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Evidence of a *s*-wave subdominant order parameter in $YBa_2Cu_3O_{7-\delta}$ from break-junction tunneling spectra

A.I. Akimenko[1,2], F. Bobba[1], F. Giubileo[1], V.A. Gudimenko[2], S. Piano[1], and A.M. Cucolo[1]

[1] *Physics Department, CNR-Supermat Laboratory, University of Salerno, Via S. Allende; 84081 Baronissi, Italy*

[2] *B. Verkin Institute for Low Temperature Physics and Engineering of the National Academy of Sciences of Ukraine 47 Lenin Ave., Kharkov 61103, Ukraine*
E-mail: akimenko@ilt.kharkov.ua

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The tunneling spectra of $YBa_2Cu_3O_{7-\delta}$ break-junctions have been investigated for the tunneling direction close to the node one. The behavior of the zero-bias conductance peak (ZBCP) and Josephson current have been studied with temperature and magnetic field. The observed deep splitting of ZBCP which starts at $T_S < 20$–30 K is in agreement with the theory for the $d_{x^2-y^2} \pm is$ order parameter [Y. Tanuma, Y. Tanaka, and S. Kashiwaya, *Phys. Rev.* **B64**, 214519 (2001)]. We also observed that a low (0.04 T) magnetic field significantly depresses such splitting. The $1/T$ temperature dependence of maximum Josephson current that goes to saturation at $T < T_S$ also confirms the mixed order parameter formation.

PACS: **74.72.–h** Cuprate superconductors;
74.20.Rp Pairing symmetries (other than *s*-wave);
74.50.+r Tunneling phenomena; Josephson effects.

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For the d-wave superconductors, theory predicts specific quasiparticle bound states (Andreev bound states) near scattering structures such as surfaces, interfaces and other defects [1]. In these areas an order parameter (OP) may change significantly and subdominant OP may appear leading to a mixed OP (such as $d_{x^2-y^2} \pm is$ or $d_{x^2-y^2} \pm id_{xy}$) with the spontaneous breaking of time reversal symmetry (BTRS) [2–5].

Andreev bound states manifest themselves in different tunneling spectra as a zero-bias conductance peak (ZBCP) in agreement with the theory for the $d_{x^2-y^2}$-wave pairing [1]. In the case of breaking of time reversal symmetry (due to magnetic field or subdominant OP), splitting of ZBCP was predicted [5–7] and also observed in several experiments [8–12]. Theory shows the different kind of splitting for the is and id_{xy} subdominant OP [5,13]. This question has not been studied in tunneling experiments up to now.

The Josephson critical current may also give information about subdominant OP presence. Its temperature dependence is predicted to saturate at temperatures $T < T_S$ (T_S is the critical temperature for subdominant OP) [14].

It is interesting to note that some theories predict splitting of ZBCP without any subdominant OP [15] and even BTRS [16].

To solve the problem, we have investigated the $S-I-S$ Josephson junctions. The break-junction method was applied to a thin film and the tunneling spectra with the deep splitting of ZBCP at temperatures up to 20–30 K were registered. Indeed this method is very appropriate when dealing with high T_c superconductors since exposition of fresh and clean surfaces is achieved. The analysis of the temperature and magnetic field dependencies says in favor of the *is*-subdominant order parameter presence. The maximum Josephson current also saturates at $T < T_S$.

The tunnel junctions were produced by applying the modified break-junction technique [17] to highly biepitaxial c-axis oriented $YBa_2Cu_3O_{7-\delta}$ thin films (thickness ≈ 200 nm), dc sputtered on (001) $SrTiO_3$ substrates [18]. Electrical transport characterization showed critical temperatures T_c $(\rho = 0)$ = 91 K and $\Delta T_c < 1$ K. To determine the lateral lattice alignment between the films and the substrates the x-ray pole figure analysis was used [19]. The stripe-like samples (with the [110]-direction long side) were glued to a metallic bending plate by the epoxy glue. A special epoxy cover over the whole sample was applied to make the construction stable with the time and temperature change (more details see in Refs. 17 and 19). As a result we were able to investigate a single break-junction in about a weak time, with the only small change in its resistance in the temperature range 4.2–120 K [20]. To maximize the tunnel current along the node direction a straight groove was scratched in the central part of the covered sample (perpendicular to [110] direction), where the bending is maximum. By bending with a differential screw at low temperature, it is possible to crack the substrate together with the film along the groove and smoothly adjust the junction resistance by gently approaching the two cracked electrodes by a micrometer screw. The optical microscope study showed that the fracture direction can deviate from the straight line only about 10°. The break-junction method we used may also give a flat fracture surface [17]. Thus, we could get the high quality tunnel junctions.

