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Schottky Currents in Dielectric Films

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The Frank-Simmons theory of emission-limited conduction in insulators has been extensively explored. The current-voltage data for Mylar, SiO, and Ta_2O_5 insulating films have been calculated and the parameters needed to fit the data compare well with experimental values. The parameters needed to fit the I-V data for Mylar were a work function (Φ) of 1.40–1.45 eV, a dielectric constant (ϵ) of 2.44, and a mobility-trapping factor product ($\mu\theta$) of 10^{-14} to 10^{-18} $m^2 \times V^{-1}$ sec^{-1}. For Ta_2O_5, the parameters found were $\Phi = 0.775$ eV, $\epsilon = 3.2$, and $\mu\theta = 1 \times 10^{-13}$ $m^2 \times V^{-1}$ sec^{-1}. In fitting these SiO data, it was found necessary to employ a temperature-dependent work function which varied from 0.66 eV at 195 °K to 0.80 eV at 297 °K more or less uniformly with a slope of approximately 1.4×10^{-3} eV °K^{-1}. The model of Frank and Simmons is analyzed in terms of both the Poole-Frenkel effect and hot-electron effects, and equations containing both terms are presented.

I. INTRODUCTION

The hot-electron concept of Frohlich,[1] and the resulting field-dependent conductivity, was recently employed by O'Dwyer[2] to predict current-voltage characteristics for insulators. We have investigated[3] the applicability of O'Dwyer's theory for the case of Mylar films and found that fair agreement with experiment could be achieved if appropriate values were chosen for the material parameters. The agreement was only qualitative, however, because (a) the necessary values for the bulk properties of the insulator did not agree well with generally accepted values; (b) the curvatures of the calculated Schottky graphs (logarithm of the current density versus the square root of the voltage) were different from those of the experimental graphs. In particular, experimental Schottky graphs for many insulators are linear at high-field strengths, and the slopes are often indicative of Schottky emission from the cathode with no space charge present in the sample. This pure Schottky behavior is not predicted by the O'Dwyer theory, as the calculations did not yield a linear Schottky graph at high fields.

The incorrect high-field predictions of O'Dwyer's theory can be traced to the failure of his model to remove the space charge from the sample. The only mechanism provided for this is the difference between the effective electron temperature T_e and the lattice temperature T. The relationship between these is taken to be

$$1/kT - 1/kT_e = E^2/E^{*2}\Delta V \quad , \tag{1}$$

where k is Boltzmann's constant, E is the electric field, E^* is the breakdown field of the dielectric, and ΔV is the energy range below the conduction band covered by isolated shallow traps. Equation (1) is only valid for $E \ll E^*$; therefore, if ΔV is taken as several (kT), it is easy to see that the maximum difference between T_e and T is a small fraction of T. If the deep traps of the insulator are on the order of 1 eV below the conduction band, only a small fraction of the space charge will ever be removed from the traps. This behavior is typical of neutral trapping centers (i.e., centers which are uncharged when empty), which might be expected to be of primary importance in organic solids such as Mylar.

However, if the deep traps are positively charged when empty (as in the case of a donor atom), then it is necessary to account for the field lowering of the trap depth, commonly known as the Poole-Frenkel effect.[4] In this case the trap depth is taken as

$$U = U_0 - e(eE/\pi\epsilon_0 K)^{1/2} \quad , \tag{2}$$

where U_0 is the depth of the trap below the conduction band in the absence of an applied field, K is the dielectric constant, and e, π, and ϵ_0 have their usual significance. If E is in the neighborhood of 10^8 V/m, it is seen that the field lowering of the trap depth can be of the order of an eV for reasonable dielectric constants. Thus, if U_0 is of the order of an eV, the Poole-Frenkel effect can obviously be a very efficient mechanism for releasing the space charge out of the sample.

In two recent papers,[5,6] Simmons has discussed possible sources of appropriate donor centers in films of SiO and Ta_2O_5, and has used Eq. (2) to develop a model for bulk-limited conduction in those systems. It is more difficult to justify the existence of donors or of charged trapping centers in Mylar films. At this time, the only obvious possibility appears to be metal im purities from the catalysts employed in the polymerization process. At any rate, we shall assume throughout this paper that the deep traps of im-

portance in Mylar are subject to the Poole-Frenkel effect. This assumption is made so that we can accomplish the primary aim of this paper, the further testing of an emission-limited model to describe current-voltage characteristics in dielectric films.

The model we employ is basically the emission-limited model of Frank and Simmons.[7] Before discussing our calculations, however, we briefly reanalyze the model in Sec. II. The reason for doing this is to clarify the difference between the models suggested by O'Dwyer[2] and by Frank and Simmons.[7] In fact, we redevelop the model accounting for both the hot-electron effect of Eq. (1) and the Poole-Frenkel effect of Eq. (2). We shall find that, for electric fields which are well below breakdown, the approximate equations developed by Frank and Simmons provide a very good approximation to the generalized results. That is, in this range of electric fields, hot-electron effects are essentially negligible.

