Effects of carrier concentration on the superfluid density of high-T_c cuprates

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The absolute values and temperature, T, dependence of the in-plane magnetic penetration depth of $La_{2-x}Sr_xCuO_4$ and $HgBa_2CuO_{4+\delta}$ have been measured as a function of carrier concentration. We find that the superfluid density ρ_s changes substantially and systematically with doping. The values of $\rho_s(0)$ are closely linked to the available low-energy spectral weight as determined by the electronic entropy just above T_c, and the magnitude of the initial slope of $[\rho_s(T)/\rho_s(0)]$ increases rapidly with carrier concentration. The results are discussed in the context of a possible relationship between ρ_s and the normal-state (or pseudo)energy gap. [S0163-1829(99)04546-4]

Superconductivity arises from the binding of electrons into Cooper pairs, thereby forming a superfluid with a superconducting energy gap Δ in the single-particle excitation spectrum. In high-temperature superconductors (HTS's), Δ has essentially $d_{x^2-y^2}$ symmetry in k space with $\Delta_k = \Delta_0 \cos 2\phi$,[1] where $\phi = \arctan(k_y/k_x)$ and Δ_0 is the superconducting gap amplitude which will in general be ϕ dependent. Changes in carrier concentration have unusually strong effects on the superconducting[2–8] and normal-state[4–7] properties of HTS's. There is evidence[3–7] that in addition to the superconducting gap Δ_k there is a normal-state (or pseudo) gap Δ_N in the normal-state energy excitation spectrum in underdoped and optimally doped samples and that this decreases as the doping is increased. The maximum superconducting gap amplitude Δ_0 seems to show little variation with underdoping even though T_c is reduced,[3–8] in disagreement with the standard mean-field Bardeen-Cooper-Schrieffer (BCS) theory. This unusual behavior is probably linked in some way to the presence of Δ_N.[7] However, fundamental problems such as the origin of Δ_N and its possible effect on the superfluid density ρ_s have not been clearly resolved.

The physical quantity most directly associated with ρ_s is the magnetic penetration depth λ because in the London model $1/\lambda^2 \propto \rho_s$. The materials studied here, $La_{2-x}Sr_xCuO_4$ and $HgBa_2CuO_{4+\delta}$, are particularly appropriate systems to investigate ρ_s as a function of doping. Both have a simple crystal structure with one CuO_2 plane per unit cell, can have their carrier concentration controlled, and there is experimental evidence suggesting the presence of a normal-state gap Δ_N which closes with increasing doping.[3–5,9] Here we report in-plane penetration depth, λ_{ab}, measurements for high-quality $La_{2-x}Sr_xCuO_4$ (LSCO) with $x = 0.10$, 0.15, 0.20, 0.22, 0.24 measured by the ac-susceptibility (acs) and muon spin relaxation (μSR) techniques and for $HgBa_2CuO_{4+\delta}$ (Hg-1201) with $\delta = 0.10$, 0.37 measured only by μSR. We find systematic changes in ρ_s with carrier concentration and a correlation with Δ_N.

Single-phase polycrystalline samples of LSCO were prepared in Cambridge using solid-state reaction procedures. No other phases were detected by powder x-ray diffraction and the phase purity is thought to be better than 1%. Lattice parameters were in good agreement with published work.[10] High-field magnetic susceptibility measurements showed no signatures of excess paramagnetic centers. The measured T_c's are 30, 37.7, 36, 27.5, and 20.3 K for $x = 0.10$, 0.15, 0.20, 0.22, and 0.24, respectively. These values are also in very good agreement with previous measurements.[10] μSR experiments as a function of T were performed on the same powders for $x = 0.10$ and 0.15. Although unoriented powders

