

Tunneling Conductance of Asymmetrical Barriers

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The voltage-dependent tunneling conductance of trapezoidal potential barriers has been calculated using two extreme models of (1) the WKB approximation and (2) perfectly sharp boundaries between the metal electrode and the insulator. We show that for both models the conductance-voltage plot is roughly parabolic at low voltages ($\lesssim 0.4$ V). The minimum conductance is not at zero bias unless the barrier is symmetrical and identical Fermi energies are chosen for the two metal electrodes. The inclusion of image forces does not radically alter the shape of the conductance-voltage dependence. Using reasonable barrier shapes, the asymmetry of the calculated conductance about $V=0$ is not as large as we frequently observe experimentally. We point out that this extreme asymmetry appears to be associated with the presence of organic impurities in the oxide layer of the junction.

INTRODUCTION

Electron tunneling through a thin insulating layer between two normal metal electrodes has been studied theoretically and experimentally for almost 40 years. Calculations based on rectangular and trapezoidal potential barriers using various approximations have shown that the tunneling current should be directly proportional to the applied voltage for voltages very much less than the barrier height and should increase exponentially with voltage when the voltage becomes comparable to the barrier height. These Ohmic and exponential behaviors were observed by Fisher and Giaever¹ in their pioneer experiments using a thermally grown oxide of one of the metals as the insulating layer. However, much more powerful techniques than the simple measurement of current and voltage are now available for the study of normal metal tunneling. These are the determination of derivatives of the current with respect to voltage as a function of voltage. The first derivative technique was first used in tunneling by Hall *et al.*² and the second derivative by Chynoweth *et al.*³ in their studies of semiconductor p - n diodes. The same techniques have been used extensively in studies of tunneling between superconductors where finer detail in the characteristic has necessitated considerable improvement in equipment.⁴ It is now fairly common, for example, to measure conductance (dI/dV) to an accuracy better than 1 part in 10^3 . These derivative techniques have been applied to some extent to normal metal tunneling but interest has been mostly confined to small abrupt changes in conductance occurring over narrow energy ranges. Typical measurements are of the conductance anomaly within a few millivolts of zero bias⁵ and the conductance increases, due to excitation of impurity molecules in the oxide layer, which are observed at discrete voltages up to 500 mV.⁶ However, these structures are relatively small and are superimposed on a voltage dependent conductance which results from changes in the barrier shape with applied bias. This "background" conductance has received relatively little experimental attention and no attempt has been made to calculate it using "realistic"

models of the barrier. One of us has pointed out⁷ that at relatively low voltages (<1 V) the conductance versus voltage for Al-I-M ($M=\text{Pb, Sn, In}$) and Pb-I-Pb junctions appears roughly as a parabola and that the minimum conductance commonly occurs at a finite voltage (in the M positive bias for Al-I-M junctions). Similar observations have been made by Kuhn⁸ and by Hauser and Testardi.⁹ It was suggested that the "offset" of this parabola from $V=0$ is simply the result of asymmetry in the potential barrier shape.⁷ However, in the past two years, there has not been any investigation of whether reasonable barrier asymmetries do result in offsets of the magnitude commonly observed (50–250 mV).

In an attempt to investigate this suggestion further, in this paper we present calculations of the conductance versus voltage for idealized models of the potential barrier. We take a trapezoidal barrier shape and consider the two extreme cases of (1) the WKB approximation and (2) perfectly sharp boundaries between metal and insulator. We show that the minimum conductance is not at zero bias unless the barrier is symmetrical and similar Fermi energies are chosen for the two metals. The inclusion of image forces does not radically alter the shape of the conductance-voltage dependence.

A detailed comparison with experiment is not possible as the barrier height and thickness have not been determined independently. However we will show that for junctions showing small offsets (~ 50 mV) the conductance plot can be fit satisfactorily using reasonable barrier parameters. We conclude that the large offsets (~ 250 mV) can not be the result of barrier asymmetry alone and present the suggestion that there is a correlation between the magnitude of the offset and the number of organic impurity molecules trapped in the junction.

