Electron Interaction Cross Sections in Al and Al_2O_3; Calculations of Mean Free Paths, Stopping Powers, and Electron Slowing-Down Spectra*

J. C. Ashley, C. J. Tung and R. H. Ritchie**
Health Physics Division, Oak Ridge National Laboratory, Oak Ridge, TN 37830

INTRODUCTION

The deposition of energy in materials due to the presence of an ionizing radiation field continues to be an important area of study for communication and space applications. The primary electrons generated lose energy mainly by inelastic collisions with other electrons in the medium resulting in an energy distribution, or slowing-down spectrum, ranging from the primary electron energy down to essentially thermal energy.

We have been interested for many years in a study of slowing-down spectra and interaction cross sections for many materials, mainly metals and alloys. More recently the work has been extended to include semiconductors[1] and insulators.[2] An area of concern has been the lack of adequate theoretical information on electron interaction probabilities in solids, particularly in the electron energy region below a few keV where experimental measurements of these quantities are rather difficult. We have developed models to calculate differential interaction cross sections and derived quantities such as inelastic mean free paths and

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** On temporary leave to Churchill College, Cambridge University, Cambridge CB3 ODS, England.

stopping powers for this low energy region.[3,4] We will discuss here new calculations of cross sections for ionization by electrons of inner shells in Al and O atoms and a model insulator theory used to describe the valence band in Al_2O_3. These new calculations have been combined with earlier work to provide an improved description of electron interactions in Al metal and in the insulator Al_2O_3. Some examples of mean free path, stopping power, and electron slowing-down flux calculations for these two materials will be described.

INNER SHELL CROSS SECTION CALCULATIONS

To treat the excitation of electrons from the inner shells of atoms in solids we make two main assumptions: (a) that the binding energies of the tightly bound inner shells are not changed substantially from the values appropriate for isolated atoms, and (b) transitions occur only between the particular bound level and states in the continuum. With these assumptions we may take advantage of atomic oscillator strength calculations to derive differential inverse mean free paths (DIMFP's) by simply multiplying the cross section per atom for the given process by the density of atoms in the solid. In first Born approximation, the differential cross section for excitation of an electron from the subshell $n\ell$ to a continuum energy level between ε and $\varepsilon + d\varepsilon$ by an incident electron of energy E is given by

$$\frac{d\sigma_{n\ell}}{d\varepsilon} = \frac{4\pi a_o^2}{(E/R)(\Delta E/R)} \int_{(ka_o)_{min}}^{(ka_o)_{max}} f_{n\ell}(k,\varepsilon)\, d\ln(ka_o)^2 \qquad (1)$$

where $a_o \equiv \hbar^2/me^2$ is the Bohr radius, $R \equiv e^2/2a_o = 13.6$ eV is the Rydberg energy, and $\hbar\vec{k}$ is the momentum transfer. Here $f_{n\ell}(k,\varepsilon)$ is the

generalized oscillator strength (GOS) for the transition, the energy loss of the incident electron is given by $\Delta E = \varepsilon + |\varepsilon^B_{n\ell}|$ where $\varepsilon^B_{n\ell}$ is the binding energy of the $n\ell$ subshell, and the limits on the integration are given from energy and momentum conservation by

$$(ka_o)^2_{\substack{max \\ min}} = \{2E - \Delta E \pm 2\,[E(E-\Delta E)]^{1/2}\}/\mathcal{R} \,. \qquad (2)$$

Generalized oscillator strengths for the ionization of L shell electrons in Al have been calculated by Manson[5] using a nonrelativistic Hartree-Slater central field model of the atom for ionized-electron energies from threshold to 128 $\mathcal{R}$. These values have been used as input for numerical evaluation of the integral over momentum transfer in Eq. (1) to obtain differential cross sections for the 2s and 2p subshells of Al. As an example of these calculations we show in Fig. 1 the differential cross section for the transition of a 2p electron from an Al atom to an energy level ε in the continuum due to incident electrons of various energies. The energy loss of the incident electron is $\Delta E = \varepsilon + 5.947\mathcal{R}$. In Fig. 2 we show the DIMFP $d\mu/d\omega$ for incident electrons of several energies for removing an electron from the 2s subshell in Al, as a function of energy transfer ω. The density of Al atoms corresponds to that in Al_2O_3 and all quantities are expressed in atomic units (e.g., energies in units of $2\mathcal{R}$ = 27.2 eV). Here ω (atomic units) = $(\varepsilon + 8.715\mathcal{R})/2\mathcal{R}$.

Similar calculations have been done for the 1s shell in oxygen. The values of the GOS for excitation to the continuum from the 1s shell of O were taken from the work of McGuire.[6] Generalized oscillator

strengths for the K shell in Al are not available using the Hartree-Slater method used by Manson[5] or McGuire[6] partly due to the length of the computer calculations. However, it is widely accepted that for the K shell of Al, hydrogenic calculations should be quite good. Consequently, we have used the GOS calculated using hydrogenic wave functions[7] to determine the cross section for excitation of electrons from the K-shell of Al to the continuum.

