Superconductivity enhancement in thin films of niobium in superconducting double-barrier structures

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We report the experimental results on the investigation of superconducting double-barrier structures with niobium interlayer and Al₂O₃ barriers. The experimental results can be explained by an extreme enhancement of superconductivity in the interlayer in accordance with the quasiparticle balance model, where the energy dependence of the inelastic relaxation time of quasiparticles with energy close to the superconducting gap is taken into account. [S0163-1829(96)06742-2]

I. INTRODUCTION

The enhancement of superconductivity has been observed in thin films of Al, Sn, and In both under microwave radiation and quasiparticle injection. It is explained by the changing of a quasiparticle energy distribution function (via an electron-phonon interaction) from the Fermi form under external influence (see, for example, Refs. 1 and 2). There have been no any observations of the enhancement of superconductivity in thin films of superconductors with a short mean free path (l<10 nm), for example, Nb. In this case the quasiparticle energy distribution function keeps the Fermi form, because of a strong electron-electron interaction in the film.³

As was analyzed by Parmenter,⁴ the enhancement of superconductivity could be intensified in the thin film of superconductor S′, placed between two superconducting electrodes S (a superconducting double-barrier structure S/S′/S, where the solidus represents a tunnel barrier). This effect is caused by the injection of quasiparticles with high energy into superconductor S′ and extraction of quasiparticles with small energy from it. The latter process is equal to an effective cooling of the interlayer. Zaitsev made a theoretical analysis of this effect in superconducting double-barrier structures (SDBS’s) on the basis of a microscopic theory.⁵ Calculations on the basis of the quasiparticle balance model were carried out by Heslinga and Klapwijk.⁶ The first experimental detection of the extreme enhancement of superconductivity was observed in the Nb/Al/Nb structure, where a sufficiently high degree of the nonequilibrium state of quasiparticles in an Al interlayer is realized: The injection (extraction) rate of quasiparticles through the tunnel barrier \( \Gamma = v_F D/d \) (\( d \) is the thickness of the Al interlayer, \( v_F \) is the Fermi velocity, and \( D \) is the barrier transparency) is greater than the reverse of the inelastic relaxation time of quasiparticles in the interlayer \( \tau^{-1}_{\text{in}} \) (\( \Gamma \tau_{\text{in}} > 1 \)). For temperatures higher than the critical temperature of the interlayer, \( T_c' \), the experimentally observed superconducting gap in Al increases up to 60% of the gap at \( T = 0 \).⁷⁻⁹

In this paper we present the results of the experimental investigation of the \( I-V \) curve of Nb/Nb′/Nb SDBS’s at various temperatures. The inelastic relaxation time in Nb is about 20 times smaller than in Al. So the case of a low degree of the nonequilibrium state of the interlayer in SDBS’s is realized, although the tunnel barriers transparency in Nb/Nb′/Nb SDBS’s are the same order of magnitude as for the Nb/Al/Nb one. The obtained temperature dependence of the gap in the interlayer could be explained by the enhancement of superconductivity in a thin niobium film even at a low degree of the nonequilibrium state (\( \Gamma \tau_{\text{in}} < 1 \)).

II. EXPERIMENTAL RESULTS

Nb/Nb′/Nb SDBS’s with Al₂O₃ tunnel barriers of 2–3 nm thicknesses are made as described elsewhere.¹⁰ Briefly, a five layer structure consisted of Nb and Al₂O₃ layers evaporates in situ. A lift-off technique, plasma and chemical etching, anodization processes, and SiO evaporation are used to define the area of the SDBS (\( S = 10 \mu \text{m}^2 \)). \( I-V \) curves and voltage dependences of the differential resistance \( R_d \) are measured at the temperatures \( T = 4.2–10 \) K for the thicknesses of the interlayer \( d = 10 \) and 20 nm. An electromagnetic screening and filtration of the biasing circuits decrease the external noise.

Table I presents some parameters of the investigated samples. The normal resistance of the SDBS is measured as an asymptotic of \( R_d \) at high voltages \( V \gg \Delta/e \). The procedure for the determination of the critical temperatures of the electrodes \( T_c \) and the interlayer \( T_c' \) will be discussed later. We estimate the transparency of the barriers \( D \) using an equation for the normal resistance of SDBS’s.⁸⁻¹¹

