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Distinguishable effects of oxygen and rhenium in $HgBa_2CuO_{4+\delta}$ superconductors

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Muon spin relaxation and magnetization experiments on $Hg_{1-x}Re_xBa_2CuO_{4+\delta}$ ceramics with different oxygen and Re contents allow us to discern the effects of the incorporation of both elements in this superconductor. For all compositions, T_c and $\sigma \sim \lambda^{-2}$ are strongly dependent on δ and display a maximum at optimal doping. The optimal T_c and σ values are nearly independent of *x*, thus indicating that the Re substitution has only second order effects on the basic superconducting properties of this compound, near optimal doping. Samples with Re have higher oxygen contents, due to the high valence of this cation. However, the shift of the $T_c(\delta)$ and $\sigma(\delta)$ curves toward higher δ values as *x* increases suggests that the oxygen atoms coordinating the Re cations are less effective to doping than the excess oxygen atoms present in Re-free samples.

INTRODUCTION

Cuprate superconductors containing mercury, with general formula $HgBa_2Ca_{n-1}Cu_nO_{2n+2+\delta}$, have been the subject of considerable interest since their late discovery,[1] due to their high critical temperatures—particularly, the $n=3$ member of the series displays the highest T_c ever reported for a superconductor, $T_c=134$ K. These high T_c's have been ascribed to the large Cu–O apical bond distances, and to the absence of buckling of the Cu–O planes.[2]

Studies of these materials soon revealed two important hindrances: their unstability and difficult synthesis, and their large anisotropy,[3] and consequently, low bulk pinning.[4] Considerable efforts have been therefore devoted to searching for chemical substitutions[5–7] which would both stabilize the phases and reduce their anisotropy. The most promising result was reported by Shimoyama and co-workers,[8] who observed that the partial substitution (10%–15%) of Hg for Re not only stabilized the compounds but also resulted in a marked increase of their irreversibility line.[9] This effect has been associated with a possible decrease of the superconductor anisotropy $\Gamma \equiv m_c/m_{ab}$, which would be due to a metallization of the blocking layers.[9,10] This would be induced by the oxygen atoms incorporated in the $Hg_{1-x}Re_xO_\delta$ planes because of the high valence of Re. Magnetic measurements on grain aligned (Hg, Re)-1223 powders point indeed to an effective anisotropy decrease.[11,12] Nevertheless, no conclusive determinations of Γ have been reported, due to the difficulties in obtaining single crystal samples of these compounds.

Besides providing one of the best ways to estimate the magnetic field penetration depth λ, muon spin relaxation (μSR) experiments constitute a useful technique to detect possible interlayer metallization effects. In fact, Uemura *et al.*[13] observed that $T_c(\sigma)$ of most high-temperature superconductors (HTS's) under and close to optimal doping fall on a unique line, the so-called Uemura line, which is thought to reflect the behavior of the individual superconducting CuO_2 planes. Here $\sigma \sim 1/\lambda^2 \sim n_s/m^*$ is the muon relaxation rate, n_s the density of Cooper pairs, and m^* the effective mass of the charge carriers. Deviations to the right of this line have been observed for cuprates with coupled CuO_2 planes (Y-123)[14,11] and for overdoped compounds,[15] and are therefore considered an indication of interlayer metallization. Recent convincing evidence of the progressive anisotropy reduction with overdoping[16] support this interpretation.

μSR measurements on Hg-based superconductors are scarce. Two Re-doped ceramics ($n=1,2$) were studied by Tallon *et al.*;[10] these samples displayed a marked deviation from the Uemura line, and therefore gave ground to the arguments of the interlayer metallization induced by Re. Nevertheless, one of these samples was not optimally doped; according to the above paragraph, overdoping could account for the observed deviation. New μSR experiments were carried out recently[17] on two Re-doped powders of composition $Hg_{0.82}Re_{0.18}Sr_2CaCu_2O_{6+\delta}$ and $Hg_{0.82}Re_{0.18}Ba_2Ca_2Cu_3O_{8+\delta}$, close to optimal doping. The measured $T_c(\sigma)$ data are in quite good agreement with the Uemura line and thus do not reveal any evidence for interlayer metallization induced by Re. Data on the unsubstituted compounds have just been reported for the $n=1$ member of the series.[18]

These scarce and controversial results therefore require for more systematic studies of these interesting materials. For instance, other still open, important questions regard the effect of the Ba substitution for Sr, and the effect of excess oxygen atoms. It was reported that oxygen treatments may sensibly modify T_c and, especially, the irreversibility line of these superconductors;[9,19] furthermore, it has been recently argued that the *c*-axis shortening resulting from the Sr substitution might have consequences as important as those due to Re.[17,20] It is important also to stress that the effects of Re, Ba/Sr, and oxygen are not independent, since the cell parameters and presence of Re modify the oxygen sites occupancy.