Section III describes our attempts to fit some actual current-voltage data. For Mylar, SiO, and Ta_2O_5 films, we find very good agreement between theory and experiment. As Frank and Simmons have already pointed out[7] this theory predicts a transition to emission-limited conditions in every case at moderate applied fields. There are some experimental data available which do not exhibit this transition, and these constitute discrepancies for the model.

II. MODEL

Frank and Simmons did not include hot-electron effects in their theory. Their main contribution was to account for the field lowering of the trap depth via Eq. (2). Thermal equilibration of the electrons between the traps and the conduction levels was assumed, and the continuity equation

$$J = n_c e \mu E \tag{3}$$

was solved together with Poisson's equation

$$dE/dx = ne/K\epsilon_0 \ . \tag{4}$$

The symbols not previously defined are J, the current density; n_c, the density of conduction electrons; n, the total space-charge density; μ, the electron mobility; and x, the distance from the cathode. With the assumption that $n_c \ll n$ for good insulators, the working equation developed was

$$dE/dx = J \exp[-\beta E^{1/2}/(K\epsilon_0)\theta_0 \mu E] \ , \tag{5}$$

where $\beta = (e/kT)(e/\pi K\epsilon_0)^{1/2}$ and θ_0 is the ratio of conduction electrons to trapped electrons in the absence of a field, i.e.,

$$\theta_0 = (\rho_c/\rho_t)\, e^{-U_0/kT} \ , \tag{6}$$

where ρ_c and ρ_t are the density of states in the bottom kT of the conduction band and the density of traps, respectively.

The only difference between the above model and that developed by O'Dwyer is in the method of distributing the electrons between conduction levels and traps. O'Dwyer employed the hot-electron concept, but assumed that the trap depth was field independent, whereas Frank and Simmons relied on the Poole-Frenkel effect to obtain an appropriate distribution. Since O'Dwyer's model was at least qualitatively successful, it is appropriate to redevelop the Frank-Simmons model taking into account the hot-electron concept. This turns out to be straightforward.

We follow O'Dwyer's development and introduce n_0 as the space-charge-free electron density over all traps and conduction levels. This is really the density of electrons in the isolated insulator which become available for conduction when the trap depth is decreased by Eq. (2). If these electrons are removed from the traps, a positive charge is left behind, and this is the situation to which Eq. (2) applies. This number n_0 was never explicitly introduced in the development of Frank and Simmons, but its value could easily be deduced from the relationships between the electron mobility μ the zero-field conductivity σ_0 and the zero-field trapping factor θ_0 of Eq. (6).

In the absence of an electric field, the density of conduction electrons n_{co}, is given by the Boltzmann ratio,

$$n_{co} = (n_0 - n_{co})\theta_0 \ , \tag{7}$$

and the zero-field conductivity is

$$\sigma_0 = n_{co} e \mu = (n_0 - n_{co}) e \mu \theta_0 \ . \tag{8}$$

When a strong electric field is impressed, let the space-charge density be n. The total density of electrons to be distributed between traps and conduction levels is then equal to (n_0+n), and of these the density n_c will be in conduction levels. Generalizing Eq. (7), we have

$$n_c = (n_0 + n - n_c)\theta \ , \tag{9}$$

where $\theta = (\rho_c/\rho_t)\, e^{-U_0/kT_e}$, (10)

and the electron temperature T_e may be different from the lattice temperature T. If Eqs. (1) and (2) are inserted into (10), the ratio becomes

$$\theta = \theta_0 \exp[f(E)] \ , \tag{11}$$

with $f(E) = (U_0/\Delta V)E^2/E^{*2} + \beta E^{1/2} - (kT\beta/\Delta V)E^{5/2}/E^{*2}$. (12)

The first term in this equation corresponds to O'Dwyer's result, and the second term is what resulted from the development by Frank and Simmons. The third term is a new one that has not been employed previously. It arises because both the hot-electron concept and the field lowering of the trap depth have been utilized.

The generalization of Eq. (8) in the presence of the field is

$$\begin{aligned}\sigma &= n_c e\mu \\ &= (n_0 + n - n_c)e\mu\theta \\ &= \sigma_0 \exp[f(E)](n_0 + n - n_c)/(n_0 - n_{c0}).\end{aligned} \tag{13}$$

If we now use the continuity equation in the form

$$J = \sigma E \ , \tag{14}$$

it becomes possible to solve Eq. (13) for the space-charge density:

$$n = n_0\left[\left(\frac{1+\theta_0\exp[f(E)]}{1+\theta_0}\right)\frac{J\exp[-f(E)]}{\sigma_0 E} - 1\right]. \tag{15}$$

This is very similar to that obtained by O'Dwyer. The factor in parentheses did not appear in O'Dwyer's result because he introduced the approximation that $n_c \ll (n+n_0)$ and we did not. And, more importantly, the present $f(E)$ is the entire expression (12) rather than just the first term as obtained by O'Dwyer.

