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June 1973

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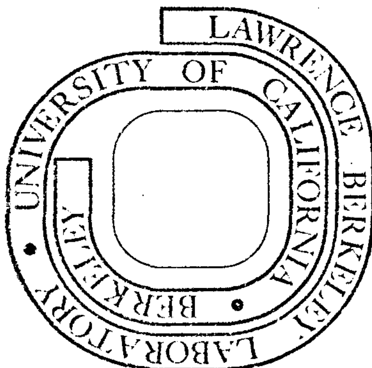

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CHARACTERIZATION OF METAL OXIDE TUNNEL JUNCTION BARRIERS

Duncan McBride, Gene Rochlin, and Paul Hansma*

June 1973

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Characterization of Metal Oxide Tunnel Junction Barriers

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ABSTRACT

A technique has been developed for determining unambiguously the barrier height ϕ and thickness S of the insulating layer in a metal-insulator-metal tunnel junction. The quantity $S\phi^{3/2}$ is measured from the slope of the Fowler-Nordheim plot. The experimental data are then compared with a family of calculated current-voltage curves by using various pairs of S and ϕ with $S\phi^{3/2}$ fixed at the measured value, in order to determine the pair which gives the best fit. The method has been tested on the best understood tunnel barrier, that grown thermally on an Al film, and is found to give good agreement with independent measurements. The tunnel barrier seems adequately described by a trapezoidal barrier model of nearly uniform height but with thickness variations on a microscopic scale over the junction area. The use of the method is illustrated on barriers grown thermally on Cr, thin

film V, and bulk V. We present a series of graphs at varying S and ϕ for six different values of $S\phi^{3/2}$ to facilitate the determination of approximate barrier parameters without lengthy computer calculations.

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I. INTRODUCTION

One of the outstanding problems in the study of electron tunneling is the characterization of the barrier region. This has become especially important in light of recent observations of collective excitations of the barrier region by inelastic electron tunneling.[1] Different groups, preparing their barriers by different techniques, need to be able to compare barrier parameters to discuss meaningfully the differences and similarities in observed inelastic tunneling spectra.

We have developed a technique for unambiguously obtaining these barrier parameters within the framework of a simple model. First the quantity $S\phi^{3/2}$, where S is the barrier thickness and ϕ is the barrier height, is experimentally determined from the slope of a Fowler-Nordheim plot[2] ($\log I/V^2$ vs $1/V$). With $S\phi^{3/2}$ fixed at this value, a family of current-voltage curves for different pairs of S and ϕ is plotted from a model calculation of the tunneling current through a one-dimensional trapezoidal barrier. Comparison of this family of curves with the experimental data allows an unambiguous separation of S and ϕ.

As a test of the method, we have applied it to the most extensively studied tunneling barrier: that grown thermally in air on an aluminum electrode. The method gives $\phi = 2.62 \pm .12$ eV, in agreement with recent independent measurements by other workers.[3] The effective barrier thickness S, of course varies with junction resistance, but the value of

20 Å for a 1300 Ω/cm^2 junction is also in agreement with measurements by other workers.[4] The value of the average barrier thickness obtained from capacitance measurements was then compared with S to obtain an estimate of microscopic barrier nonuniformity. This comparison shows that the standard deviation of the barrier thickness is approximately 5 Å.

We have also applied the method to three less well-studied tunneling barriers, those grown thermally on thin film Cr and on bulk and thin film V. Our results for the barrier heights are approximately 0.6 eV for Cr, 0.2 eV for bulk V, and 0.1 eV for thin film V, with barrier thicknesses in the 12-30 Å range. The differences between the values for barriers grown on bulk vs. thin film V will be discussed and illustrate the importance of knowing barrier parameters before comparing results on systems that might *a priori* be assumed to be identical.

For the convenience of experimentalists in obtaining a quick estimate of barrier parameters, we have plotted the family of S-ϕ curves for six values of $S\phi^{3/2}$. Although these curves cannot, of course, give as accurate results as the full calculation for the exact value of $S\phi^{3/2}$, they should be useful as a rough guide.

II. THE MODEL AND CALCULATION

The simplest useful model of a tunnel junction is that of two free-electron-gas metals separated by a thin insulating layer (Fig 1a).

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The potential barrier, formed by the forbidden gap of the insulator, is parameterized by its thickness S and height ϕ. Image force corrections are neglected. At nonzero bias (Fig. 1b) the Fermi levels of the metals are offset and a net tunneling current flows through the barrier. In order to make the model as simple as possible we show the barrier as rectangular at zero bias, even though most real barriers are probably trapezoidal. This point will be discussed further in Section V-C.

This simple model makes no pretense of describing the details of a real insulator a few atomic layers thick grown on a metal surface and cannot, of course, be expected to reproduce fine structure in the tunnel current. It may in fact introduce some artificial structure because of the discontinuous potential changes at the edges of the barrier. Nevertheless, this simple model will be shown to need only a minor modification--assumption of a microscopically nonuniform barrier with standard deviation of $\approx$ 5Å--to reproduce the current-voltage curves of real junctions over five orders of magnitude of tunnel current.

The details of this model have been worked out in other sources (e.g., ref. 5) and will not be reproduced here. For free electrons which tunnel independently, the tunnel current density through the barrier at zero temperature can be written as[6]

$$j = \frac{4\pi me}{h^3}\left[eV\int_0^{\varepsilon_f - eV} D(E_x)dE_x + \int_{\varepsilon_f - eV}^{\varepsilon_f} (\varepsilon_f - E_x)D(E_x)dE_x \right] \quad (1)$$

where ε_f is the Fermi level in the metal electrodes, $E_x = \hbar^2 k_x^2/2m$, and $D(E_x)$ is the probability of an electron with x-component of k-vector k_x tunneling through the barrier. $D(E_x)$ implicitly contains all the information about the model barrier. Gundlach and Simmons[7] have shown that the WKB approximation is an accurate approximation to the functional form of the tunneling probability for a trapezoidal barrier as long as the barrier is sufficiently thick and high ($S\phi^{1/2} \gtrsim 6$ Å$(eV)^{1/2}$). Using this further simplification, $D(E_x)$ can be expressed analytically for our model as:

$$D(E_x) = \begin{cases} \exp\left[-\frac{4}{3}\frac{(2m)^{1/2}S}{\hbar}\frac{(\varepsilon_f+\phi-E_x)^{3/2} - (\varepsilon_f+\phi-E_x-V)^{3/2}}{V}\right]; & E_x<\varepsilon_f+\phi-V \\ \exp\left[-\frac{4}{3}\frac{(2m)^{1/2}S}{\hbar}\frac{(\varepsilon_f+\phi-E_x)^{3/2}}{V}\right]; & E_x \geqslant \varepsilon_f+\phi-V \end{cases} \quad (2)$$

The integration of Eq. (1) with this expression for $D(E_x)$ was done numerically on a large digital computer to give the tunnel current density as a function of voltage.

III. EXPERIMENT

The Al, Cr, and thin film V junctions were fabricated by electron beam evaporation of high purity metals in an ion-pumped vacuum chamber at a pressure of approximately 10^{-7} torr. The bulk V junctions were fabricated on 2 gm ingots that were melted in a vacuum of 10^{-7} torr, then masked with GE 7031 insulating varnish. The Cr films were oxidized in air in an oven at 200°C for approximately 30 minutes. Both types of V samples were oxidized in air in an oven at 100°C for approxi-

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mately 20 minutes. The Al films were oxidized in air saturated with water vapor at room temperature for approximately 8 hours. The junctions were completed with evaporated second strips of Ag, Al, or Pb.

Junction area varied from 0.015 to 0.05 mm^2 on the same slide and, except for occasional shorted junctions, low-voltage junction resistance on a particular slide scaled with junction area. Junctions made with lead counter-electrodes had excess currents below the gap which were less than 0.5% of the normal state current, and the lead phonon structure was present with the proper magnitude[8] in junctions of low enough resistance so that this region could be investigated. There was, however, a problem in making V-I-Ag junctions with less than 50% excess currents. Thus, data from these samples is not included.

Current-voltage curves were graphed on an x-y recorder. Data were taken to as high a voltage as possible: that is, until the junction either shorted or changed its resistance irreversibly. The former failure, the more usual, is probably caused by junction heating. Therefore all data were taken with the samples immersed in liquid helium. In order to observe the superconducting characteristics of the Pb, V, and Al electrodes, most data were taken at about 1°K.

IV. ANALYSIS

In the limit of high bias ($V>>\phi$) Eq. (1) can be approximated by:

$$j \propto V^2 \exp\left\{-\frac{2}{3}\frac{\alpha S\phi^{3/2}}{eV}\right\} \qquad (3)$$

$$\alpha = \frac{2(2m)^{1/2}}{\hbar}$$

This is the well-known Fowler-Nordheim relation[2] and implies that a graph of $\ln(I/V^2)$ plotted against $1/V$ for a junction described by Eq. (1) is asymptotic at high biases to a straight line of slope $-\frac{2}{3}\alpha S\phi^{3/2}$. This number, experimentally determined for each junction, is the starting point for our analysis. A representative Fowler-Nordheim plot for an Al-I-Al junction is shown in Fig. 2; it results in a value for $\alpha S\phi^{3/2}$ of 81 eV.

