Universality of transport properties of ultrathin oxide films

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New Journal of Physics 14 (2012) 023025 (13pp)
Received 30 September 2011
Published 13 February 2012
Online at http://www.njp.org/
doi:10.1088/1367-2630/14/2/023025

Abstract. We report low-temperature measurements of current–voltage characteristics for highly conductive Nb/Al–AlOₓ–Nb junctions with thicknesses of the Al interlayer ranging from 40 to 150 nm and ultrathin barriers formed by diffusive oxidation of the Al surface. In a superconducting state these devices have revealed a strong subgap current leakage. Analyzing Cooper-pair and quasiparticle currents across the devices, we conclude that the strong suppression of the subgap resistance compared with conventional tunnel junctions is not related to technologically derived pinholes in the barrier but rather has more fundamental grounds. We argue that it originates from a universal bimodal distribution of transparencies across the aluminum oxide barrier proposed earlier by Schep and Bauer (1997 Phys. Rev. Lett. 78 3015). We suggest a simple physical explanation of its source in the nanometer-thick oxide films relating it to strong local barrier-height fluctuations in the nearest to conducting electrode layers of the insulator which are generated by oxygen vacancies in thin aluminum oxide tunnel barriers formed by thermal oxidation.
1. Introduction

In the past decade, there has been considerable progress in understanding the fundamental
electronic properties of ultrathin dielectrics, in particular, charge transport across disordered
oxide layers. The increasing interest in such materials has been motivated by their promising
applications as a gate dielectric in metal-oxide–semiconductor transistors instead of SiO$_2$
and a blocking dielectric for new-generation flash memory cells (see the papers [1, 2] and
references therein) as well as a potential barrier for current carriers in superconducting
tunnel junctions [3, 4]. In particular, it relates amorphous nanometer-thick alumina AlO$_x$.
Unfortunately, in practice, high demands on the ultrathin oxide films as current-blocking high-$k$
dielectrics with minimal dissipation are not usually fulfilled and a strong leakage current
through the amorphous oxides is emerging as a potential limitation for most novel applications
[1, 2, 4]. This finding has often been attributed to microscopic defects in the tunnel barrier with
greatly enhanced local transparency [5], commonly known as ‘pinholes’. It has been assumed
that the pinholes shunt the tunnel current and thus dominate the charge transport across the
junctions. But a recent extensive analysis of differential subgap conductance in highly resistive
superconducting tunnel junctions with aluminum oxide barriers [4] has ruled out pinholes as
the origin of the excess current, at least, in samples with normal conductance $G_N$ per geometric
area $A$ between $10^4$ and $10^7$ (ohm cm$^2$)$^{-1}$. Hence, the identification of the charge transport
mechanism in ultrathin oxide films as well as the nature of defects in amorphous dielectrics
such as AlO$_x$ continues to be of much scientific and practical interest.

In this paper, we study charge transport across extremely thin AlO$_x$ layers sandwiched
between two metallic electrodes with specific normal conductance ranging from $2 \times 10^7$ to
$7 \times 10^7$ (ohm cm$^2$)$^{-1}$. This means that in our case the barrier thicknesses were significantly
reduced compared to those in [4] and, hence, the presence of pinholes was more probable.
Below, we not only reject the hypothesis about a dominating role of microscopic pinholes in
ultrathin oxide layers but also use our experimental findings for four-layered Nb/Al–AlO$_x$–Nb
junctions to show that the subgap resistance suppression in a superconducting state compared to
conventional tunnel junctions originates from a universal bimodal distribution of transparencies
across the oxide barrier.

