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Sputtering coefficient calculation during light atoms bombardment of the solid targets

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A method for calculating the sputtering coefficient of targets during bombardment with light atoms is proposed. It is shown that taking into account the energy spectrum of reflected particles makes it possible to adequately describe the behavior of the sputtering coefficient near the threshold. At high energies, the particle reflection coefficient decreases greatly and the cascade sputtering mechanism proposed by Sigmund is added. Taking into account the contribution of both mechanisms makes it possible to achieve a quantitative description of the dependence of the sputtering coefficient in a wide range of energies of incident particles.

Keywords: sputtering thresholds, sputtering coefficient, ion bombardment, ITER tokamak.

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Introduction

Sputtering of materials with ion bombardment is widely used for the manufacture of thin films in modern microelectronics for creating a relief, for cleaning surfaces and for analyzing the composition of the surface. The destruction of electrodes due to sputtering plays an important role in electrical engineering. The interaction of plasma with the wall in modern thermonuclear installations leads to the sputtering of structural materials and the entry of impurities into the plasma, which affects the discharge parameters. For instance, the entry of tungsten impurities with a concentration of $10^{-3}-10^{-4}$ into the plasma from the plasma concentration because of large radiation losses makes it impossible to achieve the planned efficiency of a thermonuclear reactor.

The sputtering of solids is discussed in detail in Ref. [1–7]. The sputtering theory proposed by Sigmund played an important role [8]. However, Sigmund's theory does not describe the behavior of the sputtering coefficient near the sputtering threshold. The case of sputtering of heavy targets by light particles still does not have a reliable theoretical description. Attempts to modify Sigmund's theory were made in the papers of Falcone [9]. The contribution of various mechanisms to sputtering, in addition to the above reviews, was analyzed in Ref. [10,11]. Currently, the study of sputtering is intensively conducted by many scientific groups [12–17]. The impact of nano-roughness and surface relief on the sputtering coefficient was considered in the papers [18–21]. Sputtering by light ions of the first tokamak wall was considered in Ref. [22–24].

We consider in this paper the sputtering by light atoms in a wide range of bombarding particle energies for the cases H−Be and H−W, which characterize different mass ratios of the colliding particles.

1. Difference between the mechanism of sputtering by light and heavy ions

The use of computer modeling makes it possible to verify a number of concepts embedded in the theory of Sigmund and Falcone.

Let us consider the contribution of various mechanisms leading to sputtering, using the classification proposed Ref. [25] (Fig.1). We will distinguish between the processes of formation of sputtered atoms that take place during penetration of the bombarding particle deep into the target (index in), and similar processes during movement of the back-reflected bombarding particles (index out). The bombarding particle penetrating deep into the solid can knock out the target atom, which after several collisions can leave the solid which is called a PKA-in process (PKA — Primary Knock-on Atom). A similar contribution can be made by backscattered primary particles which is called a PKA-out process. knocked out target atoms can

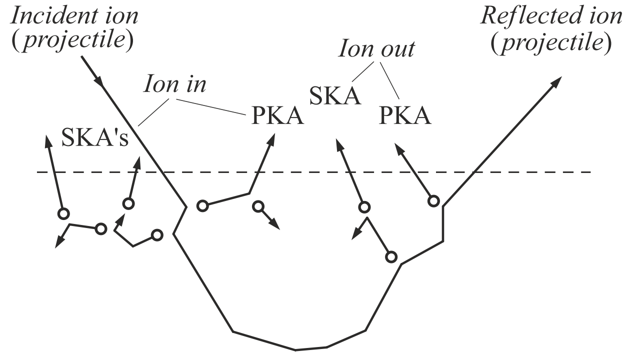

Figure 1. Diagram of sputtering processes. The figure is taken from Ref. [25].

