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A MONTE CARLO CALCULATION OF HIGH-ENERGY SPUTTERING

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Neutron Physics Division

A MONTE CARLO CALCULATION OF HIGH-ENERGY SPUTTERING

D. T. Goldman*
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ABSTRACT

A machine calculation using an IBM-704 High-Speed Calculator was used to determine the sputtering produced by high-energy ($\sim$ 100-kev) deuterons incident on copper metal. The initial displacement interaction between incident and lattice particles was taken to be Coulomb, and subsequent lattice ion - lattice ion interaction potential to be hard sphere. The value of the sputtering ratio in this energy range was found to be in agreement with that predicted by a theory of Goldman and Simon. Various suggestions are given to improve the machine calculations and to include a screened Coulomb initial interaction which would enable the code to be used for lower energy incident particles.

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I. INTRODUCTION

As remarked in a previous paper,[1] most of the theoretical treatments of ion sputtering have been concerned with incident ion energies of less than a few kilovolts. In Ref. 1 a theory was proposed to deal with the region of ion energies above about 50 Kev, where the range of the incident ion is much greater than that of the knock-on particles. This simplifying feature meant that it was possible to obtain a closed expression for the sputtering ratio, although it was conjectured that its accuracy was probably limited to the dependence of the ratio on gross quantities such as the energy, mass, and incident angle of the bombarding ion. In the absence of experimental data in this energy range, it was proposed to check the validity of the results given in Ref. 1 by means of a Monte Carlo calculation. In addition, such a code may be useful for direct computation in the lower energy ranges. In Section II of this report a general description of the theory of the Monte Carlo calculation is given and in Section III the details of the machine code are given. Section IV contains some suggested improvements and modifications in the code, which, however, have not as yet been carried out. The results of the machine calculation are given in Sections V and VI and these are compared with those of Ref. 1. The conclusions are given in Section VII.

II. GENERAL THEORY OF THE MONTE CARLO CALCULATION

We begin by considering a sufficiently large number of relatively high-energy ions incident at some given angle on the surface of a solid. In all of the following, for illustration, the incident ions were chosen to be deuterons and the bombarded material chosen to be copper. Since the displacement mean free path of the incident deuterons is much greater than that of the primaries produced by the interaction between the deuterons and the lattice particles,[1] it is necessary to consider only the first primary produced by the incident ions. At the energies of interest (above 50 kev),

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1. D. T. Goldman and A. Simon, Phys. Rev. 111, 383 (1958).

the deuteron-primary interaction can be assumed to be pure Coulomb (the screening due to electrons is not important at these high energies). We neglect elastic scattering of the incident ions without the production of a primary particle. Hence, we have a natural cut-off to the interaction, corresponding to a minimum energy transferred to the primary. The distribution function of this interaction (properly normalized) is given by

$$f(x)dx = 2\left(\frac{1 - c}{1 + c}\right) \frac{dx}{(1 - x)^2}, \quad -1 \leq x \leq c \tag{1}$$

where x is the cosine of the scattering angle in the center-of-mass and c is the cosine of the minimum scattering angle, equal to $c = 1 - (2E_d/T_m)$. E_d is the energy which must be transmitted to the struck particle to allow it to escape from the surface and T_m the maximum energy transferable,[1]

$$T_m = \frac{4M_1M_2E}{(M_1 + M_2)^2}$$

The distribution of primary particles is symmetric in the azimuthal plane.

The energy and direction of the daughter particles were determined by the appropriate distribution function (Eq. 1) by treating the lattice ions as free particles. The energy was then tested to see if it was greater than the binding energy of the lattice. The approximation we use is to subtract the lattice particle binding energy from the energy determined on the basis of the distribution function but retain the direction of the particle as previously determined. The new energy and the direction now characterize the ions. The primary ion then undergoes elastic collisions with the lattice ions, which, because of the low average energy of the primaries ($\sim$ 100 ev), are taken to be hard sphere in nature, isotropic in the center-of-mass system. If these collisions transfer sufficient energy to the bound particles to overcome the binding energy of the lattice, then a secondary particle is produced whose history must also be followed. The mean free path between

collisions of the copper atoms is computed using particle energy of 200 ev, corresponding to the average energy of primary particles displaced by a beam of 500-kev deuterons. For a more proper choice of this mean free path see Section IV.