Concerning the latter statement, we should note that in the general case, quantum transport
through a mesoscopic physical system is determined by different factors such as the system
dimensionality, its size and shape, carrier density and other sample-specific features. Universal
transport properties, if they exist, should be independent of the microscopic details of particular
materials and may include only a limited number of macroscopic characteristics. Inter alia, the
universal can be expected for ‘dirty’ systems with a very large spread of random potentials
scattering current carriers. In the seminal paper by Schep and Bauer [6] the authors studied
quantum transport properties of a single perfectly disordered insulating (I) interface whose conductance is much smaller than the conductance of a ballistic conductor of the same cross-section and whose thickness \( d \) is sufficiently shorter than the Fermi wavelength \( \lambda_F \) in metallic bulks around it. They found that the distribution \( \rho(D) \) of eigenvalues \( D \) of a transfer matrix connecting incoming and outgoing electronic modes (it is the product of the related transmission amplitude matrix and its Hermitian conjugate) does belong to a universality class and is given by an expression

\[
\rho(D) = \frac{\hbar G_N}{e^2} \frac{1}{D^{3/2}(1 - D)^{1/2}}
\]

with \( G_N \) the disorder-averaged conductance in a normal (N) state. For a sample with such an interface any physical quantity \( f \) described by linear statistics \( f(D) \) (such as conductance or shot noise power) can be calculated as

\[
\int_0^1 f(D) \rho(D) \, dD.
\]

The best way to study the distribution \( \rho(D) \) in a nanometer-thick insulating film is to place it between two metallic layers and to measure current–voltage (I–V) characteristics of the tunnel junction at very low temperatures when one or both electrodes are transformed into a superconducting (S) state. The reason is that the shape of quasiparticle I–V curves for SIS trilayers with a single quantum channel is extremely sensitive to the transmission probability [7] and, if the total current across the junction can be represented as a sum of independent contributions from individual transverse modes, its voltage dependence would be definitely determined by the function \( \rho(D) \).

A first analysis of the \( \rho(D) \) distribution in subnanometer-thick AlO\(_x\) layers using a superconducting heterostructure was performed by Naveh et al [8] who measured current–versus–voltage and differential conductance–versus–voltage characteristics of planar highly conductive Nb–AlO\(_x\)–Nb trilayers at 1.8 K. In spite of the presence of an AlO\(_x\) barrier, the quasiparticle I–V curves did not exhibit behavior of typical conventional tunnel junctions [7] with \( R_{sg} \gg R_N \). Here \( R_{sg} \) is the junction differential resistance at voltages \( |V| < 2\Delta/e \), known as a subgap resistance, \( \Delta \) is the energy gap of a superconducting electrode, in this case niobium, and \( R_N \) is the trilayer normal-state resistance. In contrast, the ratio \( R_{sg}/R_N \) in the state when the Josephson critical current was totally suppressed by magnetic fields was near unity. Moreover, conductance–voltage curves exhibited well-pronounced subharmonic gap structure known as a fingerprint of multiple Andreev reflections in highly-transparent SIS samples [9–11]. All these findings were explained [8] as the impact of a broad double-peak distribution of aluminum oxide barrier transparencies described by equation (1).

It should be noted that the presence of a striking, polarity-independent deviation from standard single-particle tunneling I–V curves in SIS junctions is well known from the 1960s [12]. The subgap leakage was observed not only in highly conductive junctions [3] but also in low-transmission Nb–AlO\(_x\)–Nb trilayers [13] and in both cases evidence of an important role played by multiple Andreev-reflection processes has been reported [4, 8] although until now it has not been clear if their nature was identical or not [14]. In recent years, there has been a resurgence of interest in this effect since enhanced quasiparticle subgap conductance within individual junctions can cause decoherence effects in Josephson-effect-based superconducting quantum devices [15]. The ‘quality factor’ of an SIS junction, which is usually defined by the ratio \( R_{sg}/R_N \), is also important for low noise performance of SIS mixers and detectors where it is required to be at least \( \sim 15 \) for low noise performance [16]. At the same time, to achieve a broad rf-bandwidth, the junction must have a very high critical-current density \( J_c \)
or a small $R_N A$ product, where $A$ is the junction area [17]. The two requirements contradict each other because of a strong positive correlation between the ‘junction quality’ and its specific conductance [3]. Another aspect of the same problem relates to the implementation of Josephson junctions operating in an overdamped mode when the ratio $R_{sg}/R_N$ should be suppressed as much as possible [18]. Since thermally oxidized aluminum oxide tunnel barriers provide the most reliable junction fabrication technology due to its reasonably good properties and comparatively easy fabrication [3], more fundamental knowledge about the possibility of changing the ratio $R_{sg}/R_N$ in a controllable way is needed in order to provide more insight into the intrinsic structure of the AlO$_x$ insulating layer [19] as well as into the nature of current fluctuations in related superconducting devices [20, 21].