We do not believe that the importance of the calculation lies in the numbers that can be obtained by its comparison with experiment. Rather we wish to point out that the calculated dependence of conductance on voltage, for simple barriers, does appear roughly as a parabola offset from $V=0$. Since the asymmetry is due

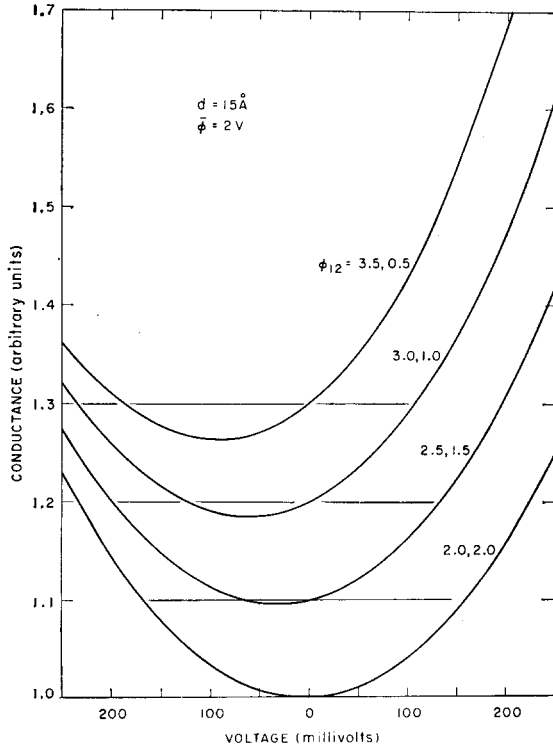


FIG. 1. Calculated conductance versus voltage (using model I) for a trapezoidal barrier with $d=15$ Å, $\bar{\phi}=2$ V. The successive plots for $\Delta\phi=0, 1.0, 2.0, 3.0$ V have had conductances normalized to 1.0 at zero voltage and then offset by 0.1 for clarity.

to the exponential weighting factor in the WKB approximation, we feel that further refinements of the model of the barrier will do little to change the overall shape of the conductance plot. By "roughly parabolic" we mean that one immediately recognizes the parabolic nature, in contrast to linear or exponential dependences which have been observed in some chromium¹⁰ and doped¹¹ junctions or at high voltages. Generally a parabola can be fitted to the calculated and experimental plots of this paper within 5% at ± 200 mV and within 20% at voltages where the conductance doubles its minimum value. The only accepted experimental proof of the occurrence of tunneling in any junction is the observation of a superconducting energy gap in one or both of the electrodes. However, this test is confined to low voltages and the observation of a parabolic dependence of conductance *as well* should suggest that tunneling is taking place up to relatively large voltages. An alternative, but more tedious, test of tunneling is to measure the I - V characteristics as a function of temperature.¹² However, this can be affected, for low resistance junctions, by the resistance of the metal films changing with temperature.¹³

CALCULATION OF BARRIER CONDUCTANCE

Before presenting the details of our calculation we will comment briefly on the approach taken by others. Consider two metals a and b separated by an arbitrary

potential barrier $\varphi(x)$. Assuming the WKB approximation inside the barrier the tunneling current density is given by¹⁴

$$j = \frac{4\pi e}{\hbar} \sum_{\mathbf{k}_t} \int_{-\infty}^{\infty} dE_x \rho_a(E) \rho_b(E-eV) P(E_x) \times [f(E) - f(E-eV)], \quad (1)$$

where $\rho_a(E)$ and $\rho_b(E)$ are the density of states for a given transverse momentum $\mathbf{k}_t$ and total energy E for system a and b respectively. The $f(E)$ is the usual Fermi distribution function. E_x is the total energy in the direction perpendicular to the barrier. $P(E_x)$ is the tunneling probability which has the form

$$P(E_x) = A \exp\left(-\frac{2}{\hbar} \int_0^d \{2m[\varphi(x', V) - E_x]\}^{1/2} dx'\right), \quad (2)$$

where d is the barrier thickness and $\varphi(x, V)$ is the barrier height at the voltage V and the position x in the barrier. The preexponential factor A may depend on E_x as we will discuss below. In order to simplify the calculation of (1) two different approaches have been used in the past and these have been discussed in detail by Hartman.¹⁵ The approximation used by Simmons¹⁶ is to replace the barrier $\varphi(x, V)$ by an average barrier $\bar{\varphi}(V)$. Hence $\bar{\varphi}(V) - E$ is a constant through