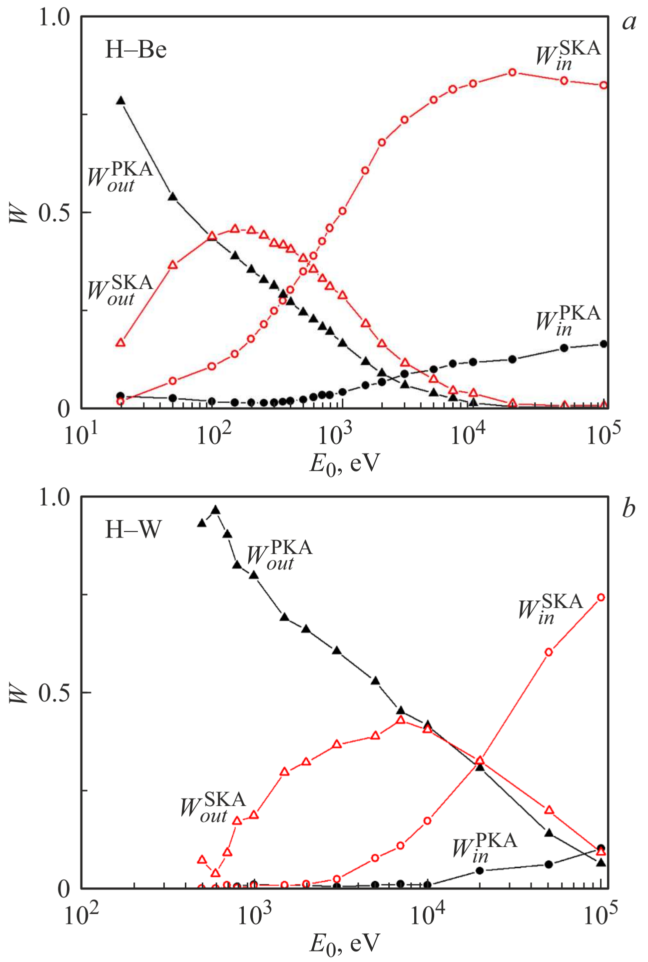

Figure 2. Contributions of various mechanisms to sputtering for the cases H–Be (*a*) and H–W (*b*).

transfer energy to secondary target particles, forming a cascade of collisions. We will distinguish between cases when secondary atoms were formed during penetration of bombarding particles deep into the target referred as SKA-in (SKA — Secondary Knock-on Atoms) and backscattered bombarding particles referred as SKA-out. The probabilities of the contribution of various mechanisms are denoted as $W_{in}^{\rm PKA}$, $W_{in}^{\rm SKA}$, $W_{out}^{\rm PKA}$, $W_{out}^{\rm SKA}$.

Figure 2 shows the contributions of various mechanisms to sputtering for the cases H–Be and H–W, calculated using our program [26–28].

Fig. 2 shows that the mechanism of sputtering of surface layers by a stream of backscattered bombarding particles prevail in case of low energies and the mechanism proposed by Sigmund contributes in case of high energies. In this case, the bombarding ions transfer their energy to the target atoms, and some of the atoms change the direction of the pulse towards the surface after a cascade of collisions and can leave the solid. This conclusion confirms the generally accepted ideas.

The knocked out target atom should overcome the surface potential barrier at the surface–vacuum interface. Two types of surface potential barriers are usually considered. A spherical potential barrier is applied to a surface consisting of atomic-sized points. In this case, the energy of the sputtered particle ε should exceed the energy of the surface bond, which is often taken as the sublimation energy U_s [29]. A planar potential barrier is used for a smooth surface. In this case, the perpendicular component of the energy of the atomized particle should exceed U_s, i.e. $E_2\cos^2\theta > U_s$, where θ is the angle of departure of the sputtered particle relative to the normal to the surface.

The position of the sputtering energy thresholds for both cases of the surface potential barrier is considered in detail in our paper [30]. The authors limit themselves in this paper to considering the cases of a spherical potential barrier and a normal beam incidence on a target.

The sputtering threshold $E_{\rm th}$ in the case of a mechanism of sputtering by a flux of backscattered bombarding particles can be calculated using the formula

$$E_{\rm th} = \frac{U_s}{\gamma(1-\gamma)^{0.5}} + \frac{dE}{dx}\,2d. \quad (1)$$

Here U_s — sublimation energy, dE/dx — electronic energy loss, d — average distance between target atoms. The value of the parameter γ is calculated using the formula

$$\gamma = \frac{4M_1M_2}{(M_1+M_2)^2}, \quad (2)$$

where M_1 and M_2 — the mass of the incident ion and the target atom, respectively. It is taken into account that the incident ion travels the distance $2d$, where d the average distance between the target atoms, and loses energy due to electron deceleration. The energy of the backscattered atom is $E_1 = (1-\gamma)^{0.5}E_0$ in case of scattering by an angle of the order of 90°, where E_0 is the initial energy. The maximum energy transferred to the target atom is $E_2 = \gamma E_1 = \gamma(1-\gamma)^{0.5}E_0$.

