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Oxygen isotope effects in $La_{2-x}Sr_xCuO_4$: evidence for polaronic charge carriers and their condensation

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Abstract. Magnetization measurements have been performed on the oxygen-isotope exchanged samples (^{16}O and ^{18}O) of the one-layer cuprate superconductors $La_{2-x}Sr_xCuO_4$ ($0.06 \leqslant x \leqslant 0.15$). From magnetization measurements on the fine-grained and decoupled samples in the Meissner state, we find substantial oxygen-isotope effects on both the penetration depth $\lambda(0)$ and T_c. From normal-state susceptibility measurements, we are able to show that there is a negligible oxygen-isotope effect on the carrier density n. The combined results strongly suggest that there is an oxygen-isotope effect on the effective supercarrier mass m^{**}, which is huge for $x = 0.06$, and reduced to a smaller value for $x = 0.15$. We discuss the isotope effects, supercarrier mass anisotropy, normal-state gap, in-plane penetration depth, and mid-infrared spectra for $x \leqslant 0.09$ on the basis of the small polaron theory of superconductivity. We find that the agreement between the calculated and experimental results is excellent without any adjustable parameters.

1. Introduction

The microscopic pairing mechanism for high-T_c superconductivity (HTSC) is one of the most controversial issues in condensed matter physics. Eleven years after the discovery of the high-T_c cuprate superconductors by Bednorz and Müller [1], there have been no microscopic theories that can describe the physics of high-T_c superconductors completely and unambiguously. The high values of T_c (>30 K) in the cuprate superconductors were not expected from the conventional phonon-mediated pairing mechanism, so many non-phonon mediated mechanisms have been proposed [2–5]. A great number of experimental results appear to support some of these unconventional mechanisms. On the other hand, there is increasing experimental evidence that a strong electron–phonon coupling is present in cuprates [6], pointing towards an important role of phonons in the pairing mechanism. Therefore, it is quite likely that both strong electron–phonon interaction and strong electron–electron Coulombic correlation play important roles in the physics of high-temperature superconductors.

In order to develop a correct microscopic theory for high-temperature superconductivity in cuprates, one has to understand many unusual physical properties observed (e.g., the normal-state gap [7–11], novel isotope effects [12–22], and very large supercarrier mass anisotropy [23, 24]). It appears that the non-phonon mediated pairing mechanisms can partly explain these unusual properties, but have some difficulties to account for the observed isotope effects. Recently, Zhao *et al* [25, 26] have demonstrated polaronic charge carriers in the magnetoresistive manganites which share many common features with the cuprate superconductors: both systems, for example, show a strong Jahn–Teller effect. The formation of polaronic charge carriers in manganites arises from a strong Jahn–Teller effect which is linked with a strong electron–phonon interaction [27, 28]. The experimental demonstrations for the presence of polaronic charge carriers in manganites may imply that such polaronic charge carriers should also exist in cuprates. Several independent experimental results have indeed pointed towards this possibility [17–19, 29, 30].

There is an important relevant question concerning the oxygen-isotope experiments: whether the two isotope samples have identical carrier concentrations. Although this issue has been partly addressed in several of our previous publications [17–21], many researchers still do not believe that the observed isotope effects in cuprates are intrinsic. In this paper, we report a systematic study of the oxygen-isotope effects in the one-layer cuprate superconductors $La_{2-x}Sr_xCuO_4$ (LSCO). From the systematic study, we are able to show that the carrier concentrations for the two isotope samples are indeed the same, and that the effective mass of supercarriers depends strongly on the oxygen-isotope mass in the deeply underdoped regime.

2. Experimental details

Samples of $La_{2-x}Sr_xCuO_4$ were prepared by a conventional solid-state reaction using dried La_2O_3 (99.99%), $SrCO_3$ (99.999%), and CuO (99.999%). The powders were mixed, ground thoroughly, pressed into pellets, and fired in air at 1000 °C for ∼96 h with three intermediate grindings. To obtain samples with small grains and sufficient porosity for the isotope experiments, we reground the samples thoroughly, pressed them into pellets, and annealed them in air at 900 °C for 12 h. Each pellet was broken in half, and the halves were then subjected to ^{16}O and ^{18}O isotope diffusion. The diffusion was carried out for 40 h at 900 °C and oxygen pressure of about 1 bar. The cooling time to room temperature was 4 h. The oxygen-isotope enrichment was determined from the weight changes of the ^{16}O and ^{18}O samples. The ^{18}O samples had ∼90% ^{18}O and ∼10% ^{16}O. Back-exchange was carried out at 850 °C for 8 h in air. X-ray diffraction (XRD) and scanning electron microscopy (SEM) have been used to characterize the samples. Both ^{16}O and ^{18}O samples are single-phase without any observable impurity phases. The lattice constants for the two isotope samples are the same within the experimental accuracy. The oxygen content is close to 4.0 as determined by titration.