INTERACTION OF ELECTRONS WITH Al METAL

For the interaction of electrons with Al metal the inner shells are treated as described above while the conduction band electrons are described as a group by the electron gas model. The details of the electron gas model and the usefulness of this model in the description of the interaction of charged particles with real metals have been discussed elsewhere.[3,4] The response of the electron gas to a given energy and momentum transfer is described by the dielectric response function $\epsilon(k,\omega)$.[8] The DIMFP is then derived from the equation

$$\frac{d\mu}{d\omega} = \frac{2}{\pi v^2} \int_{v-\sqrt{v^2-2\omega}}^{v+\sqrt{v^2+2\omega}} \frac{dk}{k} \operatorname{Im}\left(\frac{-1}{\epsilon_{k,\omega}}\right) \qquad (3)$$

where v is the speed of the incident electron and all quantities are measured in atomic units. For our purposes here we note that the interaction of an electron with an electron gas divides naturally into two parts: (a) incident electron-single electron interactions and (b) interaction of the incident electron with the collective modes of the electron gas, i.e., plasmon excitation. A DIMFP for each of these two processes may thus be calculated from the electron gas model.

The set of DIMFP's for the various interactions between an incident electron and the given medium forms the basic information for the calculation of mean free path, stopping power, electron slowing-down spectra, etc. If we integrate the DIMFP over allowed energy transfers we get the inverse mean free path. In Fig. 3 we show these results for Al. The inverse mean free paths (in units of "per Angstrom") are shown for removing electrons from the inner shells of Al and for electron-electron interaction and plasmon excitation as functions of incident electron energy. Note that the main contribution to the mean free path of electrons in Al for this energy range is due to interaction with the conduction band electrons.

The contribution to the stopping power for a particular process is obtained by multiplying the DIMFP for the process by the energy transfer and integrating over allowed energy transfers. In Fig. 4 we show the contributions to the stopping power of Al for electrons from interactions with the 1s, 2s, 2p shells and due to plasmon excitation and electron-electron interactions in the conduction band. The total stopping power agrees quite well at 10 keV with published stopping power calculations based on the Bethe-Bloch theory.[9] Agreement of these calculations with experimental measurements of both mean free path and stopping power, where available, is quite reasonable.[4,10] We should also note that the results presented here do not include electron exchange. This effect will change slightly the shape of the inverse mean free path and stopping power curves at the lower energies but leave the higher energy portion substantially unchanged. A more complete account of this work, including the effects of exchange, will be published elsewhere.

MODEL INSULATOR THEORY APPLIED TO Al_2O_3

Since Al_2O_3 is a good insulator with a band gap of about 9 eV[11] it is desirable to represent the response of its valence electrons on the basis of a rather different model than that used for the conduction band in Al. The model which we have developed for this purpose is related to that employed by Fry[12] in which the ground state wave function of the valence electrons is described in the tight-binding approximation, while excited states are represented by orthogonalized plane waves (OPW). In our use of the model to obtain a dielectric response function we fix the normalization of the OPW excited states by requiring that the sum rule $\int_0^\infty d\omega\ \omega \mathrm{Im}[\varepsilon(k,\omega)] = (2\pi^2 e^2/m) \cdot n$, where n is the density of electrons in the valence band. In addition we assume that the solid is uniform and homogeneous. The dielectric response function corresponding to this model solid is convenient and flexible for use, can be fitted to the optical dielectric function in the limit of very long wavelengths ($k \to 0$), and describes the single-particle properties of excited electrons. The existence of plasma oscillations emerges naturally as one studies the response of the system to longitudinal electric perturbations.[4] For the calculations we will describe here, the free parameters in the model have been chosen to match the optical response function of Al_2O_3.[11]

INTERACTION OF ELECTRONS WITH Al_2O_3

Given the dielectric response function of the model insulator it is straightforward to compute the DIMFP function from Eq. (3) and the inverse mean free path by calculating the integral of Eq. (3) over all possible energy transfers. Figure 5 shows a plot of the mean free path itself computed from the Al_2O_3 model dielectric response function compared with the recently obtained experimental results of Battye et al.[13]

determined from Auger electron escape depth measurements. The agreement is fairly good over the rather limited energy range for which experimental data exists. It is also straight-forward to determine slowing-down spectra from the valence electron DIMFP functions for source energies low enough that inner shell excitation is unimportant.[4] Figure 6 shows a plot of two such spectra computed for monoenergetic electron sources of 100 and 200 eV energy. It is seen that the flux distribution corresponding to a source energy of 200 eV shows local maxima corresponding to electrons which create one plasmon of energy $\sim$ 28 eV directly upon being emitted from the source, then emit one more plasmon. A third peak may be seen faintly, showing that three plasmons in succession may be created. At lower energies both spectra tend to rise very rapidly, showing a divergence at the edge of the band gap.

Figure 7 shows a plot (solid dots) of a measured electron slowing-down flux in Al_2O_3 due to beta rays from the decay of implanted ^{165}Dy. For comparison we show the spectrum computed by Monte Carlo methods using semiclassical binary collision cross sections. These results have been described previously.[2,4] The solid line is the result of our calculation based on the model insulator theory to describe the valence band, and inner shell cross sections derived from atomic GOS values. Some improvement is found in the agreement of theory with experiment using the new cross section calculations compared with the results of the earlier theoretical calculation (the histogram)[2]. The theoretical result in the region above ~2000 eV still falls somewhat below the experimental spectrum. We feel that this is due, in part at least, to the way in which the inner shell cross sections, calculated from GOS values up to energy transfers of $\sim$1800 eV, were extrapolated to larger values of energy transfer.

