Anisotropy of the penetration depth in $La_{2-x}Sr_xCuO_4$ in underdoped and overdoped regions

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Abstract. We present the results of measurements of the penetration depth anisotropy in pulverized, ceramic $La_{2-x}Sr_xCuO_4$. The measurements were carried out for $x = 0.08$, 0.1, 0.125, 0.15 and 0.2. The powdered samples, immersed in wax, were magnetically oriented in a static magnetic field of 10 T. The penetration depth in the a–b plane, λ_{ab}, and perpendicular to it, $\lambda_\perp$, were derived from alternating-current susceptibility measurements. For underdoped samples they both vary linearly with temperature (for the low-temperature region), while for the samples from the overdoped region the measured points can be fitted by an exponential function. These results support Uemura's picture (Uemura Y J 1997 *Physica* C **282–287** 194) of crossover from Bose–Einstein condensation to a Bardeen–Cooper–Schrieffer mechanism of superconductivity. The penetration depth values extrapolated to $T = 0$ may be described by a quadratic function of the strontium concentration (for both λ_{ab} and $\lambda_\perp$). The anisotropy of the penetration depth as a function of the substitution shows a similar dependence to the critical temperature $T_c(x)$.

1. Introduction

For conventional, low-temperature superconductors the phonon mechanism is responsible for the existence of Cooper pairs. According to BCS theory [2], in this case the gap function is isotropic and the pairing is called s-wave type. It is already well established that for high-temperature superconductors (HTSC) also, Cooper pairs are responsible for carrying the charge [3]. But up to now there has remained controversy regarding whether the symmetry of the gap function is isotropic [4–7] or anisotropic [8,9]. In some experiments, mixed (s + d)-type symmetry was also observed [10]. Anisotropic pairing with the nodes on the Fermi surface should lead to power-law dependence of various thermodynamic functions [11].

One such function, whose temperature dependence may reflect the symmetry of the order parameter, is the magnetic penetration depth. It has been studied, employing different methods, for different families of high-temperature superconductors [12–15].

Although the lanthanum-based 214-type family may be treated as a model for HTSC, there are only few papers dealing with penetration depth studies, and these are mainly concerned with the in-plane component λ_{ab} [16–18].

We are aware of only one paper in which a study of the penetration depth tensor for $La_{2-x}Sr_xCuO_4$ was presented [19]. The authors applied a surface impedance method to single crystals with typical dimensions equal to $1 \times 1 \times 0.2$ mm^3, with the large faces parallel or perpendicular to the a–b planes. They found that the anisotropy of the penetration depth

decreases with the increase of strontium content from about 25 for the strontium content equal to $x = 0.09$ to about 10 for $x = 0.19$. The magnitude of $\lambda_c(0)$ was in agreement with that obtained from the Josephson-coupled-layer model and the temperature dependence of λ_c was different from the local-limit BCS formula.

Because of limited resolution of their apparatus, the authors of [19] were not able to present the penetration depth behaviour for low temperatures, so they could not establish whether this dependence may be described by an exponential or a power law. This was the main reason for which we decided to present our results of measurements of the penetration depth anisotropy of $La_{2-x}Sr_xCuO_4$ obtained by an AC susceptibility method for the magnetically aligned powders.

2. Experiment

Polycrystalline $La_{2-x}Sr_xCuO_4$ (where $x = 0.08$, 0.1, 0.125, 0.15 and 0.2) samples were prepared using the standard solid-state reaction method, which has been described elsewhere [20].

