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Technical Report **R 575**

SCHOTTKY EMISSION IN THIN-FILM DIODES

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SCHOTTKY EMISSION IN THIN-FILM DIODES

Technical Report R-575

Z-R011-01-01-113

by

R. D. Hitchcock

ABSTRACT

Temperature-dependent current-voltage (I-V) characteristics have been observed in a new type of thin-film diode, consisting of Al, Al_2O_3, Mn, Mn_xO_y, and Pb. Plots of $\ln I - V^{1/2}$, $d \ln I/dV^{1/2} - 1/T$, and $\ln(I/T^2) - 1/T$ (where T is temperature in $^{\circ}$K) can be fitted by straight lines which show that Schottky emission is the dominant current-flow mechanism over the temperature range of 190°K to about 350°K. Barrier thicknesses were determined by high-frequency capacitance measurements and found to lie between 100 Å and 250 Å. Relative work functions, between the aluminum and lead films, were found to lie between 0.25 and 0.50 ev. One of the diodes was tested at 60 Hertz for 124 hours without causing a significant change in the shape of the I-V characteristic. It is concluded that nominal refinement of the fabrication technique will lead to a varistor thin-film diode which is inexpensive and which has long-term stability.

EVALUATION SHEET

Technical Report R-575

Title SCHOTTKY EMISSION IN THIN-FILM DIODES

Author R. D. Hitchcock

Task No. Z-R011-01-01-113

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INTRODUCTION

This report describes experimental work at the Naval Civil Engineering Laboratory (NCEL) on five-layer thin-film structures with Schottky-emission current-voltage (I-V) characteristics. The thin-film devices were constructed by vacuum deposition and are of the metal-insulator-metal type. The insulator structure cannot be precisely described at this time, but for discussion purposes it is presumed to be a triple-layer system which consists of a metal film sandwiched between two metal-oxide films. Hence, the complete diode system will be designated by the five-symbol array M/I/M/I/M. The work was done for the purpose of developing an improved thin-film voltage regulator of the metal-barrier-metal (M/B/M) type. The M/B/M diode that has a pure oxide barrier has neither long-term stability nor high current-carrying capabilities, but it does have a symmetrical I-V characteristic. This advantageous characteristic requires two back-to-back, conventional metal-oxide-semiconductor diodes. Hence, the development of an M/B/M diode that does have long-term stability and high current-carrying capabilities will result in increased component density for integrated circuitry at a reduced overall cost.

The attempt to develop an improved thin-film voltage-regulator diode was inspired by the discovery in fiscal year 65 of a new type of thin-film system with a symmetrical voltage-regulator I-V characteristic[1] which could be approximated by V^x, where $x \sim 2$. This thin-film system consisted of Al, Al_2O_3, Mn, Mn_xO_y, and Pb. No attempts have been made as yet to determine the manganese-oxide structure by electron or X-ray diffraction patterns, hence the x and y subscripts for the stoichiometry. Although other simpler thin-film systems, particularly the M/I/M structure with Al_2O_3 as insulator, had symmetrical voltage-regulator I-V characteristics, only the five-film system, $Al/Al_2O_3/Mn/Mn_xO_y/Pb$, could be driven at 60 Hertz and at a useful current density for a relatively long time without causing the I-V pattern to become ohmic or a dead short.

During the fiscal year 65 investigation[1] many of the $Al/Al_2O_3/Pb$ diodes exhibited symmetrical Fowler-Nordheim I-V patterns under DC excitation. These patterns could be reproduced only a limited number of times, but it was believed that development of the five-film system with Mn_xO_y would lead to a symmetrical Zener-type thin-film diode with long-term stability under continuous AC operation. As yet, stable voltage-regulator behavior with the pronounced Fowler-Nordheim or Zener knee has not been achieved. However, vacuum-deposition techniques have been refined to the point where a relatively high percentage of the five-layer diodes have highly stable I-V characteristics which are temperature dependent and consistent with the theory of Schottky emission into vacuum.[2] In most cases, the I-V patterns are nearly symmetrical at 60 Hertz, and at room temperature exhibit a higher degree of bending than a space-charge-limited characteristic ($I = \text{const} \times V^2$) for a comparable current and voltage range. Figure 1 compares the I-V characteristic of one of the Schottky thin-film diodes with the theoretical I-V patterns for space-charge-limited and Fowler-Nordheim behavior.

DIODE FABRICATION

Five-layer sandwiches were constructed by first evaporating a 1-mm-wide aluminum film strip onto a glass substrate cut from a microscope slide. The aluminum was oxidized for a few minutes in air at room temperature and atmospheric pressure; then, at a pressure around 8×10^{-5} torr, a 3-mm-wide strip of manganese was evaporated over and perpendicular to the aluminum strip.

The manganese deposition was carried out after a preheat period of 1 or 2 minutes, during which no metal deposition was detectable by the current-measuring system connected to the two indium patches, one on each side of the substrate. The upper limit of measurement was 3×10^6 Ω/square. Manganese was deposited by evaporating manganese powder from a small graphite crucible that was cut from a 1/4-inch spectrographic rod and was resistance-heated by a tantalum basket. During the preheat and deposition periods the substrate was positioned a few centimeters above the source; temperature, measured by a thermocouple attached to the opposite side of the substrate, did not exceed 350°K.

Immediately after deposition of the manganese film, the manganese slowly oxidized in the residual air of the bell jar, which was at a pressure between 2×10^{-5} and 8×10^{-5} torr. This oxidation caused the film resistance to increase from a minimum of about 3×10^4 Ω/square to 7×10^4 Ω/square. The pressure in the bell jar was raised to 760 torr, which caused further oxidation and an increase in film resistance to 3×10^6 Ω/square or greater This two-step oxidation process appeared to be necessary for stable I-V curves having a shape indicative of Schottky emission. Too rapid oxidation of the manganese (by venting immediately after deposition) almost always produced a thin-film diode with either ohmic I-V behavior or non-Schottky traces which quickly changed to ohmic. Manganese film strip resistance as a function of time is given in Figure 2; the length of the strip, between the inner edges of the indium patches, is approx.. ıately 10 mm.

