TEMPERATURE DEPENDENCE OF ELECTRON TUNNELING

by

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Date August, 1966
The variation with temperature of electron tunneling through thin insulating films of Al$_2$O$_3$ between aluminum film electrodes was studied at liquid helium temperatures (1.18°K to 4.2°K) and liquid oxygen temperatures (65°K to 90°K).

Samples were prepared in a vacuum evaporating system using the method developed by Fisher and Giaever (1961). The aluminum oxide films were grown in air at room temperature.

Resistance was measured as a function of voltage by means of a d.c. Wheatstone bridge for sample currents in the region of $10^{-7}$ amps. Conduction due to electron tunneling was indicated by the non-ohmic voltage dependence of resistance and by the fact that the resistance increased with a decrease in temperature.

The voltage dependence of the tunneling resistance was found to be in good agreement with the theory as derived by Simmons (1963), and was parabolic for low voltages.

Reliable and reproducible data for the temperature dependence of resistance were difficult to obtain because of instabilities in the samples and a general increase in resistance due to aging. Comparison of the observed temperature dependence with the theory of Simmons (1964) showed the variation to be approximately twice as great as predicted.
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INTRODUCTION

When two metal electrodes are separated by an insulator, classical electrons can cross the junction only if they have enough energy to surmount the potential energy barrier of the insulator and flow in its conduction band. For a good insulator and normal applied voltages, the conduction band energy is so much greater than the average energy of the electrons in the metal that the current is essentially zero. According to classical physics, an electron of energy $E$ approaching a potential barrier of height greater than $E$ cannot possibly cross the barrier; it has no choice but to be reflected.

However, quantum mechanics predicts that this is not strictly true; the electron has a finite probability of crossing the barrier even if it does not have the energy to "climb over the top". The electron impinges on one side of the barrier and simply reappears on the other side. This remarkable phenomenon, known appropriately as "tunneling", arises from the wave nature of the electron.

Theoretically, the finite probability of tunneling applies not just to electrons but to any object approaching any barrier. There must obviously be some quantitative restriction which ensures that for example, an automobile approaching a brick wall at fifty miles per hour has a truly negligible chance of emerging undamaged on the other side, in agreement with empirical fact. This restriction is imposed by the quantum mechanical transmission coefficient which determines the probability of tunneling and depends exponentially on the height and width of the barrier. If the height of the barrier is much greater than the energy of the electron and the width is more than a few tens of angstroms the tunneling probability
rapidly becomes infinitesimal.

It was this stringent requirement on the width of the insulating barrier that, until fairly recently, made experimental metal-insulator-metal tunneling junctions impossible. The general theory, however, was first worked out in 1933 by Sommerfeld and Bethe, who considered the cases of very low and very high voltages applied across two metal electrodes separated by a thin insulating film. Holm (1951) developed the theory for intermediate voltages.

In 1960, Fisher and Giaever (1961) developed a technique for producing experimental tunneling junctions by evaporating an aluminum film onto a glass substrate, allowing the film to oxidize and then evaporating another layer of aluminum over it. The aluminum oxide film, being about 50 Å thick, was a thin enough potential barrier to allow a measurable tunneling current for applied potentials of about 1 volt.

This development sparked a great interest in tunneling junctions, both theoretically and experimentally. Simmons (1963) extensively refined the theory, developing an accurate treatment of the shape of the barrier potential under the influence of image forces, generalizing the results for a barrier of arbitrary shape, and correcting several discrepancies in Holm's theory. He also expressed the tunneling current explicitly in terms of the measurable parameters such as applied voltage, barrier height and oxide film thickness.

Experimentally, Morris and Pollack (1964) improved on the sample preparation and current-measuring techniques of Fisher and Giaever and verified Simmons' theory for the current-voltage characteristics of asymmetric junctions (those in which the electrodes are of different metals).
over a current range of nine orders of magnitude. Their measurements, taken at 77° K, were in good agreement with Simmons' theory for 0° K.

Soon afterwards, Simmons (1964) extended his theory to the case of finite temperatures and derived expressions for the temperature dependence of the tunneling current. This work was the basis for the experiment described here: the study of the temperature dependence of the tunneling current for symmetric junctions at low temperatures (1° K to 90° K).
THEORY

The work of Simmons (1963, 1964) is the most complete and accurate study of the theory of tunneling junctions to date. Like the analyses of Sommerfeld and Bethe (1933) and Holm (1951), it is based on the WBK approximation of the transmission coefficient of an electron through a potential barrier. The analysis is first carried out for an assumed temperature of 0°K and is then extended to the case of non-zero temperatures.

Notation

\begin{align*}
  m &= \text{mass of electron} \\
  e &= \text{charge of electron} \\
  \hbar &= \text{Planck's constant} \\
  s &= \text{insulating film thickness} \\
  s_1, s_2 &= \text{limits of barrier at Fermi level} \\
  \Delta s &= s_2 - s_1 \\
  \eta &= \text{Fermi level energy} \\
  f(E) &= \text{Fermi function} \\
  \psi &= \text{work function of metal electrodes} \\
  \% &= \text{height of rectangular barrier} \\
  \bar{\psi} &= \text{mean barrier height} \\
  \epsilon &= \text{permittivity of insulating film} \\
  \kappa &= \text{dielectric constant of insulating film} \\
  J &= \text{tunneling current density} \\
  V &= \text{applied voltage} \\
  R &= \text{tunneling resistance} \\
\end{align*}

**Tunneling Current at Absolute Zero of Temperature**

The probability that an electron whose \( x \) component of energy is \( E_x = \frac{1}{2} m v_x^2 \) will tunnel through the barrier from left to right is given by the transmission coefficient

\[
D(E_x) = \exp \left\{ -\frac{\psi}{\hbar} \int_{S_1}^{S_2} \left( V(x) - E_x \right)^{\frac{1}{2}} \, dx \right\} \tag{1}
\]

The number of electrons tunneling from left to right is

\[
N_1 = \int_0^{\psi m} n(v_x) \, D(E_x) \, dv_x
\]

Since \( E_x = \frac{1}{2} m v_x^2 \) and \( dE_x = m v_x \, dv_x \),

\[
N_1 = \frac{1}{m} \int_0^{\psi m} n(v_x) \, D(E_x) \, dE_x
\]
FIG. 1 The potential barrier due to an insulator between two metal electrodes.