A simplified formula for estimating the sputtering coefficient for the mechanism of knocking out of surface atoms by a flux of backscattered particles can be written as

$$Y_{out} = \sigma(E_{\rm th}, E_0)n_tR_N\lambda. \quad (3)$$

Here $\sigma(E_{\rm th}, E_0)$ — recoil particle formation cross section with energy greater than U_s with the energy of the incident particle E_0, n_t — target density, R_N — reflection coefficient, λ — characteristic escape depth of the sputtered particles.

Let's estimate the cross-section of the process when energy exceeding the threshold energy is transferred to the target particle. The dependence of the scattering angle in the center-of-mass system on the impact parameter b is given by the expression

$$\theta(b) = \pi - 2b\int_0^{1/r_0} \frac{1}{\sqrt{1-\frac{U(1/g)}{E_{cm}} - b^2g^2}}\,dg, \quad g = \frac{1}{r}. \quad (4)$$

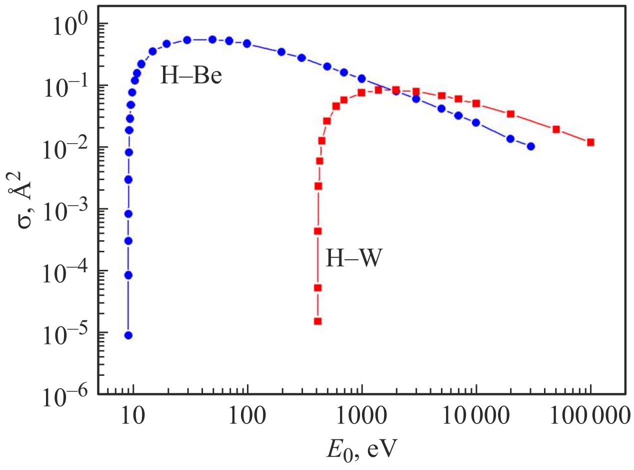

Figure 3. The cross-sections of transfer of energy exceeding the threshold for the cases H–Be and H–W for the DFT potential.

Here r_0 is the trajectory turning point. The energy transferred in case of scattering by an angle θ is determined by the expression

$$E_2 = E_0 \gamma \sin^2\left(\frac{\theta}{2}\right). \tag{5}$$

The value of $\theta_k = 2\arcsin\left([U_s/(E_0\gamma)]^{0.5}\right)$ is determined from the condition $E_2 = U_s$, and the parameter b_k from the ratio (4) is determined from the condition $\theta(b_k) = \theta_k$. The cross section is calculated as

$$\sigma = \pi b_k^2. \tag{6}$$

Fig. 3 shows the calculation of the cross section for the systems H–Be and H–W using the potential obtained in the density functional theory [31] (potential DFT).

It should be noted that the cross section of the process and, consequently, the sputtering coefficients depend on the type of potential used. Fig. 4 shows the calculation of the cross section for various types of interatomic interaction potentials. Our estimates show that the results of calculating the cross section for DFT and ZBL potentials give a difference of more than 30%. This is not surprising, since the ZBL potential describes the scattering of light particles less accurately [32]. We use the DFT potential as the most accurate.

Figure 5 shows the reflection coefficients for the cases H–Be and H–W, obtained in Ref. [33].

The energies of the knocked out target atoms can be calculated using the data on the average energies of the sputtered particles presented in Ref. [26,27]. Let us take into account that the average energy of the knocked out target atoms in a solid is greater than the average energy of the sputtered particles by the value of the surface barrier (U_s).

We obtained the dependence of the range on the energy of Be atoms in Be target and dependence of the range on the energy of W atoms in W target from the analysis of the range data from the SRIM database [34]. These dependencies in the case of Be–Be are described by the formula

$$\lambda[\text{Å}] = 0.841 \cdot \varepsilon[\text{eV}]^{0.6}, \tag{7}$$

and the case of W–W they are described by the formula

$$\lambda[\text{Å}] = 0.705 \cdot \varepsilon[\text{eV}]^{0.379}, \tag{8}$$

where ε — the energy of the knocked out target atoms. We obtain the dependence $\lambda\left(\varepsilon(E_0)\right)$ substituting the average energy of the knocked out atoms with a specific initial energy E_0 as ε. The values of $\lambda\left(\varepsilon(E_0)\right)$ are shown in Fig. 6.