Earlier investigations of subgap processes have mainly concentrated on symmetric S–I–S [3] and asymmetric S–I–N [22] junctions. At the same time, it was stressed [23] that experimental studies of asymmetric $S_1$–I–$S_2$ devices can provide more information since the structure of $I$–$V$ curves at voltages below $(\Delta_1 + \Delta_2)/e$ is richer and more pronounced [24, 25]. The aim of this work is to reexamine the conclusion of [8] about very large variations of local barrier transparencies in nanometer-thick AlO$_x$ layers, using asymmetric Josephson junctions with a proximity-coupled SN bilayer as one of the device electrodes and studying the thermal effect in SNIS devices. Compared with our previous experiments [18, 26, 27], we have enlarged the temperature interval studied, performing precise measurements of dissipative transport characteristics at temperatures significantly below and above 4.2 K. We have succeeded in extracting corresponding $R_{sg}/R_N$ values and comparing them with our numerical simulations, which are based on equation (1) applied to a more complicated than before four-layered junction geometry. Besides, we present the data of novel rf-measurements proving the Josephson character of charge transport across our samples in a superconducting state. The rest of the paper is devoted to a simple physical interpretation of the Schep–Bauer distribution (1), the primary focus of the paper, relating it to strong local barrier-height fluctuations generated by oxygen vacancies in extremely thin aluminum oxide films.

2. Experimental

We present experimental results obtained on asymmetric Nb/Al–AlO$_x$–Nb junctions developed at Instituto Nazionale di Ricerca Metrologica [26]. The main differences from standard Nb technology [28] are the following: the thickness of the Al interlayer $d_{Al}$ was increased from 5–10 nm up to 40–150 nm, whereas the exposure dose, the product of the oxygen pressure and the oxidation time, was decreased from more than 1000 Pa s down to 150–500 Pa s (note that in [4] oxygen doses varying from $1.7 \times 10^3$ to $8 \times 10^5$ Pa s produced extremely thick barriers). Because of comparatively small exposure doses in our case, specific normal conductances of the Nb/Al–AlO$_x$–Nb devices were as high as $(2–7) \times 10^7$ (ohm cm$^2$)$^{-1}$, which is of the same order of magnitude as those of highly conductive symmetric SIS Josephson junctions studied by Naveh et al [8]. Further details of our 25 $\mu$m$^2$-area junctions can be found elsewhere [18].

Electrical measurements have been carried out with a conventional four-terminal dc technique below critical temperatures of Nb/Al bilayers, which were changed from 8 to 9 K for different $d_{Al}$. All samples have exhibited supercurrents with values of 6–15 mA at 1.7 K, 3–8 mA at 4.3 K and 1–5 mA at 6.0 K (left insets in figure 1). A typical current–voltage single-valued characteristic of a Nb/Al–AlO$_x$–Nb junction at finite temperatures is shown in the main
Figure 1. Dissipative current–voltage characteristics of a representative Nb/Al–AlO$_x$–Nb junction with $d_{Al} = 110$ nm at 1.7 K (a) and 4.3 K (b) measured in external magnetic fields of about 50 mT applied to the tunnel sample through a suitable coil. Left insets demonstrate hysteretic behavior of the superconducting current with switching $I_c$ and retrapping $I_r$ values, whereas the main panels show quasiparticle $I_{qp}$–$V$ characteristics with a suppressed Cooper-pair contribution (dotted curves). Right insets exhibit the same curves on an enlarged scale near $V = 0$. Solid and dashed-dotted straight lines correspond to Ohm’s laws with $R_{sg} = 95$ mohm and $R_N = 69$ mohm at 1.7 K (a) and $R_{sg} = 71$ mohm and $R_N = 64$ mohm at 4.3 K (b).