can be used to determine λ_{ab} by μSR,[2] the acs technique requires the powders to be magnetically aligned.[11] Grain agglomerates can be a cause of poor degree of alignment, and to eliminate these, powders were ball milled in ethanol and dried after adding a defloculant. Scanning electron microscopy confirmed the absence of grain boundaries and showed that the grains were approximately spherical with average grain diameter ~ 5 μm. The powders were mixed with a 5-min curing epoxy and aligned in a static field of 12 T at room temperature. Debye-Scherrer x-ray scans showed that $\sim 90\%$ of the grains had their CuO_2 planes aligned to within $\sim 2.0°$. Low-field susceptibility measurements were performed down to 1.2 K using an ac field $H_{ac} = 1$ G rms (parallel to the c axis) and a frequency $f = 333$ Hz. Details of the application of London's equations for deriving λ from the measured low-field ac susceptibility in HTS's can be found in an earlier publication and references therein.[11] Transverse-field-cooled μSR experiments were performed at the ISIS, Rutherford-Appleton Laboratory on unaligned powders in a field of 400 G. The field produced a flux-line lattice whose field distribution was probed by muons. The depolarization rate $\sigma(T)$ of the initial muon spin is proportional to $1/\lambda_{ab}^2(T)$ [i.e., $\sigma(\mu s^{-1}) = 7.086 \times 10^4 \times \lambda_{ab}^{-2}$ (nm)].[2,12] Checks were made to ensure that the values of λ_{ab} obtained were independent of the applied field and the σ values used to estimate $1/\lambda^2$ had the mean high-temperature value of $\sigma(T > T_c)$ subtracted. The Hg-1201 [$\delta = 0.10$ ($T_c = 60$ K) and 0.37 ($T_c = 35$ K)] samples were prepared in Houston by the controlled solid-vapor reaction technique.[13]

The values of $\lambda_{ab}(0)$ derived from the acs data for LSCO are 0.28, 0.26, 0.197, 0.193, and 0.194 μm for $x = p = 0.10$, 0.15, 0.20, 0.22, and 0.24, respectively, where p is the hole content per planar copper atom. The maximum error for $\lambda_{ab}(0)$ of the acs technique is less than $\pm 15\%$. However, given that all our samples were prepared under the same conditions, the size and shape of the grains were the same for all Sr concentrations, and the $\lambda_{ab}(0)$ values measured by the acs and μSR techniques are in excellent agreement [Fig. 1(a)], we believe the actual error is significantly lower than the above estimate. $\lambda_{ab}^{-2}(0)$ was also measured for Sr $= 0.22$ using μSR and agrees with the acs value. We thus find that $\lambda_{ab}^{-2}(0)$ is suppressed on the underdoped side, including optimal doping, but there is no suppression with increasing overdoping (up to $p = 0.24$) in contrast to reports for $Tl_2Ba_2CuO_{6+\delta}$ (Tl-2201) Ref. 14 and for $Y_{0.8}Ca_{0.2}Ba_2Cu_3O_{7-\delta}$ (Y:Ca-123).[2] Values of $\lambda_{ab}(0)$ as obtained by μSR for Hg-1201 are 0.194 and 0.148 μm for $\delta = 0.10$ and 0.37, respectively. We note that $\delta = 0.10$ and 0.37 in Hg-1201 correspond to $p = 0.075$ and 0.22, respectively.[15]

The T dependence of λ_{ab} for LSCO is shown in Fig. 1(a) as a plot of $[1/\lambda_{ab}(T)]^2 \propto \rho_s(T)$. $\lambda_{ab}^{-2}(T)$ data for $x = 0.10$ and 0.15 obtained by μSR are also included for comparison. Overall there is good agreement between the results from the two techniques. From the acs data we find that the existence of an initial linear term in $\lambda_{ab}(T)$, characteristic of a clean d-wave superconductor, persists up to the highest doping measured ($x = 0.24$), in agreement with electronic specific heat studies on polycrystalline LSCO samples from the same batch as those studied here.[16] Figure 1(b) depicts data for