Fig. 6 shows that the characteristic exit depth of the knocked out particles slightly varies depending on the initial energy, and the contribution of two or three near-surface layers prevails.

The application of the estimation formula (3), as shown in Figure 7, yields the correct value of the sputtering coefficient in the region of the maximum, but does not correctly

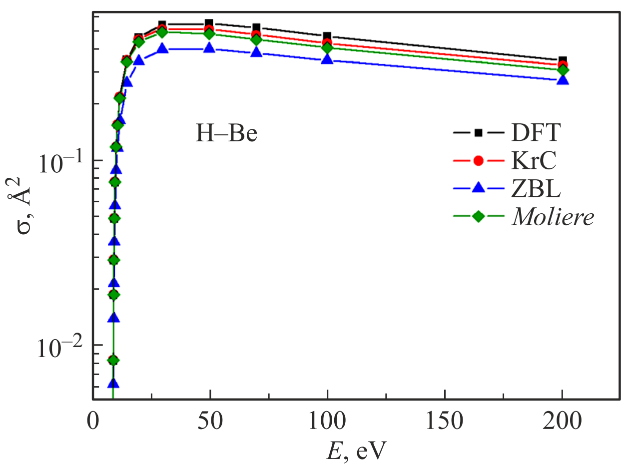

Figure 4. The cross-sections of transfer of energy exceeding the threshold for the case of H–Be for different potentials.

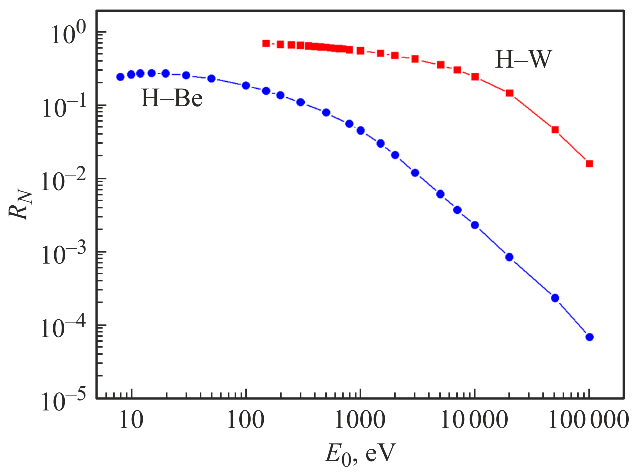

Figure 5. Reflection coefficients R_N depending on the energy of the bombarding particles for the cases H–Be and H–W [33].

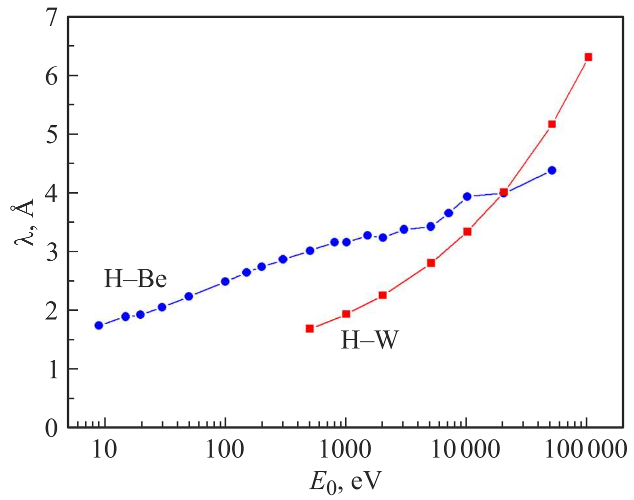

Figure 6. Characteristic exit depth of the sputtered particles λ depending on the energy of the bombarding particles E_0.

describe the energy dependence of the sputtering coefficient near the threshold. It is necessary to take into account the correction for the energy spectra of reflected particles dN/dE, which are shown in Fig. 8 for the cases H$-$Be [26] and H$-$W [28].