After each collision the energy and the direction of the primary particle and a possible secondary are computed. When the energy is degraded below the surface potential (5 ev), the particle is considered to have died, and we turn our attention to the history of the next particle in sequence. This is continued until all particles, primary and secondary, are accounted for from birth to death.

After the histories of all the displaced particles are computed, separate analysis routines are used to determine the sputtering ratio, that is ,the number of particles which escape from the surface of the material, relative to the number incident. However, a discussion of these analyses is sufficiently detailed to be postponed until the next section.

III. DESCRIPTION OF THE CODE

A. Histories

Figure 1 is a flow-chart depicting the history of the primary and all the secondary particles produced by a single deuteron as programmed for the IBM-704. The data for the particles resulting from a collision, which are stored for later analysis on Tape 3, are

1. Number of the deuteron from which each particle is descended.
2. Geneology number which specifies the intermediate particles from which each particle is descended.
3. Z, the distance in the direction of the surface from the birth of the primary to the point of collision (see Fig. 2).
4. Z_m, the maximum penetration in the direction of the surface of the particle or of any of its ancestors (see Fig. 2).

5-7. X, Y, and Z components of velocity.

8. Speed of the particle.

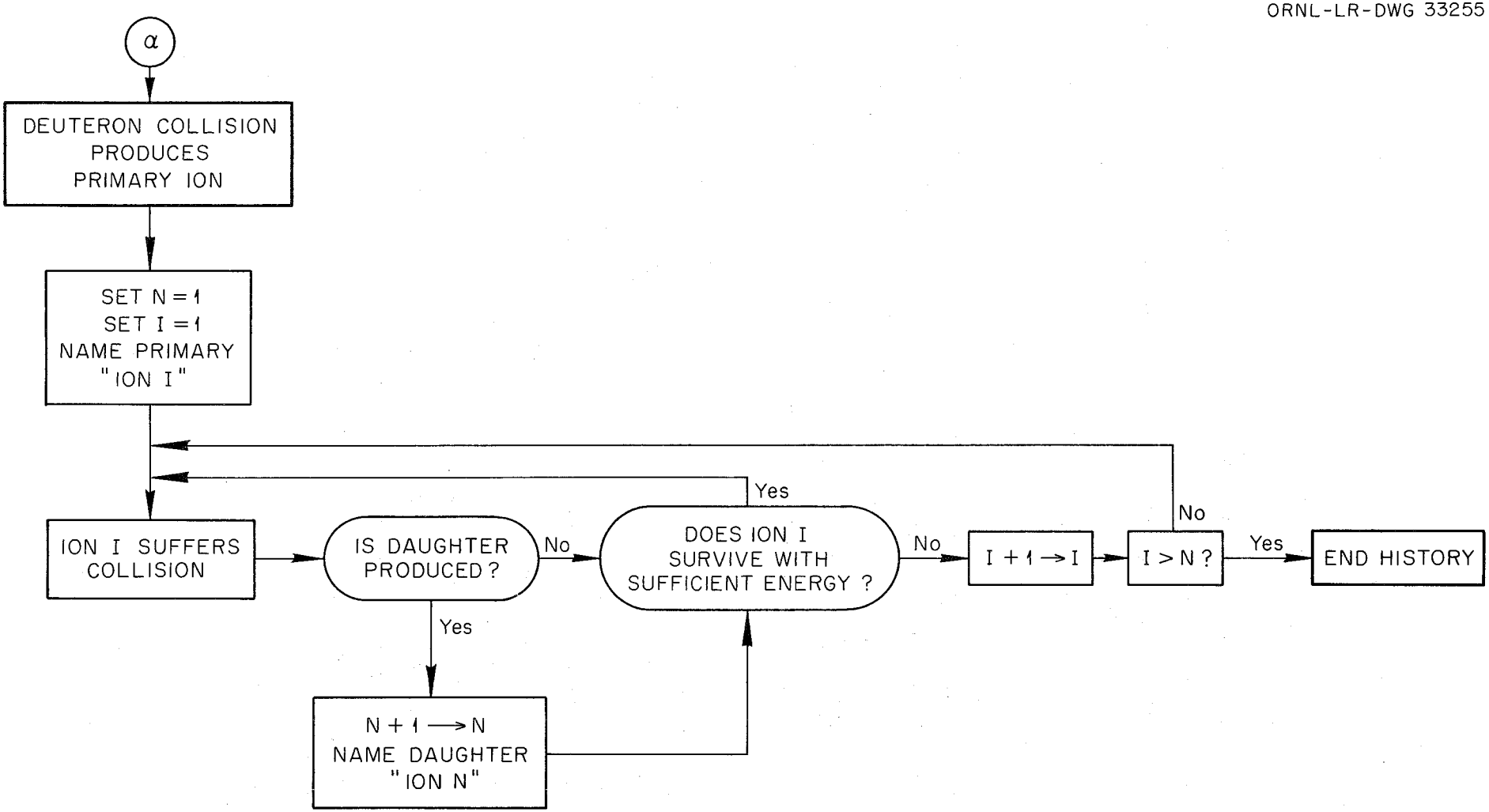

Fig. 1. Flow Chart for the Monte Carlo Calculation of the Histories of the Displaced Particles.

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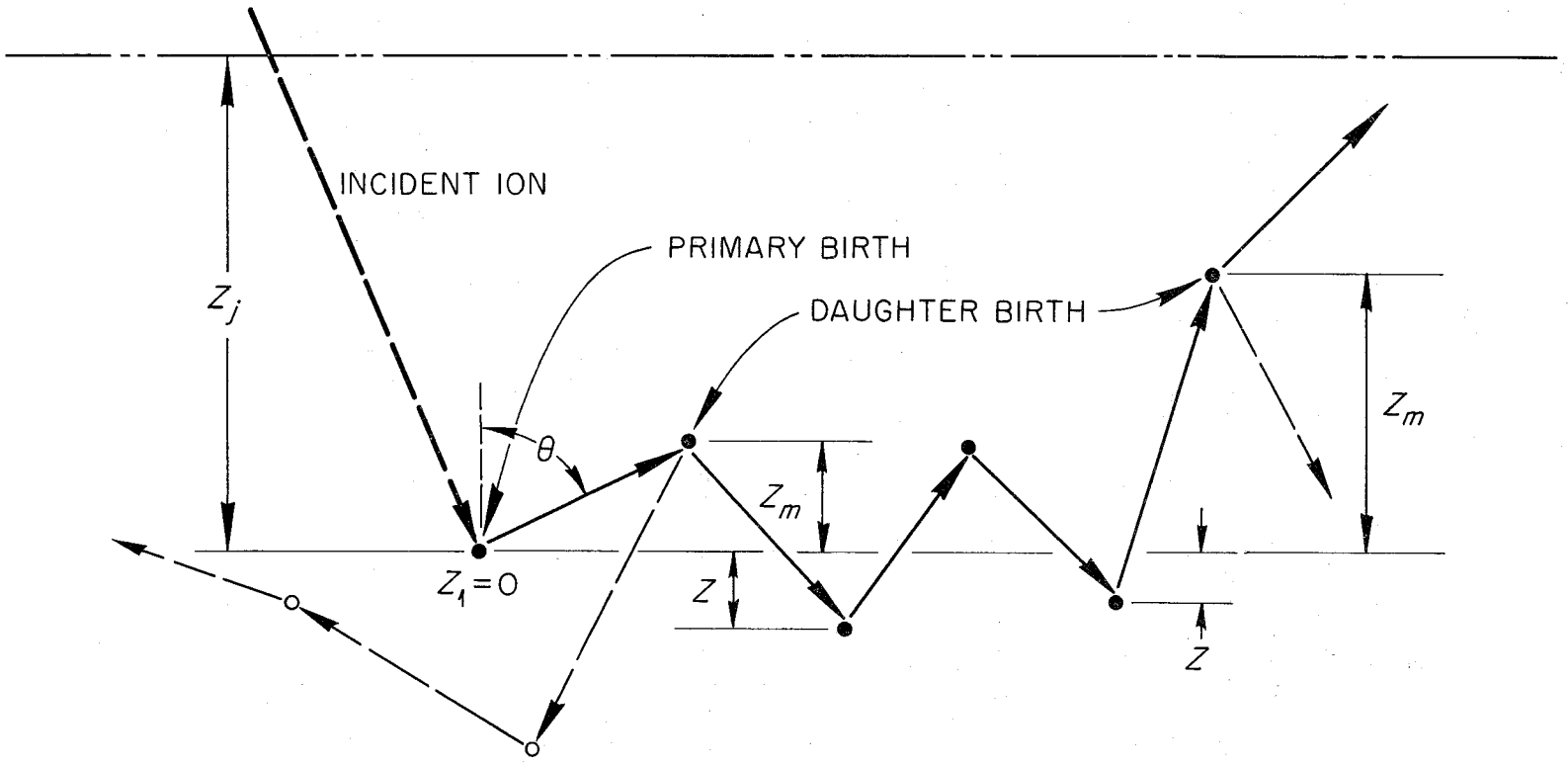

