Tunneling Through Thin Insulating Layers

J. C. Fisher, and I. Giaever

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and reflectivity data together with the given estimate of the energy difference between the top of the filled band and the vacuum level, they estimated the width of the forbidden band to be between 8 and 9 ev, with the best estimate being 8.7 ev. This estimate also implies that the electron affinity of magnesium oxide is less than 1 ev.

The analysis of the thermionic emission data in Sec. 3 strongly suggests the existence of a donor level 3.4 ev below the vacuum level. It is probable that this donor level is also the one from which the electrons originate in the enhanced photoelectric emission at 3.1 ev. It is important to point out that the 3.1-ev energy was purposely chosen in a region of low photoelectric yield in order that the enhancement effects would be more clearly evident. However, even though the photoelectric yield in the region of 3.4 ev and above was much more pronounced, it was too unstable to observe any definite structure in the curves that would aid in revealing the exact location of the emission center. This lack of stability was probably due to the enhancement effect to be discussed shortly.

As was pointed out in the foregoing, the electron affinity of magnesium oxide is probably less than 1 ev. This indicates that the donor level discussed in the last paragraph is located slightly more than 2.4 ev below

the bottom of the conduction band. It is suggested that these donors should possibly be identified with the color centers which are produced by heating magnesium oxide in magnesium vapor and which are responsible for the optical absorption band at about 2.35 ev first reported by Weber.[14] The implication is that an energy of about 0.1 ev to 0.2 ev would be required for the thermal release of electrons from the excited states of these centers. In the data obtained by both Peria[8] and Day[15] there are suggestions of a peak in their photoconductivity data in the region around 2.35 ev, however, the evidence is far from being conclusive.

Peria[8] has observed a peak in the photoconductivity of magnesium oxide centered at 5 ev. By means of a special experiment to distinguish the sign of the charge carrier, he was able to identify this peak as being due to the release of electrons to the conduction band. The enhancement data presented in this paper are positive confirmation of this identification. The interpretation here is that electrons released to the conduction band from the 5-ev level are trapped by the level responsible for the photoelectric emission at 3.1 ev. This leads to the enhancement of this emission.

[14] H. Weber, Z. Physik **130**, 392 (1951).
[15] H. R. Day, Phys. Rev. **91**, 822 (1953).

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J. C. FISHER AND I. GIAEVER
General Electric Research Laboratory, Schenectady, New York
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The resistance of thin aluminum-oxide films has been measured as a function of the voltage across the film and of the film thickness as calculated from its capacitance. All films showed ohmic behavior at low voltages, and exponential rise of current at higher voltages, in qualitative agreement with R. Holm's theoretical calculations for tunneling through thin vacuum layers. However, the resistance was several orders of magnitude lower than either the bulk value for Al_2O_3 or the calculated value for tunneling through vacuum. By making use of an effective mass in the oxide equal to about $\frac{1}{8}$ of the electron mass, the calculated values for tunneling can be brought into line with the experimental results.

INTRODUCTION

ELECTRONS can pass through thin layers of insulating materials by the quantum mechanical process called tunneling. Insulating layers thicker than a few tens of angstroms are nearly impenetrable, as the probability of getting through falls off exponentially with thickness. For this reason, very thin films are required, and the tunneling process provides a means for studying their formation and properties.

SAMPLE PREPARATION

Thin films of aluminum oxide sandwiched between aluminum metal films were prepared by depositing

aluminum films from the vapor, oxidizing their surfaces, then depositing another aluminum film over the oxide. In more detail, 99.999% pure aluminum was evaporated through a mask onto carefully cleaned microscope glass slides, forming a film 3/64 in. wide and several hundred A thick along each slide. Different aluminum films then were oxidized in different ways: some were oxidized in air, some in oxygen atmospheres at various temperatures, and some in distilled water, forming an oxide layer several tens of angstroms thick. After the oxide layer had formed, five aluminum strips from 1/64 in.–5/64 in. wide were deposited across it, sandwiching it between metallic films as shown in Fig. 1. The resistance