In Fig. 1 we show the low-bias tunneling spectrum dI/dV vs V of the YBCO break-junction at $T = 10$ K measured by standard modulation technique. One can observe the simultaneous presence of two peak structures. Indeed, a well developed, narrow peak (with the width $W_J \approx \pm 1$ mV) centered at zero energy (see also Fig. 3) appears superimposed to a less pronounced, wider double-

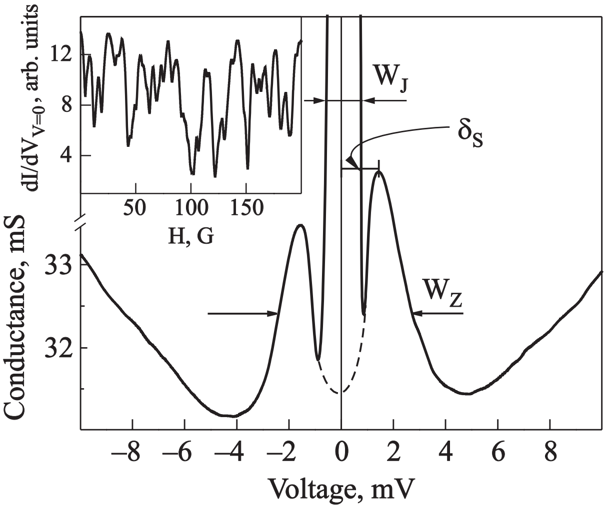

Fig. 1. Tunneling spectrum (dI/dV vs V) of YBCO break-junction at $T = 10$ K (solid line). The dashed line around $V = 0$ drawn by hand. The Josephson peak (with the width $W_J \approx \approx \pm 1$ mV) superimposes on the double peak structure ($W_Z \approx \approx \pm 2.5$ mV). δ_S shows the position (from $V = 0$) of the peak in the double peak structure. To understand the relative intensity of the peaks see Fig. 4. Inset: magnetic field dependence of the zero-bias conductance. The external field was applied parallel to the *c*-axis direction. The dependence is similar for the opposite directions of the field applied.

peak structure $W_Z \approx \pm 2.5$ mV. In addition to these, the wide gap-related maxima (or the bound states with nonzero energy [1]) around ± 15 mV are observed that shifts towards lower biases for increasing temperature and disappears at $T \to T_c$ [20]. The similar peak structure with ZBCP (without splitting) and gap-related peak were also found in Ref. 21 for the close to [110] direction tunneling in the $N-I-S$ ramp-edge junctions.

The narrow peak centered at $V = 0$ is mostly due to the Josephson direct current though it corresponds to a smeared current step at $V = 0$. The more the junction resistance the less its relative intensity. However, the most decisive argument in favor of the Josephson tunneling is the magnetic field dependence of the conductance at $V = 0$ shown in inset of Fig. 1. A similar oscillating behavior was also found for the Josephson critical current in junctions with the nonuniform current-density distribution [22]. In our case, the nonuniform current can be due to small deviations from the planar configuration of the junction. We do not have a satisfactory explanation for the finite slope of the Josephson critical current. The similar current step with the finite conductance at $V = 0$ was earlier observed in the YBCO grain boundary junctions [23] as well as in the YBCO and Nb break-junctions [19,24]. Around junction T_c, thermal and external fluctuations can induce the nonzero resistance since the Josephson coupling energy $E_J = = hI_c/2e$ is comparable with the thermal energy k_BT. However, at least for our low resistance junctions ($R_N = = 20$–$100\ \Omega$) at liquid helium temperature E_J was greater than k_BT by a factor of 20.

