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## Doping dependence of the superconducting gap by Andreev reflection in Au/La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> point-contact junctions

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### Abstract

We studied the doping dependence of the superconducting gap in La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> (LSCO) by means of Andreev reflection measurements in Au/LSCO point-contact junctions. The Andreev reflection features were found to disappear at  $T_c^A$  close to the bulk  $T_c$ . The fit of the conductance curves with the BTK–Tanaka–Kashiwaya model gives good results if a (s + d)-wave gap symmetry is used. The low-temperature dominant isotropic gap component  $\Delta_s$  follows very well the  $T_c$  vs.  $x$  curve, while the gap-like features observed by angle-resolved photoemission spectroscopy and tunneling scale with  $T^*$ . This confirms the different origin of these two energy scales at  $T < T_c$ . © 2002 Published by Elsevier Science Ltd.

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Within the scenario proposed two years ago by Deutscher [1] two distinct energy gaps exist in HTSC. The first,  $\Delta_c$ , is related to the onset of superconductivity, i.e. to the formation of a phase-coherent condensate. Hence, this gap is detected by experimental tools sensitive to the phase coherence, such as Andreev reflection and Josephson tunneling. The second energy scale,  $\Delta_p$ , is the single-particle excitation energy observed by angle-resolved photoemission spectroscopy (ARPES) and tunneling. There are several indications that these two energy scales almost coincide in the overdoped regime, as they do in conventional superconductors, while in underdoped samples  $\Delta_p$  can be much greater than  $\Delta_c$  and is also observed above  $T_c$ , where it coincides with the pseudogap. The absence of any dramatic change in the gap amplitude measured by tunneling and ARPES when  $T_c$  is crossed suggests that, at least in underdoped samples,  $\Delta_p$  has no relation with  $T_c$ . Therefore, in a certain sense,  $\Delta_c$  can be thought of as the true superconducting gap, even though whether the pseudogap is a precursor of superconductivity or has a different (i.e. magnetic) origin is still a matter of debate.

In the particular case of La<sub>2-x</sub>Sr<sub>x</sub>CuO<sub>4</sub> (LSCO), few

experiments have been performed to investigate the Andreev gap [2–4], while some tunneling and ARPES evidences exist supporting a monotonical increase in the low-temperature gap amplitude at the decrease of doping [5–7].

In this paper we present a thorough study of the doping dependence of the superconducting (phase coherence) gap in LSCO extracted from the conductance vs. voltage curves of point-contact junctions between Au tips and LSCO polycrystalline samples with six different Sr concentrations ranging from strongly underdoped ( $x = 0.08$ ) to slightly overdoped ( $x = 0.20$ ).

Details of the sample preparation are given elsewhere [8, 9]. The good quality of the LSCO samples was evidenced by XRD powder diffraction [10] and EDS microprobe analysis. AC susceptibility and resistivity measurements gave bulk critical temperatures in good agreement with the standard curve of  $T_c$  vs.  $x$  for LSCO [11]. The Au tip was obtained by electro-chemical etching with HNO<sub>3</sub> + HCl of a 0.2 mm diameter Au wire.

Fig. 1(a) and (b) show some representative low-temperature  $I$ – $V$  characteristics (solid lines) obtained in samples with  $x = 0.08$  and 0.12, together with the  $dI/dV$  vs.  $V$  curves (dashed lines).