Fig. 8, a shows that the spectrum is of the same type in the range of initial energies $E_0 = 15-70$ eV. Peaks associated with single scattering at an angle of more than 90° are observed with the energy of reflected particles $E_1 = (1-\gamma)^{0.5}E_0$. Faster particles are present in the spectrum to the right of the values corresponding to scattering by 90° due to double and triple collisions, and they affect the shift of the energy threshold of sputtering towards lower energies. The energy spectrum data were taken from Ref. [28] in the case of the system H$-$W (Fig. 8, b). The value $E_1 = 0.8E_0$ in the case H$-$Be and $E_1 = 0.989E_0$ in the case H$-$W. The absence of atoms with an energy greater than E_1 in the spectrum of reflected particles results in a shift of the sputtering threshold towards higher values. This shift is insignificant in the case H$-$W.

The coefficient R_N includes the entire spectrum of reflected particle energies and is proportional to the integral in the denominator of the formula (9). Only particles with energy $E > E_{\mathrm{th}}$ should be taken into account. The cross section $\sigma(E_{\mathrm{th}}, E_1)$ and the escape depth of the knocked out atoms $\lambda(\varepsilon(E_1))$ depend on the energy of the reflected ions. Let's make a correction for the spectrum of reflected particles, taking into account the weight of particles with energy E_1 in the spectrum:

$$Y_{out}(E_0) = n_t R_N(E_0)\,\frac{\int\limits_{E_{\mathrm{th}}}^{E_0} \sigma(E_{\mathrm{th}}, E_1)\lambda\big(\varepsilon(E_1)\big)\frac{dN}{dE}(E_1)dE_1}{\int\limits_{0}^{E_0} \frac{dN}{dE}(E_1)dE_1}. \tag{9}$$

The normalization integral in the denominator is proportional to $R_N(E_0)$ and is inserted to account for the normalization of the spectrum dN/dE. The formula (9) does not take into account the contribution of the SKA-out channel and the dependence of the range of the knocked out particles on the escape angle relative to the normal to the surface. Estimates show that these two factors cancel each other out.

Fig. 7 shows estimates of the sputtering coefficient without correction for the spectrum of reflected particles (formula (3)) and with correction (formula (9)). Fig. 7 shows that correction for the spectrum of reflected particles