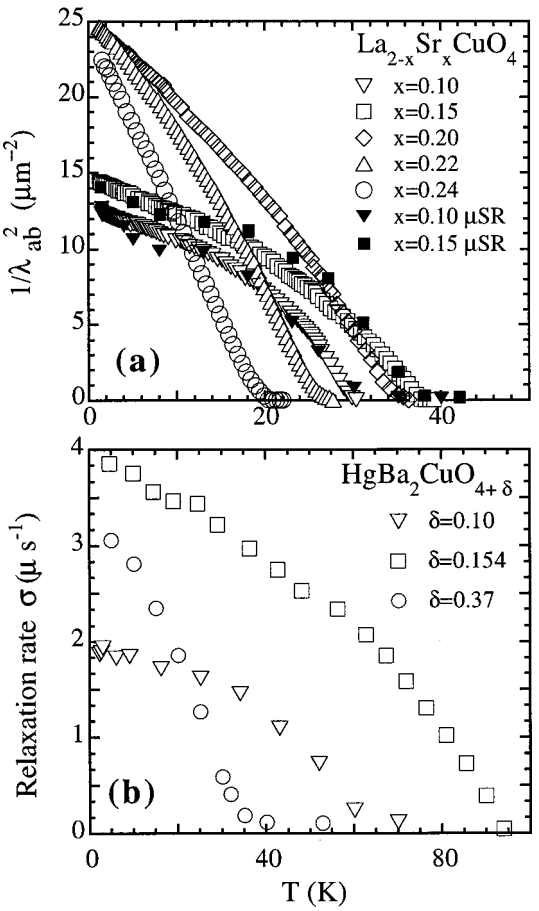

FIG. 1. (a) $\lambda_{ab}^{-2}(T)$ obtained by the ac-susceptibility technique for grain-aligned $La_{2-x}Sr_xCuO_4$ (LSCO). Data obtained by μSR for unoriented LSCO powders are also included (solid symbols). (b) $\sigma(T) \propto \lambda_{ab}^{-2}(T)$, for $HgBa_2CuO_{4+\delta}$ unoriented powders. The data for $\delta = 0.154$ are taken from Ref. 17.

Hg-1201 powders measured only by μSR, including data from Ref. 17 for a Hg-1201 sample (also from Houston) with $\delta = 0.154$ ($p = 0.17$). As in LSCO, we observe a change in the shape of $\sigma(T) \propto [1/\lambda_{ab}(T)]^2$ of Hg-1201 with doping and a suppression in $\lambda_{ab}^{-2}(0)$ with underdoping. In the underdoped region $[1/\lambda_{ab}(T)]^2$ shows a more pronounced downward curvature. Taking the magnitude of the slope of the low-T linear term to be proportional to $\rho_s(0)/\Delta_0$ (Ref. 8), the observed trend of $[1/\lambda_{ab}(T)]^2$ with p would imply that Δ_0 remains approximately constant in the underdoped region and then decreases rapidly with overdoping.

Figure 2 shows a comparison of the present results for LSCO with specific heat data taken on the same samples[16] where a finite Δ_N was observed for $x = p < 0.19$. In the inset we observe a good correlation between the x dependence of $[1/\lambda_{ab}(0)]^2$ and $[S/T(T_c) - S/T(2K)]$ where $S(T)$ is the electronic entropy obtained by integrating the electronic specific heat coefficient $\gamma(T) \equiv C_{el}/T$ from 0 to T. The quantity $[S/T(T_c) - S/T(2\,\mathrm{K})]$ is a measure of the energy-dependent normal-state electronic density of states (DOS), $g_n(E)$, averaged over $\pm 2k_BT_c$ around the Fermi energy E_F. The effect of an energy-dependent DOS on the London penetration depth λ_L, or $\rho_s(0)$, is not usually considered in the standard theory, which implicitly assumes a constant DOS and a parabolic $E(\mathbf{k})$ dispersion relation. It has been argued elsewhere[5]

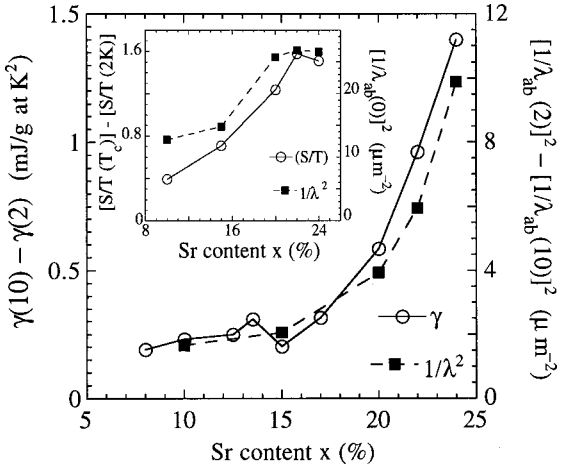

FIG. 2. Low-$T\lambda_{ab}^{-2}(T)$ for $La_{2-x}Sr_xCuO_4$ (LSCO) versus x compared with the low-T specific heat coefficient γ (Ref. 16). Inset: $\lambda_{ab}^{-2}(0)$ for LSCO compared with $[S/T(T_c)-S/T(2\,\mathrm{K})]$ (Ref. 16).