Fig. 2 shows an example of the low-temperature conductance curves (open circles) for the six values of  $x$

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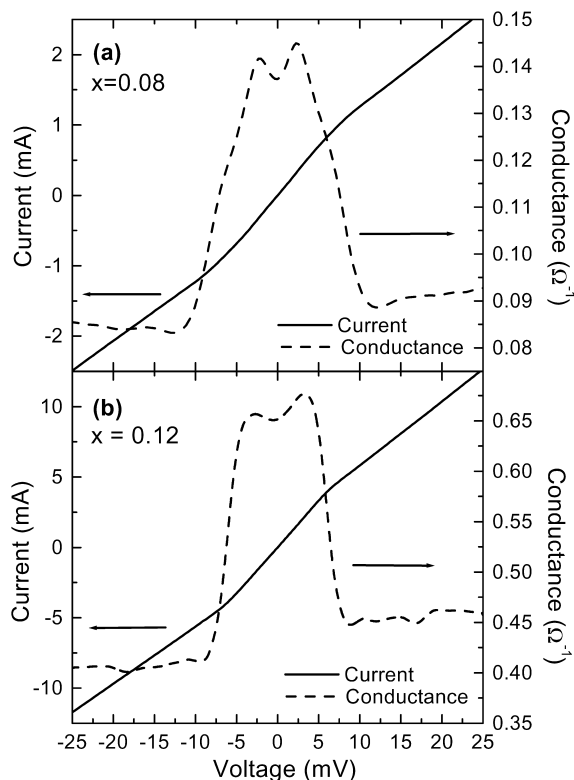


Fig. 1. (a) An example of the  $I$ - $V$  characteristics of a point-contact junction between the Au tip and a LSCO sample with  $x = 0.08$  (solid line), together with the relevant  $dI/dV$  vs.  $V$  curve (dashed line). (b) The same as (a) but in the case of  $x = 0.12$ .

considered here, normalized so that  $dI/dV = 1$  in the normal state, and vertically shifted for clarity.

These curves and all the others presented in the following were actually selected among a large number of data sets. In particular, we required the absence of any voltage-dependent heating effect in the  $I$ - $V$  curve and the thermal stability of the contact (i.e. the constancy of the normal-state conductance with the increase in temperature). Moreover, comparing the conductance curves with those predicted by the BTK model [12] provides by itself a good probe for the fulfillment of the ideal measurement conditions. Our experimental curves are indeed fairly similar to the ideal BTK ones obtained with a small finite potential barrier, although their maximum value is less than that expected and the shape is not always compatible with a pure s-wave gap symmetry. The oscillations of  $dI/dV$  at  $|V| \geq 10$  mV are not 'classic' as well, but are often observed in high- $T_c$  compounds [13].

The absence of heating effects is a key point of our discussion. It is well known that point-contact measurements give reliable spectroscopic information provided the contact radius  $a$  is smaller than the mean free path  $\ell$  in both materials (ballistic regime). Since any control on the contact dimension is impossible from the experimental point of

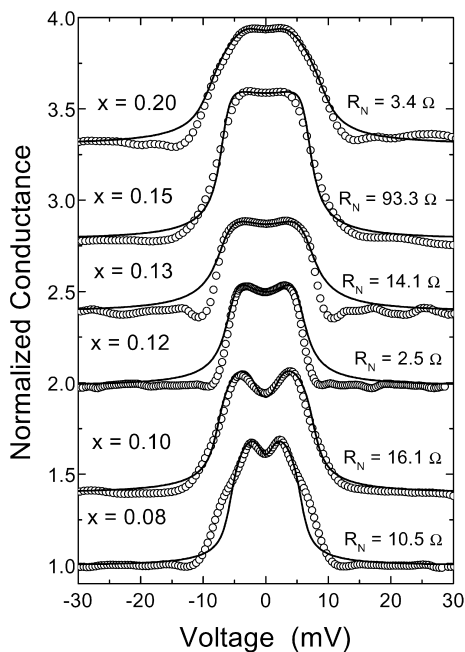


Fig. 2. The normalized conductance curves (open circles) of Au/LSCO point-contact junctions for various doping levels ( $0.08 \leq x \leq 0.2$ ) at low temperature ( $4.22 \leq T \leq 5.61$  K). The curves are vertically displaced for clarity. The solid lines represent the best-fit curves calculated by using the BTK-T-K model. The parameters of each theoretical curve are indicated in Table 1.