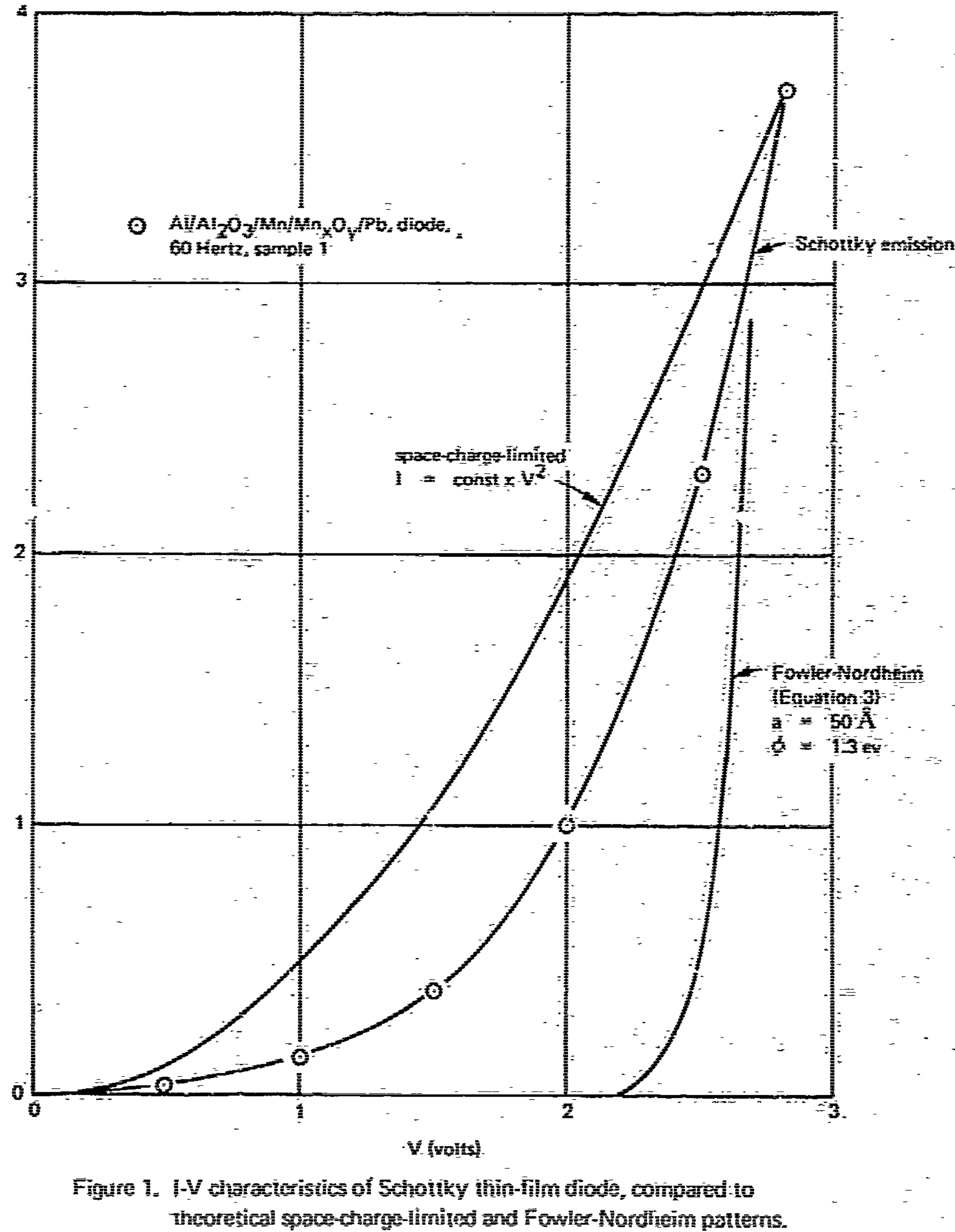

Figure 1. I-V characteristics of Schottky thin-film diode, compared to theoretical space-charge-limited and Fowler-Nordheim patterns.

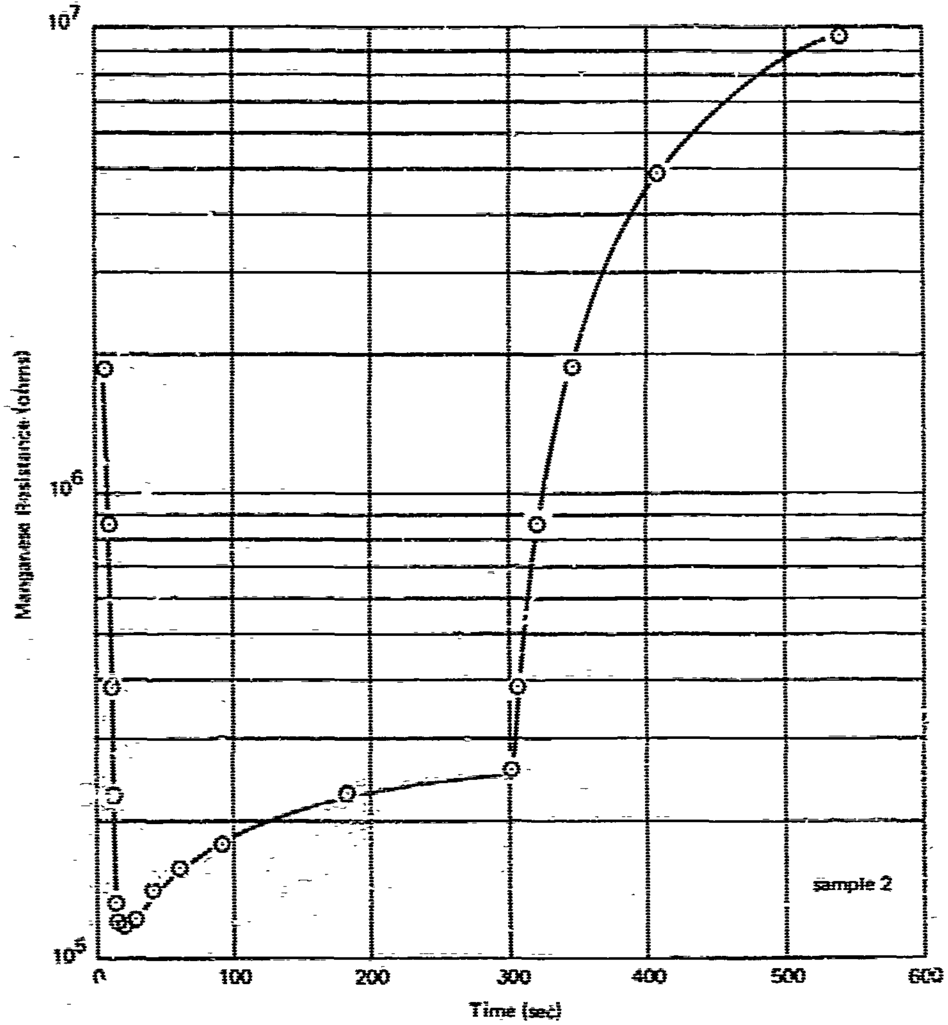

Figure 2. Manganese resistance versus time.

The five-layer M/I/M/I/M diode was completed by vacuum-depositing a 1-mm-wide strip of lead on and parallel to the manganese strip so as to prevent overlapping onto the aluminum oxide. To ensure electrical contact to the lead, the indium patches were smeared over the ends of the lead strip with a soldering iron. Figure 3 is a photograph of a completed five-layer diode with Schottky I-V characteristics.

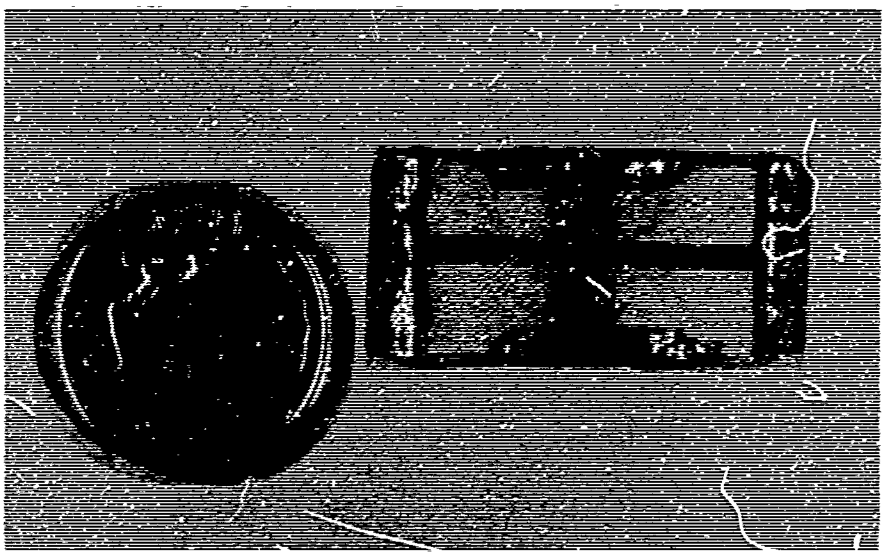

Figure 3. Five-layer M/I/M/I/M Schottky diode.

