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Role of interfaces in the proximity effect in anisotropic superconductors

E. Polturak, G. Koren, D. Cohen, and O. Nesher
Physics Department, Technion - Israel Institute of Technology, Haifa 32000, Israel

R. G. Mints and I. Snapiro
School of Physics and Astronomy, Tel Aviv University, Tel Aviv 69978, Israel

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We report measurements of the critical temperature of $YBa_2Cu_3O_7$-$YBCu_{3-x}Co_xO_{7+\delta}$ superconductor–normal-metal (S-N) bilayer films. Depending on the morphology of the S-N interface, the coupling between S and N layers can be turned on to depress the T_c of S by tens of degrees, or turned down so the layers appear almost totally decoupled. This effect can be explained by the mechanism of quasiparticle transmission into an anisotropic superconductor. [S0163-1829(98)52722-1]

The system of a high-T_c superconductor and a normal-metal conductor in proximity received much attention, not least since one of the practical high-T_c Josephson junction types is the superconductor–normal-metal–superconductor (SNS) device.[1] One aspect which so far did not receive enough attention is the characteristics of S/N interfaces which are an integral part of such junctions. In contrast to conventional s-wave superconductors, transport properties across interfaces in anisotropic superconductors are predicted to be very sensitive to the details of the morphology of the S/N interface.[2] To check this idea experimentally, we decided to compare the superconducting properties of S/N bilayers differing only in their interfaces. We know how to control the morphology well in a configuration of c-axis-oriented films. The bilayer films consist of a thin layer of $YBa_2Cu_3C_7$ (YBCO) grown on (100) $SrTiO_3$, capped by a much thicker layer of $YBCu_{3-x}Co_xO_{7+\delta}$. $YBCu_{3-x}Co_xO_{7+\delta}$ was chosen as the normal-metal material since it grows epitaxially on YBCO, has a negligible interdiffusion, and was used as a barrier in SNS junctions by several groups.[3,4] The expected influence of the normal-metal conductor on the superconductor would extend to a distance of a coherence length from the interface, as predicted by the conventional proximity effect.[1] Accordingly, the S layer should be as thin as possible in order to produce an observable effect on T_c of the bilayer. Starting from that point, we eventually extended the experiments to include thicker S layers. All of these experiments are presented below.

The films, deposited using laser ablation, are epitaxial with c axis perpendicular to the substrate. Bilayers were prepared with a thickness of YBCO between 60 and 550 Å, and a thicker $YBCu_{3-x}Co_xO_{7+\delta}$ cap, between 1000 and 1500 Å. Here we present data for $YBCu_{3-x}Co_xO_{7+\delta}$ with $x=0.15$, which is normal down to about 50 K. To maintain a clean interface between the S and N layers, each bilayer was grown in one deposition run. Different S/N interfaces were produced by changing the deposition rate. We refer to a rate of deposition of 6.5 Å/sec as fast growth, while bilayers grown at a rate 2.5 times slower, with the growth interrupted every 30 sec of deposition for a 60 sec pause, we call slowly grown. The fast growth rate produces films which grow by screw dislocations and their surface shows rounded smooth features, similar to those obtained by Schlom *et al.*[5] In contrast, the slow growth produces films which grow in the Stranski-Krastanov mode, namely layer by layer, up to a critical thickness of about 150 Å, and then by a two-dimensional (2D) island growth.[6] The bilayers were characterized by transport, ac susceptibility, and atomic force microscopy (AFM). Overall, the reproducibility of data was excellent over a period of a year or so during which the experiments were done. A summary of the T_c's of all these bilayers plotted against the thickness of the YBCO layer is shown in Fig. 1. In order to have only one variable, we chose to work in the regime where T_c of the bilayers becomes independent of the thickness of the cap layer, and thus inde-