Magnetization was measured with a Quantum Design SQUID magnetometer. A magnetic field of 2 T was used for the measurements of the normal-state susceptibility. The background of the sample holder is small (∼5–10% of the sample signal), and has been subtracted from all the data shown. The Meissner effect was measured in a magnetic field of 1 mT after the samples had been cooled from the normal state in the field. The magnetic field remained unchanged during each series of measurements.

3. Results

Figure 1 shows the temperature dependence of the Meissner effect for the ^{16}O and ^{18}O samples of $La_{1-x}Sr_xCuO_4$ with $x = 0.06$ (figure 1(a)), $x = 0.09$ (figure 1(b)), $x = 0.12$ (figure 1(c)). The results for other compositions ($x = 0.105$, 0.110, 0.115, and 0.15) were shown in [17, 19]. For $x = 0.06$, the T_c of the ^{18}O sample is lower than that of the ^{16}O sample by ∼1.0 K, and the low-temperature Meissner fraction of the ^{18}O sample is reduced by 23(2)% relative to the ^{16}O sample. The exponent of the oxygen-isotope

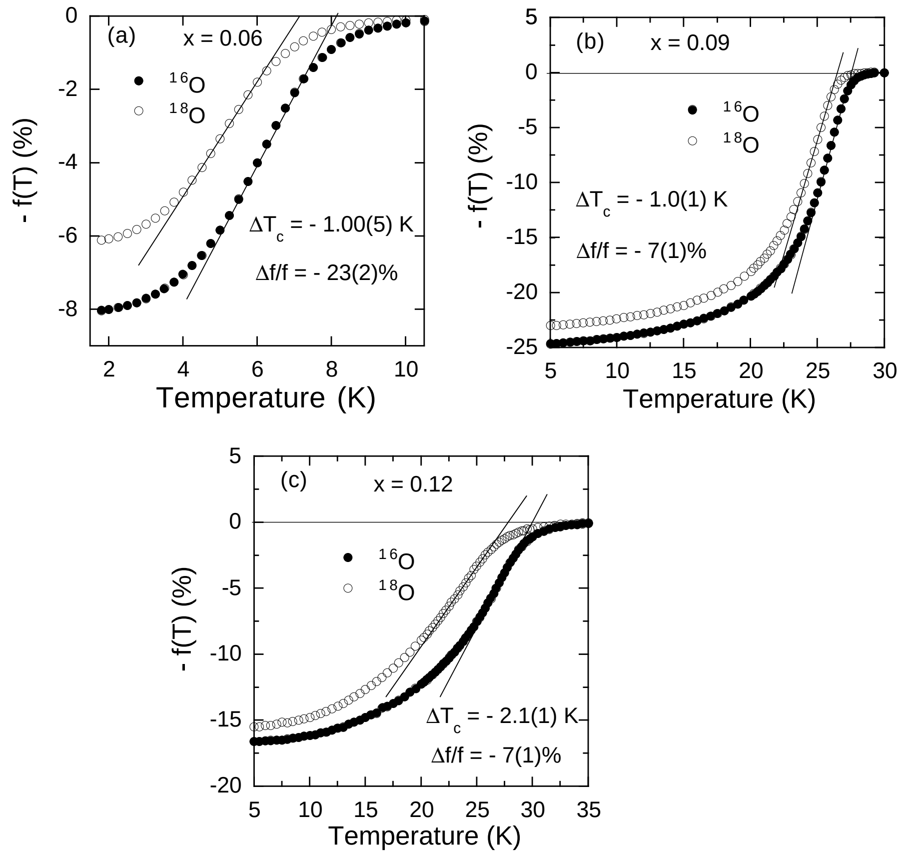

Figure 1. Temperature dependence of the Meissner effect for ^{16}O and ^{18}O samples of $La_{2-x}Sr_xCuO_4$ with $x = 0.06$ (a), and $x = 0.09$ (b), and $x = 0.12$ (c).

Table 1. Summary of the oxygen-isotope effects in $La_{2-x}Sr_xCuO_4$.

x	$T_c(^{16}O)$ (K)	$T_c(^{18}O)$ (K)	α^O	f(%)	$\Delta f/f$ (%)
0.06	8.0	7.0	1.05(5)	8.0	−23(2)
0.09	27.5	26.5	0.34(1)	25	−7(1)
0.105[a]	27.8	26.3	0.50(2)	25	−7(1)
0.110[a]	26.1	24.5	0.54(2)	22	−5(1)
0.115[a]	27.3	24.7	0.90(2)	20	−8(1)
0.120	30.2	28.1	0.62(2)	17	−7(1)
0.15[b]	37.0	36.5	0.12(1)	14	−3.7(5)

[a] [19].
[b] [17].

