Variation of the temperature dependence of the penetration depth in $HgBa_2CuO_{4+\delta}$ with oxidation

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The penetration depth λ has been deduced from the reversible magnetization of three $HgBa_2CuO_{4+\delta}$ samples with doping δ's at and near the optimal value of 0.22. The obtained superfluid density $n_s \propto 1/\lambda^2$ at temperature T follows a parabolic law $[1-(T/T_c)^2]$ for the underdoped sample below T_c over the entire temperature region examined, where T_c is the superconducting transition temperature. However, n_s depends on T linearly for the optimally doped and slightly overdoped samples below 0.5 T_c. The observation suggests that $\lambda(T)$ of cuprate superconductors depends on oxidation in a rather complicated fashion and the interpretation of $\lambda(T)$ in terms of the superconducting pairing symmetry should be taken with care.

The temperature dependence of the in-plane penetration depth $\lambda(T)$ of a high-temperature superconductor (HTS) has recently attracted great attention, in an attempt to determine the pairing symmetry of a HTS. This is because $\lambda(T)$ is related to the superfluid density n_s as $n_s \propto 1/\lambda^2(T)$ and the temperature dependence of n_s is determined by the superconducting energy gap $\Delta(T)$ and thus the pairing symmetry of a superconductor. At high temperatures, e.g., above 0.5 T_c, Δ/T is small and can easily be affected or even dominated by other factors such as the superconducting coupling strength, carrier scattering, and the shape of the Fermi surface. The T dependence of λ consequently may vary greatly in this high-temperature region,[1-3] e.g., $1/\lambda^2(T) \propto (1-t^\alpha)$ with α ranging from 2 to 4, regardless of the pairing symmetry, where $t \equiv T/T_c$. However, at low temperature of $t \ll 0.5$ where Δ/T is large, $\lambda(T)$ appears to be a good measure of the low-lying excitations and, hence, the pairing symmetry in a superconductor. According to the BCS theory, $\lambda(T)$ of an s-wave superconductor depends on T exponentially and is given[4] by

$$\lambda(T) \sim \lambda(0)[1-3.3(T_c/T)^{1/2}\exp(-\Delta_0/T)]$$

or

$$\Delta\lambda(T) \equiv \lambda(T)-\lambda(0) \sim -3.3\lambda(0)(T_c/T)^{1/2}\exp(-\Delta_0/T)$$

if $\Delta_0/T \gg 1$, where $\lambda(0)$ is $\lambda(T)$ at 0 K and Δ_0 is the smallest value of Δ over the Fermi surface at 0 K. On the other hand, $\Delta\lambda(T) \propto T$ at low temperatures has been predicted[5] for a d-wave superconductor; where nodes or lines of nodes exist in the gap.

Different T-dependent $\Delta\lambda$'s on high quality $YBa_2Cu_3O_7$ samples with similar T_c ranging from linear[6] to quadratic[7] and even to exponential[8] have been reported. The difference has been attributed either to possible impurity scattering[9] which turns the linear T dependence characteristic of a d-wave superconductor to a quadratic T dependence, or possible proximity effects[8] which gives rise to a quasilinear T dependence in an s-wave superconductor. The effect of strong resonant scattering on $\lambda(T)$ of a d-wave superconductor has been examined theoretically.[10] It has been suggested[10] that $\Delta\lambda \propto T^\alpha$ where $\alpha=1$ for $T^* < T \ll T_c$ and $\alpha=2$ for $T<T^*$, where T^* is the so-called crossover temperature and increases with the concentration of the resonant scattering centers. Almost all experiments reported were carried out on the nearly optimally doped $YBa_2Cu_3O_{7-\delta}$ with maximum T_c. However, significant structural defects associated with a nonzero δ and the proximity effect on the CuO chains may still exist without notable change of the maximum T_c. We have therefore investigated by magnetization measurements the oxygen-doping effect on $\lambda(T)$ of $HgBa_2CuO_{4+\delta}$ with δ at and near the optimal value of 0.22. Although the impurity scattering seems to be quite small in the nearly optimally doped Hg-based compounds[11,12] and proximity effect is unlikely an issue in these compounds, we have observed an unambiguous variation of the T dependence of $1/\lambda^2$ along the a-b plane with doping δ, i.e., $1/\lambda^2 \propto 1-(T/T_c)^\alpha$ with $\alpha=2$ for the underdoped sample and $\alpha=1$ for the optimally doped and the overdoped samples below 0.5 T_c. The observation cannot be understood in terms of the impurity scattering effect in a d-wave superconductor only,[10] suggesting factors in addition to the pairing symmetry also influence α.