MEASUREMENT TECHNIQUES

Manganese resistance, as a function of time, was determined from a strip-chart recording of the voltage across a known resistance in series with the manganese film strip, a battery, and another known resistance. The values of the fixed resistors were such that not more than 45 μa flowed through the manganese strip.

The I-V characteristics of the finished diodes were determined by the usual four-wire current-source technique[1] and were recorded by means of either a DC X-Y recorder or an oscilloscope with a Polaroid-camera attachment. Room-temperature measurements were made with the diode exposed to air at atmospheric pressure.

To measure the temperature dependence of the diode characteristics, a cold-finger system was built, consisting of a copper plate attached to a copper cylinder immersed in one of three fluids: ice bath, dry-ice plus methyl alcohol, or liquid nitrogen. The diode substrate was attached to the copper plate by means of silicone grease, with the film side of the substrate facing up. To prevent destruction of the diode by condensation of water vapor from the air, the crossover junction and the Pb/Mn_xO_y strip were covered with a thick layer of silicone grease. The copper-constantan thermocouple for measuring the diode temperature was embedded in the cold-finger directly beneath the copper plate; some measurements were also made with the thermocouple indium-soldered to the top of the glass substrate a millimeter or so from the crossover area. Heat to the copper cold-finger was supplied by a chromel coil wrapped around the cylinder and electrically insulated from it by glass tape.

In order to obtain an indirect measurement of the diode-barrier thickness, high-frequency capacitance measurements were made between the aluminum and lead films. An RF reactance-bridge was used with a standard signal generator and RF voltmeter. Frequencies were between 0.40 and 2.0 MHz.

RESULTS OF MEASUREMENTS

Figure 4 shows the 60-Hertz I-V characteristic of a five-layer diode at room temperature. In Figure 5 a plot of **ln I** versus $V^{1/2}$ is given for the diode of Figure 4. The linearity of the **ln I**-$V^{1/2}$ plot is consistent* with Schottky emission, for which the theoretical I-V relation[2] is the following:**

$$I = AST^2 \exp\left(-\frac{\phi}{kT}\right) \exp\left[\frac{\alpha\left(\frac{V}{\epsilon a}\right)^{1/2}}{T}\right] \qquad (1)$$

where A = Richardson's constant, theoretical value = 120 amp/cm^2 $^{\circ}K^2$

S = active area of diode, assumed to be 10^{-2} cm^2

T = temperature, $^{\circ}$K

ϕ = relative work function between aluminum and lead, ev

k = Boltzmann's constant ev/$^{\circ}$K

α = constant, 4.339 for V in volts and a in cm

ϵ = dielectric constant (pure number) of barrier

a = barrier thickness, cm

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* A straight-line plot of **ln I** versus $V^{1/2}$ is not conclusive evidence of Schottky emission. Temperature-independent current flow, such as that due to tunneling, can yield very nearly linear **ln I**-$V^{1/2}$ plots.

** The reader is referred to the nomenclature on a foldout page at the end of the report.

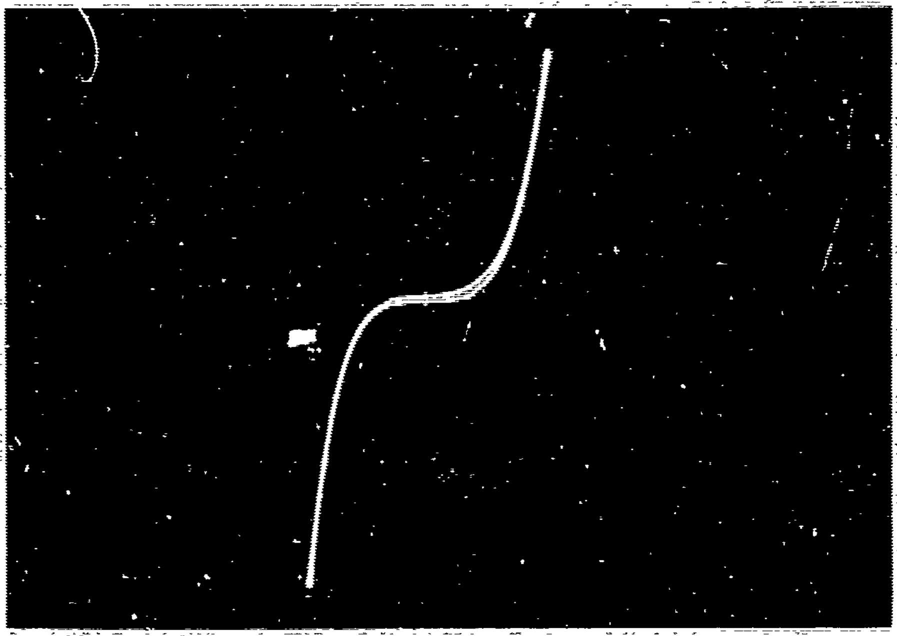


Figure 4. 60-Hertz I-V characteristic of five-layer Schottky diode. Horizontal scale, 5 v/cm; vertical scale, 1 ma/cm; $T \sim 300^0K$ (sample 3).

From Equation 1, the slope of $\ln I\text{-}V^{1/2}$ is $\alpha(1/\epsilon a)^{1/2}/T$, which yields the value of ϵa. Thus, from the diode capacitance, given approximately by

$$C = \frac{\epsilon S}{a} \tag{2}$$

both ϵ and a can be computed. For a typical five-layer diode, ϵ was found to be independent of T over the measurement range of 190^0K to about 350^0K and to be approximately 5. Barrier thickness, a, was found to be approximately 200 Å.

An estimate of the upper limit of the aluminum oxide thickness was made by comparing the I-V characteristics of $Al/Al_2O_3/Pb$ diodes with I-V plots computed from the Fowler-Nordheim relation:[3]

$$I = \left(\frac{1.535 \times 10^{-8}\, V^2}{\phi a^2}\right) \exp\left(-\frac{6.780 \times 10^7\, m^{1/2}\, a\, \phi^{3/2}}{V}\right) \text{ampere} \tag{3}$$

where m = the electronic mass, grams. The numerical quantities are based on a crossover area of 10^{-2} cm^2.

