UNIVERSALITY OF CHARGE TRANSPORT ACROSS DISORDERED NANOMETER-THICK OXIDE FILMS

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Abstract

Theoretical and experimental analysis of electron transport across ultra-thin, homogeneously disordered oxide layers is presented with particular regard to the question of how much the effects are universal. We show that (i) distribution of transparencies across dirty subnanometer-thick insulating films is bimodal and (ii) conductance-voltage characteristics of oxide layers with thicknesses increased up to several nanometers are power functions with an index near 1.3. The universality of transport properties is explained as an effect of strong local barrier-height fluctuations generated by the presence of oxygen vacancies.

1. Introduction

Transport characteristics of a mesoscopic system are usually linked to complicated dynamical processes such as impurity scattering, inter-particle interactions, etc. and, in general, are determined by the system dimensionality, geometry, as well as by other sample-specific parameters. Universal transport properties, if they are, should be independent on microscopic details of particular materials and may include only a limited number of characteristics averaged over the sample. Inter alia, the universal behavior of physical quantities can be estimated for homogeneously disordered films with a very large spread of microscopic parameters.

Increasing interest in the ultra-thin amorphous oxide layers is motivated by their promising applications as a gate dielectric in metal-oxide-semiconductor transistors with higher dielectric constant than that of SiO_2, as well as a blocking dielectric for new-generation flash memory cells (Kingon 2000, Robertson 2006). Another field of their applications relates multilayered junctions with quantum-mechanical tunneling as the main physical mechanism for electron transport across them (Wolf 2011). Most such devices are fabricated using aluminum due its superconducting properties and tendency to form a native oxide AlO_x that can be employed as a tunnel barrier. But in both cases significant leakage current, which strongly limits applications of ultra-thin oxide films in semi- and superconducting devices, has been found (Kingon 2000, Greibe 2011). Thus, identification of the physical origin of the extra current across ultra-thin oxide films is of great scientific and practical importance.

The needed information can be obtained from measurements of current (*I*) – voltage (*V*) characteristics of tri-layered structures with an ultra-thin insulating (I) layer placed between two metallic (M) electrodes. The tunnel current across such a system often exhibits unconventional behavior which does not fit into any theoretical picture. The best way to show it is to transform one or both electrodes from a normal (N) state to a superconducting (S) one. In NIS and even more in SIS trilayers the shape of quasiparticle *I-V* curves with a single quantum channel is extremely sensitive to the transmission probability *D* (Wolf 2011). Because of it, such experiments can provide valuable knowledge concerning the distribution of transparencies $\rho(D)$ in the samples studied. First analysis of the $\rho(D)$ .function in subnanometer-thick AlO_x layers was done in the paper (Naveh 2000) by measuring current-vs-voltage and differential conductance-vs-voltage characteristics of planar highly conductive Nb-AlO_x-Nb trilayers at 1.8 K. In spite of the presence of an Al-oxide interlayer, the quasiparticle *I-V* curves did not exhibit typical for conventional superconducting tunnel junctions subgap resistance R_{sg} (it is measured at voltages $|V| < 2\Delta/e$, Δ is the superconducting energy gap of the order of 1 meV) much greater than the normal-state resistance R_{N}. Moreover, R_{sg} was of the order of R_{N} (Naveh 2000). Another unexpected finding is the shape of *I-V* characteristics measured for normal-state tunnel junctions with a several nm-thick oxide interlayer in a voltage range of several hundred millivolts. It was found to be a power function with a power index near 7/3.

The figures of merit that we address in this work are the origin of unusual *I-V* characteristics in heterostructures with disordered ultra-thin oxide films with the thickness *d* and the universality of the phenomena discussed.