<table>
<thead>
<tr>
<th>Sample</th>
<th>J6N1</th>
<th>J6N3</th>
<th>J6N5</th>
<th>H3N1</th>
</tr>
</thead>
<tbody>
<tr>
<td>( d ) (nm)</td>
<td>20</td>
<td>20</td>
<td>20</td>
<td>10</td>
</tr>
<tr>
<td>( R_n(\Omega) )</td>
<td>6.5</td>
<td>6.0</td>
<td>6.1</td>
<td>15.3</td>
</tr>
<tr>
<td>( T_c ) (K)</td>
<td>8.5</td>
<td>8.5</td>
<td>9.2</td>
<td>9.2</td>
</tr>
<tr>
<td>( T_c' ) (K)</td>
<td>7.0</td>
<td>7.0</td>
<td>7.4</td>
<td>9.0</td>
</tr>
<tr>
<td>( D(10^{-6}) )</td>
<td>4.1</td>
<td>4.4</td>
<td>4.4</td>
<td>1.7</td>
</tr>
<tr>
<td>( \Gamma \tau_{\text{in}}(10^{-2}) )</td>
<td>3.1</td>
<td>3.3</td>
<td>3.3</td>
<td>1.3</td>
</tr>
</tbody>
</table>

Table I. Parameters of the investigated SDBS samples: \( d \), the thickness of the Nb′ interlayer; \( R_n \), the resistance of SDBS’s in the normal state; \( T_c \) and \( T_c' \), the critical temperatures of the superconducting electrodes and the interlayer correspondingly; \( D \), the barrier transparency; and \( \Gamma \tau_{\text{in}} \), the nonequilibrium parameter of the Nb′ interlayer.
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FIG. 1. (a) The $I$-$V$ curves of the SDBS $J6/N5$ with the thickness of the Nb interlayer $d=20$ nm at $T=4.2$ K. The dotted line presents the hysteresis part of the $I$-$V$ curve. $I_c$ is the critical current of SDBS’s without an external magnetic field. The temperature dependences of the gap features for the same sample are shown in the inset: 1, $V=V_{+}=\left(\Delta_1+\Delta_2+2\Delta’\right)/e$; 2, $V_{-}=\left(\Delta+\Delta’\right)/e$; 3, $V_{-}=\left(\Delta_1+\Delta_2-2\Delta’\right)/e$; 4, $V_{-}=\left(\Delta-\Delta’\right)/e$. Two lines for each feature caused by the limited precision in the determination of $T_c$. The size of the marks for the experimental points in the inset (0.2 mV) corresponds to the experimental precision in determination of the gap features position from the dip on the derivative curve $R_d(V)$. (b) $R_d(V)$ dependences for the same SDBS’s at different $T$: 1, 7.46 K; 2, 7.28 K; 3, 6.97 K; 4, and 6.72 K.

$$R_N = \frac{d\rho}{S} \left(1 + \frac{2l}{3dD}\right), \quad (1)$$

where $l=d$ and $\rho$ is the resistivity of the interlayer Nb film ($\rho=10^{-7} \Omega \text{cm}$).

The $I$-$V$ curve of the SDBS $J6/N5$ ($d=20$ nm) at $T=4.2$ K is shown in Fig. 1(a). The form of the $I$-$V$ curve corresponds to two superconducting tunnel junctions Nb/Nb’, connected in series: At $V=0$ we observe a critical current due to the Josephson effect; as the current increases a hysteresis on the $I$-$V$ curve (a dotted line in Fig. 1) is replaced by a gap feature at $V=4.5$ mV. A linear part of the $I$-$V$ curve is observed at higher voltages. The essentially vertical character of the gap feature indicates the small tolerance for parameters of tunnel Nb/Nb’ junctions in SDBS’s. The value of this gap feature ($V=4.5$ mV) is approximately twice that of the gap for a single Nb/Nb’ junction. There are also two other features on the $I$-$V$ curve at voltages $V_{+}=\left(\Delta+\Delta’\right)/e$ and $V_{-}=\left(\Delta_1+\Delta_2-2\Delta’\right)/e$, where $\Delta_1=\Delta_2=\Delta$ and $\Delta’$ are the superconducting gaps of the electrodes and interlayer, respectively. Furthermore, as the critical current is decreased by an external magnetic field or temperature, the gap feature at $V_{-}=\left(\Delta-\Delta’\right)/e$ is observed. The gap features $V_{+}$ and $V_{-}$ could be explained by the existence of subharmonic gap features $V_{+}=V_{-}/2$ and $V_{-}=V_{-}/2$ in Nb/Nb’ junctions. They could be presented on the $I$-$V$ curve because of a multiparticle tunneling and intensified by a multiple Andreev reflection in superconducting tunnel junctions with a high transparency of the barrier, $R_N S \approx 100 \Omega \mu \text{m}^2$. The barriers in our SDBS’s answer this condition. The temperature dependences of all features confirm the suggestion of their gap nature [see the inset to Fig. 1(a)].