Fig. 2. Diagram Illustrating the Creation of a Primary and a Secondary Particle and Depicting the Quantities Z, Z_m, Z_j and θ.

9. Energy of the particle.
10. Cosθ, the directional cosine to the normal to the surface (see Fig. 2).

Items 1-10 at a particle's birth and at its death only are also stored for later analysis on Tape 4.

The program calls for various items of input information to be loaded into the machine on IBM cards. These items include number, energy, and direction of the deuterons to be considered, masses and mean free paths of incident and bombarded particles, lattice binding energy and surface potential and numerous additional instructions, all of which can be changed to modify the program in accordance with the suggestions of the next section.

B. Analysis

Two types of analysis codes were run with approximately equal results. In the more simple trial, the births and deaths recorded on Tape 4 were utilized. Increasing distances of the surface from the primary birth point were chosen in an arithmetic progression and for each distance the data for the birth and death of each particle were tested to determine if Z_m was greater than the chosen distance. If the answers were no and yes, respectively, then if the particle were produced at the depth, it would have escaped. The probability of production depends, of course, on the mean free path of the deuterons but because this is so large compared to that of the primary particle, we can assume uniform production in depth. The sputtering ratio was then determined in a straightforward manner.

A second analysis uses the data for each collision stored on Tape 3. In this case, for each selected distance, Z_j, the particle after each collision might escape with a probability $e^{-(z-z_j/\lambda\cos\theta)}$, $(Z > 0)$, where λ is the mean free path of the copper atom. These probabilities are summed and weighted as described in the previous paragraph. This analysis also has provisions for determining the angular and energy distributions of the sputtered particles.

IV. SUGGESTED MODIFICATIONS

Perhaps the most important change in the program which is needed in order to extend its validity to lower incident energies and lighter weight targets is to change the first interaction from an ordinary Coulomb to a screened Coulomb interaction. This involves mainly replacing the normalized distribution function of Eq. 1 by the corresponding

$$f_{sc}(x)dx = \frac{(1 + k)(1 - kc)}{(1 + c)} \frac{dx}{(1 - kx)^2} , \tag{2}$$

k being the screening "constant." Note that in the limit of high energy $k \rightarrow 1$ and $f_{sc} \rightarrow f$. An analysis using this new distribution function has been started but only preliminary results have been obtained at this time.

It may be entirely possible to extend the sputtering calculation in this manner to much lower energies than 50 kev and thus to be able to check the results of the calculation with available experimental information. To be sure, the assumption of only one primary produced per deuteron would not be valid at low incident ion energies but this modification has been built into the code.

