Comparative study of the characteristic length scales and fields of Hg-based high-T_c superconductors

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The equilibrium magnetization was studied in magnetically aligned Hg-based cuprate superconductors with a crystal structure containing one, two, and three adjacent Cu-O layers. The mixed state magnetization was measured with the magnetic field applied perpendicular to the layers, along with low field Meissner effect and normal state susceptibility studies for volume correction factors. The internally consistent analysis is used to identify systematics as the number ''n'' of Cu-O layers increases. The study reveals a linear increase in the slope of upper critical field $-dH_{c2}/dT$ with ''n''. Most importantly, we find a nearly constant carrier doping per Cu-O layer. This result is compared with other studies, e.g., Rietveld analysis of neutron diffractometry, and its implications are discussed. [S0163-1829(96)04534-1]

INTRODUCTION

A ubiquitous feature of high-T_c superconductors is the presence of copper-oxygen layers in the material crystal structure. These sheets stack with one, two, three, or even more adjacent layers to form a set of planes. Between the plane sets are separator or ''blocking'' layers of Hg-O, Bi-O, or Tl-O, for example, which complete the structure and provide charge carriers for the conductive Cu-O planar sets. This stacking of building blocks gives existence to a homologous series of structures, i.e., families of superconductors.

In this work, we make a systematic study of the Hg-based cuprate family containing one, two, or three adjacent Cu-O layers per set. Studies of the equilibrium magnetic properties give internally consistent measures of the superconducting length scales: the coherence length ξ and the London penetration depth λ. We use a convenient available theoretical method—the formalism of Hao-Clem *et al.*[1]—to analyze experimental studies of magnetically aligned powders, dispersed in epoxy. In all cases, we consider the well defined and conceptually simple configuration with the magnetic field applied perpendicular to the Cu-O planes; we avoid too the additional complexity of angular averages associated with random polycrystalline materials. With a tetragonal crystal structure and comparatively stable oxygen stoichiometry, the Hg-based cuprate superconductors constitute a nearly ideal case. Hence we have made a systematic study of the first three members, to determine what trends may exist in their characteristic length and magnetic field scales.

The first of the Hg-based cuprates, $HgBa_2CuO_{4+\delta}$ (Hg-1201) possesses just one Cu-oxide layer per unit cell and exhibits a remarkably high transition temperature T_c near 95 K.[2] Hg-1201 is the first member ($n=1$) of the homologous series of $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$, similar to the mono-Tl-O layered series $TlBa_2Ca_{n-1}Cu_nO_{2n+3}$. The $n=2$ member of these Hg compounds was first formed as part of a nonsuperconductive multiphase mixture[3] and later the superconducting compound $HgBa_2CaCu_2O_{6+\delta}$ was synthesized.[4-6] Hereafter we denote the two layer Hg-Ba-Ca-Cu oxide as Hg-1212. The $n=3$ member, $HgBa_2Ca_2Cu_3O_{8+\delta}$(Hg-1223), was first synthesized by Schilling *et al.*[7] and found to have the unprecedentedly high T_c of 133 K. Structurally, all of these materials have conceptually simple tetragonal crystallography. A useful set of idealized perspective drawings of these structures, as well as those of many other high-T_c materials, has been compiled by Shaked *et al.*[8]

EXPERIMENTAL ASPECTS

Bulk samples of Hg-based high-T_c cuprates were prepared by solid-state reaction from stoichiometric mixtures of 99.998% HgO, 99.997% BaO, 99.97% CaO, and 99.999% CuO, as described previously.[9] The resulting polycrystalline materials were checked for structure by x-ray diffraction and their superconductive properties were precharacterized using dc magnetic methods. We then formed aligned composite samples by grinding the materials to monocrystalline powders (~ 7 μm diameter), dispersing the powder in ''45 min'' liquid epoxy, and magnetically aligning[10] the particles in a 5 T field. Typically the volume fraction of superconductor in the epoxy matrix was $\sim$10%. To quantify the alignment of the particles, we performed x-ray diffraction with the scat-

