PHYSICAL REVIEW B VOLUME 52, NUMBER 23 15 DECEMBER 1995-I

Theory of sputter yield fluctuations and calculation of the variance

I. Pázsit
Department of Reactor Physics, Chalmers University of Technology, S-412 96 Göteborg, Sweden

A. K. Prinja
Chemical and Nuclear Engineering Department, University of New Mexico, Albuquerque, New Mexico 87131
(Received 4 April 1995; revised manuscript received 2 August 1995)

A backward-type master transport equation is derived for the distribution of sputtered particles in a multispecies medium due to one incident foreign particle. The formalism is suitable for determining both angular and energy densities and correlations, but in this paper it is only used for the distribution of the total yield. The equations for the mean (densities and yields) are equivalent with standard theory. The solution of the equation for the second moment can be given through integrals of first-moment (the flux and its adjoint) functions. Some specific cases and corresponding formal solutions and solution methods are discussed in detail. For illustration, an explicit solution is given for the mean and the variance of the total sputtering yield, as a function of the energy of the incident particle, in a semi-infinite homogeneous single-species medium employing a simple collision model.

I. INTRODUCTION

Atomic collision cascades in an amorphous medium are traditionally described by means of linear transport theory.[1–3] In a medium with a free surface, a collision cascade leads to sputtering or reflection of the bombarding particle, which can also be described by means of linear transport theory.[4] Although most often the cascade is described by mean values, fluctuations in a collision cascade are also a matter of interest. One such situation is when one wants to confirm that statistical deviations from the mean may be disregarded. A second reason is that the fluctuations may carry information on the physics of the cascade or properties of the host material which is not contained in the mean values.

The first studies of fluctuations in particle cascades, those in connection with electron-photon cosmic showers, had their incentives in the first class of reasons above.[5–8] This may in fact be the case in general regarding fluctuations in atomic collision cascades.[9–14] On the other hand, in the field of neutron transport in a multiplicative medium, it was the second type of argument that motivated a stochastic theory. That is, it was found that the variance of the number of detector counts was a more sensitive function of several nuclear parameters than the mean value. Moreover, the variance-to-mean ratio is independent of detector efficiency or the strength of the external neutron source. Thus measurement of the variance-to-mean ratio provided an effective tool for determining absolute values of certain physical parameters without additional calibration.

One possible use of a thorough understanding of the fluctuations in collision processes could be the determination of material properties from fluctuation data. Such a study was performed by Vicanek and Urbassek[15] in connection with fluctuations in light ion-induced kinetic electron emission. One objective of the present paper is to contribute to the development of this field with a transport theoretical treatment of sputter yield fluctuations.

The above objective is endorsed by the fact that sputter fluctuations are known to be much larger than fluctuations in, e.g., specific ionization[9–11] and defect creation in an infinite medium.[16] This fact has already been pointed out theoretically in Ref. 17, and has been confirmed via Monte Carlo simulations Refs. 18 and 19. In Ref. 19, a uniform quantitative dependence of the various moments on energy was found. The authors also derived empirical scaling laws for the asymptotic behavior at high energy.

To our knowledge, this is the first report on quantitative results from analytical solutions regarding sputter yield fluctuations. A general theory for fluctuations in collision cascades was given, e.g., in Ref. 20. In the present paper this theory is adapted for treating sputtering problems. A general solution for the variance of the total-energy-dependent sputtering yield is given in the form of an integral over first-moment functions. Particular cases such as a single-species medium will be discussed in some detail, and useful forms of the integral expressing the variance are given.

One explicit solution is presented and evaluated quantitatively. It is based on a simple one-dimensional model with constant cross sections and forward-backward scattering for a single-species host medium with the same type of bombarding particle. The variance of the total sputtering yield and other related quantities such as the relative variance and relative standard deviation are displayed and discussed. The results show that two energy regions with markedly different characteristics can be distinguished. For low energies, up to about $E=40E_d$ (where E_d is the displacement threshold), there exists a nearly constant, less than unity Fano factor (relative variance). For higher energies the relative variance starts to diverge, while the relative standard deviation becomes approximately constant. These results are in a very good agreement with the numerical findings in Ref. 19, indicating that this very simple model is capable of reconstructing basic properties of the sputtering statistics.

It will be seen that evaluation of the analytical expression of the variance, as well as the transport theory calculation of the particle densities required for this evaluation, is rather involved even in the oversimplified collision model used here. In a real three-dimensional (3D) case with realistic cross sections, it is not possible to calculate even the first-moment densities analytically. It would thus appear that Monte Carlo methods are easier and more straightforward to use. However, if the first-moment densities in the variance integral can be determined numerically, evaluation of the variance integral will not be more complicated than in the present case. Analytical-numerical methods can thus be expected to yield good quality data with modest computing time. In addition, analytical scaling laws may be derived from the closed-form solutions.

