Influence of defects on the energy gap in $YBa_2Cu_3O_{7-x}$ as measured with the help of Andreev reflection

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The influence of radiation-induced defects on the energy gap Δ in the compound $YBa_2Cu_3O_{7-x}$, as measured with the help of Andreev reflection, was investigated. It was found that Δ decreases with increasing flux of irradiating particles at a much lower rate than the kinetic temperature T_c. It is shown that Δ remains different from zero for $\Phi > \Phi_c$, where Φ_c is the critical flux density corresponding to $T_c=0$. The results are interpreted as a transition of a Bose condensate of Cooper pairs into a localized state.

INTRODUCTION

The behavior of the energy gap Δ in the single-particle excitation spectrum of high-T_c oxide superconductors is, in many respects, different from that predicted by the BCS theory:[1] The ratio $2\Delta(0)/kT_c$ is much greater than 3.5; the temperature dependence of the energy gap $\Delta(T)$ is anomalous;[2–5] and so on. The reasons for these deviations are not entirely understood: Are they associated with experimental difficulties (short coherence length, low oxygen mobility, etc.) or peculiarities of the superconductivity mechanism in these compounds? For this reason it is of interest to investigate the character of the variation of Δ and T_c in the same sample under the external action of some independent parameter. The most convenient procedure is to produce radiation-induced defects by irradiating a high-T_c sample with fast particles, since the defect-formation process can be controlled in this case. It is well known that radiation-induced defects strongly influence T_c, driving T_c to zero at some critical concentration $N=N_c$ (see the review Ref. 6).

This is apparently the first attempt to investigate the influence of radiation-induced defects on the value of Δ measured with the help of Andreev reflection.[7]

Another reason for this investigation is to check the theoretical prediction, made by one of us,[8] that a localized Bose condensate of Cooper pairs exists if $N > N_c$, when the critical temperature is already zero. The existence of a localized condensate (i.e., $\langle|\Delta|^2\rangle \neq 0$) should result in characteristic features in the Andreev reflection spectra, while the superconducting current and therefore also T_c, determined by the onset of resistance, will be zero.

The choice of the experimental method is based on the fact that structures with clean N–S contacts (N is a normal metal and S is a superconductor) have many advantages over traditional tunnel junctions.[9] Moreover, the experimental characteristics of N–S structures are stable and the measurements are completely reproducible.[10–13]

EXPERIMENTAL PROCEDURE

We employed samples of high-quality epitaxial (001)-oriented, 0.2–0.3 μm thick $YBa_2Cu_3O_{7-x}$ thin films prepared by the laser-sputtering method.[14] Silver contact pads, 0.1–0.2 μm thick, were deposited on the film immediately after the sputtering. It is well known that a transitional layer is not formed when silver is deposited on freshly prepared samples of $YBa_2Cu_3\ O_{7-x}$.[15] The geometry of the samples is shown in Fig. 1 and is determined by two factors. First, the films must be irradiated over the entire length, including the contact pads. Second, the thickness of the prepared Ag and $YBa_2Cu_3O_{7-x}$ films must be checked, which is most simply done by interference.

The Andreev reflection effect was investigated, using the method described in Ref. 10, with the help of Au—bilayer (Ag–$YBa_2Cu_3O_{7-x}$) point contacts formed directly at low temperature with the help of a precision mechanical coupling.[16] Gold wires 40 μm in diameter and sharpened to a point by electrochemical etching in an HNO_3–HCl solution, were used as normal counter electrodes. The needle point radius was less than 1 μm. The current-voltage characteristics of the contacts were measured by a standard four-contact method. Curves of the differential resistance $R_d=dV/dI$ versus the voltage V were recorded automatically by the modulation technique. The contact-making system was placed in the gap of a toroidal superconducting solenoid. The magnetic field was oriented parallel to the film plane. The experiments were performed in a cryostat with a gaseous heat-exchange medium.

The samples were irradiated at room temperature, in a cyclotron accelerator, with 1.2 MeV He^+ ions with particle-flux density Φ ranging from $\Phi=0$ to $\Phi=2.3\times10^{16}$ cm^{-2}. The overheating of the sample under irradiation did not exceed 50 K. The geometry of the experiment made it possible to irradiate films along their entire length, including the contact pads, and the presence of the Ag layer on the investigated part of the film guaranteed that the surface of the $YBa_2Cu_3O_{7-x}$ films would remain clean. Irradiation produced a uniform defect structure, since the total thickness of the bilayers did not exceed 0.5 μm, which is significantly less than the path length of He^+ ions in both Ag ($L\simeq1$ μm) and $YBa_2Cu_3O_{7-x}$ ($L\simeq3$ μm).

