

# Surface quantum sputtering of solid bodies by light ions

A. I. Agafonov, É. A. Manykin, and M. A. Selifanov

*Institute of Superconductivity and Solid State Physics, Kurchatov Institute, 123182 Moscow, Russia*

(Submitted 1 July 1994)

Zh. Éksp. Teor. Fiz. **107**, 832–841 (March 1995)

We study sputtering of heavy targets by protons and deuteron beams, mediated by elastic scattering of ions off the surface atoms and accompanied by a transition of the nucleus of the atom from the ground state. It is shown that for proton energies  $\epsilon_{\mathbf{k}0} \leq 10$  keV and charged deuteron energies  $\epsilon_{\mathbf{k}0} \leq 7$  keV the sputtering coefficient obtained for sputtering via the surface quantum mechanism is in quantitative agreement with experimental data on the sputtering of gold targets. At low energies  $\epsilon_{\mathbf{k}0} \leq \epsilon_{\text{eff}}$  ( $\epsilon_{\text{eff}} = 4590$  eV for Au/H<sup>+</sup> and 2794 eV for Au/D<sup>+</sup>) inelastic ion scattering channels in the target cause an abrupt decrease in the sputtering coefficient in comparison with its value determined only by the reaction cross section on the surface of the target. © 1995 American Institute of Physics.

## 1. INTRODUCTION

At the present time the theory of ion-beam sputtering of solid-state targets is based on transport theory of collision cascades generated by the primary ions.<sup>1–4</sup> The energy of an ion colliding with the solid-state target is dissipated in elastic and inelastic scattering channels in the target near the surface, causing a cascade of elastic collisions of the target atoms. The dynamics of the collision cascades is governed by the Boltzmann equation with account of pairwise atomic collisions, which are treated in the classical representation. Here it is assumed that a target atom is at rest before the collision, and its binding energy to the solid-state lattice is neglected in comparison with the mean energy transferred in the collisions. Accounting for the surface forces reduces to introducing a flat potential barrier. Sputtering of the target is effected by atoms whose energy is greater than the height of this barrier. The intersection of the collision cascades with the target surface determines the sputtering coefficient and the energetic and angular characteristics of the sputtered particles. These particles are primarily neutral atoms in the ground state. The relative fraction of the ionic component and atom clusters is usually of the order of  $10^{-2}$ .

The theory of collision cascades is inapplicable in a number of important cases.<sup>1–3</sup> Among them we may mention sputtering of heavy targets by light ions. The primary ions lose their energy primarily through the inelastic channels of electronic excitations in the target, while the elastic cross sections of the ion–atom collisions are relatively small.<sup>2</sup> Consequently, cascades cannot develop. In Refs. 2, 3, and 5–7 a mechanism of light-ion sputtering was connected with the backscattering of ions in the near-surface layers by large angles of the order of  $\pi$ . Some of the ions, moving in U-shaped paths, can be scattered toward the surface and dislodge some of the surface atoms. These atoms can be ejected from the surface if their energy is high enough to overcome the surface potential barrier. The regime of this mechanism is called the regime of primary direct knockout.<sup>2</sup>

Here it should be noted that at moderate ion energies corresponding to velocities  $v \leq Z^{2/3} e^2 / \hbar$ , the sputtering coefficient is substantially influenced by the inelastic ion scat-

tering channels in the target.<sup>1</sup> If they are not correctly accounted for, it is difficult to obtain a reasonable value of the coefficient.<sup>2,3</sup> Using classical mechanics to describe the elastic ion–atom collisions in the target is not justified since the binding energy of an atom  $\epsilon_b$  can be comparable with the mean energy transferred to the atom in the collisions.

In Ref. 8 a theoretical study was carried out of the interaction channel between the target atoms and incident electron and photon beams, which is accompanied by bound–free transitions of the nuclei of the surface atoms from bound states on the surface to the continuum while preserving the electronic configurations of the atoms. The latter is connected with the fact that a significant fraction of the emitted particles, during desorption as well as sputtering, consists of neutral atoms in the ground state.<sup>1,9</sup> For an electron beam the interaction potential is determined by the central field of the nucleus, which is screened by atomic electrons. In the case of an incident electromagnetic wave, these transitions can be treated as a photoelectric effect for the surface ions.