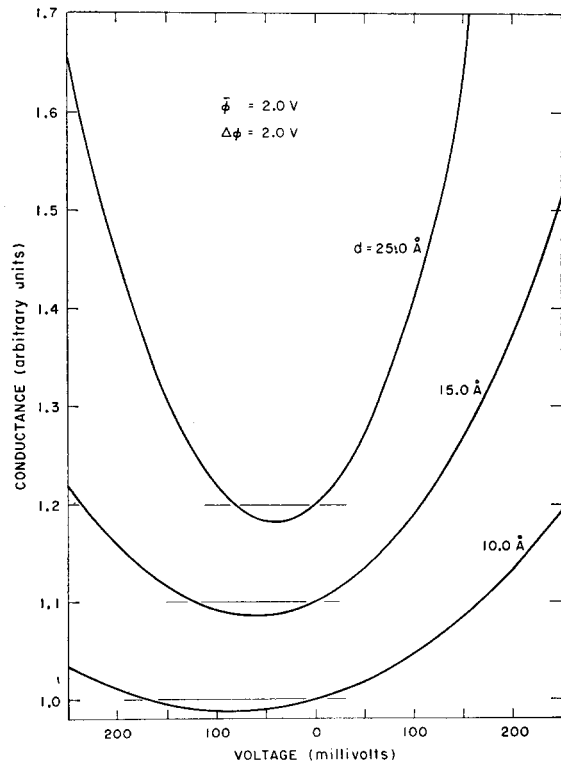


FIG. 2. Calculated conductance versus voltage (using model I), showing the effect of barrier thickness variation. The fixed parameters are $\bar{\phi}=2.0$ V, $\Delta\phi=2.0$ V.

the barrier and the tunneling probability⁷ is simply reduced to

$$P[E_x] \approx A \exp \left\{ - \left(\frac{8m}{\hbar^2} \right)^{1/2} \beta [\bar{\varphi}(V) - E_x]^{1/2} \int_0^d dx' \right\}. \quad (3)$$

The correction factor β is assumed independent of E_x in order to perform the integration. By applying this approximation of an average barrier height, all information about barrier asymmetry is lost.

The approach taken by Stratton¹⁷ and others¹⁸ is to expand $\ln P[E_x]$ for electrons lying close to the Fermi energy, i.e., $[\eta - E_x]$ is small. This leads to rather awkward expressions for the current in terms of expansion coefficients but it has been pointed out¹⁵ that the current-voltage characteristic is only symmetrical about $V=0$ if the barrier is symmetrical.

In order to avoid the unnecessary approximations made previously we have calculated the tunneling current for a trapezoidal barrier numerically by computer and found the conductance by direct differentiation of the calculated current with respect to voltage. We have considered the two extreme cases first discussed by Harrison.¹⁴

(1) In the WKB approximation, which implies that the band structure of the metal-insulator-metal system varies only slowly compared to the electron wave-

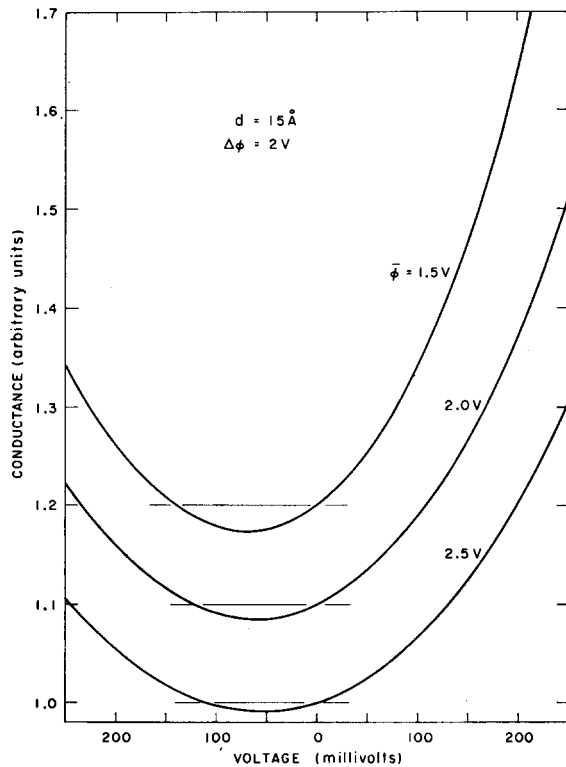


FIG. 3. Effect of variation in average barrier height on the conductance versus voltage for a trapezoidal barrier (model I). Fixed parameters are $d=15$ Å, $\Delta\phi=2$ V.