TABLE I. Dimensions of the oxide films on a sample oxidized for 11 days in air at room temperature.
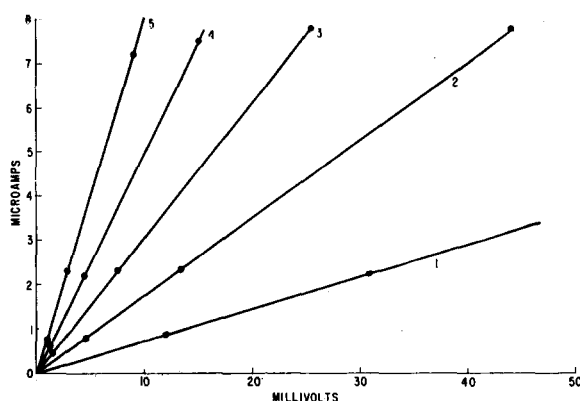
Film No.	Relative area [units of $4.72(10)^{-3}$ cm^2]	Thickness in A (measured by capacitance)
5	5	48
4	4	45
3	3	48
2	2	47
1	1	48

of each sandwiched oxide layer could be measured by making the proper electrical connections to the aluminum films on either side of it. When necessary, contact resistance was minimized by using a four-terminal method, as shown in Fig. 1.
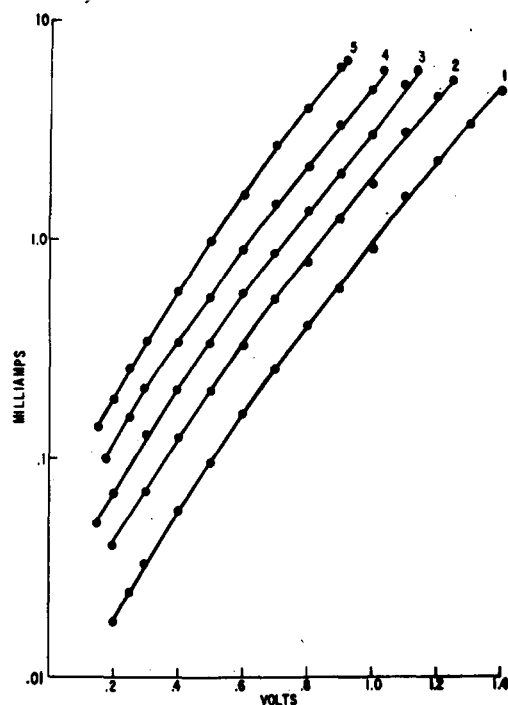
EXPERIMENTAL RESULTS

For small voltages the current through the oxide film is proportional to the voltage, demonstrating that the low-voltage resistance is ohmic, as shown in Fig. 2(a) for the five oxide films described in Table I. The five curves correspond to the five sandwiches on a single slide, and the numbers on each curve correspond to the relative areas of the oxide films. For greater voltages, the current increases exponentially with voltage, as shown in Fig. 2(b). In both figures the currents are roughly proportional to the film areas. All films showed the characteristics of ohmic behavior at low voltages, an exponential rise of current at higher voltages, and an approximate proportionality of current to film area.

Figures 3(a), (b), and (c) show that the resistance of the oxide films tends to increase exponentially with their thickness, the data in each figure corresponding
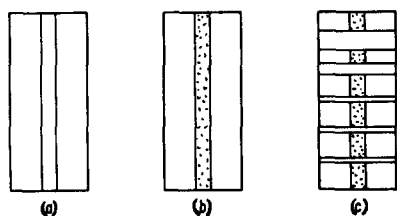

