Tunneling through Al/AlOx/Al junction: analytical models and first principles simulations

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We study from first principles the transport properties of Al/AlOx/Al tunnel junctions. On this basis, we analyze the reliability of two analytical models for the conductance, namely the trapezoidal potential barrier model and a tight-binding model. Our findings show that (i) the interface width used in the models is determined by the electronic density profile, and it is shorter than the width one expects from the atomic arrangements; (ii) the effective mass, found to be about on third of the free electron mass, can be determined from the oxide band-structure calculations, and (iii) the barrier height is given by one fourth of the bandgap in the oxide, which explains the apparently small values found for these junctions experimentally.

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I. INTRODUCTION

Tunneling of electrons through aluminum-aluminum oxide (Al/AlOx/Al) junctions is one of the prototypical examples of quantum-mechanical tunneling in solid state physics. Fisher and Giaever in their pioneering work demonstrated the tunneling character of the transport of electrons through this interface and by comparing their results with the predictions of Hohn2 for tunneling through vacuum gap, they initiated the interpretation of tunneling measurements through thin metal-insulator-metal junctions using potential barrier model. The minimal form of this model contains two parameters - the barrier width $d$, which indicates the physical width of the oxide, and its height $W$, given by the energy difference between the Fermi energy and the bottom of the conduction band in the oxide. In practice, several other parameters enter the model: the electron's effective mass in the oxide or the dielectric constant of the oxide used within an additional image-charge potential. Further parameters are used for fine-tuning the shape of the barrier, e.g. its asymmetry. Clearly, having a large set of parameters, it is no surprise that the simple barrier model can be fitted to the experimental current-voltage characteristics well, but at the same time, it rises questions about the relevance of the model itself. For example, the inclusion of the image potential can have a significant effect on the effective barrier width, but its presence depends on the time scales of the tunneling electrons and the interface plasmons in the metal.

On the other hand, much more detailed and parameter-free models of the interface can be constructed using first principles calculations, even though the size of the modeled interfaces is somewhat restricted due to the numerical cost of these calculations. Nevertheless, in many experiments, the studied interfaces have widths within the reach of ab initio simulations so that the accuracy of the potential barrier model to the interpretation of tunneling data can be tested. Specifically, Jung et al.15 presented such a study comparing the character of the equilibrium projected density of states of the Al/AlOx/Al interface obtained by a first principles simulation with the potential barrier model. They found that the parameters of the potential barrier model fitted to the experimental data are in qualitative agreement with the parameters of the first principles calculations. The potential barrier model included the image potential and hence also the dielectric constant which effectively narrowed and lowered the potential barrier.

In this work we test the performance of the potential barrier model by comparing the predicted conductance to ab initio calculations. We test this for Al/AlOx/Al junctions of four different widths $d$ and show that it is essential to use an effective mass in the oxide and an effectively shorter width of the tunneling region within the potential barrier model. We also present an analytical tight-binding model for the conductance that describes the ab initio results more accurately than the potential barrier model. The parameters of the latter are extracted from the ground state ab initio calculations of the junction. In Sec. [II] and [III] we introduce the analytical details of the models. The ab initio results for ground state properties of the studied junctions are presented in Sec. [IV] and [V] together with the computational parameters used in the calculations. Finally in Sec. [VI] we compare the conductances obtained using the ab initio calculations and the conductances obtained from the analytical models.

II. POTENTIAL BARRIER MODELS OF THE INTERFACE

The starting assumption of the potential barrier model is that inside the metallic electrodes, on the left and right of the insulator, the electrons behave like free quasi-particles with their energy being in a separable form:

$$E = E_z + E_\parallel = k_z^2 / 2 + k_\parallel^2 / 2$$

where $k_z$ is the component of electron’s momentum perpendicular to the interface and $k_\parallel$ the component of momentum parallel to the interface. The current density, induced by an infinitesimal bias voltage, consists of a sum of contributions from the electrons occupying states in the energy window around the Fermi energy $E_F$, with their momentum...
the insulator of width \( \Delta d \) into account for the description of ultra-thin interfaces: (1) the features of the potential barrier models that need to be taken respect to the Fermi energy of the metal.