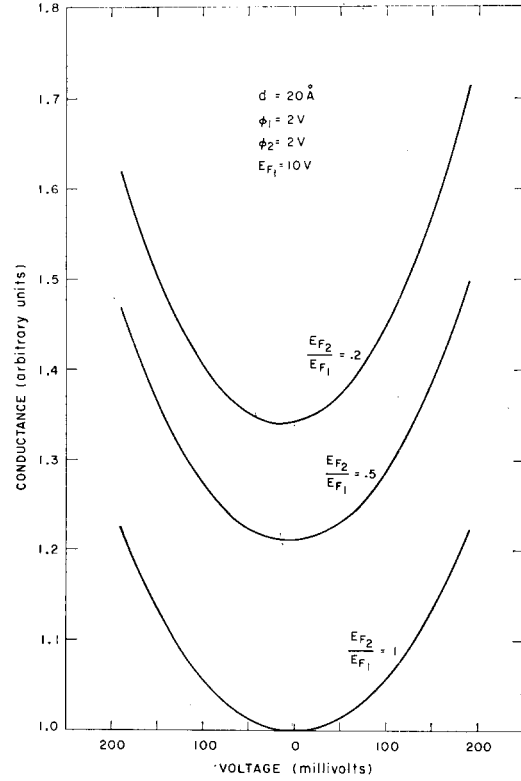


FIG. 4. Conductance versus voltage calculated using the sharp barrier approximation (model II). The parameters are $d=20$ Å, $\phi_1=2$ V, $\Delta\phi=0$, $E_{F1}=10$ V. Successive plots show the effect of changing the Fermi energy E_{F2} . The conductance at $V=0$ for $E_{F2}/E_{F1}=1.0$, the shifts of the other two curves show actual changes in relative conductance.

length, the current density is given by

$$j = \frac{2e}{h} \sum_k \int_{-\infty}^{\infty} \exp \left(- \frac{2}{\hbar} \int_0^d \{ [\varphi(x, V) - E_x] 2m \}^{1/2} dx \right) \times [f(E) - f(E - eV)] dE_x. \quad (4)$$

Here

$$\varphi(x, V) = \varphi_1 + (x/d) (\varphi_2 - eV - \varphi_1) \quad (5)$$

and φ_1 and φ_2 are the barrier heights on sides a and b with zero applied voltage. As pointed out by Harrison,¹⁴ energy-dependent terms in the preexponential term A in Eq. (3) cancel those in the density of states in (1) and the tunneling current is not density-of-states-dependent in this model.

The integrals involved in Eq. (4) were carried out numerically by computer, assuming $T=0^\circ\text{K}$ for simplicity, and the conductance was obtained by numerical differentiation. Expression (4) was also expanded in powers of the voltage and approximate expressions for the coefficients of the linear and quadratic terms in the conductance were obtained. The expression, accurate to roughly 10% when the barrier thickness is greater than 10 Å and when $\Delta\phi/\bar{\varphi}$ is less than one is

$$\frac{G(V)}{G(0)} = 1 - \left(\frac{A_0 \Delta\phi}{16\bar{\varphi}^{3/2}} \right) eV + \left(\frac{9}{128} \frac{A_0^2}{\bar{\varphi}} \right) (eV)^2, \quad (7)$$

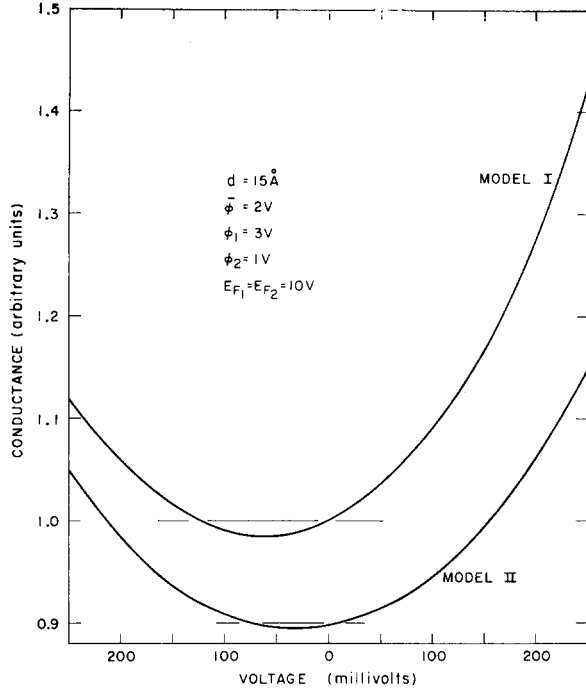


FIG. 5. A comparison of conductance versus voltage calculated for the same barrier parameters but using the two models of diffuse and sharp barrier boundaries. The conductances are normalized to 1.0 at $V=0$ but are offset for clarity. The parameters are $d=15$ Å, $\bar{\phi}=2$ V, $\Delta\phi=2$ V, $E_{F1}=E_{F2}=10$ V.

where $\Delta\phi=\phi_2-\phi_1$, $A_0=4(2m)^{1/2}d/3\hbar$, and $G(0)=(3.16\times 10^{10}\bar{\phi}^{1/2}/d)\exp(-1.025\bar{\phi}^{1/2})$. The conductance minimum occurs at

$$eV_{\min}=\frac{4}{9}(\Delta\phi/A_0\bar{\phi}^{1/2})=0.649(\Delta\phi/d\bar{\phi}^{1/2}), \quad (8)$$

where d is the barrier thickness in Å and the potentials are in volts. From (7) it is easy to see that the sum of the two voltages where the conductance doubles its minimum value is given by $11\bar{\phi}^{1/2}/d$.