FIG. 1. Sample preparation. (a) An aluminum film is deposited on a glass slide; (b) a superficial oxide film is formed; (c) aluminum films are deposited across the oxide; (d) current and voltage connections are made.
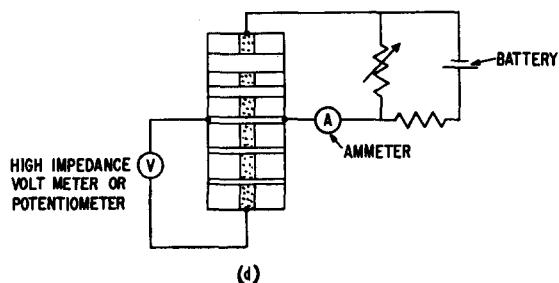

FIG. 2. (a) At low voltages, the current through thin oxide films is proportional to voltage and to film area. Curves shown are for the five films described in Table I, with areas in the proportions 5:4:3:2:1 as indicated. (b) At higher voltages, the current increases exponentially with voltage.

to a particular oxidation method. Though the scattering is rather violent, there still remains a significant trend of exponentially increasing resistance with thickness, the slope of the curves depending somewhat upon the oxidation method. The thickness values were calculated from low-frequency capacitance measurements by using a dielectric constant of $\epsilon = 8$.

Figure 4 shows that some films tend to rectify, one direction being favored at room temperature and the other at liquid nitrogen temperature, and that the current decreases slightly with decreasing temperature. The rectifying effect was not apparent in films oxidized at room temperature. By carefully annealing the

sandwiches that do rectify, the change in rectification with temperature could be made to disappear, only the low-temperature direction of rectification remaining. It should be remarked that the current for voltages

above about 0.5 v requires 5–10 sec to reach a steady-state value at room temperature. At constant voltage the current increases when the oxidized electrode is negative, and decreases when the oxidized electrode is positive.

Finally, a photovoltaic effect is easily observable for the films oxidized in air at 400°C. The maximum open circuit voltage was 35 mv, and the last-deposited electrode was always positive, no matter from which side the light came.

DISCUSSION

By referring the observed characteristics to the tunneling of electrons through the aluminum-oxide layer, most of the observed phenomena can be easily explained. It first will be demonstrated that electronic rather than ionic conduction is involved, and that the oxide film is continuous (although inhomogeneous, one face differing from the other). Then the experimental results will be interpreted in terms of electron tunneling and the structure of the oxide film.

