superconductivity. In fact normalized plots of $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ versus T/T_c were the same, and like $YBa_2Cu_3O_7$ ($YBCO_7$) (Ref. 6) agreed very well with mean-field (MF) theory for a weak-coupling d-wave superconductor. However, recent angle-resolved photoemission spectroscopy[7] (ARPES) and tunneling[8] data strongly suggest that for underdoped samples the superconducting gap Δ_0 remains constant, or even increases slightly, while T_c falls and so large deviations from MF theory might be expected. We have therefore extended our investigation to deoxygenated (underdoped) pure $YBa_2Cu_3O_{7-\delta}$ using the same ac susceptibility technique to measure the penetration depth.[1,6,9,10]

We report experimental results for the values and temperature dependences of λ_{ab} and λ_c of high-quality c-axis grain-aligned orthorhombic[11] $YBa_2Cu_3O_{7-\delta}$ (which has two CuO_2 planes per unit cell as well as Cu-O chains) with $\delta=0.0$, 0.3, and 0.43, and compare them with tetragonal[12] Hg-1201 with one CuO_2 plane per unit cell and tetragonal[13] Hg-1223 with three CuO_2 planes per unit cell. We find that the presence of the linear term in $\lambda_{ab}(T)$ is independent of the number of CuO_2 planes per unit cell, carrier concentration, crystal structure, anisotropy and the presence of chains. Surprisingly our data show good agreement with weak-coupling d-wave theory, and the linear term in $[\lambda_{ab}(T)/\lambda_{ab}(0)]$ appears to scale with T/T_c. This result highlights the need for detailed consideration of the relationship between superconducting and normal-state energy gaps in underdoped cuprates.

Sample preparation was carried out by the standard solid-state reaction process using high-purity (99.999%) Y_2O_3, $BaCO_3$, and CuO oxides. Electron probe microanalysis and x-ray diffraction showed that all samples were single phase within an accuracy of $\sim1\%$. The fully oxygenated, $\delta=0.0$ ($YBCO_7$, $T_c=92$ K), samples were prepared by annealing bulk pieces in pure oxygen atmosphere at 380 °C for 24 h and then slowly cooling to room temperature. (Hereafter T_c represents the temperature where the onset of superconductivity occurs in the ac susceptibility data for a measuring field $H_{ac}=3$ G rms and frequency $f=333$ Hz.) The $\delta=0.3$ ($YBCO_{6.7}$, $T_c=66$ K) sample was prepared by annealing in pure oxygen atmosphere at 650 °C for 12 h and then quenching in liquid nitrogen, while the $\delta=0.43$ ($YBCO_{6.57}$, $T_c=56$ K) sample was annealed in 0.2% O_2/N_2 atmosphere at 550 °C for 12 h and also quenched into liquid nitrogen. The final oxygen contents were determined from the weight change of a fully oxygenated reference sample. The $\delta=0.0$ bulk piece was lightly ground and sedimented in acetone to obtain a well-defined grain size distribution. The sedimented powders were then heat treated to repair any structural damages to the surface of the grains.[14] For $\delta=0.3$ and 0.43, on the other hand, a bulk piece for each δ was lightly ground and sieved through a 20 μm sieve in an argon glove box to obtain a well-defined grain size distribution[10] and to avoid surface degradation of the crystallites.[14] The collected powders were then kept in argon atmosphere for 30 min before being aligned. All powders, $\delta=0.0$, 0.3, and 0.43, were magnetically aligned in epoxy as described earlier.[1,6,10] The average grain diameters corresponding to the 50% cumulative volume point were 5 and 10 μm for the fully oxygenated and the oxygen deficient samples, respectively. The fraction of the unoriented powder in all grain-aligned samples was estimated to be $<5\%$. Rocking curve analysis of the $\delta=0.0$ and $\delta>0.0$ samples gave a full width at half maximum of $\pm1.4°$ and $\pm1°$, respectively.[15] Low-field susceptibility χ measurements were performed using commercial equipment (down to 4.2 K) for samples with $\delta=0.0$, 0.3, and 0.43. The sample with $\delta=0.43$ was also measured down to 1.2 K using a home built susceptometer. Details of the experimental technique and the application of London's model for deriving λ from the measured χ in cuprate superconductors can be found in earlier publications.[1,6,9,10,16]