The temperature dependences of the gap features near $T_c$ are measured from $R_d(V)$ [see Fig. 1(b)]. From Fig. 1(b) we can conclude that the difference between the critical currents (or the normal resistances) for two barriers in our SDBS’s is small; at $V>V_{-}$ both tunnel junctions are in a

FIG. 2. The deviation of the experimental values of the superconducting gap in the interlayer ($\Delta’$) from the BCS theory one ($\Delta_0$) at different voltages for the investigated samples with the thicknesses of the interlayer: $d=20$ nm ($J6/N1$, $J6/N3$, and $J6/N5$ samples) and $d=10$ nm ($H3/N1$ sample). The experimental results for the SDBS $J6/N5$ at $T=7$ K are marked by a solid circle for $V=V_{+}=\left(\Delta+\Delta’\right)/e$, and by the open circles both for $V=V_{+}=\left(\Delta_1+\Delta_2+2\Delta’\right)/e$ and $V=V_{-}=\left(\Delta_1+\Delta_2-2\Delta’\right)/e$. The calculated curves for $\tau_{\text{ad}}(E)=\text{const}$ and $\tau_{\text{ad}}(E)=\tau_{\text{ad}}(1+\left[\Delta’/(\Delta-\Delta’)^2\right])$ at $\Gamma \tau_{\text{ad}}=10^{-2}$ are shown by dashed and solid lines, respectively.
resistive state. There is no sharp increase of $R_d$, which is observed if one of the tunnel junctions goes into resistive state. $T_c$ and $T'_c$ are determined from $R_d(V)$ at various $T$: $T_c = 9.2$ K is determined from the deviation of $R_d(V)$ from $R_d = R_N$ [see curve 1 in Fig. 1(b)], and $T'_c = 7.4$ K from the onset of the critical current on the $I$-$V$ curve $R_d < R_N$ at $V = 0$ [see curve 2 in Fig. 1(b)]. In the temperature range $T'_c < T < T_c$ the electrodes are superconductors, but the interlayer is in the normal state. $R_d(V)$ has the maximum value at $V = 0$, and it corresponds to two superconductor/normal-metal (Nb/Nb') tunnel junctions connected in series. It is a well-known fact that the gap features for superconductor/superconductor junctions are more pronounced than for superconductor/normal-metal junctions. Thus, the absence of experimental data for gap features in the inset to Fig. 1(a) is caused by a smearing of the gap features and uncertainty in the determination of the gap near $T_c$. At $T < T'_c$ the interlayer is a superconductor, and so all gap features appear on $R_d(V)$ [curves 2–4 in Fig. 1(a)]. The temperature dependences of $V_{-}$, $V_{+}$, $V_{++}$, and $V_{-}$ are shown in the inset to Fig. 1(a). The solid lines correspond to the calculations from the Bardeen-Cooper-Schrieffer (BCS) theory of superconductivity (two lines for each feature caused by a limited precision in the determination of $T_c$: 0.1 K). We use the following approximation for $\Delta_b(T)$:

$$\Delta_b(T) = \begin{cases} (1 - T^{2.75})^{0.5}(0.9847 + 0.1577T - 0.0953T^2), & 1 > T > 0.7, \\ (1 - T^{3.3})^{0.5}(0.971 + 0.1786T - 0.2035T^2), & 0.7 > T > 0.36, \\ 1 - 1.89T^{0.5} \exp(-1.76/T), & 0.36 > T. \end{cases}$$

The experimental data for $(\Delta + \Delta')/e$ at $6K < T < 7.4$ K obviously exceed the BCS theory, and so the enhancement of the superconducting gap $\Delta'$ is observed in the interlayer Nb'.

Figure 2 presents the deviations of $\Delta'(V)$ from the BCS theory, obtained from experimental data for $V_{-}$, $V_{+}$, $V_{++}$, and $V_{-}$ for all investigated samples. The suppression of the gap is not presented in Fig. 2. According to the theoretical calculations, there is no enhancement of superconductivity for voltages $V > V_{++}$ and it is small for $V < V_{--}$ even in the case of strong quasiparticle injection. In our experiment, the enhancement of $\Delta'$ occurs in the range $2(\Delta - \Delta') < eV < 2(\Delta + \Delta')$ in good agreement with Refs. 5 and 6. The gap enhancement $(\Delta' - \Delta_b)$ is less than in the theory due to the absence of strong nonequilibrium state in the interlayer, as is suggested in Refs. 5 and 6. In our experiment $\Delta' - \Delta_b / \Delta_b < 1$ at $\tau_{\text{in}} = 5 \times 10^{-10}$ s at $T = 7$ K for Nb. The inelastic relaxation of the quasiparticles, injected to the interlayer of Nb', goes through electron-electron and electron-phonon interactions ($\tau_{\text{e-e}}^{-1} + \tau_{\text{e-ph}}^{-1}$), but in the case of tunnel injection (see estimations in Refs. 14–16) the electron-electron interaction is small in the Nb interlayer of SDBS's.