effect on T_c ($\alpha^O = -\mathrm{d}\ln T_c/\mathrm{d}\ln M_O$) is calculated to be 1.05(5). The oxygen-isotope effects for other compositions are summarized in table 1 and are shown in figure 2. It is clear that α^O does not increase monotonically with decreasing x (see figure 2(a)); there is an additional large contribution to α^O near $x = 0.115$. This additional contribution has been explained by Pickett *et al* [31] who showed a strong anharmonicity of phonon modes

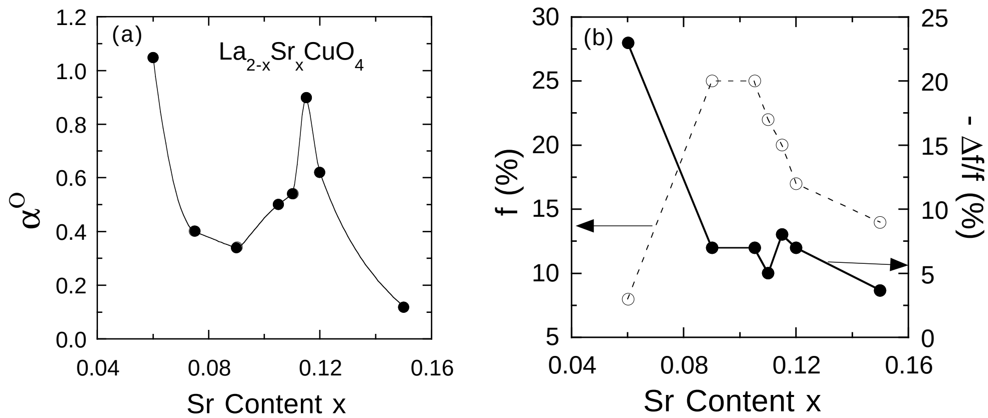

Figure 2. Dependence of α^O on the Sr content x for ^{16}O and ^{18}O samples in $La_{2-x}Sr_xCuO_4$ (a); and dependence of $\Delta f/f$ and of f on x (b). The data for $x = 0.075$ are taken from [12], the data for $x = 0.100$, 0.105, 0.110, 0.115 are from [19] and the data for $x = 0.15$ are from [17].

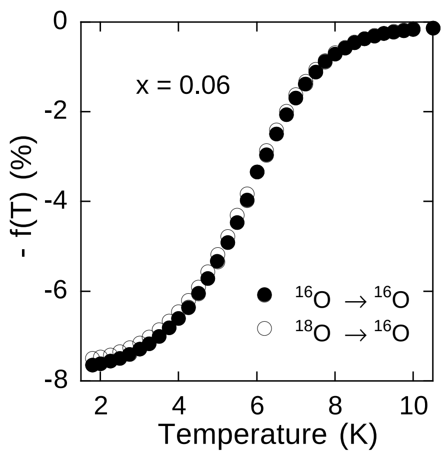

Figure 3. Temperature dependence of the Meissner effect for a pair of samples ($x = 0.06$) which have been back-exchanged to the ^{16}O isotope (one from ^{18}O to ^{16}O, and another from ^{16}O to ^{16}O) by annealing them together in air for 8 h.

near this composition. On the other hand, the oxygen-isotope effect on the Messner fraction seems to have a weak dependence on the composition for $0.09 \leqslant x \leqslant 0.12$ (see figure 2(b)).

In figure 3 we show the temperature dependence of the Meissner effect for a pair of samples ($x = 0.06$) which have been back-exchanged to the ^{16}O isotope (one from ^{18}O to ^{16}O, and another from ^{16}O to ^{16}O) by annealing them together in air for 8 h. It can be seen that the two samples have nearly the same T_c and Meissner fraction after they have been exchanged back to the same oxygen isotope. This indicates that the observed large oxygen-isotope effects on both T_c and Meissner fraction (figure 1(a)) are caused only by the exchange of the oxygen isotope. By comparing the results shown in figure 1(a) and figure 3, one can also see that both T_c and the Meissner fraction for the sample annealed in 1 bar $^{16}O_2$ environment are slightly larger than that for the same sample annealed in air (0.2 bar $^{16}O_2$ environment). This is due to a very small difference in the hole concentration of the sample which was annealed in very different oxygen partial pressures. Therefore, the hole concentration of this system is very insensitive to the oxygen partial pressure used for annealing or diffusion.