A natural objection against sputter fluctuation studies is that currently individual realizations of sputter yields cannot be measured and thus no comparison with measurements is possible. This obstacle may eventually be removed by using time-resolved measurements with stationary ion beams. The theory for fluctuations in collision cascades induced by a steady source was also described in Ref. 20. Moreover, there are experimental values available regarding secondary electron yield distributions.[15] The present methods can be applied for treating such problems as well.

II. STOCHASTIC THEORY OF SPUTTERING

The sputtering process will be treated with the following assumptions. We consider a semi-infinite polycrystalline or amorphous target containing n different atomic species. Channeling effects and focused collision chains are thus not included. Also, we treat the cascade in the linear regime; that is, we assume a low density of both the energetic recoils and vacancies as well as negligible surface erosion,[21] although this may not be a good approximation at the low-energy tail of the cascade.[22] Recoil and defect production will be described by the damage model of Khinchin and Pease,[23] which assumes a deterministic displacement energy E_d^k for species k. Only binary collisions are considered, with a macroscopic collision cross section $d\sigma_{ij}=\sigma_{ij}(\underline{r},\underline{v}\rightarrow\underline{v}',\underline{v}'')d\underline{v}'d\underline{v}''$. As was remarked, e.g., in Ref. 4, this scattering function can account for both elastic and inelastic scattering, including electronic stopping. However, as noted in Ref. 24, if one is not interested in following up the history of the electrons, it is simpler to treat electronic stopping with a continuous energy-loss model. It is this method which we will follow in this paper. The energy-loss mechanism will then be a deterministic process; that is, it will not directly contribute to the fluctuation of the sputtering process. This is not a restrictive assumption since the overwhelming part of the fluctuations is due to discrete collisions. Nevertheless, the existence of electronic stopping, whether deterministic or not, influences the statistics, and thus cannot be completely neglected.

The quantity of interest in this paper is the probability distribution $P_j^k(N|x,E,\eta)$, which is the probability that there will be altogether N sputtered atoms of type k with energies between E and E_d^k, as a result of one projectile at depth x and with energy E and directional cosine η and type j. Here, as in all later work, it will be assumed that the analyzing detector has azimuthal symmetry; thus slab geometry can be used.

A backward-type master transport equation for P can then be derived by writing P as the sum of mutually exclusive possibilities of the projectile suffering and not suffering a collision in $ds=dx/\eta$, respectively:

$$P_j^l(N|x,E,\eta)=\left[1-\frac{dx}{\eta}\sigma_j\right]P(N|x+dx,E-dE,\eta)+\frac{dx}{\eta}\sum_m\int d\sigma_{jm}\sum_{n=0}^{N}P_j^l(N-n|x,E',\eta'')\times P_m^l(n|x,E'',\eta'')\ , \tag{1}$$

where $\sigma_j=\sum_m\sigma_{jm}$ is the total cross section of species j. This equation can be converted into an integrodifferential equation as

$$\eta\frac{\partial}{\partial x}P_j^l(N|x,E,\eta)-S_j(E)\frac{\partial}{\partial E}P_j^l(N|x,E,\eta)=\sum_m\int d\sigma_{jm}\left\{P_j^l-\sum_{n=0}^{N}P_j^l(N-n|x,E',\eta')\times P_m^l(n|x,E'',\eta'')\right\}\ , \tag{2}$$

where

$$S_j(E)=\eta\frac{dE}{dx}=\frac{dE}{ds} \tag{3}$$

is the electronic stopping power for species j. The boundary condition on Eq. (2) is

$$P_j^l(N|0,E,\eta)=\delta_{j,l}\Theta(E-U_j)(\delta_{N,1}-\delta_{N,0})+\delta_{N,0}\ ,\quad \eta<0\ , \tag{4}$$

where U_j is the surface barrier energy. As was remarked, e.g., in Ref. 20, the equation would have exactly the same form if instead of the yield of one species, the angular and/or energy-dependent joint distribution of several particle types were sought, or even the joint distribution of sputtered particles and energetic particles in the medium. The type of distribution sought would only affect the boundary condition in Eq. (4).