It seems certain that the major part of the current is the result of electronic rather than ionic processes. The weak temperature effect indicates this, as does the maximum current density of approximately 1 amp/cm^2. If this current were the result of the electrolysis of water dissolved in the oxide, approximately 10^{-4} g/sec of water would be required, far in excess of the maximum possible amount.
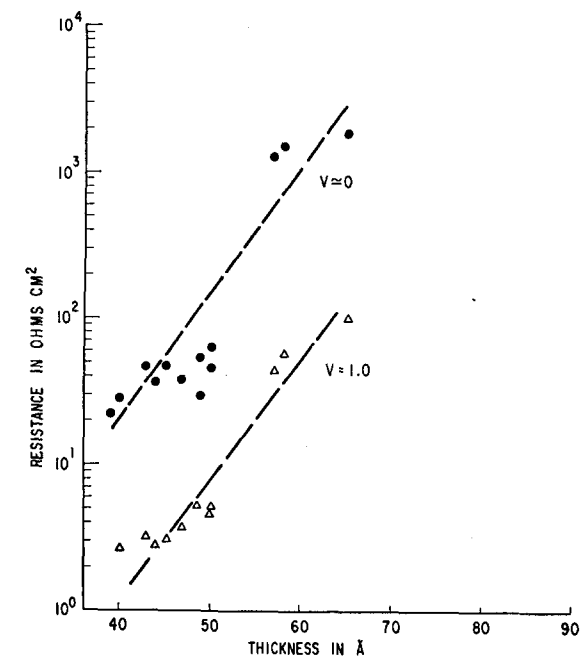
(a)
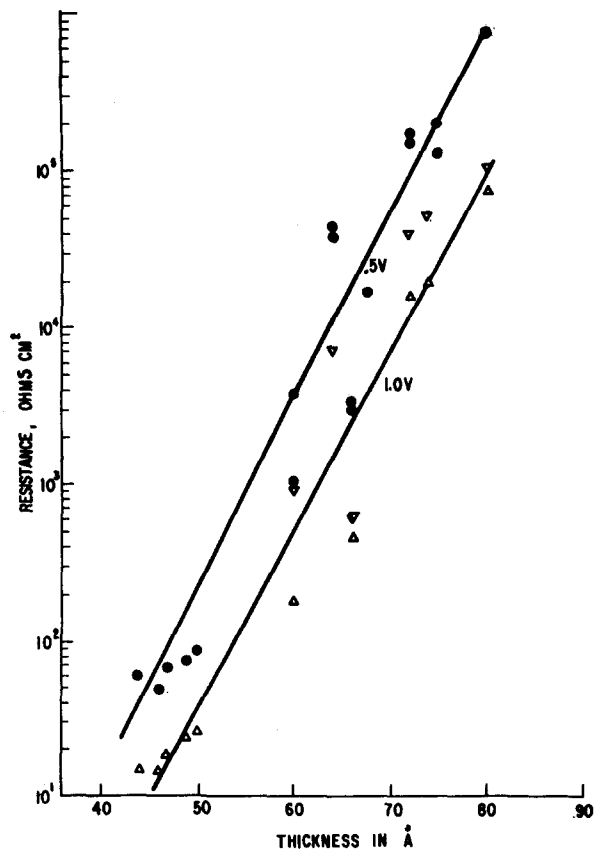
(b)
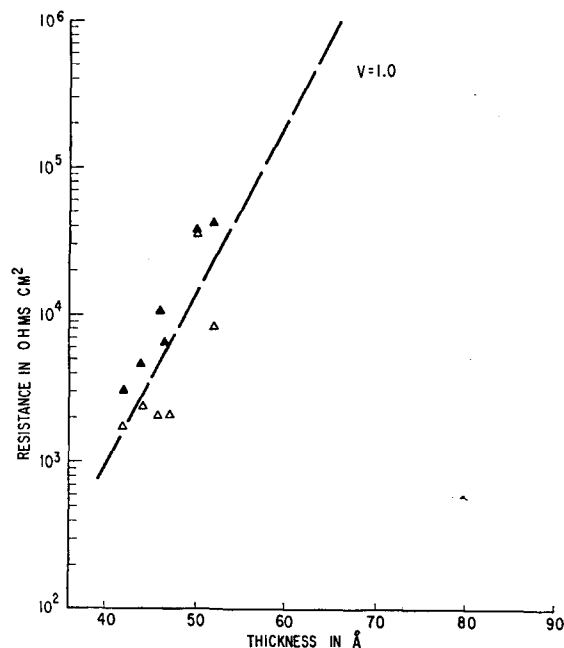
(c)

FIG. 3. (a) The resistance of oxide films tends to increase exponentially with thickness (films formed in air at room temperature). (b) Exponential increase of resistance with thickness (films formed in air at room temperature and then at 400°C). (c) Exponential increase of resistance with thickness (films formed in air at 400°C).

One difficulty with thin films is that small metal bridges often are responsible for conduction across them. This form of conduction is ruled out qualitatively for our films by the presence of nonlinear current-voltage characteristics, rectifying properties, the slight increase of resistance with decreasing temperature, the observed sharp breakdown at high voltages, and the inverse proportionality of resistance to film area. Furthermore, quantitatively, on assuming a resistivity of 10^{-6} ohm-cm for metal bridges (if in existence) and a film thickness of 100 A, the total area of the bridges would be of the order of 10^{-12} cm^2, giving an unreasonably high local current density.