X-ray data proved our samples to have single-phase K_2NiF_4-type structure. The ceramic samples were reground in an agate mill for about 100 min. The powdered samples were mixed with molten Okerin wax and magnetically aligned in a 10 T static magnetic field. The wax was cooled through the melting point whilst the magnetic field was maintained. Blocks, of dimensions $2 \times 2 \times 10$ mm^3, were cut out from the composite using a saw, with their c-axes parallel and perpendicular to the longest block dimension. Scanning electron microscope (SEM) photographs were taken of different parts of these blocks and, after digitizing them, the grain size was determined for all samples by means of a 'home-made' computer program. From these results, data histograms were made and their shape was approximated by a grain distribution function. This function was arbitrarily chosen, with the only criterion being that of producing the closest functional description of the histograms. The same distribution function was used for every sample. The parameters in this function changed even for samples cut from the same batch; this was caused mainly by some sedimentation. The procedure for preparing and characterizing the samples is described elsewhere [21].

AC measurements were carried out using a commercial Lake Shore AC susceptometer, with an amplitude of the varying magnetic field of 0.1 mT and a frequency of 111.1 Hz. In spite of the fact that from the SEM photographs, it was known that the individual grains were well separated, we made additional tests by measuring the linearity of the output voltage of the susceptometer signal as a function of the AC-field amplitude and frequency. The voltage was linear within $\pm 0.1\%$, proving that there are no links between the individual grains.

The temperature dependence of the penetration depth was derived from AC measurements by the method used earlier by Porch *et al* [22] making use of the formulae

$$\frac{\chi_i}{\chi_0} = \frac{2}{3} \frac{v_{AC}}{[V_S N - v_{AC} f(\frac{1}{3} - D)]}$$

$$\frac{\chi_i}{\chi_0} = \left[\int \left(1 - \frac{3\lambda_i}{r} \coth \frac{r}{\lambda_i} + \frac{3\lambda_i^2}{r^2} \right) r^3 g(r) \, dr \right] \Big/ \left(\int r^3 g(r) \, dr \right)$$

where χ_i is the measured susceptibility in the a–b plane or perpendicular to it, χ_0 is the susceptibility of a perfectly diamagnetic spherical grain, v_{AC} is the measured AC voltage (after subtraction the signal from the empty holder), V_S is the volume of the superconductor, N is the calibration factor of the apparatus used, f is the volume of superconductor divided by the total volume of the composite, D is the demagnetizing factor of a grain, r is the radius of a grain and $g(r)$ is the measured grain size distribution function.

Employing the method described above, we were able to evaluate the penetration depth in the *a–b* plane, which we describe as 'in plane' and call λ_{ab}, and the one which is the mean value for all crystallographic directions, which we will describe as 'out of plane' and call $\lambda_\perp$. When the magnetic field is perpendicular to the CuO planes, screening currents flow within these planes and the measured penetration depth is the true value of λ_{ab}. For magnetic field parallel to the CuO planes, screening currents flow in some places within the planes and in some places perpendicularly to them, so the measured penetration depth is some complicated mean of λ_{ab} and λ_c—whose true magnitude cannot be easily derived from the measurements. To avoid forgetting this, we called this measured value $\lambda_\perp$.

3. Results and discussion

Great care was taken to ensure that the whole sample volume consists of properly aligned grains. Ball milling rather than grinding was employed to obtain powder with single-domain grains, with shapes close to spherical. Powdering ceramic samples with an agate mortar results in the grains being shaped mainly as platelets.