\[ \psi(x) \]

\[ \varphi(x) \]

\[ V(x) \]

NEGATIVE ELECTRODE

INSULATOR

POSITIVE ELECTRODE

\[ 4\pi me \frac{e^2}{\hbar^2} f(E) \]

\[ Y(E_x) = 4\pi me \int_{E_x}^{\infty} [f(E) - f(E+eV)] dE \]

FIG. 2 The function \( Y(E_x) \) is the integral of the difference between two Fermi functions.
Em is the maximum energy of the electrons in the electrodes, \( v_m \) is the corresponding velocity, and \( n(v_x) \) is the number of electrons per cm\(^3\) with velocities between \( v_x \) and \( v_x + dv_x \), and is given by
\[
\eta(v_x) = (2m^3/\hbar^3) \int f(E) dv_x \int_0^\infty f(E) dv_y \int_0^\infty f(E) dv_z
\]
\[
\therefore \eta(v_x) = (2m^3/\hbar^3) \int_0^\infty \int_0^\infty f(E) dv_y dv_x
\]
If we substitute \( v_x^2 = v_y^2 + v_z^2 \) and \( E_r = \frac{mv_x^2}{2} \)
\[
\eta(v_x) = (4\pi m^2/\hbar^3) \int f(E) dE_r
\]
\[
\therefore \eta(v_x) = (4\pi m^2/\hbar^3) \int D(E_x) dE_x \int_0^\infty f(E) dE_r
\]
Similarly, the number of electrons tunneling from right to left is
\[
N_2 = (4\pi m^2/\hbar^3) \int D(E_x) dE_x \int_0^\infty f(E + eV) dE_r
\]
If the right-hand electrode is positive, the Fermi function is written \( f(E + eV) \). The transmission coefficient is the same in both directions.
The net number of electrons crossing from left to right is then
\[
\mathcal{J} = eN = e (N_1 - N_2)
\]
\[
= \int_0^{E_m} D(E_x) dE_x \left\{ (4\pi me/\hbar^3) \int \left[ f(E) - f(E + eV) \right] dE_r \right\}
\]
Let \( Y(E_x) = (4\pi me/\hbar^3) \int \left[ f(E) - f(E + eV) \right] dE_r \)
\[
= (4\pi me/\hbar^3) \int_0^\infty \left[ f(E) - f(E + eV) \right] dE
\]
Then
\[
\mathcal{J} = \int_0^{E_m} D(E_x) Y(E_x) dE_x
\]
\( Y(E_x) \) is the integral of the difference between two Fermi functions, the shaded area in the diagram of Fig. 2.
\[
Y(E_x) = \begin{cases} 
(4\pi me/\hbar^3)eV & \text{for } 0 < E_x < \eta - eV \\
(4\pi me/\hbar^3)(\eta - E_x) & \text{for } \eta - eV < E_x < \eta \\
0 & \text{for } E_x > \eta
\end{cases}
\]
From Fig. 1 we see that \( V(x) = \eta + \varphi(x) \). Then the transmission coefficient becomes
\[
D(E_x) = \exp\left\{ -(4\pi/\hbar)(2m)^{1/2} \int_0^{S_x} \left[ \eta + \varphi(x) - E_x \right]^{1/2} dx \right\}
\]
This can be written approximately as

\[ D(E_x) \approx \exp \left[ -A \left( \eta + \Phi - E_x \right)^{1/2} \right] \]  

where \( A = \left( \frac{4\pi m \Delta s}{h} \right) (2m)^{1/2} \) and \( \Phi \) is a correction factor \( \approx 1 \). (See appendix in Simmons (1963)).

Then the expression for the current density becomes

\[ J = (4\pi m e/h^3) \left\{ \frac{eV}{\Phi} \int_{\eta}^{\eta-eV} \exp \left[ -A \left( \eta + \Phi - E_x \right)^{1/2} \right] dE_x \right. 
+ \left. \int_{\eta-eV}^{\eta} \exp \left[ -A \left( \eta + \Phi - E_x \right)^{1/2} \right] dE_x \right\} \]  

If we make some suitable approximations, Eq. (6) integrates to

\[ J = \frac{2\pi h}{(\Phi \Delta s)} \left\{ \Phi \exp(-A \Phi^{1/2}) - (\Phi+eV)\exp(-A(\Phi+eV)^{1/2}) \right\} \]  

The Low Voltage Limit \( (V \approx 0) \)

In this limit we simplify Eq. (7) making the approximation that \( eV \ll \Phi \), (\( \Phi \) is usually 1 or 2 electron volts), and also making use of the fact that \( A \Phi^{1/2} \gg 1 \). (For typical values of barrier thickness, the value of \( A \Phi^{1/2} \) lies between 10 and 50). We then obtain

\[ J = \frac{e^2 (2m)^{1/2} \Phi^{1/2}}{\hbar^2 \Delta s} \exp(-A \Phi^{1/2}) \]  

The important point to be noted here is that \( J \) is linear in \( V \), i.e. the tunneling junction is ohmic for very low voltages.

Eq. (8) can be further simplified by noting that for very small voltages, the barrier shown in Fig. 1 is almost rectangular. Then \( \Delta s = s_2 - s_1 \approx s \) and the mean barrier height \( \Phi \) can be considered equal to the rectangular barrier height \( \Phi_b \).

Then (8) becomes

\[ J = \left\{ \frac{3(2m \Phi_b)^{1/2}}{2s} \left( \frac{e}{h} \right)^2 V \right\} \exp\left[ -\frac{4\pi s}{h} (2m \Phi_b)^{1/2} \right] \]  

The Intermediate Voltage Range \( (V \leq \Phi_b/e) \)

In this range \( \Delta s = s \)

and \( \Phi = (\Phi - eV/2) \)
Also, for voltages below 0.75 \( \frac{V_0}{e} \), the proportionality factor \( \beta \) can be set equal to unity with negligible error.