The double-peak structure with the width W_Z looks like the expected Andreev bound states structure for the case of subdominant OP presence [1]. The background conductance underlying the Josephson peak as inferred from the low temperature data is showed by thin dashed line in Fig. 1. This procedure is often used in the literature when dealing with high T_c superconductors or if it is not possible to separate different effects. Indeed, due to the extremely high value of the critical field H_{c2} for these compounds, it is not possible to observe experimentally the «intrinsic» condensate normal state at low temperatures, which knowledge is needed to normalize the superconducting conductance data at low temperatures. In our case we cannot depress the Josephson current by magnetic field without essential change of the double-peak structure. This effect will be discussed below in detail.

In Fig. 2 we show the temperature evolution of the structures of Fig. 1. One can see that the underlying double peak, observed at low temperatures, disappears with the temperature raise between 20 and 30 K transforming into the single wide peak. Such splitting of ZBCP with deep minimum for decreasing temperatures is only predicted for the $d_{x^2-y^2} \pm is$ order parameter (left inset of Fig. 2) [5,13]. The experimental temperature dependence is similar to that calculated in Ref. 5 (right inset in Fig. 2). We should note that for the $S-I-S$ junctions investigated

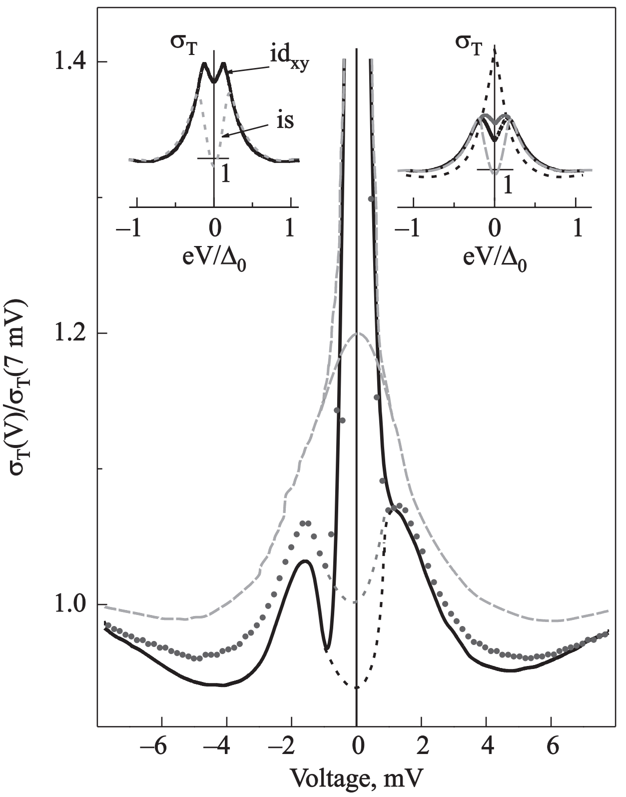

Fig. 2. Temperature dependence of normalized tunnel conductance $\sigma_T(V)$ at low temperatures T. Full line — 13 K, dotted line — 20 K, dashed line — 30 K. Normalizations of $\sigma_T = dI/dV$ is done for T = 13 K at V = 7 mV. The thin dashed line is the inferred background conductance (see text). Left inset: comparison of calculated tunneling conductance $\sigma_T\ (eV/\Delta_0)$ of $N-I-S$ junction for the node direction tunneling and for the $d_{x^2-y^2} \pm is$ and $d_{x^2-y^2} \pm id_{xy}$ order parameters [5]. Bath temperature T/T_c = 0.05. T_S/T_c = 0.2. Right inset: calculated $N-I-S$ junction tunneling conductance $\sigma\ _T\ (eV/\Delta_0)$ for the $d_{x^2-y^2} \pm is$ state for the node direction tunneling [5]. T/T_c = 0.05, 0.10, 0.12, 0.13 starting from bottom. T_S/T_c = 0.2.

here, the relative intensity of extremums must be more than that for the $N-I-S$ junctions calculated in Ref. 5 [25]. Nevertheless, the alternative $d_{x^2-y^2} \pm id_{xy}$ order parameter will not give so deep splitting that are found in our experiments.