Following standard procedure, we introduce the generating function

$$G_j^k(Z,x,E,\eta)\equiv\sum_{N=0}^{\infty}Z^NP_j^k(N|x,E,\eta)\ , \tag{5}$$

which satisfies

$$\eta\frac{\partial}{\partial x}G_j^l(Z,x,E,\eta)-S_j(E)\frac{\partial}{\partial E}G_j^l(Z,x,E,\eta)$$
$$=\sum_m\int d\sigma_{jm}\{G_j^l(Z,x,E,\eta)-G_j^l(Z,x,E',\eta')G_m^l(Z,x,E'',\eta'')\}\ . \tag{6}$$

The boundary condition for (6) is obtained from (4) as

$$G(Z,0,E,\eta)=\delta_{j,l}\Theta(E-U_j)(Z-1)+1,\quad \eta<0\ . \tag{7}$$

The moments of the distribution can be obtained as derivatives of the generating function. Thus, defining $N_j^l(x,E,\eta)\equiv\langle N\rangle$, i.e., the expected number of l-type particles escaping the medium due to a fast ion of type j at (x,E,η), one can show that

$$N_j^l(x,E,\eta)=\sum_{N=0}^{\infty}NP_j^l(N|x,E,\eta)=\frac{\partial G_j^l}{\partial Z}\bigg|_{Z=1}\ . \tag{8}$$

Applying (8) to (6) and (7), the equation for the mean number of sputtered particles becomes

$$\eta\frac{\partial}{\partial x}N_j^l(x,E,\eta)-S_j(E)\frac{\partial}{\partial E}N_j^l(Z,x,E,\eta)$$
$$=\sigma_j(E)N_j^l-\sum_m\int d\sigma_{jm}\{N_j^{l'}+N_m^{l''}\}+\delta(x)\theta(E-U_l)\eta\theta(-\eta)\delta_{jl}\ , \tag{9}$$

where the boundary condition has been incorporated as a source.

Defining the angular density of energetic recoils of type l at (x_0,E_0,η_0), initiated by a fast ion of type j at (x,E,η) as $\phi_j^l(x,E,\eta\rightarrow x_0,E_0,\eta_0)$, which is also a Green's function, it can be easily shown that N_j^l can be expressed as

$$N_j^l(x,E,\eta)=-\int_{U_l}^{E}\int_{-1}^{0}\phi_j^l(x,E,\eta\rightarrow 0,E_0,\eta_0)\eta_0 dE_0 d\eta_0\ . \tag{10}$$

Equation (10) shows again that N_j^l is the (positive) number of particles leaving the free surface in the negative direction; N is thus a current, whereas ϕ is an angular density.

Similarly, now defining M_j^l as

$$M_j^l(x,E,\eta)\equiv\langle N(N-1)\rangle\ , \tag{11}$$

then it follows from (5) that

$$M_j^l(x,E,\eta)=\frac{\partial^2}{\partial Z^2}G_j^l(Z,x,E,\eta)\bigg|_{Z=1}\ , \tag{12}$$

which for the variance of the sputtering yield gives

$$\sigma_N^2=\langle N^2\rangle-\langle N\rangle^2$$
$$=M_j^l(x,E,\eta)+N_j^l(x,E,\eta)-N_j^l(x,E,\eta)^2\ , \tag{13}$$

and for the variance-to-mean ratio (Fano factor)

$$F(x,E,\eta)=\frac{M_j^l}{N_j^l}+1-N_j^l\ . \tag{14}$$

With a double differentiation, from Eqs. (6) and (7) one obtains

$$\eta\frac{\partial}{\partial x}M_j^l(x,E,\eta)-S_j(E)\frac{\partial}{\partial E}M_j^l(x,E,\eta)=\sigma_jM_j^l-\sum_m\int d\sigma_{jm}\{M_j^l(x,E',\eta')+M_m^l(x,E'',\eta'')\}$$
$$-2\sum_m\int d\sigma_{jm}N_j^l(x,E',\eta')N_m^l(x,E'',\eta'')\ , \tag{15}$$

with

$$M_j^l(0,E,\eta)=0,\quad \eta<0\ . \tag{16}$$

As was shown, e.g., in Ref. 20, the solution of Eq. (17) for $M_j^l(0,E,\eta)$ with a positive η can easily be given as

$$M_j^l(0,E,\eta)=2\sum_k\int dx_0dE_0d\eta_0\phi_j^k(0,E,\eta\rightarrow x_0,E_0,\eta_0)$$
$$\times\sum_m\int d\sigma_{km}N_k^l(x_0,E_0',\eta_0')N_m^l(x_0,E_0'',\eta_0'')\ . \tag{17}$$

This expression can be further expanded if in the second integral the sputtering yields are also generated by the Green's function ϕ_j^l:

$$M_j^l(0,E,\eta)=2\sum_k\int dx_0dE_0d\eta_0\phi_j^k(0,E,\eta\rightarrow x_0,E_0,\eta_0)$$
$$\times\sum_m\int d\sigma_{km}\phi_k^l(x_0,E_0',\eta_0'\rightarrow 0,E',\eta')\phi_m^l(x_0,E_0'',\eta_0''\rightarrow 0,E'',\eta'')\eta' d\eta' dE'\eta'' d\eta'' dE''\ . \tag{18}$$

Thus the variance is completely determined once the Green's function for the mean is known.