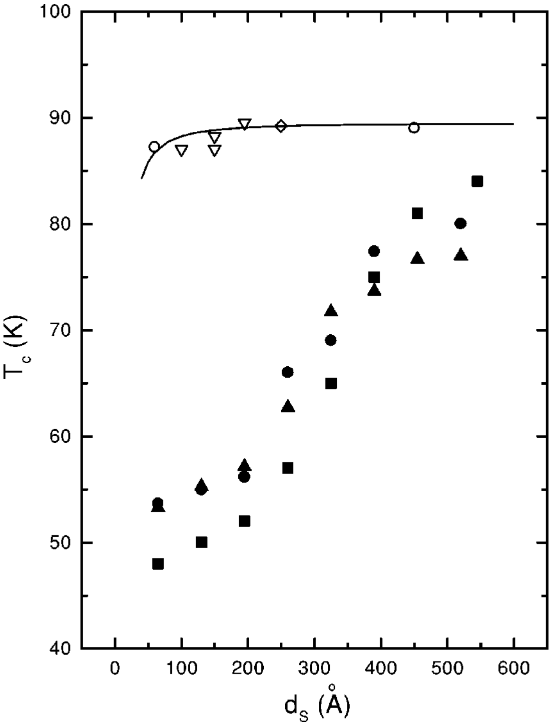

FIG. 1. T_c of the S/N bilayers vs the thickness of the YBCO. Closed symbols refer to fast grown bilayers and open symbols refer to slowly grown bilayers. The solid line is T_c expected from the proximity effect in the case of a rough interface.

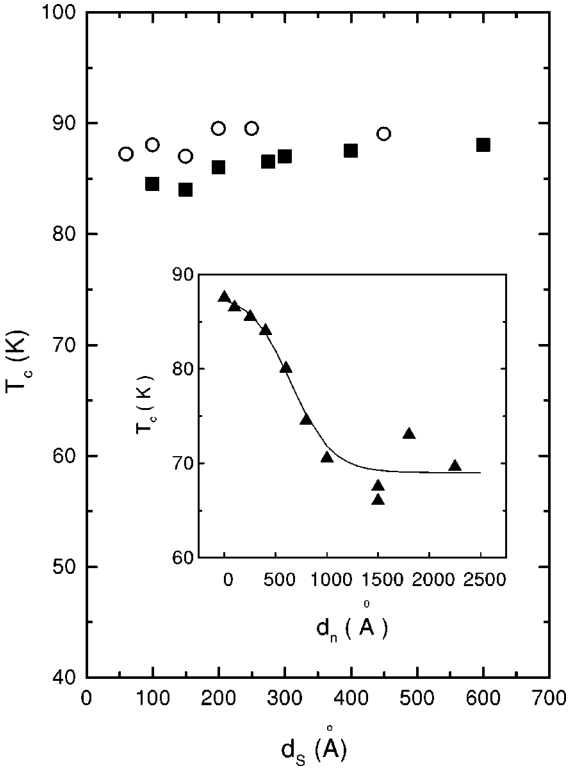

FIG. 2. T_c of single-layer reference YBCO films vs their thickness. Closed squares refer to fast grown films and open circles to slowly grown films. The inset shows the dependence of the T_c of a bilayer on the thickness of the $YBCu_{2.85}Co_{0.15}O_{7+\delta}$ cap. The thickness of the (fast grown) YBCO film in these bilayers is 250 Å. The solid line is a guide to the eye.

pendent of the top surface of the bilayer. One can see in the inset of Fig. 2 that this happens once the thickness of the cap exceeds about 1000 Å. All the data presented here were obtained in this regime. One can see that there is an enormous difference between the T_c's of the fast grown (solid symbols) and the slowly grown (open symbols) bilayers. In contrast, the difference between the T_c's of single-layer YBCO films grown as reference at these two deposition rates was no more than 2–3 K, as shown in Fig. 2. Furthermore, T_c of single-layer fast grown films as thin as 100 Å is near 85 K, which shows that there are no problems with the films of this thickness being discontinuous. Thus, the lowering of T_c of the bilayers is definitely associated with the presence of the normal-metal layer.

Since the effect was much larger than expected, our first thought was that the reduction of T_c is caused by migration of oxygen from the YBCO into $YBCu_{3\text{-}x}Co_xO_{7+\delta}$, leaving the YBCO oxygen deficient, and hence with a lower T_c. Fully oxygenated $YBCu_{3\text{-}x}Co_xO_{7+\delta}$ contains more oxygen than YBCO, with the excess amount increasing with x.[7] If not enough oxygen is supplied during growth, then some oxygen may subsequently migrate from the YBCO into the $YBCu_{3\text{-}x}Co_xO_{7+\delta}$, lowering the T_c of the YBCO. To check this possibility, we grew bilayers under different methods of oxygen loading as follows: (a) increased the oxygen ambient pressure during deposition by a factor of 2; (b) increased the time length of the post deposition oxygen loading by a factor of 2; (c) increased the Co composition of the capping layer from $x=0.15$ to $x=0.3$ and then to $x=1$. Test (a) increases the flux of oxygen atoms during growth. Test (b) allows the film to absorb more oxygen during postdeposition oxygen loading. Both (a) and (b) should increase the amount of oxygen in the film, and thus increase T_c. In contrast, test (c) increases the amount of excess oxygen needed by $YBCu_{3\text{-}x}Co_xO_{7+\delta}$ over that of YBCO,[6] so if the oxygen migrates from the YBCO into $YBCu_{3\text{-}x}Co_xO_{7+\delta}$, T_c should be further lowered. The results were that T_c's of the bilayers made in these various methods were the same. Thus, oxygen doping is not the reason for the reduction of T_c.