SUMMARY

Efforts to compute electron slowing-down spectra, stopping power, etc., over a wide range of energy have been hampered in various degrees by a lack of reasonably accurate inner shell inelastic cross sections. The prospects for obtaining better theoretical calculations of experimentally interesting electron interaction effects are now considerably improved through the development of improved models for valence electron excitation in simple insulators and through the availability of theoretical atomic generalized oscillator strengths for inner shell electron excitation in several low-Z atomic systems. We have described briefly the way in which this information may be used to calculate mean free paths, stopping powers, and electron slowing-down spectra in Al metal and the insulator Al_2O_3.

FOOTNOTES AND REFERENCES

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FIGURE CAPTIONS

Fig. (1) The differential cross section for the transition of an electron from the 2p subshell in Al to a continuum energy level ε for several incident electron energies.

Fig. (2) The DIMFP times incident electron energy for an energy transfer ω to a 2s electron in Al. The density of Al atoms corresponds to Al_2O_3. All quantities are measured in atomic units.

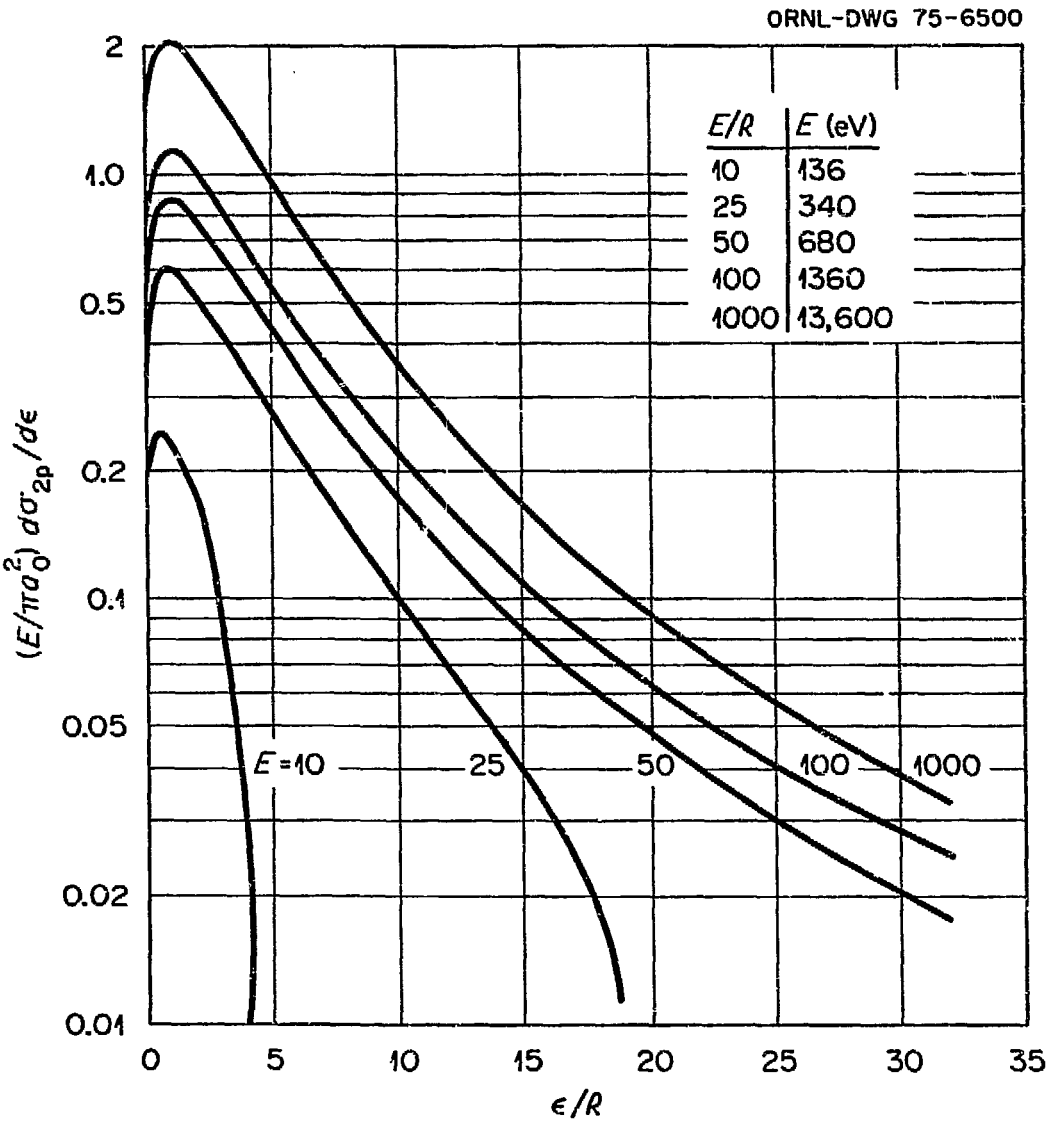
Fig. (3) The inverse mean free paths for various processes as a function of electron energy for Al metal.

