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New fit formulae for the sputtering yield

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Abstract

New fit formulae for the energy and angular dependencies of the sputtering yield are proposed. Though they are empirical they give a better description of yield data, especially near the threshold for the energy dependence and at low mass ratios for the angular dependence. The new formula for the energy dependence was applied to determine threshold energies for different mass ratios.

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1. Introduction

The aim of this paper is to present new formulae for the sputtering yield, which describe the energy and angular dependence in the whole energy and angular range. New is a better description at low projectile energies close to the threshold. It has become obvious, that previous formulae for the energy and angular dependence fail near the threshold as demonstrated in [1]. The Bohdansky formula with two fit parameters is not able to fit the whole energy range if data below 10^{-3} for the sputtering yield are taken into account. The sputtering threshold behavior is of importance for fusion devices because of the plasma impurity problem [2]. Most particles hitting the first wall, especially in the divertor, have low energies and their flux is strongly increasing at low energies [2]. Also a new formula for the threshold energy at normal incidence is given, and for the first time a few examples for the angular dependence of the threshold energy are provided. The fitting is performed for pure elemental targets, but the new formulae presented may be valid for compounds, too, if for the target charge and mass the mean values are applied. They may not be used for projectiles in a high charged states and for clusters.

2. Simulation

The simulations are performed with the Monte Carlo program TRIM.SP (version trvmc95) [3,4]. The Kr–C interaction potential [5] is used for elastic collisions, an equipartition of the local Oen–Robinson [6] and of the nonlocal Lindhard–Scharff [7] models for the inelastic energy loss. The heat of sublimation is applied for the surface binding energy in the planar surface binding model.

3. New fitting formulae

In contrast to earlier fitting formulae a new approximation is used. In [8] the revised Bohdansky formula was applied to describe the energy dependence of the sputtering yield at normal incidence

$$Y(E_0) = Qs_n(\varepsilon)\left[1-\left(\frac{E_{th}}{E_0}\right)^{2/3}\right]\left(1-\frac{E_{th}}{E_0}\right)^2, \qquad (1)$$

where $s_n(\varepsilon)$ is the nuclear stopping. The remaining two terms containing the threshold energy E_{th} were added in

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order to describe the threshold behavior empirically according to data available at the time the formula was introduced [9]. In the revised Bohdansky formula the Kr–C interaction potential, which is a good mean potential for many species, is used to describe the nuclear stopping.

$$s_{\mathrm{n}}^{\mathrm{KrC}}(\varepsilon) = \frac{0.5\ln(1 + 1.2288\varepsilon)}{\varepsilon + 0.1728\sqrt{\varepsilon} + 0.008\varepsilon^{0.1504}} \tag{2}$$

with the reduced energy

$$\varepsilon = E_0 \frac{M_2}{M_1 + M_2} \frac{a_{\mathrm{L}}}{Z_1 Z_2 e^2} = E_0 \varepsilon_L. \tag{3}$$

Z_1 and Z_2 are the atomic numbers, and M_1 and M_2 the masses of the projectile and the target atom, respectively. e is the electron charge. The Lindhard screening length a_{L} is given by

$$a_{\mathrm{L}} = \left(\frac{9\pi^2}{128}\right)^{1/3} a_{\mathrm{B}} \left(Z_1^{2/3} + Z_2^{2/3}\right)^{-1/2}, \tag{4}$$

where a_{B} is the Bohr radius. E_{th} is the threshold energy for sputtering, and E_0 is the incident energy of the projectile. Q and E_{th} are used as parameters. Some discrepancies in calculated values originate from the fact, that TRIM.SP uses the Lindhard screening length, whereas ACAT [10] applies the Firsov screening length (the exponents of the charge term are exchanged), which can differ for the same system by 4–12%. Yamamura and coworkers applied a small correction to the screening length in many cases, to get better agreement with experimental data, which was not done in the TRIM.SP calculations.

