calculated maximum separation factor. For xenon there is actually good agreement between the observed and calculated optimum gas pressures, although the observed maximum factor is smaller than calculated. Most likely this discrepancy arises from the failure of the theory to include any temperature variation of α_0, which for xenon is quite large ($\alpha_0=0.08$ at 350°K and approximately 0.16 at 700°K). Requirements (1) and (2) then are mutually contradictory; the gases which are more nearly Maxwellian also display the greatest temperature variation of the thermal diffusion factor, while the gases for which α is more nearly constant are the least Maxwellian.

Finally, why does this column performance for normal argon at $T_1=288$°K agree fairly well with that calculated from the theory, while there is no comparable agreement for the argon analyzing 9.70% A^{36} under similar conditions? Since the shape of the calculated $2A$-*versus*-p curve depends in a sensitive manner on the values of D (self-diffusion) and η (viscosity), these coefficients must be known to say one percent for accurate comparison. In calculating the performance curve for the argon enriched in A^{36}, we used the coefficients given in the literature for normal argon, but corrected for the change in the average molecular mass. Small but appreciable errors may be present in any or all of these coefficients. It is quite possible that the good agreement for the case of normal argon is the fortuitous consequence of slightly inaccurate gas coefficients. Or, of course, argon may represent a compromise in satisfying requirements (1) and (2) of the theory.

The introduction of a remixing factor K_p will often improve somewhat the fit between the theoretical and experimental separation-factor curves, of course, and one must admit that there may be small parasitic convection currents in any column operation. Examination of Fig. 1, however, shows that this cannot be a general solution of the problem.

We conclude that measurements of the performance of a thermal diffusion column are not at present a good way to determine thermal diffusion factors with any accuracy. The next step to be taken, if column performance data are to be so used, is to extend the theory to include other than Maxwellian molecules as well as a temperature dependence for the thermal diffusion factor.

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Theory of Sputtering by High-Speed Ions

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The theoretical treatment of ion sputtering at energies above about 50 kev is simplified by the fact that the emergent particles originate at depths in the material which are small compared to the range of the incident particles. The displacement rate is nearly constant over this region and this enables one to obtain relatively simple solutions of the diffusion problem for emission.

The emission problem is reduced to an effective one-velocity diffusion calculation by an artifice. The volume displacement rate is increased by the factor $\bar{\nu}$, where $\bar{\nu}$ is the average number of secondaries, and the macroscopic absorption and scattering cross sections are adjusted to make the average number of collisions of each particle equal to the actual average over the secondaries. The resultant sputtering ratio, $\mathfrak{R}$, varies with incident energy E, incident angle ψ, and mass ratio $\mu=M_1/M_2$ (where $M_1=$mass of incident particle, $M_2=$mass of target particle), as $\mathfrak{R}\propto\mu(\ln E/E)\sec\psi$.

I. INTRODUCTION

MOST of the theoretical treatments of ion sputtering appear to be limited to the region below a few kilovolts.[1] In this case, the ions have a mean free path of only a few atomic layers and the phenomenon involves a complicated analysis of surface interactions. As the energy of the bombarding ion increases above about 50 kev, however, a simplifying feature emerges. The range of the incident particle increases greatly ($\cong 10^{-4}$ cm at 500 kev) while the knock-on particles maintain an approximately constant mean free path ($\cong 10^{-7}$ cm). Hence, only a small portion of the initial track length of the incident particle produces knock-on particles which can re-emerge from the surface. The displacement rate is nearly constant over this region and this enables one to obtain relatively simple solutions of the diffusion problem for emission.

In Sec. II of this report an expression is obtained for the volume density of primary particles produced by the incident beam. The mean free path for primary production is calculated in Sec. III, as well as the average energy of the primaries. The average number of secondaries is calculated in Sec. IV, the diffusion problem for re-emission is solved in Sec. V and the results summarized in Sec. VI.

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[1] D. E. Harrison, Phys. Rev. **102**, 1473 (1956); E. B. Henschke, Phys. Rev. **106**, 737 (1957).