Potential barrier , the WKB approximation for the transmission energy integration in Eq. (3) are sufficient for an accurate approximate expressions for the transmission as well as for the energy in Fig. 1.

On the other hand, in Sec. VI we will also demonstrate that these two requirements can be fulfilled by using a specific shape of the potential barrier. In this work we use a trapezoid potential barrier (TB) as defined in Fig. [1](#).

On the other hand, in Sec. VI we will also demonstrate that approximate expressions for the transmission as well as for the energy integration in Eq. (3) are sufficient for an accurate evaluation of the model conductance. For the trapezoid potential barrier, the WKB approximation for the transmission gives

\[
T(E - E_\parallel) = \exp \left\{ -F(E - E_\parallel) \right\},
\]

\[
F(E - E_\parallel) = 2 \int_{-d(E_\parallel)}^{d(E_\parallel)} 2m_{\text{eff}} |W(z) - E_\parallel|dz,
\]

where \( m_{\text{eff}} \) is the effective mass of the electrons in the insulator, \(-d(E_\parallel)\) and \(+d(E_\parallel)\) give the region where \( |W(z) + E_\parallel| \geq 0 \), and \( W(z) \) is the trapezoid potential profile. Accounting only for the largest contribution from the states close to the Fermi energy in the integral in Eq. (3), \( E_\parallel \sim E_F \), we obtain the following simple analytical expression:

\[
g \approx \frac{e^{-F(E_F)}}{2\pi^2 F'(E_F)},
\]

where

\[
F(E_F) = 2\sqrt{2m_{\text{eff}}W (d_W + \frac{2}{3}\Delta d_{EF})},
\]

\[
F'(E_F) = -\frac{2}{\sqrt{2m_{\text{eff}}W}} (d_W + 2\Delta d_{EF}).
\]

We will refer to Eq. (6) as the TB\(^{A,m_{\text{eff}}} \) model (\( A \) stands for “analytical” as compared to the numerically calculated transmission for the trapezoid potential barrier - TB\(^{N} \)). We note that the introduction of two transition regions of width \( \Delta d \) adds to the exponent of the transmission amplitude only a small fraction of \( \Delta d \), namely \((2/3)\Delta d_{EF} \). This results in a substantial increase of the conductance which is needed for the agreement of the TB model and \( ab \text{ initio} \) results (see Sec. VI).

### III. ATOMIC \( sp \) MODEL OF THE INSULATOR

It is typically assumed that the barrier height in the potential barrier model corresponds to energy distance between the Fermi energy and the closest among the valence or conduction bands of the insulator, or even to its whole bandgap. However, fits of the potential barrier model to experimental data often lead to unphysically small values if one follows this interpretation. Various arguments like interface roughness or image potential have been suggested to correct for this underestimation, but perhaps the most important one – the principal difference in the energetic spectrum of the real insulator and the vacuum gap – received less attention.

To account for a more realistic electronic structure of the insulator we consider a minimal tight-binding model of a \( sp \)-like insulator with rock-salt crystal structure. For our purposes, the cation with \( s \)-like orbital plays the role of aluminum and the anion with \( p \)-like orbital the oxygen atom. While this is different from the true structure of alumina, this model works surprisingly well even for the disordered aluminum oxide found in our interfaces, as will be shown in Sec. IV.

The \( sp \) model has four parameters: the onsite atomic energies of the cation (\( E_t \)) and anion (\( E_p \)), the hopping matrix element between the two atoms (\( t \)), and the length of the edge of the conventional unit cell (cube) \( a \). A standard calculation leads to valence (\( v \)) and conduction (\( c \)) band energies

\[
E_{v/c}(k) = E_{v/c}^{\infty} \pm \frac{E_g}{2} \sqrt{1 + \frac{m_{\text{eff}}}{m_p a^2} \sum_{i=1}^{3} \sin^2(k_i a/2)}
\]

where \( m_{\text{eff}} = m_{c}/(2m_p a^2) \) is the effective mass of the electrons close to the conduction band minimum, equal in magnitude that of the valence band maximum. The two bands are separated by the bandgap \( E_g = E_p - E_v \), and the energy in the middle of the gap is

\[
E_{F} = \frac{E_p + E_v}{2}.
\]
In the tunneling regime, the current is carried by the electronic states in the bandgap, \(27-29\), i.e., the evanescent Bloch states with imaginary wavenumber \(k_z = i\kappa\):

\[
\phi_{k_z\parallel}(r) \sim e^{-\kappa z} e^{\imath k_{\parallel} \cdot r} u_{k_z\parallel}(r). \tag{11}
\]