The mutual influence of N and S as expressed in Figs. 1 and 2 is reminiscent of the conventional proximity effect.[8] We first consider which of our observations are generally consistent with this picture and which are not. To begin, we discuss the ''saturation thickness,'' namely the thickness of N above which the influence of N on S saturates (see inset of Fig. 2). In the proximity effect, T_c of a bilayer reflects the balance between the number of quasiparticles transmitted from N into S and pairs transmitted in the opposite direction. Pairs penetrate into N a distance of several normal coherence lengths, ξ_N. It is therefore plausible that the depth from which quasiparticles in N will reach the interface and penetrate into S should be quite similar. Values of ξ_N of 270 Å have been measured, for example, in c-axis-oriented Pr-Ba-Cu-O films,[9] so that the ~1000 Å that we find as the thickness of N at which the influence of N on S saturates appears to be within several such ξ_N. Thus, the ''saturation thickness'' of N seems within the bounds of what has been observed by other groups (the broader issue of comparing these values of ξ_N to what one would expect in the conventional proximity effect was discussed in Ref. 1). Three other observations reported here do not fit the conventional proximity effect. First, the depression of T_c of the bilayers does not depend on the Co doping level of the $YBCu_{3\text{-}x}Co_xO_{7+\delta}$, and hence on the T_c of the normal-metal material. This effect is not understood at present. Second, the depression of T_c of the bilayers seems to be very different for fast grown and slowly grown bilayers. Third, a large depression of T_c's of the fast grown bilayers occurs for YBCO layers much thicker than the coherence length of S. The remainder of the paper is devoted to discussing possible explanations of the last two effects mentioned.

We first turn to discuss the reason for the difference between the slowly grown and fast grown bilayers. The morphology of the surface of slowly and fast grown single-layer films, measured by AFM, is shown in Fig. 3 (these surfaces are the interface in the bilayers). There is indeed a striking difference between the features visible on the surface of the fast grown film, which are rounded and isotropic, while the slowly grown film shows a very regular array of pyramids, reflecting the symmetry of the underlying lattice. Despite the difference in the details, the averaged interface roughness of the fast grown and slow grown bilayers is quite similar. For example, the rms roughness of a slowly grown film 250 Å thick is 24 Å, while for a fast grown film of the same thickness it is 30 Å. Consequently, if all that mattered for the coupling between S and N layers was the area of the interface, the reduction of T_c should be very close for the two types of films. Clearly, this is not the case. We propose that the difference between these two types is unique to anisotropic superconductivity.