After each irradiation the curves $R(T)$ were measured by the four-contact method, the temperature dependences of the diamagnetic susceptibility in a weak constant mag-

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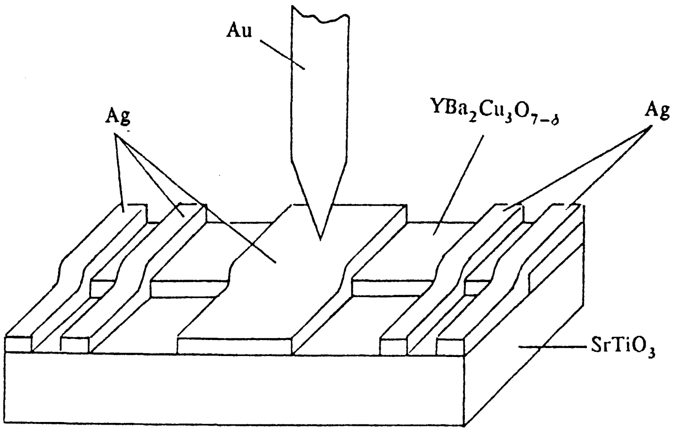

FIG. 1. Arrangement of the contact pads on the high-T_c superconducting samples.

netic field were measured with the help of a SQUID magnetometer,[17] and the characteristics of at least four point contacts were investigated.

Two $YBa_2Cu_3O_{7-x}$ films with $T_c=91$–92 K and superconducting transition width $\Delta T_c<1$ K were irradiated and investigated.

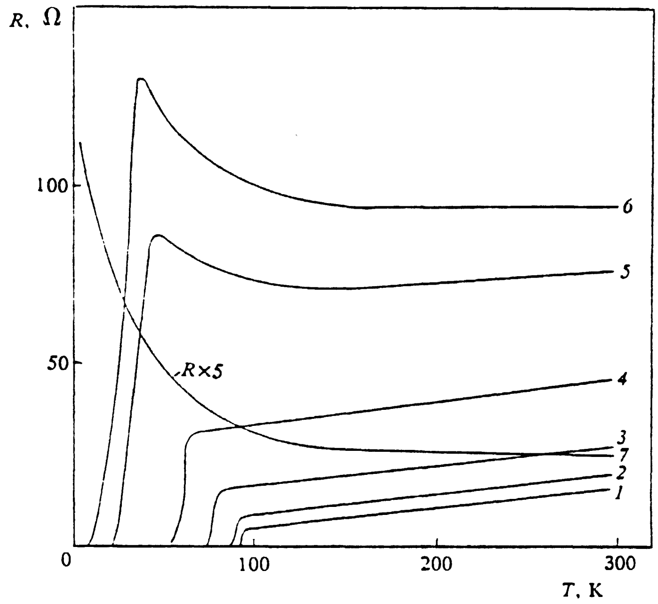

FIG. 3. Temperature dependences of the electric resistance $R(T)$ of sample No. 2 with different irradiation fluxes: $\Phi=0$ (*1*), 5×10^{15} (*2*), 10^{16} (*3*), 1.5×10^{16} (*4*), 1.9×10^{16} (*5*), 2.05×10^{16} (*6*), and 2.3×10^{16} cm^{-2} (*7*).

EXPERIMENTAL RESULTS

Figure 2 displays the differential resistance of a point contact (sample No. 2) as a function of the voltage in the absence of irradiation ($\Phi=0$) in the temperature range 4.2–82.2 K. These voltage dependences are typical for our films and are similar to the voltage dependences obtained in Refs. 12–13. The energy gap, determined from the length of the flat section of the $R_d(V)$ curve in the c-axis direction is $\Delta=10$ meV, which also agrees with the results of Refs. 12–13. The shape of the $R_d(V)$ curves at $T=4.2$ K shows that the effect of the potential barrier of the N–S boundary is weak, i.e., the parameter $Z=0$.[18] A characteristic feature is the strong blurring of the flat section of $R_d(V)$ near $V=0$ with increasing temperature.