Making these substitutions we obtain

\[
J = \left( \frac{e}{2\pi h} \right) \left\{ (\gamma_0 - \frac{eV}{2}) \exp \left[ -\frac{4\pi s}{h} (2m)^{1/2} \left( \gamma_0 - \frac{eV}{2} \right)^{1/2} \right] - (\gamma_0 + \frac{eV}{2}) \exp \left[ -\frac{4\pi s}{h} (2m)^{1/2} \left( \gamma_0 + \frac{eV}{2} \right)^{1/2} \right] \right\}
\]

Expanding the exponentials to third order terms we obtain

\[
J \approx \frac{e}{2\pi h} \exp(-c) \left\{ (c-2) x - \frac{1}{6} c (1+c-\frac{1}{3} c^2) x^3 \right\}
\]

where \( c = A \gamma_0^{1/2} \) and \( x = eV/2 \gamma_0 \).

The junction resistance in ohm \( \cdot \) cm² is then \( R = V/J \).

\[
R = \frac{4\pi h s^2 \exp(c)}{e^2 (c-2)} \left\{ 1 + \frac{c(1+c-\frac{1}{3} c^2)}{8 (c-2)} x^2 \right\}
\]

If \( c \gg 2 \), then

\[
R \approx \frac{h^2 s \exp(A \gamma_0^{1/2})}{e^2 \left( \frac{2}{r_i \gamma_0} \gamma_0 \right)^{1/2}} \left\{ 1 - \frac{A^2 e^2}{96 \gamma_0^2} V^2 \right\}
\]

This expression is valid only if \( \frac{A^2 e^2}{96 \gamma_0^2} V^2 < 1 \). Typically, this condition is satisfied if \( V \) is less than about 0.3 volts.

Then from Eq. (12) it can be seen that the resistance drops off parabolically from a low voltage limiting value. This behaviour is illustrated in Fig. 3.

**The Image Force**

So far we have considered the tunneling barrier to be rectangular. This is assumed in the approximation leading to Eq. (9) and is implied in the expression for the transmission coefficient (1). Actually, the barrier is slightly rounded by the image potential as indicated in Fig. 1.

The image potential is a hyperbolic function which, when included in the integral of (1), makes it soluble only by numerical methods. One alternative is to approximate the image potential by a symmetric
FIG. 3 Voltage dependence of tunneling resistance for various barrier thicknesses and a barrier height of 1 eV.
From Simmons (1963).
parabola as has been done by Sommerfeld and Bethe and by Holm. However, this approximation is rather crude and works well only for low voltages, since an applied voltage makes the barrier asymmetric.

Simmons has overcome this difficulty by approximating the true image potential by a simpler hyperbolic function which is quite accurate and can also be readily solved. Taking the image potential into account results in a corrected value for the barrier height \( \varphi \), which is now a function of barrier thickness and the dielectric properties, \( K \) and \( \varepsilon \), of the insulator. The limits of the barrier at the Fermi level, \( s_1 \) and \( s_2 \) are also functions of \( \varepsilon \) and \( \varphi_0 \).

In the low voltage case, the corrected value of barrier height reduces numerically to

\[
\varphi = \varphi_0 - \left[ 11.5/(Ks - 12/\varphi_0) \right] \ln(5K\varphi_0/6 - 1) = \varphi_L
\]

This value of \( \varphi \) would then be used in Eq. (8) to obtain the low voltage current density.

Similarly, a corrected expression for \( \varphi \) applicable to the intermediate voltage range would be used in Eq. (7).

Simmons (1964) notes that in practice, especially for low or moderate voltages, the actual barrier is practically rectangular if \( K > 4 \). (For \( Al_2O_3 \), \( K \) varies from 8 to 10). Consequently, the image force will be neglected in our analysis.

The Tunneling Current at Finite Temperatures

In the foregoing discussion we assumed a temperature of \( 0^\circ K \). The tunneling theory for finite temperatures is formally identical with that set out in the previous section, except that the Fermi functions in Eq. (2) are no longer degenerate.
In general, the integral of the Fermi function is
\[ \int f(E) \, dE = \int \frac{dE}{1 + \exp((E - \eta)/kT)} = -kT \ln \left[ 1 + \exp((\eta - E)/kT) \right] \]

Then \[ \int_{E_x}^{\infty} f(E) \, dE = kT \ln \left[ 1 + \exp((\eta - E_x)/kT) \right] \]

Then the function \( Y(E_x) \) of (3) becomes
\[ Y(E_x) = (4 \pi m e / h^3) \int_{E_x}^{\infty} \left[ f(E) - f(E + eV) \right] dE = (4 \pi m e / h^3) \ln \left\{ \frac{1 + \exp \left[ \left( \eta - E_x \right)/kT \right]}{1 + \exp \left[ \left( \eta - E_x - eV \right)/kT \right]} \right\} \]

Using the expression for \( D(E, \eta) \) given in (5), we have
\[ J(V, T) = (4 \pi m e kT / h^3) \int_{E_x}^{\infty} \ln \left\{ \frac{1 + \exp \left[ \left( \eta - E_x \right)/kT \right]}{1 + \exp \left[ \left( \eta - E_x - eV \right)/kT \right]} \right\} X \exp \left[ -A \left( \eta + \bar{\phi} - E_x \right)^{1/2} \right] dE_x \]

With the help of some simplifying approximations, (13) reduces to
\[ J(V, T) = \frac{4 \pi m e}{h^3 B^2} \left[ \frac{\pi B kT}{\sinh(\pi B kT)} \right] \exp(-A \bar{\phi}^{1/2}) \left[ 1 + \exp(B eV) \right] \]

where \( B = A/2 \bar{\phi}^{1/2} \)

When \( T = 0 \), (14) becomes
\[ J(V, 0) = \frac{e}{2 \pi \hbar (\beta \Delta s)^2} \left[ \bar{\phi} \exp(-A \bar{\phi}^{1/2}) - \bar{\phi} \exp[A(\bar{\phi} + eV)^{1/2}] \right] \]

Comparing (15) with (7), we see that they are identical except that (7) has a factor of \( (\bar{\phi} + eV) \) in the last term where (15) has a factor of \( \bar{\phi} \).

This discrepancy arises because of an approximation used in deriving (15).

Eq. (7) is the correct form because it was obtained without resorting to this approximation, but since \( eV \ll \bar{\phi} \), the difference is not significant.