III. SELF-SPUTTERING OF A MONATOMIC TARGET

To simplify numerical work we restrict our considerations to a single species. One can drop all indices and use N and M for the mean and $\langle N(N-1)\rangle$, respectively. The relevant equations are then

$$\left[\eta\frac{\partial}{\partial x}-S(E)\frac{\partial}{\partial E}\right]N(x,E,\eta)=\int d\sigma\{N(x,E,\eta)-N(x,E',\eta')-N(x,E'',\eta'')\} \tag{19}$$

and

$$M(0,E,\eta)=2\int dx_0 dE_0 d\eta_0 \phi(0,E,\eta\rightarrow x_0,E_0,\eta_0)\int d\sigma\, N(x,E',\eta')N(x,E'',\eta'')\ . \tag{20}$$

Using the single-species version of (10) to express N, one obtains

$$M(0,E,\eta)=2\int dx_0 dE_0 d\eta_0 \phi(0,E,\eta\rightarrow x_0,E_0,\eta_0)$$
$$\times\int d\sigma \int_U^E\int_{-1}^0 \phi(x_0,E_0',\eta_0'\rightarrow 0,E',\eta')\int_U^E\int_{-1}^0 \phi(x_0,E_0'',\eta_0''\rightarrow 0,E'',\eta'')\eta' d\eta' dE' \eta'' d\eta'' dE''\ . \tag{21}$$

It is without any doubt that even in possession of the Green's function, evaluation of (21) is a very complicated task in view of the fact that it contains an 11-fold integration. Due to energy conservation, the scattering kernel usually contains a function $\delta(E-E'-E'')$ which makes evaluation of one integral possible. For that matter, if $N(x,E,\eta)$ can be directly determined without using the Green's function, the evaluation of (20) may be simpler, since the number of integrals is reduced. In view of the complexity of the expressions (20) or (21), Monte Carlo techniques may be more promising to calculate the variance. However, the closed-form solutions above may be evaluated by approximations or simplification of the scattering kernel, making some insight possible into the physics of the problem. One such case is analyzed below in some detail.[25]

Following Dederichs,[26] and denoting the energy E'' transferred to the recoil by T, the scattering function can be expressed as

$$\sigma(E_0,\underline{\Omega}_0\rightarrow E',\underline{\Omega}',E'',\Omega'')$$
$$=\sum_{l,m}\sigma(E_0,T)a_{lm}(E_0,T)P_l(\eta_0')$$
$$\times P_m(\eta_0'')\delta(E-E'-T)\ , \tag{22}$$

where

$$a_{lm}(E_0,T)=P_l\left[\left[1-\frac{T}{E_0}\right]^{1/2}\right]P_m\left[\frac{T}{E_0}\right]^{1/2}\ , \tag{23}$$

$\eta_0'\equiv\underline{\Omega}_0\cdot\underline{\Omega}'$ and $\eta_0''\equiv\underline{\Omega}_0\cdot\underline{\Omega}''$, and $P_l(\eta)$ is the Legendre polynomial of degree l. Assuming an expansion of the mean sputtered yield in the form

$$N(x,E,\eta)=\sum_l N_l(x,E)P_l(\eta)\ , \tag{24}$$

and a similar expansion for the Green's function as

$$\phi(0,E,\eta\rightarrow x_0,E_0,\eta_0)$$
$$=\sum_k n_k(x_0,E\rightarrow E_0)P_k(\eta)P_k(\eta_0)\ , \tag{25}$$

after the usual manipulations, for the variance one obtains

$$M(0,E,\eta)=2\sum_k\sum_l\sum_m P_k(\eta)A_{klm}\int dx_0\int_U^E dE_0 n_k(x_0,E\rightarrow E_0)\int dT\,\sigma(E_0,T)a_{lm}(E_0,T)N_l(x_0,E_0-T)N_m(x_0,T)\ , \tag{26}$$

where

$$A_{klm}=\int_{-1}^1 P_k(\eta)P_l(\eta)P_m(\eta)d\eta \tag{27}$$

is a Wigner factor. In case of isotropic scattering in the center-of-mass (CM) system and energy-independent cross sections, $\sigma(E_0,T)=\sigma=$const, and the evaluation of (26) simplifies further. It is thus seen that by application of the spherical harmonics expansion, the expression for M is significantly reduced. If a few term series expansion for N and ϕ can be employed, such that the sums in k, l, and m can be truncated after a few terms, then using (26) can be a powerful way of evaluating the variance.[27,28]