II. VOLUME DENSITY OF PRIMARY SOURCES; SPUTTERING RATIO

Consider an initial beam of monoenergetic ions incident at an angle ψ to a solid surface as shown in Fig. 1. The particle flux in the beam is denoted by I and the cross-sectional area of the beam, measured normal to its direction, is denoted by A. The number of primaries produced per unit volume in the solid is then given by

$$q \cong IS(R)/A\lambda_i(E), \tag{1}$$

where $S(R)$ is a step function which is equal to unity from 0 (the surface) down to R (the incident particle's range) and is zero thereafter. The mean free path for production of a primary by an ion of energy E is denoted by $\lambda_i(E)$ and E is the energy at a given depth r. The basic assumption is that the displaced particles which emerge originate from a depth which is small compared to R. Hence, q is effectively a constant over this region with E equal to the incident energy E_i. Thus

$$q \cong I/A\lambda_i(E_i). \tag{2}$$

Owing to the linear nature of the problem, the current density of sputtered atoms leaving the surface will be directly proportional to the volume source strength. Hence, one may write

$$J=\gamma q, \tag{3}$$

where J is the emitted current density normal to the surface and γ is to be determined later. The total emitted current is then (see Fig. 1)

$$J_{\text{tot}}=\gamma q A_s. \tag{4}$$

The sputtering ratio $\mathfrak{R}$, defined as the ratio of total current of sputtered atoms to incident current, is obtained by dividing Eq. (4) by the incident current I and substituting from Eq. (2). The result is

$$\mathfrak{R}=\gamma A_s/A\lambda_i(E_i)=\gamma[\lambda_i(E_i)\cos\psi]^{-1}. \tag{5}$$

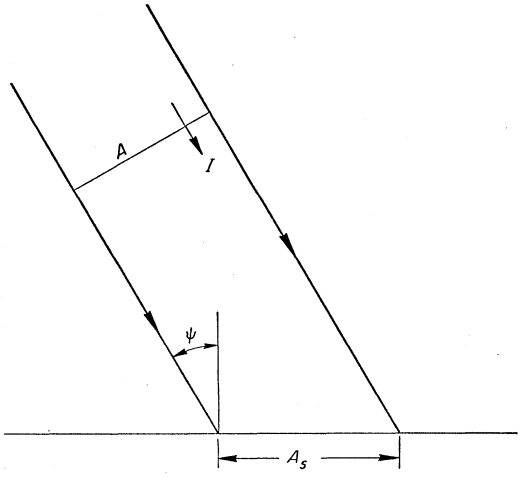

FIG. 1. Incident ion beam.

The mean free path for primary production, λ_i, will be evaluated in the next section.

III. MEAN FREE PATH FOR PRIMARY PRODUCTION

Following Seitz and Koehler,[2] the differential cross section for a Coulomb collision in which an energy between T and $T+dT$ is transferred is given by

$$d\sigma=\frac{\pi b^2}{4}T_m\frac{dT}{T^2}, \tag{6}$$

where the maximum energy transferable is denoted by T_m and

$$T_m=4M_1M_2E/(M_1+M_2)^2. \tag{7}$$

The energy of the incident particle of mass M_1 is denoted by E, M_2 is the target particle mass, b is the classical distance of closest approach defined by

$$b=z_1z_2e^2/E_r, \tag{8}$$

and E_r is the relative energy in the center-of-mass system. We assume that the screening is weak, as it will be for energetic ions. The total cross section for producing a displacement, σ_d, is the integral of $d\sigma$ between E_d and T_m, where E_d is the energy which must be transmitted to the struck particle to cause it to be removed from the lattice structure. Hence

$$\sigma_d=\frac{\pi b^2}{4}\left(\frac{T_m}{E_d}-1\right)=\frac{\pi b^2 T_m}{4E_d}, \tag{9}$$

since E_d ($\cong$25 ev) is such smaller than T_m. Substituting in Eq. (9) from Eqs. (7) and (8), one obtains

$$\sigma_d=\pi\frac{M_1}{M_2}\frac{z_1^2z_2^2e^4}{EE_d}. \tag{10}$$

The mean free path for primary production is then

$$\lambda_i=(n_0\sigma_d)^{-1}, \tag{11}$$

where n_0 is the particle density in the medium.

For illustration in this and succeeding sections, we shall consider the case of a beam of 500-kev deuterons striking a copper target. Then

$$\sigma_d=6.95\times10^{-14}/E=1.4\times10^{-19}\ \text{cm}^2,$$

$$\lambda_i=0.86\times10^{-4}\ \text{cm}.$$

The mean energy transferred to the primary particle

[2] F. Seitz and J. S. Koehler, in *Solid State Physics*, edited by F. Seitz and D. Turnbull (Academic Press, Inc., New York, 1956), Vol. 2, p. 321.

is readily obtained from Eq. (6). The result is

$$\bar{T}=E_d \ln(T_m/E_d), \quad T_m \gg E_d.$$

In our illustrative case, $\bar{T}_d=195$ ev. In fact, owing to the slow variation of the logarithm, the mean energy transferred is less than 500 ev at all energies of interest.