The temperature dependence of the tunneling current at a given voltage is found from Eq. (14):
\[ J(V, T) \approx J(V, 0) \left\{ 1 + \left[ 3 \times 10^{-9} \frac{(\Delta s T)^2}{\bar{\phi}} \right] \right\} \]

Here \( J \) is expressed in amps/cm², \( \Delta s \) in angstroms, \( T \) in degrees Kelvin and \( \bar{\phi} \) in volts.

Eq. (16) is the general result for any barrier shape or voltage
range. To apply it to a particular voltage range with or without correction for the image potential, the appropriate expression for $J(V, 0)$ is used.

Discussion

We have seen that the tunneling resistance at a given temperature is ohmic for very low voltages, and for $V < \frac{\varphi_0}{e}$ drops off roughly parabolically with increasing voltage. Similarly, at a fixed voltage, the temperature dependence of the resistance is also parabolic as can be seen from Eq. (16). However, the dependence is rather weak: for typical values of barrier thickness $\Delta s$ and barrier height $\varphi$, the tunneling current is expected to change by only about 6% between the temperatures of 0°K and 77°K.

The resistance is exponentially dependent on barrier thickness and height and furthermore is strongly affected by the dielectric constant of the insulator, since this influences the image force and hence the shape and effective thickness of the barrier. (See Fig.'s 7 and 9 of Simmons (1963)). Thus the tunneling current depends not only on the properties of the electrodes but also on those of the insulator.
Thin Aluminum Oxide Films

The electrical properties of thin oxide films are the subject of intensive investigation and are still not well understood. Quite often samples are irreproducible in their electrical behaviour and investigators will differ in their observations. The reason for this appears to be that the properties of an oxide film are critically dependent on many factors influencing it during its preparation.

As was mentioned earlier, Fisher and Giaever (1961) pioneered the technique of producing tunneling junctions by allowing a thin film of evaporated aluminum to oxidize and then evaporating another film of aluminum or other metal over it. Obviously such considerations as pressure, temperature and cleanliness of the vacuum system are important but other apparently minor factors, such as the rate of deposition of both films, also affect the properties of the junction.

Furthermore, the junctions exhibit a marked aging effect which also varies with the method of preparation of the sample and the conditions under which it is subsequently stored.

Growth of Aluminum Oxide Films

Hunter and Fowle (1956) have made a study of the growth of \( \text{Al}_2\text{O}_3 \) films under various conditions. Film thickness was measured by making the sample (a piece of oxidized aluminum) the anode in an electrolytic bath and noting the potential necessary to produce a normal leakage current. The applied potential is a direct measure of the oxide thickness. However, this method measures only that thickness of oxide which is electrically insulating and it is known that in general these
films consist of an insulating "compact" or "barrier" layer covered by a porous, electrically permeable layer.

The explanation of this is as follows: If a piece of aluminum is oxidized in dry air or oxygen, the oxide very quickly builds up to a limiting thickness which varies as the temperature, confirming that the depth of penetration of the oxidation is determined by thermal motion of the metal atoms. This oxide layer is all of the compact type. If the sample is now exposed to moist air, the water vapour attacks the surface of the oxide and breaks it down into a porous layer. As the barrier is broken down, more of the metal oxidizes until finally, after months or even years, an equilibrium barrier thickness is reached. This thickness is determined by the temperature of the environment independently of the temperature at which the original barrier layer was formed. If the sample is oxidized in moist air, the oxidizing and hydrating effects go on simultaneously. The final barrier thickness is the same as that formed in dry air at the same temperature but it is now covered with a porous layer. It is thus apparent that the maximum possible barrier thickness is a function of temperature only.

When a junction is made by evaporating a second layer of metal several hundred å thick over the newly formed oxide, the deterioration of the barrier oxide by water vapour should be greatly inhibited. This is confirmed by the fact that the resistance of a metal-oxide-metal sandwich almost invariably increases with age, whereas one would expect it to decrease if the insulating oxide layer were being decomposed by water vapour. Furthermore, this increase has been observed even when the sample is stored in an inert atmosphere or a vacuum. The aging effect is much
smaller and the characteristics of the sample are more stable and reproducible if it has been oxidized at temperatures of 200°C to 350°C.

Experimental Behaviour of Tunneling Junctions

The original tunneling samples of Fisher and Giaever were made by evaporating a strip of aluminum about 3/64" wide and several hundred angstroms thick along a glass microscope slide, allowing it to oxidize, and then evaporating several strips of aluminum of varying widths at right angles across it. Electrical contacts were made to the ends of the strips and d.c. J-V characteristics were measured with a volt-meter and ammeter at room temperature and at liquid nitrogen temperature. Voltages up to 1.4 volts were applied and currents up to about 10 milliamps were observed.

It was found that the junctions were ohmic at very low voltages, the current increasing exponentially at higher potentials and being roughly proportional to the junction area. Samples oxidized at high temperatures (400°C) were found to be slightly rectifying and the preferred direction reversed if the sample was cooled to 77K. These samples were also photovoltaic, developing a maximum open circuit voltage of 35 mv. Neither of these effects was observed in samples oxidized at room temperature. The thickness of the oxide film was calculated from the electrical capacitance of the junction. Results agreed qualitatively with the theory of Holm but the currents were consistently higher than those predicted, usually corresponding to the theoretical resistivity of an oxide film 1/3 of the measured thickness.

In the course of verifying experimentally the improved tunneling theory developed by Simmons (1963), Morris and Pollack (1964) made several important discoveries concerning the preparation of tunneling junctions
and the measurement of their characteristics.

Firstly they found that for samples oxidized at 23°C, a significant portion of the current came from edge emission at the places where the counter-electrode passed over the edges of the oxidized electrode. This had the effect of placing in parallel with the oxide resistance a short-circuiting resistor of about 4 ohms per mm. of edge, and was detected by the fact that the resistance of the samples depended on edge length and not on junction area.

In order to overcome this difficulty, a film of SiO about 1000 Å thick was evaporated over the edges of the oxidized electrode, leaving a central strip bare for contact with the counter-electrode. These shields also made it possible to apply higher voltages to the sample without edge break-down.