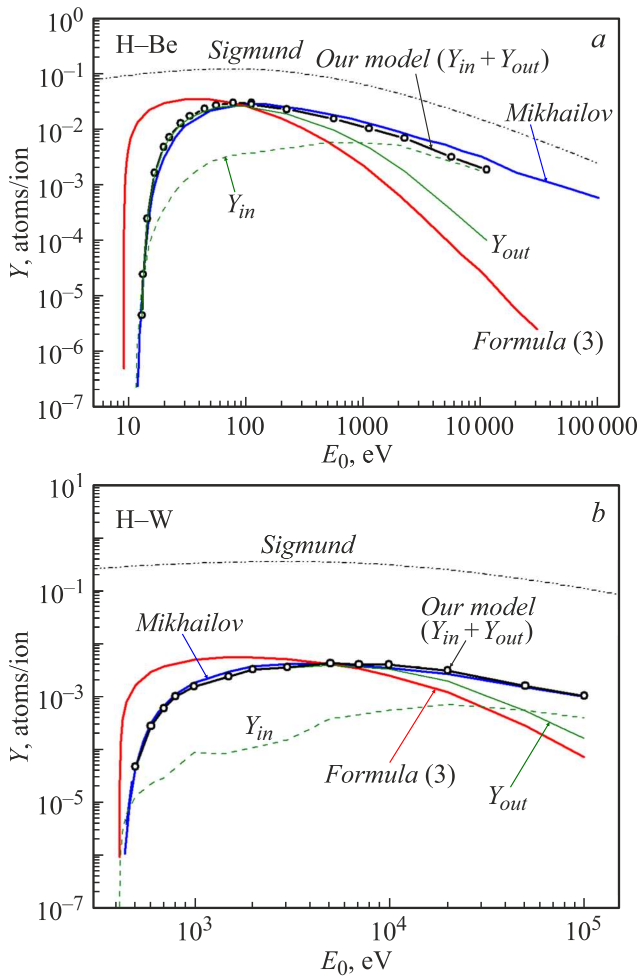

Figure 7. Dependence of the sputtering coefficient on the energy of the bombarding particle for the cases H$-$Be (a) and H$-$W (b). The computer calculation for a spherical potential barrier is shown by a blue curve [26,27]. Red curve shows the estimate Y without correction for the energy spectrum of reflected particles, formula (3). Solid green line shows the estimation of the contribution of the mechanism of knocking out of surface atoms by a flux of backscattered particles, taking into account the energy spectra of backscattered ions Y_{out}, formula (9). Green dashed curve denotes the accounting for the contribution of the cascade mechanism Y_{in}. The total contribution $Y_{in} + Y_{out}$ is shown by the open circles. The curve obtained by the Sigmund's formula is shown for comparison [8].

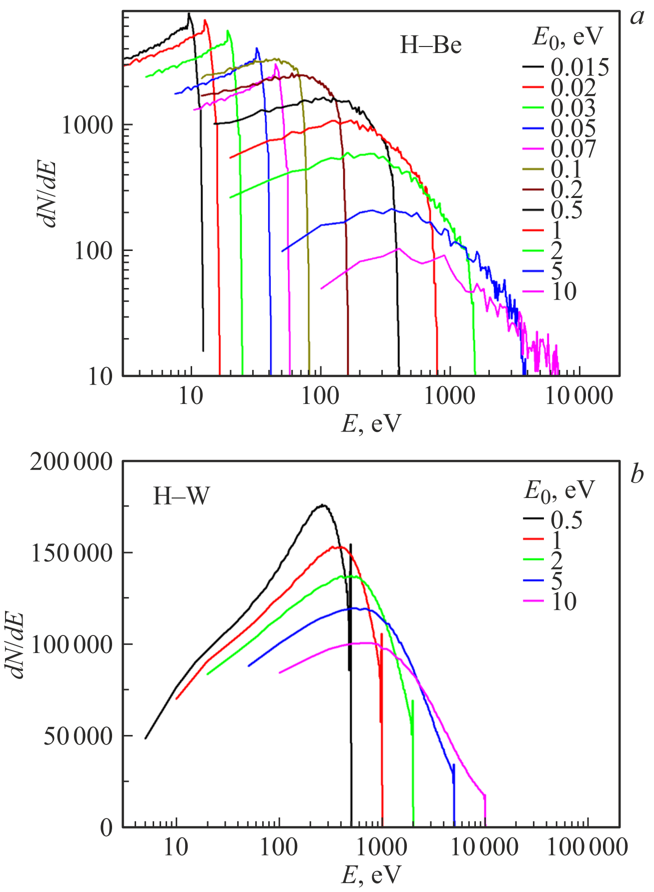

Figure 8. Spectra of reflected particles for the cases H–Be [26] (*a*) and H–W [28] (*b*) for various initial energies E_0.

significantly changes the path of the curve and correctly reflects the dependence of the sputtering coefficient on the energy of bombarding ions near the threshold.

The contribution of the Sigmund's cascade mechanism can be estimated as:

$$Y_{in} = Y_{out} \frac{W_{in}^{\mathrm{PKA}} + W_{in}^{\mathrm{SKA}}}{W_{out}^{\mathrm{PKA}} + W_{out}^{\mathrm{SKA}}}. \qquad (10)$$

Fig. 2 shows the probabilities of the contribution of various mechanisms.

Fig. 7 shows that the total contribution of both mechanisms is in good agreement with the data obtained by computer modeling of multiple collisions of bombarding particles with target atoms for the cases H–Be [26] and H–W [27]. Fig. 7 also shows the results of the evaluation using the Sigmund's formula [8], which yield greatly overestimated values of the sputtering coefficient and do not reflect the functional dependence $Y = f(E_0)$ near the sputtering threshold.

The mechanism of knocking out of surface atoms by a flux of backscattered particles prevails in a wide range of energies up to the energy of 30 keV in the case of H–W. In our opinion, the decrease of the contribution of the mechanism of knocking out of surface atoms by a flux of backscattered particles with an increase of the energy of the bombarding particles is associated with a decrease of the reflection coefficient. The reflection coefficient decreases in the case H–W with relatively high energies compared to the case H–Be (Fig. 5).