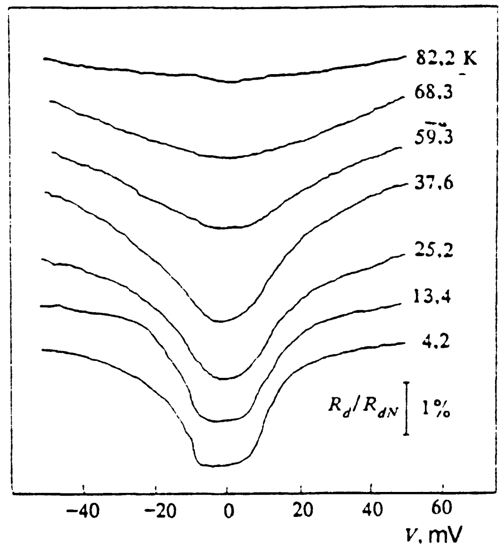

FIG. 2. Differential resistance R_d of a point contact ($R_{dN}=31.2\ \Omega$) for sample No. 2 versus the voltage V at different temperatures in the absence of irradiation ($\Phi=0$).

Figure 3 displays the temperature dependences of the resistance $R(T)$ of sample No. 2 with different irradiation fluxes right up to $\Phi=2.3\times10^{16}$ cm$^{-2}>\Phi_c=2.13\times10^{16}$ cm^{-2}. The curves are in good agreement with the well-known results of Ref. 6. Indeed, the critical temperature, defined as $R(T_c)=0$, decreases with increasing flux and vanishes at $\Phi=\Phi_c$. The critical flux density was determined by extrapolating the $T_c(\Phi)$ curve to zero and was equal to $\Phi_c=1.4\times10^{16}$ cm^{-2} and 2.13×10^{16} cm^{-2} for samples No. 1 and No. 2, respectively. The electric resistance increases and the character of the temperature dependence changes. Curves of the reduced critical temperature T_c/T_{c0} versus the reduced irradiation flux Φ/Φ_c are displayed in Fig. 4 for both experimental samples, and they exhibit universal behavior.[6]

The most interesting result was obtained by measuring the voltage dependences $R_d(V)$ of point contacts at different irradiation fluxes. These curves for sample No. 2 are shown in Fig. 5 (the curves for sample No. 1 are similar). It is evident that as Φ increases all features typical of Andreev reflection are preserved, and Δ decreases continuously but much more slowly than T_c (see Figs. 4 and 6). We note that the flat sections of $R_d(V)$ near $V=0$, which correspond to nonzero Δ, continue right up to the critical flux Φ_c, when T_c is already zero. For $\Phi>\Phi_c$ ($\Phi=2.3\times10^{16}$ cm^{-2}) the excess current continues to exist, exhibiting a gap singularly, though the characteristic plateau near $V=0$ is strongly blurred.

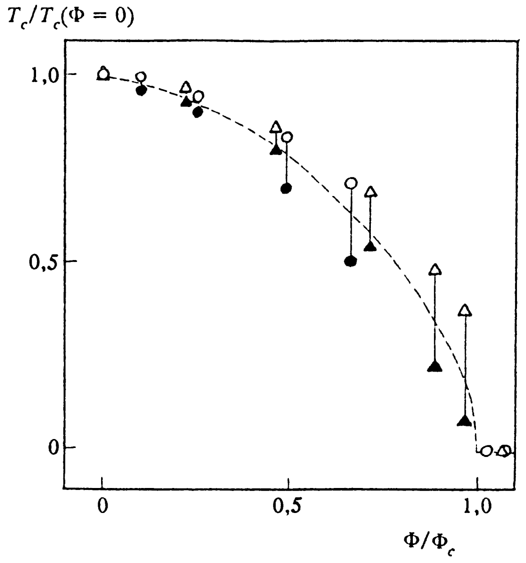

FIG. 4. Reduced critical temperature T_c/T_c ($\Phi=0$) versus the reduced irradiation flux Φ/Φ_c for samples No. 1 (○) and No. 2 (Δ).