In the present paper we investigate sputtering of heavy targets by H<sup>+</sup> and D<sup>+</sup> beams at normal incidence to the surface, due to elastic scattering of the ions by the surface atoms. The microscopic mechanism of sputtering is connected with the elastic collision of the incident ion ( $C_{\mathbf{k}0}$ ,  $\mathbf{k}_0$  is the wave vector in the initial state) with a surface atom, accompanied by a transition of the nucleus of the atom from the ground (and bound) state ( $A_0$ ) on the surface to a state of the continuum with wave vector  $g(A_g)$ :

$$(A)_0 + C_{\mathbf{k}0} \rightarrow A_g + C_{\mathbf{k}}. \quad (1)$$

At low ion energies it is important to take account of the influence of fast, inelastic ion scattering channels on the sputtering cross section.

The spectrum of the continuous states as well as the discrete surface states for an atomic nucleus is determined by the surface potential, which in turn is determined in the adiabatic approximation by the quantum state of the target. We will investigate reaction (1) at the surface only for interactions of the surface atoms with incident ions in the initial state, neglecting the contribution to the sputtering from ions scattered by the target and secondary electrons. Thus, the

surface potential is determined by the ground state of the target, and the evolution of the initial state of the ion is due to scattering processes in the target.

As a rule, the flux of sputtered particles is proportional to the flux of ions bombarding the target.<sup>1-3</sup> Therefore we will limit ourselves here to a calculation of the sputtering cross section.

## 2. BOUND-FREE TRANSITION AMPLITUDE

The Hamiltonian of the system can be represented in the form

$$H = H_A + H_B + H_C + V_{AC} + V_{BC}, \quad (2)$$

where  $H_A$  is the unperturbed Hamiltonian for a surface atom nucleus, including its core electrons, which determines the spectrum of states in the field of the stationary surface potential;  $H_B$  and  $H_C$  are the unperturbed Hamiltonians of the solid body and the incident ion, respectively;  $V_{AC}$  and  $V_{BC}$  are the surface atom-ion ( $A-C$ ) and ion-solid body ( $B-C$ ) interaction operators. The eigenstates  $\Phi_A^A$ ,  $\Phi_B^B$ , and  $\Phi_C^C$  of the Hamiltonians  $H_A$ ,  $H_B$ , and  $H_C$  are assumed to be known.

Let the entire system at the initial instant of time  $t = t_0$  ( $t_0 \rightarrow -\infty$  in the final formulas) be described by the wave function  $\Phi_i = \Phi_0^A \Phi_0^B \Phi_0^C$ , where  $\Phi_0^A$ ,  $\Phi_0^B$ , and  $\Phi_0^C$  are the initial states of the bound surface atom, target, and ion, respectively. In the final state  $\Phi_f = \Phi_g^A \Phi_f^B \Phi_f^C$  the wave function corresponds to the state of the nucleus of the surface atom in the continuum with wave vector  $\mathbf{g}$ .

For scattering channel (1) the scattering amplitude can be represented in the form

$$A_{fi}(t, t_0) = (\Phi_f, S(t, t_0) \Phi_i), \quad (3)$$

where the  $S$ -matrix for the total system corresponds to the actual Hamiltonian (2) in the interaction picture with unperturbed Hamiltonian  $H_0 = H_A + H_B + H_C$ :

$$i\hbar \frac{\partial}{\partial t} S(t, t_0) = (\mathcal{H}_{AC}(t) + \mathcal{H}_{BC}(t)) S(t, t_0).$$

Since we are calculating the cross section of process (1), we can limit ourselves to first order in  $V_{AC}$  for the transition amplitude  $A_{fi}$ . As a result, we have

$$A_{fi} = -\frac{i}{\hbar} \int_{t_0}^t dt_1 (\Phi_f, S_{BC}(t, t_1) \mathcal{H}_{AC}(t_1) S_{BC}(t_1, t_0) \Phi_i). \quad (4)$$