III. DISCUSSION

In the calculations $\tau_{\text{in}}$ is considered as a constant, independent of the energy of the quasiparticles in the interlayer. But it is known that the relaxation is slow at $E \sim \Delta$ due to the factor of coherency and divergence in the quasiparticle density of states (DOS). The following equation for $\tau_{\text{in}}$ takes into account these factors phenomenologically,

$$\tau_{\text{in}}(E) = \tau_{\text{in}0} \left[1 + \left(\frac{\Delta'}{|E - \Delta'|}\right)^{3.5}\right]^{-1},$$

under assumptions of a linear dispersion for phonons and a suggestion that the square of the matrix element for the electron-phonon interaction is proportional to the phonon wave vector. At $E - \Delta' \gg \Delta'$, $\tau_{\text{in}}(E)$ is the same as the electron-phonon relaxation time in normal metal, $\tau_{\text{in}0} = 5 \times 10^{-10}$ s at $T = 7$ K for Nb. The inelastic relaxation of the quasiparticles, injected to the interlayer of Nb', goes through electron-electron and electron-phonon interactions ($\tau_{\text{e-e}}^{-1} + \tau_{\text{e-ph}}^{-1}$), but in the case of tunnel injection (see estimations in Refs. 14–16) the electron-electron interaction is small in the Nb interlayer of SDBS's.}

FIG. 3. The experimental temperature dependence of the deviation of the superconducting gap in the interlayer $(\Delta')$ from the BCS theory $(\Delta' - \Delta_b)$ at $V = V_{+} = (\Delta + \Delta')/e$. The solid line presents the calculated results for $V = V_{+}$ at $\tau(\Delta / T) = \tau(1 + [(\Delta'/|E| - \Delta'|)^{3.5}].$ $\tau(\Delta / T) \approx 10^{-2}.$
The numerical calculation of the gap in the interlayer is made on the basis of the quasiparticle balance model: The number of quasiparticles injected into the interlayer (from the first electrode) must be equal to the total number of quasiparticles extracted from it (to the second electrode) and quasiparticles, relaxed to the Fermi level in the interlayer. The energy distribution function in the interlayer has the form

$$f'(E) = \frac{N_1(E-eV/2)f_0(E-eV/2) + N_2(E+eV/2)f_0(E+eV/2) + f_0(E)/\Gamma \tau_{in}(E)}{N_1(E-eV/2) + N_2(E+eV/2) + 1/\Gamma \tau_{in}(E)},$$

where $N_i (i=1,2)$ are the DOS functions in the electrodes, normalized to the value at the Fermi level in the normal state, $f_0(E)=[\exp(E/kT+1)]^{-1}$ is the Fermi function, and the relaxation process is supposed to be a linear function of $\tau_{in}$. Due to the condition $d\approx h\nu T D/4\Delta(0)$, we can neglect the proximity effect in SDBS's. To find $\Delta'$ we should solve the self-consistent equation

$$\int_0^{\infty} dE \left[ f'(E)\Theta(|E| - \Delta') - \frac{1}{E^2} \text{th} \left( \frac{E}{2kT} \right) \right] = 1 - \frac{T}{T_c},$$

where $\Theta(x)$ is the Heaviside step function. The results of the numerical solution of Eqs. (4) and (5) are shown in Fig. 2 (solid line) for $d=20$ nm at $T=7$ K. Three points for the SDBS J6/66 ($d=20$ nm) for $T=7$ K (which correspond with the calculation conditions) are marked in Fig. 2. There is good agreement between the calculated and the experimental results for $\Delta'$ obtained from $V_+$ (solid point in Fig. 2). The gap value of the interlayer at $V_+$, $T=7$ K is approximately 93% of the equilibrium one at $T=0$ K.

The temperature dependence of $(\Delta'-\Delta_n)$, obtained from $V_+$, is shown in Fig. 3. The experimental points are very close to ones obtained from the numerical calculations with one fitted parameter $\tau_{in}$. The ratio of the nonequilibrium gap to the equilibrium one increases with $T$. It could be explained by the fact that the deviation of the quasiparticle distribution function from the Fermi form grows with the increase of $T$. The experimental values of $\Delta'$, obtained from $V_+$, $V_-$, and $V_-$, differ from the calculated ones possibly due to the phenomenological character of our dependence of $\tau_{in}(E)$.

IV. CONCLUSION

The experimental results of the investigation of the superconducting double-barrier structure Nb/Nb$^'/$/Nb indicate the enhancement of superconductivity in thin Nb$^'/$/Nb films. Reasonable agreement is found of our experimental results to the quasiparticle balance model with the energy dependence of the inelastic relaxation time.

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