In this paper we undertake the study of the simplest member of the series, with a single CuO_2 plane. Through the measurements of $Hg_{1-x}Re_xBa_2CuO_{4+\delta}$ samples with different oxygen doping and Re content, we analyze the distinct effects of these two parameters on two basic superconducting properties: T_c and λ. $T_c(\delta)$ and $\sigma(\delta)$ are found to shift with x, indicating that the oxygen atoms in the $(Hg, Re)O_\delta$ planes bound to the Re atoms provide a reduced charge transfer to the superconducting CuO_2 planes, as compared with the excess oxygen atoms present in Re-free samples. The oxygen content is observed to have significant effects on $\sigma \sim n_s/m^*$, whereas the σ values of optimally doped samples are independent of their Re content, thus evidencing that this cation has little effect on the superconducting properties of this phase.

EXPERIMENT

High quality $Hg_{1-x}Re_xBa_2CuO_{4+\delta}$ ceramics with $x=0$, 0.10, and 0.15 were synthesized by the sealed quartz tube technique, as described elsewhere.[21] X-ray and neutron diffraction as well as scanning electron microscopy (SEM) did not show any trace of segregated or impurity phases. Electron dispersion spectroscopy indicated that within the experimental resolution the grains have an homogeneous cationic composition, which is close to the nominal one. Additionally, the cell parameters and bond lengths, determined from Rietveld refinements of neutron diffraction data, display a monotonic variation with Re content, thus indicating a gradual incorporation of this cation to the structure.[22] Profile refinements of the neutron diffraction data do not reveal the existence of inhomogeneities of the Re content. No traces of eventual superlattice ordering were observed in Re-substituted phases.

The oxygen content of the ceramics was modified by different treatments under oxygen pressures within the range 180 and 1.7×10^{-9} atm, and temperatures between 180 and 550 °C: high pressure annealings under pure oxygen, annealings under He–O_2 atmospheres with controlled O_2 partial pressure, and under ultra high vacuum. The annealing procedures had to be varied depending on the Re content of the sample. As-grown samples were progressively overdoped as their Re content increased; at the same time, oxygen gradually lost mobility. Therefore, as x increased higher temperatures and lower oxygen pressures were required to obtain optimally and underdoped samples.[21]

The oxygen content was determined by thermogravimetric and neutron diffraction analyses.[21] The critical temperature was obtained from the diamagnetic onset in the low-field temperature-dependent dc magnetization of ceramic bars, measured with a commercial superconductor quantum interference device magnetometer. Further characterization of the superconducting properties of these samples with different oxygen and Re contents will be published elsewhere.[23]

Transverse field μSR experiments were performed at ISIS (Rutherford-Appleton Laboratory, United Kingdom). The powders were cooled in a closed-cycle cryostat, after applying a field $H=500$ Oe. This value was shown in a previous work[17] to be high enough to ensure that the intervortex spacing in the superconductor is at most of the order of the penetration depth λ, and thus the observed muon relaxation rate, due to this modulated field distribution, is $\sigma \sim 1/\lambda^2 \sim n_s/m^*$.

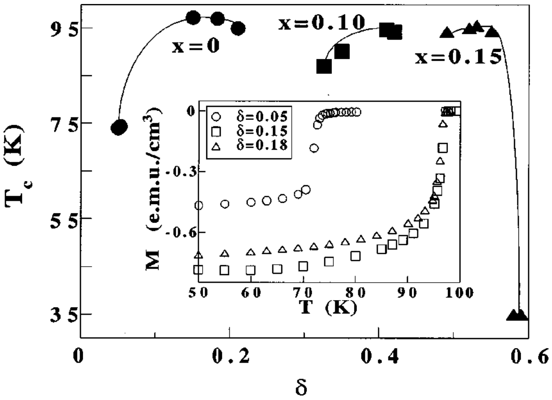

FIG. 1. Evolution of the critical temperature with oxygen excess δ, for samples with different Re content x. (Inset) Low field diamagnetic transitions of $x=0$ samples with different oxygen doping.