Here  $S_{BC}$ —the matrix for the  $BC$  subsystem—is determined by the unperturbed Hamiltonian  $H_B + H_C$  and the interaction operator  $V_{BC}$ . Allowing only for the interaction of the surface atoms with the ions in the initial state with wave vector  $\mathbf{k}_0$  and energy  $\varepsilon_{\mathbf{k}_0}$ , from Eq. (4) for the transition amplitude we have

$$\begin{aligned} A_{fi}(t) = & -\frac{i}{\hbar} \sum_{\mathbf{k}} \int_{t_0}^t dt_1 (\Phi_f^B \Phi_f^C, S_{BC}(t, t_1) \Phi_0^B \Phi_{\mathbf{k}}^C) \\ & \times (\Phi_g^A \Phi_{\mathbf{k}}^C, \mathcal{H}_{AC}(t_1) \Phi_0^A \Phi_{\mathbf{k}_0}^C) \\ & \times (\Phi_0^B \Phi_{\mathbf{k}_0}^C, S_{BC}(t_1, t_0) \Phi_0^B \Phi_{\mathbf{k}_0}^C). \end{aligned} \quad (5)$$

The last factor in expression (5) is the probability amplitude that the initial states of the solid body and ion, prescribed by the wave function  $\Phi_0^B \Phi_{\mathbf{k}_0}^C$  at time  $t_0$ , will be preserved at time  $t_1$ . This amplitude can be represented in the form

$$\begin{aligned} (\Phi_0^B \Phi_{\mathbf{k}_0}^C, S_{BC}(t_1, t_0) \Phi_0^B \Phi_{\mathbf{k}_0}^C) = & \exp\left(\frac{i}{\hbar} \left( \Delta \varepsilon_{\mathbf{k}_0} - \frac{1}{2} i \Gamma_{\mathbf{k}_0} \right) \right. \\ & \left. \times (t_1 - t_0) \right), \end{aligned} \quad (6)$$

where  $\Delta \varepsilon_{\mathbf{k}_0}$  is the energy shift of the  $BC$  subsystem due to the interaction  $V_{BC}$ , and the quantity  $\Gamma_{\mathbf{k}_0}$  can be considered the inverse lifetime, associated with scattering in the target, of the initial state of the ion. Here  $\Delta \varepsilon_{\mathbf{k}_0}$ ,  $\Gamma_{\mathbf{k}_0} \ll \varepsilon_{\mathbf{k}_0}$ .

The matrix element for elastic scattering of the ion by a surface atom, accompanied by a bound-free transition of the latter, has the form

$$\begin{aligned} (\Phi_g^A \Phi_{\mathbf{k}}^C, \mathcal{H}_{AC}(t_1) \Phi_0^A \Phi_{\mathbf{k}_0}^C) \\ = M^{AC}(\mathbf{g}, \mathbf{k}; 0, \mathbf{k}_0) \exp\left(\frac{i}{\hbar} t_1 (\varepsilon_g + \varepsilon_{\mathbf{k}} + \varepsilon_b - \varepsilon_{\mathbf{k}_0})\right), \end{aligned} \quad (7)$$

where the energy  $\varepsilon_g$  corresponds to the state of the nucleus of the surface atom in the continuum with wave vector  $\mathbf{g}$ ;  $\varepsilon_0$  is its binding energy in the potential well on the surface in the initial state;  $\varepsilon_{\mathbf{k}}$  is the energy of the scattered ion.

The first factor under the integral in Eq. (5) is connected with scattering of the ion in the target and is the transition amplitude from the state  $\Phi_0^B \Phi_{\mathbf{k}}^C$  at time  $t_1$  to the state  $\Phi_f^B \Phi_f^C$  at time  $t$ . For the state  $\Phi_f^B \Phi_f^C \neq \Phi_0^B \Phi_{\mathbf{k}}^C$  we have

$$\begin{aligned} (\Phi_f^B \Phi_f^C, S_{BC}(t, t_1) \Phi_0^B \Phi_{\mathbf{k}}^C) \\ = -\frac{i}{\hbar} \int_{t_1}^t dt_2 (\Phi_f^B \Phi_f^C, \mathcal{H}_{BC}(t_2) S_{BC}(t_2, t_1) \Phi_0^B \Phi_{\mathbf{k}}^C). \end{aligned} \quad (8)$$