The quality of the alignment of the powders may be easily inferred from the x-ray diffraction patterns. They are depicted in figure 1. As can be seen, almost all of the reflections present are 00*l* reflections and their intensities are about 40 times higher than for non-oriented samples. Traces of reflections other than 00*l* ones result from the volume of non-aligned material. To evaluate the percentage of non-aligned grains, we have applied the procedure described in reference [23] and found that in all of our samples the amount of non-oriented material was less than 15%. The unit-cell parameters were used to calculate the density of superconducting material and these are presented in table 1. They are within the ranges of values presented by other authors [24]. The volumes of the unit cells, together with the results of density measurements for powders immersed in wax, were used to calculate the volume of
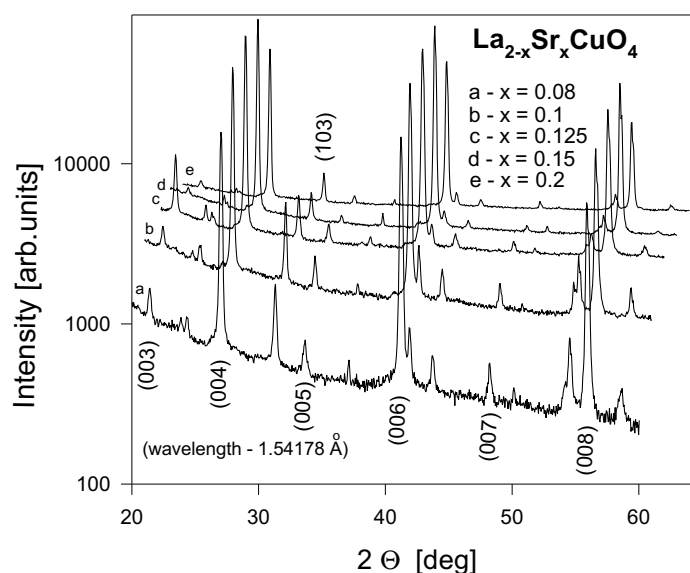

Figure 1. X-ray diffraction patterns for aligned powders of La$_{2-x}$Sr$_x$CuO$_4$ for different strontium contents *x* (successive curves are shifted by one degree along the horizontal axis and 1000 arbitrary units along the vertical axis for clarity).

Table 1. Critical temperatures, lattice constants, unit-cell volumes and penetration depths of the compounds studied.

Compound	T_c (K)	a, b (nm)	c (nm)	V (nm^3)	$\lambda_{ab}(0)$ (nm)	$\lambda_{\perp}(0)$ (nm)
$La_{1.92}Sr_{0.08}CuO_4$	26.5	0.37728	1.31634	0.187373	327	431
$La_{1.9}Sr_{0.1}CuO_4$	36.9	0.37769	1.32021	0.188331	304	449
$La_{1.875}Sr_{0.125}CuO_4$	34.8	0.37832	1.32321	0.189383	563	693
$La_{1.85}Sr_{0.15}CuO_4$	39.5	0.37949	1.32143	0.190302	234	547
$La_{1.8}Sr_{0.2}CuO_4$	33.9	0.37696	1.32241	0.187914	252	497

superconducting material in the measured samples.

The distribution of the grains was obtained from SEM photographs of different parts of the samples. The histograms obtained were fitted using the same function for all samples. One of the histograms, together with the distribution function used, is presented in figure 2 as an example. The diameter of the grains varied between 0.5 and 10 μm, with an average of about 1 μm for all samples. Although, from the SEM pictures, it was known that there are some grains with the shapes of platelets and needles, for evaluation of the penetration depth the demagnetizing factor for a sphere was used in the calculation.
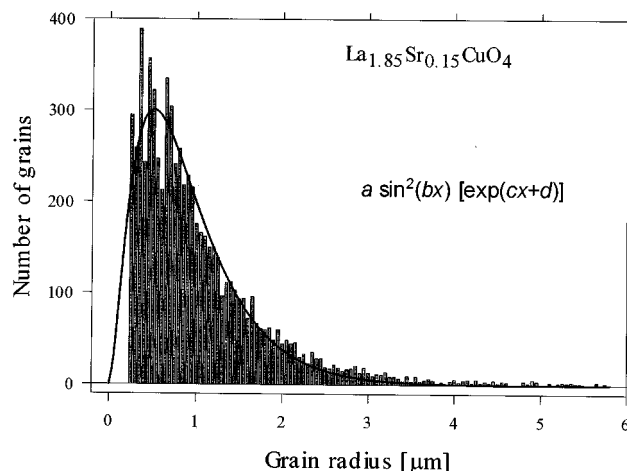
Figure 2. An example of a histogram and the grain size distribution function for $La_{2-x}Sr_xCuO_4$.

We should add that the log–normal plot usually used to approximate the shape of the powder grain size distribution gave a much worse approximation than the function used by us (especially for small grain size).