The observed large oxygen-isotope effects in these cuprate superconductors will place strong constraints on the microscopic pairing mechanism of HTSC only if one can convincedly prove that there is a negligible oxygen-isotope effect on the hole concentration. There are no direct experimental techniques which are accurate enough to clarify whether the hole concentrations for the two isotope samples are identical. This leaves room for people to have a reasonable doubt about the observed large oxygen-isotope effects. Very often, most researchers just ignore the observed isotope effects and thus the role of the electron–phonon interaction in the pairing mechanism. This is because they speculate that the difference in the hole concentrations of the two isotope samples should be large enough to mimic the observed isotope effects. Although such a speculation is not completely unreasonable, one needs experiments to prove this. There are several indirect experiments that can determine a very small difference in the hole concentrations of the two isotope samples. The first experiment is the accurate measurement of the oxygen-isotope effect on the structural-phase transition temperature T_s in $La_{1-x}Sr_xCuO_4$, since T_s in this system is very sensitive to the hole concentration. We have shown [17] that T_s is nearly independent of the oxygen-isotope mass, which implies that the hole concentrations of the two isotope samples differ by less than 0.1%. The second experiment is the accurate measurement of the normal-state susceptibility for the two isotope samples. The results are shown in figure 4(a). It is clear that the normal-state susceptibility is independent of the oxygen-isotope mass. Figure 4(b) shows the normal-state susceptibility for a ^{16}O sample ($x = 0.06$) which was annealed in 1 bar oxygen partial pressure, and for the same sample which was annealed in air. The very different oxygen partial pressures for annealing the same sample lead to a very small difference in T_c (<0.2 K), but a noticeable difference in the normal-state susceptibility. From the difference in T_c, and the dependence of T_c on the hole concentration (see later), we can estimate that the difference in the hole concentration of the sample annealed in 1 or 0.2 bar oxygen partial pressure is less than 0.0002 per cell. Therefore, the results shown in figure 4 vividly demonstrate that the difference in the hole concentrations of the two isotope samples must be far less than 0.0002 per cell. Such a difference is too small to account for the huge oxygen-isotope effects on both T_c and the Meissner fraction in the underdoped region, especially for $x = 0.06$.

In figure 5, we plot the dependence of T_c on the Sr content for both ^{16}O and ^{18}O samples. It is important to note that the T_c values of the ^{18}O samples are always lower than those for the ^{16}O samples, independent of whether the slope ($\mathrm{d}T_c/\mathrm{d}x$) is positive or negative, and that the maximum T_c shift does not correspond to a composition where there is a maximum slope. This result gives additional evidence that the observed large oxygen-isotope shifts are not caused by any difference in the hole concentration, because such a difference would lead to a parallel shift of the T_c against x curves for the two isotope samples. Therefore, these results consistently indicate that there is a negligible oxygen-isotope effect on the hole concentration in this one-layer cuprate system.

4. Data analysis

Since there is no difference in the hole concentrations of the two isotope samples, the observed large oxygen-isotope effects on both T_c and the Meissner fraction must be caused by a strong electron–phonon interaction. By analogy to the manganite system, a strong Jahn–Teller effect in cuprates manifests a strong electron–phonon coupling, and may lead

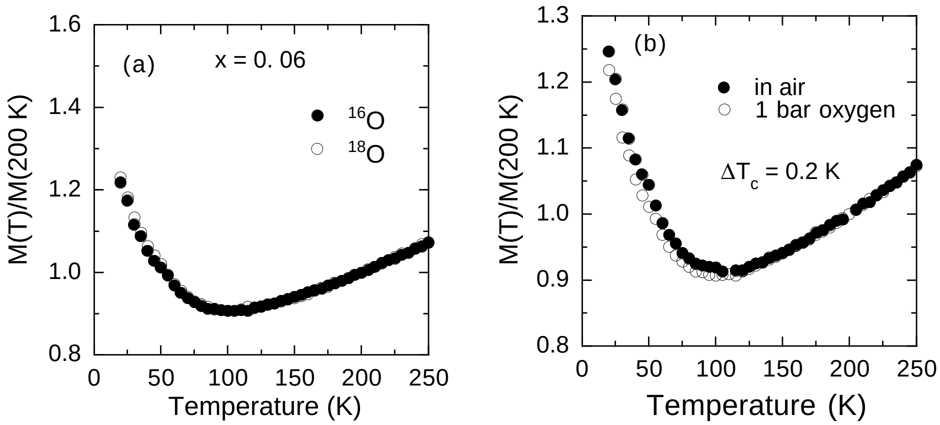

Figure 4. Normal-state susceptibility for the ^{16}O and ^{18}O samples of $x = 0.06$ (a); normal-state susceptibility for a ^{16}O sample ($x = 0.06$) which was annealed in 1 bar oxygen partial pressure, and for the same sample which was annealed in air (b).

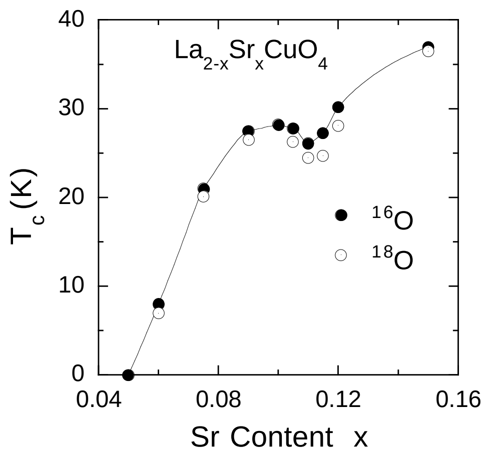

Figure 5. Dependence of T_c on the Sr content x for ^{16}O and ^{18}O samples in $La_{2-x}Sr_xCuO_4$. The data for $x = 0.075$ are taken from [12], the data for $x = 0.100$, 0.105, 0.110, 0.115 are from [19], and the data for $x = 0.15$ are from [17]. We have set $T_c = 0$ for $x = 0.05$.