We have also found that in relatively low magnetic field ≈ 0.04 T the depth of the minima around Josephson peak and distance between peaks $2\delta_S$ essentially decrease (see Fig. 3). It is reasonable because such magnetic field may effect as a strong depairing factor on the s-wave pairing. On the other hand, magnetic field can effect on the Andreev bound states shifting their energies to the higher values (with proper increase of δ_S) due to Doppler effect [8]. This effect was observed earlier. It seems difficult to distinguish these two opposite effects if the splitting is small (smeared due to roughness of the junction interface, see for instance [1]). Nevertheless, looking carefully on the results in Ref. 11, one can find the systematic decrease of δ_S with the low magnetic field increase too.

Thus, the comparison of both temperature and magnetic field dependencies of our data with the most accredited theories, appears in favor of the *is*-subdominant OP from tunneling measurements. The similar, in our opinion, but not so evident conclusion was done in Ref. 26 after analysis of the Andreev reflection point-contact spectra.

The maximum strength of the subdominant *s*-wave pairing from our measurements is $T_S/T_c \approx 0.24$ much higher than $T_S/T_c \approx 0.10$ earlier reported [8]. Theory [4] predicts $T_S/T_c = 0.16$.

In addition to this, when the is -wave (or id_{xy} -wave) subdominant pairing realizes, theory predicts the saturation of the maximum Josephson current at $T < T_S$ due to the decrease of the density of Andreev bounds states at Fermi level which transfer the Josephson current [14]. A similar behavior is observed in our experiments as reports in Fig. 3. To infer these data, we have integrated the Josephson conductance peak taking into account the inferred background (see the curve at T = 13 K). The temperature dependence of such current demonstrates clear saturation at T_S < 20–30 K. We have also observed the close to $1/T$ dependence in large temperature range in agreement with the experimental results for the ZBCP intensity in $S-I-S$ junction in Refs. 27, 28 and the theory for the node direction tunneling in the junctions with the same order parameter orientation in both electrodes [29]. Such junctions are most probably realized in our experiments.

In summary, the specific form of the tunneling spectrum with the deep splitting, the predicted temperature

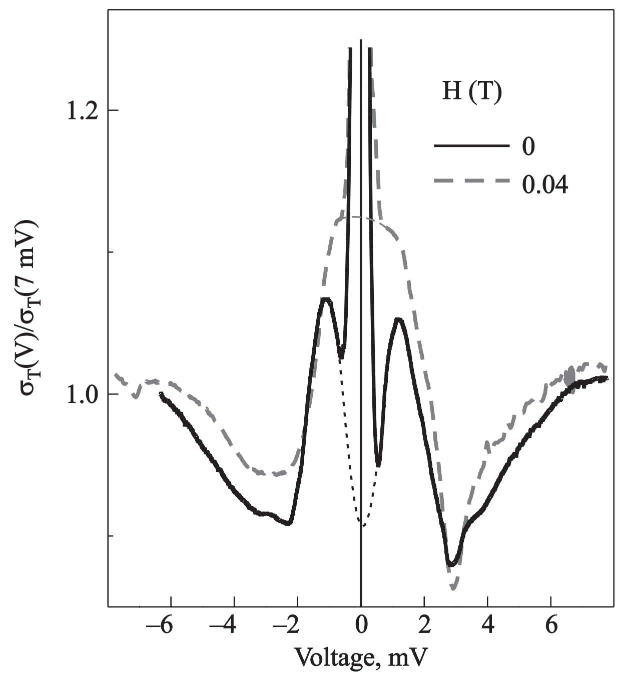

Fig. 3. Effect of magnetic field H = 0.04 T on the tunnel conductance.