view, one usually evaluates  $a$  from the normal-state junction resistance  $R_N$  by supposing that a single contact is established [14,15]. In our case, the values of  $R_N$  shown in Fig. 2 give  $146 \leq a \leq 800$  Å, whereas the (evaluated) mean free path  $\ell$  ranges from 40 to 70 Å from underdoped to overdoped. These values would rather indicate that the contact is in the thermal (Maxwell) regime, characterized by strong heating phenomena. Nevertheless, in the curves we chose, the variation of the normal-state conductance with bias is very small and well within that expected in the ballistic regime [16]. The exceedingly low contact resistances are thus very likely to be due to the presence of several parallel ballistic contacts between sample and tip [17]. Anyway, there is no doubt that the features shown in Figs. 1 and 2 can only be produced by Andreev reflection at the SN interface.

Even at a first glance, the curves in Fig. 2 show that the gap amplitude increases up to a maximum and then decreases again when one moves from underdoped to overdoped samples. The simplest way of evaluating the gap is to identify its edges with the positions of the conductance maxima. The resulting gap values are shown in Fig. 3 as a function of doping, together with those measured below  $T_c$  by tunneling [5,6]. The two measures almost coincide in the overdoped region, but differ more and more when the doping is reduced.

To investigate in greater detail the doping dependence of

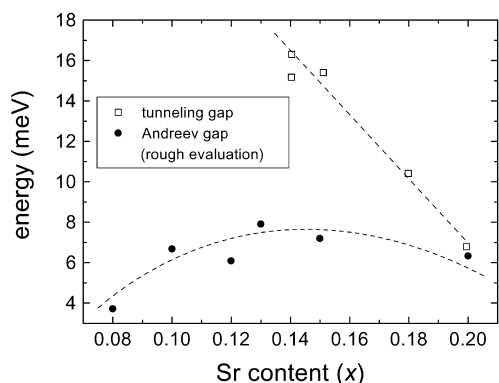


Fig. 3. Qualitative doping dependence of the superconducting Andreev gap (solid circles) roughly evaluated as described in the text. The trend of the superconducting gap is compared to that of the tunneling gap reported in literature [5,6].

the Andreev gap, we compared the experimental curves with the theoretical ones predicted by the BTK–Tanaka–Kashiwaya (BTK–TK) model [18–20], in which we also introduced the quasiparticle lifetime broadening  $\Gamma$ . We tried different possible gap symmetries: *s*, *d*, *s + d*, *s + id*, and *extended s*. If one restricts the analysis to the low-temperature data in Fig. 2, all these symmetries apart from the pure *d*-wave and the extended *s*-wave give curves which agree almost equally well with the experimental ones (for a comparison of the low-temperature fits with different symmetries, see Refs. [8,9]). One could object that this result is in contrast with the general belief that the gap symmetry in HTSC is predominantly *d*-wave, or at least has nodes. Actually, this is not demonstrated in LSCO, where many experimental evidences support instead a fully gapped symmetry. Incidentally, the fit of our conductance curves with other nodeless symmetries (such as *anisotropic s* and *d + id*) will be presented in a future paper.

Further information about which gap symmetry is the most suitable for the fit can be found in the temperature dependence of the conductance curves. An example of this dependence for three different doping contents ( $x = 0.08$ ,

0.10, and 0.20) is shown in Fig. 4(a)–(c), which actually report for clarity only some of the curves we measured.

First of all, a result worth to mention is that the Andreev features always disappear at a temperature  $T_c^A$  close to the bulk  $T_c$  or slightly lower. At  $T > T_c^A$ , the conductance curves are identical to those expected in a N–N point-contact ballistic junction [16]. The fact that Andreev reflection gives no evidence of gap above  $T_c$  confirms that it actually measures the phase-coherence gap  $\Delta_c$ .