Conclusion

The mechanism of knocking out of surface atoms by a flux of backscattered particles make a prevailing contribution to sputtering at low energies up to $E_0 \approx 100E_{\mathrm{th}}$. The cascade mechanism proposed by Sigmund is added at high energies.

The characteristic exit depth of the knocked out atoms is calculated depending on the initial energy, and it is found that it equals to 2–3 surface layers.

The behavior of the curve near the sputtering threshold can be quantitatively described by taking into account the energy spectrum of reflected atoms.

A good quantitative agreement has been achieved between the data of the presented calculation and the results of computer modeling both in terms of the absolute value of the sputtering coefficient and a good description, in particular, near the sputtering threshold of the functional dependence of the sputtering coefficient on the energy of the incident particles.

The proposed model accurately describes the collision cases H–Be and H–W for a wide range of the ratio of the masses of the incoming particle and the target particle and the collision energies.

Conflict of interest

The authors declare that they have no conflict of interest.

References

[1] R. Behrisch. *Sputtering by Particle Bombardment I. Physical Sputtering of Single-Element Solids* (Springer, Berlin, 1981), DOI: 10.1007/3-540-10521-2

[2] R. Behrisch. *Sputtering by Particle Bombardment II. Sputtering of Alloys and Compounds, Electron and Neutron Sputtering, Surface Topography* (Springer, Berlin, 1983), DOI: 10.1007/3-540-12593-0

[3] R. Behrisch. *Sputtering by Particle Bombardment III. Characteristics of Sputtered Particles, Technical Applications* (Springer, Berlin, 1991), DOI: 10.1007/3-540-53428-8

[4] R. Behrisch, W. Eckstein. *Sputtering by Particle Bombardment* (Springer, Berlin, 2007), DOI: 10.1007/978-3-540-44502-9

[5] N.V. Pleshivtsev, A.I. Bazhin. *Fizika vozdejstviya ionnykh puchkov na materialy* (Vuzovskaya kniga, M., 1998) (in Russian).

[6] E.S. Mashkova. *Sovremennye tendencii v issledovanii raspyleniya tverdyh tel.* V kn.: *Fundamental'nye i prikladnye aspekty raspyleniya tverdykh tel*: Sb. statey 1986–1987: Per. s angl. E.S. Mashkova (Mir, Moscow, 1989) (in Russian)

[7] E.S. Mashkova, V.A. Molchanov. Rad. Eff., **108**, 307 (1989). DOI: 10.1080/10420158908230319