Basavaiah, Eldridge, and Matisoo[9] have suggested that in Pb-PbO-Pb junctions it may be necessary to define a tunneling effective mass m* of approximately 0.5 m, where m is the free electron mass, in order to obtain agreement between the trapezoidal barrier model and experiment. While we cannot duplicate their experiment directly, we obtain a satisfactory picture of a junction using the free electron value which occurs naturally in our simple model, and we know of no compelling theoretical reason for assuming otherwise. With this value, α in practical units is 1.025 $Å^{-1}$ $(eV)^{-1/2}$, giving a value for $S\phi^{3/2}$ in the example above of 79 Å $(eV)^{3/2}$.

In this way we obtain an experimentally determined value for $S\phi^{3/2}$. However, we cannot yet determine either S or ϕ itself. In order to do this, a family of calculated current-voltage curves is produced from Eq. (1) using different values of S and ϕ for each,

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chosen so that the product $S\phi^{3/2}$ is kept constant. Part of the family for $S\phi^{3/2} = 79$ is shown in Fig. 3. Then the experimental data are compared with each member of the calculated family until one is found which most closely matches the data. Since a relatively small change in the barrier parameters assumed for the calculation results in a considerable change in both the shape and magnitude of the calculated curves, the fit is a sensitive one. Fig. 4 shows a comparison of experimental data and the calculated curve with which we obtained the best fit for $S\phi^{3/2} = 79$, giving S = 20 Å and ϕ = 2.50 eV. Fig. 5 shows the same data compared with the next member of the family, with S = 22 Å and ϕ = 2.35 eV. The fit of the latter is obviously much poorer, and it is shown merely to illustrate the sensitivity of the method.

In doing the fitting in Figs. 4 and 5, the vertical axis of the calculated curves has been relabeled by adding an arbitrary constant to the logarithm of the current so that the curves lie approximately on the experimental data. This is equivalent to allowing an arbitrary constant multiplying Eq. (1). It is therefore the shape of the calculated curve and not its absolute magnitude that we compare with the data. This is necessary because the absolute current through a tunnel junction apparently cannot be calculated adequately in the coherent-elastic-tunneling scheme used to derive Eq. (1), although it appears that Eq. (1) describes relative currents quite accurately.[10]

V. RESULTS

A. The Test Case: Al-I-Al

The fit shown in Fig. 4 for Al-I-Al is excellent over the whole range of tunnel current, and from it we can assign an effective tunneling thickness of 20 Å and an effective tunneling barrier height of 2.50 eV to this particular junction. We obtain similar fits for other Al-I-Al junctions with thicknesses in the range 12-22 Å and barrier heights of 2.50-2.75 eV. All these measurements were made with oxidized electrode biased negative. In the same way one can obtain tunneling thicknesses and barrier heights for junctions with other barrier materials.

1. Barrier thickness. The parameters S and ϕ which we determine can of course be measured in other ways. A measure of the barrier thickness can most simply be obtained by measuring the capacitance of the junction and its area. By assuming a value for the dielectric constant, a thickness can then be derived. The film thickness can also be measured using ellipsometry, with identical results,[11] although we have not performed these measurements ourselves. Capacitance and ellipsometry measurements always give a measure of the barrier thickness greater than the thickness determined from tunneling measurements. For example, capacitance measurements on the Al-I-Al junction discussed above, together with an assumed static dielectric constant of 8.8[12] yield a barrier thickness of 33 Å.

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This discrepancy leads us to a refinement in our model for the barrier. In both the tunneling and capacitance calculations we have assumed a plane parallel geometry for the junction. This is certainly not the case in a real junction. The grown oxide does not cover the oxidized electrode in a layer uniform on the scale of a few angstroms, and thus there are some areas which are thinner and some which are thicker.

Using the Josephson interference pattern, Dynes and Fulton[13] have shown that current flow is distributed almost uniformly over the area of a thermally oxidized Sn-I-Sn junction. It is likely that our junctions, oxidized in a similar way, have a similarly uniform current density distribution over the junction area. However, the Fourier transform technique which Dynes and Fulton used is limited in resolution by the number of lobes of the Josephson interference pattern which can be measured. Their measurements effectively averaged the current density over strips of about 0.01 mm by 0.2 mm on a junction 0.2 mm square. Thus their experiment could not resolve a variation in barrier thickness which occurred over distances much smaller than .01 mm.

In fact, there must be some variation in the thickness of the oxide layer, since thermal growth certainly cannot produce an oxide a few atomic layers thick which is uniform in thickness to within a single atomic layer. This variation apparently occurs on a scale small enough so that no macroscopic region of the junction carries a predomi-

nently larger current than any other. The sample can be thought of schematically as a set of microscopic tunnel junctions, with the same barrier height but with barrier thickness distributed about a mean, which are connected in parallel and arranged randomly over the actual junction region.

This distribution of thicknesses affects the tunneling current and the junction capacitance in different ways, because of their different dependence on thickness. The capacitance varies as 1/S, while the tunneling current very crudely can be approximated by an exponential e^{-S/S_o}, where S_o (although voltage dependent) is a characteristic length on the order of 1 Å. Therefore, a distribution of thicknesses has a much larger effect on the tunneling current than on the capacitance of the junction, and this effect tends to make the apparent tunneling thickness smaller than the capacitance thickness. In fact, if we assume a Gaussian distribution of thicknesses around a mean,[14] the current-voltage curve calculated above for a 20 Å uniform barrier can be closely approximated by that for a barrier with a mean thickness of 33 Å and a standard deviation of about 5 Å, while the capacitance is hardly affected by this distribution (3% increase).[15]

Therefore, we conclude that a thermally oxidized tunneling barrier is microscopically nonuniform and that the capacitatively determined thickness is to a good approximation the mean thickness of the insulating barrier. The thickness derived from tunneling measurements is,

however, the thickness which must be used to determine a barrier height from a Fowler-Nordheim value of $S\phi^{3/2}$, not the capacitative thickness, which results in a considerable underestimate of ϕ.

2. Barrier height. The most convincing independent measurement of barrier height is obtained from photoemission over the barrier.[16] However, attempts to observe a photoresponse in our junctions have been unsuccessful. Gundlach and Hölzl[17] have measured the barrier height in thermally oxidized aluminum oxide barriers and obtain, for the oxidized electrode negative, about 2.4 volts from photoresponse data and about 2.6 volts from measurement of the logarithmic derivative of the I-V curve. Chang, Stiles, and Esaki[18] report a value of about 2.3 volts based on the temperature dependence of the tunneling current. These are sufficiently close to the values we obtain to give us confidence in our procedure.

Further, in Fig. 1 we show the Fermi level halfway between the valence and conduction bands in the insulator. This is true for an insulator without impurities in our simple model and predicts that the tunneling barrier is just one-half the optical band gap in the insulator. While nothing is so simple in a real junction, there seems to be a remarkably close correspondence in the aluminum oxide barrier. Our values of ϕ lie between 2.5 and 2.75 volts, while half the band gap in amorphous Al_2O_3 is between 2.5 and 3 volts.[19,20]

B. Other Materials: Cr and V

We have applied the model developed here to barriers grown on thin film Cr and on thin film and bulk V. Representative values for $S\phi^{3/2}$ are 5.9 for thin film Cr, 0.52 for thin film V, and 2.75 for bulk V, and Fig. 6 shows data from three junctions and calculated curves for these values. In the case of bulk V (Fig. 6c) an excellent fit is obtained for a thickness parameter of 25 Å and a barrier height of 0.23 eV. For thin film Cr and V, the fit cannot be made as precisely (probably due to the inadequacy of the model), and three calculated curves have been drawn, bracketing the data. The middle curve in each case seems the most reasonable, and results in a barrier thickness and height of 13 Å and 0.59 eV for Cr and 17 Å and 0.097 eV for thin film V.

Note that the barrier is much lower for our thin film V than the bulk V. This is probably due to the large amounts of dissolved oxygen in the V films and the complex chemistry of the vanadium-oxygen system. This clearly illustrates that tunneling junctions prepared by different techniques may have quite different parameters due to variations either in the barrier material or the nature of its contact with the electrodes.

C. Discussion

The results we have shown have been obtained for particular junctions with the oxidized electrode negative in each case. However, from the analysis of many samples it is possible to draw some more

general conclusions about the barriers of the tunnel junctions we have made.

First, the barrier height we obtain is practically constant for a given barrier grown in the same way on the same material, and therefore we conclude that we are measuring an intrinsic property of the material. Our range of measurements has been 2.50-2.75 eV for Al, 0.52-0.60 eV for Cr, 0.20-0.25 eV for bulk V, and 0.090-0.110 eV for thin film V. The barrier thickness, on the other hand, varies considerably from sample to sample, and its value means little except for the individual sample.

Second, when data are used from the opposite bias polarity, that with the oxidized electrode positive, the thickness parameter obtained is almost exactly the same, but the barrier height obtained is sometimes different. In the case of Al-I-Al junctions, it corresponds to that obtained previously (e.g., ref. 3), roughly 2 eV, for the oxidized electrode positive. Cr-I-Pb junctions show a similar asymmetry, but in both bulk and thin film V-I-Pb junctions the current-voltage curves are virtually symmetric in bias polarity, and this results in the same measure of barrier height. Thus, although the model we use is exact only for a symmetric barrier, it seems to give reasonable estimates of the barrier heights of asymmetric barriers as well.