The muon precession was measured at temperatures between 10 K and above T_c. The recorded data display two contributions: a dominant one with fast amplitude decay plus a nearly constant one, which becomes apparent at longer times and is present above T_c. The latter, with precession frequency corresponding to the external field, is likely due to some amount of nonsuperconducting material in the sample, or nonscreened Hg-1201 phase. The first contribution arises from the vortex lattice in the superconductor, as evidenced by its slightly lower precession frequency, which results from the negative magnetization of the superconductor.

In order to obtain the muon relaxation rate associated to the superconducting state, $\sigma \propto \lambda^{-2}$, the recorded signal was thus fitted to a two-component function; the first component, a Gaussian $\exp(-\sigma^2 t^2/2)$, accounts for the decay due to the field modulation inside the superconductor; the second, minor, component accounts for the background contribution, observed also in the normal state. The obtained fits and σ values were verified to not depend on this background contribution.

When measuring rapidly decaying signals, care must be taken to ensure that the determined relaxation rate σ is not limited by the finite width of the muon pulse (40 ns at ISIS). In our measurements, the asymmetry signal recorded during the experimental runs providing the largest σ values was not different from that obtained in runs corresponding to significantly smaller relaxation rates. Thus, the maximum σ values obtained in this study are not limited by the finite width of the muon pulse. Further support to this conclusion is provided by the fact that the temperature dependence of σ is essentially the same-within the error bars—for all the measured samples, in spite of their rather different values of T_c and σ at low temperatures (see Fig. 2 and related discussion below).

RESULTS

Figure 1 shows the evolution of the critical temperature T_c with the oxygen excess δ, for the measured samples. T_c is obtained from the low field ($H=10$ Oe) magnetic transition. The inset in Fig. 1 displays some of these transitions, having

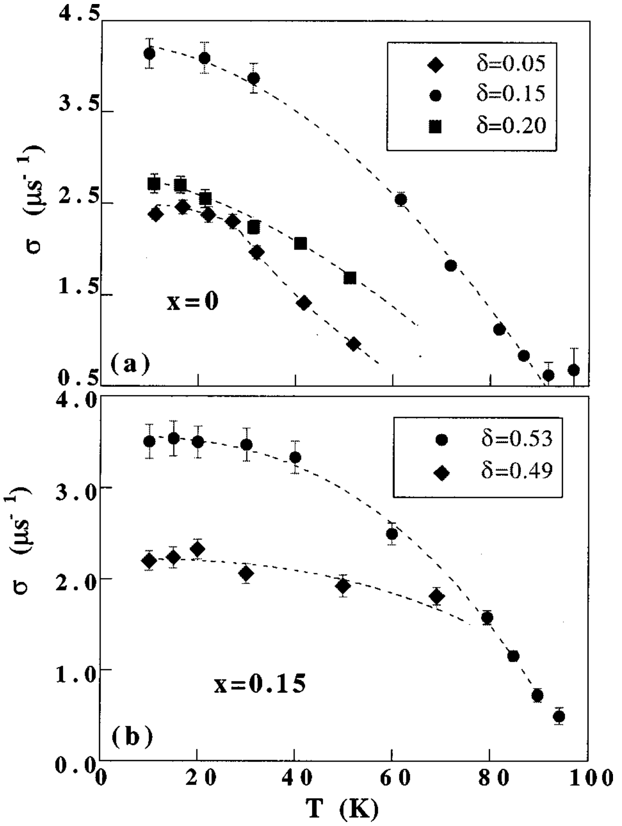

FIG. 2. Temperature dependence of the muon relaxation rate σ, for Hg-1201 ceramics with different doping level, for $x=0$ (a) and $x=0.15$ (b).

typical widths of a few K. The powdered samples showed only partial field screening, as usually reported for Hg-based superconductors.[24,25] The dependence of the superconducting volume fraction on δ and x will be further addressed below.