Newer calculated sputtering yields [8,11] give values below the threshold obtained from the fit with the revised Bohdansky formula. For this reason a new fit formula was developed.

$$Y(E_0) = q s_{\mathrm{n}}^{\mathrm{KrC}}(\varepsilon) \frac{\left(\frac{E_0}{E_{\mathrm{th}}} - 1\right)^{\mu}}{\lambda + \left(\frac{E_0}{E_{\mathrm{th}}} - 1\right)^{\mu}}. \tag{5}$$

In the new formula a free parameter q and the nuclear stopping term is kept, but now the threshold term appears in the nominator and in the denominator to make sure that the high energy behavior is not affected by the threshold term. Contrary to the Bohdansky formula an empirical exponent μ is assigned to this term in order describe the strength of the decrease near the threshold energy. An additional third parameter λ triggers the onset of the decrease, as was already proposed in [12].

The reason for changing also the formula for the angular dependence of the sputter yield is, that the often used Yamamura formula [13] does not represent the available data for all cases, especially for problems that occur at low incident energies and for selfbombardment, see [8,11]. The Yamamura formula is given by

$$Y(E_0, \alpha) = Y(E_0, 0)[\cos(\alpha)]^{-f} \exp\left\{ f \left[1 - \frac{1}{\cos\alpha}\right] \sin(\eta) \right\} \tag{6}$$

with

$$\eta = \pi/2 - \alpha_{\mathrm{opt}}, \tag{7}$$

where the angle of incidence α is counted from the surface normal, and α_{opt} is the angle of incidence for which the sputter yield has a maximum. f and η are used as fit parameters. The new fit formula

$$Y(E_0, \alpha) = Y(E_0, 0)\left\{ \cos\left[\left(\frac{\alpha}{\alpha_0}\frac{\pi}{2}\right)^c\right]\right\}^{-f} \times \exp\left\{ b\left(1 - 1\Big/\cos\left[\left(\frac{\alpha}{\alpha_0}\frac{\pi}{2}\right)^c\right]\right)\right\} \tag{8}$$

keeps most of the original Yamamura formula, but introduces additional physical information, that incident atoms (projectiles) may experience a binding energy E_{sp}, which creates an acceleration and a refraction towards the surface normal [4], so that an incidence angle of 90° is never reached. The parameter η is not used anymore, but a new parameter c is chosen. The new value α_0 is given by

$$\alpha_0 = \pi - \arccos\sqrt{\frac{1}{1 + E_0/E_{\mathrm{sp}}}} \geqslant \frac{\pi}{2}, \tag{9}$$

where the binding energy of projectiles, E_{sp}, has to be provided. For selfbombardment E_{sp} is equal to the surface binding energy E_{s} of target atoms; for noble gas projectiles $E_{\mathrm{sp}} = 0$, for hydrogen isotopes $E_{\mathrm{sp}} = 1$ eV is assumed.

4. Numerical method

In contrast to older publications Bayesian probability theory [14] was employed in order to determine the free parameters of Eqs. (5) and (8). In this framework the parameters are calculated as expectation values $\langle\mu\rangle$ over the posterior probability distribution $p(\mu \mid \vec{Y}^{\mathrm{data}}, I)$, i.e. the conditional probability of μ in the light of the data $\vec{Y}^{\mathrm{data}}$

$$\langle\mu\rangle = \frac{\int \mathrm{d}\mu\, \mu p(\mu \mid \vec{Y}^{\mathrm{data}}, I)}{\int \mathrm{d}\mu\, p(\mu \mid \vec{Y}^{\mathrm{data}}, I)}. \tag{10}$$

The I gives formal notion that there is additional information not explicitly stated in the formulae (e.g. positiveness of data, Gaussian distributed measurement uncertainty, ...). The second moment $\langle\mu^2\rangle$ of Eq. (10) is used in order to determine the error $(\langle\mu^2\rangle - \langle\mu\rangle^2)^{1/2}$. Marginalization over all remaining parameters $\vec{\theta}$ (e.g. in Eq. (5) it is $\vec{\theta}^{\mathrm{T}} = (q, \varepsilon, \lambda)$)

$$\langle\mu\rangle = \frac{\int \mathrm{d}\mu\,\mathrm{d}\vec{\theta}\,\mu p(\mu, \vec{\theta} \mid \vec{Y}^{\mathrm{data}}, I)}{\int \mathrm{d}\mu\,\mathrm{d}\vec{\theta}\, p(\mu, \theta \mid \vec{Y}^{\mathrm{data}}, I)} \tag{11}$$