2. Universal distribution of transparencies in dirty subnanometer-thick oxide films

Let us start with a subnanometer-thick oxide barrier which can be modeled by a set of disordered short-range scatterers with strength γ_k at position vectors $\boldsymbol{\rho}_k$ randomly distributed within a plane interface between the two metallic electrodes which is perpendicular to the transport direction. The scattering characteristic of an ultra-thin interface can be calculated from the standard Schrŏdinger equation with a localized potential $V(\mathbf{r}) = V(x, \boldsymbol{\rho}_k) = \sum_k \gamma_k \delta(x)\delta(\boldsymbol{\rho} - \boldsymbol{\rho}_k)$ where the *x*-axis is orthogonal to the interface and, hence, parallel to the current direction. After some algebra we obtain that the probability of an electron to be transmitted through the disordered ultra-thin interface for states at the Fermi energy E_F is a sum of local transparencies $D_k = (1 + Z_k{}^2)^{-1}$ with $Z_k = Z(\boldsymbol{\rho}_k) = k_F \int_0^d V(x, \boldsymbol{\rho}_k) dx / E_F$, k_F is the Fermi wave vector. If the parameter Z_k is a uniform random variable ranging from zero to infinity, then its distribution function $\rho(Z) = 2\hbar\bar{G}/e^2 = \mathrm{const}$. Here $\bar{G} = \int_0^\infty \rho(Z)G(Z)dZ$ is the disorder-averaged macroscopic conductance, $G(Z) = \frac{2e^2}{h}D = \frac{2e^2}{h}\frac{1}{1+Z^2}$ is the conductance of a normal-state one-dimensional tunnel junction with a scattering parameter $Z = Hdk_F / E_F$, where *H* is the barrier height of an ultra-thin potential barrier (Blonder 1982). With the parametrization $D = (1+Z^2)^{-1}$ we can transfer to the distribution function of local transparencies $\rho(D)$ which is bimodal with two peaks at $D = 0$ and $D = 1$ $\rho(D) = \hbar\bar{G}\left[e^2 D^{3/2}(1-D)^{1/2}\right]^{-1}$. This result was obtained, first, by Melsen and Beenakker for a three-dimensional clean M-I-M-I-M structure (Melsen 1994), then by Schep and Bauer for a dirty interface in an M-I-M trilayer (Schep 1997) and its universality was many times questioned. It can be shown analytically that the transparency of the double-barrier system is also a Lorentzian $D(\theta) = \left[1 + \tilde{Z}(\theta)^2\right]^{-1}$ with a single parameter $\tilde{Z}(\theta)$, a rapidly oscillating function, which changes periodically from zero (for resonance conditions) to very high values during incident angle θ variations from -π/2 to -π/2. This behavior which is very similar to that in disordered ultra-thin insulating films discussed above is just the reason why the two distributions for physically different systems do coincide.

The $\rho(D)$ formula contains only a single parameter $\bar{G}$. Our aim was to compare the theory with experimental data using no free parameters. It can be realized by transferring metallic electrodes in the tunnel junction into a superconducting state and dividing the measured I-V curves over those obtained in normal-state experiments. Whereas *G*-vs-*Z* dependence for an N-I-N junction is very simple (see above), it is not so for S-I-S devices due to Andreev reflections at the S-I interface when an incident electron (hole) with a probability amplitude $a(\varepsilon)$ is retroreflected into a hole (electron) of the same energy ε (ε will be calculated from the Fermi energy E_F) and almost the same momentum which is travelling in the opposite direction to the incoming charge (Wolf 2011). Our experimental situation was even more complicated since one of the electrodes was a Nb/Al bilayer and, hence, we were dealing with an asymmetric S_1-I-S_2 junction where S_1 stands for an S/N bilayer. In proximity with Nb a nano-scaled Al layer becomes superconducting and it modifies the standard equation for a homogeneous superconductor (for example, Nb) where $a_\mathrm{Nb}(\omega) = i\left(\omega - \sqrt{\omega^2 + \Delta_\mathrm{Nb}{}^2}\right)/\Delta_\mathrm{Nb}$ (with ω, the Matsubara frequency) to a more general case with a function $\Phi(\omega)$, which is the ratio of a modified and normal Green's functions in a superconductor $a_\mathrm{Al}(\omega) = i\left(\omega - \sqrt{\omega^2 + \Phi_\mathrm{Al}{}^2(\omega)}\right)/\Phi_\mathrm{Al}(\omega)$. In the calculations we have used a simplest approximation for $\Phi_\mathrm{Al}(\omega)$ in the S/N bilayer derived by Golubov *et al.* (Golubov 1995) $\Phi_\mathrm{Al}(\omega) = \Delta_\mathrm{Nb} / \left(1 + C\sqrt{\omega^2 + \Delta_\mathrm{Nb}{}^2} / \Delta_\mathrm{Nb}\right)$ and they were based on a numerical method which uses the bimodal distribution $\rho(D)$ and was developed earlier in a few publications; see, for example, the paper (Averin 1995).