It is possible to rewrite Eq. (15) in terms of mobility rather than conductivity, and one obtains

$$n = J\exp[-f(E)]/e\mu\theta_0 E + J/e\mu E - n_0 \ . \tag{16}$$

This equation now looks like the result of Frank and Simmons. The first term is identical to their result, implicit in Eq. (5), except that their relation contained only the $E^{1/2}$ term of $f(E)$. The second and third terms were really neglected by them, again by the assumption that $n_c \ll (n_0+n)$.

If one inserts reasonable numbers for all the parameters in Eq. (12), he is soon convinced that so long as the electric field E does not approach too closely to the breakdown field E^* the only term of importance is the one containing $E^{1/2}$, i.e., the one used by Frank and Simmons. It should be clearly noted, though, that this is not an additional restriction: It has already been pointed out that if E approaches E^* then Eq. (1) ceases to be a valid approximation. For the calculations described in Sec. III, the maximum fields in the insulators were always well below the assumed breakdown field. As a result, we conclude that Eq. (5) obtained by Frank and Simmons is an extremely good approximation to the more complete result obtained by combining Eq. (15) or (16) with Poisson's equation (3). In fact, we have made a very large number of calculations using both procedures, and in no case did we obtain any detectable difference.

III. CALCULATED RESULTS

In determining the emission current, Frank and Simmons accounted for the image force by writing the thermionic emission equation as

$$J = AT^2 e^{-\Phi_m/kT}, \tag{17}$$

where $A = 1.2\times 10^6 \mathrm{A/m^2}$ and $\Phi_m = \Phi_0 + \Delta\Phi$. Here Φ_0 is the difference in energy between the Fermi level in the cathode and the bottom of the conduction band in the dielectric. $\Delta\Phi$ is the increase in the potential barrier due to both the applied field and the image force,

$$\Delta\Phi = \int_0^{x_m} E(x)dx = e/16\pi\epsilon_0 K x_m \ . \tag{18}$$

The distance x_m from the cathode to the barrier maximum is the distance at which the applied field equals the image field. The gradient of Φ, the applied field, was taken to be constant between the cathode and x_m, a reasonable assumption because of the small range of the image force. Upon choosing a current, the field E_0 at the cathode was obtained from the Schottky equation

$$\begin{aligned}J &= (4mek^2T^2/h^3) \\ &\quad\times\exp[-(\Phi_0 - e^{3/2}E_0^{1/2}/4\pi K^{1/2}\epsilon_0^{1/2})kT] \ ,\end{aligned} \tag{19}$$

where m is the electron mass and h is Planck's constant. Then Eq. (18) was used to obtain the actual potential barrier between the cathode and x_m. From x_m on, calculations were done by numerically integrating Poisson's equation as described below.

Frank and Simmons integrated Poisson's equation to obtain an implicit equation for $E(x)$. We have found this to be too complicated for practical use, and have simply proceeded by numerically integrating Eq. (5) or the equivalent obtained by combining Eq. (3) with either Eq. (15) or (16). The potential across the dielectric was also obtained numerically from

$$V(x) = \int_0^x E(x')dx' \ . \tag{20}$$

To test our method of calculation, we began by recalculating the sample Schottky graphs given by Frank and Simmons. In every case, the agreement was perfect. We then proceeded to fit some actual experimental data for Mylar, silicon oxide, and tantalum oxide.

The parameters that must be specified in each case are the work function, Φ_0; the dielectric constant, K; and the product, $\mu\theta_0$, of electron mobility and trapping factor. The work function determines the magnitude of the emission current from the cathode, the dielectric constant controls

the slope of the Schottky graph in the space-charge-free (linear) region, and the product $\mu\theta_0$ determines the onset of emission-limited conditions. With this knowledge, it is fairly straightforward to determine the parameter values needed to optimally match an experimental curve. The approach taken was purely one of trial and error and then the optimum parameter values were compared with values from other sources whenever possible.