and using Bayes theorem

$$p(\mu, \vec{\theta} \mid \vec{Y}^{\mathrm{data}}, I) = \frac{p(\vec{Y}^{\mathrm{data}} \mid \mu, \vec{\theta}, I)p(\mu, \vec{\theta} \mid I)}{p(\vec{Y}^{\mathrm{data}} \mid I)} \tag{12}$$

results finally in an integral over probability distributions which are easy to assign:

$$\langle\mu\rangle = \int \mathrm{d}\mu\,\mathrm{d}\vec{\theta}\,\mu \frac{p(\vec{Y}^{\mathrm{data}} \mid \mu, \vec{\theta}, I)p(\mu, \vec{\theta} \mid I)}{\int \mathrm{d}\mu'\,\mathrm{d}\vec{\theta}'\, p(\vec{Y}^{\mathrm{data}} \mid \mu', \vec{\theta}', I)p(\mu', \vec{\theta}' \mid I)}. \tag{13}$$

The first one is the likelihood function

$$p(\vec{Y}^{\mathrm{data}} \mid \mu, \vec{\theta}, I) = \frac{1}{Z_l} \exp\left\{ -\frac{1}{2} \sum_i \frac{(Y_i^{\mathrm{data}} - Y_i^{\mathrm{res}})^2}{\sigma_i^2} \right\} \tag{14}$$

with $\vec{Y}^{\mathrm{res}}$ as the result from Eq. (5) or Eq. (8). The maximum error σ of the numerical calculations is 5% and is used for all cases. Finally we assign a constant prior to $p(\mu, \vec{\theta} \mid I)$ since no estimation about the parameters can be given in advance. The emerging integrals are not accessible analytically and the Markov chain Monte Carlo method had to be used, with the fraction in Eq. (13) as the sampling density.

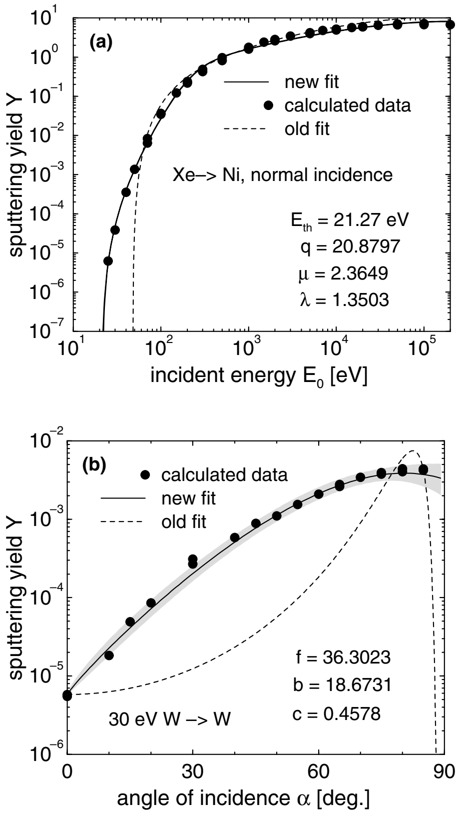

Fig. 1. Comparison of the new fit formulae with calculated values of the sputtering yield and the older fit with the revised Bohdansky formula [11] in (a) and the Yamamura formula [13] in (b). (a) Xe bombardment of Ni at normal incidence, (b) 30 eV W bombardment of W. The gray shaded region is the error margin.

5. Results

The new formulae (5) and (8) were already used in an IAEA publication [15], where all available experimental as well as calculated data for Be, C and W targets have been taken into account. In this paper we rely completely on data calculated with TRIM.SP. The projectile species range from D to Xe and selfbombardment, and the target species from Be to Au. Examples of an energy and angular dependence of the sputtering yield are shown in Fig. 1(a) and (b), which clearly demonstrate the better description with the new fit formulae. The most obvious point in Fig. 1(a) is, that the new fit gives a lower threshold and a less steep decrease of the yield at low energies. This seems to be a general behavior for small mass ratios as found for many examples. The angular dependence for small mass ratios and especially for selfbombardment (see Fig. 1(b)), at low energies shows a completely different behavior as given by the Yamamura formula; this was already observed in [11].