Fig. (4) The contributions to the stopping power of Al for electrons due to various processes as described in the text.

Fig. (5) Mean free path of electrons in Al_2O_3 based on the model insulator theory. Also shown are calculations based on other models and some experimental data from Ref. 13.

Fig. (6) Electron slowing-down spectra in Al_2O_3 generated by monoenergetic electron sources of 100 and 200 eV based on the model insulator theory.

Fig. (7) Electron slowing-down spectrum generated in Al_2O_3 by beta particles from the decay of ^{165}Dy. The solid dots represent values obtained experimentally. The histogram indicates some results found using a Monte Carlo approach with cross sections based on a classical binary collision model (see Ref. 2). The solid curve is the result of a calculation based on the model insulator theory to describe the valence band, and inner shell cross sections derived from atomic GOS values.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2
1.0
0.5
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0.05
0.02
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5
10
15
20
25
30
35
$(E/\pi a_0^2)\, d\sigma_{2p}/d\epsilon$
ϵ/R
E/R | E (eV)
10 | 136
25 | 340
50 | 680
100 | 1360
1000 | 13,600
E=10
25
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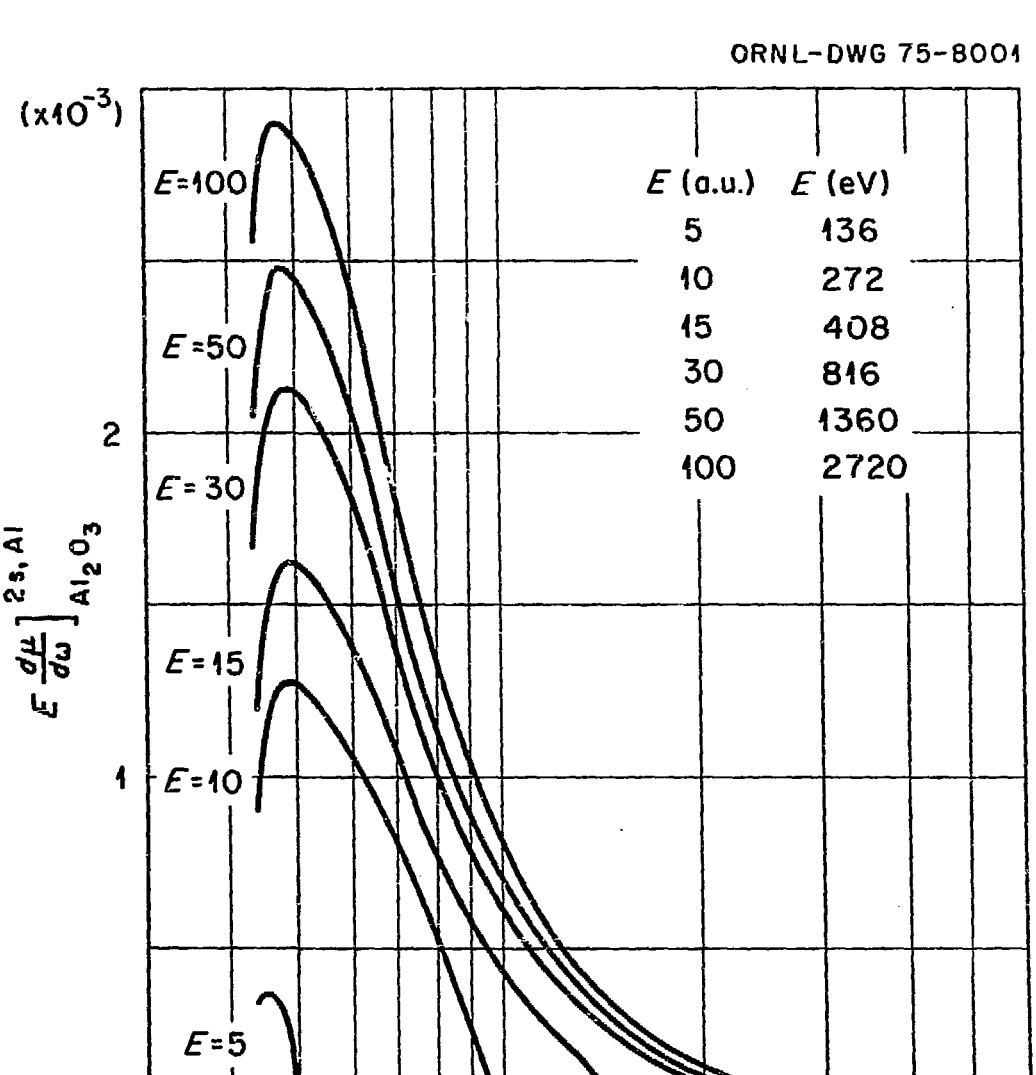
100
1000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(x10⁻³)
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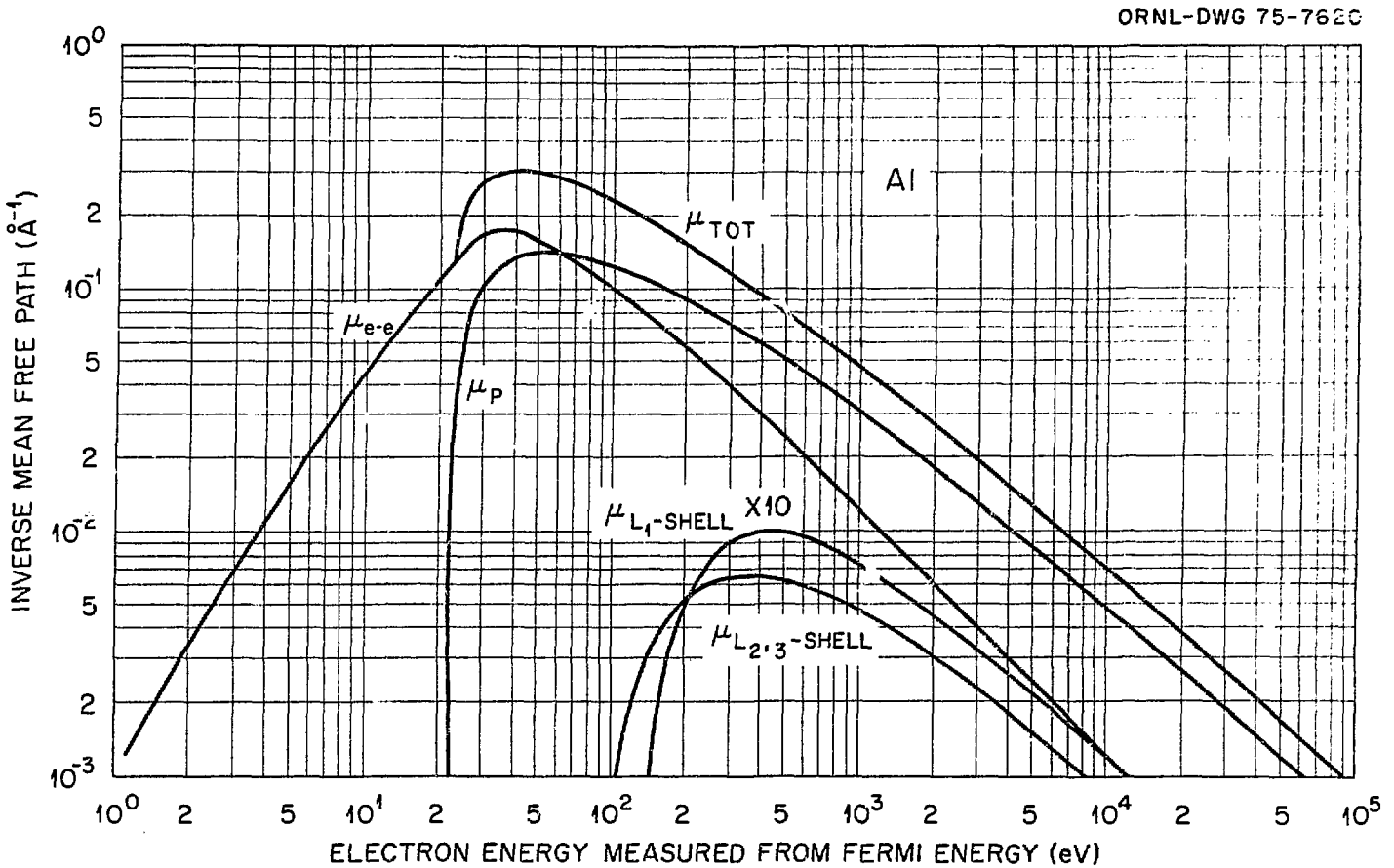
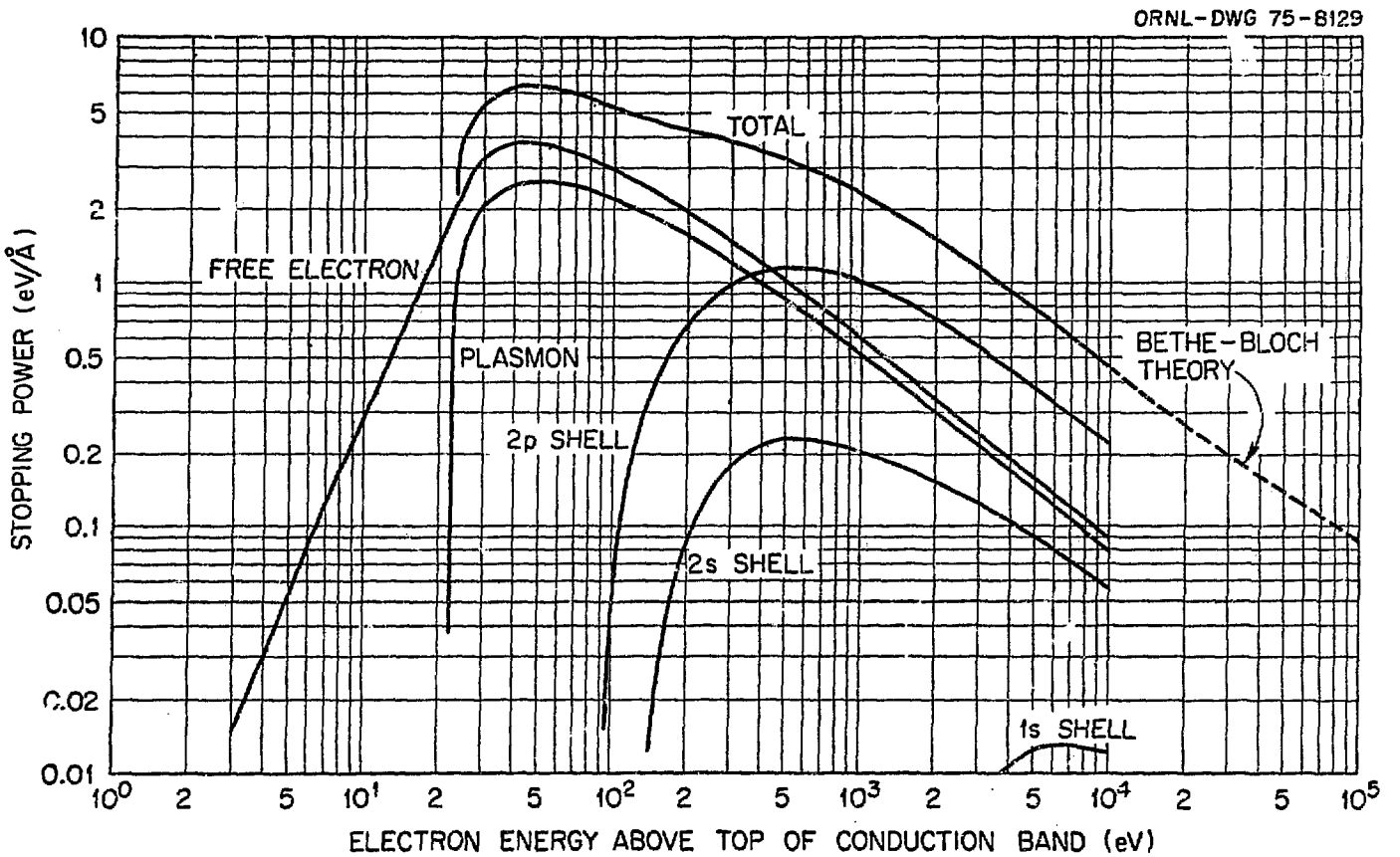
E=50
E=30
E=15
E=10
E=5
E (a.u.) E (eV)
5 136
10 272
15 408
30 816
50 1360
100 2720
$E\frac{d\mu}{d\omega}\Big]^{2s,Al}_{Al_2O_3}$
0
1
2
5
10
20
50
ω, ENERGY TRANSFER