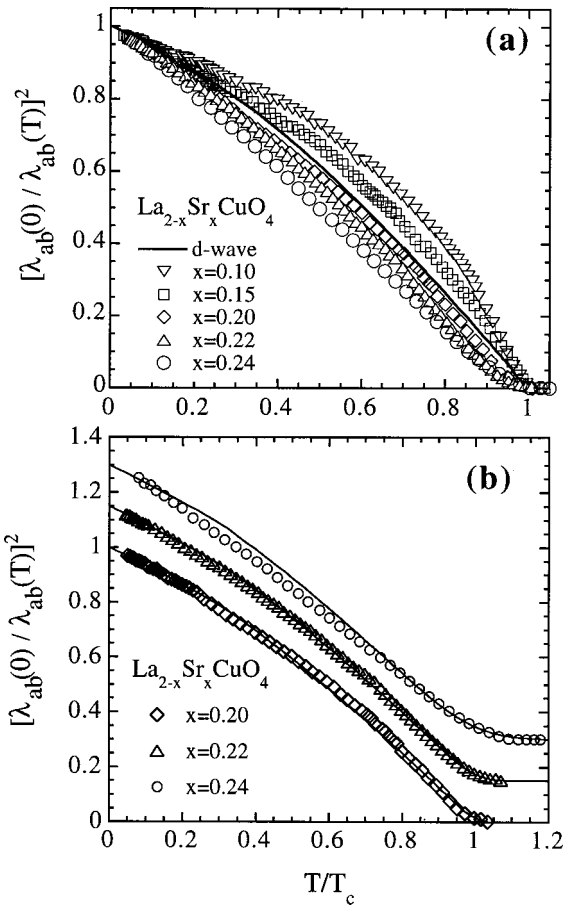

FIG. 3. (a) $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ obtained by the ac-susceptibility technique for grain-aligned $La_{2-x}Sr_xCuO_4$ compared with the weak-coupling BCS theory (solid line) for a d-wave superconductor (Ref. 19). (b) The $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ data for $x=0.20$, 0.22, and 0.24 shown in panel (a), but corrected for a distribution of T_c values (see text for details). The solid lines are the BCS d-wave T dependence shown in panel (a) corrected for the respective distribution in T_c's. The curves for $x=0.22$ and 0.24 are shifted vertically for clarity.

that $\rho_s(0)=4\pi^2\langle v_x^2 g_n(E)\rangle/e^2$, where v_x is the Fermi velocity and the average is taken over an (anisotropic) energy shell, $E_F\pm\Delta_0$. Note that this result agrees with the standard expression for the normal-state conductivity and the usual relation between $\lambda_L(0)$ and the real part of the frequency-dependent electronic conductivity in the normal and superconducting states $\sigma_1^n(\omega)$ and $\sigma_1^s(\omega)$, respectively. Namely, $\rho_s(0)\propto\lambda_L^{-2}(0)$ (Ref. 18) is determined by the area under the $[\sigma_1^n(\omega)-\sigma_1^s(\omega)]$ curve in the frequency range $0<h\omega/2\pi<2\Delta_0$. Thus the inset to Fig. 2 suggests that the strong decrease of $\rho_s(0)$ with x from $x=0.20$ to 0.10 is related to the suppression of spectral weight with energy range $E_F\pm\Delta_0$, which is believed to be due to the presence of the normal-state gap.[4] Conversely, the fact that $\rho_s(0)$ for LSCO does not fall on the overdoped side as in Tl-2201 (Ref. 14) and Y:Ca-123 (Ref. 2) may well be associated with the observed occurrence of a low-energy peak in the DOS, unique to LSCO,[16] which grows with overdoping.

The main panel in Fig. 2 shows a correlation between the doping dependence of the slopes of $\lambda_{ab}^{-2}(T)$ and $\gamma(T)$ both quantities being related to the number of excited quasiparticles, $n_e(T)$. For low values of x, $n_e(T=10\,\mathrm{K})$ is much smaller than expected from the T_c value, and this probably implies that the average value of $\Delta_0(\phi)$ is significantly larger than T_c. The rapid rise above $x=0.20$ may arise from the combined effects of the closure of Δ_N at $x=0.19$ (Refs. 4 and 16) and the decreasing T_c values, plus the fact that for LSCO there is significant pileup of states near E_F in the overdoped region $0.20<x<0.35$.[16]