Finally we wish to note our mixed results in checking the measurements of the barrier heights made here with those determined by the method of observing a peak in the logarithmic derivative $\frac{d(\ln I)}{dV}$ of the current-voltage curves.[3] Our calculations show that the peak depends sensitively on the shape chosen for the barrier: It can be either above or below the actual barrier height by as much as 20%, and for some barrier shapes it does not occur. In the case of our aluminum samples, the logarithmic derivative rises without peaking to our highest bias. For the model we use and the junction shown in Fig. 4, we estimate the peak would occur at about 3 volts. However, the junction would then be dissipating more than 200 watts/cm^2. In the thin film Cr and V samples, the logarithmic derivative decreases monotonically, even though we have data to more than twice the barrier height. Only in the junctions made on bulk V do we see a peak, and then only on some samples. For the sample shown in Fig. 6c, we obtain a peak at 0.2 eV, in good agreement with our measurements.

D. Graphs for Various Values of $S\phi^{3/2}$

Fig. 7a-f show families of curves for various S-ϕ pairs for six different values of $S\phi^{3/2}$. We present these as a rough guide for determining barrier parameters when more exact information is unavailable or unnecessary. For each value of $S\phi^{3/2}$, the family includes more than the full range of barrier thicknesses which might reasonably be expected in oxidized barriers with enough values so that interpolation is

possible.

Because the voltage (horizontal) scale is different for the different members of Fig. 7, they bear a superficial resemblance to each other. However, curve shapes are actually quite different between the various families, and we have been able to find no means of parameterizing the curves into one universal family.

VI. CONCLUSION

Our general method is to determine the combination of parameters $S\phi^{3/2}$ for a particular junction from a Fowler-Nordheim plot of the experimental data. We then perform a model calculation using several values of S and ϕ with the product $S\phi^{3/2}$ held equal to the experimentally determined one. Experimental data are compared to the calculated curves and barrier parameters are assigned based on the curve to which the data corresponds most closely.

We have tested the method with an Al-I-Al junction and one bias polarity. The method is general, however, and does not seem to be affected appreciably by a barrier whose height is not the same on both sides at zero bias. That is, barrier thickness determined seems to be independent of polarity and the barrier height seems to correspond to those determined by others for the corresponding polarity, even though our calculation is done for a symmetric barrier model. Although our model is simple, we obtain fits to data which are both excellent over a large range and highly sensitive to the choice of barrier parameters.[21]

Further, from our analysis, we believe this simple model of an insulating tunnel barrier to be as detailed as is necessary to consider from an analysis based on macroscopic properties (tunnel current and capacitance). The trapezoidal model seems to be a reasonably adequate description of the barrier. The barrier heights on the two sides of the barrier are often unequal, but the barrier height does not appear to vary over the junction area. The barrier thickness is not uniform, but the variation, which is on a microscopic scale, can be approximated, at least for the purposes of calculation, by a Gaussian distribution with a standard deviation which is a small fraction of the mean.

We illustrate the use of the method with three other types of tunneling barriers: those grown thermally on thin film Cr, thin film V, and bulk V. Finally, we have plotted a series of curves with different values of $S\phi^{3/2}$ for use by others in estimating barrier parameters without doing lengthy computer calculations.

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FIGURE CAPTIONS

Fig. 1a. A simple model of a potential barrier of height ϕ and thickness S formed between two metals by the presence of an insulating layer.

b. When a voltage V is applied between the two metals the Fermi levels are offset by eV and a net tunneling current flows.

Fig. 2. A Fowler-Nordheim plot for an Al-I-Al junction. Since $I \alpha V^2 \exp\left(-\frac{4(2m)^{1/2}}{3\hbar}\frac{S\phi^{3/2}}{eV}\right)$ for large V, the data points asymptotically approach a straight line toward the left of the graph. The slope of this straight line gives $S\phi^{3/2} = 79$ Å $(eV)^{3/2}$.

Fig. 3. A family of curves for various values of S with the product $S\phi^{3/2}$ fixed at 79 Å $(eV)^{3/2}$. Note that the shape of the curves is a sensitive function of S, permitting unambiguous determinations by comparison to the experimental data.

Fig. 4. The best fit values of S and ϕ for an Al-I-Al junction with $S\phi^{3/2} = 79$ Å $(eV)^{3/2}$. The agreement is excellent over five orders of magnitude of current. The slight hump in the data points near V = 1 volt is not understood.

Fig. 5. The data points compared with the computer generated curve for S 10% away from the best fit value shown in Fig. 4. The magnitude of disagreement illustrates the sensitivity of this technique for determining barrier parameters.

Fig. 6. Experimental data and computer generated curves for tunneling barriers grown on (a) thin film Cr, (b) thin film V, and (c) bulk V. Though the fits in (a) and (b) are not as good as in the case of the Al junctions, the barrier parameters can be bracketed as shown.

Fig. 7a. Computer generated curves for $S\phi^{3/2} = 2.0$. The curves in (a) through (f) can be used for quickly approximating barrier parameters of a new material.

7b. Curves for approximating barrier parameters for a junction with $S\phi^{3/2}$ near 4.0.

7c. Curves for approximating barrier parameters for a junction with $S\phi^{3/2}$ near 8.0.

7d. Curves for approximating barrier parameters for a junction with $S\phi^{3/2}$ near 16.

7e. Curves for approximating barrier parameters for a junction with $S\phi^{3/2}$ near 32.

7f. Curves for approximating barrier parameters for a junction with $S\phi^{3/2}$ near 64.

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21. Unfortunately, this sensitivity occurs only at high bias voltage, and our method shares with all other tunneling methods for determining the barrier height the practical difficulty of requiring experimental data to biases greater than the barrier height.

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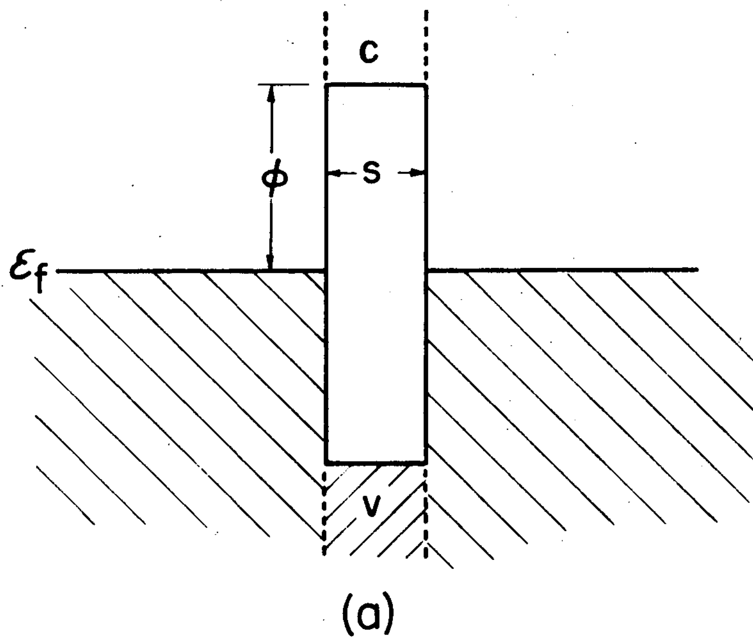
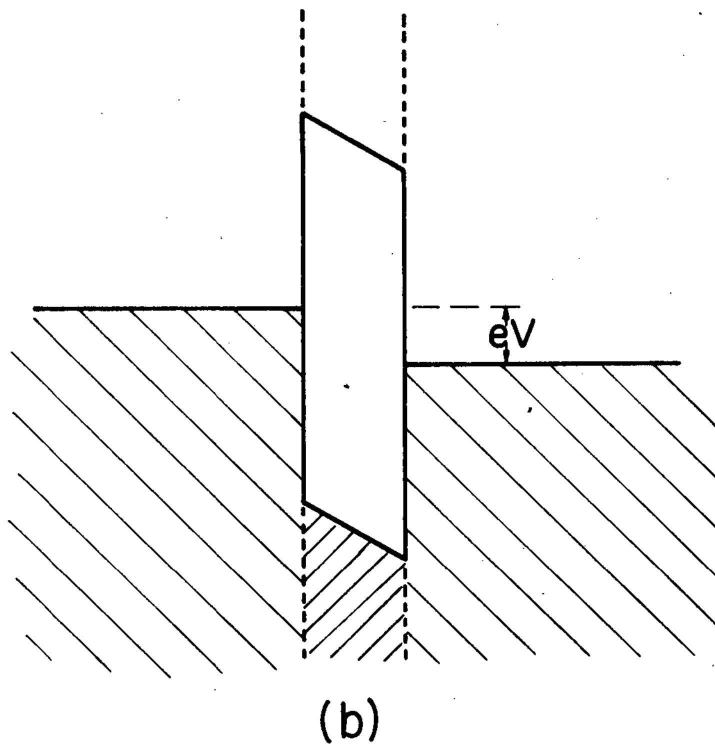
However, we have been able to make satisfactory parameterizations for some junctions for which we could not reach a high enough bias to observe a peak in the logarithmic derivative of the IV curve. The voltage difference is small in these cases; the current difference is enormous.