The calculated yield values presented in [16] and a few additional ones are used to redetermine the threshold energies with the new formula for the energy dependence of the sputtering yield. The Bayesian method delivers the threshold energy, E_{th}, and the constants λ, q and μ. As a result from all examples μ is approximately two with a slight decrease with an increasing mass ratio, M_2/M_1. The other parameters λ and q vary much stronger for the different examples and show a correlation only for a fixed mass ratio. The result is given in Fig. 2, where $\gamma E_{\mathrm{th}}/E_{\mathrm{s}}$ is plotted versus the target mass divided by the projectile mass. γ is the energy transfer factor $4M_1M_2/(M_1+M_2)^2$. This presentation has the advantage, that for large mass ratios $E_{\mathrm{th}}/E_{\mathrm{s}}$ approaches $1/\gamma$ and the value of $\gamma E_{\mathrm{th}}/E_{\mathrm{s}}$ approaches unity. For large mass ratios the results are close to the earlier values presented in [1], but for heavy targets the threshold is lowered by about a factor of two compared with the older data. The reason is the availability of newer yield values close to the threshold. The fit curve shown in Fig. 2 approaches unity at large mass ratios in contrast to formula (28) in [11]; the following formula

$$\gamma E_{\mathrm{th}}/E_{\mathrm{s}} = (0.3198 M_2/M_1)^{-0.5279} + 1 \tag{15}$$

gives a good fit to the calculated values. The fit values in Eq. (15) are determined with the above described procedure using individual errors for E_{th}, which are typically a few percent. The threshold values given here are usually somewhat lower than the values given in [15].

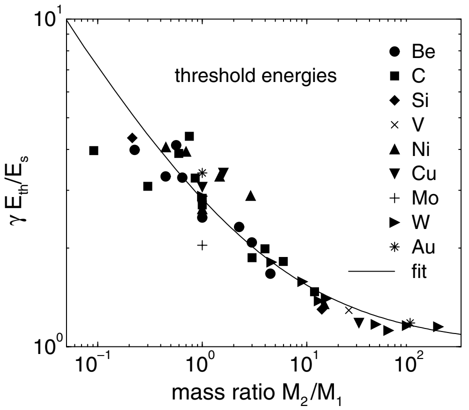

Fig. 2. The value $\gamma E_{th}/E_s$ versus the mass ratio M_2/M_1 for several ion-target combinations. The target species are indicated by different symbols. The fit curve is given in Eq. (15).

The reason for this discrepancy is the use of calculated and experimental values in [15]. Experimental values are generally higher near the threshold due to target surface structures and the energy width of the incident beam.

In Fig. 3 we depict the resulting parameter values for Eq. (5) for a few examples. Unfortunately it is not possible to give an overall functional behavior for these parameters. μ is approximately 2 ± 1 for all examples, whereas the other parameters λ and q depend on target and projectile mass. For a fixed target mass q increases with a positive power of the projectile mass. For λ no simple functional behavior can be given.

The new formula for the energy dependence has the advantage, that also the energy dependence at oblique angles of incidence can be fitted. For small target masses the fit works well near the threshold but shows deviations at large angles of incidence (grazing incidence) and higher energies. The probable reason is the neglect of the inelastic energy loss in the fit formula. But in every case, the threshold dependence can well be fitted. The results show (see Fig. 4) that the threshold energy is nearly independent of the angle of incidence for large mass ratios M_2/M_1 with a slight increase at grazing incidence in most cases. For $M_2/M_1 = 1$, the threshold energy decreases with increasing angle of incidence, whereas for