In order to gain some insight through quantitative results, a solution will be given here for the energy dependence of the variance in a very simple model. Electronic stopping will be neglected. The surface barrier energy will be assumed to be equal to the displacement energy E_d. The transport will be assumed to be purely one dimensional, such that particles can only move in forward and backward directions along the x axis. This model has been used in studies of neutron transport and criticality calculations,[29,30] radiation damage,[31] and sputtering studies.[32] The model has clearly nonphysical features; nevertheless, it allows solutions to be obtained with reasonable effort and may still display some basic trends of the proper solution.

One important advantage of the model is that it allows the energy and angular dependences to be decoupled. This enables the angular integrals to be evaluated in the transport equation and also in the expression of the variance. The corresponding transport equation can then be Laplace transformed in lethargy $u=\ln(E/E_d)$, and it will formally be equivalent to a one-speed equation. For such cases, as is mentioned in Ref. 20, the optical reciprocity theorem holds,[33] and this fact can be utilized to simplify the calculation of the variance, as will be seen below.

The scattering model we shall consider here uses constant cross sections and hard-sphere scattering, combined with the one-dimensional forward-backward scattering model. Thus one has

$$\sigma(E,\eta \to E',\eta',E'',\eta'') = \frac{\delta(E-E'-E'')}{4E}[\delta(\eta-\eta')+\delta(\eta+\eta')] \times [\delta(\eta-\eta'')+\delta(\eta+\eta'')] , \quad (28)$$

with $\eta = \pm 1$ as only possible values. Since the sputtering yield is invariant to a scaling of the mean free path, $\sigma_T = \int d\sigma = 1$ was chosen in (28).

We shall assume a homogeneous semi-infinite monatomic target, with one fast atom of the same type as the host material impinging on the free surface with $\eta = +1$ and energy E. We assume a displacement energy E_d equal to the surface binding energy, which will be accounted for by setting all transport functions (sputtering yield and Green's function) to zero below E_d. We shall seek the expected value and the variance of the total number of sputtered particles.

The mean sputtering yield is then given by $N(0,E,\eta=1)$. Solution of Eq. (19) with the scattering model (28) was given, e.g., in Ref. 34, with the result

$$\langle Y \rangle \equiv N(u) \equiv N(0,E,\eta=1) = \int_0^u \frac{I_1(u')}{u'} du' , \quad (29)$$

where I_1 is the modified Bessel function of first kind and order 1, and $\langle Y \rangle$ is the mean sputtering yield.

As it was shown in Ref. 35, in this special case all that is needed to evaluate the variance is the Green's function

$$\bar{\phi}(x,u) \equiv \int \phi(u;x,\eta \to 0, \eta_0 = -1) d\eta \equiv \int \phi(u;0,\eta = +1 \to x, \eta_0) d\eta_0 ,$$

where, due to reciprocity, $\bar{\phi}(x,u)$ is both the density of the escaping particles due to an isotropic source at x and also the scalar (angularly integrated) particle density at x due to one projectile at the surface. The solution for $\bar{\phi}$ was given by Lux and Pázsit[32] as

$$\bar{\phi}(x,u) = \frac{e^{-x}}{E} \sum_{j=0}^{\infty} L_j(2x)[\mathcal{F}_j(u) + \delta_{j,0}\delta(u)] , \quad (30)$$

where

$$\mathcal{F}_j(u) = \frac{(-1)^j e^u}{u}[jI_j(u) + 2(j+1)I_{j+1}(u) + (j+2)I_{j+2}(u)] . \quad (31)$$