The $T_c(\delta)$ points belonging to the Re free samples describe a well defined bell-shaped curve, as commonly found in these[24,26] and the other HTS's.[27] The data points of samples with Re appear to also describe a bell-shaped curve, with nearly the same maximum T_c, but shifted $\Delta\delta\approx 0.23$ and 0.37 for $x=0.10$ and $x=0.15$, respectively, with regard to that of Re-free compounds. Superconducting Re-substituted phases display thus higher oxygen contents than the unsubstituted ones. Moreover, Fig. 1 reveals that samples with Re, with oxygen contents equal to those of heavily overdoped Re-free samples, have T_c values corresponding to optimal doping.

Figure 2 display the evolution with temperature of the muon depolarization rate for some of the measured ceramics, with different Re content and doping level. The transition of σ is much broader than that of dc magnetization [compare for instance Fig. 2(a) with the inset on Fig. 1, corresponding to the same samples]; this is a feature commonly observed in HTS materials, and attributed to their d-wave nature. Nevertheless, a tendency toward saturation of σ at low temperatures may be clearly appreciated in Fig. 2. The dependence of $\sigma(T)$ may be described by a law $\sigma(T)\sim 1-(T/T_c)^n$, with $n\approx 2-3$ for all the samples studied; and, particularly, the enhancement of σ when decreasing the temperature from 20 to 10 K is lower than the error bars of σ, in all our samples. This saturation trend contrasts with data reported in Ref. 18 for Re-free Hg-1201 ceramics. In this work none of the measured samples showed any indication of saturation; for instance, the change of σ for the optimally doped sample between 20 and 10 K is about 0.33 μs^{-1}, much larger than that observed in our equivalent sample, and significantly above our error bars. It should be remarked, however, that the experimental incertitude in the σ values on Fig. 2 allows for a residual temperature dependence of σ at low temperatures; these data are thus not in contradiction with the likely d-wave nature of these materials.

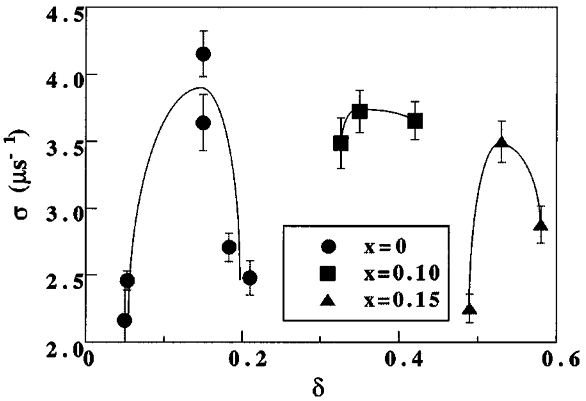

FIG. 3. Values of the low temperature ($T=10$ K) muon relaxation rate σ for superconducting Hg-1201 samples with different Re and oxygen contents.

Figure 3 displays the values of the muon depolarization rate of the $(\mathrm{Hg,Re})\mathrm{Ba_2CuO_{4+\delta}}$ ceramics recorded at 10 K, as a function of their excess oxygen. The $\sigma(T)$ dependence, shown in Fig. 2 and discussed in the previous paragraph, indicates that these low temperature values are a good approximation to those corresponding to $T=0$. Any monotonic enhancement of $\sigma(x,\delta)$ from its 10 K value toward $T=0$ will not modify any of the systematic trends and behavior discussed in the following.

The data belonging to the Re-free compounds clearly display a maximum at optimal doping. The Re-substituted phases seem to display the same kind of behavior, but with the $\sigma(\delta)$ curve shifted progressively to higher δ values. The maximum value of σ is nearly insensitive to the Re content. In fact, $\sigma(\delta)$ behaves much like $T_c(\delta)$: both have a maximum at optimal doping, fairly independent of x, and display identical shifts in δ as x increases.

The nonmonotonic dependence of σ with doping is in agreement with that reported for Re-free Hg-1201 ceramics in Ref. 18. However, we should mention that although the σ values for the optimally doped samples are nearly coincident in both works, some discrepancies exist for the overdoped samples. Indeed, the σ value of our $\delta=0.20$ sample is significantly smaller than that reported in Ref. 18 for a heavily overdoped sample with $\delta=0.37$. The reason for such a difference is not presently understood; however, the reported extreme sensitivity of T_c to the charge carrier density in the overdoped regime[24] may be of relevance.