Temperature dependences of the penetration depths derived from the AC susceptibility measurements for the samples from the underdoped and optimally doped regions (excluding the samples with the $x = 0.125$ composition) are presented in figure 3(a). This dependence is linear below about $0.5T_c$ for both λ_{ab} and $\lambda_{\perp}$ for all of the samples, as is expected for the material with nodes on the Fermi surface. Such linear behaviour was already observed for LaSrCuO by the muon-spin-rotation technique [16]. It was also observed for other high-temperature superconductors [5, 13].
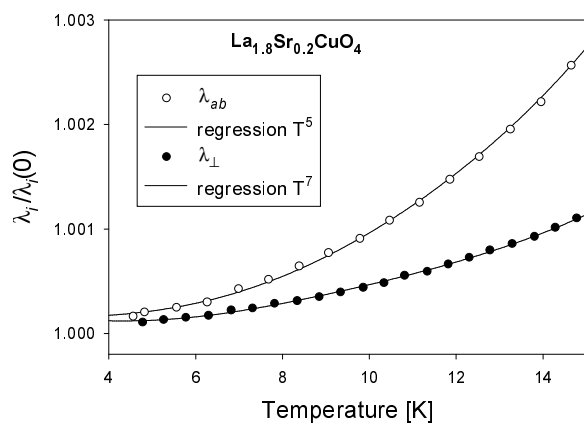
For the samples with strontium content equal to $x = 0.125$, linear behaviour is not observed down to the lowest temperature used—i.e. about 4 K. The low-temperature behaviour of both
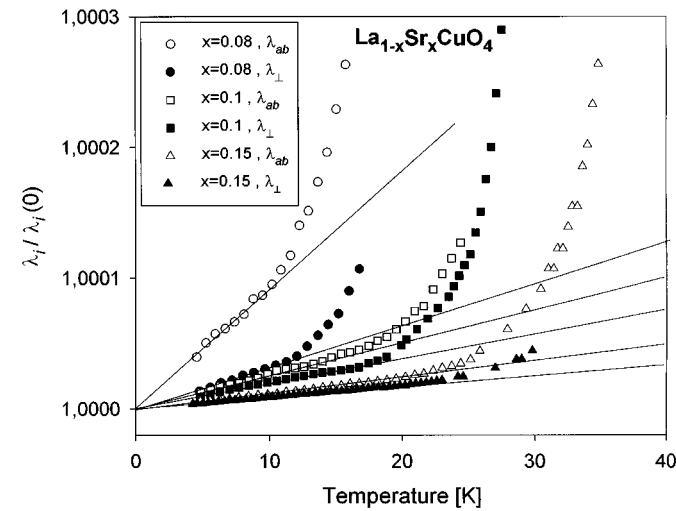
(a)
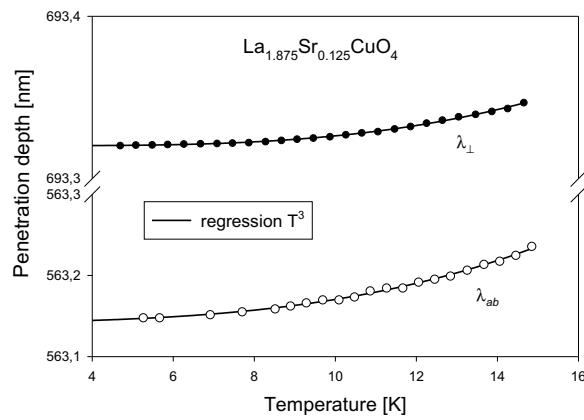
(b)

(c)

Figure 3. The temperature dependence of the reduced penetration depth of La$_{2-x}$Sr$_x$CuO$_4$ for: (a) $x = 0.08$, 0.1 and 0.15; (b) $x = 0.125$; and (c) $x = 0.2$. For (b) and (c), polynomial fits to the measured points are also presented.

the in-plane and the out-of-plane penetration depths can be satisfactorily fitted by a polynomial of the third order. This is depicted in figure 3(b).