Our experimental results were obtained on asymmetric Nb/Al-AlO$_x$-Nb junctions developed at INRiM (Lacquaniti 2007) with the Al-interlayer thicknesses d_{Al} ranged from 40 to 150 nm and the exposure dose, the product of the oxygen pressure and the oxidation time, from 150 to 500 Pa·s. Electrical measurements were performed below critical temperatures of Nb/Al bilayers about 8-9 K for different d_{Al} with a conventional four-terminal dc technique. The samples exhibited supercurrents with values of 6-15 mA at 1.7 K and for the measurements of quasiparticle *I-V* curves we have applied magnetic .fields *B* up to 50 mT through a suitable coil. Normal-state resistance R_{N} were determined from a linear fit to *I-V* curves with and without supercurrents at voltages above 1 mV and the results of both estimations were in a good agreement with each other. Subgap Ohmic resistances R_{sg} were extracted from experimental data as a 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 linearly with *V.* The R$_{\mathrm{sg}}$/R_{N} ratio obtained after averaging over five Nb/Al-AlO$_x$-Nb samples with different d_{Al} and R_{N} (measured at 1.7 K) is given in Tab. 1 together with theoretical outputs for N-I-N, N-I-S, S-I-S, and S/N-I-S devices.

Table 1. Comparison of theoretical and experimental data for superconducting heterostructures

	Nb/Al-AlO$_x$-Nb(exper)	NIN (calc)	NIS (calc)	S/NIS (calc)	SIS (calc)
R$_{\mathrm{sg}}$/R_{N}	1.26	1.0	1.40	1.17	0,68

Reasonable agreement between ratio R$_{\mathrm{sg}}$/R_{N} calculated for an S/N-I-S structure and that measured experimentally proves that, independently on the Al interlayer thickness, the distribution of transparencies across the disordered oxide layer is universal and quantitatively well describes the experimental results.

3. Universal current-voltage characteristic for disordered several nm-thick insulating layers

When the thickness of a dirty insulting layer is increased up to several nanometers, the tunnel barrier in an M-I-M trilayer cannot be more described with a delta function in the transport direction and its internal structure should be taken into account in order to explain unusual bias dependence of the tunnel current which is markedly different from that predicted by the standard tunneling model (Xu 1995). This observation is ascribed to the presence of localized states within noncrystalline materials. Assuming that in this case the dominant mechanism for electronic conduction is hopping, Glazman and Matveev (Glazman 1988) proposed a microscopic model for charge transport across two and more localized states forming optimal conduction chains. In some experiments (see, *e.g.*, Svistunov 2008, Belogolovskii 2011) it was found that the Glazman-Matveev theory does well describe experimental data for tunneling into complex oxides like manganites, independently on whether a barrier was due to an oxygen-depleted layer at the oxide surface (Svistunov 2008), or thin insulating layers inside manganite single crystals resulted from a percolative nature of the transition between charge ordered insulating and metallic ferromagnetic states (Belogolovskii 2011). At the same time it should be noticed that the Glazman-Matveev theory which takes into account inelastic tunneling via pairs of the localized states was based on a sound-like approximation for the phonon dispersion relation valid only for extremely small wave vectors in the complex oxides. It raises the natural questions about why the theory works in the materials studied and why it is so universal. In this section we answer the questions by developing a general theoretical framework for inelastic processes arising when an electron is hopping across localized states inside thin amorphous films.

If a tunneling charge transfers classically forbidden region elastically (without energy loss), the probability of such process exponentially depends on the tunneling distance *l*: $D_{\mathrm{el}} \propto \exp(-2\kappa l)$, where κ^{-1} is the localization length, and the differential conductance $G(V) = dI(V)/dV$ is proportional to V^2 (Wolf 2011). But as the barrier thickness *d* increases, hopping along chains containing localized states is favored, since in this case it is not necessary to transfer quantum-mechanically the whole distance between the electrodes, but rather to jump from one of them to a first nano-island, then transfer to the second one and, after all hopping events, to jump to the opposite electrode. The electron jumps can be as elastic, as inelastic, with emitting a phonon of the energy ε. Due to the strong electron-phonon interaction for localized states, the latter processes which reduce the electron energy from E_1 to $E_2 = E_1 - \varepsilon$ are very important just in amorphous semiconductors. For fixed E_1 and E_2 the number of the inelastic tunneling events is proportional to the electron-phonon interaction function $\alpha^2 F(\varepsilon)$ whose amplitude is determined by α^2, a characteristic of the interaction strength, whereas the shape of the function resembles that of the phonon density of states $F(\varepsilon)$ (Wolf 2011). Then the total probability of electron inelastic tunneling through the distance *l* with the energy decrease from zero to ε is proportional to $\exp(-2\kappa l)\int_0^{\varepsilon} \alpha^2 F(\omega) d\omega$. Taking into account that optimal conductance chains correspond to the case when transmission probabilities of all hoppings are almost identical (Glazman 1988, Xu 1995), we can analytically calculate *I-V* curves for any microstructure of the insulating nano-scaled layer without any assumption concerning the phonon spectrum. As an example, it can be a process of inelastic tunneling across the dielectric through two localized states coupled elastically to the nearest electrodes discussed earlier (Glazman 1988, Xu 1995). For this configuration we find that the inelastic contribution to the differential conductance $G_{\text{inel}}(V) \sim \exp\left(-\frac{2}{3}\kappa d\right)\int_0^{eV}\left(\int_0^{\varepsilon}\alpha^2 F(\omega)d\omega\right)^{1/3} d\varepsilon$. The factor $\exp(-2\kappa d/3)$ reflects the presence of two-step tunneling events across the barrier.