It might also prove beneficial to be able to write the mean free path of the knock-on particles as an algebraic function of the energy and thus have the code compute the mean free path of these particles after each collision, instead of assuming an average mean free path corresponding to an average energy, as has been done heretofore. Preliminary work has been accomplished in this direction also.

An additional analysis routine has been suggested which would analyze the energy and angular distribution of the sputtered particles in terms of appropriate Legendre functions.

V. RESULTS

The program was run on the IBM 704 located at the Oak Ridge Gaseous Diffusion Plant. In each of three cases, 500 deuterons were allowed to bombard the surface of metallic copper at normal incidence. The energy of the incident deuterons was varied in the three cases. The sputtering ratio and displacement mean free path at each energy are tabulated in Table 1. No exact value of the variance of the sputtering ratio as a function of the random number was obtained. However, preliminary results indicate that this variance, for a choice of 500 incident particles, was no greater than a few percent.

Table 1

E (Mev)	Sputtering Ratio	Displacement Mean Free Path (cm)
30	4.22×10^{-3}	$.0516 \times 10^{-4}$
100	1.85×10^{-3}	$.172 \times 10^{-4}$
300	6.72×10^{-4}	$.516 \times 10^{-4}$

Some work was also done on determining the energy and angular distribution of the sputtered particles. These results, which are preliminary in nature, nevertheless are interesting and are given in Table 2. The averaged results of five different runs of fifty normally -incident deuterons of 100 Kev energy were analyzed using the second routine described in Section III B. Table 2 contains the percentage of sputtered particles with energy and angle of emergence in given energy and angle range as a function of the total number of sputtered particles. We note that 90% of the particles possess energy less than 200 ev, and approximately 80% of the particles emerge at an angle of less than 45 deg with the normal.

It would also prove beneficial to determine the dependence of the sputtering ratio on the angle of incidence and mass of the incident particle,

Table 2

A. Number of Sputtered Particles with Energies in 200 ev Energy Ranges as a Percent of the Total Number of Sputtered Particles (100 Kev Incident Deuterons)

Energy	Percent of Total
0-200 ev	90.0
200-400	3.9
400-600	1.9
800-1000	0.9
Above 1000	1.2

B. Number of Sputtered Particles with Directional Cosines in 2 Ranges as a Percent of the Total Number of Sputtered Particles (100 Kev Incident Deuterons)

Cosθ	Percent of Total
0-.2	3.2
.2-.4	9.9
.4-.6	27.8
.6-.8	30.4
.8-1.0	28.7

and on the mass of the target particles. Such work, however, has not as yet been undertaken, although the code makes provision for this extension.

VI. COMPARISON WITH THEORY

In Reference 1, the energy dependence of the sputtering ratio, R, was predicted to be

$$R \propto \frac{\ln E}{E} \tag{3}$$

The exact expression for R is given by Ref. 1, Eq. 22. The predicted sputtering ratio as a function of energy is plotted as the upper line of Fig. 3. For comparison we have also included the values of the sputtering ratio determined by the computer calculation and joined these by a smooth curve. We note that at the high-energy end the computed curve can be so constructed as to give the energy dependence predicted by Eq. 3. At the two points corresponding to higher energies, the computed curve is consistently about 2/3 that of the theoretical one. If one takes into account the assumptions involved in obtaining the theoretical prediction of the sputtering ratio, this agreement is quite satisfactory. The discrepancy between the two calculations can best be explained as the fact that fewer secondary particles are produced by each primary than determined from the theory as detailed in Ref. 1. An analysis of the computer data tends to confirm this hypothesis.

VII. CONCLUSION

At this time there would appear to be reasonably good agreement between the theory and machine computed results for the sputtering ratio at high energies, both as to the energy dependence of the sputtering ratio and the absolute magnitude of it. Further work should be done at higher energies to determine if this agreement persists. The modifications suggested in the program will be used to extend the calculations to a lower energy and in an effort to get direct comparison with experiment.