Figure 6 shows the I-V plot of an $Al/Al_2O_3/Pb$ diode for which the aluminum was oxidized 60 minutes in air at ambient temperature and ambient pressure. For 1.2 ev $\leqslant \phi \leqslant$ 2.0 ev and a = 50 Å, the Fowler-Nordheim plots would lie between the two calculated Fowler-Nordheim curves shown in Figure 6. Nelson and Anderson[4] found the photothreshold to be around 2.0 ev for light absorption in the overlayer of $Al/Al_2O_3/Al$ diodes with ~ 50 Å aluminum oxide layers. It is reasonable to assume, therefore, that the diode of Figure 6 has an oxide thickness not greater than 50 Å. This is to be expected, since 50 Å is well-known to be close to the upper limit on tunneling thickness; and the experimental plot of Figure 6 is clearly a tunneling curve and not consistent with some other current-flow mechanism which could occur in thicker oxide layers. The conclusion that the aluminum oxide layers in either the three-layer or five-layer diodes are not thicker than 50 Å is in agreement also with the findings of Hart.[5] According to Hart, aluminum oxide, produced by oxidation in pure oxygen at room temperature and atmospheric pressure, reaches a limiting thickness of 35 Å to 45 Å in a period of days. The data of Eley and Wilkinson[6] indicate that in air, at 300°K and 760 torr, a 10 Å layer of aluminum oxide forms in a period of a few seconds. Hence, the few-minute exposure of the aluminum film in the five-layer diodes is assumed to have produced aluminum oxide layers with thicknesses between 10 Å and 45 Å.

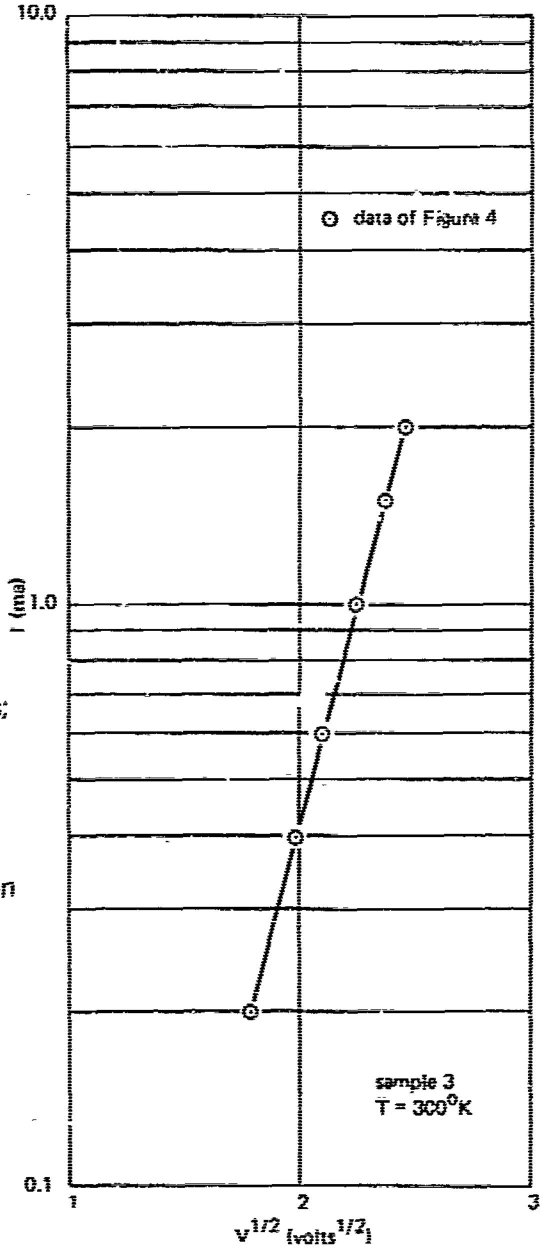


Figure 5. ln I versus $V^{1/2}$ for data of Figure 4.

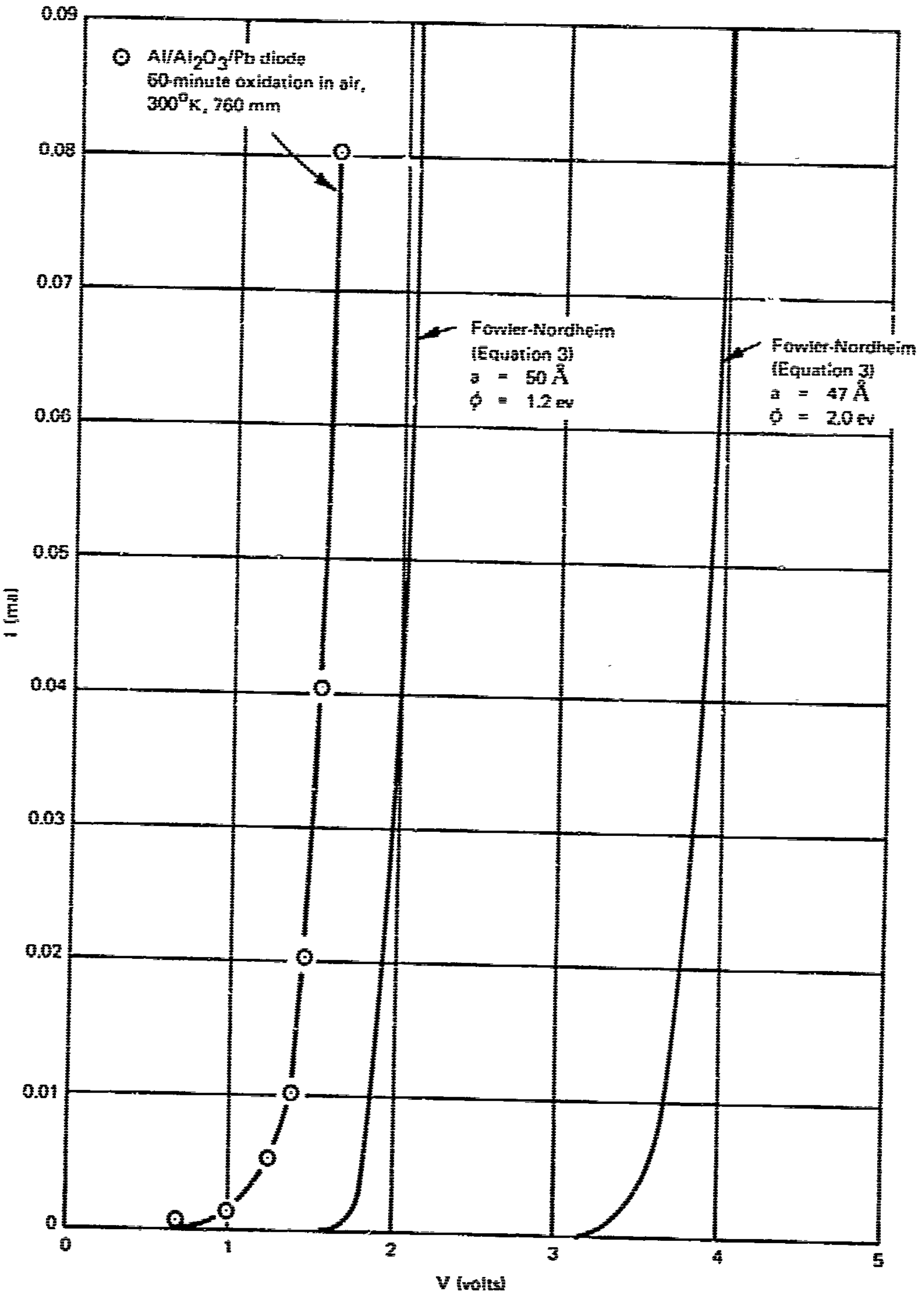


Figure 6. I-V characteristic of $Al/Al_2O_3/Pb$ thin-film diode compared to Fowler-Nordheim plots.