The information on the gap symmetry we were looking for can be obtained by fitting the conductance curves in the whole temperature range (from 4.2 K up to  $T_c^A$ ) with the BTK–TK model. The fitting function is quite complicated and it would not be useful to report it here. It can be found in Refs. [18–20]. The free fitting parameters, in the general case of mixed symmetry, are: the isotropic and anisotropic gap components ( $\Delta_{is}$  and  $\Delta_{an}$ ), the parameter  $Z$  (proportional to the potential barrier height), the lifetime broadening  $\Gamma$  and the angle  $\alpha$  between the *a* axis and the normal to the S–N interface [18–20] ( $\alpha$  is unknown because our samples are polycrystalline). Actually, some constraints reduce the number of adjustable parameters. First, since  $R_N$  changes very little with  $T$ , we assumed  $Z$  to be constant and we extracted it from the fits in the various symmetries of the lowest-temperature curves. We obtained very low values of  $Z$  ( $\leq 0.3$ ), which make the choice of  $\alpha$  have very little influence on the values of  $\Delta_{is}$  and  $\Delta_{an}$  determined by the fit, independent of the symmetry used [8,9]. Therefore, we could choose  $\alpha = 0$  without loss of generality. The remaining parameters  $\Delta_{is}$ ,  $\Delta_{an}$ , and  $\Gamma$  were varied in order to fit the data, but always keeping  $\Gamma$  as small as possible.

A good agreement between the theoretical curves and the experimental data *in the whole temperature range* is only obtained if a (*s + d*)-wave gap symmetry is used [8,9]. This provides the missing information about the symmetry to be used for the fit at low temperature, and allows us to refine the rough evaluation of the doping dependence of the gap shown in Fig. 3. The (*s + d*)-wave theoretical curves which best fit the low-temperature data in Fig. 2 are shown in the same figure as solid lines. The results are consistent with

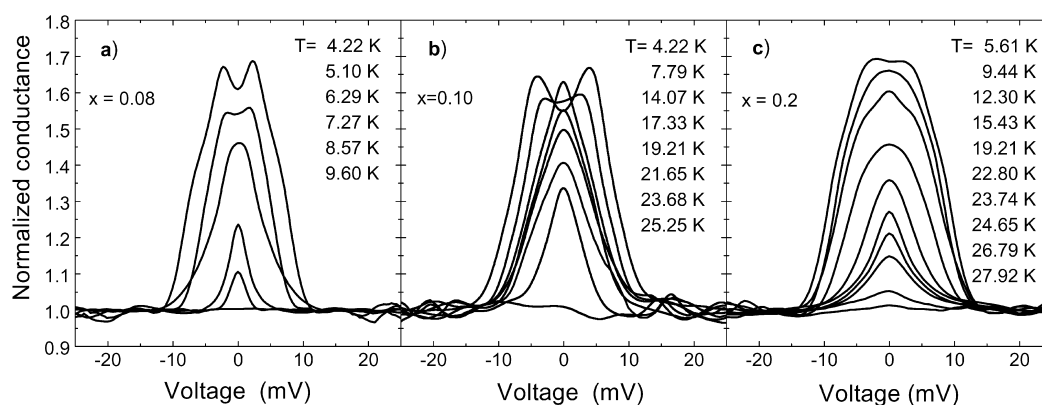


Fig. 4. Temperature dependence of the normalized Andreev conductance in LSCO samples with  $x = 0.08$  (a), 0.10 (b), and 0.20 (c).