The WKB-like result for the transmission takes then the form

\[
T_{k_{\parallel}}^{sp}(E) \sim |\phi_{k_z\parallel}(d)|^2 \sim e^{-2\kappa(E,k_{\parallel})d}, \tag{12}
\]

where \(d\) is a vector normal to the interface with the length given by the width of the interface \((d) \sim d\). \(\kappa(E,k_{\parallel})\) can be obtained from Eq. (9) using the substitution \(k_z = i\kappa\) therein. The transmission can be then used in the calculation of the conductance in Eq. (3). The largest contributions to the conductance come only from \(\kappa d/2 < 1, k_{\parallel} d/2 < 1\), so that the \(\sin (\cdot)\) functions in the dispersion can be expanded in Taylor series. Keeping only the first two terms we find \(34\)

\[
\kappa(E,k_{\parallel}) = \sqrt{\nu(E)m_{\text{eff}}E_g/2 + k_{\parallel}^2 + k_z^2}, \tag{13}
\]

\[
= \sqrt{2 \left[\nu(E)m_{\text{eff}}E_g/4 + E_f\right]}, \tag{14}
\]

where we have introduced a multiplicative factor \(\nu(E)\) accounting for the relative distance of the energy \(E\) from the middle of the gap,

\[
\nu(E) = 1 - 4 \left(\frac{E - E_F}{E_g}\right)^2, \tag{15}
\]

which is close to 1 for \(E \sim E_F\). We note that by using the Taylor expansion the model becomes independent of the size of the conventional cell \(a\). The transmission \(T_{k_{\parallel}}^{sp}(E)\) is similar to the WKB result for a potential barrier \([\text{Eqs. (4)}]\) for a constant barrier height \(W\). Hence, making the same approximations as in Sec. [II] and substituting \(W \rightarrow \nu(E_F)E_g/4\) we find an analytical expression for the transmission through a \(sp\) insulator of width \(d\) precisely of the form of Eq. (6), where

\[
F_{sp}(E_F) = 2\sqrt{\nu(E_F)m_{\text{eff}}E_g/2 \, d}, \tag{16}
\]

\[
F'_{sp}(E_F) = -\frac{2}{\sqrt{\nu(E_F)m_{\text{eff}}E_g/2}} \, d. \tag{17}
\]

This represents one of the main results of our paper: the potential barrier height \(W\) is related to the bandgap through the relation \(W = \nu(E_F)E_g/4\). Since the Fermi energy in our junctions is close to the center of the gap (Sec. [IV] where we have \(\nu(E_F) \sim 1\), we expect that the bandgap is about four times larger than the barrier height obtained from the fits to the experimental data. This explains the typical situation in Al/AlO\(_x\)/Al junctions where \(W\) can be as small as 2eV or less, which is to be compared with the bandgap of alumina being about 7–9eV. Further comparisons will be made in the Sec. [VI] where the \(sp\) model is compared to the \textit{ab initio} calculation of the conductance.

\[\text{FIG. 2: (Color online) The 4L structure (above) and the corresponding averaged electronic density of the occupied transmitting states. The dashed lines indicate the positions of the metal-oxide boundary obtained according to Eq. (18).}\]