Now we shall summarize Holm's analysis[1] of tunneling through a thin vacuum layer between similar plane metal surfaces, as sketched in Fig. 5. Holm finds the number of electrons N tunneling through the vacuum to be

$$N = (4\pi m/h^3) \int_0^\infty D(E_x) Y(E_x) dE_x, \qquad (1)$$

where

$$Y(E_x) = \int_{E_x}^\infty [f(E) - f(E+eV)] dE \qquad (2)$$

and

$$D(E_x) = \exp\left(-(4\pi/h) \int_0^s \{2m[\phi(x) - E_x]\}^{\frac{1}{2}} dx\right), \qquad (3)$$

and where $h =$ Planck's Constant; e and m are the electron charge and mass; V is the applied potential; E and E_x are the total electron energy $p^2/2m$ and "energy in the x direction" $p_x^2/2m$; $f(E)$ is the Fermi distri-

FIG. 5. Potential energy diagram for analysis of tunneling through a thin vacuum layer.

bution function of electron energy; $\phi(x)$ is the electronic work function; and s is the film thickness.

Assuming an absolute temperature $T=0$, Holm carried out the integration by using reasonable assumptions. At low voltages he found the flux of electrons to be

$$N = (eV/h^2s)(2m\phi)^{\frac{1}{2}} \exp[-(4\pi s/h)(2m\phi)^{\frac{1}{2}}] \qquad (4)$$

electrons/cm^2 sec. At high voltages $(eV \geqslant \phi + \eta$ in Fig. 5) he found that

$$N = (e^2 V^2/8\pi h\phi s^2) \exp[-(8\pi s/3heV)(2m)^{\frac{1}{2}}\phi^{\frac{3}{2}}]. \qquad (5)$$

We note here particularly that the first equation is linear in the applied field, while the second equation is the well-known Fowler-Nordheim equation.

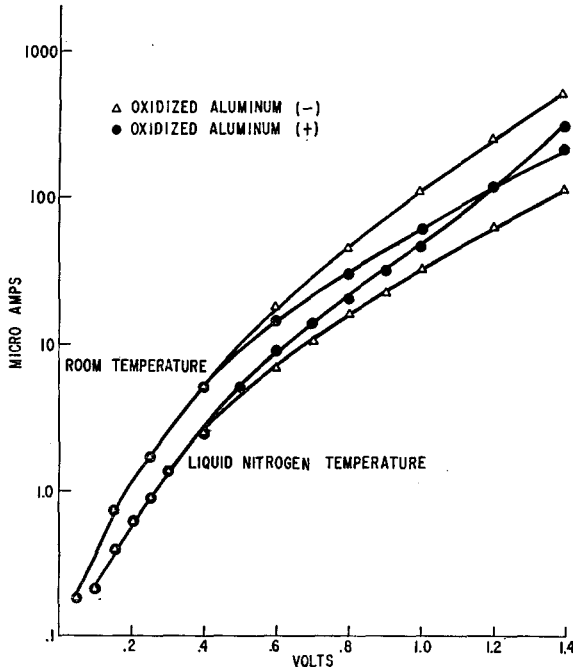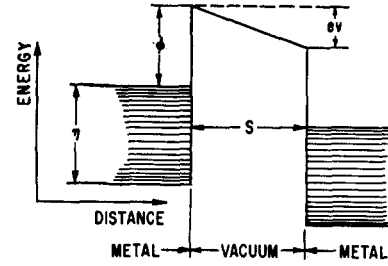For intermediate fields $(0 < eV < \phi + \eta)$, the electron

