Energy gap in $YBa_2Cu_3O_x$ determined from the data on tunnel experiments with single crystals

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The tunnel spectra of $YBa_2Cu_3O_x$ single crystals with various oxygen concentrations were studied using a scanning tunneling microscope in the orientations perpendicular to and parallel to the basal *ab* plane. Two gap-related structural features were observed in the tunnel spectra in the orientation at right angles to the *ab* plane. The plot of the energy gap $\Delta(x)$ is similar to the plot of $T_c(x)$. The distribution of Δ in the depth of the sample was found to be systematically nonuniform.

In the present letter we report the results of an experimental study of single crystals of $YBa_2Cu_3O_x$ with various oxygen concentrations. The study was carried out with use of a scanning tunneling microscope. The single crystal samples of $YBa_2Cu_3O_x$ with various oxygen concentrations were synthesized by a method described in Ref. 1.

The results of experimental studies of some structural, optical, and superconduct-

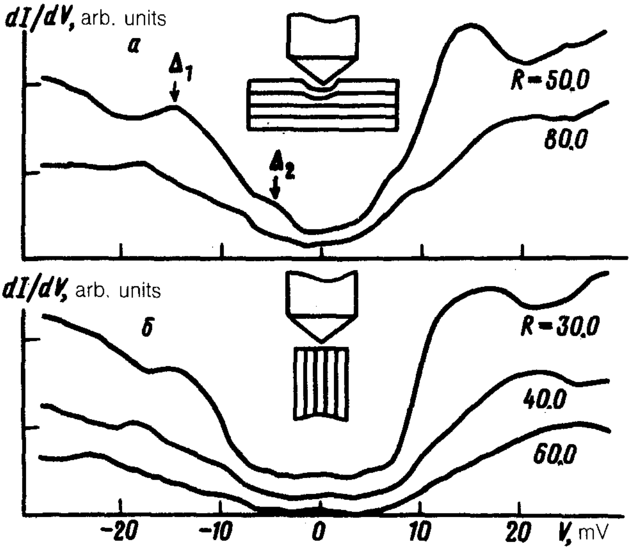

FIG. 1. Fragments of a tunnel spectrum (derivatives of the current-voltage characteristics) obtained with use of a scanning tunneling microscope for two orientations of the needle relative to the basal plane (a, b) of the single crystal for various resistances $R(M\Omega)$ of the tunnel gap (various spacings between the needle and the sample). (a) At right angles to ab; (b) perpendicular to ab.

ing properties of the samples were reported in Refs. 1–3. The design of the scanning tunneling microscope was described in Ref. 4.

The tunnel spectra were measured in two orientations of the crystal relative to the needle of the scanning microscope (Fig. 1). In the orientation a the needle was directed normal to the ab plane of the single crystal, and in the orientation b it was directed parallel to this plane. During the measurements in the a orientation (the needle is at right angles to the ab plane), the nonconducting surface layer of the single crystal was destroyed by the needle.[5] Measurements in the b orientation (the needle is parallel to ab) were performed using a freshly cleaved surface obtained *in situ* at 4.2 K.

The vacuum gap between the needle and the surface of the sample was controlled by measuring the tunnel current as a function of displacement of the needle.[5] The temperature dependence of the tunnel spectra was measured only at low temperatures, $T < 20 - 40$ K, when the thermal expansion of the structural elements of the microscope is negligible.

Traces of the fragments of the tunnel spectra which we measured are shown in Figs. 1a and 1b. We see that the tunnel spectra in each orientation exhibit certain characteristic features which can be linked with the superconducting energy gap. In the orientation a we see two such features.