### IV. FIRST PRINCIPLES CALCULATIONS OF THE AL/ALO\(_x\)/AL INTERFACES

The Al/AlO\(_x\) thin film is well known for its difficulties to be grown in an ordered form. \(30-33\) The process of oxidation consists of a quick chemisorption of oxygen on a clean Al surface which is followed by a complex diffusion process leading to various widths of the interface which is typically disordered. \(29-29\) The model that we consider is on the other hand relatively simple and ordered. We followed Jennison at constructing chemisorbed layer of oxygen on an ideal Al(111) \(\sqrt{3} \times \sqrt{3}\) surface (three Al atoms per layer), modelled as a slab 6 layers thick (left electrode). Next we were adding Al and O atoms and relaxed the geometry until we found a stable interface having two layers of oxygen atoms (2L). Finally we enclosed the interface with four ideal Al(111) layers (right electrode) and connected it with the left electrode through periodic boundary conditions. Performing this procedure two different geometries of the interfaces were identified: (1) an asymmetric structure, corresponding the ultra-thin AlO\(_x\) layer investigated by Jennison, and (2) an symmetric structure which did not contain the layer of chemisorbed oxygen next to the bottom Al electrode. More details on the differences between the asymmetric and symmetric structures can be found elsewhere \(33\). In our present work we will consider only structures derived from the asymmetric geometry.

Motivated by the geometry of the asymmetric 2L interface model we have constructed thicker Al/AlO\(_x\)/Al by adding one (3L), two (4L) or three (5L) full oxygen layers sandwiched between monoatomic (Al1) or diatomic (Al2) layers of aluminum. The resulting geometries were optimized until the forces on the atoms were smaller than 0.002 Ha/\(\AA\), while the Al atoms beyond the first layer of bulk metal were kept fixed. An example of the resulting geometric structure of 4L is shown in Fig. [2]. We should mention that these models are not necessarily the only ones possible for the interface of the concerned width. Due to the above described tendency of AlO\(_x\) systems towards disorder, we expect that many different vari-
TABLE I: Values of the interface widths, band-gaps, Fermi energy shifts and the shift factor $\nu(E_F)$ obtained from ab initio calculations.

| System | $d$ [Å] | $E_g$ [eV] | $\Delta E_F$ [eV] | $\nu(E_F)$ |
|--------|---------|-----------|------------------|-----------|
| 2L     | 4.5     | 7.0       | 1.5              | 0.82      |
| 3L     | 5.5     | 6.5       | −0.25            | 0.99      |
| 4L     | 7.8     | 6.5       | −1.0             | 0.91      |
| 5L     | 9.8     | 6.5       | −1.0             | 0.91      |

The second important parameter of the potential barrier and the atomic $sp$ models is the insulator band gap $E_g$. It can be extracted from the projected density of states (PDOS), where the Kohn-Sham eigenstates of the interface are projected on atomic orbitals. Figure 4 shows the PDOS for the 4L interface, where the PDOS of atoms in each layer are added together, green lines corresponding to the Al layers and the red lines to the oxygen layers. The oxide bandgap is estimated as the energy distance between the onset of the valence bands on the oxygen atoms below the Fermi energy, and the onset of the mixed Al and O bands above the Fermi energy. From the PDOS we can also obtain the energy distance between the midgap energy and the Fermi energy, $\Delta E_F$, needed for the $sp$ model. The calculated bandgaps and $\Delta E_F$ for all studied interfaces are collected in the Table I. Interestingly, in spite of the well known bandgap problem of the DFT[30,32], these bandgaps appear to be in very good agreement with recent experimental results for the Al/Al$_2$O$_3$ interfaces[33,34] which found $E_g = 6.4$ eV.

While the bandgap stays roughly the same for all of the studied interfaces $2L - 5L$, the Fermi energy shifts with respect to the middle of the gap from positive (conventionally called the electron tunneling regime) to negative values (hole tunneling). However, the factor $\nu(E_F)$ stays close to one in all the cases (see Table I), as anticipated already in Sec II. The energy difference between the bottom of the conduction band and the Fermi energy determined experimentally[31] was found to be $E_c - E_F = 2.9 \pm 0.2$ eV which is 1 eV smaller than the DFT value found here for 4L and 5L, but on the other hand, in good agreement with 2L and 3L, which perhaps indicates larger sensitivity of this quantity on the particular system.
V. ELECTRONIC STRUCTURE OF AN IDEAL INSULATOR