Since the incident particle loses only a small portion of its kinetic energy, on the average, the direction of the primary recoil after the collision will be almost perpendicular to the direction of the incident particle. This follows from the relationship between the c.m. scattering angle θ and the energy transferred:

$$\sin^2(\theta/2)=T/T_m,$$

as well as that of the recoil angle ψ in the laboratory system:

$$\psi=(\pi-\theta)/2.$$

In our illustrative case, $\bar{\psi}=86.6°$.

IV. AVERAGE NUMBER OF SECONDARIES PRODUCED

Since the primary particles are not very energetic, their interaction with the lattice ions reduces to hard-sphere collisions. The total cross section is πR^2, where R is determined from the relationship

$$\frac{z_2^2 e^2 \exp(-R/a')}{R}=E', \tag{12}$$

and where, after Seitz,

$$a'=a_0/z_2^{\frac{1}{3}},$$

with a_0 being the Bohr radius. The radius R is not a very sensitive function of the energy, E'. In our illustrative case, $R \cong 1.35a_0=0.7\times10^{-8}$ cm. The corresponding mean free path in copper is then 7.4×10^{-8} cm.

The average number of all secondary atoms dislodged from the lattice by the primary atom may be determined by the method described in Seitz and Koehler.[3] The result is

$$\bar{\nu}=\left[0.885+0.561 \ln\left(\frac{x_m+1}{4}\right)\right]\frac{x_m+1}{x_m}, \tag{13}$$

where

$$x_m+1=\frac{4M_1M_2}{(M_1+M_2)^2}\frac{E}{E_d}. \tag{14}$$

For 500-kev deuterons impinging on copper, $x_m=2.4\times10^3$ and $\bar{\nu}=4.5$.

V. DIFFUSION SOLUTION FOR THE RETURN CURRENT

The exact solution of the propagation of knock-on particles through the material to the surface involves a complex multiplication problem which cannot easily be solved by analytic means. Instead, we shall replace the correct source term q, of Eq. (2), by an effective source $\bar{\nu}q$, where $\bar{\nu}$ is given by Eq. (13), thus accounting for the multiplication process directly at the primary source. In addition, the angular distribution of primaries is considered to be isotropic instead of peaked at right angles to the incident particle's direction. This artifice allows one to deal with an effective one-velocity diffusion problem. The absorption and scattering mean free paths, λ_a and λ_s, will be adjusted so that the average number of collisions made by the source particles will be equal to the average number of collisions suffered by all the knock-ons.

Since the collision of a secondary particle with a lattice particle is an isotropic hard-sphere scattering, there will be equal probability for all possible energy transfers. Hence, the ratio of scattering with displacement to scattering without displacement will be approximately

$$R \cong (E-E_d)/E.$$

When E is large compared to E_d, there are very few scatterings without displacement. On the other hand, when E approaches E_d, it takes only one or two collisions for the particle to become trapped itself and effectively absorbed. It seems reasonable to assume that the number of scatterings without either displacement or absorption can be neglected compared to those resulting in either a knock-on or absorption.

The average number of collisions (including absorption) of all the knock-ons, $\bar{N}$, is equal to the sum of the number of scatterings without displacement, N_s, plus the number of displacement collisions, N_d, plus the total number of absorptions, N_a, all divided by the mean number of secondaries. Thus

$$\bar{N}=(N_s+N_d+N_a)/\bar{\nu}.$$

However, by the discussion in the previous paragraph, N_s may be neglected. In addition

$$N_d=\bar{\nu}-1,$$

and

$$N_a=\bar{\nu}.$$

Hence

$$\bar{N}=2-(1/\bar{\nu}) \cong 2.$$

It is quite unlikely that $\bar{N}$ can be much larger than this value. It will be shown at the end of this section that a value of $\bar{N}=3$ does not change the numerical results by a large factor.

This value of $\bar{N}$ may now be used to determine the ratio of scattering to absorption. It is clear that the average number of collisions $\bar{N}$, including absorptions, is

$$\bar{N}=(\Sigma_s+\Sigma_a)/\Sigma_a.$$

If $\bar{N}=2$, this requires that $\Sigma_s=\Sigma_a$ and this is the choice that is made in the diffusion calculation which follows.