It is well known [29] that the subgap current $I_{sg}$ is a sum of three terms: a quasiparticle part $I_{qp}$, a superconducting pair current $I_c$ and an interference contribution. The latter two contributions can be made arbitrarily small if the critical current $I_c$ is reduced by applying a magnetic field $B$. Then $I_{sg} \approx I_{qp}$. In our measurements of dissipative current–voltage characteristics, magnetic fields $B$ up to 50 mT have been applied to the Nb/Al–AlO$_x$–Nb samples through a suitable coil. The $I_{qp}$–$V$ curves have not been changed for two configurations of the magnetic fields (parallel and orthogonal to the current flow). The normal resistance $R_N$.

New Journal of Physics 14 (2012) 023025 (http://www.njp.org/)
Figure 2. Typical current–voltage characteristics of a Nb/Al–AlO$_x$–Nb junction with a 110 nm-thick Al interlayer (the main panel) and that of a binary-divided array of the same 8192 SNIS junctions under 73 GHz microwave irradiation (the inset). Critical currents $I_c$ and the products $V_c = I_c R_N$ were about 2.2 mA and 0.2 mV, respectively, at the operating temperature of 6.3 K.

was determined from a linear fit to current–voltage curves with and without supercurrents at voltages above 1 mV (see figure 1) and the results of both estimations were in good agreement with each other. It also agrees with the data for Nb-based trilayers [3] where the specific conductance of thermally oxidized AlO$_x$ tunnel barriers was above $10^7$ (ohm cm$^2$)$^{-1}$ for the lowest oxygen exposure doses similar to ours. Subgap Ohmic resistance $R_{sg}$ was extracted from experimental data as the slope of a best-fit linear regression line for quasiparticle curves in the interval from 0 to 0.2 mV where the subgap current increases near linearly with $V$ (the right insets in figure 1). The ratios $R_{sg}/R_N$ at 4.2 K were always of the order of unity, as was found recently in our work on Nb/Al–AlO$_x$–Nb junctions with ultrathin barriers [18] and before that for high-transparency symmetric Nb–AlO$_x$–Nb samples in [3]. Complementing our previous 4.2 K data for $R_{sg}$ in SNIS junctions [18], in this work we have measured dissipative $I_{qp}$–$V$ curves also at 1.7 and 6.0 K and increased the upper value of $d_{Al}$ to 150 nm.

Below, we analyze experimental data for five representative SNIS junctions with very different values of $d_{Al}$, $R_N$, $I_c$ and products $I_c R_N$. It means that the samples had dissimilar Nb/Al interface resistances, as well as oxide barrier parameters, and our aim was to compare measured ratios $R_{sg}/R_N$ with those calculated using the universal distribution (1). Table 1 summarizes experimental characteristics of the samples, whereas the main panel of figure 1 shows measured $I$–$V$ characteristics for one of the SNIS devices (dots) with two linear fits corresponding to $I = V/R_N$ and $I = V/R_{sg}$.

3. Discussion

Conventional theory of tunneling effects in superconducting junctions [7] leads to the following conclusions relating the $I$–$V$ curves: firstly, at 1.7 K for $V \approx 0.2$ mV the ratio $R_{sg}/R_N$ should
Table 1. Measured parameters for Nb/Al–AlO$_x$–Nb junctions with different Al thicknesses.

| $d_{Al}$ (nm) | $I_c (B = 0)$ $R_N$, 1.7 K (mA) | $R_S$, 1.7 K (mohm) | $R_N$, 4.3 K (mohm) | $R_{sg}/R_N$, 1.7 K (mohm) | $R_{sg}/R_N$, 4.3 K (mohm) | $R_{sg}/R_N$, 6.0 K (mohm) |
|---------------|---------------------------------|---------------------|---------------------|--------------------------|--------------------------|--------------------------|
| 45            | 7.01                            | 136                 | 136                 | 132                      | 1.39                     | 1.36                     | 1.14                     |
| 57            | 6.13                            | 177                 | 177                 | 175                      | 1.31                     | 1.29                     | 1.07                     |
| 110           | 11.92                           | 69                  | 64                  | 63                       | 1.38                     | 1.11                     | 1.05                     |
| 115           | 14.37                           | 82                  | 80                  | 80                       | 1.30                     | 1.15                     | 1.03                     |
| 142           | 6.87                            | 181                 | 178                 | 177                      | 1.27                     | 1.13                     | 1.03                     |