The function  $\exp[-i/\hbar t (H_B + H_C)] S_{BC}(t, t_1) \Phi_0^B \Phi_{\mathbf{k}}^C$  in the Schrödinger picture describes the state of the subsystem  $BC$  with Hamiltonian  $H_B + H_C + V_{BC}$  at time  $t$ , which arises from the state  $\Phi_0^B \Phi_{\mathbf{k}}^C$  at time  $t_1$ . We introduce the eigenfunction  $\Phi_{0\mathbf{k}}^{(+)}$  of the Hamiltonian  $H_B + H_C + H_{BC}$ :

$$\Phi_{0\mathbf{k}}^{(+)} = \exp\left(\frac{it}{\hbar} (\varepsilon_0^B + \varepsilon_{\mathbf{k}}^C - H_B - H_C)\right) S_{BC}(t, t_1) \Phi_0^B \Phi_{\mathbf{k}}^C, \quad (9)$$

which is defined on the energy surface  $\varepsilon_0^B + \varepsilon_{\mathbf{k}}^C$  (Ref. 10). From Eq. (9) it is easy to show that  $\Phi_{0\mathbf{k}}^{(+)}$  satisfies the equation

$$\begin{aligned} \Phi_{0\mathbf{k}}^{(+)} = & \Phi_0^B \Phi_{\mathbf{k}}^C + \frac{1}{\varepsilon_0^B + \varepsilon_{\mathbf{k}}^C - H_B - H_C + i\delta} V_{BC} \Phi_{0\mathbf{k}}^{(+)}, \\ & \delta \rightarrow +0. \end{aligned} \quad (10)$$

Solutions of the integral equation (10) include both diverging waves and bound states of the ions in the solid-state target.

Employing Eq. (9), we bring expression (8) into the form

$$\begin{aligned}
& (\Phi_f^B \Phi_f^C S_{BC}(t, t_1) \Phi_0^B \Phi_k^C) \\
&= -\frac{i}{\hbar} (\Phi_f^B \Phi_f^C, V_{BC} \Phi_{0k}^{(+)}) \\
&\times \int_{t_1}^t dt_2 \exp\left(\frac{i}{\hbar} t_2 (\varepsilon_f^B + \varepsilon_f^C - \varepsilon_0^B - \varepsilon_k^C)\right). \quad (11)
\end{aligned}$$

### 3. BOUND-FREE TRANSITION CROSS SECTION

The transition cross section for a surface atom can be represented in the form

$$\sigma_g = 2 \frac{\Omega}{v_{k_0}} \sum_{f(BC)} \operatorname{Re} \lim_{t \rightarrow \infty} A_{\tilde{f}}^*(t, t_0) \frac{\partial}{\partial t} A_{\tilde{f}}(t, t_0), \quad (12)$$

where  $v_{k_0} = (2\varepsilon_{k_0}/m)^{1/2}$  is the velocity of the ion, and  $\Omega$  is the normalized volume of the  $\Phi_{k_0}^C$  state. The sum is over the final states of the  $B$  and  $C$  subsystems.

Substituting expression (5) into Eq. (12), differentiating with respect to  $t$  in the upper limit of the integral for the scattering amplitude does not contribute to the cross section, because of damping in the system (6). Differentiating the integrand in expression (5) with respect to  $t$  taking Eq. (11) into account and substituting Eqs. (6), (7), and (11) in Eq. (12), we obtain for the transition cross section

$$\sigma_g = \sum_{\mathbf{k}} \frac{v_{\mathbf{k}}}{v_{k_0}} \frac{\Omega}{L_{\mathbf{k}}} \frac{|M^{AC}(\mathbf{g}, \mathbf{k}; 0, \mathbf{k}_0)|^2}{(\varepsilon_g + \varepsilon_{\mathbf{k}} + \varepsilon_b - \varepsilon_{k_0} - \Delta\varepsilon_{k_0})^2 + \frac{1}{4} \Gamma_{k_0}^2}, \quad (13)$$