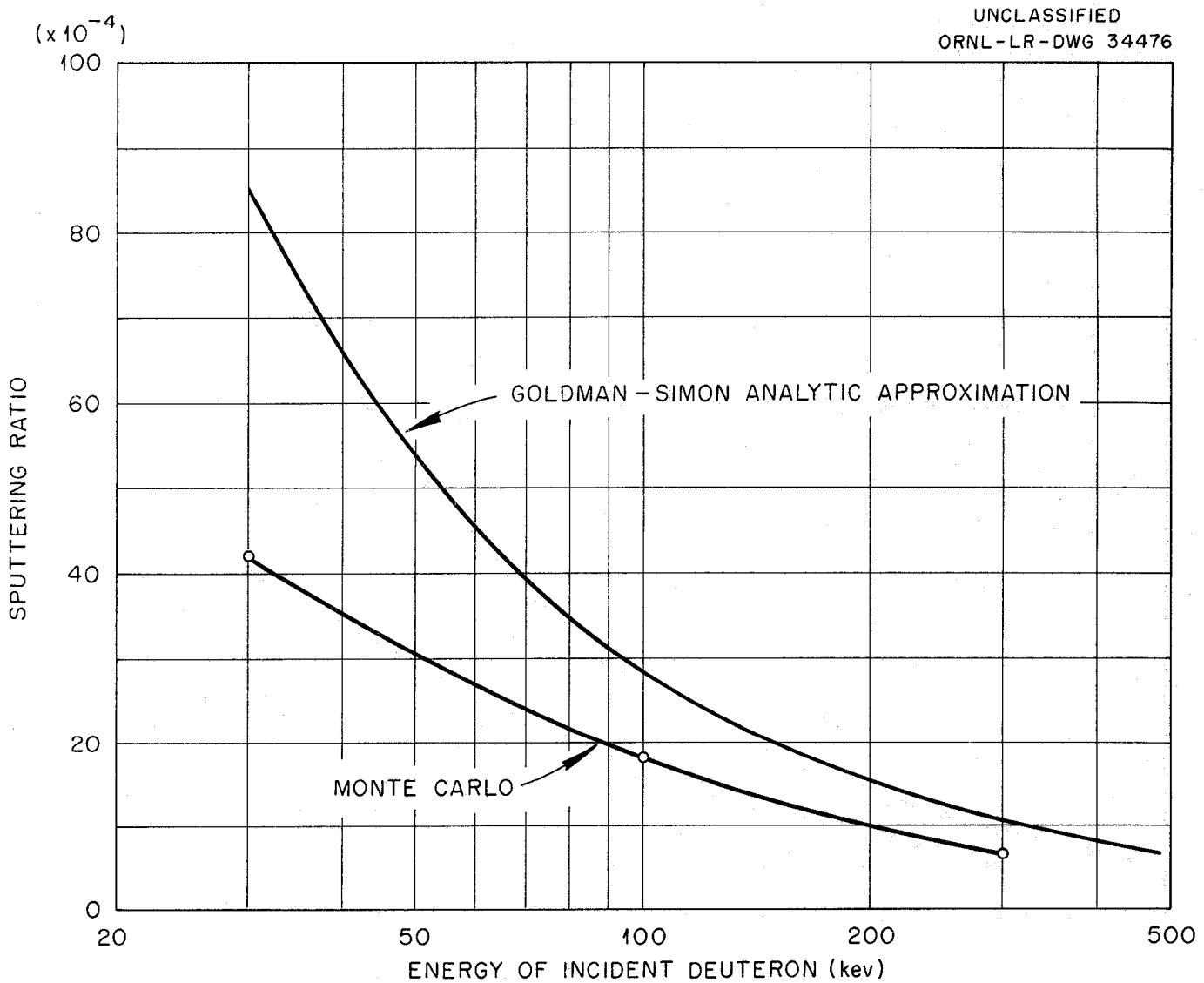

Fig. 3. The Sputtering Ratio as a Function of Energy in the Range 30–500 kev.

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