In the absence of a model-based theory of tunneling in high-T_c superconductors, it is difficult to correctly determine the magnitude of the energy gap. As a measure of the gap Δ, we have therefore used half the distance between the corresponding structural features of a tunnel spectrum. The problem is complicated by the fact that the energy position of the structural features, Δ_1, strongly depends, as was initially noted in Ref. 6, on the magnitude of the tunnel gap (Fig. 1). In Ref. 6 this phenomenon was attributed to the proximity effect. This phenomenon could be the result of the particular properties of the vacuum tunnel junction established in the scanning microscope. The structural features which set this tunnel junction apart from the standard clamp-on tunnel junctions are, aside from uniquely high degree of localizability (< 1 nm), an appreciably higher tunnel resistance R (several tens or even several hundreds of $M\Omega$), very low effective capacitance of the needle [the sample's capacitance is $c \sim 10^{-18}$ F (Ref. 7)], and an unambiguous dependence of these quantities on the tunnel gap. A low capacitance C may cause a "Coulomb blocking" of the tunneling, depending on the value of C (and hence on the value of R), which would shift the energy position of Δ_1 in the tunnel spectrum.[7] Furthermore, despite regular monitoring, one should not completely rule out the possibility that a mechanical contact can be established between the needle and the sample which, when attempting to change the tunnel gap, would account for the strong, variable pressure the needle exerts on the sample, thereby causing the tunnel spectrum to change. In any case, the observed functional dependence $\Delta(R)$ requires further study. The values of Δ obtained in various experiments with use of a tunneling microscope can be compared quantitatively by using tunnel spectra which were recorded when the spacing between the needle and the sample was identical and the tunnel resistance R was also identical. Our quantitative estimate of Δ was based on a tunnel spectrum corresponding to a current of 1 nA (with the needle held at $V \sim 100$ mV; $R \sim 50$–100 $M\Omega$). The structural feature of the tunnel spectrum which corresponds to the lower energy Δ_2 (Fig. 1a) is generally barely discernible and the effect the tunnel gap has on it is difficult to determine.

Figure 2 is a plot of Δ_1 and Δ_2, which correspond to the structural features of the tunnel spectra, as a function of the oxygen content of the sample. The experimental results for the orientation a are represented by the vertical segments which demonstrate the spread of the local values of Δ (from the maximum to the minimum value)

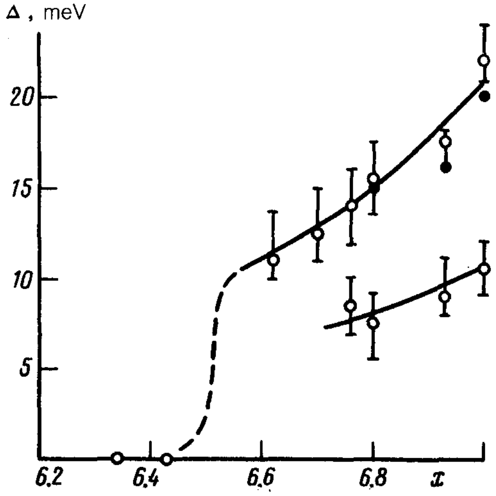

FIG. 2. Energy gaps Δ_1 and Δ_2 versus the oxygen concentration x in the sample. The filled circles are for the orientation parallel to *ab*.

obtained on a square grid with a 100-nm spacing and a total area of 1 μm^2. The point inside the line is the grid-averaged value of Δ. The spatial distribution of Δ in sections of the samples with different values of x is similar in nature to that reported to Ref. 8.

The filled circles in Fig. 2 represent the values of Δ obtained in the b orientation. A comparison of the results obtained for the a and b orientations requires that account be taken of the fact that cleavage in the plane at right angles to the ab plane exposes the deep-seated regions of the crystal which are subjected to conditions during processing different from those at the surface.