The polycrystalline samples of $Hg_{1-x}Ba_2CuO_{4+\delta}$ studied were prepared by the solid/vapor reaction technique. Hg-deficiency appears to help achieve pure $HgBa_2CuO_{4+\delta}$ as indicated by x-ray diffractions. The oxygen content δ of the samples was altered by heating the samples under different O_2-partial pressures at ~300 °C. Details have been published elsewhere.[12] The magnetization (M) of the samples was measured at various temperatures as a function of field (H) during field increasing as well as field decreasing, employing

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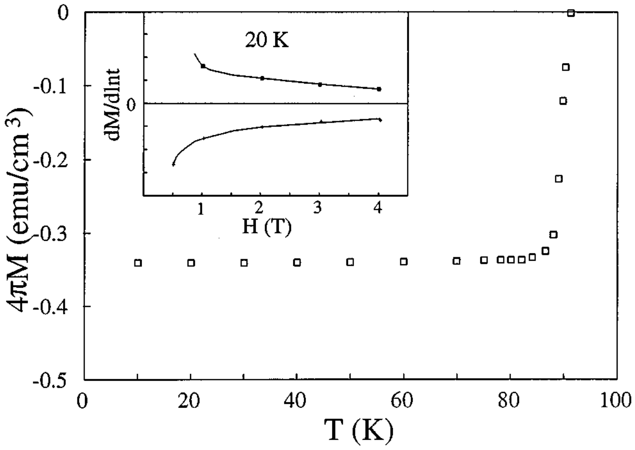

FIG. 1. The magnetization $M(T)$ of the overdoped $HgBa_2CuO_{4+\delta}$ with $\delta=0.28$ at 5 Oe determined in the field-cooled mode. Notice the sharp transition and the T-independent M below T_c. Inset: $+dM_+/d\ \ln t$ and $dM_-/d\ \ln t$ at 20 K for the same sample.

a Quantum Design superconducting quantum interference device (SQUID) magnetometer. Below the irreversibility line, the reversible M is taken as the average of the M's measured during H-increasing (M_+) and H-decreasing modes (M_-), i.e., reversible $M=(M_++M_-)/2$.

It has been shown that $HgBa_2CuO_{4+\delta}$ exhibits a large T_c variation through anion-doping with a minimum perturbation to the structural integrity.[12] By increasing δ from 0 to 0.4, T_c can be altered parabolically from 0 K to its maximum value of 97 K at optimal doping $\delta_{op}\sim0.22$ and back to 20 K. The compound is known as underdoped when $\delta<0.22$ and overdoped when $\delta>0.22$. For the present study, we have chosen to examine three samples with $\delta=0.16$, 0.23, and 0.28, and T_c's=88, 97, and 88 K, respectively. This is because of their high sample homogeneity as evident from their sharp transitions, and flat low field M determined in the field-cool (FC) mode below T_c, as shown in Fig. 1 for the $\delta=0.28$ sample. Samples show a general broadening of the transition width and a less flat M, presumably due to the inhomogeneous distribution of oxygen, as δ moves further away from 0.22.

Four theoretical models have been used to extract λ from the reversible M. According to the London model,[13] which considers only the electromagnetic energy in a type-II superconductor with H parallel to the c axis, $dM/d\ \ln H=\phi_0/(32\pi^2\lambda^2)$, where ϕ_0 is the flux quantum. By adding the free energy associated with the vortex cores to the London model, Hao and Clem[14] obtained $dM/d\ \ln H=\beta\phi_0/(32\pi^2\lambda^2)$, where β changes with H from ~0.7 for $0.1\leqslant H/H_{c2}\leqslant0.3$ to 0.84 for $0.02\leqslant H/H_{c2}\leqslant0.1$. Bulaevskii *et al.*[15] included the fluctuations effect and proposed $dM/d\ \ln H=\phi_0/(32\pi^2\lambda^2)-(M^*/T^*)T$ for H above a crossover field, where M^* and T^* are determined by the crossing point of $M(T)$'s at different H's. Near T_c, λ can also be deduced from the two-dimensional scaling as proposed by Tesanovic *et al.*[16] The values of $1/\lambda$ obtained from different models are shown as functions of temperature in Fig. 2(a) for the sample with $\delta=0.28$. The agreement between models appears to be good. Extending these models to lower temperature is debatable. However, the related uncertainty may not be significant at $H\leqslant5$ T, since the free energy of vortex is mainly determined by the energy of the magnetic field.