Presumably the barrier in these five-layer diodes consists of the thin-film system $Al_2O_3/Mn/Mn_xO_y$, although most of the manganese may have oxidized. If pure manganese was initially deposited, the minimum value of Ω/square yields a value of ρ_F, the manganese-film resistivity, around 4.8×10^{-2} Ω-cm. This is based on a film thickness of 160 Å, which, in turn, is derived on the basis of a 40 Å aluminum oxide layer. This value of ρ_F is approximately 10^4 times larger than the bulk resistivity of manganese, ρ_o; and it cannot be explained by the theory of thin-film resistivity in which ρ_F varies inversely with thickness as the result of the boundary scattering of electrons.[7] Since the pressure during manganese deposition was around 10^{-4} torr, it is more likely that not pure manganese, but rather a granular system of manganese, Mn_xO_y, and residual gases was deposited. The high value of ρ_F could be accounted for by the theory of thin-film resistivity which is based on the concept of agglomeration and the formation of potential walls throughout the film.[8] The agglomeration theory predicts a relation for ρ_F/ρ_o of the form

$$\frac{\rho_F}{\rho_o} = \text{const}\exp\left(\frac{\text{const}}{a_g}\right) \quad (4)$$

where a_g = the linear dimension of an average-size metal granule. Thus, it is possible, theoretically, for ρ_F/ρ_o to be about 10^4. On the other hand, in the boundary-scattering theory, ρ_F/ρ_o is given by

$$\frac{\rho_F}{\rho_o} = \frac{\text{const}\left(\frac{\lambda_o}{a}\right)}{\log\left(\frac{\lambda_o}{a}\right)} \quad (5)$$

where λ_o = the bulk electronic mean-free-path. For a in the 100 Å to 200 Å range, λ_o/a cannot cause ρ_F/ρ_o to approach 10^4.

The temperature dependence of the 60-Hertz I-V characteristics of these five-layer diodes is illustrated in Figure 7. The temperature range is 285°K to 344°K. Figures 8a and 8b show plots of **ln I-V$^{1/2}$** at different temperatures for two five-layer diodes operated by DC. The temperature dependence of **d ln I/dV$^{1/2}$** is plotted in Figures 9a and 9b for the data of Figures 8a and 8b. The **d ln I/dV$^{1/2}$ - 1/T** plot in Figure 9a can be fitted by a straight line which extrapolates to the origin within experimental error. The linearity and extrapolation to the origin is in agreement with Equation 1. Over the range of temperatures at which measurements were taken the slope of a straight-line fit to the **d ln I/dV$^{1/2}$ - 1/T** points was found, in general, to change from one reading of the raw data to another. To properly demonstrate that **d ln I/dV$^{1/2}$ - 1/T** extrapolates to zero would have required the determination of I-V characteristics at temperatures well above 350°K. This could not be done because of unstable I-V behavior above about 355°K.

The **d ln I/dV$^{1/2}$ - 1/T** plot in Figure 9b cannot very well be fitted by straight line. It is possible to show, however, that if a small tunneling current is superimposed on the Schottky current, the **d ln I/dV$^{1/2}$ - 1/T** plot will behave approximately like the plot in Figure 9b. If the total current is expressed as the sum of a Schottky current and a Fowler-Nordheim current, then

$$I = c_1 \exp\left(c_2 V^{1/2}\right) + c_3 V^2 \exp\left(-c_4 V^{-1}\right) \qquad (6)$$

where $c_1 = A S T^2 \exp(-\phi/k T)$ ampere

$c_2 = \alpha/(\epsilon a)^{1/2} T \text{ (volt)}^{-1/2}$

$c_3 = 1.535 \times 10^{-8}/\phi a^2 \text{ amp} - v^{-2}$

$c_4 = 6.780 \times 10^7 (a \phi^{3/2}) \text{ volt}$

For V = 1 volt, $190 \leq T < \infty$ °K, and **A S** equal to the experimentally derived value, 1.2×10^{-4} amp/°K^2, Equation 6 leads to

$$\frac{d \ln I}{dV^{1/2}} \cong b_1 T^{-1} + b_2 T^{-2} \exp\left(b_3 T^{-1}\right) \qquad (7)$$

where b_1, b_2, and b_3 = positive quantities dependent on the relative work function, ϕ, and barrier thickness, a.

Equation 7 is plotted in Figures 9a and 9b for comparison with the experimental plots of **d ln I/dV$^{1/2}$ - 1/T**. Equation 7 does not apply to temperatures below about 190°K because tunneling current increases relative to Schottky current, and the approximations made in deriving Equation 7 are no longer permissible. Furthermore, the plots of Figures 9a and 9b are for V = const = 1 volt; actually the Fowler-Nordheim relation, Equation 3, causes **d ln I/dV$^{1/2}$** to be voltage dependent. Thus, for V > 1 volt, the theoretical plot of **d ln I/dV$^{1/2}$ - 1/T** would have a smaller degree of bending than those in Figures 9a and 9b for the same range of temperatures.