Using the above, a simple but lengthy calculation yields, for $M(u)$,

$$M(u) = \frac{1}{2} \sum_i \sum_j \sum_k A_{ijk} \{\alpha_{ijk}(u) + \delta_{i,0}\beta_{jk}(u)\} , \quad (32)$$

where

$$A_{ijk} = \int_0^\infty dx\, e^{-3x} L_i(2x) L_j(2x) L_k(2x) = \frac{1}{3} \sum_{l=0}^{i} \sum_{m=0}^{j} \sum_{n=0}^{k} \begin{bmatrix} i \\ l \end{bmatrix} \begin{bmatrix} j \\ m \end{bmatrix} \begin{bmatrix} k \\ n \end{bmatrix} \left[-\frac{2}{3} \right]^{l+m+n} \times \frac{(l+m+n)!}{l!m!n!} , \quad (33)$$

$$\alpha_{ijk}(u) = \int_{\ln 2}^{u} du' \mathcal{F}_i(u-u') \times \int_0^{\ln(e^{u'}-1)} du'' \mathcal{H}_j(u'') \times \mathcal{H}_k[\ln(e^{u'}-e^{u''})] e^{-(u'-u'')} , \quad (34)$$

$$\beta_{jk}(u) = \int_0^{\ln(e^{u'}-1)} du' \mathcal{H}_j(u') \times \mathcal{H}_k[\ln(e^u - e^{u'})] e^{-(u-u')} , \quad (35)$$

and

$$\mathcal{H}_j(u) = \int_0^u \mathcal{F}_j(u) + \delta_{j,0} . \quad (36)$$

From $M(u)$ the variance $\Delta Y^2 \equiv \sigma^2(u)$, the relative variance $F(u) \equiv \Delta Y^2 / \langle Y \rangle$ and the relative standard deviation $\sigma(u)/N(u) \equiv \Delta Y / \langle Y \rangle$ of the sputtering yield can be calculated.

IV. NUMERICAL RESULTS AND DISCUSSION

First we discuss the behavior of the mean $\langle Y \rangle$ and the relative variance $F(u)$. Calculated values are shown in Fig. 1 as functions of u. The numerical values of both the mean and variance were checked with Monte Carlo calculations[35] with a very good agreement, as seen in the figure.

The behavior of $\langle Y \rangle = N(u)$ is well known and rather simple. It is monotonically increasing, due to the fact that there will be more and more recoils created by in-

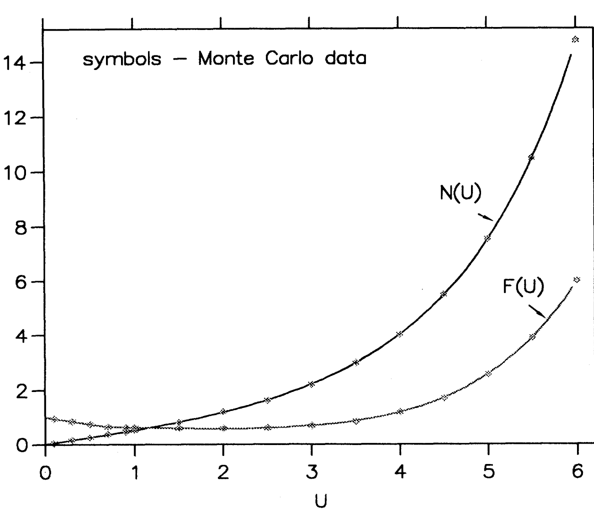

FIG. 1. Dependence of the total yield $N(u)$ and the variance-to-mean ratio $F(u)$ on the lethargy $u = \ln(E/E_d)$.

creasing u. It may be interesting to make a comparison with the number of Frenkel pairs N_F created in an infinite medium:[16,23]

$$N_F = \frac{E}{2E_d} = \frac{e^u}{2} . \tag{37}$$

Comparing (37) with an upper bound on (29), obtained from an asymptotic expansion for large u,

$$\langle Y \rangle \sim \frac{e^u}{u^{1/2}} , \tag{38}$$

shows that the expected value of the sputtering yield increases much slower with energy than the number of defects created. The reason is clearly that in the Khinchin-Pease model, all particles that slowed down past E_d lead to a defect; on the other hand, in sputtering, only a fraction of the energetic recoils will escape (those created near to the surface), and even the total number of generated recoils is lower, due to the leakage process.

The behavior of $F(u)$ displays another pattern, in which two distinct areas can be identified. For $u < \ln 2$, $F(u)$ is decreasing. This is because for projectile energies $E < 2E_d$, at most one particle can leave the collision site; thus one has $P(N \geq 2) = 0$ and $\langle N \rangle^2 = N$. Thus, for $u < \ln 2$, Eq. (29) yields

$$F(u) = 1 - \langle Y \rangle \sim 1 - \frac{u}{2} . \tag{39}$$

Above $u = \ln 2$, the probability of two sputtered particles becomes greater than zero, and at that point $F(u)$ has a break. It can be shown that for $u = \ln 2 + \varepsilon$ and $|\varepsilon| \ll 1$, $M(u) \sim \varepsilon/6$. This results in a nearly constant $F(u)$ for u slightly above ln2. $F(u)$ remains nearly constant and less than unity up to $u \sim 3$. It reaches unity at $u = 3.7$; that is, at $E \approx 40E_d$, or at ≈ 1 keV with $E_d = 25$ eV.