$$j(V)=\frac{2e}{\hbar}\sum_k\int_{-\infty}^{\infty}(\text{Prefactor})\exp\left(-\frac{2}{\hbar}\int_0^d\{[\varphi(x,V)-E_x]2m\}^{1/2}dx\right)[f(E)-f(E-eV)]dE_x, \quad (9)$$

where the Prefactor term is given by

$$=16(E_x-\Delta E_F)^{1/2}/E_x^{1/2}\left[\frac{(\varphi_2-E_x)^{1/2}}{(\varphi_1-E_x)}+\frac{[(\varphi_2-E_x)(\varphi_1-E_x)]^{1/2}}{E_x}+\frac{(E_x-\Delta E_F)}{[(\varphi_2-E_x)(\varphi_1-E_x)]^{1/2}}+\frac{(\varphi_1-E_x)^{1/2}}{(\varphi_2-E_x)}\left(\frac{E_x-\Delta E_F}{E_x}\right)\right].$$

where $\Delta E_F=E_{F2}-E_{F1}$, the difference in Fermi energies of the two metals. This preexponential factor implies that in this model the tunneling current depends on the Fermi energies and also on the densities of states of the metals in an indirect fashion. The choice of Fermi energies therefore has an effect on the shape of these conductance curves. This point is clearly illustrated in Fig. 4 where the effect of differences in the Fermi energies

To illustrate the results of our calculations the computed conductances are presented in Figs. 1–3. First we fix the mean barrier height $\bar{\phi}=2$ V and the thickness $d=15$ Å. By varying $\Delta\phi$ from 0 to 3 V the minimum conductance of Fig. 1 moves from $V=0$ to 90 mV. In each case the conductance has been normalized to 1.0 at zero voltage and successive curves have been offset by 0.1 for clarity. In the extreme case of $\Delta\phi=3.0$ V the conductance decreases by nearly 4% from $V=0$ to 90 mV.

The thickness and mean height of the barrier also affect the offset of the conductance as they affect the symmetrical and asymmetrical conductance terms differently in Eq. (7). The rather striking effect of thickness variation is shown in Fig. 2. On increasing the thickness from 10 to 25 Å the parabolic term becomes much stronger and the offset decreases from 85 to 40 mV (for $\bar{\phi}=2.0$ V, $\Delta\phi=2.0$ V). Finally in Fig. 3 the relative insensitivity of the minimum position to changes in $\bar{\phi}$ is shown. For $d=15$ Å, $\Delta\phi=2.0$ V this position moves from 55 to 70 mV on changing $\bar{\phi}$ from 2.5 to 1.5 V. The results of Figs. 1–3 all agree with Eqs. (7) and (8) within the 10% accuracy stated earlier. It should be noted that the plots of Figs. 1–3 are not truly parabolic or symmetrical about the minimum conductance, as can be inferred from Eq. (7) which could be continued to include higher-order terms. For example the 25-Å barrier of Fig. 2 deviates by ~4% from parabolic behavior at 200 mV. In addition, the minimum conductance occurs at 40 mV whereas a conductance of $G=1.4$ is reached at -238 and $+144$ mV, which means the asymmetry has increased to 47 mV.

(2) Our second model for the barrier assumes that the boundaries between the metals and the oxide are sharp. Harrison¹⁴ and Davydov¹⁹ show that by matching suitable boundary conditions the current density in this case is given by

of the two electrodes is reflected in shifts of the asymmetry of the conductance. In this figure the conductance at $V=0$ for $E_{F2}/E_{F1}=1$ is normalized to 1 and the shifts of the two other curves represent the actual changes in relative conductance produced by varying the Fermi energies.

To illustrate the differences between the two extreme models considered here, we show in Fig. 5 a comparison

of the calculated conductances of models (1) and (2) using the same parameters ϕ_1 , ϕ_2 , and d . From this figure it is clear that, although there are differences between the two curves, the general parabolic behavior and the basic asymmetry of the plots remains.

THE INCLUSION OF IMAGE FORCES

The calculations of model (1) have been repeated taking into account image forces in the barrier using the approximations introduced by Simmons²⁰ for the shape of the reduced barrier. Instead of the trapezoidal shape of Eq. (5), the image force potential tends to round off the corners of the barrier, thus making it narrower and lower. This has the overall effect of increasing the conductance between the electrodes but, as shall be seen, does not qualitatively change the overall shape of the $G(V)$ curve.