Table 1  
Best-fit parameters and temperatures for the curves of Fig. 2

Doping	$T$ (K)	$\Delta_s$ (meV)	$\Delta_d$ (meV)	$\Gamma$ (meV)	$Z$	$T_c^A$ (K)	$2\Delta_s/k_B T_c^A$
0.08	4.22	3.4	2.5	0.19	0.20	9.6	8.2
0.10	4.22	4.8	3.1	0.27	0.23	25.3	4.4
0.12	4.22	5.6	0	0.92	0.18	26.0	5.0
0.13	4.22	6.8	0	1.50	0.17	29.1	5.4
0.15	4.65	6.8	0	0.44	0.08	35.3	4.5
0.20	5.61	6.0	3.5	1.00	0.13	27.9	5.0

those obtained in LSCO by Deutscher et al. [3]. Although the general symmetry is  $s + d$ , for some values of  $x$  the weight of the  $d$  component is zero — that is, the actual symmetry is pure  $s$ -wave (but *only* at low temperature). In all cases the  $s$ -wave component is dominant and thus is the more representative one. Also notice that it is a very robust parameter, since its value would change very little if other gap symmetries were considered. Table 1 lists all the values of the parameters related to the curves in Fig. 2.

Fig. 5 shows the doping dependence of the *low-temperature*  $\Delta_s$  (solid circles) obtained from our fit, compared to those of the gap determined by tunneling measurements (open squares) [5,6] and of the ARPES leading-edge shift (LE) [7] (open circles), both measured below  $T_c$ . The doping dependence of the isotropic gap component  $\Delta_s$  determined by the fit of our Andreev data confirms our previous evaluation. In fact, it follows surprisingly well the  $T_c$  vs.  $x$  curve (thick solid line) [11]. On the contrary, both the ARPES LE and the tunneling gap increase monotonically with the decrease in  $x$  and, in the underdoped region, reach values very larger than those of the superconducting gap. As a further support to our results, the Andreev gap almost coincides with the tunneling gap in

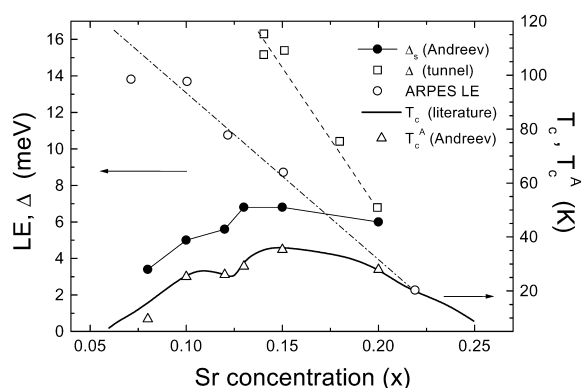


Fig. 5. Doping dependence of the ARPES leading-edge shift (open circles, from Ref. [7]), of the tunneling gap (open squares, from Refs. [5,6]) and of our point-contact Andreev isotropic gap  $\Delta_s$  (solid circles) in LSCO. All the data were taken below  $T_c$ . The temperatures  $T_c^A$  at which the Andreev features disappear in our samples are also reported (up triangles) and compared to the  $T_c$  vs.  $x$  curve from Ref. [11] (solid line).

overdoped samples as it happens in  $\text{Bi}_2\text{Sr}_2\text{CaCu}_2\text{O}_{8+\delta}$  (BSCCO) and  $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$  (YBCO) [1].

In conclusion, we found that the gap measured by Andreev reflection spectroscopy in LSCO closes at  $T_c$ , and we obtained a spectroscopic information supporting a mixed  $s + d$ -wave symmetry for the order parameter in this compound. Finally, we found that the doping dependence of the isotropic component of the low-temperature Andreev gap clearly follows the  $T_c$  vs.  $x$  curve, in contrast to both the tunneling gap and the ARPES LE.

These results clearly show that, in LSCO as in other cuprates [1], the gap measured by Andreev reflection is different from that measured by tunneling and ARPES below  $T_c$ , and is related to the presence of a phase-coherent condensate. This is in agreement with the picture proposed by Deutscher [1] according to which the pseudogap is a property of the non-superconducting state of LSCO, independent of its origin. Although we make no hypothesis about the mechanisms which govern the opening of the pseudogap, our results are very well described by some models which appeared in literature [21,22].

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