where in the statistical limit  $\Omega \rightarrow \infty$ ,  $L_{\mathbf{k}} = \Omega/\sigma_{\mathbf{k}}^{bC}$  is a finite quantity, and has dimensions of length. Here we have introduced the notation

$$\begin{aligned}
\sigma_{\mathbf{k}}^{BC} &= \frac{2\pi}{\hbar} \frac{\Omega}{v_{\mathbf{k}}} \sum_{f(BC)} |(\Phi_f^B \Phi_f^C, V_{BC} \Phi_{0k}^{(+)})|^2 \delta \\
&\times (\varepsilon_f^B + \varepsilon_f^C - \varepsilon_0^B - \varepsilon_{\mathbf{k}}^C) \quad (14)
\end{aligned}$$

for the total cross section of elastic and inelastic scattering, including ion capture processes in the solid body.

Usually, the binding energy  $\varepsilon_b$  amounts to several eV, and the mean energy of the sputtered atoms  $\langle \varepsilon_g \rangle \sim 10$  eV. For moderate ion energies,  $\varepsilon_{k_0} \gg \varepsilon_g, \varepsilon_b, \Delta\varepsilon_{k_0}, \Gamma_{k_0}$ . Assuming  $L_{\mathbf{k}}(\varepsilon_{\mathbf{k}})$  to be a slowly varying function (on the scale of the energies  $\max\{\langle \varepsilon_g \rangle, \Delta\varepsilon_{k_0}, \Gamma_{k_0}\}$ ), the transition cross section is given approximately by

$$\begin{aligned}
\sigma_g &= \frac{2\pi\Omega}{L_{k_0}\Gamma_{k_0}} \sum_{\mathbf{k}} |M^{AC}(\mathbf{g}, \mathbf{k}; 0, \mathbf{k}_0)|^2 \delta \\
&\times (\varepsilon_g + \varepsilon_{\mathbf{k}} + \varepsilon_b - \varepsilon_{k_0} - \Delta\varepsilon_{k_0}). \quad (15)
\end{aligned}$$

Here  $L_{k_0}\Gamma_{k_0}$  is a function of the ion energy  $\varepsilon_{k_0}$ . It is easy to obtain the asymptotic form of this function at high ion energies. Since the total width of the inelastic scattering channels in the target decreases with increasing  $\varepsilon_{k_0}$ , starting at some  $\varepsilon_{k_0}$  the influence of the inelastic channels on the bound-free transition amplitude of the atom can be neglected. In this case the surface quantum sputtering coefficient

of the target is determined only by the elastic process (1) on the surface. In this case, setting  $(\Phi_0^B \Phi_{k_0}^C, S_{BC}(t_1, t_0) \Phi_0^B \Phi_{k_0}^C) = 1$ , we have the following asymptotic form at high energies for the function  $L_{k_0}\Gamma_{k_0}$ :

$$L_{k_0}\Gamma_{k_0} \rightarrow \hbar v_{k_0} \quad \text{for } \varepsilon_{k_0} \rightarrow \infty. \quad (16)$$

In principle, the function  $L_{k_0}\Gamma_{k_0}$  is determined by the contribution of all inelastic ion scattering channels, as well as the elastic channel. Therefore it would be advisable, without calculating this function from theoretical first principles, to consider it empirically for the given target and ion. A reasonable parametrization of  $L_{k_0}\Gamma_{k_0}$  should reflect the asymptotic form (16) at high energies, which, as we will show, within the framework of the proposed model of surface quantum sputtering of solid-state targets by light ions, does not contain any fitting parameters.