ORNL-DWG 75-7620
Al
μ_{TOT}
$\mu_{e\text{-}e}$
μ_P
$\mu_{L_1\text{-SHELL}}$ X10
$\mu_{L_{2,3}\text{-SHELL}}$
INVERSE MEAN FREE PATH (Å^{-1})
ELECTRON ENERGY MEASURED FROM FERMI ENERGY (eV)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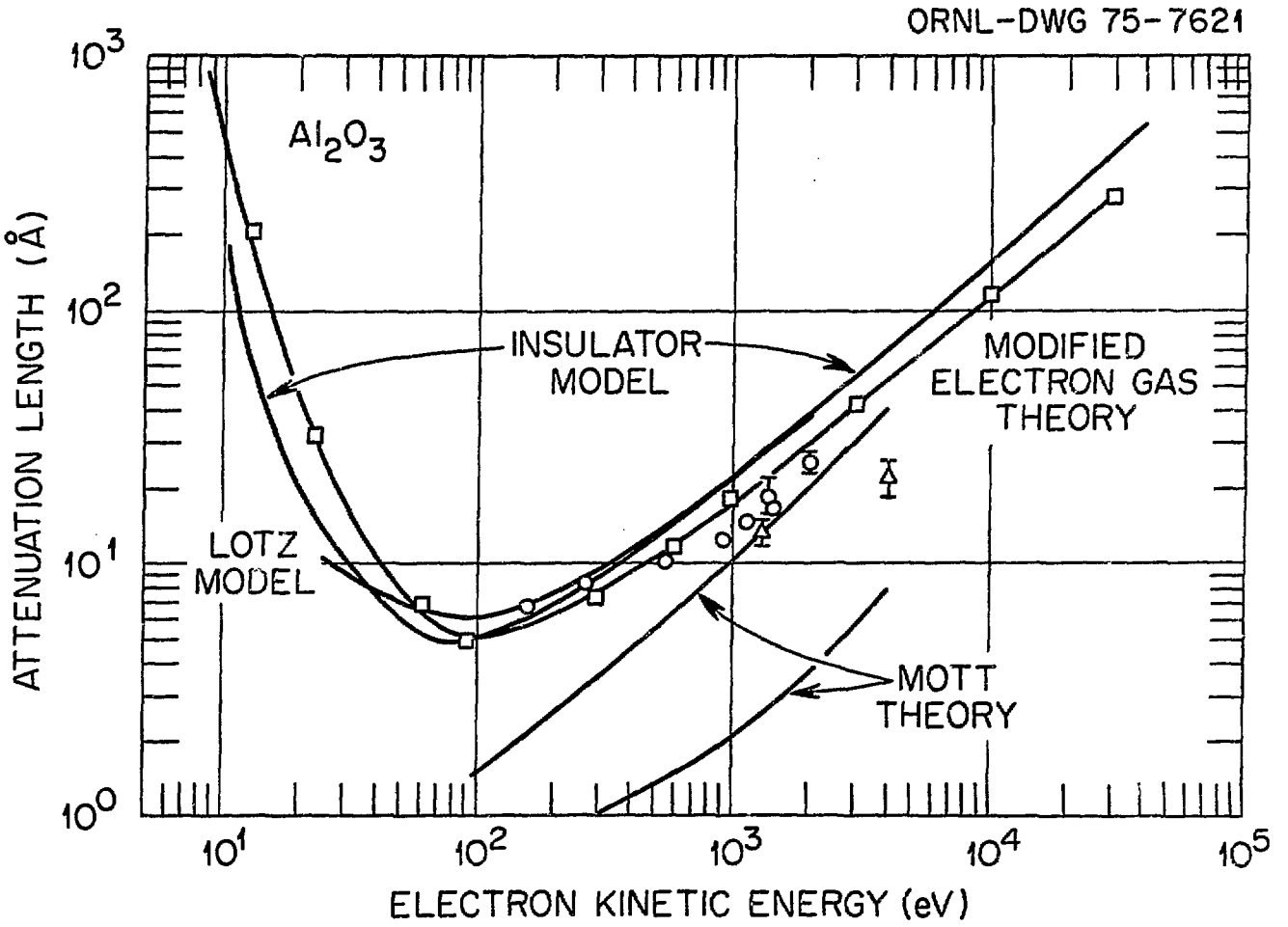
STOPPING POWER (eV/Å)
10
5
2
1
0.5
0.2
0.1
0.05
0.02
0.01
FREE ELECTRON
TOTAL
PLASMON
2p SHELL
2s SHELL
1s SHELL
BETHE-BLOCH THEORY