DISCUSSION

The shift of the $T_c(\delta)$ curve in Fig. 1, roughly proportional to the Re content, indicates that the superconducting phases with Re have higher oxygen contents than the Re-free ones. In fact, this is what should be expected, since each Re cation tends to form ReO_6 octahedra. In order to fulfil them, four oxygen atoms should be incorporated into the $(Hg, Re)O_\delta$ plane for each Re atom.[19] Indeed, the oxygen excess δ in as-grown Re-substituted samples is found to be very close to $4x$.[21,22] Neutron diffraction data have provided evidence that in Re-substituted samples nearly all the oxygen atoms in the $(Hg, Re)O_\delta$ plane are bound to the Re cations.[22]

Figure 1 shows also that: (i) the shift of the $T_c(\delta)$ curves for samples with different Re is lower than $4x$ ($\Delta\delta \approx 0.23$ and 0.37 for $x=0.10$ and 0.15, respectively), and (ii) the oxygen excess corresponding to optimal doping deviates progressively from $4x$ [$\delta_{opt}(x=0.10) \approx 0.42$, and $\delta_{opt}(x=0.15) \approx 0.53$], i.e., as grown samples become overdoped as x increases. These two facts reveal that the oxygen atoms in the $(Hg, Re)O_\delta$ plane bound to Re must contribute to the doping of the compound, but their role must be different from that of the excess oxygen atoms present in the HgO_δ plane of Re-free phases. Following the crystallographic notation,[22] and in order to lighten the text, we will refer to these two oxygen sites as O(4) and O(3), respectively. The O(3) atoms are located at the center of the Hg-square network, whereas the O(4) ones are displaced toward the Re cations.

As stated above, Re-free phases only have O(3) oxygen atoms, whereas in Re-substituted phases almost all the excess oxygen atoms are O(4). On the other hand, the optimal doping of the Re-free Hg-1201 structure corresponds to a hole concentration $p_{opt} \approx 0.16$.[24,26] Preliminary Seebeck measurements on our Re-substituted samples indicate that this value is not modified by the Re presence,[22] as the invariance of the optimal T_c already suggests. These observations allow an estimate of the effective valence of the excess oxygen atoms O(3) and O(4), as well as their charge transfer to the superconducting CuO_2 planes. Assuming that when $\delta=0$ the compound is an insulator, which is true for the Re-free phase,[28] and knowing that excess oxygen atoms occupy a single site [either O(3) or O(4)] at fixed x, one can roughly estimate the charge transfer of the O(3) and O(4) atoms by dividing P_{opt} by the excess oxygen corresponding to optimal doping $\delta_{opt}(x)$. This simple calculation reveals that in Re-free samples each O(3) atom provides nearly one hole to the CuO_2 planes. This value is in very good agreement with both theory and detailed Seebeck measurements, which indicate that the effective valence of the O(3) atoms lies between 0.7 and 1.[24,28] For the $x=0.10$ and 0.15 samples, our estimate indicates that each O(4) provides 0.38 and 0.30 holes, respectively, to the superconducting planes. The O(4) atoms appear therefore less effective to doping than the O(3) ones, i.e., there is a reduced charge transfer from the oxygen atoms coordinating the Re cations in the $(Hg, Re)_\delta$ plane, as compared to that of excess oxygen atoms in the Re-free compound.

An alternative estimate may be obtained by simply making a charge balance, assuming that the effective valence of Re is roughly 6+, as the Re–O bond length summation suggests.[22] The O(4) valence obtained in such a way is roughly 1.3, both for $x=0.10$ and 0.15. Facing this and the above estimates, it seems clear that each O(4) atom borrows one electron from the Re cation and about 0.3 from the CuO_2 planes. Such a conclusion is fully consistent with the preferred octahedral coordination of Re, and the four oxygen atoms brought by each Re cation to the structure. Thus, although the valence of the O(4) atoms may be higher than that of the O(3) atoms, they transfer less holes to the superconducting planes, because they are tightly bound to the high valence Re cations. This reduced hole doping per oxygen atom is overbalanced by the higher content of oxygen in Re-substituted samples, and thus as-grown samples with increasing Re contents are progressively overdoped.