For larger values of u the relative variance starts to increase, and it diverges asymptotically with the same rate as the mean. That is, asymptotically $F(u) \sim \langle Y \rangle$. From the latter it follows that the relative standard deviation is asymptotically constant for high energies. This latter is illustrated in Fig. 2, which shows the mean, the variance, and the relative standard deviation on a logarithmic scale. The data in Fig. 2 are in a very good qualitative agreement with the energy dependence of the same quantities found by Conrad and Urbassek (see Fig. 1 in Ref. 19), who found the asymptotically constant relative standard deviation in Monte Carlo simulations with realistic cross sections. This somewhat surprisingly good agreement between the forward-backward model and the realistic 3D model of Ref. 19 shows that the former is actually capable of reconstructing the most important properties of the sputtering statistics.

It is thus seen that the second moment has two differing domains of behavior. For low energies, in the present model up to $u \sim 3.5$, there exists a sputter Fano factor; that is, a nearly constant, less than unity relative variance. For higher energies, the relative variance increases and hence no Fano factor exists, but instead one can define a relative Fano factor (relative variance divided by the mean) since the relative standard deviation becomes constant.

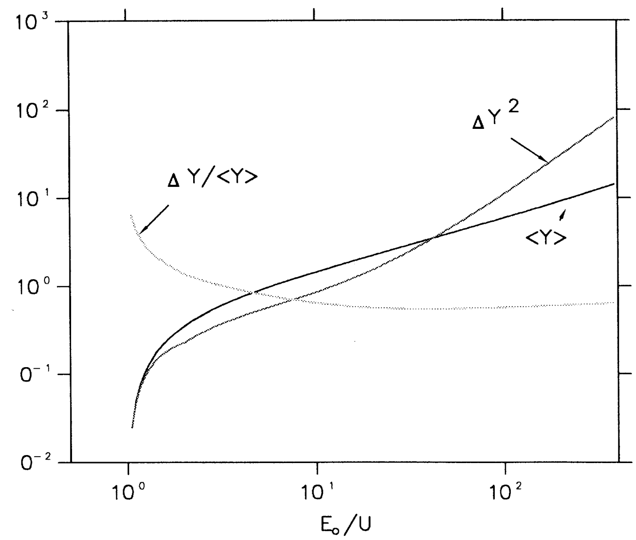

FIG. 2. Mean yield $\langle Y \rangle$, variance ΔY^2, and relative standard deviation $\Delta Y / \langle Y \rangle$ of the sputtering yield distribution.

Again, it can be useful to make a comparison with the variance-to-mean ratio of the number of Frenkel pairs F_F in an infinite medium in the same model. This was calculated by Leibfried[16] with the result $F_F \approx 0.15$ for energies larger than $4E_d$. In other words, the relative variance of the defect creation in an infinite medium is asymptotically constant and significantly below unity.

It is thus seen that the fluctuations in the sputtering process are asymptotically much higher than in other collision processes, such as defect creation. This was already noticed in Refs. 17 and 19 and interpreted as being due to fluctuations in the energy deposited in low-energy recoil motion close to the surface. In the case of ion-induced secondary electron emission, the high fluctuations were explained with the fluctuations in the ion trajectory.[15] A complementary view of the phenomenon is as follows. Whether in a cascade (leading to defects or sputtered particles) $F > 1$ or $F < 1$ depends on the correlations between particles with different energies or spatial points. The former ($F > 1$) prevails if positive correlations dominate, and vice versa. In the sputtering process, for $u < \ln 2$, there are negative correlations, since occurrence of a sputtered particle excludes the possibility of another particle. For $u > \ln 2$, there may be more than two sputtered particles, often from the same or successive collisions, and thus there will be positive correlations. Further, collisions near to the surface will likely lead to several sputtered particles, whereas collisions deep in the medium will not lead to sputtering at all. Thus the site of any collision, but especially the location of the first collision, has a large significance on the total yield. All this leads to large fluctuations. On the other hand, in the defect creation process, at least in an infinite medium, negative correlations will dominate. A recoil whose energy has decreased below E_d, and which thus becomes an interstitial, cannot lead to further defects, and this gives a negative correlation.

V. CONCLUSIONS AND FURTHER WORK

A closed-form solution for the variance of the sputtering yield was given and evaluated in a simple concrete case. The solution is in agreement with previous work, both theoretical and numerical.