A. Mylar

In Figs. 1 and 2 are shown the best fits obtained to the data of Lilly and McDowell[8] on 5-mil and 1-mil Mylar films. In both figures an attempt has been made to fit the data with one set of parameters. However, the low-temperature experimental curves did not have a slope close to the theoretical "pure-Schottky" value so the theoretical curves are far off, particularly in the 101°C curve of Fig. 2. In the case of the other curves in Figs. 1 and 2 the theory fits the data in a reasonable fashion and if small changes were made in Φ and $\mu\theta$ for the individual curves the agreement could be made even better. It is interesting to note that the parameters that give the best fit, $\Phi = 1.4$–1.45 eV and $\epsilon = 2.44$, are the same as obtained in using the O'Dwyer "hot-electron" model,[3] although the general agreement is much better here. The value of $\mu\theta$ ranging from 10^{-14} to 10^{-18} $m^2 V^{-1} sec^{-1}$ does not seem unreasonable for a low-mobility good-trapping material such as Mylar film. Lilly and McDowell also published data on Mylar films of 10 mil thickness. As they indicated, however, the high-field linear portions of the Schottky graphs did not exhibit pure Schottky coefficients in that case. It naturally follows that the present model cannot describe those data, and they have therefore been omitted.

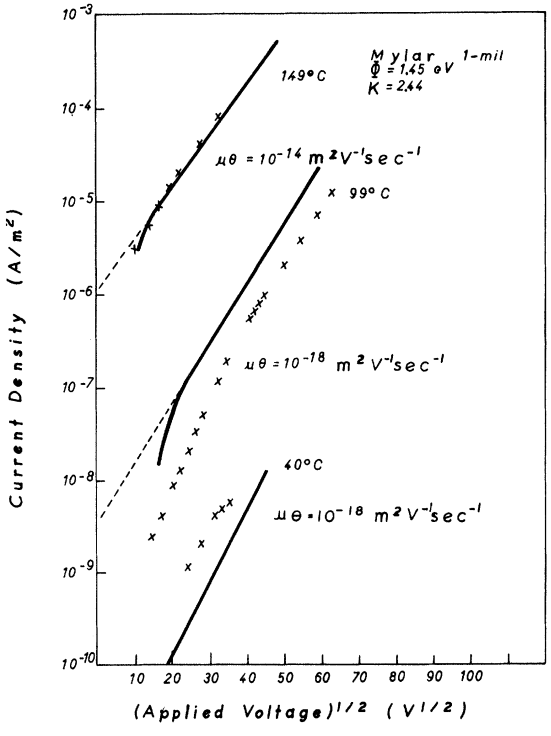

FIG. 1. Calculated I-V curves (solid lines), superimposed on the experimental data (X) for 1-mil Mylar at these temperatures. The $\mu\theta$ value for the 40 °C calculated curve is 10^{-18} m^2 V^{-1} sec^{-1}.

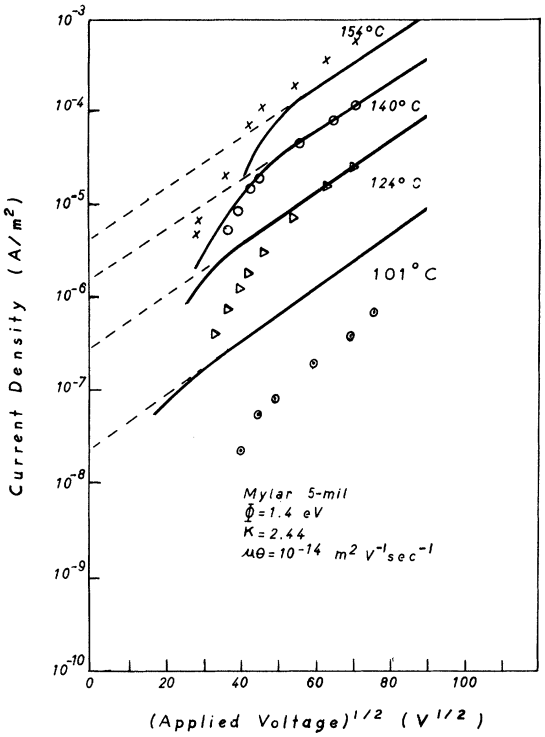

FIG. 2. Experimental (points) and theoretical (solid lines) I-V curves for 5-mil Mylar are shown above for four temperatures.

It should be mentioned that the recent model — developed by Simmons[6] for the transition from electrode-to bulk-limited conduction in the case of a blocking contact at the cathode surface – does indeed lead to a dependence on film thickness. However, it is clear that this theory cannot reconcile the 10-mil Mylar data with the 1- and 5-mil data either, because the thickness dependence predicted is actually in the opposite direction from that observed. Thus one has to invoke the anomalous Poole-Frenkel effect[5] in order to explain the Schottky slopes observed for the 1- and 5-mil films, but the 10-mil experimental data[8] agree much more closely with the normal Poole-Frenkel

effect. It is clear that the 10-mil Mylar presents an anomaly in either case, and that is where we prefer to leave the matter. A possibility that cannot be overlooked is that films of different thickness are manufactured differently, and that the conduction mechanisms are in fact different due to the presence of different impurities or different amounts of the same impurities.