be of the order of $10^5$–$10^6$ in Nb–I–Nb samples and $10^2$–$10^3$ in Nb/Al–I–Nb junctions, and secondly, the ratio has to decrease (exponentially for SIS contacts) with increasing temperature up to $\geq 10^2$ and $\leq 10^2$ at 4.3 K in Nb–I–Nb and Nb/Al–I–Nb devices, respectively. Our samples do not follow this behavior and exhibit (i) the ratio $R_{sg}/R_N$ of the order of unity at all temperatures and (ii) a very weak temperature effect.

To conclude that these observations cannot be attributed to pinholes, we should prove a sinusoidal current–phase relation $I_s(\phi)$ for our junctions. One of the ways of checking it is to detect Shapiro steps in the $I$–$V$ curves under microwave irradiation. Our experiments on SNIS single junctions rf radiated with a frequency of 73.5 GHz have displayed a clear harmonic step structure in the current–voltage characteristics up to 8.3 K without any subharmonic features [30]. In the inset of figure 2 we show Shapiro steps at $T = 6.3$ K for binary divided series arrays of 8192 Josephson SNIS junctions with $d_{Al} = 110$ nm, a part of the samples used for subgap resistance measurements.

It should be noted that similar results for the ratio $R_{sg}/R_N \simeq 1$ and its universality have already been observed in symmetric Nb–AlO$_x$–Nb junctions and interpreted in the paper [8] following calculations [10, 11] where the validity of the Schep–Bauer distribution (1) was assumed and the effect of multiple Andreev reflections important for high transparencies $D$ was taken into account. In particular, it follows from figure 2 of [11] that in SIS trilayers the $R_{sg}/R_N$ value averaged over the voltage interval $|V| < \Delta_{Nb}/e$ should be near 0.7 (the dashed line in figure 3). Experimental data for Nb–AlO$_x$–Nb samples (see figures 1 and 2 in [8]) did agree well with this estimate. Our aim was threefold: (i) to introduce an additional Al interlayer in order to modify the $R_{sg}/R_N$ ratio and to compare measured values with a calculated one; (ii) to change the thickness of the interlayer and to reveal in the experiment whether the distribution (1) comes from the disordered oxide layer or is generated by diffusive charge transport in the N region; and (iii) to investigate for the first time the temperature dependence of the $R_{sg}/R_N$ ratio.

In figure 3, we show experimental data for five representative samples which agree well with each other after normalization to $R_N$. In contrast to the observations for Nb–AlO$_x$–Nb devices [8], the subgap resistance in our samples was higher than the normal-state value. To explain the findings, in figure 3 we show linear fits to results of such numerical calculations for two limiting cases of NIS and SIS junctions (dotted and dashed lines, respectively). The $I_{qp}$–$V$ curve for an NIS trilayer was computed by us and that for an SIS structure was taken from [11]. For comparison we plot the current–voltage characteristic in a normal state as well (dashed-dotted line).
Figure 3. Quasiparticle current–voltage characteristics of five Nb/Al–AlO$_x$–Nb junctions with different thicknesses of the Al interlayer (indicated in the figure) at 1.7 K (symbols) compared with linear fits to calculated dependences for NIS, SIS [11], SNIS and NIN junctions with a disordered barrier layer (dotted, dashed, solid and dashed-dotted lines, respectively).