To go further, we need an exact dependence of the phonon spectrum on energy. In general, it is very complicated but for complex oxides of transition metals we are interested in, it can be approximated as $F(\varepsilon) \approx \text{const}\,\theta(\bar{\varepsilon} - \varepsilon)$ with $\bar{\varepsilon}$, the cutoff phonon energy (see, for example, neutron scattering data for $La_{0:625}Ca_{0.375}MnO_3$ polycrystalline samples (Adams 2004). With this approximation for a double-state configuration of the defects inside the disordered complex-oxide layer we obtain the following conductance-vs-voltage dependence $G_{\text{inel}}(V) \sim V^{4/3}$ which should be universal for different complex-oxide materials. To check it, we suggest the following procedure which has no fitting parameters. If the differential conductance of an inhomogeneous thin insulating layer is a power function of the voltage bias $G(V) = G_0 + \text{const} \cdot V^k$, we can find the index *k* and, hence, to distinguish between different transport mechanisms by calculating the function $k(V) = d\ln(G(V) - G_0)/d\ln V$.

In Fig. 1 we have applied the proposed procedure to experimental data for point-contact junctions formed by a sharp Ag tip with $Nd_{0.67}Sr_{0.33}MnO_3$ (NSMO) thin films (Svistunov 2008) and obtained a clear transition from elastic tunneling behavior with $k = 2$ at very low biases to inelastic one with $k \simeq 4/3$ for voltages increased up to several tens of millivolts.

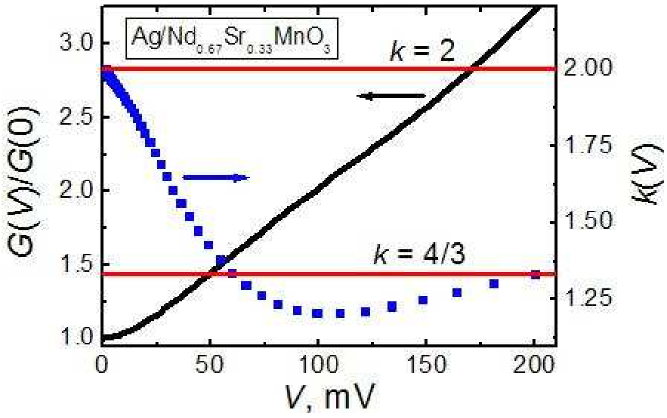

Fig 1. Differential conductance and the power index k dependencies on voltage for an Ag/NSMO point contact

3. Conclusions

Resuming, we have analyzed two phenomena in transport characteristics of strongly disordered oxide films: universal bimodal distribution of transparencies across subnanometer-thick layers for voltages about several millivolts and universal current-voltage characteristic of layers with increased thicknesses up to several nanometers and voltages up to several hundreds of millivolts. We believe that the universalities arise due to strong local barrier-height fluctuations caused by oxygen vacancies (Kim 2011). Just these fluctuations generate huge variations of the parameter Z in ultra-thin oxide films and formation of localized states inside thicker layers. If the transport property depends only on a single parameter and the corresponding analytical relation is mathematically simple (like Lorentzian in subnanometer-thick interlayers), the result obtained after averaging over the uniform random variable is of a general character bearing no relation to sample-specific details of particular objects and including only a limited number of macroscopic parameters or, by other words, is universal.

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