The second discovery was that at room temperature an applied potential of a few volts across the sample created a field that was more than enough to cause aluminum ion migration through the oxide. This obviously changes the sample and means that the current can no longer be attributed to electron tunneling. However, this ion current required a finite length of time to set in, the time varying inversely with temperature. If the measurements were taken quickly enough, reproducible tunneling characteristics could be obtained. For currents below 10^{-7} amps at 77°C, the change in current with time was so small that d.c. measurements could be made. For measurements at higher currents, the sample was placed across one arm of a Wheatstone bridge whose output voltage was directly proportional to the sample current. A triangular input voltage lasting from 2 to 60 seconds was applied and the output was automatically recorded.
on an X-Y plotter. If two consecutive identical input pulses produced the same output trace it was assumed that the conduction was due predominantly to tunneling and not ion migration. At higher currents and temperatures the time required for ion migration to begin was so short that electronic pulse techniques were used.

The ion migration at room temperature was found to produce hysteresis loops on a J-V graph and this anomalous behaviour could be quenched in by cooling the sample to liquid nitrogen temperature within 20 minutes of taking the measurement at room temperature. Consequently, although the sample is not permanently damaged by inducing ion migration at room temperature, care should be taken not to apply a d.c. voltage to it immediately before cooling.

The energy-barrier model proposed by Morris and Pollack includes an n-type semiconducting transition region between the oxide and the electrode on which it was grown. This region has a small step-like barrier of about 0.1 eV but since it is small compared to the oxide barrier height (about 2 eV), its main effect is to reduce the thickness of the insulating barrier, and most of the voltage applied to the sample appears across the oxide barrier. The counter-electrode is known to penetrate the oxide layer to a depth of about 5 Å, but the electronic barrier interface is considered to be quite sharp.

It was found that even for symmetric junctions in which both electrodes were aluminum, the barrier was trapezoidal rather than rectangular with a difference in height of 0.92 eV between the two metal-oxide interfaces.

Morris and Pollack found, as had Fisher and Giaever, that the
value of oxide thickness determined by capacitive measurement was greater by a factor of 1.5 to 2 than the value calculated from the tunneling characteristics.

In view of the built-in barrier asymmetry of 0.92 eV, the thickness measurements of Hunter and Fowle (1956), which were determined from the applied potential required to produce a certain leakage current, would have to be corrected. This would mean that for a given oxidation temperature, the barrier oxide thickness would be greater than originally calculated. This corrected value agreed fairly well with the thickness determined capacitively and the large discrepancy between these values and the thickness calculated from tunneling is still unexplained.

Conclusions

From the foregoing discussion we can summarize several important considerations to be kept in mind during the preparation and measurement of tunneling junctions:

1. In order to produce a compact oxide layer, the sample should be oxidized in dry air and stored in a dry atmosphere or vacuum.
2. Shields of evaporated SiO should be used to cover the edges of the oxidized electrode to prevent edge emission.
3. If d.c. measuring techniques are to be used, currents must not exceed $10^{-7}$ amps and temperatures should not be much more than that of liquid nitrogen ($77^\circ$K), otherwise aluminum ion migration begins to set in.
4. Samples can be tested at room temperature without harming them as long as measuring currents are kept low and the sample is left unbiased for at least 20 minutes before cooling to low temperatures.
EXPERIMENT

Sample Preparation

Samples were prepared in a Veeco 400 evaporator belonging to the Electrical Engineering department. This versatile system contained six evaporating chambers. The five masks used during the production of the samples were etched photographically from thin shim-stock copper sheets and each one was positioned over a separate evaporating chamber about 8" above the filament. The glass substrate was placed in a holder which could be rotated from outside the vacuum system so as to position the substrate over each of the masks in turn. The clearance between the substrate and the masks was about 1 mm.

The steps in the production of samples are illustrated in Fig. 4. First, nine strips of aluminum about 800 Å thick are evaporated onto a 2 x 2" piece of optically flat substrate glass (a). Next, a gold film from 400 Å to 800 Å thick is deposited over the "lands" at the ends of the strips in order to prevent these areas from oxidizing later, and to provide a good surface for soldering electrical leads (b). Silicon oxide shields about 1500 Å thick are evaporated over the edges of the strips (c), and the system is opened to the atmosphere to oxidize the aluminum (d).

After a typical oxidation time of 2 hours, the system is evacuated again and the nine counterelectrodes are deposited and their ends covered with a gold film, (e) and (f). The electrodes are of varying widths so as to produce junctions with areas of 1, 2, 4 and 8 mm$^2$. This method produces nine samples under identical conditions. The samples are then cut apart with a glass-cutter and leads are attached.
FIG. 4  Steps in the preparation of a tunneling junction.
Substrates and Cleaning

The substrates used were special Corning Pyrex 7059 borosilicate substrate glass 2" square. These plates are optically flat to within 60 Å. The manufacturer recommends only very simple cleaning if any, since the substrates are supposed to be exceptionally clean even if taken straight from the package. Small lint particles can be removed with a brush and the glass may be washed in de-ionized water.

During the course of the experiment, several cleaning methods were tried. Sometimes the substrate was simply brushed free of lint before using, sometimes it was further cleaned by means of a glow discharge in the vacuum system just prior to evaporation, and on one occasion it was cleaned by means of ultra-sonic vibration in distilled water. The difference in cleaning techniques did not seem to have any marked effect on the properties of the samples.

Evaporating Sources

The gold and aluminum used (both 99.999% pure) were in the form of wire and were wound on straight filaments of stranded tungsten. In all cases, the substrate was shielded from the source while the metal was melted, so as to wet the filament and remove any surface contaminants, like grease or aluminum oxide. The substrate was then exposed and the metal evaporated by passing 50 to 70 amps through the filament for 3 min. Usually the metal was completely evaporated within 2 min., signifying an evaporation rate of 5 to 7 Å per sec. for aluminum and 4 or 5 Å per sec. for gold.

The silicon oxide was in the form of pellets which were crushed and placed in a tantalum foil box with a tungsten filament inside it. A
current of 30 amps was passed through the filament for 5 to 10 min., evaporating about 1500 Å of oxide at a rate of 2.5 to 5 Å per sec.

The pressure in the system varied from $5 \times 10^{-6}$ to $2 \times 10^{-5}$ torr during the evaporations, as measured by an ionization gauge situated a few inches above the substrate.