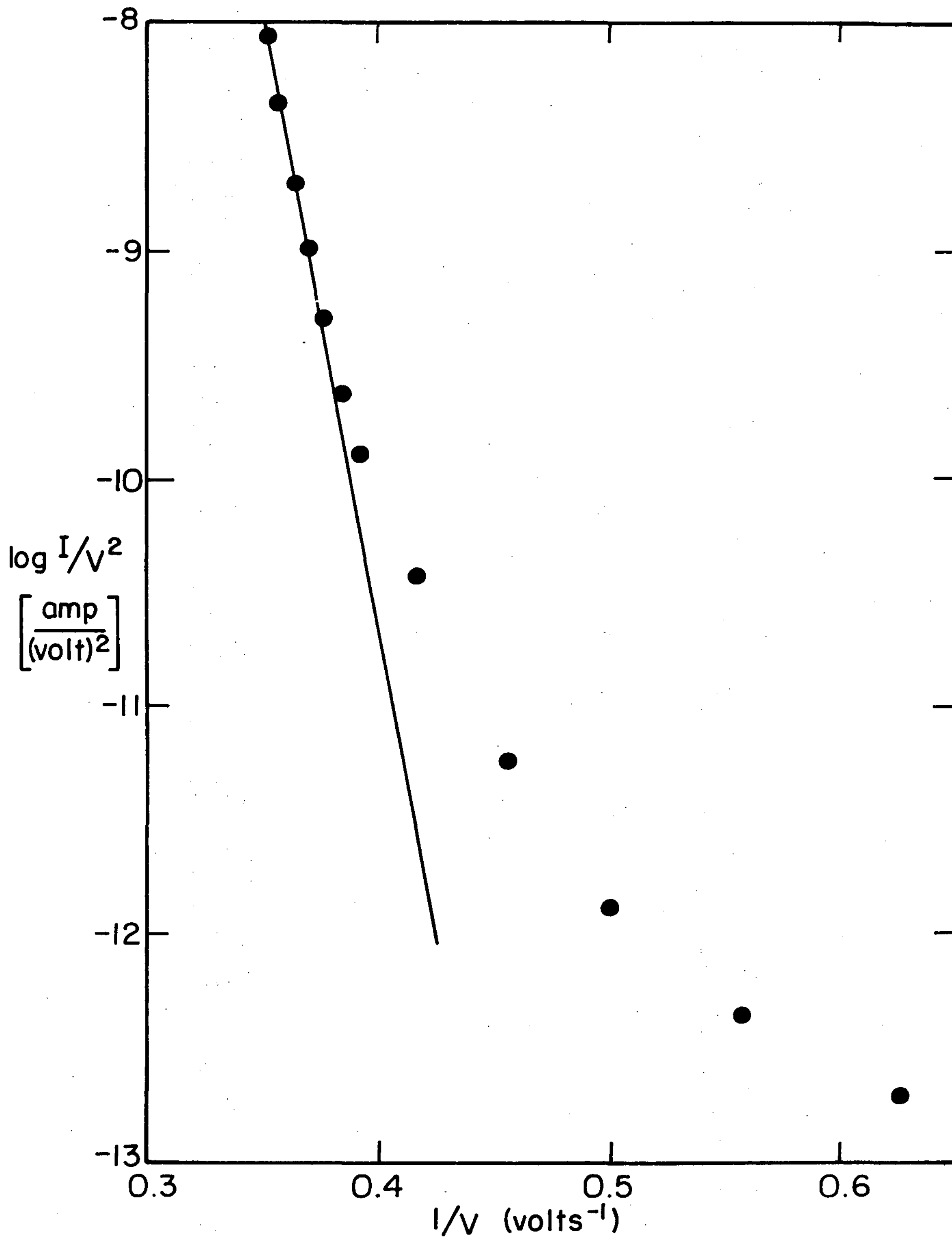




XBL733-5840

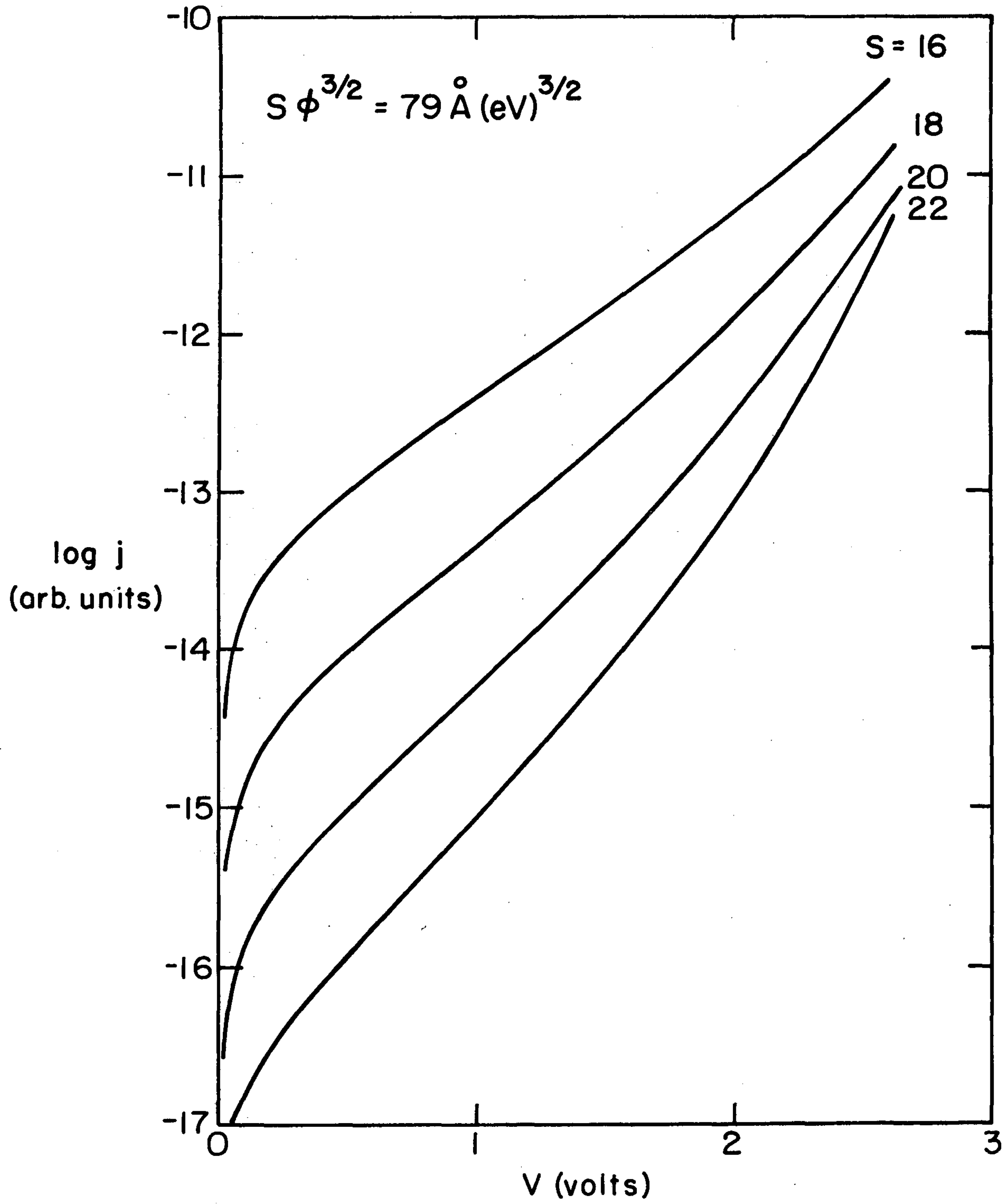
Fig. 1

0 0 0 0 3 9 0 0 9 4 3



XBL 733-5841