The potential barrier model as well as the sp model also rely on the knowledge of the effective mass $m_{\text{eff}}$ of the electrons in the insulator or barrier region. To calculate it we have considered a first principles model of the insulator extracted from the geometry of the 4L junction. Namely, it consists of a supercell of length $l = 8.11a_B$ in the $z$ direction and with identical dimensions in the two remaining in-plane directions [i.e. $\sqrt{3} \times \sqrt{3} \text{Al}(111)$]. The supercell contains two layers of oxygen and two layers of $2/3$ filled Al planes. (the 3rd and 4th oxygen layers in Fig. 2 from the left and their immediately following Al layers respectively). This way, the chemical composition actually corresponds to alumina, Al$_2$O$_3$.

The DFT ground state calculation has been done with the PWCOND program [Eqs. 1-7] that is capable of obtaining the so called complex band-structure, i.e. energy bands for imaginary as well as real Bloch $k$-vectors. We have checked that calculations of the band-structure for real $k$-vectors using the Quantum Espresso and the PWCOND gave identical results so that the parameters involved in the PWCOND program were correctly chosen.

The band-structure along the direction normal to the interface ($z$) is shown in Fig. 5. First of all we note that the bandgap obtained here, $E_{\text{gap}} \approx 4$eV (in agreement with the previous DFT-PBE results for bulk $\gamma$-Al$_2$O$_3$ [Eqs. 1-7]), is significantly smaller than the bandgap extracted from the PDOS of the full junction ($\sim 6.5$eV). Interestingly, the experimental value of this phase of alumina is $E_{\text{gap}}^{\text{exp}} = 7$eV, which can be obtained also computationally if the DFT-PBE result is followed by a GW calculation [Eqs. 8].

The DFT band structure in Fig. 5 is fitted with two model dispersions. The TB model uses a free-electron like dispersion $E_c(k) = E_x + k^2/(2m_{\text{eff}})$ which after fitting gives the effective mass $m_{\text{eff}} = 0.35$. The atomic sp model [Eq. 9] in the approximation $k\ell/2 < 1$, which is used in the analytic expression for the conductance, gives (for $k_x = k_y = 0$) the dispersion:

$$E_{c/\gamma}(k) = E_F \pm \frac{E_g}{2} \sqrt{1 + \frac{2k_z^2}{m_{\text{eff}}E_g}}. \quad (19)$$

The parameters of the fit given in Fig. 5 are $m_{\text{eff}} = 0.35$, $E_x = 4.0$eV and $E_g = 1$eV. Our value of the effective mass is to be compared with the electron’s mass obtained from DFT calculations for ideal $\alpha$-Al$_2$O$_3$ crystal, $m_{\text{eff}} \approx 0.147$ [Eqs. 10-11] and fits to experimental $I-V$ characteristics, $m_{\text{eff}} \approx 0.234$ [Eqs. 12].

We see that the sp model works very well for real as well as imaginary band-structure close to $k = 0$. While both models give the same effective mass, the values of $\kappa$ for the free-electron like dispersion are larger by $\sim 50\%$ (as indicated by arrows) which contributes to prediction of smaller conductances within the potential barrier model given the interface width is the same, as will be shown in the following section.

VI. THE CONDUCTANCE

Transport properties of the junctions were obtained using the transfer matrix method [Eqs. 13-16] implemented in the PWCOND code [Eqs. 17], using plane-wave basis and ultra-soft pseudopotentials. For the given self-consistent Kohn-Sham potential (ob-
The horizontal error bars accompanying the \textit{ab initio} conductances, \(\Delta d\approx 2\text{ Å}\), indicate the width of the transition region between the metal and the insulator, which is taken from the averaged density profile, Fig. 2.

First we consider the potential barrier model with effective mass equal to one, where the calculation of the transmission as well as its energy integration (Eq. 3) are done numerically exactly (TB\(^N\)). The potential barrier is of the form given in Fig. 1 where \(d_w = d - \Delta d\). The conductances are shown as the blue crosses, where the height of the energy barrier \(W = 0.5neV\), \(n = 1, 2, 3, 4, 5\) is increasing from top to bottom. The pink-dotted line is the conductance of the same potential barrier of width \(W = 2eV\), but evaluated using the approximate formula [Eq. (6)]. As anticipated in Section II, we see that in view of the overall differences, the approximate but analytic formula is very satisfactory and the numerical calculation of the transmission of its energy integration is not really needed.