**Measurement of Evaporated Film Thickness**

The thickness of the evaporated films was monitored by means of a quartz crystal with a resonant frequency of about 5 megacycles per sec. The crystal was driven by a transistorized oscillator and was situated just below and to one side of the substrate so as to be exposed to the evaporating source. As the metal film built up on the crystal, its mass increased and its resonant frequency correspondingly decreased. The change in frequency was registered on a Beckman digital display pulse counter. The crystals had been calibrated so that, by means of a simple conversion factor, the change in frequency during evaporation could be translated into film thickness in angstroms.

**Oxidation**

According to the study made by Hunter and Fowle (1956), an oxide-layer formed on high purity aluminum in dry air at room temperature reaches an ultimate thickness of about 10 Å in two hours. If allowance is made for the barrier asymmetry as mentioned earlier, this value increases to about 20 Å.

In the present experiment, samples were usually oxidized in ordinary air at room temperature for a period of 1 to 2 hrs., although times as short as 15 min. and as long as 60 hrs. were tried. In the former case the resistance of the junctions was essentially zero, and in the
latter case it was many megohms and could not be measured with our equipment. It became apparent that the optimum time for producing resistances of $10^4$ to $10^5$ ohms was about 2 hrs.

There was, however, an extremely high failure rate. In a batch of nine samples it was not unusual to find six of them short circuited. At first it was thought that this was due to dust particles on the substrate, which left pin-holes in the oxide film and short circuited the junction. Accordingly, the various cleaning techniques mentioned above were tried, none of which appeared to have any effect on the failure rate.

Even among samples that were not shorted, there was often an order of magnitude difference in the resistances of junctions from the same batch and there did not seem to be any correlation between resistance and junction area. Results were the same for samples oxidized in ordinary air and in dry air. A satisfactory explanation of these effects is still lacking.

**Filamentary Bridges**

One of the possible reasons for the failure of a sample is that one or more filaments of aluminum have penetrated the oxide layer through pin-holes or other faults in the structure, forming a conducting bridge between the electrodes. The presence of these filaments can be ruled out if the samples are strongly non-ohmic, if the resistance increases with decreasing temperature, or if the resistance is above a certain minimum.

The last criterion is the easiest to check quickly with one room temperature resistance measurement and is fairly reliable. For if we assume the filament has the resistivity of bulk aluminum, a thickness of only 10 Å, and a length equal to the thickness of the oxide layer, say
50 Å, then its resistance is only about 300 ohms. If the filament is thicker than 10 Å, or if there is more than one, the resistance will be even smaller.

Furthermore, Fisher and Giaever point out that if conduction were due to such small filaments, it would create unreasonably high local current densities. Consequently, we can assume that a sample is free of filaments if its resistance is greater than about 300 ohms.

**Electrical Leads**

Electrical leads in the form of #37 bare copper wire were attached to the gold films at the ends of the aluminum electrodes using indium as a solder. If the surface of the gold film is clean, the indium will wet it quite readily without the use of any kind of flux. Indium also wets clean glass and usually diffuses through the metal films and sticks to the substrate, forming a strong, low resistance joint which is reliable at low temperatures.

Since the samples are in the form of a cross with provision for making electrical contact at both ends of each electrode, it was a simple matter to measure the resistance due to the aluminum film, the soldered joint, and the wire leads. This resistance was always about 2 ohms.

**Measuring Technique**

The electrical circuit used to measure the sample resistance is shown in Fig. 5. It consists of a Wheatstone bridge powered by an adjustable d.c. current supply. The voltage applied to the circuit is measured by means of a Leeds and Northrup potentiometer. Since this instrument does not measure potentials greater than 75 mV, a precision voltage divider of 990 ohms and 10 ohms is used. The applied voltage is
FIG. 5 Circuit used for measuring sample resistance.
then exactly 100 times the potentiometer reading.

The resistor of $10^7$ ohms in series with the bridge limits the bridge current to a maximum of $3 \times 10^{-7}$ amps. The output of the bridge is amplified by a Guildline type 9460 photocell galvanometer amplifier, and a Hewlett-Packard model 425A micro-ammeter is used as a null detector. Since the balance of a Wheatstone bridge is most sensitive when all four bridge resistors are equal, resistors $R_1$ and $R_2$ are adjustable by decades from 10 ohms to $10^5$ ohms by means of a low resistance double-pole, five-way switch. Thus, although $R_1$ is always equal to $R_2$, their value can be selected, after a rough balance is obtained, to be within half an order of magnitude of the sample resistance for values up to 0.5 megohms.

The resistance of the leads between the bridge and the sample is cancelled out by means of the arrangement shown, whereby the sample leads are placed in different arms of the bridge so that their resistances balance.

One of the problems encountered in using a d.c. measuring circuit is that thermal emf's affect the bridge balance. The effect of these stray voltages is overcome by use of the current reversal switch. The bridge is adjusted so that reversing the current produces a minimum deflection of the null detector.

However, this method of balancing the bridge also masks any asymmetries in the sample if they should exist; the resistance so determined will be the average of the forward and reverse resistances of the junction.

The sample current and voltage can be calculated from a knowledge of the input voltage as determined by the potentiometer, and the
values of the bridge and current limiting resistors.

The Cryostat

Samples were maintained at low temperatures in a conventional $^4$He cryostat by direct contact with a bath of liquid $^4$He or liquid $^2$O$_2$. The temperature of the bath could be reduced below the normal boiling point of the liquid by pumping on the vapour above the bath with a Stokes pump in the case of liquid $^4$He, or a Cenco Megavac pump in the case of liquid $^2$O$_2$. In this way, liquid $^4$He temperatures from 4.2°K to 1.18°K, and liquid $^2$O$_2$ temperatures from 90°K to 62°K could be attained.

When pumping pure oxygen with a rotary vacuum pump, there is always the danger that the oxygen and pump oil vapour will form an explosive mixture which can ignite under compression in the pump. Therefore, the ordinary pump oil was drained from the Cenco Megavac pump and replaced with pure tricresyl phosphate (TCP), which is non-explosive.

The vapour pressure above the bath was measured by a mercury or oil manometer and the temperature was determined from vapour pressure vs. temperature tables for liquid helium and oxygen.
RESULTS AND DISCUSSION

As was mentioned earlier, our method of sample preparation resulted in a discouragingly high proportion of failures. In all, 108 samples were made, of which perhaps half a dozen had resistances in the range of $10^{-3}$ to $10^6$ ohms. The high failure rate and non-uniformity of results from one evaporation to the next could have been caused by contamination of the vacuum system. This was possible because the evaporator was being used almost continuously by several different experimenters on various projects. However, these same circumstances made it very difficult to determine or eliminate the sources of contamination if they existed.