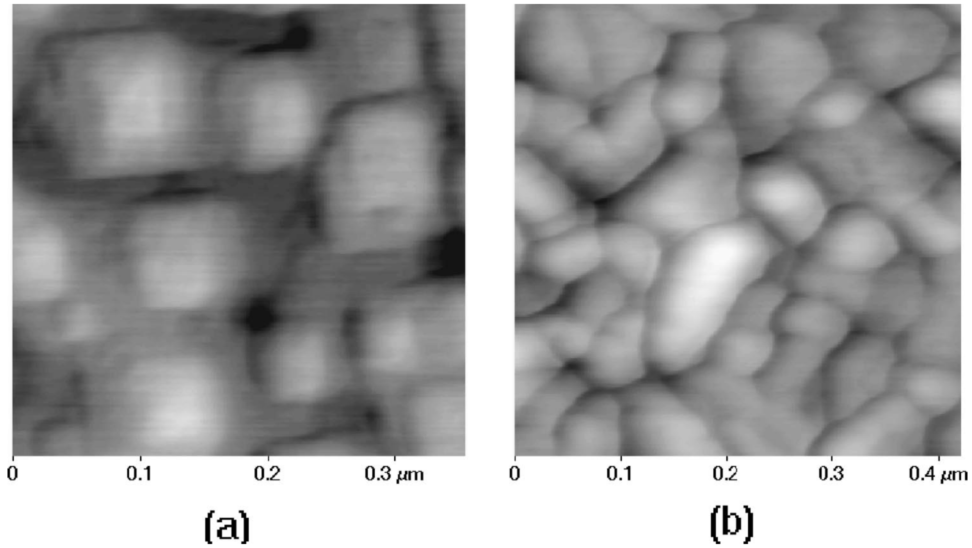

FIG. 3. Atomic force microscope pictures of the surface of the single-layer slowly grown film (a) and the fast grown film (b). These surfaces are the interfaces in the bilayer geometry. The average thickness of both films is about 500 Å, and the scale of the features shown represents ±10% thickness variation.

In the context where S and N are in proximity, the decrease of T_c of the S layer is due to the presence of an excess number of normal quasiparticles, transmitted from N into S.[8] When the order parameter is isotropic, the orientation of the interface is not an important factor in the transmission of quasiparticles from N into S. However, in the case of an anisotropic order parameter (d-wave, or $s+d$), Tanaka and Kashiwaya[2] have predicted that the transmission coefficient is strongly dependent on θ, the angle between the high symmetry crystalline directions (to which the order parameter is locked) and the normal to the interface. Essentially the same conclusion was reached by Barash *et al.*[10] In particular, the transmission through the interface should be anomalously large in the directions along which the order parameter has a minimum ($\theta=\pi/4$, where the interface normal is parallel to the diagonal between **a** and **b** in the case of a d-wave or $s+d$ order parameter). Depending on the value of interfacial potential barrier, the transmission along these diagonal directions can be tens of times larger than along the high symmetry directions.[2,11] It was shown recently that this effect is responsible also for the zero bias anomalies found in high-T_c tunnel junctions.[12] On the other hand, when the interface normal is parallel to **a** or **b**, the transmission will be similar to that in isotropic superconductivity. The anisotropy of the transmission survives the summation over all angles at which quasiparticles are incident on the interface, because their pair potential is symmetric about the crystalline axes, and therefore is not symmetric about the normal to the interface, which means the result of the summation will depend on θ, unless this normal and one of the crystalline high symmetry directions happen to coincide.[11] On the basis of these models, two additional statements can be made regarding the interfaces in our bilayers. First, Barash *et al.*[10] argue that the anisotropy would survive the averaging over all incidence angles only when the interface scattering is specular. On the basis of the differences between the slowly grown bilayers and the fast grown ones, this condition is fulfilled by the interfaces in our experiment. Second, the transmission through the interface[2,11] depends linearly on θ, being low for small values of θ and maximum for $\theta=\pi/4$. The large difference between the fast grown and slowly grown bilayers arises since most of the interface of the slowly grown bilayers is oriented with θ close to zero, as seen in Fig. 3, while in the fast grown bilayers all crystalline directions are equally exposed. Therefore, the transmission through the interface of the fast grown bilayers is much larger and their T_c would correspondingly be lower. We find it remarkable that just by changing the morphology of the surface the film can switch from normal to anomalously high transparency. Our data therefore illustrate rather vividly the crucial role of anisotropic superconductivity.

We now discuss the question of the length scale inside S where T_c is reduced. Assuming the dirty limit and perfect matching at the interface, the reduction of T_c of a bilayer, ΔT_c, expected due to the proximity effect is given by $\Delta T_c/T_{c0}\approx 1.35\ \xi_c^2(0)/d_S^2$, where d_S is the thickness of the superconducting layer, $d_S \gg \xi_c(0)$, and T_{c0} is the transition temperature of a single-layer thick film.[8] In the case of a perfectly smooth, c-axis oriented film, $\xi_c(0)\sim 3$ Å, and the predicted decrease of T_c of a 100 Å film is about 0.1 K. If the S/N interface is not planar, some in-plane coupling will be present, extending the range of the influence of N on S. To obtain a quantitative comparison, we extended the calculation of ΔT_c for the case when the S/N interface is rough. We use the Ginzburg-Landau approach and describe this interface by a function $z_S(x,y)=d_S+f(x,y)$, where the directions of x and y are chosen so that $\hat{\mathbf{x}} \parallel \mathbf{a}$, and $\hat{\mathbf{y}} \parallel \mathbf{b}$. We also assume that $|f(x,y)| \ll d_S$, and that the value of $f(x,y)$ averaged over the interface surface $\langle f\rangle=0$. In this case $\Delta\ T_c$ is given by[13]