The values of $\lambda_{ab}(0)$ derived from our data are 0.14, 0.21, and 0.28 μm and the corresponding values for $\lambda_c(0)$ are 1.26, 4.53, and 7.17 μm for $\delta=0.0$, 0.3, and 0.43, respectively. The errors in $\lambda_{ab}(0)$ arising from a possible uncertainty of $\pm5\%$ in the alignment can be as high as $\pm25\%$, whereas those in $\lambda_c(0)$ are $\pm8\%$. However, the corresponding uncertainty in the linear term in $[\lambda_{ab}(T)/\lambda_{ab}(0)]$ is

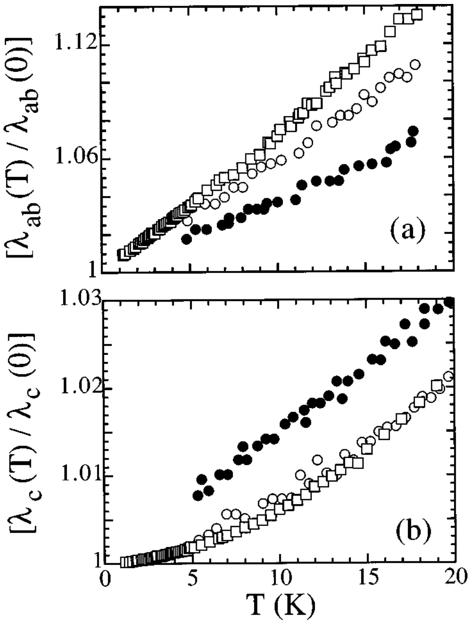

FIG. 1. Low-temperature plots of (a) $[\lambda_{ab}(T)/\lambda_{ab}(0)]$ and (b) $[\lambda_c(T)/\lambda_c(0)]$ for $YBCO_7$ (closed circles), $YBCO_{6.7}$ (open circles), and $YBCO_{6.57}$ (open squares). The T_c, $\lambda_{ab}(0)$, and $\lambda_c(0)$ values are given in the text.

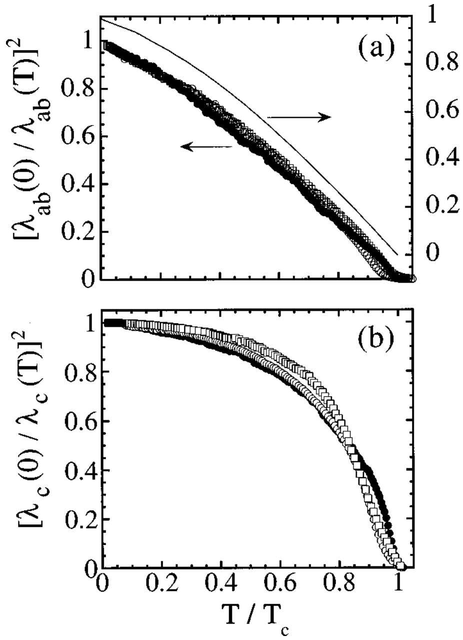

FIG. 2. Plots of (a) $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ and (b) $[\lambda_c(0)/\lambda_c(T)]^2$ as functions of T/T_c for $YBCO_7$ (closed circles), $YBCO_{6.7}$ (open circles), and $YBCO_{6.57}$ (open squares). The solid line in (a) is the theoretical prediction for the normalized superfluid density from the weak-coupling BCS theory for a d-wave superconductor (Ref. 23).

much less, at most $\pm 10\%$. The present results differ from previous work[17] in which the surfaces of the particles were probably not as clean and the degree of grain alignment was probably lower. As T_c is reduced by lowering the carrier concentration (for $\delta = 0.3$ and 0.43), $[1/\lambda_{ab}^2(0)]$ falls, a behavior which has been extensively discussed in terms of the Uemura relation.[18,19] The ratio $\gamma = [\lambda_c(0)/\lambda_{ab}(0)]$, i.e., the anisotropy, increases with oxygen deficiency.

Figures 1(a) and 1(b), show characteristic low-temperature plots of $[\lambda(T)/\lambda(0)]$ for the ab plane (measured with the applied field $H \| c$) and c axis (measured with $H \| ab$), respectively, for the three oxygen concentrations studied. The low-temperature ($T/T_c < 0.25$) linear term in $\lambda_{ab}(T)$, is 4.8 Å/K for $YBCO_7$ in good agreement with that found from microwave measurements on $YBCO_{6.95}$ single crystals.[20] As oxygen is removed from the lattice (the chains) the linear term increases to 12 and 20 Å/K for $\delta = 0.3$ and 0.43, respectively. For $YBCO_7$ we also observe a linear T dependence in λ_c at low temperatures but the relative change is about a factor of 2 smaller than in $[\lambda_{ab}(T)/\lambda_{ab}(0)]$, while $\lambda_c(T)$ of $YBCO_{6.7}$ and $YBCO_{6.57}$ obeys a T^2 behavior at low T. Details of the systematics of $\lambda_c(T)$ of cuprate superconductors can be found in Refs. 1, 6, 21, 22.