The samples exhibited an aging effect characterized by a monotonic but sometimes erratic increase in resistance, which would often double within two days after formation of the sample. This was not unexpected in view of the findings of previous workers. However, there were often large and very sudden changes in resistance which could not be easily explained. These and other effects will be described in more detail as we describe three experimental trials in chronological order.

First Trial

The first sample tested, hereafter referred to as sample 1, was made by oxidizing the aluminum for 45 min. in air at room temperature. The aluminum films were 650 Å and 850 Å thick, and the silicon oxide shields were 1800 Å thick. The junction area was 1 mm$^2$.

The sample was stored in air and three days later had a room temperature resistance of 74K ohms for a current of about $10^{-7}$ amps. Upon cooling to liquid nitrogen temperature ($77^o$K), the resistance increased
to 248K ohms. The sample was then cooled with liquid helium and two preliminary measurements were taken at 4.2°K and 2°K for a potential of 0.033 volts. At both temperatures, the resistance was 331K ohms. These measurements are represented as point A in Fig. 6. The voltage dependence of the resistance was not investigated at this time.

The next day, the sample was immersed in liquid oxygen and the voltage dependent characteristics were taken at 81°K and 65°K. The sample was kept in liquid oxygen for three days, after which the oxygen was allowed to evaporate and liquid helium was again transferred into the dewar vessel.

The value of the resistance at 4.2°K agreed fairly well with the value obtained four days previously at the same temperature and voltage (point A), but the 1.18°K measurements, taken about half an hour afterwards, were much higher than expected. According to Eq. (16), the current density, and hence the resistance at a given voltage should change by only one part in $10^5$ between 4.2°K and 1.18°K.

In view of the fact that no change in resistance was noted between these temperatures four days previously, it appears that this large increase was due to some anomalous change in the barrier parameters of the sample and is not related to the temperature dependence of the tunneling current.

Such large and unexplained increases in the resistance of samples were not uncommon; the resistance of a second sample being tested at the same time as sample 1 increased quite suddenly from 1.58 megohms to many megohms and could no longer be measured with our equipment.

Since the superconducting transition temperature for bulk alu-
FIG. 6 Experimental resistance vs. voltage characteristics for sample 1 at various temperatures.
minimum is $1.2^\circ K$, and for thin films is generally higher, the question arose as to whether the large change in resistance between $4.2^\circ K$ and $1.18^\circ K$ was caused by some superconducting phenomenon. This seemed unlikely because one would expect the tunneling resistance to decrease as the electrodes became superconducting. Furthermore, subsequent trials on other samples failed to show any variation in resistance between $4.2^\circ K$ and $1.18^\circ K$. The anomalous result therefore remains unexplained.

It is apparent from Fig. 6 however, that at all temperatures the voltage dependence of the resistance is parabolic, as predicted by Eq. (12). It can be seen that the low voltage data are on the threshold of the ohmic region. This makes the results of this trial particularly suited to analysis by Eq. (12) which is a low-voltage approximation. The detailed analysis of these data will be described in a later section.

**Second Trial**

Sample 2 was oxidized at room temperature in atmospheric air for 3 hours and had a room temperature resistance of 182K ohms for a current of $10^{-7}$ amps one day after formation. This value had increased to 330K ohms two days later. At $77^\circ K$, the resistance was 450K ohms.

The resistance vs. voltage characteristics were taken at $4.2^\circ K$, $2.4^\circ K$ and $1.18^\circ K$. The curves for these three temperatures coincided and were reproducible over the voltage range, as shown in Fig. 7. These results strengthened our suspicion that the increase in resistance of sample 1 between $4.2^\circ K$ and $1.18^\circ K$ was not a temperature effect. They also indicated that the resistance was not pressure dependent, since the pressure varied from 760 mm Hg at $4.2^\circ K$ to 0.5mm Hg at $1.18^\circ K$. Unfortunately sample 2 broke down before it could be tested at higher
Sample 2

Fig. 7 Experimental resistance vs. voltage characteristics for sample 2 at 4.2°K, 2.4°K and 1.18°K.
temperatures.

Third Trial

In preparing samples for this trial, two new techniques were used. The substrate was cleaned by ultra-sonic vibration in distilled water for 5 min., and the sample was oxidized for 2 hrs. in dry air. Of the nine samples, three were short circuited, three had resistances too high to measure, and three were in the range of 18K to 39K ohms.

The resistance of sample 3 was 21K ohms one day after formation, increased to 160K ohms at 77°K and remained fairly steady at this temperature for 12 hrs. before increasing quite suddenly to 6.35 megohms. This large jump in resistance was almost certainly due to the operation of a Tesla spark coil near the apparatus. It was observed on several other occasions that samples were quite sensitive to such external electric fields.

Because our measuring technique was to pass certain pre-selected values of current through the sample, the voltage developed across the junction was determined by its resistance. Consequently, the voltage range in which sample 3 was investigated was roughly an order of magnitude larger than for samples 1 and 2, and was well out of the ohmic region.

Characteristic curves were taken at 1.18°, 2.2°, 4.2°, 62°, 70°, 80° and 90°K. The voltage dependence at a given temperature was usually reproducible within the experimental error.

Fig. 8 shows an effect which was observed at 1.18°, 62°, 80° and 90°K: as the sample current is increased beyond a certain value, the resistance drops so sharply that the resultant voltage actually decreases. In all four cases, this decrease occurred as the sample voltage reached
FIG. 8 Experimental resistance vs. voltage characteristics for sample 3 at 1.18°K.
0.4 volts.

This effect is similar to what one would expect if the Fermi level of the positively biased electrode were depressed below the level of the conduction band of the negative electrode. (See Fig. 3(c) of Simmons, 1963). This occurs when $eV > (\varphi + \eta)$, typically for an applied potential of 1 or 2 volts. The observed value of 0.4 volts would seem to indicate a very low value for $\varphi$.