We see that in principle, we can achieve agreement between this model and the \textit{ab initio} results if we choose \(W \approx 0.5eV\), but this is in stark contrast with the estimates of the potential barrier height from the PDOS, typically taken as the distance between the Fermi energy and the nearest band in the oxide (e.g. the valence band in the oxide in 4L structure according to Fig. 4), here expected to be \(W \sim 2eV\).

The green-dashed line is a conductance corresponding to a simple square potential barrier with \(W = 2eV\) and effective mass equal to one, and we see that plain square barrier model goes in the wrong direction. The use transition regions of width \(\Delta d\) does shift the potential barrier model in the right direction, particularly for very short interfaces, where the effective mass within the insulator does not seem to play an importance role. Hence, use of the transition region between the metal and the insulator of width \(\Delta d\), given by the spatial extent of the drop if the electronic density between the metal and the oxide, is essential for the TB model.

Finally, the full black line corresponds to the atomic \(sp\) model with the effective mass \(m_{\text{eff}} = 0.35\), band gap \(E_g = 6.5eV\) and the barrier width \(d_w = d - \Delta d\). The use of this reduced width \(d_w\) is motivated by two observations: (1) in Sec. II we have seen that the linearly increasing potential at distance \(\Delta d\) contributes to the exponent of the conductance [Eqs. (6)] through a much smaller contribution \(\Delta d_{\text{Fermi}} = W/(W + E_g)\Delta d \sim 0.15\Delta d\). (2) in the TB model we have seen that the use of a shorter barrier, effectively given by \(d_w + 2/3\Delta d\) [Eq. (6)], is important to compare well with the \textit{ab initio} conductances. Hence we expect that also in the \(sp\) model, the oxide width (i.e. the equivalent of the potential barrier) needs to be reduced almost to \(d - \Delta d\), which is the value we use. As a result, the \(sp\) model is essentially on top of the \textit{ab initio} conductances. While the improvement with respect to the potential barrier model with transition region and the effective mass is not that large, it is important to stress that the parameters of the \(sp\) model \((E_g, m_{\text{eff}}, d - \Delta d)\) correspond to the characteristics of the true \textit{ab initio} model.

It is interesting to attempt a quantitative comparison between experimentally determined barrier widths and heights, and our \textit{ab initio} and \(sp\) model results (Fig. 7). As mentioned already in the introduction, there are experimental junctions that are now accessible to first principles simulations. Based on the rather unsatisfactory state of affairs in Fig. 7 we sus-
FIG. 7: (Color online) The ab initio conductances compared to selected experimental results. (a) Jung [13], (b) Gloos [14] (c) Holmqvist [15] (d) Brinkman [16] and recent experiments by Schaefer [17]. The model gives fairly rigid prediction of the conductances, even using the collected experimental results. (a) Jung [13].

VII. CONCLUSIONS

In the conclusions, we have analyzed the performance of simple analytical models in describing the conductance of ultra-thin Al/AlOₓ/Al junctions. We have compared atomistic first-principles calculations using the DFT-PBE framework combined with the Landauer formula, with the conductances obtained from a potential barrier and a tight-binding sp analytical models. We have shown that the expression for the conductance of the atomic sp model has the same form as that from the potential barrier model if the barrier height W is exchanged for \( v(E_F)E_g/4 \) with \( v(E_F) \sim 1 \), which explains the small values of W obtained frequently in the past by fitting the potential barrier model to the experimental I−V curves. The accuracy of the analytical models has been tested by using parameters derived from ground-state DFT calculations. We have found that the oxide is characterized by effective mass \( m_{eff} = 0.35 \) and bandgap \( E_g = 6.5eV \). When these parameters are used in combination with the sp model, excellent agreement with the numerically calculated conductances is found. The interface width used in the models has been shown to correspond to the width of the well-developed oxide which is shorter by about \( \Delta d \approx 2A \) compared to the geometric width of the interface d.

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