$$\frac{\Delta T_c}{T_{c0}} \approx 1.35\,\frac{\xi_c^2(0)}{d_S^2} + 0.72\,\frac{\xi_a^2(0)\langle f_a^2\rangle + \xi_b^2(0)\langle f_b^2\rangle}{d_S^2}, \qquad (1)$$

where $f_i=\partial f/\partial x_i$. A direct measurement of the roughness of the film is not viable on the scale of $\xi_a(0)$ [or $\xi_b(0)$]. However, one can set a limit using the fact that the surface of the film is composed of unit cell size steps in the **c** direction on planar terraces. The density of steps on the surface can be calculated from the macroscopic inclination of the film surface relative to the substrate. Using the AFM pictures of the films such as shown in Fig. 2, we calculated $\langle f_a^2 \rangle$ as a function of d_S. For example, for $d_S=500$ Å we find $\langle f_a^2 \rangle \sim 1.3$ for a fast grown film and 0.5 for the slow grown one. For $\langle f_a^2 \rangle \sim 1$, the second term in Eq. (1) is much bigger than the first one. The result of the calculation of $T_c = T_{c0} - \Delta T_c$ is shown in Fig. 1 as the solid line. We took for this calculation $\xi_a(0) = \xi_b(0) = 20$ Å and $T_{c0}=90$ K. We find that if one takes the interface roughness into account, then the conventional proximity effect is consistent, without any adjustable parameters, with the dependence of T_c on d_S for the slowly grown bilayers. This result is also consistent with the transmission through the interface being normal in this case, namely similar to that found in isotropic superconductors. However, the proximity effect cannot account for the data of the fast grown bilayers, where the transmission through the interface is strongly enhanced. It is therefore an open question whether the fast grown and slow grown films should be treated on the same footing (in this case, the proximity effect does not work) or whether the fast grown films should be described by a totally different theory.

In equilibrium, the length scale describing the influence of N on S is $\sim \xi_0$. Looking at T_c's of the fast grown bilayers in Fig. 1, one can see a crossover between the thin film regime, where T_c is very close to that of $YBCu_{3-x}Co_xO_{7+\delta}$, about 50 K for $x=0.15$, and the thicker film regime, where T_c approaches that of YBCO. This crossover takes place at $d_S \sim 300$ Å, which clearly is much larger than ξ_0. This is essentially why the proximity effect fails to describe this case. We know of no theory pertinent to these experimental observations. However, we may mention that in the context of the usual s-wave superconductivity, Blonder *et al.*[14] considered another characteristic length, that for the direct conversion of a current of normal-metal quasiparticles entering S into pairs. This length is given by $\Lambda_Q = \sqrt{D\tau_Q}$. Here, D is the diffusion coefficient and τ_Q is the relaxation time which characterizes the return to equilibrium of the quasiparticle distribution (''branch imbalance relaxation time''). We calculated this length both for in-plane and out-of-plane diffusion. In the plane, $D_{ab} = v_F l/3$, where v_F is the Fermi velocity, and l is the mean free path. Taking $v_F = 5 \times 10^7$ cm/sec,[15] $l \sim 6 \times 10^{-7}$ cm, and $\tau_Q = 5$ psec,[16] we find the in-plane value of $\Lambda_Q(a-b)$ is about 700 Å. Regarding the c direction, quasiparticles diffuse mainly via interlayer scattering with a diffusion coefficient $D_c \sim c^2/t_c$.[17] Here, $c=12$ Å is the interlayer distance and $1/t_c$ is the scattering rate. Taking $t_c = 1.5 \times 10^{-14}$ sec,[17] we find $\Lambda_Q(c) = 220$ Å. Experimentally, τ_Q is constant[16] throughout the temperature range of this work, and therefore Λ_Q is also temperature independent. It is evident that the calculated values of Λ_Q are comparable to the experimental crossover length of 300 Å. However, Ref. 14 describes the conversion of normal-metal current into a supercurrent in a BCS s-wave superconductor, whereas our case brings out out the dramatic difference between the isotropic and anisotropic superconductivity. Thus, altough the numbers are suggestive, it would clearly be imprudent to identify this length as characterizing our data without a model more specific to our case. We hope that this work may in fact stimulate interest to do this calculation.

In conclusion, we demonstrated a dramatic effect of the morphology of the S/N interface on the properties of the bilayers. It is obvious that one has to consider this effect in the future design of high-T_c Josephson junctions.

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