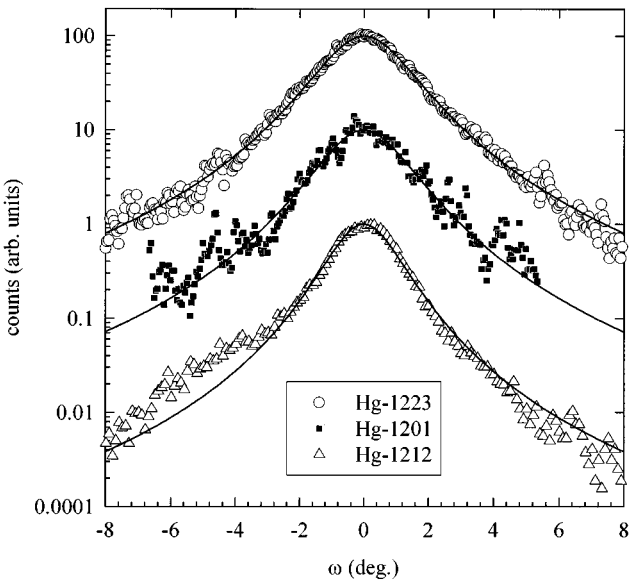

FIG. 1. Rocking curves: x-ray count rate (arbitrary logarithmic scale) versus rocking angle for the (006) reflections (Cu $K\alpha$ radiation) from three magnetically aligned samples of Hg-cuprate superconductor. Lines are fits to the expression $I=I_0(1+\omega^2/\Omega^2)^{-3/2}$.

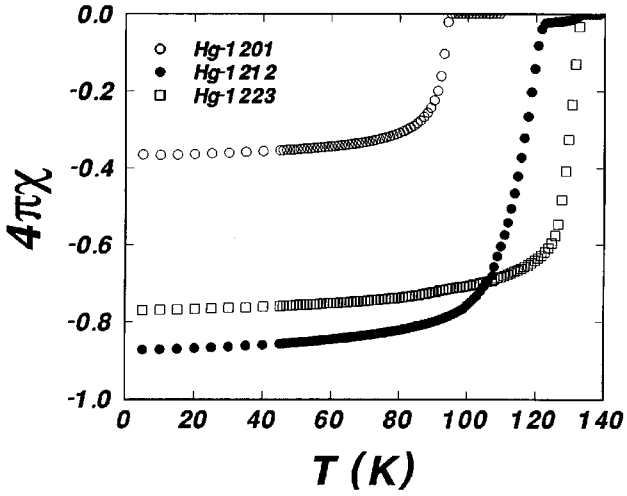

FIG. 2. The Meissner state diamagnetic susceptibility $4\pi\chi$ versus temperature T for the three samples shown.

tering vector parallel to the direction of the alignment field. The diffractograms had strong $(00l)$ reflections, but (hkl) reflections with nonzero indices h or k were immeasurably small. Figure 1 shows the rocking curves for the (006) lines of each sample. For these omega scans, a Rigaku rotating anode x-ray generator was used with a graphite monochromator to select Cu K_α radiation. It is evident that the angular alignment is good, with full widths at half maximum (FWHM) of 2.4°, 1.9°, and 2.5° for the Hg-1201, Hg-1212, and Hg-1223 materials, respectively. Previously we reported[11] on some equilibrium properties of aligned Hg-1223; the present results differ only slightly (mainly due to revised background and volume corrections) and are included to facilitate intercomparison of the three materials.

A SQUID-based magnetometer (Quantum Design model MPMS-7) was used to measure the magnetic properties of the materials, both in the low field Meissner state and in the vortex state above the lower critical field H_{c1}. In all cases, the magnetic field H was applied perpendicular to the Cu-O planes, i.e., $H\|c$ axis of the composite sample. Scan lengths of 3 cm were used to maintain a field uniformity of $<0.005\%$ in the 7 T magnet during measurement. In low field studies ($H_{\rm appl}=4$ G), we measured the magnetic shielding signal (zero field cooled=ZFC) and Meissner signal (field cooled=FC) as a function of temperature. To permit application of a precisely known field, the superconductive magnet in the magnetometer was ''reset'' by heating it above its T_c of $\sim$9 K prior to inserting the sample. The superconducting magnetization $M(T,H)$ is defined as the magnetic moment m per unit of volume of superconductor. Unfortunately, determining the superconducting volume fraction for high-T_c materials is notoriously difficult and is discussed in a later section. To proceed, we take for the volume $V_{\rm tot}=({\rm mass}/\rho_{\rm x\ ray})$ the mass of Hg-Ba-Ca-Cu oxide divided by the x-ray density of the respective superconductor. Then the experimental Meissner susceptibility is $\chi=m/(V_{\rm tot}H_{\rm eff})$, where $H_{\rm eff}=(H_{\rm appl}-4\pi DM)$ is the effective magnetic field and $D\approx 1/3$ is the effective demagnetizing factor for the roughly equiaxed particles. The results for the FC (Meissner) study are shown in Fig. 2, a plot of $4\pi\chi(T)$ versus temperature for the three materials. Generally the ZFC shielding susceptibility (not shown for clarity) was only slightly larger in magnitude, as is typically the case for small, isolated monocrystalline particles.