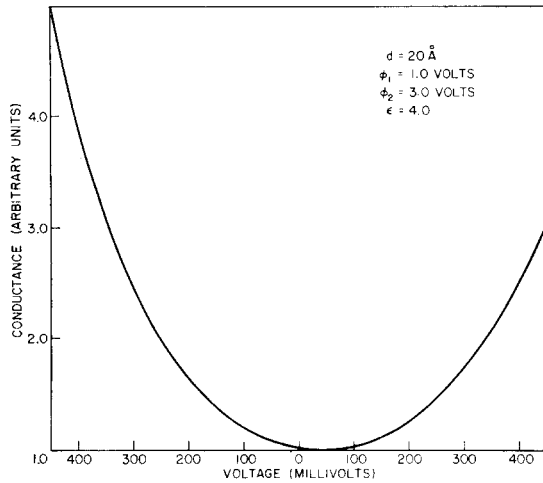


FIG. 6. Conductance versus voltage for a barrier with $d = 20 \text{ \AA}$, $\phi_1 = 1.0$, $\phi_2 = 3.0$, $\epsilon = 4.0$. The calculation used model I and included the effect of image forces.

Following Simmons, Eq. (5) can be revised to include these effects and the shape of the barrier is given approximately by

$$\varphi(x, V) = \varphi_1 + \frac{x}{d} (\varphi_2 - eV - \varphi_1) - \frac{1.15\lambda d^2}{x(d-x)}, \quad (10)$$

where $\lambda = e^2 \ln 2 / 8\pi\epsilon_0\epsilon d$, where ϵ is the dielectric constant of the barrier material. A model calculation employing this modification to the barrier shape is illustrated in Fig. 6 for $\bar{\varphi} = 2 \text{ V}$, $\Delta\varphi = 2 \text{ eV}$, $\epsilon = 4.0$, and $d = 20 \text{ \AA}$. The basic asymmetry of the conductance plot remains but a comparison with Fig. 2 tells us that the asymmetry is reduced and the conductance is rising faster for this calculation. This faster rising conductance can be understood as the result of two effects. First, the overall height of the barrier is reduced by an amount equal to the last term in Eq. (10). Second, and more important, the thickness of the barrier is a function of the bias (and of E_x), unlike the case of the

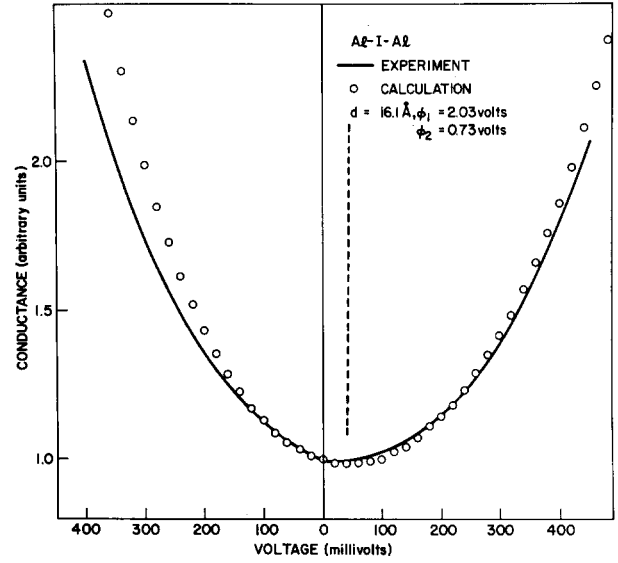


FIG. 7. The solid line is an experimental conductance-voltage plot for an Al-I-Al junction. The barrier parameters $d = 16.1 \text{ \AA}$, $\phi_1 = 2.03 \text{ V}$, $\phi_2 = 0.73 \text{ V}$ were determined as outlined in the text. The points are calculated using these parameters and the diffuse barrier model (1). The dashed line is the mean of the two voltages required to produce a given experimental conductance.

trapezoidal barrier. For example, in this model calculation, at a bias of -400 mV , the effective thickness of the barrier for electrons at the top of the Fermi surface of the negatively biased metal is $17.59 \text{ \AA}$, while at zero bias, the thickness is $18.14 \text{ \AA}$.

This, then, is the reason for the more rapidly changing conductance in this calculation, and as the thickness is a function of bias, it is expected that deviation from a parabolic behavior in the $G(V)$ curve will occur at a lower applied bias than for the trapezoidal barrier.