Figure 3 shows Richardson plots obtained with the intercepts of the straight-line portions of the theoretical Schottky graphs for 1-, 5-, and 10-mil Mylar. The slopes of these lines yield work functions of 1.44 eV for the 1-mil case and 1.37 eV for both the 5- and the 10-mil, which are very close to the values employed in the calculations. A similar treatment of the experimental data by Lilly and McDowell resulted in σ equal to 1.77, 1.86, and 2.21 eV for the 1-, 5-, and 10-mil Mylar, respectively. The discrepancies are obviously due to the lack of perfect agreement between the calculated and experimental data which has already been noted. The true work function is probably closer to the 1.4 eV obtained here, which is the same as obtained in Ref. 3.

In Fig. 4 are plots of space-charge density

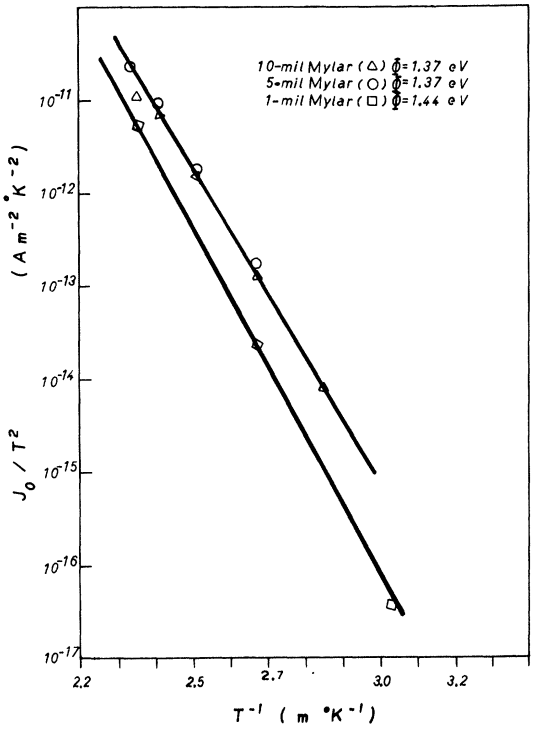

FIG. 3. Richardson plots obtained with the intercepts of the straight-line portions of the theoretical Schottky graphs for 1-, 5-, and 10-mil Mylar. The slopes of the curves in eV are shown on the graph. Here J_0 is the zero-field current density.

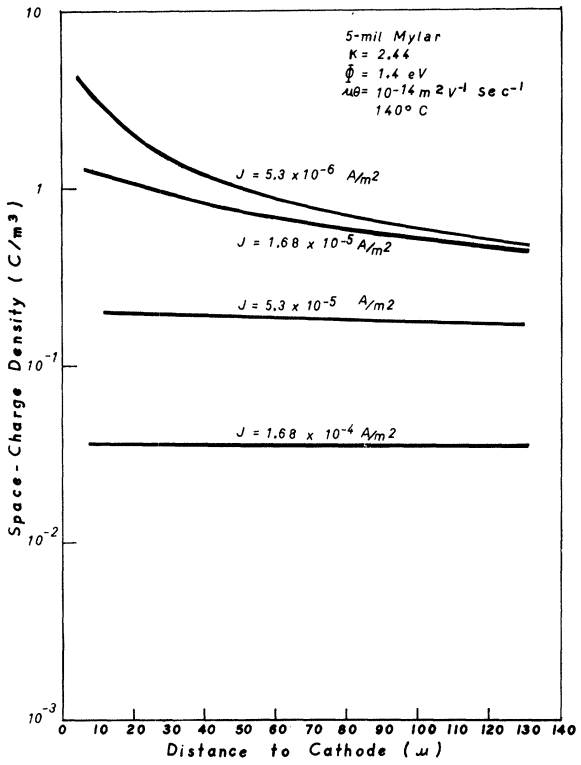

FIG. 4. Curves above are plots of space-charge density calculated as a function of distance from the cathode for several current densities at a temperature of 140 °C.

calculated as a function of distance from the cathode for several current densities at a temperature of 140 °C. Two points to be noted here are (a) the space-charge density decreases with increasing current, which is opposite to that found with the calculations[3] on O'Dwyer's model and (b) the space-charge densities obtained here are several orders of magnitude smaller than those calculated earlier.[3] Clearly, these results arise because of the much greater efficiency of the Poole-Frenkel effect, relative to the hot-electron mechanism, in removing space charge from the traps.