to the formation of polaronic charge carriers (e.g., Jahn–Teller polarons [32]). The formation of the polaronic charge carriers in the Jahn–Teller manganites has indeed been shown by both experimental and theoretical investigations [25, 27]. There is no reason why such charge carriers do not exist in similar systems such as the cuprate superconductors. Optical measurements have shown that polaronic charge carriers exist in cuprates [33], and condense into supercarriers in the superconducting state [29]. Because the effective mass of polaronic charge carriers depend on the isotope mass [25], one should be able to observe the isotope effect on the effective supercarrier mass m^{**} if polaronic charge carriers condense into supercarriers.

Since the low-temperature magnetic penetration depth $\lambda(0)$ is proportional to $\sqrt{m^{**}/n_s}$, then

$$\Delta m^{**}/m^{**} = 2\Delta\lambda(0)/\lambda(0) + \Delta n_s/n_s \tag{1}$$

where Δ means any small change of a quantity upon isotope substitution. Thus the isotope dependence of m^{**} can be determined if one can independently measure the isotope dependences of $\lambda(0)$ and of n_s.

For decoupled and fine-grained samples, the Meissner fraction $f(0)$ depends on the penetration depth $\lambda(0)$ and on the average grain radius R (precisely speaking, on the grain size distribution), as seen from the Shoenberg formula for spherical grains [34]:

$$f(T) = \frac{3}{2}\left[1 - 3\left(\frac{\lambda(T)}{R}\right)\coth\left(\frac{R}{\lambda(T)}\right) + 3\left(\frac{\lambda(T)}{R}\right)^2\right] \tag{2}$$

where $\lambda(T) = ([(\lambda_{ab}(T)]^2\lambda_c(T))^{1/3}$ for layered materials [35]. From equation (2), one can see that a change in $f(0)$ will lead to a change in $\lambda(0)$. Therefore, the observed large oxygen-isotope effect on $f(0)$ implies that $\lambda(0)$ depends strongly on the oxygen mass. Since there is a negligible dependence of the hole concentration on the isotope mass as shown above, the observed large oxygen-isotope effect on $\lambda(0)$ means a strong dependence of the effective supercarrier mass m^{**} on the oxygen-isotope mass. This gives strong evidence that polaronic charge carriers exist and condense into supercarriers in the cuprate superconductors.

Although a quantitative determination of the oxygen -sotope effect on $\lambda(0)$ from the magnetization measurements may need further measurements of the particle size distribution, one could show that equation (2) is a quite good approximation when $f(T) \leqslant 10\%$. This is because the radii of most particles in this case are comparable to and/or less than $\lambda(0)$, so that the Meissner fraction contributed from each particle is proportional to $r^2/\lambda^2(0)$ ([36]), where r is the radius of a particle. For such a simple case, one has a simple relation $\Delta f(0)/f(0) = -2\Delta\lambda(0)/\lambda(0)$. For $\Delta n_s/n_s = 0$, as shown above, one obtains $\Delta m^{**}/m^{**} = -\Delta f(0)/f(0)$. As seen from figure 1(a), $f(0)$ is less than 8% for $x = 0.06$, so one can quantitatively determine the oxygen-isotope effect on m^{**} from the above equation. If we define the exponent of the oxygen-isotope effect on m^{**} as $\beta^O = -\mathrm{d}\ln m^{**}/\mathrm{d}\ln M_O$, then we have $\beta^O = -1.9(2)$ for $x = 0.06$. For other compositions, the values of β^O determined from equation (2) may have a noticable uncertainty if the particle sizes have a wide distribution.

5. Discussion

The observed huge oxygen-isotope effect on the effective supercarrier mass m^{**} for the deeply underdoped sample ($x = 0.06$) cannot be explained by any non-phonon mediated pairing mechanisms nor by the conventional phonon-mediated mechanism. As discussed above, strong polaronic effects arising from a strong Jahn–Teller effect should be involved to account for such novel isotope effects observed in the cuprate superconductors. In the following, we will discuss the isotope effects, supercarrier mass anisotropy, in-plane penetration depth, normal-state gap, and mid-infrared (MIR) spectra of the deeply underdoped samples ($x \leqslant 0.09$) in terms of the small polaron model of superconductivity developed by Alexandrov *et al* [37–39]. This is because almost all charge carriers are of small polarons and/or bipolarons in this doping regime, as shown recently [40]. So the small polaron model should be valid in this doping regime, and indeed we find that the theory can quantitatively explain the isotope effects, supercarrier mass anisotropy, in-plane penetration depth, normal-state gap, and MIR spectra without adjustable parameters. On the other hand, the theory does not apply to the other compositions (especially for optimal doping) where there coexist Fermi-liquid and small (bi)polaronic charge carriers [40]. These two types of charge carriers are presumably present in the alternating stripes which have been shown