FIG. 4. Rectification, observed with films formed at elevated temperatures, reverses sign with temperature.
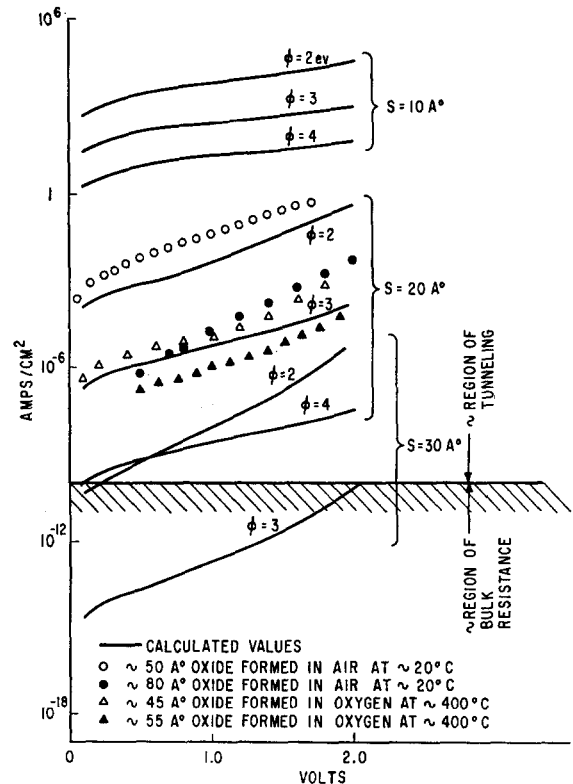
[1] R. Holm, J. Appl. Phys. 22, 569 (1951).

FIG. 6. Comparison of measured current-voltage relationships with those calculated for vacuum films. Approximate agreement is found between results measured for 45–80 A oxide films and calculated for 20 A vacuum films.
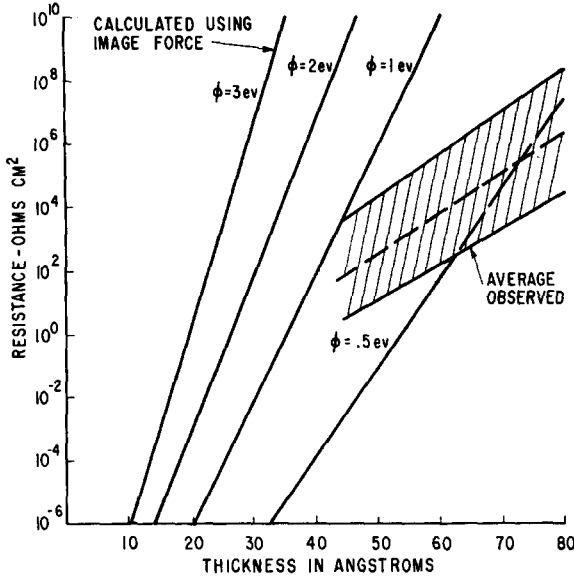
FIG. 7. Inclusion of image forces does not bring the calculation into line with experiment.

flux is

$$N = N_I + N_{II} \tag{6a}$$

$$\left.\begin{array}{l} N_I = [(2m)^{\frac{1}{2}}/h^2 s]eV(\phi+eV/2)^{\frac{1}{2}} \\ \quad \times \exp[-A(\phi+eV/2)^{\frac{1}{2}}] + [3(2m)^{\frac{1}{2}}/h^2 sA] \\ \quad \times \{(\phi-eV/2)\exp[-A(\phi-eV/2)^{\frac{1}{2}}] \\ \quad\quad -(\phi+eV/2)\exp[-A(\phi+eV/2)]\} \\ A = (4\pi s/h)(2m)^{\frac{1}{2}}, \end{array}\right\} \tag{6b}$$

$$N_{II} \approx \text{Fowler–Nordheim equation [Eq. (5)]}, \tag{6c}$$

and where the two parts of the equations are to be so understood that

$$\text{if } eV < \phi \begin{cases} N_I \text{ as written} \\ \\ N_{II} = 0 \end{cases}$$

$$\text{if } eV > \phi \begin{cases} \text{substitute } \phi \text{ for } eV \text{ in } N_I \\ \\ N_{II} \text{ as written.} \end{cases} \tag{6d}$$

For voltages less than about 0.25 v, the current-voltage characteristic calculated from this equation is linear, agreeing well with the observed values. We also see immediately that the Fowler-Nordheim domain can never be reached, as $\phi+\eta$ is approximately 14 ev for aluminum, and the films break down at about 2–3 v.

Figure 6 shows some voltage-current characteristics as calculated from Eq. (6). Some of the experimental values are also shown. The calculated and experimental slopes agree fairly well, but the current density observed for an oxide film of given thickness corresponds to that calculated for a vacuum layer only about $\frac{1}{3}$ as thick.