10^0 2 5 10^1 2 5 10^2 2 5 10^3 2 5 10^4 2 5 10^5
ELECTRON ENERGY ABOVE TOP OF CONDUCTION BAND (eV)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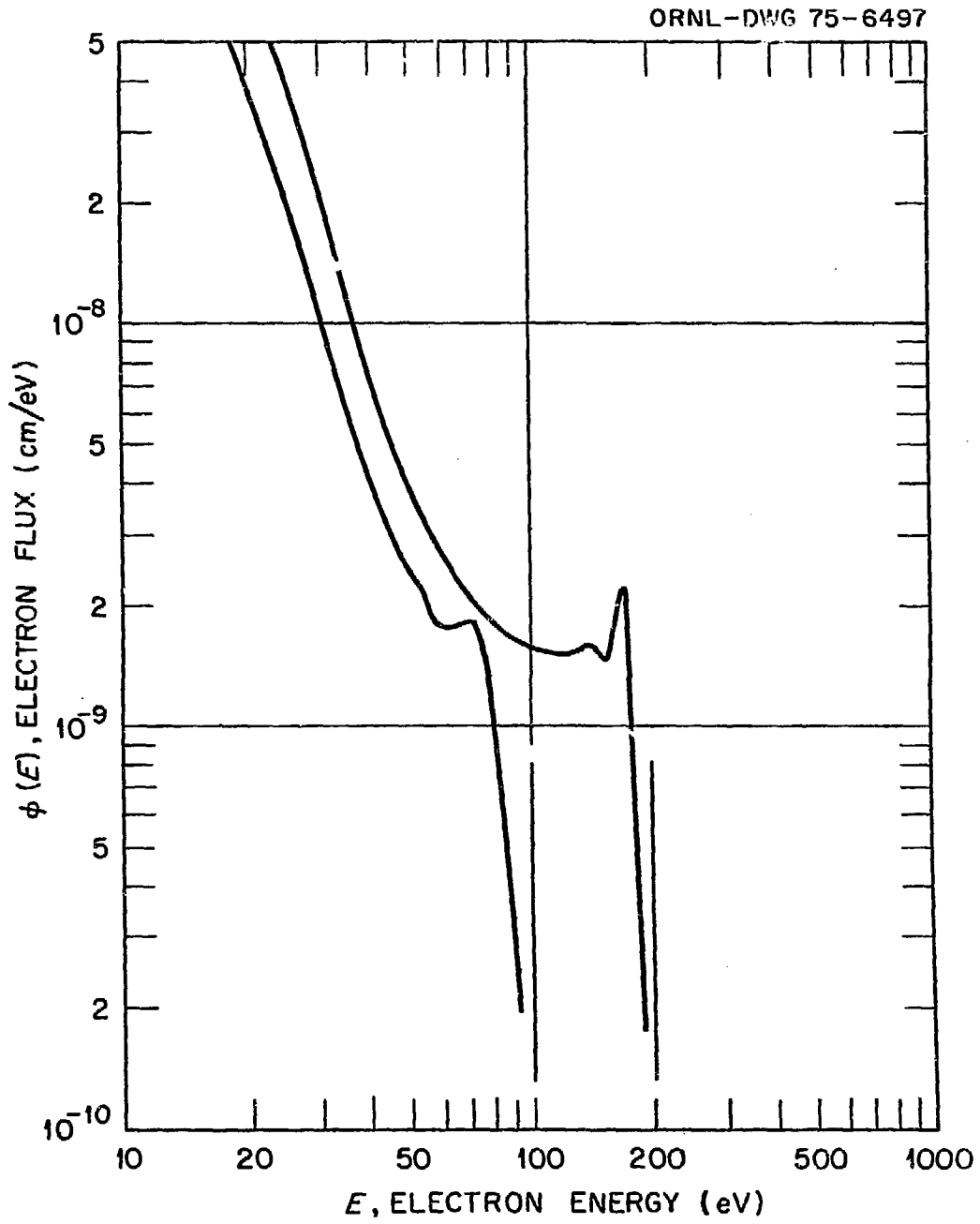
Al_2O_3
INSULATOR MODEL
MODIFIED ELECTRON GAS THEORY
LOTZ MODEL
MOTT THEORY
ATTENUATION LENGTH (Å)
ELECTRON KINETIC ENERGY (eV)
10^0
10^1
10^2
10^3
10^4
10^5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5