B. Tantalum Oxide

In Fig. 5 we show the fit made to the current-voltage data on 870-Å Ta_2O_5 given in Fig. 5 of Mead's paper.[9] Room temperature is taken to be 300°K and Mead's approximate electrode area of 10^{-3} cm^2 is used to obtain the current density. Values of the parameters of $K = 3.2$, $\Phi = 0.775$ eV, and $\mu\theta = 1 \times 10^{-13}$ $m^2 V^{-1}$ sec^{-1} give a very reasonable description of the data. The value of the dielectric constant of 3.2 is lower than that used by Mead, but as Simmons has pointed out,[5] the

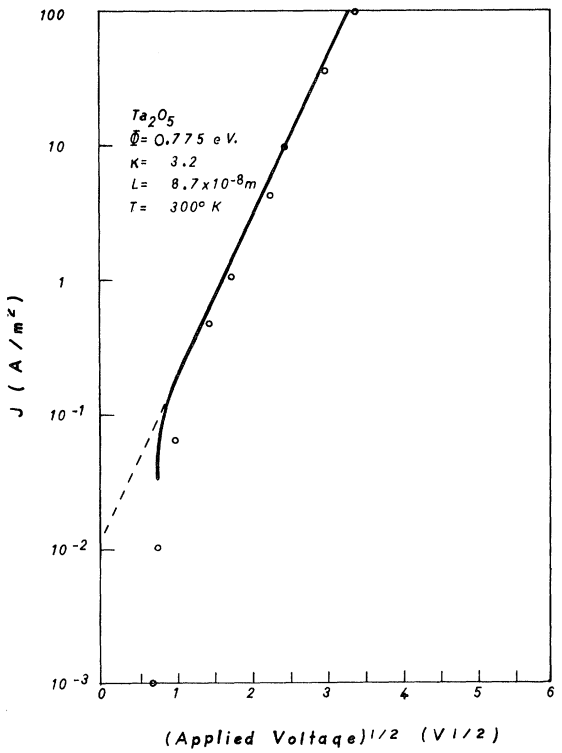

FIG. 5. *I-V* calculations from the theory (points) fitted to Mead's[9] data (solid line) on Ta_2O_5. A $\mu\theta$ of 10^{-13} m^2 V^{-1} sec^{-1} was used in the calculations.

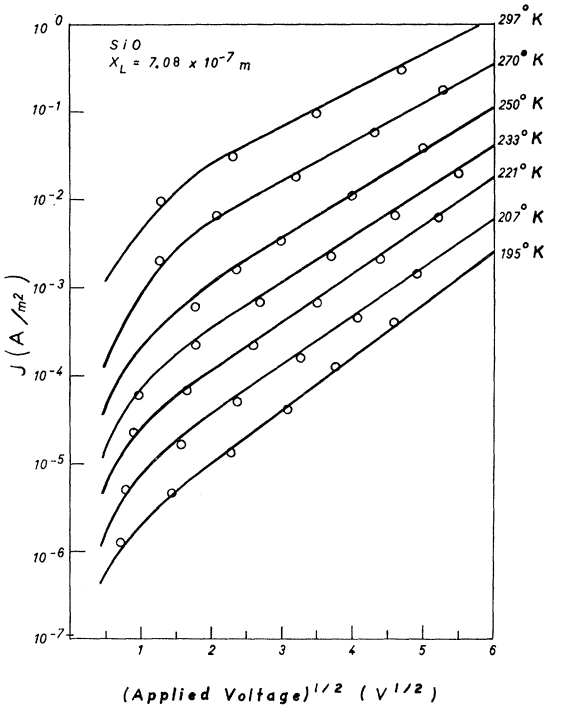

FIG. 6. *I-V* calculations from the theory (points) fitted to the data of Hartman *et al.*[10] on SiO (solid line) at different temperatures. The parameters used in the calculations are shown in Table I.

optical value of K should be used. The value of the work function obtained 0.775 eV is about the same as Mead obtained from a Fowler-Nordheim plot of a typical sample, but about twice that obtained from $\log_{10} I$-versus-$1/T$ plots in Mead's paper. Mead originally interpreted the data as a bulk-limited Poole-Frenkel conduction, but it is clear that the emission-limited conduction theory of Frank and Simmons is also a reasonable interpretation.

C. Silicon Oxide

Hartmen, Blair, and Bauer have presented[10] an extensive set of current-voltage-temperature data for SiO films, and they concluded that their data were not amenable to interpretation in terms of Schottky currents. Simmons agrees with these workers, and, in fact, has recently discussed[6] how the data might be interpreted on the basis of bulk-limited conduction. Our calculations have resulted in such good agreement with the experimental data, however, that we are led to precisely the opposite conclusion.