The image force has been neglected in these calculations. If it is included, the resistance is given by

$$\sigma = (h^3/8\pi m e^2)A^2\{1+A[\phi-(e^2/s)\ln 2]^{\frac{1}{2}}\}^{-1}$$
$$\times \exp\{A[\phi-(e^2/s)\ln 2]^{\frac{1}{2}}\} \tag{7}$$
$$A = 1.15(\pi^2 s/h)(2m)^{\frac{1}{2}}(1-e^2/2s\phi),$$

as reported by Holm and Kirchstein[2] in the limit of low voltage. These results are pictured in Fig. 7, where the calculated values still are very much out of line with the observed values.

It remains to be shown that neither surface roughness nor a very small work function for electrons escaping into Al_2O_3 can account for the discrepancy. Surface roughness would increase the electric field at some portions of the surface, thereby leading to unexpectedly high tunnel currents in the region where current increases exponentially with field. However, the discrepancies observed in these experiments are several orders of magnitude greater at low fields where current is directly proportional to field, ruling out surface roughness as the cause. A small work function for electrons escaping from aluminum to aluminum oxide would provide another possible explanation for the high currents that are observed. If the Fermi level of the aluminum lined up approximately with the middle of the forbidden band, the work function should be of the order of 5 ev; but because of impurities we might expect the Fermi level to be displaced relative to the center of the forbidden band, and from the curves we see that a work function of about 0.1 ev might explain our results. However, for such a low work function, we should have a Fowler-Nordheim relationship between current and voltage because, as soon as the applied voltage is greater than the work function, the Fowler-Nordheim part of the current is dominant. Since no Fowler-Nordheim behavior is observed, this explanation fails.

The results can be explained by introducing an effective thickness in comparing the oxide layer to vacuum, the oxide layer being equivalent to a vacuum layer $\frac{1}{3}$ as thick. Physically this can be understood by picturing the oxide as containing potential wells caused by the atoms in the lattice.[3] However, it is more common with semiconductors and insulators to introduce an effective mass for the electron. This can be understood qualitatively by examining the Kronig-Penney model of a crystal. With a vacuum layer between
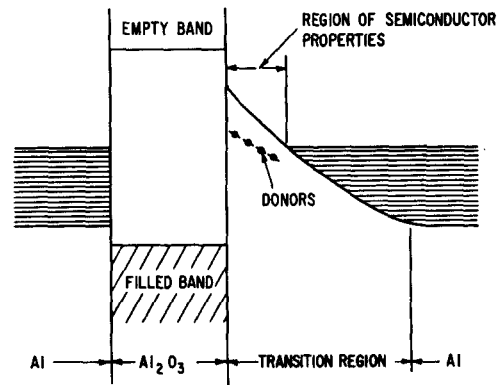

FIG. 8. Schematic model of oxide layer, with oxidized aluminum on right.

[2] R. Holm and B. Kirschstein, Z. tech. Physik 16, 488 (1935).
[3] R. Gomer, Australian J. Phys. 13, 391 (1960).

WILSON'S MODEL

SCHOTTKY'S MODEL

MODEL APPLICABLE TO THIS CASE
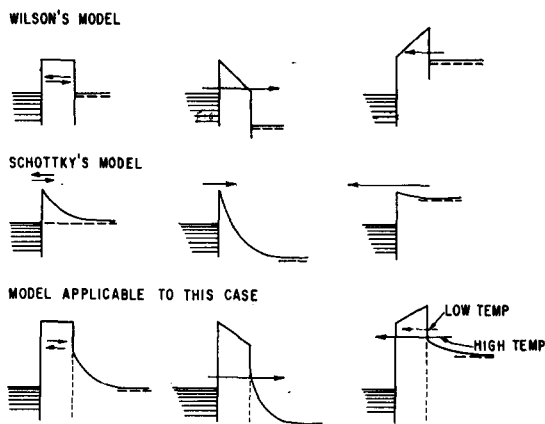
LOW TEMP
HIGH TEMP

FIG. 9. Rectification at a metal semiconductor contact according to the theories of Wilson (tunneling through a barrier gives easy electron motion toward semiconductor) and Schottky (flow over a barrier gives easy electron motion toward metal). In combination, the two theories explain the reversal of easy direction with temperature.

the two electrodes, the only permissible solution of the wave equation is an exponentially decaying plane wave. Introduction of a periodic potential between the two electrodes permits an exponentially decaying periodic solution.