Since the Meissner state susceptibility of an ideal superconducting body is $-1/4\pi$, the quantity $-4\pi\chi(T\ll T_c)$ gives an estimate for the volume fraction $V_{\rm sc}/V_{\rm tot}$ of superconductor. Values at $T=5$ K are given in Table I for each material. These values should be a lower bound on the true volume fraction, as both vortex pinning and small particle size effects[12] reduce the magnitude of expelled flux.

In the mixed state, measurements of the isothermal magnetization M were made for a set of temperatures T between 5 K and T_c. To obtain virgin magnetization curves, trapped flux was released from the sample by heating it above T_c and then recooling it in zero applied field. Prior to applying the magnetic field H, the temperature was stabilized to within ± 0.05 K of the target temperature. With the temperature stabilized, the magnetic moment m was measured for fields H in the range 0.01–6.5–0.01 T. A 10 s pause after each field change allowed rapid instrumental transients to decay.

To correct for magnetic background effects, the normal state magnetic susceptibility was measured for temperatures from 250 K down to the fluctuation region near T_c. A Curie-like behavior was found, with $m_{bkg}=H(\chi_0+C'/T)$; note that the coefficients in this expression refer to the magnetic moment and not the moment per volume. This dependence was extrapolated to temperatures below T_c and subtracted from the measured magnetic moment, in order to isolate the superconducting signal. Then the equilibrium magnetization was determined as the mean value between field-increasing and field-decreasing magnetic moment, divided by $V_{\rm tot}$. For nearly all of the results reported here, the measurements were conducted in the magnetically reversible region above the irreversibility line and these values coincided. Parenthetically, we note a potential pitfall with the averaging procedure that is related to surface barrier effects, which are fre-

TABLE I. Equilibrium properties of the high-T_c materials $HgBa_2CuO_4$ (Hg-1201), $HgBa_2CaCu_2O_6$ (Hg-1212), and $HgBa_2Ca_2Cu_3O_8$ (Hg-1223), with magnetic field $H\|c$ axis.

Material	Hg-1201	Hg-1212	Hg-1223
T_c (K) [low field]	95.1	122.5	133.5
$-4\pi\chi$=Meissner fraction$\approx V_{sc}/V_{tot}$	0.36	0.87	0.77
$[V_{sc}/V_{tot}]$ from Curie background	0.41	0.94	0.95
Values from Hao-Clem analysis			
$-dH_{c2}/dT$ [T/K]	1.2	1.6	2.0
$H_{c2}(T=0)$ [T] from WHH theory	80	135	190
$\xi_{ab}(T=0)$ [nm]	2.0	1.6	1.3
$\kappa=\lambda_{ab}/\xi_{ab}$, near T_c	130	85	120
$H_c(T=0)$ [T]	0.38	0.96	1.03
Values rescaled for V_{sc}/V_{tot}			
$\kappa_{rescaled}=\kappa\times(V_{sc}/V_{tot})^{1/2}$	78	79	103
$H_{c,rescaled}=H_c/(V_{sc}/V_{tot})^{1/2}$ [T]	0.63	1.03	1.17
$\lambda_{ab}(T=0)=\phi_0/2^{3/2}\pi H_{c,rescaled}(0)\xi_{ab}(0)$ [nm]	180	145	155

quently observed in Hg-based superconductors.[13–15] With a surface barrier, the thermally activated decay of M is much more rapid in the field-increasing branch. Consequently the mean value of substantially hysteretic signals tends to underestimate the magnitude of the equilibrium magnetization—and the severity of the effect may be field dependent. Measurements of the hysteresis loops, with H first increasing and then decreasing, made it easy to recognize regions of magnetic irreversibility at lower fields and temperatures.