In Fig. 6 is shown a comparison of our calculated results with experimental data taken from Fig. 3 of the paper by Hartman *et al.*[10] Obviously, the present model is capable of giving a very precise interpretation of these data. The parameters that were employed in our calculations are displayed in Table I. It is interesting that the appropriate value for the product $\mu\theta_0$ was constant at 10^{-13} m^2 $V^{-1}sec^{-1}$ between 195 and 250 °K, but then increases to 10^{-11} at 297 °K. The one possibly disconcerting feature of our calculations was the necessity of varying the work function from 0.66 eV at 195 °K to 0.80 eV at 297 °K. This represents a more or less uniform increase in Φ with temperature of about 1.4×10^{-3} eV/deg. This is of the order of the increase in the work function obtained with oxide cathodes. In a very recent study of Fowler-Nordheim emission into silicon

TABLE I. Parameters needed to fit Hartman *et al.*[10] *I-V-T* data for SiO.

T (°K)	Φ (eV)	$\mu\theta$ (m^2 V^{-1} sec^{-1})	K
195	0.66	1×10^{-13}	3.25
207	0.70	1×10^{-13}	3.25
221	0.70	1×10^{-13}	3.25
233	0.72	1×10^{-13}	3.25
250	0.74	1×10^{-13}	3.25
270	0.76	1×10^{-12}	3.25
297	0.80	1×10^{-11}	3.25

dioxide, Lenzlinger and Snow have found a similar change in the barrier height with temperature.[11] The dielectric constant we employed 3.25 is somewhat lower than the value of 5.8 obtained at 1592 Hz by Hartman *et al.*[10] but a reduction is to be expected at higher frequencies.

Figure 7 shows the predictions of the present model in regard to the thickness dependence of the current-voltage characteristics of SiO. These calculated curves essentially reproduce the experimental data in Fig. 2 of Ref. 10; however, the work function Φ has to be increased slightly from 0.80 to 0.82 eV as the thickness is reduced from 6800 to 1300 Å. The parameters used to fit the experimental data are shown in Table II. Although the total increase in work function of 0.02 eV is small, it could represent an important shortcoming of the emission-limited model. The primary alternative to the present emission-limited model would appear to be the bulk-limited mechanism described by Simmons.[6] As we have already stated, that model also predicts a thickness dependence. In fact, Simmons has discussed these same data in the context of his model. In particular, he pointed out that when the straight lines (of the bulk-limited regions) of the Schottky graphs are extrapolated to zero voltage, the lines corresponding to different thicknesses do not have a common intercept. He also pointed out, however, that the order of the intercepts is opposite from that observed. The results of our calculations in Fig. 7 show very clearly that the thicker films uniformly have higher intercepts, just as do the experimental extrapolations,[10] but the work function has to change slightly.

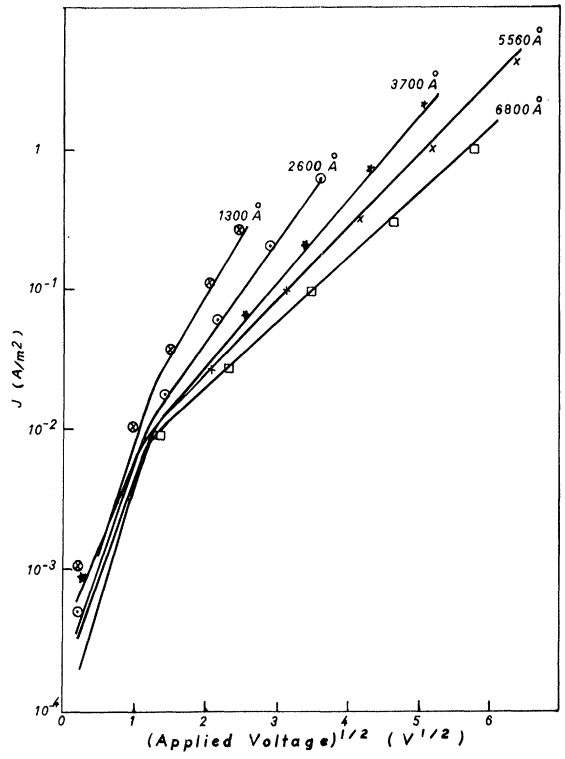

FIG. 7. *I-V* calculations from the theory (points) fitted to the data of Hartman *et al.*[10] on SiO (solid line) at different thicknesses. The parameters used in the calculations are shown in Table II.