Fig. 2



XBL 733-5842

Fig. 3

0 0 0 0 3 9 0 0 9 4 6

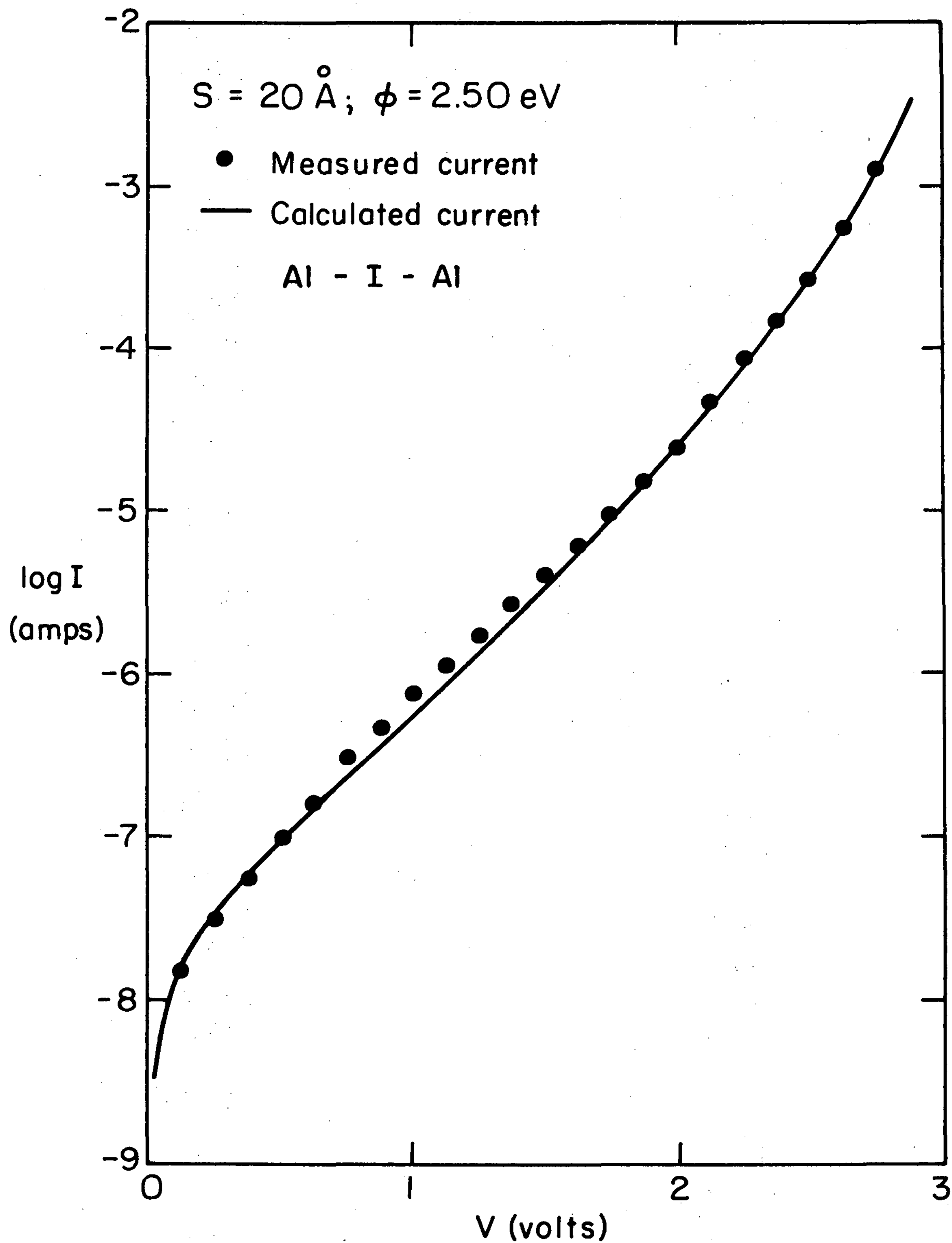
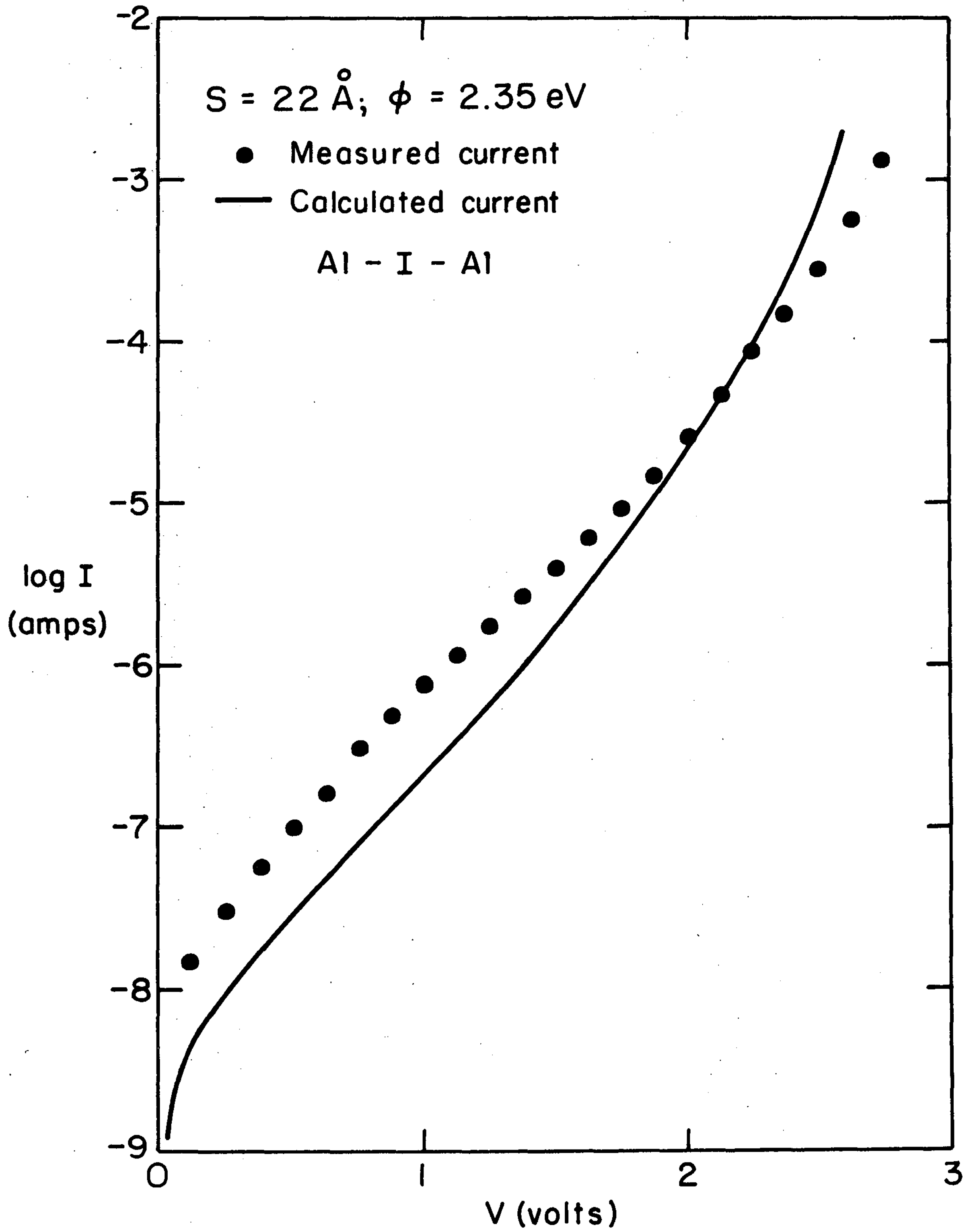