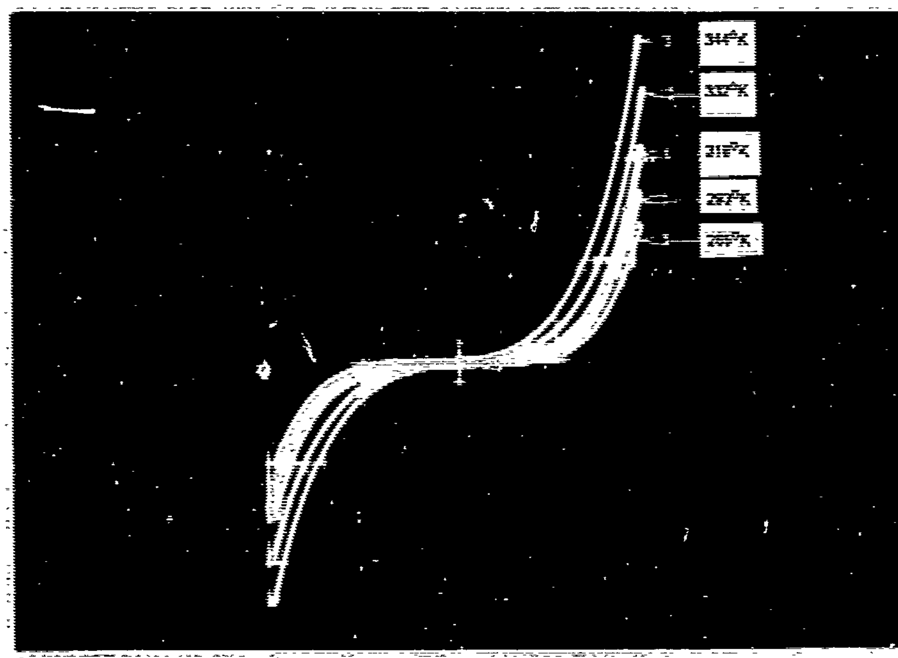


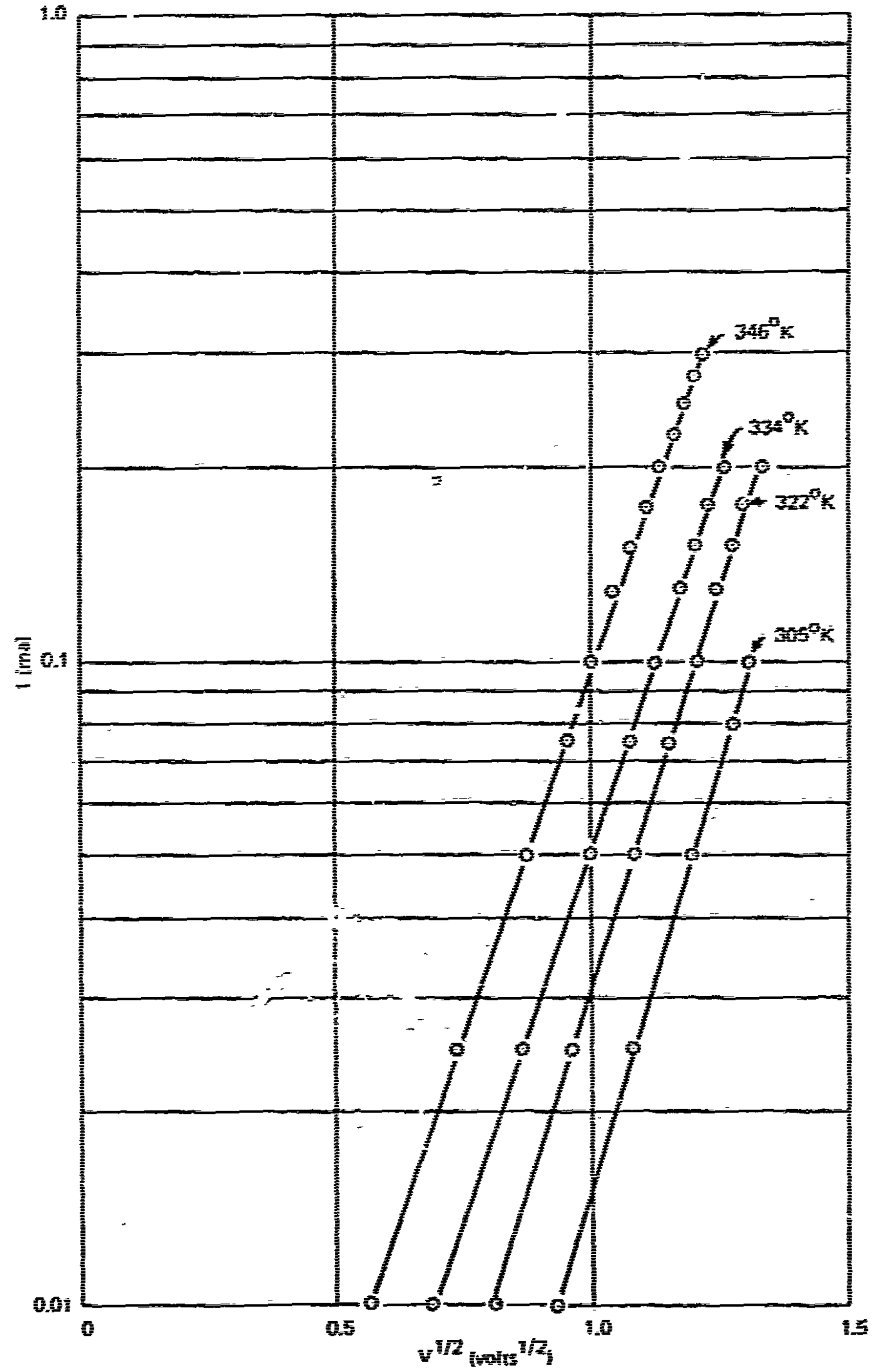
Figure 7. 60-Hertz I-V characteristics of five-layer Schottky diode.
Horizontal scale, 2 v/cm; vertical scale, 0.5 ma/cm (sample 4).

The relative work function, ϕ, can be computed from the experimental data by two methods.[9,10] One method uses the slope of the Richardson plot, **ln(I/T²)** versus **1/T**. According to Equation 1, this plot is a straight line with slope $[\alpha(V/\epsilon a)^{1/2} - \phi/k]$. The function ϕ is computed by inserting the value of ϵa, derived above. The other method of computing ϕ uses the $\ln(I/T^2)$ intercept of the Richardson plot and the **ln I** intercept of the $\ln I\text{-}V^{1/2}$ plot. The $\ln(I/T^2)$ intercept of the Richardson plot is **ln(A S)**, and from Equation 1

$$\phi = kT\ln\left(\frac{AST^2}{\beta}\right) \qquad (8)$$

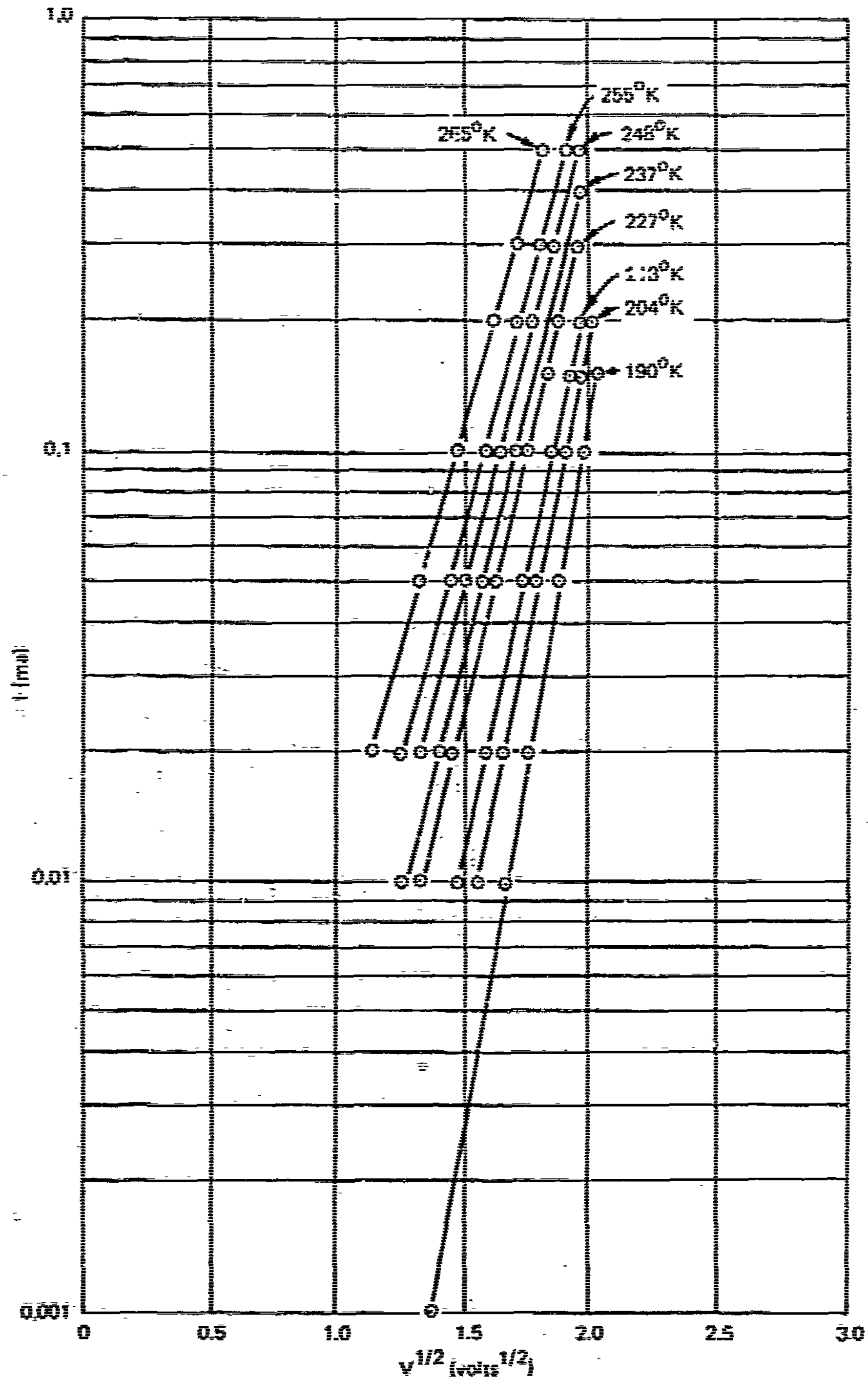
where $\ln\beta$ is the **ln I** intercept of $\ln I\text{-}V^{1/2}$.