We have also performed numerical simulations of the subgap resistance in SNIS junctions with arbitrary transparency $D$ of the interspace between two superconducting electrodes and averaged the $I_{qp}-V$ curves with the relation (1). Since our approach to the problem is based on numerical methods developed earlier [9–11, 24, 25, 31], we shall only briefly outline the main points of the calculations. The Nb/Al–AlO$_x$–Nb system studied is considered as an asymmetric S$_1$IS$_2$ junction where S$_1$ is the S/N bilayer. Proximized Nb layer induces in a thin Al film a superconducting order parameter, which in the general case is a function of $\varepsilon$, the energy of a quasiparticle excitation in a superconductor measured from its Fermi level. The proximity effect changes the probability amplitude of a process of Andreev scattering $a_1(\varepsilon)$ from Al/Nb bilayer compared with that from a bulk superconductor and this is just our main modification of the calculation scheme proposed in the papers referred to above. The probability amplitude $a_1(\varepsilon)$ can be found if the ratio of modified and normal Green’s functions $F$ and $G$ of a superconductor $\Phi_1(\omega) = \omega F(\omega)/G(\omega)$ ($\omega$ is the Matsubara frequency) is known. Note that in a bulk superconductor $\Phi_1(\omega)$ is equal to a constant order parameter $\Delta_1$ [32]. The function $\Phi_1(\omega)$ of a dirty normal metal placed in proximity to a superconductor can be calculated using the Usadel equations with corresponding boundary conditions [32]. In the following, we use the simplest approximation for the function $\Phi_{Al}(\omega)$ in the Nb/Al bilayer $\Phi_{Al}(\omega) = \Delta_{Nb}/(1 + \gamma \sqrt{\omega^2 + \Delta_{Nb}^2} / \Delta_{Nh})$ with a fitting parameter $\gamma$ and the energy gap of Nb $\Delta_{Nh}$ [33]. The value of $\gamma$ can be found by equating the gap magnitude in the Al interlayer calculated numerically to that found by us experimentally (for Al-film thicknesses studied its value at 1.7 K can be roughly estimated as 0.4 meV [27]). Then we are dealing with an S$_1$IS$_2$ junction where $a_1(\omega) = i(\omega - \sqrt{\omega^2 + \Phi_{Al}^2 (\omega)}) / \Phi_{Al}(\omega)$ and $a_2(\omega) = i(\omega - \sqrt{\omega^2 + \Delta_{Nb}^2} / \Delta_{Nh})$. At first, we consider a single-mode channel with a fixed transparency $D$ between S$_1$ and S$_2$ electrodes. Its length is assumed to be much smaller than elastic and inelastic lengths. If so, we can describe the transport across the device in terms of Andreev-reflection amplitudes.
using Zappe's proposes that, independently of the presence and the it is clear that the approximation for \( (2) \) of the junction in a superconducting state is single valued like that shown in figure \( \text{R} \). Let us stress that while the \( R_{\text{sg}}/R_N \) value is almost the same for different \( d_{\text{Al}} \) at 1.7 K, its temperature behavior, as follows from table 1, is strongly different. For the lowest \( d_{\text{Al}} \), the deviation of the ratio value at 4.3 K from that at 1.7 K is tiny, whereas for larger thicknesses the \( R_{\text{sg}}/R_N \) value tends to unity. It can be explained as a result of a very different temperature behavior of the energy gap induced in an Al interlayer. It follows from figure 3 in \[ \text{C} \] that the temperature suppression of the induced gap and, hence, the asymmetry of SNIS superconducting junctions become stronger for thicker interlayers. As a result, in the subgap voltage range a strong peak evolves in \( I-V \) curves for comparatively small barrier transparencies (see figure 5 of \[ \text{D} \]) and this thermally induced decrease of the subgap resistance is more effective for larger \( d_{\text{Al}} \).

The disappearance of a hysteretic \( I-V \) response in Nb/Al–AlO\(_x\)–Nb Josephson junctions when increasing the temperature from 1.7 to 4.3 K (compare left insets of figures 1(a) and (b)) is partly an effect of the temperature influence on the \( R_{\text{sg}}/R_N \) ratio. It is well known \[ \text{F} \] that the Josephson-device operation regime is governed by the McCumber–Stewart damping parameter

\[
\beta_c = \frac{2e^2}{h}(I_c R_N^2 C)(R_{\text{sg}}/R_N)^2
\]  