Also, for the samples from the overdoped region, a linear temperature dependence of the penetration depth is not observed down to a temperature of about 4 K. The in-plane component of the penetration depth can be satisfactorily described by a fifth-order polynomial and the out-of-plane component by a seventh-order one (figure 3(c)). Nevertheless, it should be admitted that fitting with polynomials of such high orders differs only marginally from fitting to exponential behaviour. Therefore the samples from the overdoped region may be showing BCS-type behaviour.

So whereas for underdoped and optimally doped samples the existence of d-wave pairing is supported by our measurements, BCS-type behaviour is observed instead for the samples from the overdoped region. Such behaviour agrees with expectations resulting from the picture of crossover from Bose–Einstein to BCS behaviour suggested by Uemura [1].

Using the polynomials described above, we have obtained the penetration depth extrapolated to zero temperature. Its dependence on the strontium concentration is presented in figure 4. The values obtained for $\lambda_{ab}(0)$ are similar to those presented by Locquet *et al* [17]. There are a few features of these dependencies which are not obvious.
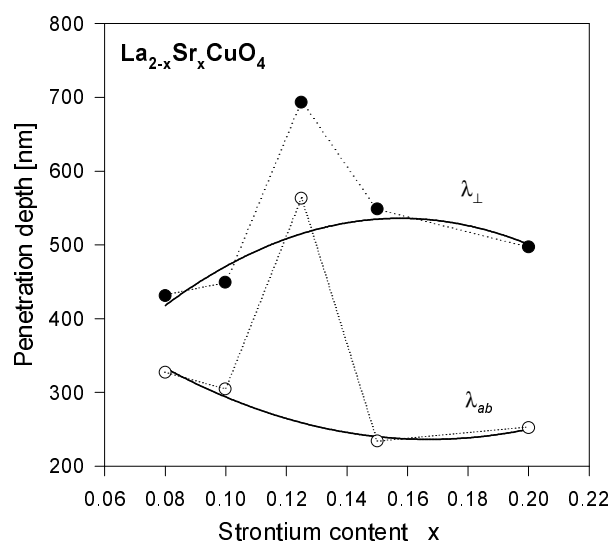

Figure 4. Penetration depth values as functions of the strontium content extrapolated to zero temperature (solid lines are quadratic fits to the measured points—except for $x = 0.125$—and are guides to the eye only).

First, both in-plane and out-of-plane penetration depths for the samples with the composition $La_{1.875}Sr_{0.125}CuO_4$ are much greater than for other compositions, reflecting the so-called 1/8 problem, i.e. the fact that just for this composition the critical temperature is suppressed. This is probably connected both with the low-temperature structural phase transition and with static order of the strip correlations of holes and spins discovered by Tranquada *et al* [25]. In the case of the existence of a regular pattern of normal and superconducting strips, it is obvious that the effective penetration depth is increased. The fact that λ_{ab} increases more strongly than $\lambda_\perp$ supports our view that the part of the evaluated $\lambda_\perp$ which is connected with the screening currents flowing within the CuO planes is rather meaningful.

For other samples the in-plane penetration depth decreases with the increase of the carrier

number, but for the overdoped region it starts to increase again. Such an effect was first reported by Locquet *et al* [17], and showed that in the overdoped state, despite the increase in the number of charge carriers, the density of superconducting carriers may decrease or their effective mass may increase or both. Such an increase of the effective mass of the carriers was postulated by us [21] to explain the results of our measurements of the penetration depth of zinc- and nickel-substituted La$_{1.85}$Sr$_{0.15}$CuO$_4$.

The out-of-plane penetration depth increases with the strontium content in the underdoped range and slightly decreases in the overdoped one. Also, for this dependence the value of $\lambda_\perp(0)$ is much greater for the sample with the strontium content equal to 1/8.

The dependencies of the penetration depth on the strontium doping, extrapolated to zero temperature, are fairly well described by the quadratic functions (of course, with the exception of that for $x = 0.125$).