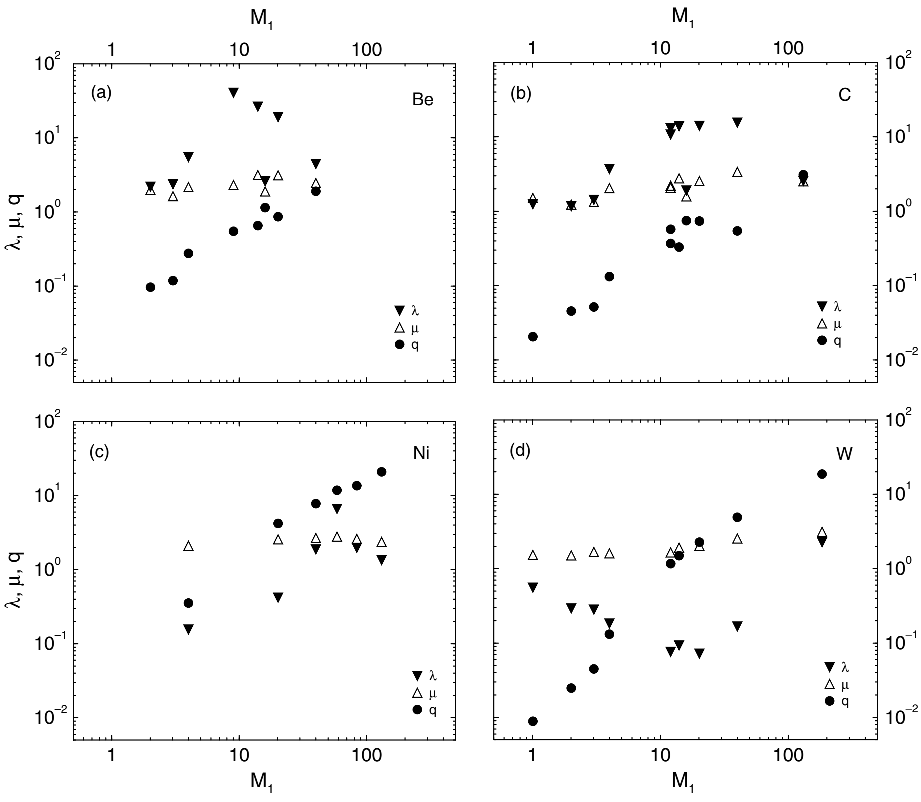

Fig. 3. Dependence of the parameters λ, μ, q on the projectile mass for the following targets: (a) Be, (b) C, (c) Ni, (d) W.

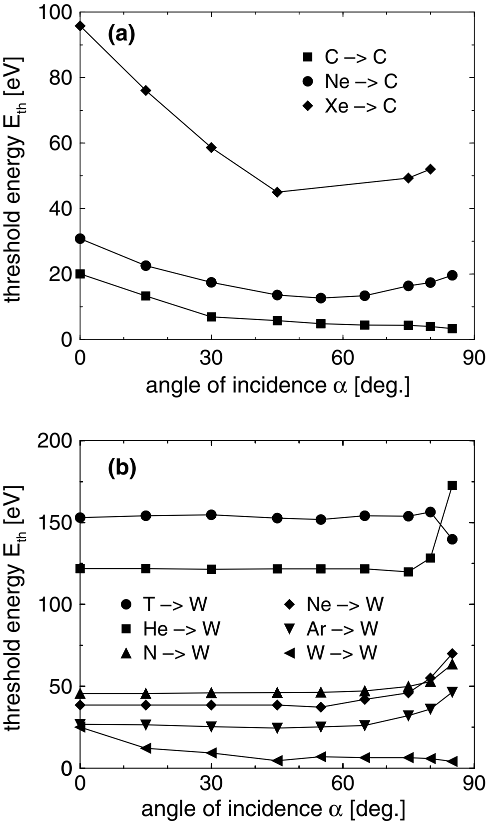

Fig. 4. The sputtering threshold energy versus the angle of incidence for several ion-target combinations. (a) C target, (b) W target. The lines are a guide to the eye and merely connect data points.

$M_2/M_1 < 1$ the threshold energy has a minimum at medium angles around 50°.

6. Conclusions

The new three parameter fit formula for the energy dependence of the sputtering yield has been demonstrated to be a good description especially near the threshold, where older fit formulae failed. The new fit formula is empirical and not based on theoretical grounds, although it is based on the revised Bohdansky formula [8]. The Yamamura fit formula for the energy dependence has been improved to account for binding energies of the bombarding species, for example self-bombardment, where large angles of incidence cannot be reached. With the use of large datasets [16] and the new fits threshold energies for different ion-target combinations and for the angle of incidence at several mass ratios are given.

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