One can also extract complementary estimates for the superconducting volume fraction from the magnetic background measurements, specifically the ''Curie constant'' C'. This temperature dependent component arises mostly from paramagnetic Cu ions, since closed shell ions such as Ba^{+2} or Ca^{+2} have small diamagnetic moments that give a temperature independent susceptibility. Then the total number of Cu ions is $N_{tot}=(N_{sc}+N_{para})$, where N_{para} is the number of Cu ions in insulating, paramagnetic phases such as barium cuprates. For a Curie law susceptibility, we have $C'=N_{para}\mu_B^2 p^2/3k_B$, where μ_B is the Bohr magneton, $p\approx1.73$ is the effective moment of an isolated $3d^9$ Cu^{+2} ion, and k_B is Boltzmann's constant. Combining these expressions, we have that $[1-3k_BC'/N_{tot}\mu_B^2p^2]=N_{sc}/N_{tot}=V_{sc}/V_{tot}$. From this procedure, an independent set of estimates for the superconducting volume fraction was obtained. The values are shown in Table I. This modeling assumes that the normal state susceptibility of the superconducting phase *per se* is nominally temperature independent, which is plausible for a system with extended ''d'' states and is observed experimentally.

EXPERIMENTAL RESULTS

To obtain information on the microscopic superconducting properties of the three Hg cuprates, we use the formulation of Hao-Clem to analyze the reversible magnetization M. The authors[1] have described in detail the basis for this theory. It accounts explicitly for the core energy of vortices, through approximate analytical expressions for their structure, and provides values for the Ginzburg-Landau parameter $\kappa(T)=\lambda(T)/\xi(T)$ and the thermodynamic critical field $H_c(T)$. In practice, the formalism is implemented using a self-consistency criterion: one analyzes the isothermal $M(H)$ at various fields (here $H=1$, 2, 3, 4, and 5 T) and finds a value for κ such that $H_c(T)$ is the same (or nearly so) at each field. Then the process is repeated at other fixed temperatures; our methodology has been reported previously.[16] The values of $\kappa(T)$ for the three samples with $H\|c$ axis are shown in Fig. 3 as a function of reduced temperature $t=T/T_c$. The results for the range $t\leqslant0.85$ are well behaved, with κ being nearly independent of temperature. For $t>0.85$, fluctuation effects cause the extracted values to increase rapidly with temperature, with qualitatively similar behavior for the three samples. As fluctuations are not incorporated in the Hao-Clem formulation, the data near T_c are excluded from the subsequent analysis.

To reduce experimental noise, we average κ over its temperature-independent interval $t<0.8$–0.85. We then recompute $H_c(T)$ at each temperature, for each sample. These resulting equilibrium parameters provide much useful information. For example, the upper critical magnetic field is $H_{c2}(T)=\sqrt{2}\kappa H_c(T)$ in Ginzburg-Landau theory. Figure 4 shows results for H_{c2} versus temperature that are reasonably

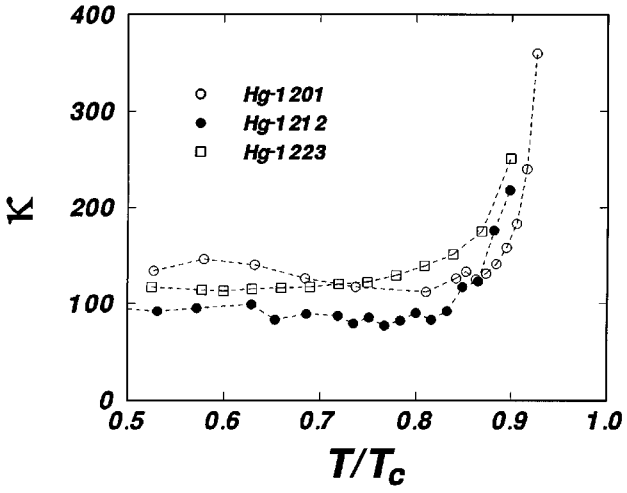

FIG. 3. Temperature dependence of the Ginzburg-Landau parameter κ, as obtained by analyzing the reversible magnetization using the Hao-Clem formulation; see text.