(2)

with \( C \) being the junction capacitance. If \( \beta_c \) is less than unity the current–voltage characteristic of the junction in a superconducting state is single valued like that shown in figure 1(b). Otherwise, the response is hysteretic with switching \( I_c \) and retrapping \( I_r \) currents (figure 1(a)). We can estimate the \( \beta_c \) value for a representative junction shown in figure 1 using Zappe’s
The Schep–Bauer formula (1) for the transparency $D$ is not limited to this assumption but is valid, independently of the physical nature, when (i) the transparency may be represented as a one-parameter Lorentzian and (ii) this parameter is uniformly distributed from very small up to very large values (see the related discussion concerning spatial distribution of barrier defects in the paper by Il’ichev et al [36]). It is interesting that before the work [6], the same relation (1) was derived for a quasi-ballistic double-barrier INI interspace with two identical uniform insulating layers [37], a system that is physically very different from a thin disordered dielectric film. In our opinion, the reason for coincidence between the two systems consists in the fact that, for a finite thickness of the N interlayer $d_N$ the transmission coefficient $D$ is also of a Lorentzian-like form. To show it, we express the transmission amplitude $\tilde{t}$ across the $I_1N_2I_2$ transition region as a sum of a geometric series

$$\tilde{t} = t_1 \exp(i\varphi)t_2(1 + r_1 \exp(2i\varphi)r_2 + \cdots) = \frac{t_1 \exp(i\varphi)t_2}{1 - r_1 \exp(2i\varphi)r_2},$$

where $\varphi(\theta) = k d_N \cos(\theta)$ is the phase shift acquired by an electron with a wave number $k$ traveling between two interlayer boundaries; $t_{1,2}(\theta)$ and $r_{1,2}(\theta)$ are the transmission and reflection amplitudes for $I_1$ and $I_2$ insulating layers, respectively; $\theta$ is the injection angle between the vector $\mathbf{k}$ and the normal to layer interfaces. Then the probability of charge transition across the double-barrier $I_1N_2I_2$ trilayer can be expressed as $D(\theta) = (1 + \tilde{Z}^2(\theta))^{-1}$ with

$$\tilde{Z} = \sqrt{2 - |t_1|^2 - |t_2|^2 - 2 \text{Re} \{r_1r_2 \exp(2i\varphi)\}}/(|t_1| |t_2|).$$

For a sufficiently large thickness $d_N$ and symmetrical INI structure with low-transmission barriers, $\tilde{Z}(\theta)$ is a rapidly oscillating function, which changes periodically from zero in the resonance state to very high values. This very wide spread of the parameter $\tilde{Z}(\theta)$ is the reason why the two distributions coincide.
Now we proceed to experimental facts supporting the statement about a broad spread of local barrier transparencies in very thin dielectric layers and explaining its origin. First, we refer to a recent paper by Welander et al [38], where two ways of aluminum oxide barrier formation in Nb/Al–AlOₓ–Nb junctions were employed. The first process based on the conventional diffusion-limited oxidation of the Al layer yielded Josephson tunnel junctions with significant subgap leakage, whereas the subgap currents in devices with layer-by-layer grown barriers agreed well with a standard tunneling theory. According to [38], the extra conductance most probably comes from defects in the diffused Al oxide caused by room-temperature thermal oxidation of Al. These defects are known to be positively charged oxygen vacancies [19]. Another work [20] supporting the idea of the decisive role of oxygen vacancies deals with low-frequency noise measurements in Al–AlOₓ–Al tunnel junctions. It was found that vacuum thermal annealing strongly reduces the 1/f noise level in the Al-based devices (sometimes up to an order of magnitude). Since the 1/f noise phenomenon in metal–insulator–metal tunnel junctions is definitely related to the slow filling and emptying of localized electron states in the barrier [39], the finding by Julin et al [20] can be understood as a reduction of the charge traps number within the AlOₓ layer by the annealing procedure.