We now move to the behavior of σ with oxygen doping and Re substitution, shown in Fig. 3. The similarity of the $T_c(\delta,x)$ and $\sigma(\delta,x)$ curves suggests that $\sigma \sim n_s/m^*$ is mainly reflecting the changes in the charge carrier density. At a given x, σ increases when going from under to optimal doping, indicating an increase of the superfluid density. This is a common observation in HTS. Hole doping is known to lead to an increase of the carrier density and a suppression of the pseudogap;[24,29] in the underdoped regime, both effects result in an enhancement of the superfluid density. As Fig. 3 reveals, σ reaches a maximum for optimal doping and then decreases again in the overdoped regime. This fall of σ for overdoped samples has also been observed in other HTS, specially in T1-based compounds.[15] It may be due to a reduction of the Cooper pair density n_s or to an enhancement of the effective mass m^* of the charge carriers within the *ab* planes. Since increasing σ above optimal doping results in a progressive increase of the normal state carrier density in Hg-1201 compounds, at least for $\delta < 0.22$,[24] a reduction of n_s would imply the existence of enhanced pair breaking in the overdoped state, as argued in Ref. 15. On the other hand, an increase of m^* cannot be disregarded as there is increasing disorder in the BaO layers when increasing the oxygen content: a displacement of Ba toward the $(Hg, Re)O_\delta$ plane takes place, induced by the strong bonds between Ba and extra oxygens in this plane.[26]

Figure 3 reveals that the changes in the oxygen content—and therefore in doping—have more important effects on σ than the substitution of Re for Hg. Even more, the maximum σ value, corresponding to optimal doping, is shown in Fig. 2 to be nearly independent of the Re content, within the error bars. This observation, together with the absence of any significant effect of Re on the optimal T_c, allows some important conclusions to be drawn. The insensitivity of the optimal T_c to the Re presence implies that the optimal density of Cooper pairs is not affected by the Re incorporation. Thus, the invariance of the optimal σ value reveals that the effective mass of the charge carriers remains also basically unaltered by Re, at least at optimal doping. That is, Re appears to have minor effects on the basic superconducting parameters of Hg-1201 compounds. In fact, *none of the Re-substituted samples has larger σ values than those corresponding to optimally doped Re-free samples*. Even more, the slightly underdoped sample with $x=0.15$ displays a σ value as small as those of more underdoped samples without Re.

The observed insensitivity of the superconducting properties of these compounds to the Re presence is to some extent unexpected, since the incorporation of this cation is known to have important effects on the crystal structure of the Hg-based phases. The high valence of Re, and much shorter length of the Re–O bond as compared to the Hg–O bond, result in structural modifications, evidenced specially in the

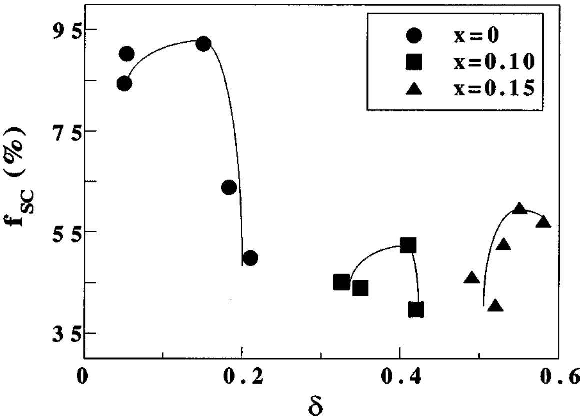

FIG. 4. Dependence of the superconducting volume fraction f_{SC} on the oxygen excess δ, for samples with $x=0$, 0.10, and 0.15.

shortening of the *c*-axis length. One would therefore expect relevant changes in the tails of the band structure of 1201 compounds. In spite of it, the present results indicate that the Re-induced structural changes do not modify the conduction band near optimal doping. In this sense, it would be interesting to perform μSR measurements on Re-substituted samples far away from optimal doping, particularly in the underdoped regime, where changes in the band structure and disorder induced localized states should be more evident. These samples, however, are hard to obtain, due to the tight Re–O bonds, which reduce considerably the mobility of oxygen atoms and make oxygen removal difficult. In view of our results, other interesting experiments should involve the study and comparison of other cation (Pb, Cr) substituted Hg-based superconductors, with depressed optimal T_c.