The possibility of an analytical solution in the present work was a consequence of a number of simplifications regarding the scattering cross sections. However, in realistic models, numerical solutions for the first-moment densities can be obtained whose integration in the variance formula will presumably not be more complicated than in the case treated here. There is also some chance that asymptotic behavior and analytical scaling laws may be derived.

One complication is that direct comparison with measurements is not possible, but one can get around the problem by calculating the sputter yield fluctuations for a steady source, as outlined in Ref. 20. A more straightforward field of application may be a study of the statistics of ion-induced electron emission,[15] since experimental data are available there.

ACKNOWLEDGMENTS

This subject was first suggested during a visit by one of us (I.P.) to QMC, London, by Professor M. M. R. Williams, whose advice is appreciated by both authors. We thank Professor P. Sigmund and Dr. M. Vicanek for useful comments and discussions.

[1]J. Lindhard, V. Nielsen, M. Scharff, and P. V. Thomson, Mat. Fys. Medd. Dan. Vid. Selsk. **33**, 10 (1963).

[2]J. Lindhard, V. Nielsen, and H. E. Schiott, Mat. Fys. Medd. Dan. Vid. Selsk. **33**, 14 (1963).

[3]J. Lindhard, V. Nielsen, and M. Scharff, Mat. Fys. Medd. Dan. Vid. Selsk. **36**, 14 (1968).

[4]P. Sigmund, Phys. Rev. **184**, 383 (1969).

[5]L. Jánossy, Proc. Phys. Soc. London Sect. A **63**, 241 (1950).

[6]L. Jánossy, Proc. R. Irish Acad. **53**, 181 (1950).

[7]L. Jánossy and H. Messel, Proc. Phys. Soc. London Sect. A **63**, 1101 (1950).

[8]H. Messel, *Progress in Cosmic Ray Physics* (North-Holland, Amsterdam, 1954), Vol. II.

[9]U. Fano, Phys. Rev. **70**, 44 (1946).

[10]U. Fano, Phys. Rev. **72**, 26 (1947).

[11]U. Fano, Phys. Rev. **92**, 328 (1953).

[12]K. B. Winterbon, Radiat. Eff. **60**, 199 (1982).

[13]H. H. Andersen, Nucl. Instrum. Methods Phys. Res. Sect. B **15**, 722 (1986).

[14]P. Sigmund, Mat. Fys. Medd. Dan. Vid. Selsk. **36**, 14 (1978).

[15]M. Vicanek and H. Urbassek, Phys. Rev. B **47**, 7446 (1993).

[16]G. v. Leibfried, Nukleonik **1**, 57 (1958).

[17]J. E. Westmoreland and P. Sigmund, Radiat. Eff. **6**, 187 (1970).

[18]W. Eckstein, Nucl. Instrum. Methods Phys. Res. Sect. B **33**, 489 (1988).

[19]U. Conrad and H. M. Urbassek, Nucl. Instrum. Methods Phys. Res. Sect. B **48**, 399 (1990).

[20]I. Pázsit, J. Phys. D **20**, 151 (1987).

[21]P. Sigmund, Nucl. Instrum. Methods Phys. Res. Sect. B **27**, 1 (1987).

[22]H. H. Andersen, Nucl. Instrum. Methods Phys. Res. Sect. B **33**, 466 (1988).

[23]G. H. Khinchin and R. S. Pease, Rep. Prog. Phys. **18**, 1 (1955).

[24]M. M. R. Willians, Proc. R. Soc. London Ser. A **358**, 105 (1977).

[25]A. K. Prinja, Phys. Rev. B **39**, 8858 (1989).

[26]P. H. Dederichs, Phys. Status Solidi **10**, 303 (1965).

[27]K. T. Waldeer and H. M. Urbassek, Nucl. Instrum. Methods Phys. Res. Sect. B **18**, 518 (1987).

[28]K. T. Waldeer and H. M. Urbassek, Appl. Phys. A **45**, 207 (1988).

[29]M. M. R. Williams, J. Phys. D **11**, 2455 (1978).

[30]I. Pázsit, A. K. Prinja, and N. S. Garis, Ann. Nucl. Energy **21**, 432 (1994).

[31]I. Pázsit, Phys. Status Solidi B **104**, 119 (1981).

[32]I. Lux and I. Pázsit, Radiat. Eff. **59**, 27 (1981).

[33]G. Bell and S. Glasstone, *Nuclear Reactor Theory* (Van Nostrand Reinhold, New York, 1970).

[34]M. M. R. Williams, Ann. Nucl. Energy **6**, 145 (1979).

[35]I. Pázsit and A. K. Prinja (unpublished).

[36]R. Chakarova (unpublished).