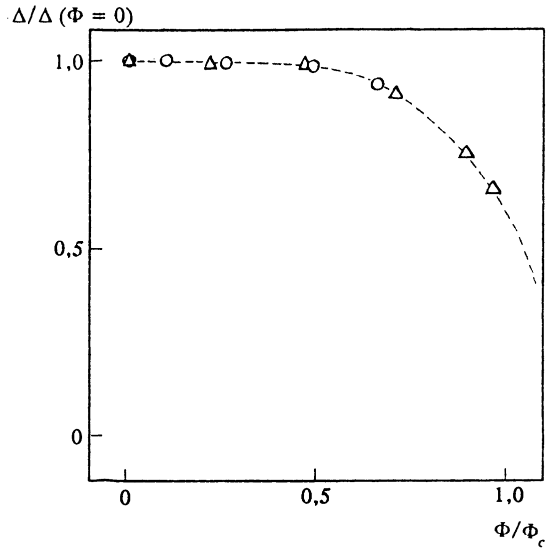

FIG. 6. Energy gap Δ versus the reduced irradiation flux Φ/Φ_c for samples No. 1 (○) and No. 2 (Δ).

To determine the reason for the excess current we employed a standard technique in which a magnetic field is applied parallel to the plane of the film.[19] It is well known that the magnetic field in this geometry displaces the trajectories of the incident and reflected (if present) particles, which results in a reduction of the excess current and, in our experiments, an increase of the differential resistance of the point contact for $|eV| < \Delta$. Figure 7 displays the $R_d(V)$ curves for sample No. 2 irradiated with flux $\Phi = 2.3\times 10^{16}$ cm^{-2} in magnetic fields $H=0$ (curve *1*) and $H=1.4$ T (curve *2*). It is evident that when a magnetic field is applied the resistance of the contact near $V=0$ increases, just as is predicted by the theory. We note that the scale of the change in the resistance for a given thickness of the N layer ($d=0.1$ μm) is in good agreement with the data of Refs. 13 and 19. For this reason, there is every basis for believing that in this case Andreev quasiparticles are responsible for the excess current.

Figure 6 displays the curves of $\Delta(T=4.2$ K) versus the reduced flux density for samples Nos. 1 and 2. It is

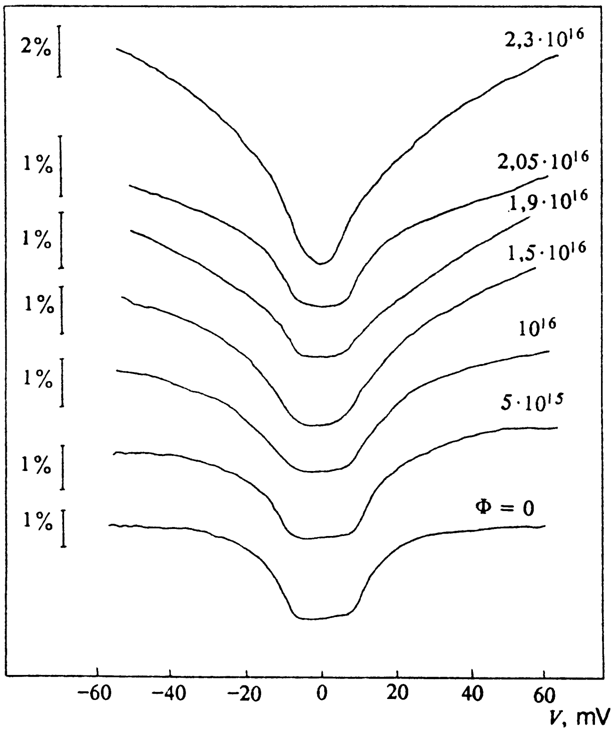

FIG. 5. Differential resistance of point contacts of sample No. 2 versus the voltage for different irradiation fluxes. The normal resistance of the contacts for the displayed curves ranges from 19 to 36 Ω.