Fig. 4



XBL733-5844

Fig. 5

0 0 0 0 3 9 0 0 9 4 7

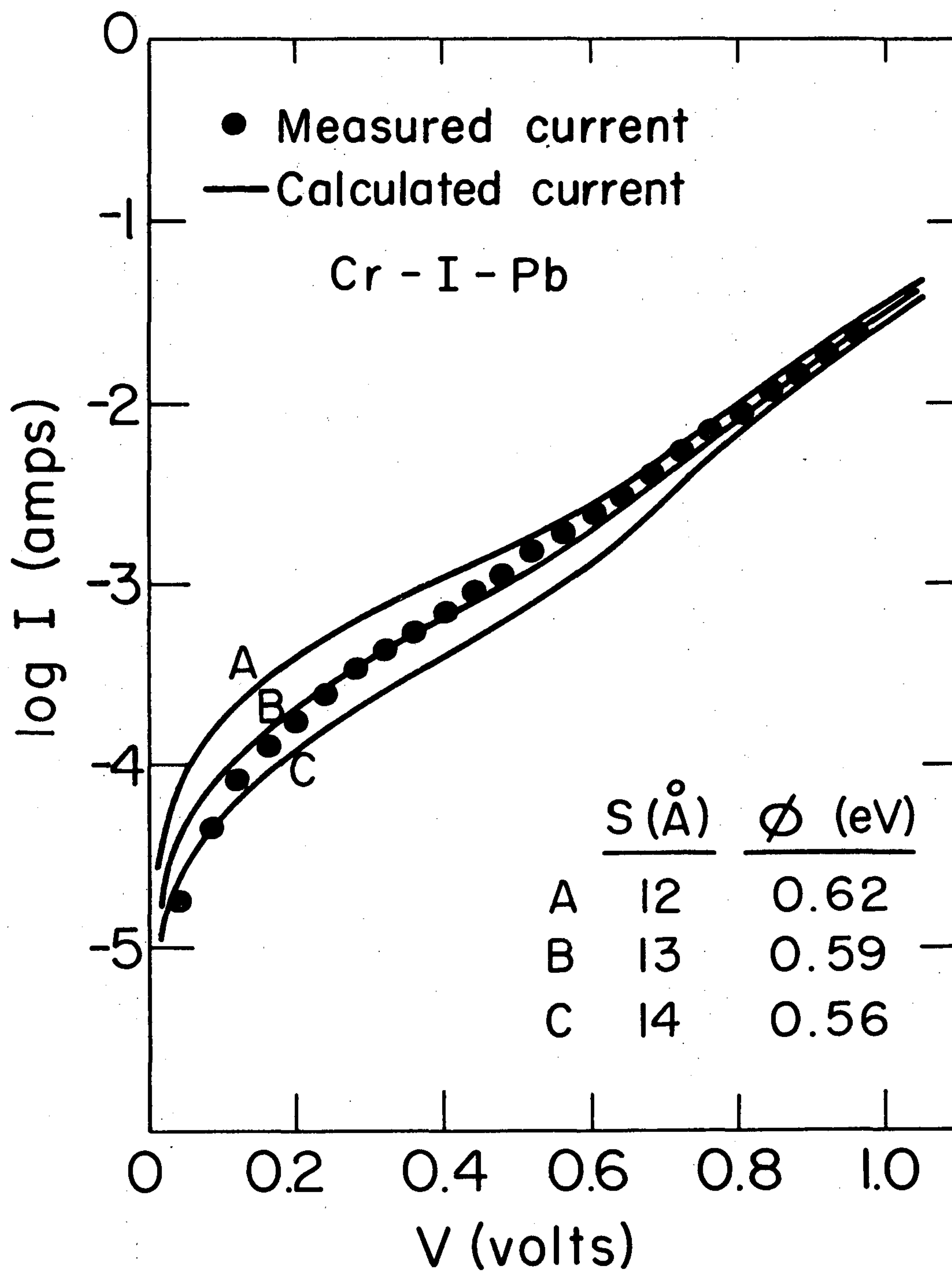
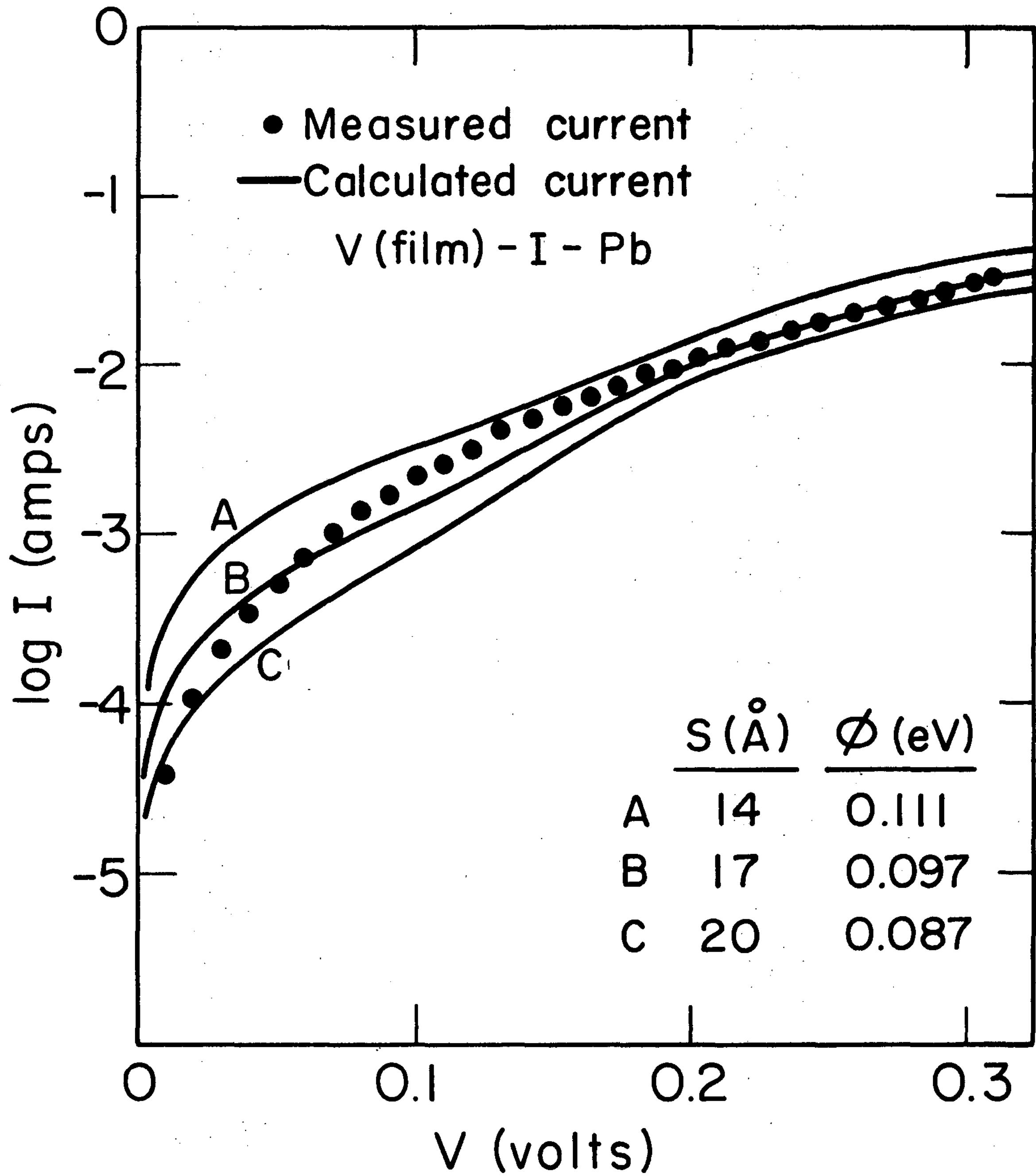