### 4. TRANSITION MATRIX ELEMENT

The bound-free transition matrix element of a surface atom with preservation of its electronic configuration has the form

$$M^{AC}(\mathbf{g}, \mathbf{k}; 0, \mathbf{k}_0) = \langle \Phi_{\mathbf{g}}^A \Phi_{\mathbf{k}}^C | V_{AC}(\mathbf{r}, \mathbf{R}, \{\mathbf{r}_e^j\}) | \Phi_0^A \Phi_{k_0}^C \rangle, \quad (17)$$

where  $\Phi_0^A = \psi_e \psi_0^A$ , where  $\psi_e$  is the wave function of the atomic electrons and  $\psi_0^A(\mathbf{R})$  is the wave function of the atomic nucleus in the ground state on the surface;  $\Phi_{\mathbf{g}}^A = \psi_e \psi_{\mathbf{g}}^A$ , where  $\psi_{\mathbf{g}}^A$  is the wave function of the nucleus in the continuum with wave vector  $\mathbf{g}$ ;  $\Phi_{k_0}^C(\mathbf{r})$  and  $\Phi_{\mathbf{k}}^C(\mathbf{r})$  are the wave functions of the ion in the initial and final states;  $\mathbf{r}$  is the radius vector of the ion;  $\mathbf{R}$  is the radius vector of the particle reckoned from the position of the minimum of the potential well on the surface;  $\{\mathbf{r}_e^j\}$  is the set of coordinates of the atomic electrons reckoned from  $\mathbf{R}$ ;  $V_{AC}(\mathbf{r}, \mathbf{R}, \{\mathbf{r}_e^j\})$  is the interaction potential of the ion with the central field of the nucleus with atomic number  $Z$ , screened by the atomic electrons.

Employing the plane-wave approximation for the incident and scattered ions, we obtain from Eq. (17) the following expression for the transition matrix element:

$$\begin{aligned}
M^{AC}(\mathbf{g}, \mathbf{k}; 0, \mathbf{k}_0) &= \frac{4\pi e^2}{\Omega_i \Delta k^2} (Z - Q(\Delta\mathbf{k})) \\
&\times \langle \psi_{\mathbf{g}}^A(\mathbf{R}) | \exp(i\Delta\mathbf{k}\mathbf{R}) | \psi_0^A(\mathbf{R}) \rangle, \quad (18)
\end{aligned}$$

where  $\Delta\mathbf{k} = \mathbf{k}_0 - \mathbf{k}$ , and  $Q(\Delta\mathbf{k})$  is the atomic form factor,

$$Q(\Delta\mathbf{k}) = \left\langle \psi_e \left| \sum_j \exp(i\Delta\mathbf{k}\mathbf{r}_e^j) \right| \psi_e \right\rangle, \quad (19)$$

which is the Fourier component of the electronic density of the surface atoms. We represent the wave function of the particle in its initial state in the form<sup>8</sup>

$$\psi_0^A(\mathbf{R}) = \pi^{-1/2} \kappa^{3/2} \exp(-\kappa R), \quad \kappa = \hbar^{-1} (2M\varepsilon_b)^{1/2}, \quad (20)$$

where  $M$  is the mass of a surface atom.

The transition matrix element on the right-hand side of Eq. (18) can be calculated in the vicinity of the minimum of the potential well with radius  $R \approx \kappa^{-1}$ . As is easy to show,

typical values of  $\kappa^{-1}$  lie in the range  $10^{-3}$ – $10^{-2}$  Å. In this spatial region, it is possible to neglect the parabolic variation of the potential well in the Schrödinger equation. Noting that  $\varepsilon_b$  is much larger than the zero-point energy of the particle, we have for the wave function

$$\psi_{\mathbf{g}}^A(\mathbf{R}) = 2^{1/2} \Omega_i^{-1/2} \chi(\mathbf{g}) \sin[\mathbf{g}_{*z} R_z] \exp(i \mathbf{g}_{*p} \mathbf{R}_p), \quad (21)$$

where  $g_*^2 = g_{*z}^2 + g_{*p}^2 = 2\hbar^{-2} M(\varepsilon_b + \varepsilon_g)$ , and  $\mathbf{g}_{*z} > 0$ . Here the  $z$  axis points in the direction of the outward normal to the surface. The function  $\mathbf{g}_*$ , defined near the bottom of the potential well, is treated as a function of the wave vector  $\mathbf{g}$ , and is determined by the shape of the potential well. In the one-dimensional case in the quasiclassical approximation we obtain a rough estimate for the function  $\chi(\mathbf{g})$ :  $\chi(\mathbf{g}) = [\varepsilon_g / (\varepsilon_g + \varepsilon_b)]^{1/4}$ .