The temperature dependence of resistance for sample 3 was inconclusive. As in the case of sample 2, no appreciable change in characteristics was observed in the liquid helium temperature range. Although there was a variation with temperature in the liquid oxygen temperature range, the characteristics for $1.18^\circ K$ were almost coincident with those for $90^\circ K$. Since the high temperature data were taken about 24 hours after the low, it appears that the temperature induced decrease in resistance was offset by an increase due to aging.

**Analysis of Data**

A quantitative analysis was carried out for the data taken from sample 1. The parabolic voltage dependence of the resistance, which is indicated in Fig. 6, is shown more strikingly in Fig. 9, in which the resistance is plotted against the square of the voltage. The data points for each temperature fall along a straight line.

Since, for reasons discussed above, the characteristic curve taken at $1.18^\circ K$ was rejected as anomalous, the $4.2^\circ K$ data were taken as an approximation to the behaviour of the sample at $0^\circ K$ and were analyzed according to the low voltage, low temperature approximation, Eq. (12), repeated here for convenience:
FIG. 9  Resistance vs. square of voltage at three temperatures, together with theoretical high temperature characteristics calculated from behaviour at 4.2°K.
Setting $V = 0$ in this equation gives an expression for $R(0, 0)$ to be equated with the extrapolated zero voltage value of resistance from the 4.2°K data in Fig. 9. If we assume $\varphi = 2eV$, which is the average barrier height for a symmetric aluminum junction as determined by Morris and Pollack (1964), the only unknown in the theoretical expression for $R(0, 0)$ is $A$, which is a function of the barrier thickness $s$:

$$A = \frac{4\pi s}{\hbar} (2m)^{1/2}$$

Thus, by equating the theoretical expression for $R(0, 0)$ with the extrapolated value, we can obtain a value for $s$ which will be quite precise because of the exponential dependence of $R(0, 0)$ on $A$.

This calculation yields

$$s = 20.5 \text{ Å}.$$  

Eq. (12) also shows that the graph of $R(V, 0)$ vs. $V^2$ is a straight line whose slope is given by

$$\frac{\hbar^2 s \exp \left( \frac{A \varphi_s^{1/2}}{2m \varphi_s} \right)}{2} \left( \frac{A^2 e^2}{96 \varphi_s} \right)$$

Equating this expression for the slope with the value obtained graphically, (again taking $\varphi = 2eV$), we can again solve for $s$. This yields

$$s = 22.2 \text{ Å}.$$  

These two approximations for $s$ are in fairly good agreement and are reasonable values, according to the corrected results of Hunter and Fowle (1956), for the thickness of an oxide film grown in air at room temperature.

The results from sample 1 are also quite consistent in that the lines in Fig. 9 all have the same slope, indicating that the voltage
dependence is the same at all temperatures, as predicted by theory.

The 4.2°K line was used to predict the two theoretical lines in Fig. 9 for the behaviour at 65°K and 81°K. Writing Eq. (12) as

\[
R(V, 0) = R(0, 0) \left\{ 1 - \frac{A^2 e^2}{96 \varphi_o} V^2 \right\}
\]

and the slope of the line as \(dR/d(V^2)\), we obtain

\[
\frac{A^2}{\varphi_o} = - \frac{dR}{d(V^2)} \times \frac{96}{e^2} \times \frac{1}{R(0, 0)}
\]

Using the graphically obtained values for \(dR/d(V^2)\) and \(R(0, 0)\) we derive a value for \(A^2/\varphi_o\) and hence a value for \(s^2/\varphi_o\). Since for small voltages, \(\Delta s \approx s\) and \(\overline{\varphi} \approx \varphi_o\), this value of \(s^2/\varphi_o\) can be used in the temperature dependent Eq. (16) for various values of \(V\) to obtain the theoretical lines for 65°K and 81°K as shown in Fig. 9.

The theoretical change in resistance between 4.2°K and 65°K was 4.6% as compared with an observed change of 8.6%. Between 4.2°K and 81°K, the theoretical and observed changes were 6.6% and 12% respectively. The variation in tunneling resistance with temperature was thus roughly twice as great as predicted. This was also true for the variation in resistance between 65°K and 81°K.
CONCLUSIONS

The purpose of this experiment was to test quantitatively the theory of the temperature dependence of the tunneling resistance in thin insulating films as derived by Simmons (1963, 1964). One of the problems encountered was the high failure rate in sample production. This did not appear to be due to dust on the substrates or moisture in the air used for oxidation. It is known that cleanliness of the vacuum system is a very critical factor in the production of metallic thin films with uniform electrical properties, and contamination may have been responsible for the poor results. Clearly, in order to ensure success, the conditions in the vacuum system during evaporation must be very strictly controlled.

The samples were found to be easily changed or damaged by such influences as external electric fields, and besides the general increase in resistance due to aging, often showed large and sudden jumps in resistance which could not be easily explained. These effects may have been due to unstable ionic configurations at the barrier interfaces (see Morris and Pollack, 1964). In order to obtain reproducible and reliable temperature dependence data, it is essential that the samples be stable and the aging effect as small as possible. This might be achieved by oxidizing the samples for extended periods of time at temperatures of several hundred degrees Centigrade.

Although d.c. techniques are the most suitable for the investigation of tunneling characteristics, provision should be made for measuring the resistance of the junction in each direction in order to obtain some idea of the barrier asymmetry even for all-aluminum samples.

The voltage dependence of the tunneling resistance was found to
agree quite well with theory and was parabolic for low voltages as predicted. Changes in resistance of the sample due to aging or anomalous jumps did not destroy the voltage dependence, although they did make it difficult to investigate the temperature dependence, since the temperature range could not be scanned as quickly and easily as the voltage range.

Analysis of the temperature dependence of resistance for sample 1 was carried out by taking the 4.2\(\text{K}\) curve as an approximation of the behaviour at 0\(\text{K}\). By graphical analysis, an experimental value for \(s^2/\rho_0\) was derived and used in the equation for the temperature dependence to predict lines for 65\(\text{K}\) and 81\(\text{K}\). The theoretical change in resistance between 4.2\(\text{K}\) and 65\(\text{K}\) was 4.6\% as compared with an observed change of 8.6\%. Between 4.2\(\text{K}\) and 81\(\text{K}\) the observed temperature dependence was also about twice as great as expected - 12\% as compared with a predicted value of 6.6\%. 