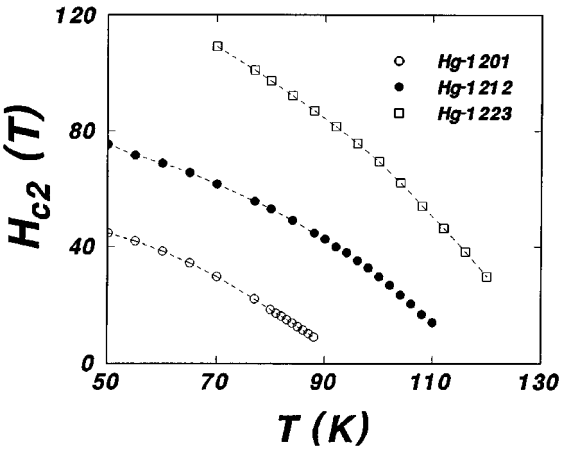

FIG. 4. The upper critical magnetic field H_{c2} versus temperature, for $HgBa_2CuO_4$, $HgBa_2CaCu_2O_6$, and $HgBa_2Ca_2Cu_3O_8$ with $H \| c$ axis.

well behaved: (1) as T decreases from T_c, H_{c2} varies approximately linearly with T and (2) it continues a smooth, monotonic increase down to the lowest temperatures where the materials were magnetically reversible. For three-dimensional (3D) materials, Werthamer, Helfand, and Hohenberg[17] (WHH) have shown that the upper critical field $H_{c2}(0)$ extrapolated to $T=0$ is proportional to the slope dH_{c2}/dT near T_c, with

$$H_{c2}(0) = 0.7 T_c [dH_{c2}/dT]_{\approx T_c}. \tag{1}$$

Table I contains values extrapolated to $T=0$, using this expression. Corresponding values for the coherence length at zero temperature, ξ_{ab}, were obtained from the standard Ginzburg-Landau (GL) expression $H_{c2}(0) = \phi_0/2\pi\xi_{ab}^2$ and are shown in Table I also. Finally, it contains values for the thermodynamic critical field H_c extrapolated to $T=0$. These nominal values were obtained by fitting the BCS expression of Clem[18]

$$\frac{H_c}{H_c(0)} = 1.7367\left(1-\frac{T}{T_c}\right)\left[1-0.2730\left(1-\frac{T}{T_c}\right) - 0.0949\left(1-\frac{T}{T_c}\right)^2\right] \tag{2}$$

which is valid for $t = T/T_c > 0.7$.

Let us now account for the influence of an incomplete volume fraction of superconductor, $f = V_{sc}/V_{tot}$. Conceptually it is simplest to consider the total condensation energy E of the sample, since the experimental measurement is performed on the total sample, no matter how it is partitioned. So far, all results have been based on the total volume V_{tot}, as obtained from the sample mass. Then one has $E \sim (H_c)^2 V_{tot}$ with the value H_c for the critical field. If the true, smaller volume of superconductor is V_{sc}, then the energy is $E \sim (H_{c,\text{rescaled}})^2 V_{sc}$ with a rescaled, larger critical field $H_{c,\text{rescaled}}$. Since the energy (and the sample) are the same in each case, we have $H_{c,\text{rescaled}} = H_c/(V_{sc}/V_{tot})^{1/2} = H_c/f^{1/2}$. Similar considerations show that the rescaled kappa is $\kappa_{\text{rescaled}} = \kappa \times f^{1/2}$. Finally, we see that with $H_{c2} = \sqrt{2}\kappa H_c$, the volumetric factors cancel; hence no rescaling is needed for H_{c2} or ξ.

Table I contains estimates for the volume fraction f obtained from the Meissner signal and from the normal state Curie background signal. The two estimates lie reasonably close together for each of the samples. The similarity lends credibility to the values obtained; this is important, for it is generally very difficult to quantify the actual amount of superconductor that is present in cuprate materials with many competing phases. In any event, the correction is relatively small for the two and three layer samples, but substantial for the single layer Hg-1201. Using the estimates for f from the Meissner signal, we obtain the rescaled values shown in the lower section of Table I. These values, together with those for H_{c2} and ξ, constitute the major quantitative results of this work. Let us now see what the study reveals in a more global way about these superconductors.