$$\begin{aligned} \frac{\partial^2 \sigma}{\partial \varepsilon_g \partial O_g} &= \frac{2^{19/2}}{\pi^2} (r_B Z R y)^2 \frac{m^{1/2} \hbar \varepsilon_b^{5/2}}{M L_{k_0} \Gamma_{k_0} \varepsilon_{k_0}^{1/2}} \frac{\varepsilon_g (\varepsilon_g + \varepsilon_b)^{1/2}}{(\varepsilon_g + 2\varepsilon_b)^6} \\ &\times \cos^2 \theta_g^* \int_0^{2\pi} d\varphi_k \int_{-1}^1 dx_k \frac{(1 + \gamma x_k)^2}{(1 + \gamma^2 + 2\gamma x_k)^2} (1 - Z^{-1} Q(x_k, \gamma, \varepsilon_{k_0}))^2 \\ &\times \frac{[1 + a[1 + \gamma^2 + 2\gamma x_k + 2\gamma b(1 - x_k^2)^{1/2} \sin \theta_g^* \cos \varphi_k]]^2}{\{[1 + a[1 + \gamma^2 + 2\gamma x_k + 2\gamma b(1 - x_k^2)^{1/2} \sin \theta_g^* \cos \varphi_k]]^2 - 4a^2 b^2 (1 + \gamma x_k)^2 \cos^2 \theta_g^*\}^4}. \end{aligned} \quad (22)$$

Here  $L_{k_0} \Gamma_{k_0}$  is a function of  $\varepsilon_{k_0}$ ,  $m$  is the mass of an ion,  $O(\mathbf{g})$  is the solid angle,  $r_B$  is the Bohr radius,  $\cos \theta_g^* = g_{*z} / g_* \geq 0$ ,  $x_k = k_z / k$ ,  $\varphi_k$  is the polar angle of the vector  $\mathbf{k}$ , and we have used the following notation:

$$\gamma = \left(1 - \frac{\varepsilon_g - \varepsilon_b - \Delta \varepsilon_{k_0}}{\varepsilon_{k_0}}\right)^{1/2}, \quad a = \frac{m}{M} \frac{\varepsilon_{k_0}}{\varepsilon_g + 2\varepsilon_b},$$

$$b = \left(\frac{M}{m} \frac{\varepsilon_g + \varepsilon_b}{\varepsilon_{k_0}}\right)^{1/2}.$$

To calculate the atomic form factor  $Q$  of atoms with large  $Z$ , we can use the Thomas–Fermi method. In a three-term approximation,<sup>11</sup>

$$Q(s) = Z^{5/3} \sum_{j=1}^3 \frac{\alpha_j \beta_j}{s^2 + \beta_j Z^{2/3}}, \quad (23)$$

where  $\alpha_1 = 0.255$ ,  $\alpha_2 = 0.581$ ,  $\alpha_3 = 0.164$ ,  $\beta_1 = 6.984 \cdot 10^{-3}$ ,  $\beta_2 = 0.10348$ ,  $\beta_3 = 2.1896$ , and, in our notation,  $s^2 = 2^{-1} \pi^{-2} \hbar^{-2} m \varepsilon_{k_0} (1 + \gamma^2 + 2\gamma x_k)$ , expressed in units of Å<sup>-2</sup>.

For  $L_{k_0} \Gamma_{k_0}$  as a function of  $\varepsilon_{k_0}$  we have used the expression

$$L_{k_0} \Gamma_{k_0} = \hbar v_{k_0} \left(1 + \left(\frac{\varepsilon_{\text{eff}}}{\varepsilon_{k_0}}\right)^n\right), \quad n > 0. \quad (24)$$

This leads to a substantial decrease in the sputtering cross section for ion energies  $\varepsilon_{k_0} < \varepsilon_{\text{eff}}$ , due to the influence of the inelastic scattering channels. At high energies  $\varepsilon_{k_0} \gg \varepsilon_{\text{eff}}$ ,

## 5. NUMERICAL CALCULATIONS OF THE SPUTTERING CROSS SECTION AND DISCUSSION OF RESULTS FOR Au/H<sup>+</sup> AND Au/D<sup>+</sup>

Here we restrict ourselves to the case of normal incidence of ions with wave vector  $\mathbf{k}_0$  on the target.