In Fig. 2 we present normalized plots of $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ and $[\lambda_c(0)/\lambda_c(T)]^2$ versus T/T_c. There is excellent agreement between the $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ curves for the three oxygen concentrations. The data in Fig. 2(a) are compared with the weak-coupling theory for a d-wave superconductor (solid line).[23] It can be seen that the d-wave curve fits the data very well. On the other hand, the $[\lambda_c(0)/\lambda_c(T)]^2$ curves do not fit the d-wave curve and also differ from each other slightly, because of the effect of the interplane coupling on $\lambda_c(T)$.[1,6,10,21,22] We also find that $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ of $YBa_2Cu_3O_{7-\delta}$ agrees with that of Hg-1201 and Hg-1223.[1,10] The behavior of $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ is generally similar to that of $YBa_2(Cu_{1-x}Zn_x)_3O_7$ ($x = 0.02$ and 0.03),[6] except at very low temperatures where a T^2 term developed in the Zn-doped samples due to impurity scattering.

The full temperature dependences of $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ for $YBa_2Cu_3O_{7-\delta}$ ($\delta = 0.0$, 0.3, and 0.43), $YBa_2(Cu_{1-x}Zn_x)_3O_7$ ($x = 0.02$ and 0.03), Hg-1201 and Hg-1223 are also in agreement with recent data of $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212) (Ref. 24) and $Tl_2Ba_2CuO_{6+\delta}$ (Tl-2201) (Ref. 25) single crystals measured by a microwave technique with $H \| c$, but they only agree with another set of microwave data ($H \| c$) for Bi-2212 single crystals[26] at $T/T_c < 0.3$. At higher temperatures the data in Ref. 26 deviate from the weak-coupling d-wave calculation. Independent evidence for the scaling behavior of $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ with T/T_c can also be found in a recent publication by Bonn *et al.*[27] who measured the relative changes in λ with temperature for underdoped, optimally doped and slightly overdoped untwinned $YBa_2Cu_3O_{7-\delta}$ crystals using a microwave technique and $H \perp c$. However, in Ref. 27 the changes of $[\lambda_{a,b}(0)/\lambda_{a,b}(T)]^2$ for $YBCO_{6.95}$, at high temperatures, and $YBCO_{6.6}$, over the whole temperature range, were smaller than ours and closer, at high temperatures, to the Bi-2212 data in Ref. 26. We do not know the precise origin of this difference but we believe that for weakly coupled layers, data taken with $H \| c$ give the best measure of the superfluid density.

Figure 3 shows plots of $\{1 - [\lambda_{ab}(0)/\lambda_{ab}(T)]^2\}$ vs T which is equivalent to $\{[n_s(0) - n_s(T)]/n_s(0)\}$, i.e., the normalized density of quasiparticle excitations, where $n_s(T)$ is the density of condensed electrons at a temperature T, for all the samples studied. It is clear that the linear term in $\{1$

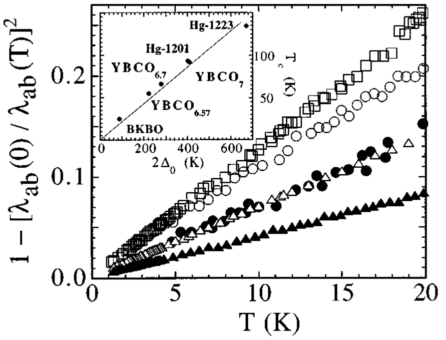

FIG. 3. Low-temperature plot of $\{1-[\lambda_{ab}(0)/\lambda_{ab}(T)]^2\}$ for Hg-1223 $[\lambda_{ab}(0)\approx 1770\pm 300$ Å$]$ (closed triangles) (Ref. 1), Hg-1201 $[\lambda_{ab}(0)\approx 1710\pm 250$ Å$]$ (open triangles) (Ref. 1), $YBCO_7$ (closed circles), $YBCO_{6.7}$ (open circles), and $YBCO_{6.57}$ (open squares). Inset: T_c versus $2\Delta_0$ as derived from the plot in the main panel (see text for details). BKBO is included for comparison. The solid line is drawn as a guide to the eye.