The dependence of the critical temperature on the strontium concentration is presented in figure 5, together with the corresponding dependence of the anisotropy of the penetration depth, defined as $\lambda_\perp(0)/\lambda_{ab}(0)$. It is seen that the dependencies of the critical temperature and the penetration depth anisotropy follow similar trends. It is well established that the critical temperature T_c is proportional to the density of superconducting carriers and inversely proportional to their effective mass, i.e. $\propto n_S/m^*$ in the underdoped region [12, 26] (this is the so-called Uemura plot). For overdoped material this proportionality breaks down, which may imply that, in addition to the increase in the density of carriers with doping, there is also an increase of their effective masses in this region. We should recall that the Uemura plot was constructed from the results obtained by the positive-muon-rotation technique, which probes only superconducting a–b planes, an so is responsible for λ_{ab} only. The increase of the effective mass of the carriers within the a–b planes is equivalent to the decrease of the penetration depth anisotropy (if defined as we have, i.e. as $\lambda_\perp(0)/\lambda_{ab}(0)$). As for the overdoping region the critical temperature also diminishes, it is not surprising that T_c and $\lambda_\perp(0)/\lambda_{ab}(0)$ have similar doping dependences.
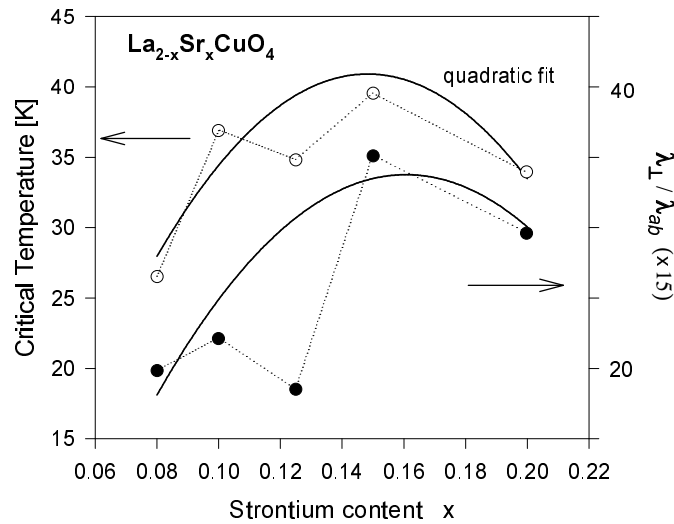

Figure 5. The dependences of the critical temperature (left-hand scale) and anisotropy of the penetration depth (multiplied by a factor of 15—right-hand scale) on the strontium content (solid lines are quadratic fits to the measured points—except for $x = 0.125$—and are guides to the eye only).

The anisotropy of the penetration depth is highest for optimally doped samples and decreases for underdoped and overdoped branches of the dependence. This is consistent with the crossover from the Bose–Einstein to the BCS picture proposed by Uemura [1]. The increase of the anisotropy with doping (there is no Bose–Einstein condensation (BEC) in purely 2D systems) decreases the temperature at which the BEC occurs and drives it close to that obtained from the BCS theory just for the optimally doped samples.

For the strontium composition equal to 1/8, the anisotropy is the smallest among those of all of the samples. Apparently, for $La_{1.875}Sr_{0.125}CuO_4$ the deviation from the optimal composition is connected with the increasing level of isotropization of the penetration depth in the material.

The first results of measurements of the penetration depth anisotropy for $La_{2-x}Sr_xCuO_4$ were presented by Shibauchi *et al* [19]. They employed the surface impedance method for measurements on small single crystals with typical dimensions $1 \times 1 \times 0.2$ mm^3, having the large faces parallel and perpendicular to the *a–b* planes (cut from bigger crystals). The critical temperatures of their samples were lower than those previously published for crystals used by them [27] and known from other papers [18]. They did not find the traces of critical-temperature suppression for their sample with the composition $x = 0.12$ which were usually found by other authors [11, 18, 28].