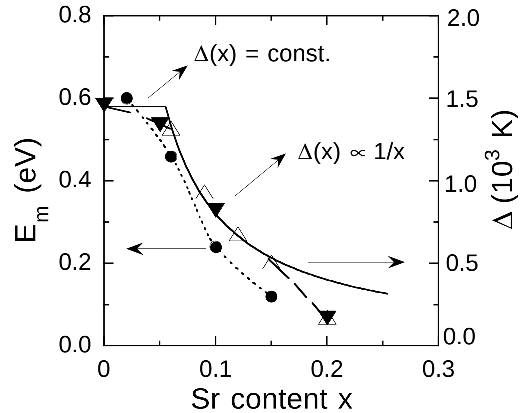

Figure 6. Composition dependences of the bipolaron binding energy Δ and $E_m \equiv 2g^2\hbar\omega$ (after [40]). The left and right scales are for the circle and triangle symbols, respectively. The bipolaron binding energy Δ precisely follows $1/x$ for $0.05 \leqslant x \leqslant 0.15$. At low doping $\Delta \simeq 0.12$ eV, which is in excellent agreement with the first-principles calculation ($\sim$0.12 eV) [37, 43].

by x-ray absorption (EXAFS), neutron scattering, and other techniques [41]. That is why most experimental results reported in literature (mainly for optimally doped samples) are not consistent with the small (bi)polaron theory of superconductivity, and this theory has not received much attention so far.

For a Jahn–Teller system, the characteristic electron–phonon coupling potential E_p should be close to the Jahn–Teller stabilization energy E_{JT}. In the La_2CuO_4 system, $E_{JT} \simeq 1.2$ eV, as estimated from the Jahn–Teller distortion [42]. It has been shown that [39] small polarons are stable if $E_p > t\sqrt{2z}/2$ (where t is the bare hopping integral and z is the number of nearest neighbours). For cuprates, $t \equiv t_{ab} \simeq 0.6$ eV (where t_{ab} is the bare hopping integral of a single particle in the planes), and $z = 4$, leading to $t\sqrt{2z}/2 = 0.85$ eV $< E_{JT}$. This implies that small polaronic states are stable in deeply underdoped cuprates. The effective mass of polarons is roughly enhanced by a factor $\exp(g^2)$, where $g^2 = \Gamma E_p/\hbar\omega$, and Γ is a constant which is less than 1 and depends on E_p/t. The high-temperature MIR spectrum of $La_{1.98}Sr_{0.02}CuO_4$ shows a maximum optical conductivity at $E_m \simeq 0.6$ eV, which corresponds to $2g^2\hbar\omega$ according to the small polaron model [39]). This leads to $\Gamma E_{JT} \simeq 0.3$ eV, and thus $g^2 = 5$ with $\hbar\omega = 0.06$ eV [37]. In addition, Alexandrov *et al* [38, 39] considered a semiclassic screening between charge carriers, and showed that interaction energies (e.g., E_p and bipolaron binding energy Δ) should be independent of the hole doping for $x \leqslant 0.05$ where the screening radius is larger than the lattice constant, but is proportional to $1/x$ for $0.05 \leqslant x \leqslant 0.15$, and decreases faster than $1/x$ for $x > 0.15$. As seen from figure 6, Δ is indeed proportional to $1/x$ for $0.05 \leqslant x \leqslant 0.15$. This is possible only if the local carrier density in the small (bi)polaronic stripes is proportional to x [40]. Since the values of g^2 are intermediate in the superconducting regime of $La_{2-x}Sr_xCuO_4$ ($1 < g^2 < 5$ for $0.05 < x < 0.15$), the polaronic carriers are not so heavy. That is why high-temperature superconductivity can be realized in cuprates with a large attractive force but an intermediate carrier mass.

Now we turn to theoretical calculations of the penetration depth, isotope effect, and supercarrier mass anisotropy for $x \leqslant 0.09$ on the basis of the small (bi)polaron theory of superconductivity. In this doping regime, almost all the charge carriers are of small polarons