In Fig. 3(a) we present the LSCO acs data as $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ versus T/T_c and compare the data with the mean-field calculation for a d-wave weak-coupled BCS superconductor with a cylindrical Fermi surface which gives $\Delta_0/T_c\sim 2.14$.[19] There appears to be a systematic deviation of the data from the weak-coupling T dependence with a greater (weaker) curvature on the underdoped (overdoped) side. The observed trend in $\lambda_{ab}^{-2}(T)$ with underdoping is in agreement with theoretical predictions[20] based on a pseudogap scenario. In the overdoped samples in Fig. 3(a), there is a positive curvature near T_c, which may arise from a small amount of doping inhomogeneity, giving a distribution of T_c values in this region where dT_c/dp is maximal.[10] The effect of this is to rescale the curves with a slightly lower value of T_c. We have modeled $\rho_s(T)$ using the weak-coupled BCS d-wave T dependence for a cylindrical Fermi surface and a normal distribution of T_c values with standard deviation of 3%, 5%, and 9% for $x=0.20$, 0.22, and 0.24, respectively. The resultant curves in Fig. 3(b) (solid lines) are in excellent agreement with the experimental data especially for $x=0.20$ and 0.22, implying that the classic d-wave T dependence is preserved in the moderately overdoped region. However, the $x=0.24$ sample still shows significant deviations that possibly reflect changes in the electronic structure. This would not be surprising given the changes in the Fermi surface with the rapid crossover from holelike to electronlike states near $x=0.27$.[21] We note that the data for $x=0.24$ are in excellent agreement with a weak-coupling d-wave calculation for a rectangular Fermi surface.[22]

In contrast to the overdoped samples, the optimal and underdoped samples both show very small rounding near T_c. The data depart significantly from the weak-coupling curve

and in the opposite direction. We note that accounting for inhomogeneities in these samples will, if anything, move the curves even further from the weak-coupling BCS fit.

A central conclusion of the present work is that there is a crossover in both $\rho_s(0)$ and $\rho_s(T)$ near $p=0.20$. Such behavior is characteristic of many other normal-state and superconducting properties which have been interpreted in terms of the presence of Δ_N for $p<0.20$.[5,23–26] The proper means of incorporating the normal-state gap effects within a realistic model and, indeed, the very nature of the normal-state gap are a matter of current debate. However, a key characteristic of Δ_N is the loss of normal-state spectral weight near E_F. As discussed above, the loss of spectral weight can cause both a strong reduction in $\rho_s(0)$ and, in a simple model, enhanced curvature in $[\rho_s(T)/\rho_s(0)]$ relative to the BCS weak-coupling *d*-wave T dependence,[27] the very features we observe for the optimal and underdoped samples.

We note that the curvatures in $\rho_s(T)$ in our data are in reasonable agreement with earlier reports for slightly underdoped, grain-aligned $HgBa_2Ca_2Cu_3O_{8+\delta}$ (Refs. 11 and 28) and single-crystal LSCO with $x=0.15$.[29] In contrast to the strong p dependence we have found in $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ for LSCO and Hg-1201, studies in $YBa_2Cu_3O_{7-\delta}$ (YBCO) (Refs. 30 and 31) showed that $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ scaled approximately with T/T_c for various dopings, at all temperatures. However, systematic changes in $[\lambda_{ab}(0)/\lambda_{ab}(T)]^2$ with p were noted at the time,[31] although these were too small to allow further analysis. This may simply be due to the fact that the YBCO samples were not as heavily underdoped as the $x=0.10$ LSCO sample. We also note that YBCO is complicated by a mixed $s+d$ order parameter[1,32] and the effect of the Cu-O chains on the total superfluid density ρ_s.[33,34] Furthermore, the high sensitivity of λ measurements on sample quality is one of the main reasons for the incomplete and inconclusive data available from earlier measurements.[35]

In summary, using the acs and μSR techniques, we have obtained consistent and systematic results on the effects of carrier concentration on ρ_s of monolayer cuprates. In the overdoped region we find a reasonably constant value of $\rho_s(0)$ (up to $p=0.24$), and $[\rho_s(T)/\rho_s(0)]$ is in good agreement with the weak-coupling *d*-wave T dependence. In the optimal and underdoped regions $\rho_s(0)$ is rapidly suppressed, and above $0.1T_c$ there is a marked departure of $[\rho_s(T)/\rho_s(0)]$ from the weak-coupling curve. Our data are entirely consistent with available specific heat data, which give evidence for a link between the behavior of ρ_s and the normal-state gap Δ_N.

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