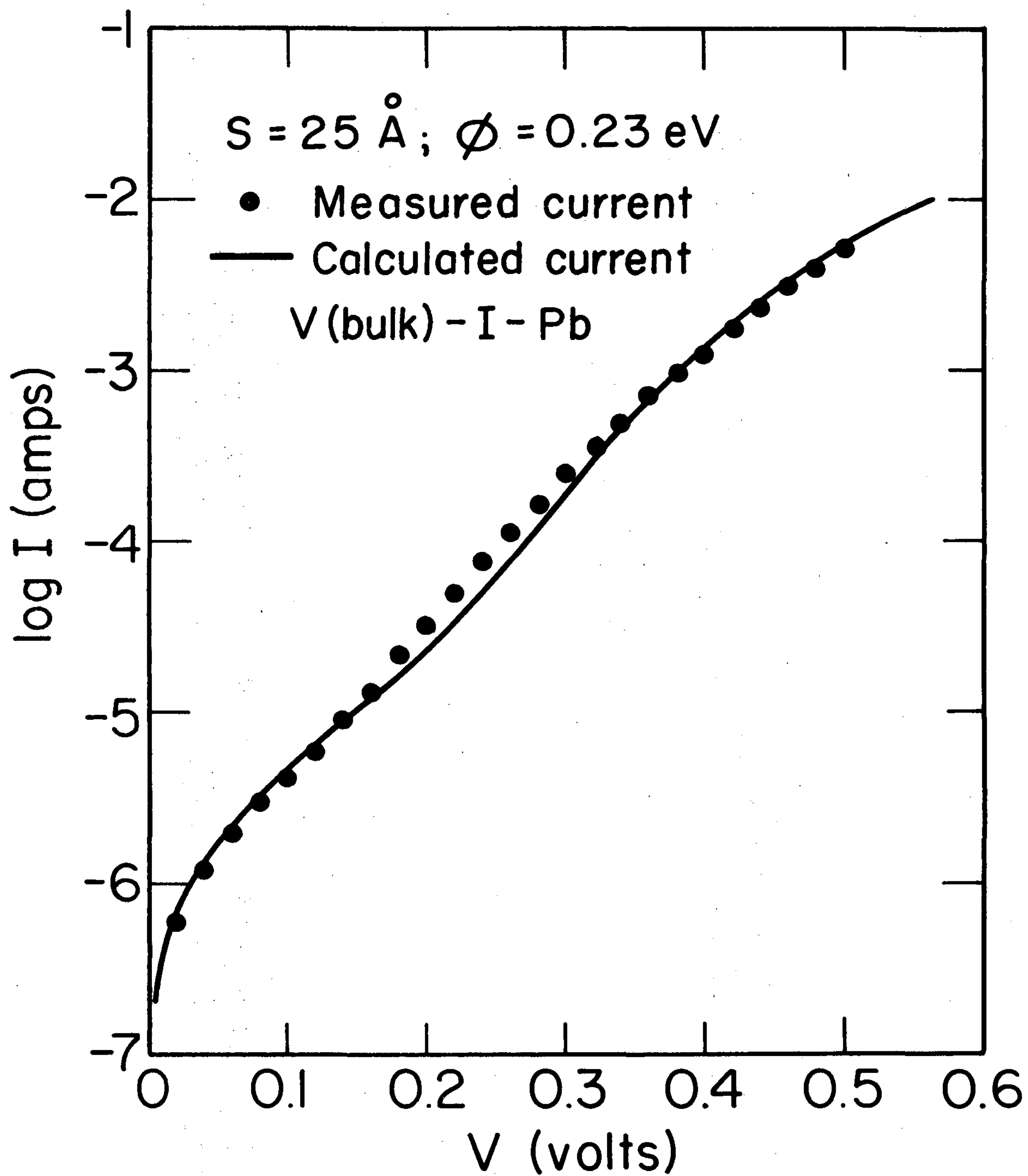
Fig. 6a



XBL 736-6341

Fig. 6b

0 0 0 0 3 9 0 0 9 4 8



XBL 736-6342

Fig. 6c

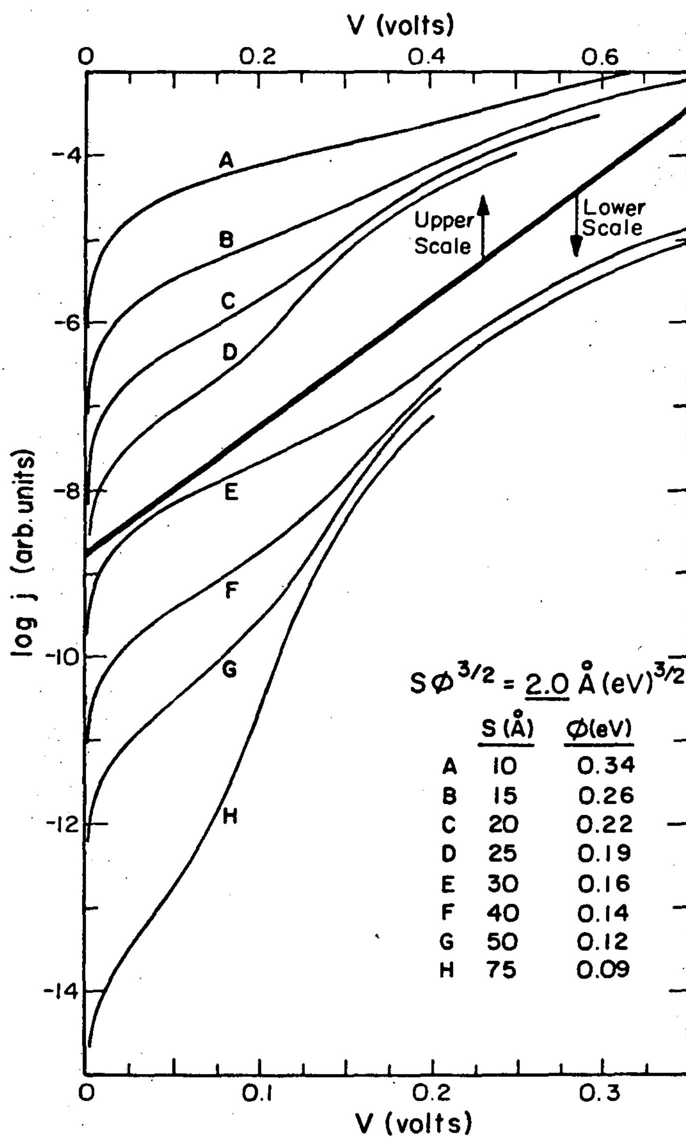


Fig. 7a

0 0 0 0 3 9 0 0 9 4 9

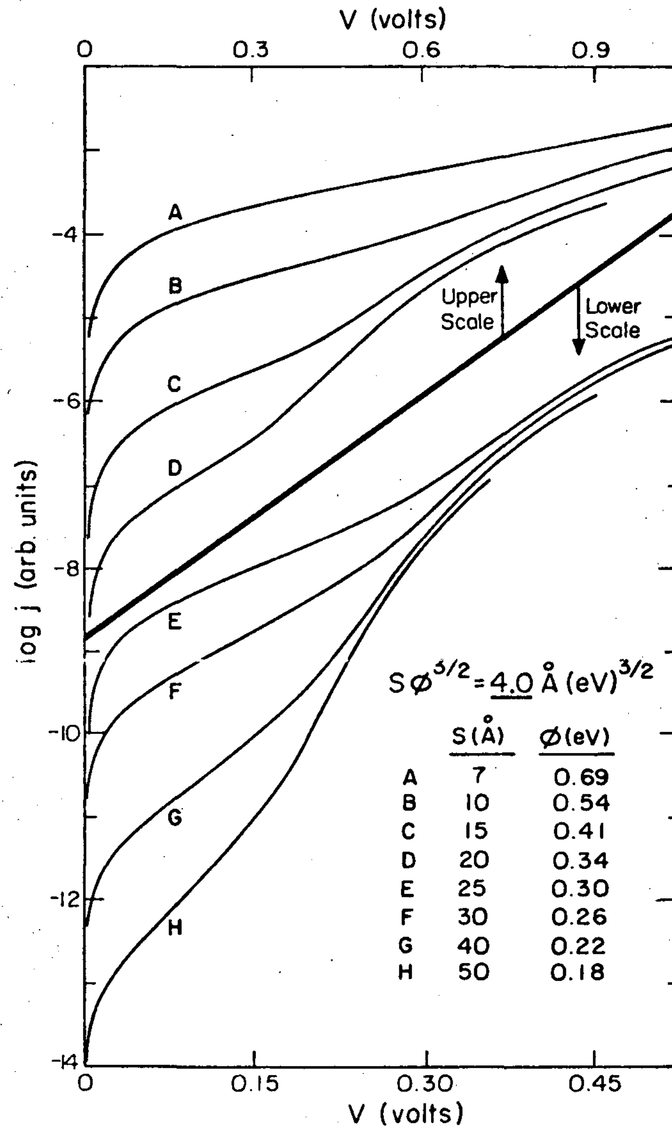


Fig. 7b