DISCUSSION

A primary objective of the present study is to develop an internally consistent, uniformly generated set of values for the length scales and characteristic fields in the Hg-based family of high-T_c cuprates. With these results, we can now look in a systematic way to see how the superconductor evolves with the number ''n'' of adjacent Cu-O layers. As background, it is well known[19] that T_c increases as n goes from 1 to 2 to 3, and this feature is evident in Table I. While materials with $n>3$ are difficult to synthesize in phase-pure form, it seems relatively clear that T_c decreases at higher n; e.g., Hg-1234 has a T_c of ~120 K. Due to the presence of multiple superconducting phases in the Hg-1234 investigated, we regard our results on this material to be unreliable and do not report them in detail.

One very clear trend in the present results is an increase in the slope of H_{c2} with layer number. As seen in Fig. 5(a), the slope rises almost linearly with n. Figure 5(b) shows the (extrapolated) zero temperature coherence length ξ_{ab}, which also varies smoothly, of course. Let us recall that H_{c2} and ξ do *not* require any correction for volume fraction; hence they should be the most well founded of the results obtained. These results can be compared with earlier studies of specific compounds. Considering first the Hg-1201 compound, we previously obtained[20] for $-dH_{c2}/dT$ the value 1.05 T/K, based on a London-limit fluctuation analysis[21,22] of random polycrystalline $HgBa_2CuO_4$. The present value, 1.2 T/K, compares well with the earlier, logarithmic accuracy determination made on different samples. Puzniak *et al.*[23] have made somewhat similar magnetic studies of aligned powder samples and obtained, in effect, two values for the slope: 1 T/K from a simple London theory analysis[24] and 1.75 T/K from an analysis similar to that described originally by Welp *et al.*[25] For the Hg-1223 material, the present values of H_{c2} slope and ξ lie in almost perfect agreement with those of Schilling *et al.*,[26] who found the slope 2.0 T/K in random polycrystalline materials. For this superconductor, Puzniak *et al.*[23] obtained values equivalent to 0.9 and 1.2 T/K using the respective procedures, as described. Other studies on individual materials have been reported by Huang *et al.*[27] and by Bae *et al.*[28]

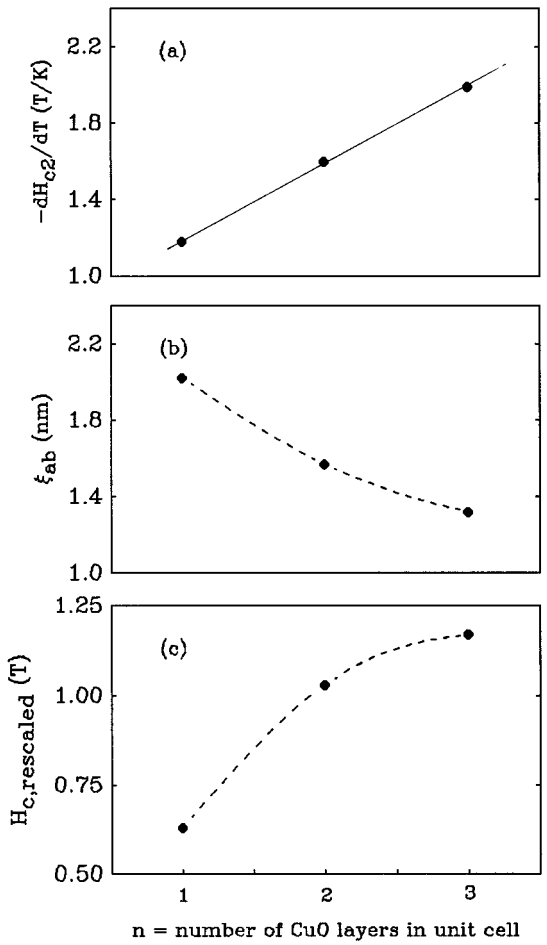

FIG. 5. Equilibrium properties versus number n of adjacent Cu-O layers in Hg-cuprates $HgBa_2Ca_{n-1}Cu_nO_{2n+2}$. (a) the deduced slope $-dH_{c2}/dT$ of the upper critical field, near T_c; (b) the coherence length ξ_{ab} in the ab (copper-oxygen) plane, extrapolated to $T=0$; (c) the thermodynamic critical field $H_{c,\mathrm{rescaled}}(T=0)$, rescaled to correct for superconducting volume fraction, as described in the text.