2
10^{-9}
5
2
10^{-10}
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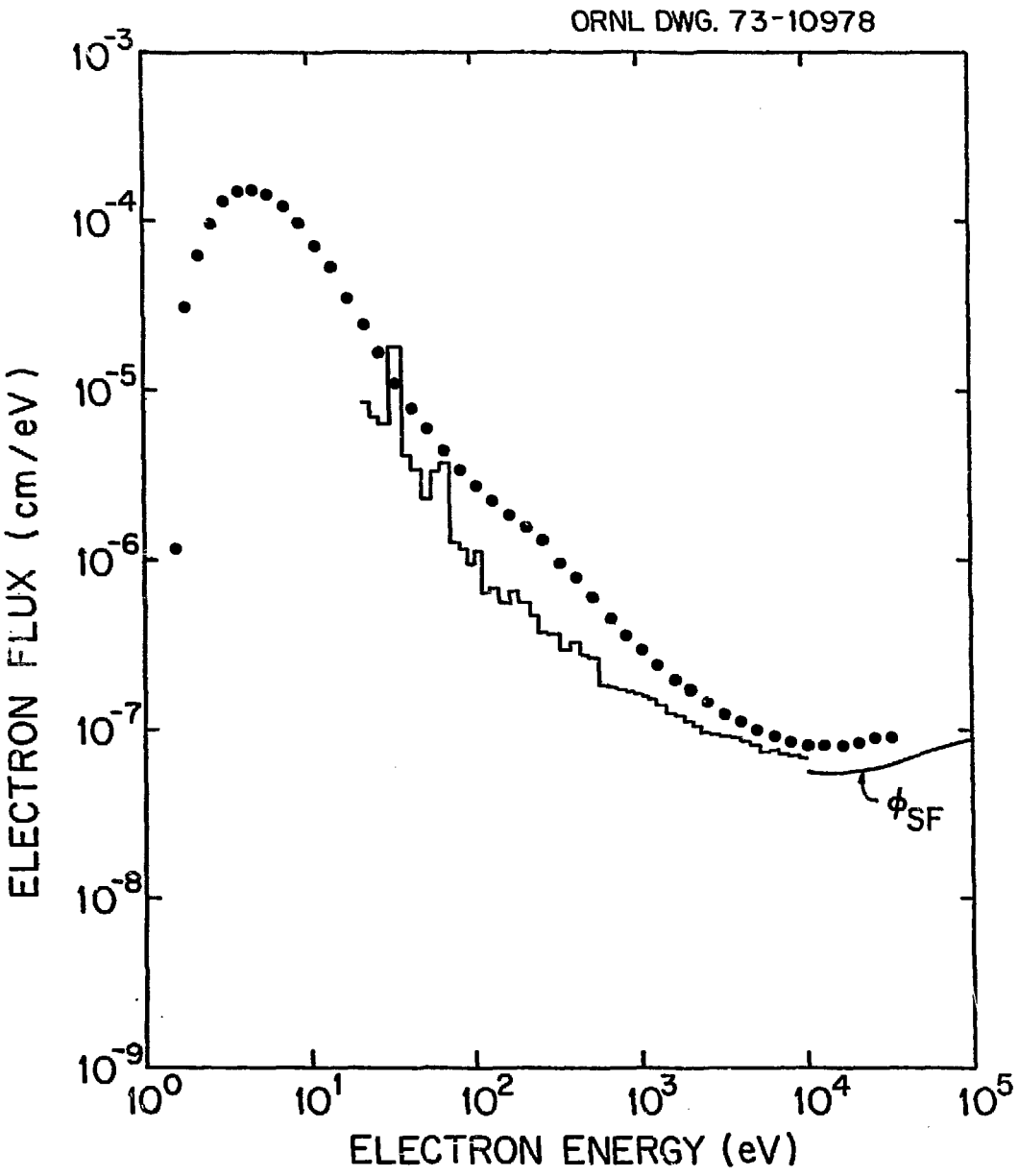
20
50
100
200
500
1000
E, ELECTRON ENERGY (eV)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ELECTRON FLUX (cm/eV)
ELECTRON ENERGY (eV)
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10^{-4}
10^{-5}
10^{-6}
10^{-7}
10^{-8}
10^{-9}
10^{0}
10^{1}
10^{2}
10^{3}
10^{4}
10^{5}
ϕ_{SF}