It had been also suggested that the *c*-axis shortening and metallic character of ReO_3 should result in an interlayer metallization, and thus a change in the dimensionality of these superconductors, which would manifest by a significant enhancement of σ.[10] The present data do not show any evidence of it, in accordance with some recent results obtained for the Hg-based compounds with 2 and 3 Cu–O layers.[17]

Finally, we would like to draw attention to the implications of the $\sigma(\delta)$ behavior shown in Fig. 3, regarding the field penetration depth and field screening response. Figure 4 displays the superconducting volume fraction f_{SC} as a function of the excess oxygen δ, for all our samples. f_{SC} is defined as the ratio between the screened and total volume of the sample, $f_{SC} \equiv V_{SC}/V$, and was obtained from the slopes of the low field $M(H)$ data in powder samples. The observation of only a partial field screening in the Meissner state is common in Hg-based superconductors.[24,25,18] Comparison of Figs. 3 and 4 reveals a close similarity of the evolution of σ and f_{SC} with doping that of the $\sigma(\delta)$. Both quantities—as well as T_c—display a maximum at optimal doping. The simplest link between σ and f_{SC} is established by the penetration depth λ. Within the London model, λ and the muon relaxation rate are related through $\lambda^{-2} \sim n_s/m^* \sim \sigma$; for polycrystalline samples with anisotropy ratio $\lambda_c/\lambda_{ab} > 5$, where λ_c and λ_{ab} are, respectively, the penetration depths along the *c* axis and in the *ab* planes, the λ value extracted from σ is roughly λ_{ab}. Therefore, the existence of a maximum value of σ as a function of δ implies that λ_{ab} displays a minimum close to optimal doping; from data in Fig. 3, this optimal value is found to be $\lambda_{ab} = 135 \pm 3$ nm for $x=0$. On the other hand, if the superconducting grains have an average size *a* comparable to λ_{ab}, the screened volume of the ceramic sample is a factor $(1-\lambda_{ab}/a)^2$ smaller than its overall volume, and thus $f_{SC} \sim (1-\lambda_{ab}/a)^2$. The correlation of f_{SC} and σ revealed by Figs. 3 and 4 suggests that, indeed, the nonmonotonic behavior of the superconducting volume fraction might be reflecting the dependence of λ with doping. Although SEM images of some of our ceramic samples indicate the existence of grains as large as 5 $\mu m \gg \lambda_{ab}$, this by no means excludes the existence of subgrain boundaries which might reduce the effective grain size: the extreme sensitivity of Hg-based compounds to mechanical stress points in this direction.[25] In summary, the fact that the effective grain size of Hg-based superconducting ceramics is comparable to λ_{ab} would allow the observation of a dependence of the superconducting volume fraction on doping, and establish a relationship between f_{SC} and the measured σ.

CONCLUSIONS

In conclusion, measurements of the muon relaxation rate in Hg-1201 superconductors with different oxygen and Re contents have revealed their distinct effects on some fundamental parameters of these superconductors. Oxygen has significant effects on σ and T_c, which display a maximum as a function of δ. Re-substituted samples display higher oxygen content, due to the high valence of the Re cation, and consequently $T_c(\delta)$ and $\sigma(\delta)$ are shifted to higher δ values. However, we have found that their optimal values are nearly insensitive to Re. Thus, the Re substitution does not appear to produce any relevant changes on the optimal Cooper pair density, nor on the effective mass of the charge carriers along the superconducting planes and the effective dimensionality of the material. It is found that the oxygen atoms bound to Re in the $(Hg, Re)O_\delta$ planes are less effective to doping than the interstitial oxygen atoms in Re-free samples; this reduced charge transfer is nearly counterbalanced by the higher oxygen content due to the Re presence, for $x \leqslant 0.10$; as-grown samples with higher Re content are progressively overdoped. On the basis of the present results early suggestions of flux pinning enhancement due to a Re-induced anisotropy reduction should be reconsidered.

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