Figure 3 shows the results of measurement of the parameter Δ in the b orientation for two samples ($x = 6.74$ and $x = 6.9$) as functions of the distance from the fracture edge of the sample of thickness ~ 30 μm along the direction at right angles to the ab plane. The circles in Fig. 3 correspond to the local measurements and they show that Δ decreases in an entirely regular manner as the needle moves from the periphery of the sample to its midpoint. Clearly, this effect characterizes the oxygen distribution in the samples and is determined by the particular features of the diffusion and sorption processes. In any case, a comparison of the values of Δ obtained in the b orientation with the results obtained in the a orientation must make use of the values of Δ measured at the periphery of the sample. It follows from Fig. 2 that the gap in the direction parallal to ab corresponds to the large gap Δ_1 in the direction at right angles to the ab plane. Looking at Fig. 2 again, we wish to emphasize that the overall behavior of the gap Δ_1, including the change in the oxygen content, is clearly similar to the behavior of the critical temperature T_c (see Refs. 1 and 3). We can accordingly identify the parameter Δ_1 as a measure of the superconducting gap which characterizes the bulk properties of the test samples. This point is clearly illustrated by the plot in Fig. 4, which was constructed from the curves for $T_c(x)$ (Refs. 1 and 3) and $\Delta(x)$ (Fig. 2). It should be noted that the curve of T_c (x) characterizes the sample as a whole and can, in principle, differ from the curve for the surface of the sample [for which a $\Delta(x)$ curve was measured]. Taking into account the considerable scatter of the local values of Δ, however, we can assume that it is legitimate to construct the plot in Fig. 4 from the given data. The lines in Fig. 4 correspond to normalized values of the energy gaps, $2\Delta_1/kT_c = 5.3$ and $2\Delta_2/kT_c = 2.7$. The measurements of the temperature depend-

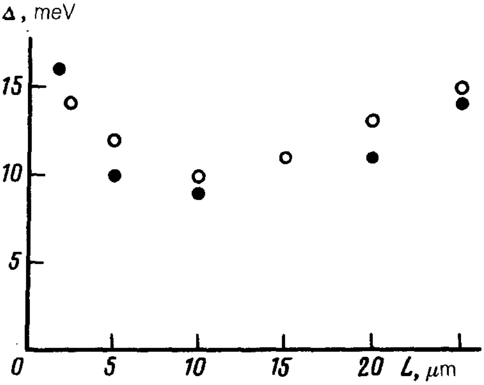

FIG. 3. Distribution of the energy gap Δ along the surface of a $YBa_2Cu_3O_x$ crystal freshly cleaved at right angles to ab. ○—$x = 6.75$; ●—$x = 6.93$.

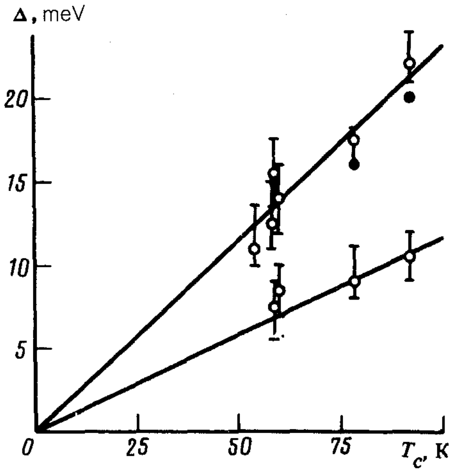

FIG. 4. Energy gap versus the critical temperature of the superconducting transition T_c. The circles on the lower part of the plots correspond to the structural feature of the tunnel spectra observed at low energies. Filled circles—Measurements in the orientation parallel to *ab*.

ence of Δ_1 in the temperature interval 4.2 – 40 K are in general agreement with the predictions of the BCS theory, although the values extrapolated to T_c are slightly lower (by 7–10 K) than those obtained from volume measurements. The small values of the energy gap Δ_2 can be explained in a trivial manner if we consider that the surface of a single crystal always has oxygen-depleted regions.[1)]

On the other hand, if we take into account the data obtained from the various tunneling and optical measurements (see, e.g., Refs. 9–11), we see that there could be other explanations of the observed "tunneling" picture. It is conceivable that the small gap Δ_2 does indeed exist. Note that the results of our observations in this case would indicate that there are two discrete quantities, Δ_1 and Δ_2, rather than point out the anisotropy of the gap. In the orientation *a* (Fig. 1a) the electron tunneling can actually occur in all directions, but in the case of an ordinary anisotropy we would have only a "smeared" structural feature in the tunnel spectrum. Unfortunately, we have no other evidence now which could substantiate the existence of a clear-cut "two-gap" feature in $YBa_2Cu_3O_x$.

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[1)] Since there is a plateau on the $T_c(x)$ and $\Delta(x)$ curves in the concentration range $x = 6.5$–6.75, we should expect that Δ_2 would correspond to ~ 10 meV.

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