Now we consider the ''volume fraction corrected'' quantities. The critical field $H_c(0)$ has fundamental thermodynamic interest and is shown in Fig. 5(c) versus layer number n. For Hg-1201, the present value 0.6 T lies between the earlier determinations of Thompson *et al.*,[20] 1.0 T, and that of Puzniak *et al.*,[23] 0.43 T. For Hg-1223, we obtain $H_c(0)=1.2$ T. This is again about 50% larger than the value of Puzniak *et al.*, 0.82 T, but compares well with the result of Schilling *et al.*,[26] 1.4 T. In fact, the latter authors obtained values for both H_{c2} and κ that are almost identical to our present results, meaning that the difference in $H_c(0)$ likely results only from a difference in extrapolation procedures. It should be noted that Schilling *et al.*[26] used a rescaling procedure, similar to that described here, to account for an incomplete superconducting volume fraction.

It is interesting to see what further trends emerge from the quantitative results. To do so, we apply simple 3D relations from BCS and Ginzburg-Landau theory. Orlando *et al.*,[29] whose notation we use in the following, provided a convenient compilation of theoretical results. For example, one can use the relation

$$H_c=4.23\gamma^{1/2}T_c(1-t) \tag{3}$$

to estimate γ, the coefficient of the linear electronic heat capacity. In this tabulation, magnetic fields are measured in Oe, and all lengths are measured in cm. From the values of H_c and T_c, this relation predicts $\gamma\approx 250$–430 erg/cm^3 K^2 for the three Hg compounds. This estimate lies much lower than simple BCS theory (which neglects sizeable fluctuation effects) would predict, $\sim$2000 erg/cm^3 K^2, from the jump in specific heat of Hg-1223 near T_c as measured by Schilling *et al.*[26] The estimate compares reasonably well, however, with the value $\gamma_{\mathrm{BCS}}\sim 600$ erg/cm^3 K^2 for BiPb-2223 deduced by Schnelle *et al.*,[30] who explicitly allowed for fluctuations in their analysis. Next one has, in the clean limit, that

$$-dH_{c2}/dT=9.55\times 10^{24}\gamma^2T_c(n^{2/3}S/S_F)^{-2}, \tag{4}$$

where n is the density of conduction electrons, S is the Fermi-surface area, and S_F is the Fermi-surface area for a free electron gas of density n. Using the slopes of H_{c2} given in Table I and setting $S/S_F=1$ for simplicity, we estimate that $n\approx 0.6$, 1.1, and 1.1×10^{21} cm^{-3} for the one, two, and three layer material, respectively.

Microscopically, the deduced carrier density corresponds to having 0.08, 0.11, and 0.09 charge carriers *per layer* in the unit cell: thus the layer doping is approximately the same in the three materials. The deduced doping level is comparable with the oxygen excess $\delta\approx 0.06$ reported for fully oxygenated Hg-1201 materials[31] and $\delta/2\approx 0.11$ for oxygenated Hg-1212 (Ref. 6). It is particularly striking to compare the present findings with the results of a recent neutron powder diffractometry study,[32] performed on materials synthesized and annealed in oxygen in exactly the same manner as those studied in the present work. Rietveld analyses yielded δ values of 0.10 ± 0.01, 0.32 ± 0.03, and 0.24 ± 0.03 for the $n=1$, 2, and 3 members, respectively. This corresponds to an excess of 0.10, 0.16, and 0.08 oxygens per layer. Thus the deduced doping level per layer is similar for the three materials, which corroborates the finding in the experimentally independent magnetic analysis. Of course, these superconductors are far more complex than simple metals and details of band structure are being ignored—we omit many unknown factors of order unity. Nonetheless, with similarly structured compounds, our treatment preserves the systematics within the Hg-based family and reveals a nominally constant number of carriers per layer in the unit cell. In fact, this conclusion is conceptually consistent with earlier studies of T_c versus hole density in other high-T_c families. In particular, Presland *et al.*[33] and Tallon and Flower[34] have argued that the maximum T_c always occurs at the same number of holes per planar Cu ion, ≈ 0.16. The present results for the Hg cuprates can be regarded as independent evidence for the general concept of a constant number of carriers per copper ion, at doping levels that maximize T_c.