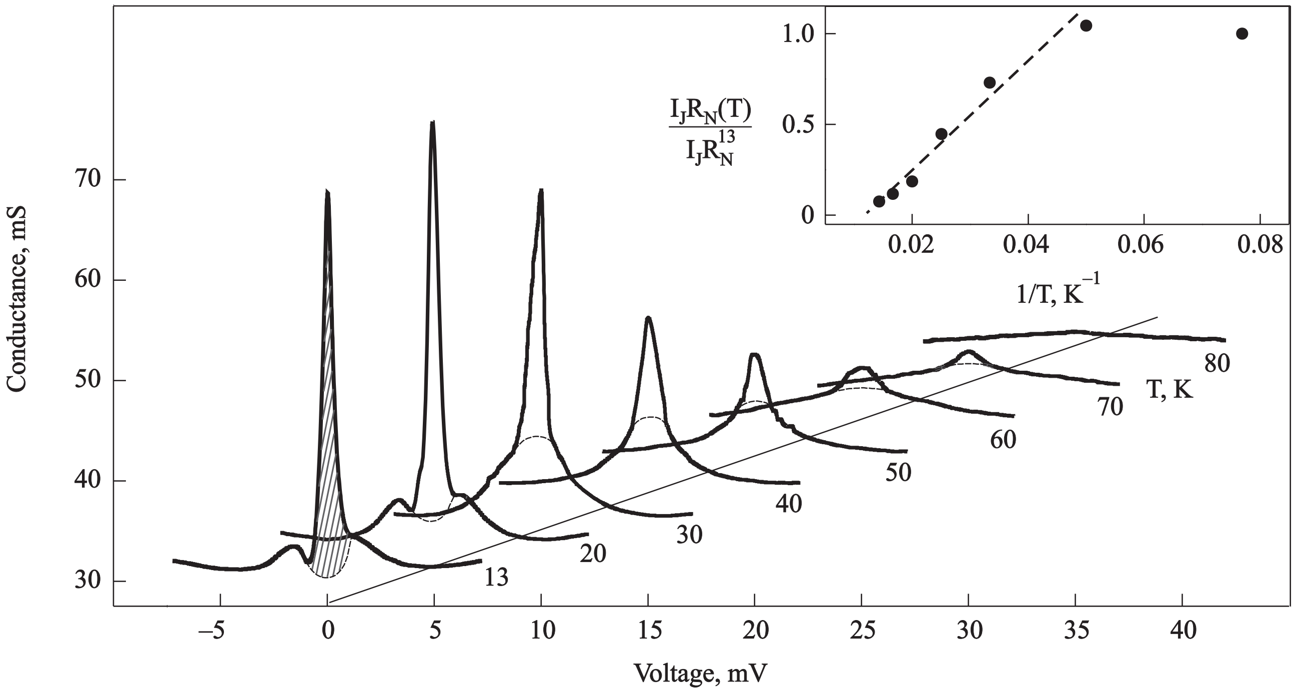

Fig. 4. Temperature dependence of tunneling spectrum dI/dV vs V. The curves at $T > 13$ K have been successively shifted along the voltage bias (with a 5 mV step) and conductance axes (along the thin solid line). The dashed line is the inferred background under Josephson peak. Inset: temperature dependence of the Josephson current $I_J R_N$ normalized to its value at $T = 13$ K, where R_N is the normal state resistance at $V = 100$ mV. The current corresponds to a square of the shaded area like shown for the curve at $T = 13$ K.

behavior of the splitted ZBCP and of the Josephson current have been observed in YBCO break-junctions giving the clear evidence for the mixed symmetry $d_{x^2-y^2} \pm is$ of order parameter near the (110) surface in contrast to the $d_{x^2-y^2} \pm id_{xy}$ OP. The deduced strength of the *is*-subdominant OP is rather high leading to the transition into the $d_{x^2-y^2} \pm is$ states at $20 < T_S < 30$.

A recent finding of *s*-wave pairing in the heavily Zn-doped YBCO [30] says also in favor of *is*-subdominant OP in the undoped YBCO.

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