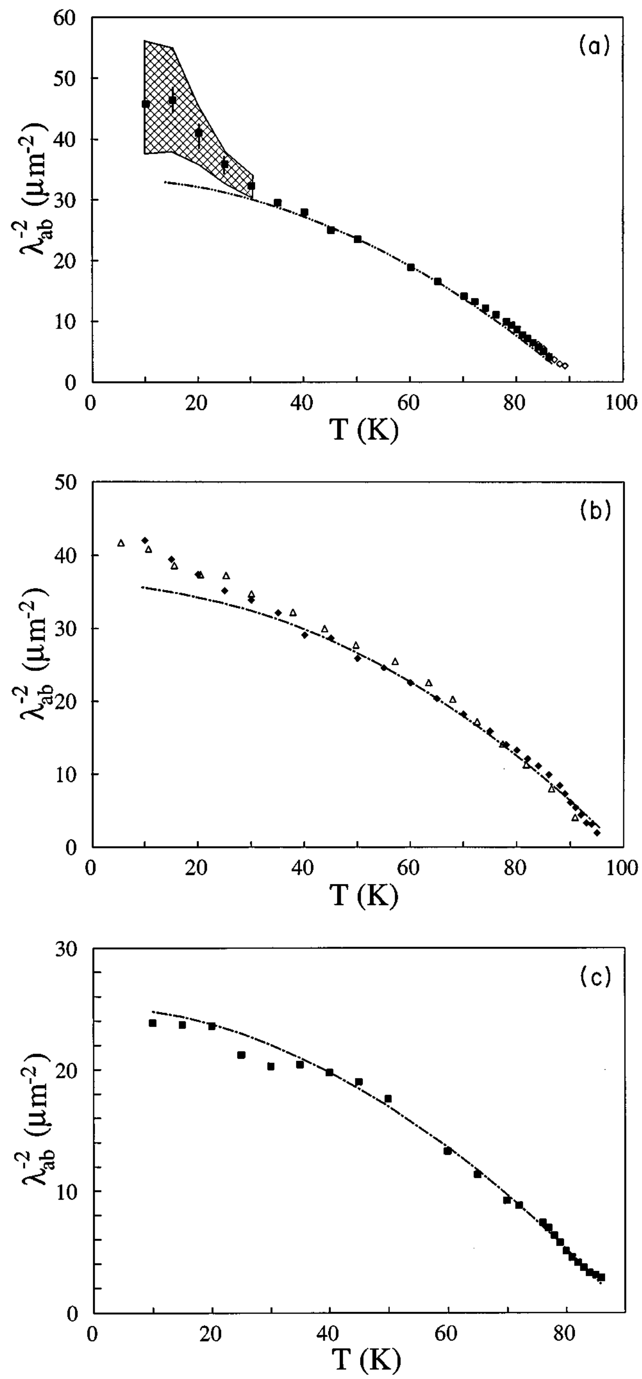

FIG. 2. (a) $1/\lambda^2$ vs T of $HgBa_2CuO_{4+\delta}$ with $\delta=0.28$: ■ London model; ◇ Bulaevskii model; ⊠ uncertainty caused by paramagnetic background; | estimated uncertainty due to the irreversible magnetization; -·- $1-(T/T_c)^2$ fit. (b) $1/\lambda^2$ vs T of the optimally doped sample: ◆ from reversible M; △ converted from μSR data (Refs. 19, 20); -·- $1-(T/T_c)^2$ fit. (c) $1/\lambda^2$ vs T of the underdoped sample: ■ from reversible M; | $(1-t^2)$ fit; -·- $1-(T/T_c)^2$ fit.

Since $\lambda(T)$ is indirectly obtained in this study from $M(H)$ at various T's, there exist several possible sources of uncertainties in the determination of the T dependence of λ. They are effects of phase impurity, doping inhomogeneity, polycrystallinity, irreversible magnetization, and paramagnetic background. Their contributions to the uncertainty of $\lambda(T)$ are assessed to be small and briefly summarized.