[40], justifying the validity of this theory. On the basis of this theory, the effective hopping integral for intersite bipolarons along the CuO_2 planes is given by [37]

$$t_{ab}^{**} = At_{ab}\exp(-g^2) \tag{3}$$

where A is a numerical constant of the order of 1 (e.g., $A = 0.25$ for the intersite bipolaron consisting of one hole from the apical oxygen and another from the in-plane oxygen [37]). However, along the c-axis, the hopping integral is [38]

$$t_c^{**} = 2t_c^2\sqrt{\frac{2\pi}{\hbar\omega\Delta}}\exp\left[-2g^2 - \frac{\Delta}{\hbar\omega}\left(1+\ln\frac{2g^2\hbar\omega}{\Delta}\right)\right] \tag{4}$$

where t_c is the bare hopping integral of a single particle along the c-axis, Δ is the bipolaron binding energy. The above equations indicate that the effective mass of bipolarons along the CuO_2 planes m_{ab}^{**} are the same order as that for a single polaron, but the effective mass along the c-axis m_c^{**} is strongly enhanced. Since $m_{ab}^{**} = \hbar^2/2a^2t_{ab}^{**}$ and $m_c^{**} = \hbar^2/2d^2t_c^{**}$ (where a is the planar lattice constant, d is the interlayer distance), then the supercarrier mass anisotropy $\gamma^2 \equiv m_c^{**}/m_{ab}^{**}$ is given by

$$\gamma^2 = \frac{\gamma_b^4 d^2}{32At_{ab}a^2}\sqrt{\frac{\hbar\omega\Delta}{2\pi}}\exp\left[g^2 + \frac{\Delta}{\hbar\omega}\left(1+\ln\frac{2g^2\hbar\omega}{\Delta}\right)\right] \tag{5}$$

where $\gamma_b = \sqrt{m_c/m_{ab}}$ is the bare carrier mass anisotropy. The in-plane penetration depth $\lambda_{ab}(0)$ for $x \leqslant 0.09$ can be also determined from equation (3),

$$\lambda_{ab}(0) = \sqrt{\frac{d\hbar^2c^2\exp(g^2)}{16\pi e^2 xAt_{ab}}} \tag{6}$$

where c is the speed of light. Substituting equation (6) into (5) and using $E_m = 2g^2\hbar\omega$, one obtains

$$\gamma^2 = \frac{x\pi e^2\lambda_{ab}^2(0)\gamma_b^4 d}{2(\hbar ca)^2}\sqrt{\frac{\hbar\omega\Delta}{2\pi}}\exp\left[\frac{\Delta}{\hbar\omega}\left(1+\ln\frac{E_m}{\Delta}\right)\right]. \tag{7}$$

Equation (7) indicates that the supercarrier mass anisotropy strongly depends on the binding energy of bipolarons which is associated with the normal-state gap observed in underdoped cuprates.

We also note that there are two empirical relations for HTSC. One is the parabolic relation, $\bar{T}_c = 2/\bar{\lambda}^2(1 - 1/(2\bar{\lambda}^2))$, where $\bar{T}_c = T_c/T_c^m$, and $\bar{\lambda} = \lambda_{ab}(0)/\lambda_{ab}^m(0)$ (where T_c^m and $\lambda_{ab}^m(0)$ are the values at optimal doping) [44]. This relation is a consequence of the three-dimensional xy critical universility for extremely type-II superconductors [44]. Another empirical relation is $\gamma(x) \propto 1/(x - x_u)$ as shown in figure 7 [45, 46]. This dependence is based on the assumption that γ diverges at the lower critical composition x_u, where superconductivity vanishes, and the system behaves like a two-dimensional (2D) superconductor [45]. Both empirical relations work well for the mercury-based, one-layer cuprate superconductors $HgBa_2CuO_{4+x/2}$ (see figure 7 and [46]). For LSCO, $T_c^m = 38.3$ K and $\lambda_{ab}^m(0) = 2300$ Å [47]. With $T_c = 27.5$ K for $x = 0.09$, one finds from the empirical relation that $\lambda_{ab}(0) = 3400$ Å, which is in good agreement with the measured value $\lambda_{ab}(0) = 3700$ Å [15]. With $x_u = 0.05$, $\gamma = 15$ for $x = 0.15$ [24], one obtains from the relation $\gamma(x) \propto 1/(x - x_u)$ that $\gamma = 50$ for $x = 0.08$, which is in excellent agreement with the measured value $\gamma = 50$–56 [24, 48]. Therefore, the two empirical relations also work well for the LSCO system. This enables us to obtain $\lambda_{ab}(0) = 7000$ Å, $\gamma = 150$ for $x = 0.06$, and $\gamma = 37.5$ for $x = 0.09$.

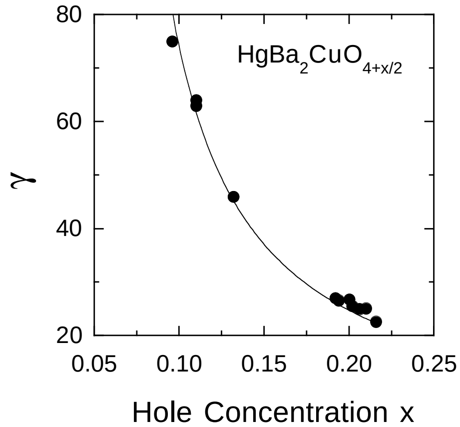

Figure 7. The supercarrier mass anisotropy γ against the hole concentration in the one-layer cuprate superconductors $HgBa_2CuO_{4+x/2}$ (after [46]). The full curve represents one-parameter fitting: $\gamma(x) = 3.7/(x - 0.05)$ with $x_u = 0.05$ being the same as for $La_{2-x}Sr_xCuO_4$.