Equation (3) shows that the exponent, which almost completely determines the current, depends linearly on the film thickness and upon the square root of the electron mass. Thus, Figs. 6 and 7 show that by assuming an effective mass of approximately $\frac{1}{9}$ of the electron mass, the measured values are brought into approximate agreement with the calculated ones.

The major quantitative features of oxide-film conduction, including ohmic conduction at low voltages, exponential rise of current at higher voltages, exponential increase of resistance with film thickness, and insensitivity to temperature, can be understood in terms of tunneling through a homogeneous oxide, using an effective thickness factor of $\frac{1}{3}$ or, equivalently, an effective electron mass factor of $\frac{1}{9}$. Minor variations from ideal behavior, including the photovoltaic effect, rectification, and the change in direction of rectification with temperature, can be understood in terms of a nonuniform oxide film. These effects suggest that the system can be idealized as follows: The oxidized aluminum does not abruptly go over to aluminum oxide; rather, there exists a thin layer with semiconductor properties between the aluminum and the oxide that formed upon it. On the top side of the oxide, however, there is a much sharper interface between the oxide and the aluminum deposited upon it. Although the transition from semiconductor to purer oxide is continuous, it can be idealized as sketched in Fig. 8. The difference between the two interfaces seems to be most pronounced when the sample is oxidized at an elevated temperature.

We may associate the observed photovoltaic effect with the semiconductor layer. Impurities, or possibly an excess of aluminum, form donor levels in the band gap. Electrons are knocked out of these levels into the conduction band, showing that the energy difference must be less than 2 ev as the process goes on with visible light. In contrast, the total band gap of aluminum oxide is at least 6 ev, and is probably closer to 10 ev. This excess of electrons makes the oxidized aluminum always negative, which is what is observed.

The change in the direction of rectification can be explained by this model. Figure 9 pictures Wilson's and Schottky's models for rectification at a metal semiconductor contract. Wilson[4] postulated that electrons could only pass the junction by tunneling through the barrier. The number of available electrons is the limiting factor in this case, and the current flows most easily when the metal is negative. Schottky[5] postulated that the electrons had to flow over the potential barrier. In this case the height of the potential hill is the limiting factor, and the current flows most easily when the metal is positive. The aluminum oxide film sketched in Fig. 8 can be pictured as Wilson and Schottky rectifiers in series. At low temperatures, only the Wilson effect can operate, so that the current flows best with the metal negative. With increasing temperature, the Schottky effect comes into the picture, and might eventually overpower the Wilson effect. This is what is observed, the change occurring somewhere between liquid nitrogen and room temperature.

CONCLUSION

It seems certain that the observed conductivity in the aluminum oxide film results from the tunneling of electrons through the film. The current-voltage characteristic, the thickness dependence, and the weak temperature dependence all point to this. Furthermore, the change in rectification and the photoelectric effect can be explained by this model. The absolute magnitude of the current for a given voltage is much greater than the value predicted for tunneling through vacuum, but one can bring the calculated values into line with the experimental results by introducing an effective mass for the motion of an electron in the oxide film. As the effective mass is closely dependent upon the structure of the film, it varies somewhat from film to film. An effective mass of about $\frac{1}{9}$ of the electron mass is a good average value.

[4] A. H. Wilson, Proc. Roy. Soc. (London) A136, 487 (1932).
[5] W. Schottky, Z. Physik 118, 539 (1942).