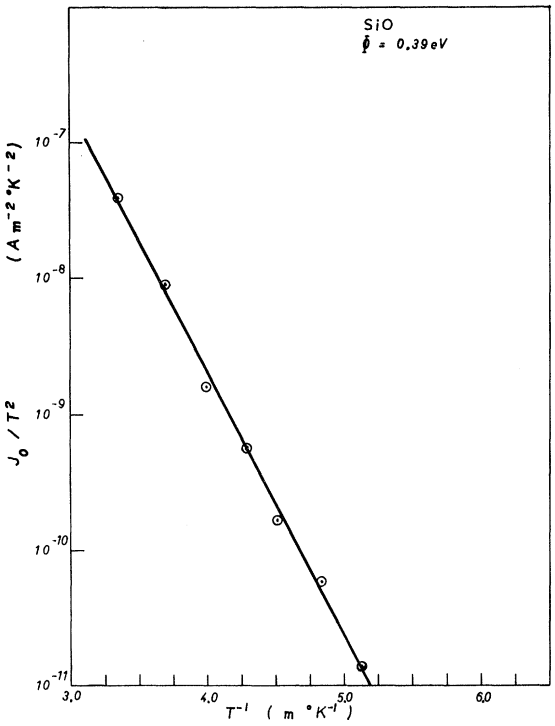

FIG. 8. Richardson plot obtained from the intercepts of the calculated curves of Fig. 6. The slope of the curve is 0.39 eV. Here J_0 is the zero-field current density.

Figure 8 is a Richardson plot obtained from the intercepts of the calculated curves of Fig. 6. Although the data were calculated using work functions in the range of 0.66—0.80 eV, the slope of the Richardson plot corresponds to a work function

TABLE II. Parameters needed to fit Hartman *et al.*[10] *I-V*-Thickness data for SiO.

Thickness (Å)	Φ (eV)	K	$\mu\theta$ (10^{-11} m^2 V^{-1} sec^{-1})	Temp (°K)
1300	0.82	3.25	1	297
2600	0.82	3.25	1	297
3700	0.808	3.25	1	297
5560	0.80	3.25	1	297
6800	0.80	3.25	1	297

of 0.39 eV. Since the calculations were performed with varying work function, it is apparently fortuitous that a straight line is obtained in the Richardson plot. The difference between the work functions employed and that obtained from the Richardson plot is not, therefore, significant. This only means that when the work function varies with temperature a Richardson-type analysis is not expected to give meaningful results. In Fig. 9 are plots of space-charge densities in SiO calculated as a function of distance from the cathode for several current densities. Note that again the space-charge density decreases with increasing current because of the Poole-Frenkel ionization of the traps.

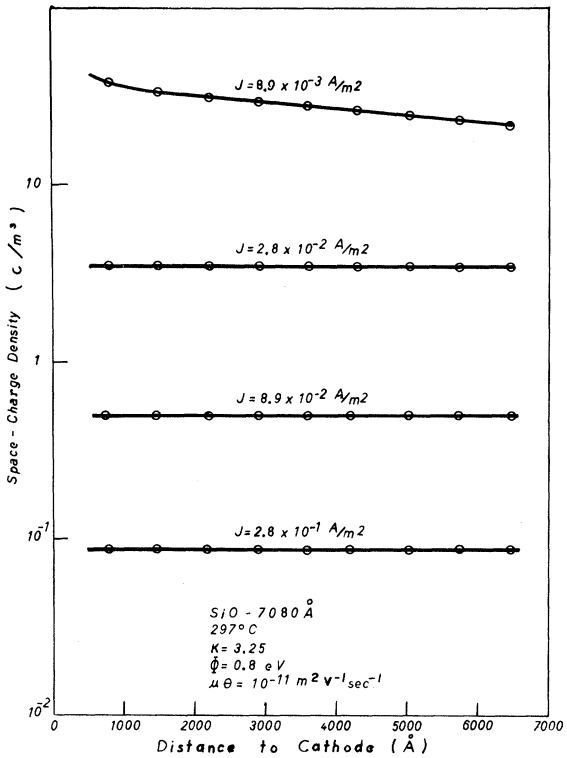

FIG. 9. Plots of space-charge density in SiO as a function of distance from the cathode for several current densities.

IV. CONCLUSION

It is clear that the present model of Schottky emission from the cathode coupled with Poole-Frenkel ionization of the traps in the bulk of the dielectric gives a very good description of current-voltage characteristics for Mylar, tantalum oxide, and silicon oxide. In regard to these data there are only a few points which will require future discussion. These are (a) the anomalous experimental results obtained[8] on the 10-mil Mylar films; (b) the nature and source of the deep traps in Mylar which are subject to the Poole-Frenkel effect; and (c) the temperature and thickness-dependent work function which was found to be necessary in explaining the silicon-oxide data.[10]

The choice between our emission-limited model and a bulk-limited model, as described by Simmons,[6] is obviously a hard one to make. Currently, we feel that the only sensible basis for a decision is detailed calculations such as the ones we have presented. Equally good, or better, results would have to be calculated by any other model before it can replace the one we employed for the systems considered.

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