Now we can check the validity of equation (7) where all the quantities are measurable. For $x = 0.06$, one has $\hbar\omega = 0.06$ eV [37], $E_m = 0.44$ eV [33], $\Delta = 1320$ K [40], $\gamma_b = 5.3$ [49] and $\lambda_{ab}(0) = 7000$ Å. Substituting these values into equation (7), we obtain $\gamma = 130$, which is in good agreement with the value ($\sim$150) deduced from the empirical relation above. For $x = 0.09$, E_m should be reduced by a factor of $0.09/0.06 = 1.5$ relative to that for $x = 0.06$ (since $E_m \propto 1/x$), so $E_m = 0.29$ eV for $x = 0.09$, which is consistent with the measured value of 0.24 eV for $x = 0.10$ [33]. With $\Delta = 930$ K [40], $\lambda_{ab}(0) = 3400$ Å for $x = 0.09$, we calculate from equation (7) that $\gamma = 34$, which is in excellent agreement with the deduced value (37.5) from the empirical relation. Since all the calculations do not include any adjustable parameters, the excellent agreement between the calculated and experimental results indicates that the small polaron theory of superconductivity is valid for deeply underdoped cuprates.

The next step is to show whether the same theory can also explain the observed isotope effects. Using equations (3) and (4), and the relations $(m^{**})^3 = (m_{ab}^{**})^2 m_c^{**}$ and $g^2 = E_m/2\hbar\omega$, we obtain the expressions for the total exponents of the isotope effects on m_{ab}^{**} and m^{**}, respectively

$$\beta_{ab} = -\mathrm{d}\ln m_{ab}^{**}/\mathrm{d}\ln M = -\frac{1}{4}E_m/\hbar\omega \tag{8}$$

$$\beta = -\frac{1}{3}E_m/\hbar\omega - \frac{1}{6}\left[\frac{\Delta}{\hbar\omega}\left(1 + \ln\frac{E_m}{\Delta}\right) - 0.5\right]. \tag{9}$$

For $x = 0.06$ which is near the critical composition $x_u = 0.05$, T_c should be proportional to $n_s d/m_{ab}^{**}$, where d is the interlayer distance. This simple relation is expected for a quasi-2D superconductor where γ is huge [45]. From this relation, one has $\beta_{ab}^O = -\mathrm{d}\ln m_{ab}^{**}/\mathrm{d}\ln M_O = \mathrm{d}\ln T_c/\mathrm{d}\ln M_O = -\alpha^O$. From the measured value of $\alpha^O = 1.05$ for $x = 0.06$, one yields $\beta_{ab}^O = -1.05$ for $x = 0.06$. Substituting $E_m = 0.44$ eV and $\hbar\omega = 0.06$ eV into equation (8), we have the total exponent $\beta_{ab} = -1.8$ for $x = 0.06$. Thus, the oxygen contributes $\beta_{ab}^O/\beta_{ab} = 57\%$ to the total isotope effect, which is in good agreement with the studies of the copper and oxygen-isotope shifts of T_c in this system [15]. Then the exponent of the oxygen-isotope effect on m^{**} is $\beta^O = 0.57\beta$. Using equation (9), we calculate $\beta = -3.1$, and so $\beta^O = -1.8$, which is in excellent agreement with the measured value ($\beta^O = -1.9(2)$). Thus, this theory can quantitatively explain the oxygen-isotope effects on both T_c and the effective supercarrier mass without any adjustable parameters.

6. Summary

In summary, magnetization measurements have been performed on the oxygen-isotope exchanged samples (^{16}O and ^{18}O) of the one-layer cuprate superconductors $La_{2-x}Sr_xCuO_4$ ($0.06 \leqslant x \leqslant 0.15$). From magnetization measurements on the fine-grained and decoupled samples in the Meissner state, we find substantial oxygen-isotope effects on both the penetration depth $\lambda(0)$ and T_c. From normal-state susceptibility measurements, we are able to show that there is a negligible oxygen-isotope effect on the carrier density n. The combined results strongly suggest that there is an oxygen-isotope effect on the effective supercarrier mass m^{**}, which is huge for $x = 0.06$, and reduced to a smaller value for $x = 0.15$. We discuss the isotope effects, supercarrier mass anisotropy, normal-state gap, in-plane penetration depth, and MIR spectra for $x \leqslant 0.09$ on the basis of the small polaron theory of superconductivity. We find that the agreement between the calculated and experimental results is excellent without adjustable parameters. The present experimental and theoretical investigations will provide important insight into the pairing mechanism of high-temperature superconductors.

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