[3] Reference 2, p. 380

The diffusion equation in plane geometry for a constant volume source $\bar{\nu}q$ is

$$-Dd^2\phi/dx^2+\Sigma_a\phi=\bar{\nu}q, \tag{15}$$

where ϕ is the particle flux and where the diffusion coefficient D is

$$D=[3\Sigma_s(1-\bar{\mu})]^{-1}. \tag{16}$$

Here $\bar{\mu}$ is the average cosine of the scattering angle in the laboratory system. For isotropic scattering in the center-of-mass system, one has $\bar{\mu}=2/3A\ll 1$ for all but the lightest elements. Hence $D\cong(3\Sigma_s)^{-1}$.

The solution of Eq. (15) is

$$\phi=(\bar{\nu}q/\Sigma_a)+Ce^{\kappa x} \tag{17}$$

where $\kappa=[\Sigma_a/D]^{\frac{1}{2}}$ and the negative solution is thrown out by the boundary condition $x=-\infty$. The usual diffusion-theory boundary condition at a vacuum interface is the vanishing of the flux at the extrapolated end point, $x=d$, where in our case $d=0.71\Sigma_s^{-1}$. Hence

$$\phi=(\bar{\nu}q/\Sigma_a)[1-e^{\kappa(x-d)}]. \tag{18}$$

The surface current density is now

$$J=-Dd\phi/dx\,|_{x=0}.$$

Hence

$$\begin{aligned} J&=\bar{\nu}q(D/\Sigma_a)\kappa e^{-\kappa d}\\ &=\bar{\nu}q(D/\Sigma_a)^{\frac{1}{2}}e^{-\kappa d}. \end{aligned} \tag{19}$$

By use of Eq. (3), one has

$$\gamma=\bar{\nu}(D/\Sigma_a)^{\frac{1}{2}}e^{-\kappa d}. \tag{20}$$

Finally, since $\Sigma_a=\Sigma_s$ in our case,

$$\begin{aligned} \gamma&=\frac{\bar{\nu}}{\sqrt{3}\Sigma_s}\exp[-(0.71)\sqrt{3}]\\ &=0.17\bar{\nu}/\Sigma_s. \end{aligned} \tag{21}$$

It should be noted that if $\bar{N}=3$, this result is changed to $0.34\bar{\nu}/\Sigma_s$ which is larger by a factor of 2.

VI. SUMMARY

Upon combining the results in Eqs. (5), (10), (11), and (21), the following expression for the sputtering ratio is obtained:

$$\mathfrak{R}=\frac{0.17\bar{\nu}}{\Sigma_s}\frac{n_0}{\cos\psi}\left(\pi\frac{M_1}{M_2}\frac{z_1^2z_2^2e^4}{EE_d}\right).$$

The macroscopic scattering cross section for the secondaries is hard-sphere scattering and is equal to $n_0\pi R^2$ with R given by Eq. (12). Hence,

$$\mathfrak{R}=\frac{0.17\bar{\nu}M_1z_1^2z_2^2e^4}{M_2EE_dR^2\cos\psi}, \tag{22}$$

with

$$\bar{\nu}=\left[0.885+0.561\ln\left(\frac{x+1}{4}\right)\right]\left(\frac{x+1}{x}\right),$$

$$x=\frac{4M_1M_2}{(M_1+M_2)^2}\left(\frac{E}{E_d}\right),$$

and with R the solution of the transcendental Eq. (12). Equation (12) is not very sensitive to the energy E' which should be chosen equal to some value between E_d and $E_d\ln x$.

At high energies, x is large compared to unity and the general behavior of the sputtering ratio is

$$\mathfrak{R}\propto\frac{M_1}{M_2}\frac{\ln E}{E}\frac{1}{\cos\psi}. \tag{23}$$

In our illustrative case, 500-kev deuterons striking a copper target at normal incidence, the numerical value of Eq. (22) is

$$\mathfrak{R}=6.5\times10^{-4}.$$

Experimental data in this energy range seem unavailable.

The numerical results obtained from the general formula of Eq. (22) are probably trustworthy only in their order of magnitude. The dependence on incident energy, angle, and mass ratio may be more reliable.

It should be noted that the possibility of focusing effects[4] has been ignored. The validity of this assumption must await experimental investigation.

Note added in proof.—In a recent paper, E. J. Sternglass [Phys. Rev. **108**, 1 (1957)] has used somewhat similar methods in analyzing secondary electron emission by high-speed ions.

[4] R. H. Silsbee, J. Appl. Phys. **28**, 1246 (1957).