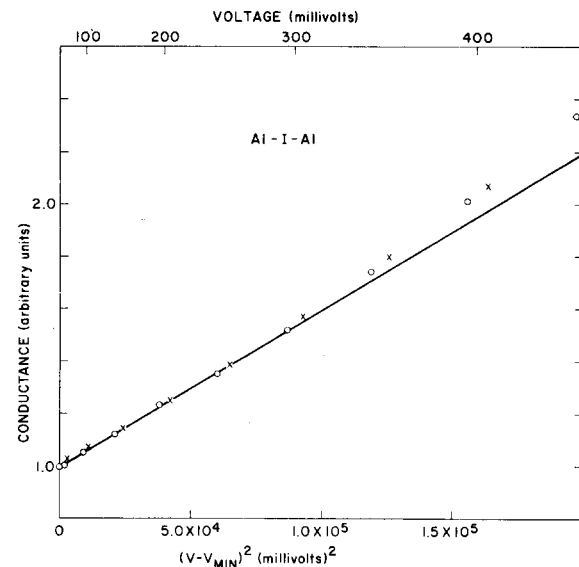


FIG. 8. The experimental data of Fig. 7, plotted as conductance versus $(V - V_{\min})^2$, where $V_{\min}$ is the asymmetry for conductances ~ 1 .

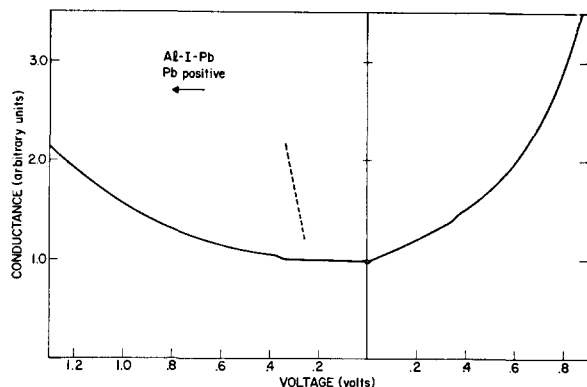


FIG. 9. A measurement of conductance versus voltage for an Al-I-Pb junction showing very large asymmetry of the characteristic. The dashed line shows the increase of asymmetry with increasing conductance.

However the differences in the overall shape of G versus V for barriers with and without the image force correction is not readily apparent from a casual examination of the plots and will only become important when all the barrier parameters are determined independently.

COMPARISON WITH EXPERIMENT

It has been our experience^{7,21} that conductance plots for junctions of the type Al-I-M ($M = \text{Pb, Sn, In, Al}$) and Pb-I-Pb do exhibit a roughly parabolic dependence on voltage with the minimum conductance offset from $V=0$. An example is shown in Fig. 7. From the minimum conductance (8.66 mho/cm^2), the sum of the voltages needed to double this conductance (784 mV), and the asymmetry (45 mV) the parameters of this junction are calculated from Eqs. (7) and (8) to be $d = 16.1 \text{ \AA}$, $\bar{\phi} = 1.38 \text{ V}$, $\Delta\phi = 1.3 \text{ V}$ ($\phi_1 = 2.03$, $\phi_2 = 0.73 \text{ V}$). The conductance-voltage plot calculated using the "diffuse barrier" model (neglecting image forces for simplicity) is compared with experiment in Fig. 7 and agrees within 20% from -300 mV to $+500 \text{ mV}$. As pointed out earlier, the asymmetry of the conductance about $V=0$ increases as the conductance increases. This is shown for the experimental data in Fig. 7 as the dashed line which locates the mean of the voltages required to produce a given conductance. It is also apparent that the experimental data of Fig. 7 is a better fit to a parabola than the calculation. This fit is illustrated in Fig. 8 where we plot G against $(V - V_{\min})^2$ where $V_{\min}$ is the asymmetry voltage (45 mV) for small conductances. Similarly good agreement for an Al-I-Sn junction, with larger asymmetry (125 mV) has been published previously.⁷ Whether the above approach is a reasonable way to parameterize a junction, when the asymmetry is rather small, can only be checked by future measurements of the barrier parameters by independent methods.