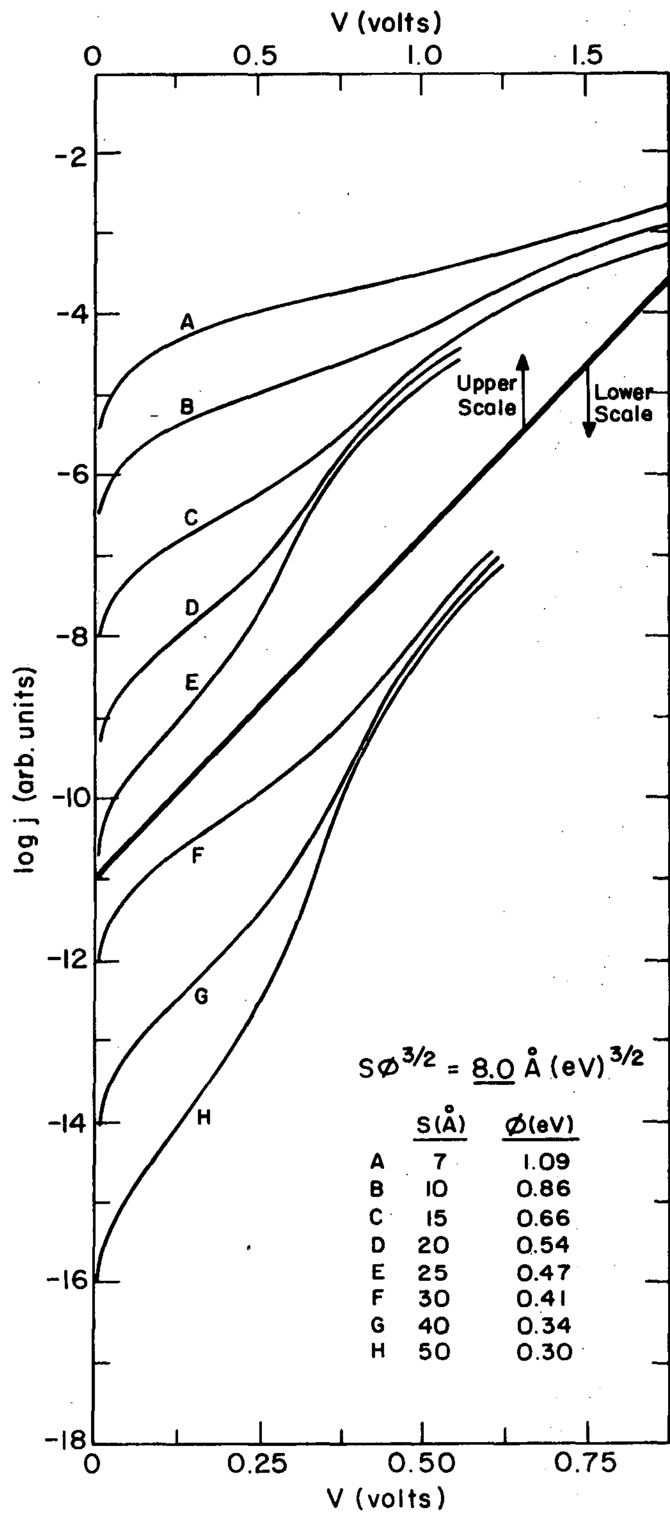


Fig. 7c

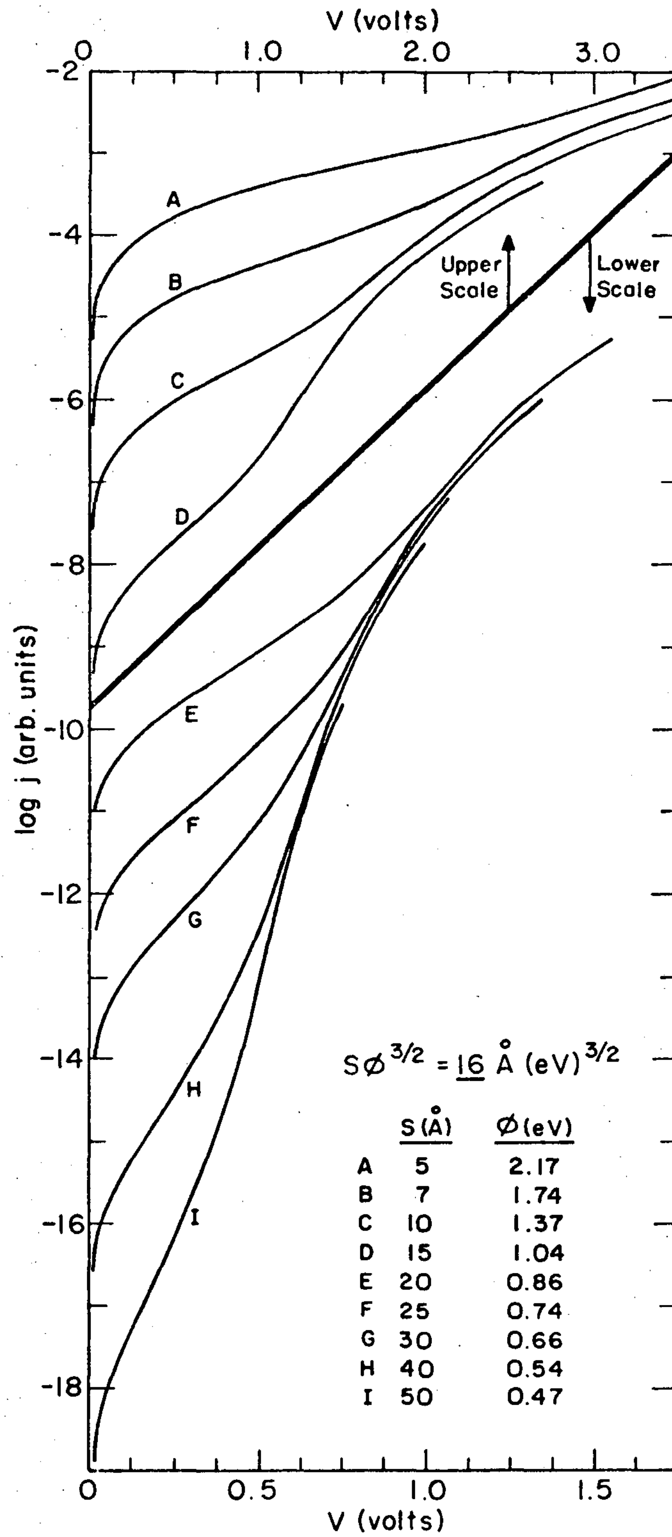


Fig. 7d

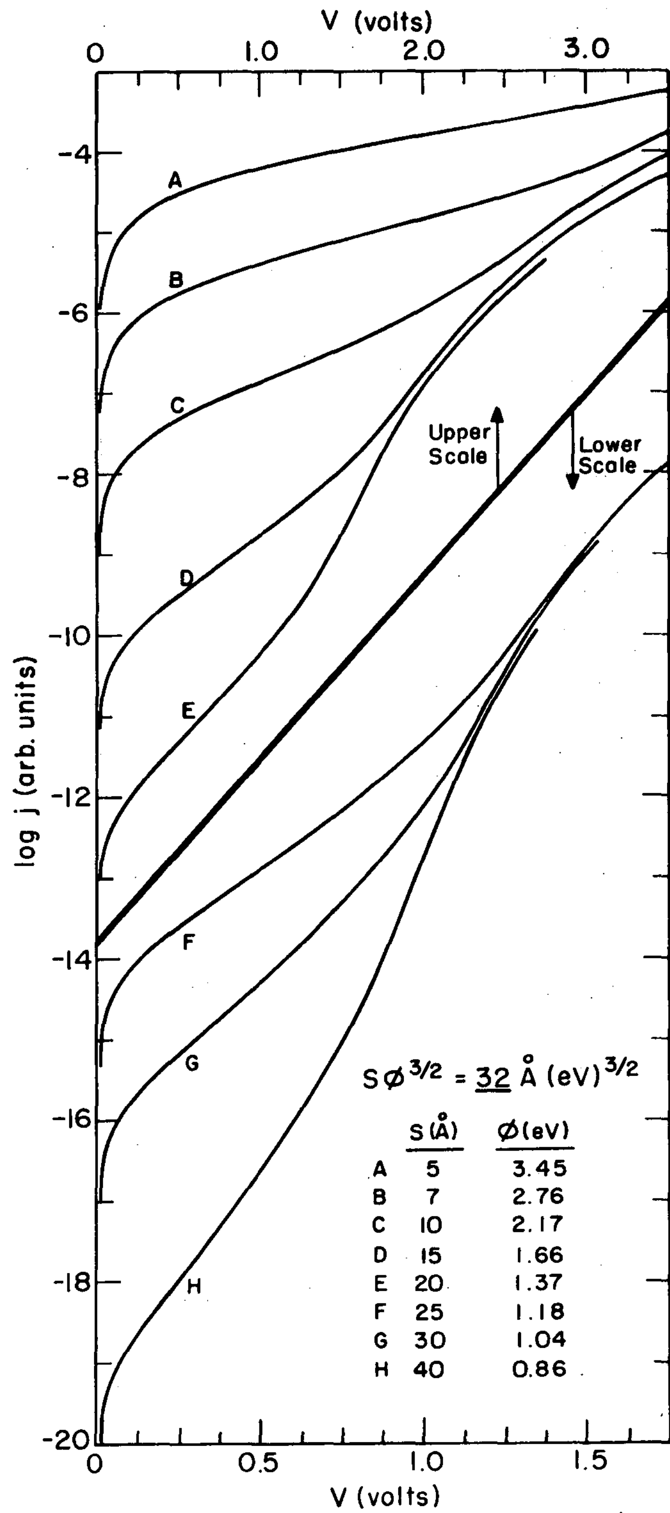


Fig. 7e

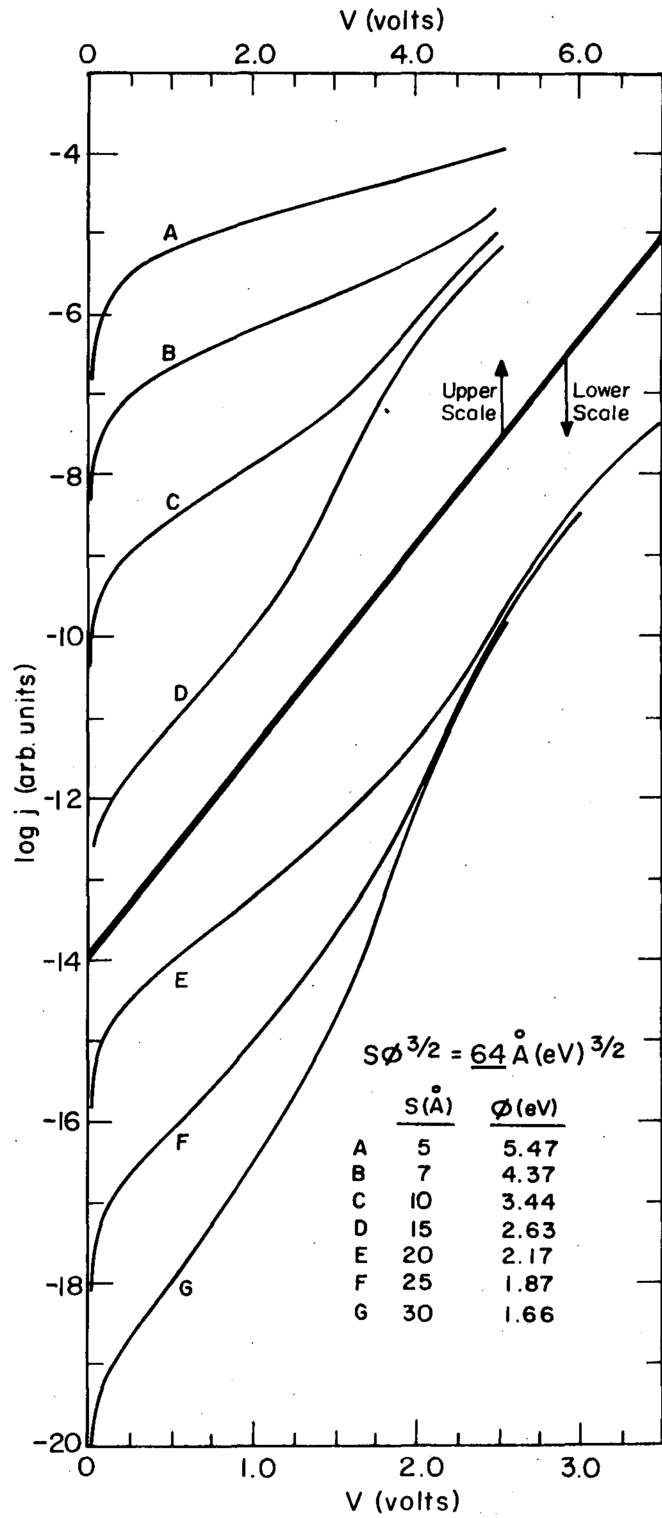


Fig. 7f

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