[8] P. Sigmund. Phys. Rev., **184**, 383 (1969). DOI: 10.1103/PhysRev.184.383
[9] G. Falcone. La Rivista del Nuovo Cimento, **13**, 1 (1990). DOI: 10.1007/bf02742981
[10] R. Behrisch, G. Maderlechner, B.M.U. Schemer, M.T. Robinson. Appl. Phys., **18**, 391 (1979). DOI: 10.1007/BF00899693
[11] J.P. Biersack, W. Eckstein. Appl. Phys. A., **34**, 73 (1984). DOI: 10.1007/bf00614759
[12] A.P. Mika, P. Rousseau, A. Domaracka, B.A. Huber. Phys. Rev. B, **100** (7), 075439 (2019). DOI: 10.1103/PhysRevB.100.075439
[13] K. Schlueter, K. Nordlund, G. Hobler, M. Balden, F. Granberg, O. Flinck, T.F. da Silva, R. Neu. Phys. Rev. Lett., **125**, 225502 (2020) DOI: 10.1103/PhysRevLett.125.225502
[14] A. Tolstogouzov, P. Mazarov, A.E. Ieshkin, S.F. Belykh, N.G. Korobeishchikov, V.O. Pelenovich, D.J. Fu. Vacuum, **188**, 110188 (2021). DOI: 10.1016/j.vacuum.2021.110188
[15] F. Duensing, F. Hechenberger, L. Ballauf, A.M. Reider, A. Menzel, F. Zappa, T. Dittmar, D.K. Bohme, P. Scheier. Nucl. Mater. Energy, **30**, 101110 (2022). DOI: 10.1016/j.nme.2021.101110
[16] A. Lopez-Cazalilla, F. Granberg, K. Nordlund, C. Cupak, M. Fellinger, F. Aumayr, W. Hauptstra, P.S. Szabo, A. Mutzke, R. Gonzalez-Arrabal. Phys. Rev. Materials, **6**, 075402 (2022). DOI: 10.1103/PhysRevMaterials.6.075402
[17] P. Phadke, A.A. Zameshin, J.M. Sturm, R. van de Kruijs, F. Bijkerk. Nucl. Instrum. Methods Phys. Res. B, **520**, 29 (2022). DOI: 10.1016/j.nimb.2022.03.016
[18] N.N. Andrianova, A.M. Borisov, E.S. Mashkova, A.A. Shemukhin, V.I. Shulga, Yu.S. Virgiliev. Nucl. Instr. Meth. Phys. Res. B., **354**, 146 (2015). DOI: 10.1016/j.nimb.2014.11.071
[19] N.N. Andrianova, A.M. Borisov, E.S. Mashkova, V.I. Shulga. J. Surf. Invest.: X-Ray, Synchrotron Neutron Tech., **10**, 412 (2016). DOI: 10.1134/S1027451016020233
[20] V.I. Shulga. J. Surf. Invest.: X-Ray, Synchrotron Neutron Tech., **14**, 1346 (2016). DOI: 10.1134/S1027451020060440
[21] N.N. Andrianova, A.M. Borisov, M.A. Ovchinnikov, R.Kh. Khisamov, R.R. Mulyukov. J. Surf. Invest.: X-Ray, Synchrotron Neutron Tech., **18**, 305 (2024). DOI: 10.1134/S1027451024020046
[22] D.G. Bulgadaryan, D.N. Sinel'nikov, N.E. Efimov, V.A. Kurnaev. Bull. Russ. Acad. Sci.: Phys., **84**, 742 (2020). DOI: 10.3103/S1062873820060064
[23] V.A. Kurnaev, D.K. Kogut, N.N. Trifonov. J. Nucl. Mater., **415**, S1119 (2011). DOI: 10.1016/j.jnucmat.2010.09.035
[24] V.S. Mikhailov, P.Yu. Babenko, A.P. Shergin, A.N. Zinoviev. Plasma Phys. Reports, **50**, 23 (2024). DOI: 10.1134/S1063780X23601682
[25] W. Eckstein. *Computer Simulation of Ion-Solid Interactions* (Springer-Verlag, Berlin, 1991), DOI: 10.1007/978-3-642-73513-4
[26] P.Yu. Babenko, V.S. Mikhailov, A.P. Shergin, A.N. Zinoviev. Tech. Phys., **68** (5), 662 (2023). DOI: 10.21883/TP.2023.05.56074.12-23
[27] V.S. Mikhailov, P.Yu. Babenko, A.P. Shergin, A.N. Zinoviev. JETP, **137**, 413 (2023). DOI: 10.1134/S106377612309011X
[28] V.S. Mikhailov, P.Yu. Babenko, D.S. Tensin, A.N. Zinoviev. J. Surf. Invest.: X-Ray, Synchrotron Neutron Tech., **17**, 258 (2023). DOI: 10.1134/S1027451023010330
[29] C. Kittel. *Introduction to Solid State Physics. 8th edition* (Wiley, NY., 2005)
[30] P.Yu. Babenko, V.S. Mikhailov, A.N. Zinoviev. Pisma v JTF, **50**, 3 (2024) (in Russian). DOI: 10.61011/PJTF.2024.12.58055.19851
[31] D.S. Meluzova, P.Yu. Babenko, A.P. Shergin, K. Nordlund, A.N. Zinoviev. Nucl. Instrum. Methods Phys. Res. B, **460**, 4 (2019). DOI: 10.1016/j.nimb.2019.03.037
[32] A.N. Zinoviev, P.Yu. Babenko, K. Nordlund. Nucl. Instrum. Methods Phys. Res. B, **508**, 10 (2021). DOI: 10.1016/j.nimb.2021.10.001
[33] V.S. Mikhailov, P.Yu. Babenko, A.P. Shergin, A.N. Zinoviev. ZhTF, **93** (11), 1533 (2023) (in Russian). DOI: 10.21883/JTF.2023.11.56484.192-23
[34] J.F. Ziegler, J.P. Biersack. SRIM. http://www.srim.org.

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