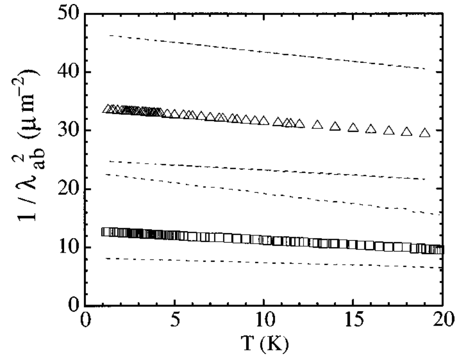

FIG. 4. Low-temperature plot of $[1/\lambda_{ab}{}^2(T)]$ (i.e., n_s) for $YBCO_{6.57}$ (open squares) and Hg-1201 (open triangles) showing the approximate parallel shift of n_s with T_c as discussed in the text. The dashed lines, immediately above and below each data set, indicate the maximum possible error in $1/\lambda_{ab}{}^2(T)$ arising from $\pm 5\%$ uncertainty in the alignment (see text).

$-[\lambda_{ab}(0)/\lambda_{ab}(T)]^2\}$ increases as T_c decreases. If we use the standard BCS result for $\lambda(T)$ of a d-wave superconductor,[28]

$$[\lambda(0)/\lambda(T)]^2 \approx 1-2(T/\Delta_0)\ln 2, \tag{1}$$

to fit the experimental data shown in Fig. 3 (at $T/T_c<0.25$), we find that Δ_0 scales approximately with T_c [Fig. 3 (inset)], giving $\Delta_0\approx 2T_c$, a value close to that expected for weak-coupling superconductivity.[28] For comparison we also include data for the s-wave perovskite $Ba_{0.6}K_{0.4}BiO_3$ (BKBO).[10] The compounds Bi-2212 (Refs. 24 and 26) and Tl-2201 (Ref. 25) would also give $\Delta_0\approx 2T_c$ on this plot. The maximum error in the linear terms, i.e., the values of Δ_0 in Fig. 3 (inset), is $\pm 20\%$.

The scaling of Δ_0 with T_c, Fig. 3 (inset), is in agreement with early tunneling spectroscopy data[29] for several cuprates as a function of carrier concentration, ranging from the underdoped to the optimally doped regimes. It is not consistent however, with more recent tunneling[8] and ARPES (Ref. 7) results for underdoped cuprates where Δ_0 was actually found to increase slightly while T_c falls. There seems to be two possible ways of accounting this discrepancy. One is that the recent spectroscopic experiments[7,8] actually measure the normal-state gap. In this scenario the effect of the normal-state gap would be to leave small pockets of holes whose superconducting properties are still described reasonably well by MF theory. The other is similar to a recent phenomenological approach.[30] As shown in Fig. 4, it is probable, that within experimental error, the *unnormalized* plots of $[1/\lambda_{ab}(T)^2]$, i.e., $n_s(T)$, versus T at low temperatures are parallel for samples with different T_c values. This would correspond to the same number of excited quasiparticles, $[n_s(0)-n_s(T)]$, at a given temperature for all T_c values—as implied by specific-heat work on underdoped YBCO.[31] Such parallel shifts give $[n_s(0)-n_s(T)]=\alpha T$, where α is independent of doping level (T_c). In combination with the well-known Uemura relation $n_s(0)\propto T_c$,[18] this gives $[1-n_s(T)/n_s(0)]=\beta T/T_c$, where β is independent of T_c. So at low T $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ versus T/T_c would still scale on to a single curve even when the MF relation $\Delta_0/T_c\approx 2$, is strongly violated.

In conclusion, we have studied $\lambda_{ab}(T)$ and $\lambda_c(T)$ of high-quality grain-aligned $YBa_2Cu_3O_{7-\delta}$ with $\delta=0.0$, 0.3, and 0.43. The values of $\lambda_{ab}(0)$, $\lambda_c(0)$, and γ were found to increase with oxygen deficiency. We find that the existence of the linear term in $\lambda_{ab}(T)$ is independent of the number of CuO_2 planes per unit cell, carrier concentration, crystal structure, anisotropy, and the presence of chains. If viewed in isolation, all the penetration depth data presented here and most of the microwave measurements for $H\|c$ appear to be in excellent agreement with mean-field theory for a weak-coupling d-wave superconductor for which $\Delta_0/T_c\approx 2$. However, recent spectroscopic data are more consistent with a different approach[30] in which there is a strong interplay between the superconducting and normal-state gaps. Clearly the relationship between these two gaps is of crucial importance for understanding superconductivity in the cuprates.

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