Richardson plots for three M/I/M/I/M diodes are shown in Figure 10. The plot for sample 3 has a slope of -4.04×10^3 °K, which for $\epsilon a = 10^{-5}$ cm, yields a ϕ of 0.47 ev. Linear extrapolation of this plot gives 1.2×10^{-2} amp/cm² °K² for the Richardson constant, the value used above in plotting Equation 7. Inserting this value into Equation 8 gives ϕ at each temperature. The average value of ϕ turns out to be 0.51 ev, with maximum and minimum values differing by less than 0.01 ev.



(a) Sample 3

Figure 8. ln I versus $V^{1/2}$ for DC I-V characteristics of a five-layer Schottky diode.



(b) Sample 5

Figure 8. Continued.

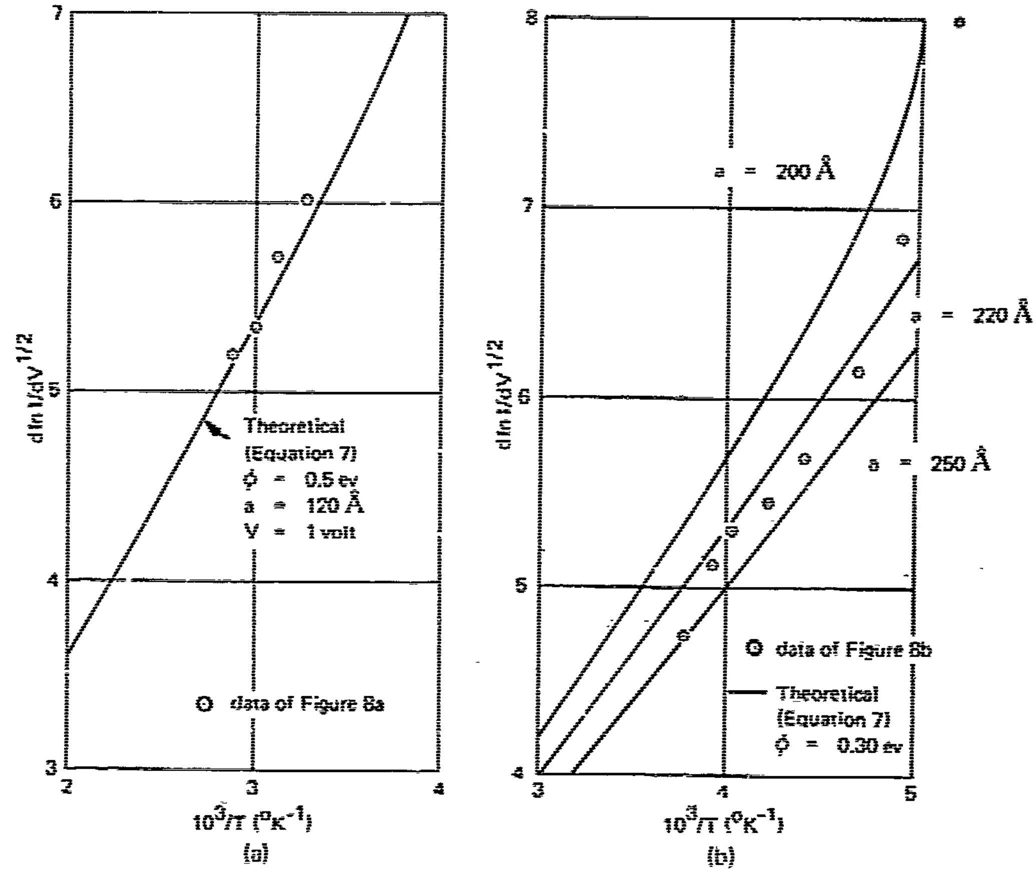


Figure 9. d ln I/dV$^{1/2}$ versus 1/T.

LONG-TERM PERFORMANCE

One of the Schottky diodes was operated at 60 Hertz for approximately 100 hours at a peak-to-peak voltage equal to 6 volts. Figure 11 compares the I-V characteristic of the diode at the beginning of the test with that which was recorded after a total operating time of 98 hours. The diode was without encapsulation of any kind and was exposed to the atmosphere. During the next 24 hours of operation, the peak-to-peak driving voltage was set at 7.6 volts, after which there was no indication that the diode characteristic was about to straighten or display dielectric breakdown. The peak-to-peak voltage was advanced to 8.4 volts, which then caused breakdown in about 6 minutes.