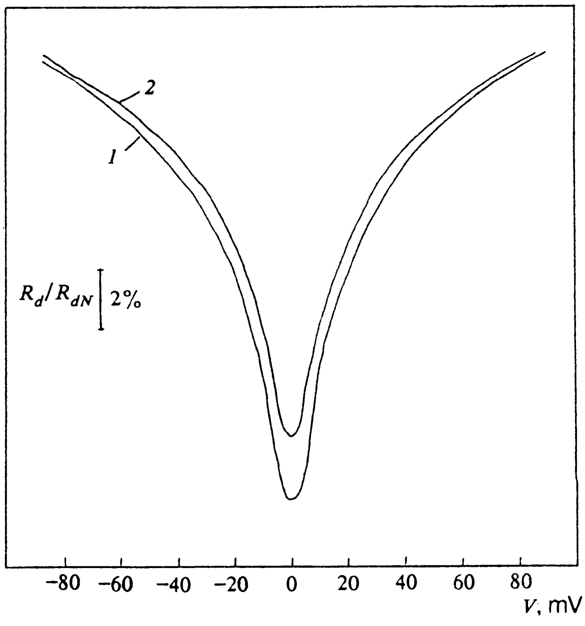

FIG. 7. $R_d(V)$ of sample No. 2 irradiated with the flux $\Phi=2.3\times 10^{16}$ cm^{-2} in magnetic fields $H=0$ (*1*) and $H=1.4$ T (*2*). The normal resistance of the contact $R_{dN}=35.5$ Ω.

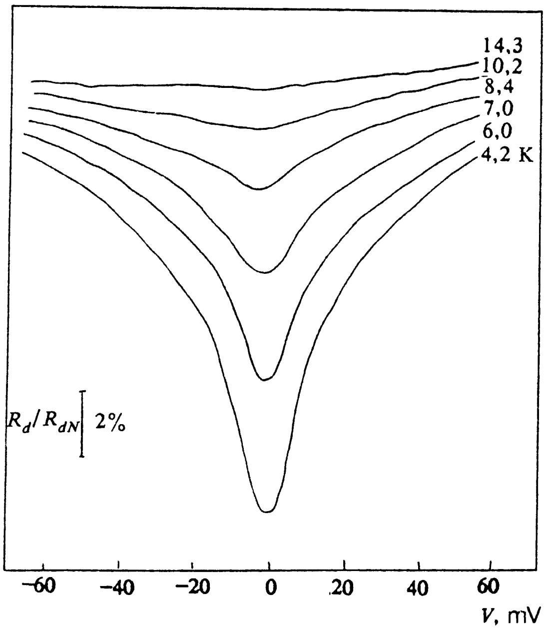

FIG. 8. $R_d(V)$ of sample No. 2 irradiated with the flux $\Phi=2.3\times10^{16}$ cm^{-2} at different temperatures. The normal resistance of the contact $R_{dN}=29\ \Omega$.

evident that, in contrast to the critical temperature T_c, the gap width Δ for $\Phi>\Phi_c$ does not vanish. We note that the experimental points for both samples fall on a single curve.

It is of interest to study the temperature dependences of $R_d(V)$ for $\Phi>\Phi_c$. As is evident from Fig. 8, the temperature rapidly reduces the excess current, and already at $T=14.3$ K the singularities produced by Andreev reflection vanish. It should also be noted that the excess current is higher for $\Phi>\Phi_c$ than $\Phi<\Phi_c$.

We also performed control measurements of the resistance of the silver films during irradiation, since a possible decrease of the mean-free path in Ag can strongly alter the character of the voltage dependence $R_d(V)$. Within the limits of measurement error, however, no growth of the resistance of Ag films was observed.

DISCUSSION

It is well known that the excess current in an N–NS point contact is due to Andreev reflection of holes from the "barrier" produced by the field of the Bose condensate. Since such a "barrier" will also be present in the case of a localized condensate (this is especially obvious if the coherence length is shorter than the localization radius), in this case similar features should be expected in the differential resistance. Therefore the observation of excess current for $\Phi>\Phi_c$, when $T_c=0$, can be regarded as evidence for the existence of a localized Bose condensate of Cooper pairs, in agreement with theoretical predictions.[8] Indeed, since the characteristic features of $R_d(V)$ near $V=0$ remain for $\Phi>\Phi_c$ (see Figs. 5 and 6) and the critical temperature is zero in this case (Fig. 4), this means that a condensate incapable of carrying undamped current is present.

The model in Ref. 8 also predicts that the localized condensate will break down with increasing temperature (Fig. 8), and explains the blurring of the flat section of $R_d(V)$ for $\Phi>\Phi_c$. The latter circumstance is associated with the strong spatial nonuniformity of the localized Bose condensate.

Of course, the experimental results presented are not a final proof of the existence of a localized condensate of Cooper pairs since it is difficult to take into account all factors in the interpretation of the curves $R_d(V)$. At present, however, we do not se an alternative explanation for the phenomena described above. It would be desirable to study the influence of radiation-induced defects on the energy gap with the help of other methods, for example, optical or ultrasonic spectroscopy.

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