A constant areal density N/A of charge carries has interesting implications. If one regards *each* Cu-O layer as a 2D electron gas, then the Fermi velocity $v_F\propto(N/A)^{1/2}/m^*$ should be the same for each of the three materials, assuming that they have the same electronic effective mass m^*. Interestingly, this conjecture regarding the Fermi velocity is implicit in the results for ξ and T_c in Table I. Let us recall that, in general, one has $\xi\propto\hbar v_F/\Delta(T=0)\propto\hbar v_F/T_c$, where we

plausibly assume that the superconducting gap ratio Δ/T_c is the same for the three Hg cuprates. Thus one has that $v_F \propto \xi \times T_c$: using the results in Table I gives the same value for this product, (187 ± 10) nm K, for each of the three materials. Hence both models, with the assumptions outlined, imply that the Fermi velocity is about the same for the three optimally doped materials. This implication should, of course, be confirmed by other more direct methods.

Continuing, then, with the formalism compiled by Orlando *et al.*,[29] one has the following relation for the London penetration depth λ_{ab}:

$$\lambda(T=0) = 1.33 \times 10^8 \gamma^{1/2} (n^{2/3} S/S_F)^{-1}. \quad (5)$$

Again eliminating the hard-to-measure quantity γ gives estimates for λ_{ab} that are larger than the Table I values by a factor of 1.7 in each case. A similar numerical difference was found previously in studies of neutron-damaged BiPb-2223 materials.[35]

Given the higher T_c and short coherence lengths of the cuprate superconductors, it has become apparent that fluctuations play a larger role than in conventional superconductors. Indeed, the apparent upturn and divergence in κ at temperatures near T_c, as observed in Fig. 3, has been accounted for, by incorporating fluctuations in a London formulation.[36,37] However, we do not use the formalism here for the following reason. A central feature of this theory is a ''crossing point'' magnetization $M^* = k_B T^* / \phi_0 s$, such that the mixed state magnetization at temperature T^* is independent of H. Here ''s'' is the spacing between sets of Cu-O planes, which can be obtained from x-ray diffraction. Values for ''s'' from magnetization studies are often larger than the x-ray values, and this fact is generally interpreted in terms of an incomplete superconducting volume fraction $f = (s_{\text{expt}}/s_{\text{x ray}}) < 1$. The difficulty in the present case is that the estimates so obtained are significantly smaller (1/4 to 1/2) than either the Meissner fraction or the estimate based on the Curie background. Given this implausible result, we do not apply here the formalism incorporating fluctuations and exclude the measurements made in the fluctuation region near T_c from our determinations of H_{c2}, H_c, etc. Similar contradictions have been noted by others.[38]

Finally, we note that previous applications of the Hao-Clem formalism have yielded unlikely results in some cases. For example, analysis[39] of $M(H,T)$ for single crystals of $Bi_2Sr_2Ca_1Cu_2O_8$ ($H \| c$) gave values for H_{c2} that are independent of temperature for T in the range 35–70 K. Kogan *et al.*[40] have shown that nonlocality (a nonlocal relation between the current density **j** and vector potential **a**) accounts well for the observed temperature independence. The phenomenon is most pronounced in very clean, high-κ superconductors. On the other hand, a modest amount of electron scattering (due to structural disorder, impurities, etc.) reduces the mean free path, so that the electrodynamics becomes local and the conventional formalism can be applied. In the present case, Fig. 4 shows that H_{c2} is well behaved to the lowest temperatures where M was reversible. Consequently, the Hao-Clem formalism is appropriately applied here.

In conclusion, we have studied the equilibrium magnetization of magnetically aligned Hg-based cuprate superconductors containing one, two, and three adjacent Cu-O layers. Internally consistent values for the characteristic lengths and fields are deduced for the three materials. Specific trends include (1) a systematic increase in the slope of the upper critical magnetic field H_{c2} with layer number n and (2) evidence that the number of charge carriers per layer is nearly the same for the three materials; the latter feature is corroborated by a Rietveld analysis of neutron diffractometry experiments. Further comparisons and implications have been presented. Specific values agree well with some previous measurements[26] for Hg-1223, but differ significantly from the findings of a another study[23] based on a simple London-limit analysis.

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