The values of the in-plane penetration depth obtained by Shibauchi *et al* [19] are very similar to those discussed in our paper. They found that the temperature dependence of λ_{ab} is roughly explained by the local, clean-limit BCS theory, while, in contrast, the λ_c-behaviour is consistent with the Josephson-coupled-layer model. The anisotropy measured by them was a decreasing function of the strontium content, and changed from about 25 for the strontium content equal to $x = 0.09$ to about 10 for $x = 0.19$.

Our results for the out-of-plane penetration depth $\lambda_\perp(0)$ are lower than those obtained by Shibauchi *et al* for $\lambda_c(0)$. We are not convinced that the contributions of λ_{ab} and λ_c to the measured out-of-plane penetration depth $\lambda_\perp$ are equal, as suggested by Shibauchi *et al* [19]. This assumption may be one of the sources of discrepancies between our results. Another source may lie in the fact that the shape of the single crystals of Schibauchi *et al* enhanced the demagnetization factor; this would be very difficult to assess, especially for the case where the larger faces of the samples were parallel to the magnetic field direction.

The anisotropy of the resistivity measured by Kimura *et al* [27] for the samples used in the study by Shibauchi *et al* was much higher than that of the penetration depth and was equal to about 4000 for strontium content $x = 0.06$ and about 160 for $x = 0.266$. So the results obtained by Shibauchi *et al* for the penetration depth anisotropy are also unexpectedly low. We think that the reasons for this are similar in their case and ours. The difference between our results may be caused by the different kinds of sample used and the different methods employed to study the penetration depth behaviour.

A much lower value of the anisotropy of the penetration depth in $La_{1.85}Sr_{0.15}CuO_4$ was also obtained by Uchida *et al* [18]—i.e. about 2–5 times lower. But those authors obtained their results by comparing the out-of-plane penetration depth derived from optical reflectivity spectra with the in-plane values measured from the positive-muon-relaxation rate by Uemura *et al* [29]. In their case, the anisotropy was also a decreasing function of the doping.

Although our values of $\lambda_\perp$ and the values of λ_c derived by Shibauchi *et al* were different, their temperature and composition dependences should be fairly similar. But we definitely did not observe BCS-type behaviour of the temperature dependence of the in-plane penetration depth—not only for optimally doped material but also for all of the underdoped ones (with the exception of that with $x = 0.125$). The anisotropy of the penetration depth measured by us is also not a decreasing function of the strontium content as had been found by Shibauchi

et al [19], but has the shape of the dependence of the critical temperature on the strontium concentration. This behaviour in fact constituted one of the reasons for carrying out the present study and presenting its results.

From the results presented above and from those of our previous paper [21], we can state that for the underdoped state, the penetration depth is connected with changes of the superconducting charge-carrier density, while for the overdoped region, interaction between the charge carriers, i.e. their effective masses, should also be taken into account.

4. Summary

In summary, we have evaluated the penetration depth in $La_{2-x}Sr_xCuO_4$ for $x = 0.08$, 0.1, 0.125, 0.15 and 0.2 from AC susceptibility measurements of magnetically aligned single-crystalline powders. We found that the temperature dependences of the in-plane and out-of-plane penetration depths are both linear functions for samples from the underdoped region. The exceptions are the samples with $x = 0.125$; their dependences of the penetration depth on the temperature may be described by a polynomial of third order. For overdoped samples, exponential behaviour of both $\lambda_{ab}(T)$ and $\lambda_\perp(T)$ is observed. Such behaviour was expected from the Uemura model [1] of crossover from Bose–Einstein condensation to a BCS-type mechanism of superconductivity. The penetration depth extrapolated to zero temperature may be described by a simple quadratic function of the strontium content, except for the concentration $x = 0.125$. Samples with this composition behave as if made from different material. The dependence of the anisotropy of the penetration depth has the same form as the dependence of the critical temperature on the strontium concentration.

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