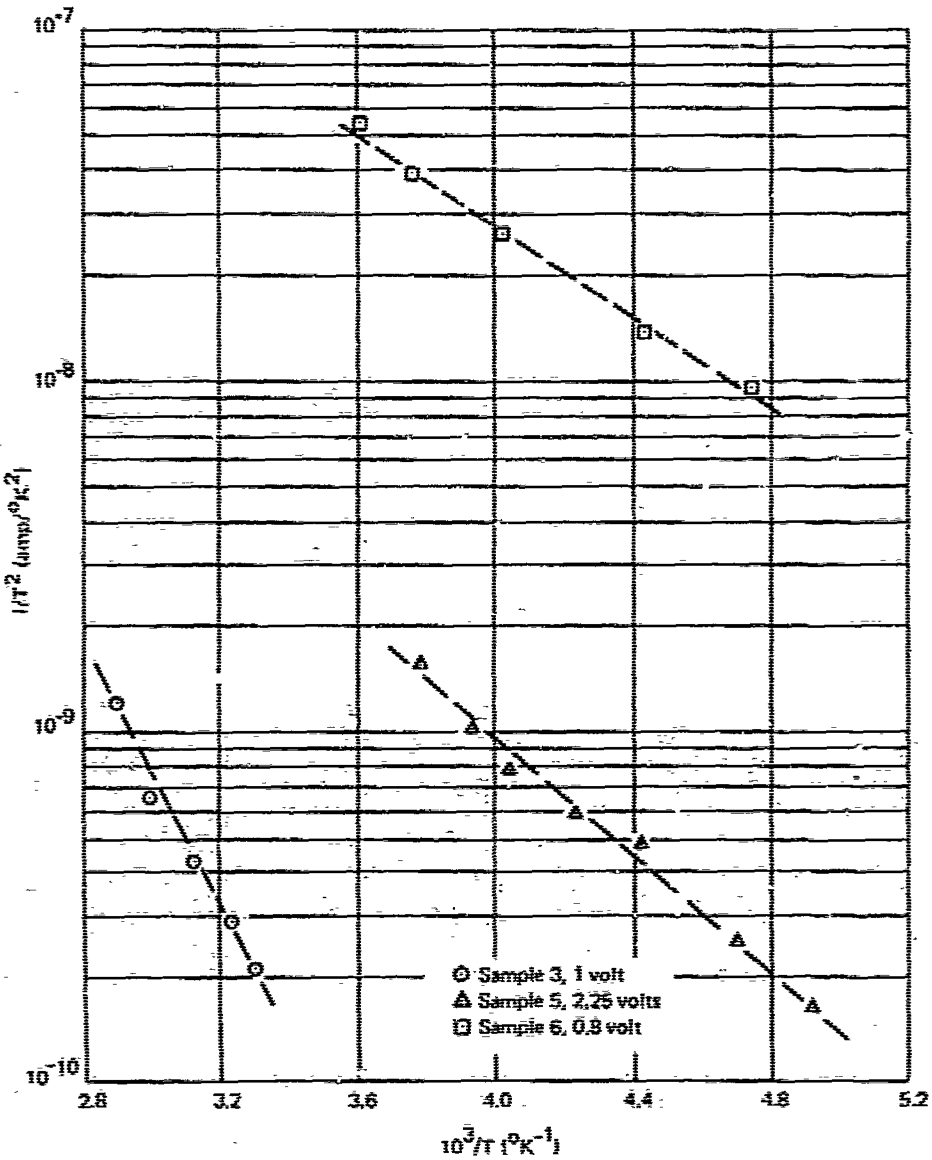


Figure 10. Ln(I/T^2) versus 1/T for DC I-V characteristics of five-layer Schottky diodes.

Only the diode of Figure 11 was tested nearly continuously for approximately 124 hours. All of the diodes with Schottky characteristics had highly stable I-V patterns during measurements, which, over the temperature range of 190°K to about 350°K, usually required between 30 and 60 minutes. A number of diodes were operated briefly at randomly chosen times over a total period of 1 or 2 days; total on-time for these diodes is estimated to be around 1 hour. In no case was the I-V characteristic, DC or 60 Hertz, observed to change significantly unless the diode temperature had been raised above ~ 355°K.

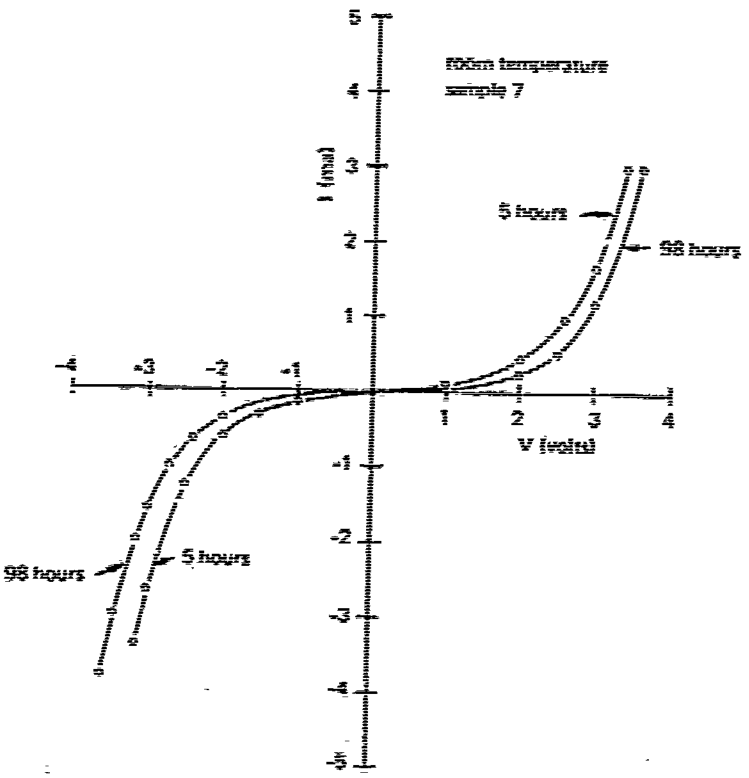


Figure 11. 60-Hertz I-V characteristics of five-layer Schottky diode, showing effect of 98-hour operation.

CONCLUSIONS

1. The data of Figures 9a, 9b, and 10 provide conclusive evidence that Schottky emission is the dominant current-flow mechanism in the thin-film system $Al/Al_2O_3/Mn/Mn_xO_y/Pb$.

2. This type of thin-film diode can be operated at 60 Hertz while exposed to the atmosphere for 124 hours before barrier deterioration occurs.

3. Although the ideal voltage-regulator I-V characteristic was not achieved with this five-layer diode, nominal refinement of the fabrication technique will lead to a thin-film varistor diode which is easy to manufacture and which has both long-term stability and a moderate current-carrying capability.

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NOMENCLATURE

A	Richardson constant, amp/cm^2 $^oK^2$
a	Barrier thickness, cm
a_g	Linear dimension of an average-size metal granule
b_1 b_2 b_3	Positive quantities dependent on the relative work function, ϕ, and barrier thickness, a
C	Diode capacitance, farads
c_1	$A S T^2 \exp(-\phi/k T)$ ampere
c_2	$\alpha/(\epsilon a)^{1/2}$ T (volt)$^{-1/2}$
c_3	$1.535 \times 10^{-8}/\phi a^2$ amp volt^{-2}
c_4	$6.780 \times 10^7 (a\phi^{3/2})$ volt
k	Boltzmann's constant, ev/^{o}K
m	Electronic mass, grams
S	Active area of diode, cm^2
T	Temperature, ^{o}K
α	Constant
$\ln \beta$	ln I intercept of ln I-V$^{1/2}$
ϵ	Dielectric constant of barrier
λ_o	Bulk electronic mean-free-path
ϕ	Relative work function between aluminum and lead, ev
ρ_F	Manganese-film resistivity, Ω-cm
ρ_o	Bulk resistivity of manganese, Ω-cm

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13. ABSTRACT

Temperature-dependent current-voltage (I-V) characteristics have been observed in a new type of thin-film diode, consisting of Al, Al_2O_3, Mn, Mn_xO_y, and Pb. Plots of ln I-$V^{1/2}$, d ln I/d$V^{1/2}$ – 1/T, and ln(I/T^2) – 1/T (where T is temperature in °K) can be fitted by straight lines which show that Schottky emission is the dominant current-flow mechanism over the temperature range of 190°K to about 350°K. Barrier thicknesses were determined by high-frequency capacitance measurements and found to lie between 100 Å and 250 Å. Relative work functions, between the aluminum and lead films, were found to lie between 0.25 and 0.50 ev. One of the diodes was tested at 60 Hertz for 124 hours without causing a significant change in the shape of the I-V characteristic. It is concluded that nominal refinement of the fabrication technique will lead to a varistor thin-film diode which is inexpensive and which has long-term stability.

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