Nonmagnetic phase impurity will underestimate M and thus overestimate the absolute value of λ. Within the resolution of x-ray and neutron-power diffraction, our samples displayed a single phase of $HgBa_2CuO_{4+\delta}$,[12] although weight-balance and wet chemistry analyses showed a Hg deficiency. Since the three samples investigated were obtained by annealing similarly synthesized $HgBa_2CuO_{4+\delta}$ under various O_2-partial pressures, the overall systematic error introduced by phase impurity was estimated to be less than 5%. A doping inhomogeneity gives rise to a distribution of compounds with different T_c's and thus can result in a false T-dependent λ. To minimize possible error associated with doping inhomogeneity, we chose samples with a narrow transition width and a T-independent M below T_c as shown in Fig. 1. The estimated error so introduced should be less than 5%. All theoretical models treat single crystalline samples with H parallel to the c axis. The samples examined were polycrystalline. Fortunately, it has been shown that the magnetization (M_s) of a single crystal HTS and that (M_c) of a polycrystalline one uniquely related as $M_s(H)=2M_c(\sqrt{e}H)$ provided the anisotropy γ of the HTS is greater than 5. Our preliminary results on aligned $HgBa_2CuO_{4+\delta}$ powder suggest a $\gamma \geqslant 10$ for the compound. This simple conversion was used for our analysis and should not lead to uncertainties in the T dependence of λ. An inhomogeneous field through which the sample moves in a SQUID magnetometer reduces the accuracy of M and hence λ. It was found to be very small and have an insignificant effect on our M, as evidenced by the negligible M variation detected when the scanning length in the SQUID was changed from 2 to 6 cm.

$HgBa_2CuO_{4+\delta}$ has a relatively high irreversibility line. The (H,T) region of interest to the present study often lies below the irreversibility line. The irreversible contribution to M can therefore be significant. Even for the granular samples of ours, the irreversible contribution to M of the optimally doped sample at 0.5 T was ~10% at 40 K and increased to ~25% at 20 K, although it was smaller for the other two samples. We have therefore determined the relaxation of the magnetization during H-increasing (M_+) and during H-decreasing (M_-) modes. We found that M_+ and M_- relaxed toward their arithmetic average $M \equiv (M_+ + M_-)/2$ at the same rate above 15 K and 1 T within a resolution of $\pm 10\%$ as shown in Fig. 1 (inset). Even if in the surface-pinning dominated cases, the observed $0.9 < (dM_+/d\ \ln t)/(dM_-/d\ \ln t) < 1.1$ will lead to the conclusion that the difference between the reversible M and $(M_+ + M_-)/2$ is 5% or less.[17] Consequently, we have used this averaged value as the reversible M for $\lambda(T)$ analysis and expect little error to be introduced to $\lambda(T)$.

A paramagnetic background M_p was detected in all of our samples above T_c for reasons yet-to-be determined. The paramagnetic background above T_c fits well the Curie-Weiss relation with a low Curie-temperature < 3 K. For an underdoped sample with a $T_c < 4.2$ K, the fit was excellent down to 30 K. However, at lower temperature and high field, M becomes H dependent and fits the Brillouin function $M=a\ \tanh(bH/T)$ where a and b are experimentally determined constants. The corrections on M below T_c were made by subtracting from the measured M the paramagnetic contribution extrapolated from the Curie-Weiss and Brillouin-function fits at different temperatures and fields. The estimated uncertainty associated with the paramagnetic background appears to be the largest among all possible sources listed above, and is shown in Fig. 2(a) as the crossed region, for example.

As a further check, we have compared the values of $1/\lambda^2$, so obtained with those determined by μSR,[18] by using a conversion factor of $(1/\lambda^2)/\sigma = 11\ \mu s\ \mu m^{-2}$,[19] where σ is the depolarization rate in μSR measurements. As shown in Fig. 2(b) for the optimally doped sample, the T dependences of the two sets of data agree excellently. The agreement suggests that our method used to determine $\lambda(T)$ in the present study is unlikely to introduce large artifact to the T dependence of λ.

It is clear from Fig. 2 that $1/\lambda^2(T)$ varies with T following a power law of $1-(T/T_c)^2$ within our experimental uncertainty for the underdoped sample below T_c. However, the observed data deviates from the square fit significantly below $T_c/2$. The difference seems to be larger than the possible uncertainties. A linear T dependence is clear below $T_c/2$ for both the optimally doped and overdoped samples. Unfortunately, the impurity scattering in the d-wave superconductors and the proximity effect may not be able to account for the observed change in the T dependence of $1/\lambda^2(T)$. All three samples were obtained by annealing the same as-synthesized $HgBa_2CuO_{4+\delta}$ at ~300 °C although at different oxygen partial pressures. Therefore, they should have similar structural imperfections except those induced by different δ's which increases by a factor of approximately 2 from the underdoped to the overdoped samples. If one regards the randomly added oxygen as scattering centers, the change of α from 2 to 1 with δ increase contradicts the prediction of the defect models of the d-wave superconductors. The observation suggests that the carrier concentration can strongly affect the T dependence of $1/\lambda^2$ in addition to impurity scattering. Similar observations have been made in μSR investigations before.[20] The interpretation of the T dependence of λ in terms of the superconducting pairing symmetry should be taken with extreme care.

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