Using the conductive atomic force microscopy technique, Kim et al [40] studied local tunneling properties of ultrathin MgO films. Topographic maps showed that MgO layers were very flat, whereas local tunnel current maps were strongly inhomogeneous with a number of current hotspots without any correlation between the two maps for the same sample. The most probable explanation can be that the tunnel current inhomogeneity arises from fluctuations in barrier height but not structural fluctuations. Comparing the maps before and after adding O₂ to the Ar plasma during MgO growth, the authors of [40] argued that the inhomogeneity originates from oxygen vacancies. Elimination of certain oxygen defect populations leads to improved barrier heights with reduced spatial variations. An additional argument relating the presence of defect states in a very thin MgO tunnel barrier comes from the nonlinearity of I–V characteristics of corresponding magnetic tunnel junctions. The presence of oxygen vacancies in MgO leads to a new tunneling transport mechanism within the film when a charge is transported across the classically forbidden region via boson-induced tunneling jumps between randomly distributed localized states [41]. The combination of direct and inelastic tunneling events through the defects should lead to anomalous voltage dependence of the differential conductance in the form of a sum of power functions of V with different non-integer exponents. Such behavior was just observed in [42] for magnetic tunnel devices with MgO barriers. Very recent examples proving the relation of anomalous properties of thin oxide layers to oxygen-originated inhomogeneities are given in [1, 2]. Together with those discussed above they show that defects in disordered alumina are definitely related to oxygen although their detailed structure remains unclear (of course, this statement is material dependent).

But which physical quantity does mainly fluctuate in the disordered nanometer-thick oxide layers—the thickness d as was assumed in [22] or the average barrier height? In other words, in what way do the oxygen defects influence the tunnel current? The authors of [43] paid attention to the role of metal-induced evanescent electronic states in the nearest to metal electrodes 2–3 layers of insulator unit cells. Inevitably random fluctuations in the electronic potential due to the formation of oxygen defects should localize a substantial fraction of the evanescent states and, in our opinion, it is just the origin of strong local barrier strength variations within the insulating barrier. Let us emphasize that this scenario is exceptionally important only for ultrathin oxide layers.
barriers like ours, since their thickness is expected to be of several unit cells and they directly contact metallic films from both sides.

In conclusion, highly conductive asymmetric Nb/Al–AlO\textsubscript{x}–Nb junctions show strong subgap leakage currents as was observed earlier in symmetric Nb–AlO\textsubscript{x}–Nb trilayers [8] with high-transparency potential barriers similar to ours. Moreover, we found that at 1.7 K the ratio of subgap to normal resistances does not greatly change for samples with Al-interlayer thicknesses ranging from 40 to 140 nm. This finding was explained as an indication of the universal distribution of barrier transparencies in ultrathin Al oxide films. A noticeably reduced subgap leakage compared with corresponding Nb–AlO\textsubscript{x}–Nb trilayers [8] is in agreement with the analogous finding for Nb/Al–AlO\textsubscript{x}–Al samples with a 5-nm-thick interfacial Al film [15]. We do agree with the authors of [15] that the magnitude of the leakage does not significantly depend on a particular material of the metallic layers. The temperature impact on the subgap junction resistance can be understood taking into account different temperature behavior of a superconducting energy gap induced in the proximized Al interlayer. Our results clearly indicate that in the case of ultrathin and disordered aluminum oxide films, the main effect comes from the distribution of local oxide transparencies. The suppression of the subgap current with the introduction of the Al interlayer is caused by the reduction of the energy gap of one of the electrodes, whereas the key factor, the universal distribution (1), remains almost the same as in Nb–AlO\textsubscript{x}–Nb structures. We have argued that its presence originates from a very broad and homogeneous distribution of local barrier heights generated by oxygen vacancies within the aluminum oxide film. If the defect distribution is more or less uniform [36], the integral of the potential barrier height along an electron path inside the barrier would be a uniform random variable. Of course, this phenomenon is not scale invariant. Since the depth of the penetration of metal-induced evanescent electronic states is limited to 2–3 atomic layers [43], leakage currents should be more pronounced for extremely thin insulating layers. One way to suppress it is to use dielectrics with a lower band forbidden gap [16]. Because the effect is self-averaging, it will be more pronounced in devices with a comparatively large junction area like ours. The latter remark explains why a noticeable reduction of the subgap current can be achieved by splitting a high-transparency micrometer-scale SIN heterostructure into several submicron sub-junctions [22].

Acknowledgments

This work was supported by a grant from Regione Piemonte. MB thanks the National Institute of Metrological Research in Turin, Italy for support during this study.

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New Journal of Physics 14 (2012) 023025 (http://www.njp.org/)