We cannot claim, however, that our simple calculation explains the conductance of many junctions where

the asymmetry is large, approaching 300 mV in some cases for thermally oxidized aluminum junctions. We show an extreme example of an Al-I-Pb junction with an asymmetry of 250 mV in Fig. 9. It can be seen in Fig. 9 that the asymmetry of 250 mV determined at conductances ~ 1 has increased to 320 mV when the conductance has doubled from its minimum value. The conductance at low voltages was 4.65 mho/cm^2 and the sum of the two voltages needed to double this conductance 1856 mV . These values give $d = 11 \text{ \AA}$ and $\bar{\phi} = 3.5 \text{ V}$ but the asymmetry of 250 mV used in Eq. (8) gives $\Delta\phi > 2\bar{\phi}$. Thus our model is not able to describe this type of tunneling barrier. We have noticed two consistent correlations in this type of junction with large asymmetries. First the conductance has a very slow dependence on voltage which, if one attempts to parameterize the junction in the way described above, results in a somewhat thin barrier (typically $10\text{--}12 \text{ \AA}$ for our junctions) but a very large barrier height (3 to 3.8 V). Second we have noticed that junctions which have large asymmetries consistently show strong structure due to molecular excitations.⁶ This is easiest to see at 360 mV , the energy of the C-H stretching modes. In the Al-Pb junction of Fig. 9 this excitation produces an increase in conductance of almost 3%; in the Al-Al junction of Fig. 7 it is unobservable against the rapidly rising conductance and is estimated to be $< 0.2\%$. It is therefore tempting to suggest that a layer of organic impurities in the barrier oxide leads to an asymmetry of the potential which is much larger than one would expect on the basis of differences in metal work functions. This organic layer could also result in a high average barrier. Arguments similar to this have been used recently to explain the rather flat conductance in some metal-semiconductor contacts.²² Whether the role of the organic impurities in the oxide junction is simply to affect the barrier shape remains to be seen; one can speculate about more exotic mechanisms involving the self-field of the junction and its effect on polarizable molecules. An examination of the experimental data of Figs. 7 and 9 reveals that, although the asymmetry is quite marked at conductances ~ 1 , the minimum con-

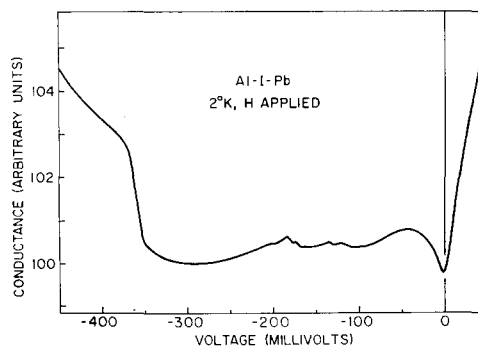


FIG. 10. A detailed measurement of conductance versus voltage for an Al-I-Pb junction. This figure illustrates the various excitation effects which change the conductance by a few percent.

ductance actually occurs at $V=0$. We believe this is because, in most cases, excitation effects "fill in" the minimum in conductance we expect to occur at some finite voltage. Occasionally the minimum does occur away from $V=0$; such an example has been published earlier (Fig. 3 of Ref. 21). A more common behavior is shown in Fig. 10 for an Al-I-Pb junction. This expansion near $V=0$ shows a 1% increase in conductance from 0–40 mV which we believe is due to excitation of phonons in the electrodes.²¹ The local increases in conductance near 120, 180, and 360 mV are due to interactions with water and organic molecules in the oxide.⁶ However, the important feature to notice is that, apart from regions of excitation, the conductance decreases ($dG/d|V|$ negative) out to nearly 300 mV. As far as we know, the only possible origin of such an effect is that the background conductance itself must be decreasing to a minimum located away from $V=0$.

DISCUSSION

In this paper we have attempted to explain the conductance characteristics of normal metal-oxide insulator-normal metal tunnel junctions using a simple model of transmission through a trapezoidal potential barrier. We have reached three conclusions.

(1) A trapezoidal barrier does lead to a roughly parabolic dependence of conductance on voltage with the minimum conductance offset from $V=0$. This may be a useful check of the existence of tunneling up to high voltages (~ 5 V) if it is observed in conjunction with a superconducting energy gap at low voltages. For junctions showing relatively small offsets of the minimum conductance (say 50 mV) satisfactory agreement can be obtained between the experimental characteristic and the calculation using reasonable junction parameters. Whether these parameters are the correct ones can only be decided by determining them independently.

(2) When a junction shows an extreme offset of the parabolic dependence (say 250 mV) our model fails to reproduce such large effects. Examination of the data from this type of junction suggests that the barrier

height must be large and the potential distorted in a more drastic way than we have considered.

(3) We believe that there is a correlation between the size of the asymmetries discussed above and the strength of the conductance increases arising from the excitation of organic molecules. It seems reasonable to postulate that the presence of these organic molecules in the junction is therefore the cause of a severe distortion of the barrier shape.

In summary we would suggest that the most important conclusion we have reached is that tunneling between normal metals is not yet a field that can be regarded as fully understood. It is obvious that much careful experimental work is necessary before the nature of the potential barrier can be understood further.

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