Substituting Eqs. (20) and (21) into Eq. (18), it is easy to calculate the matrix element  $M^{AC}(\mathbf{g}, \mathbf{k}; 0, \mathbf{k}_0)$ . Utilizing relation (15), we obtain for the differential bound–free transition cross section

where the asymptotic form (16) is valid, the differential cross section (22) does not contain any fitting parameters in the current model.

In the numerical calculations of the cross section (22), in

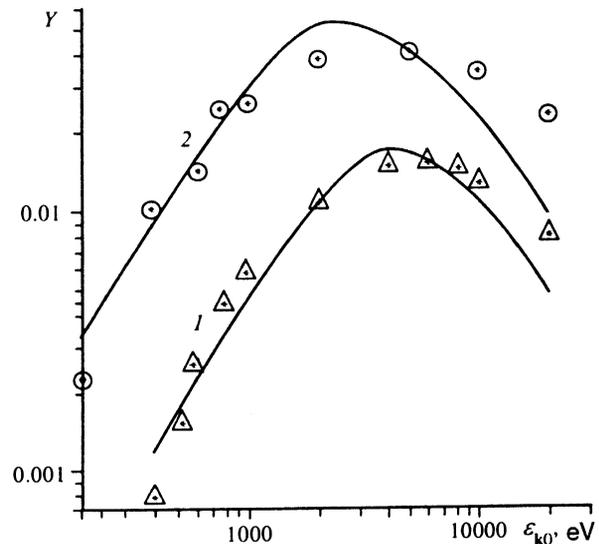


FIG. 1. Dependence of the sputtering coefficient  $Y$  on the ion energy for H<sup>+</sup> and D<sup>+</sup>. The points indicate experimental data from Ref. 12. The solid curves were calculated according to formula (22): 1—Au/H<sup>+</sup>; 2—Au/D<sup>+</sup>. For binding energy of the surface atom Au  $\varepsilon_b = 3.81$  eV (Refs. 2 and 12), and surface concentration  $1.95 \cdot 10^{15}$  cm<sup>-2</sup>.

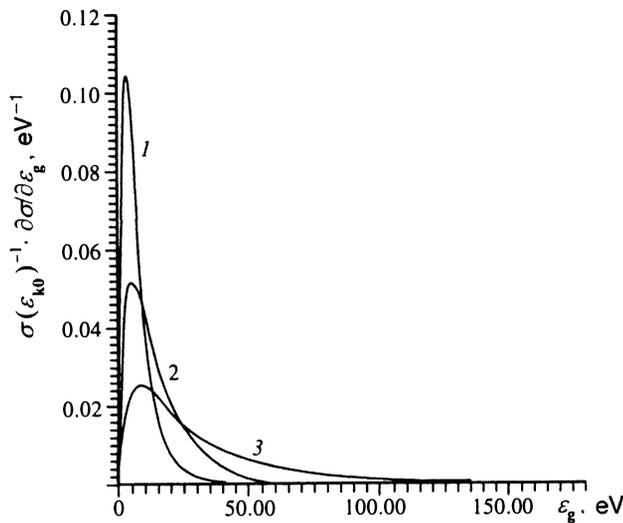


FIG. 2. Normalized dependence of the differential sputtering cross section  $\sigma(\epsilon_{k0})^{-1} \partial\sigma/\partial\epsilon_g$  on the energy of the atom Au in the continuum  $\epsilon_g$  for different ion energies (for  $H^+$ ):  $\epsilon_{k0}=400$  eV (1), 2 keV (2), 10 keV (3);  $\epsilon_b=3.81$  eV.

calculating the integral over the solid angle  $O_g$  we used the approximation  $\theta_g^* = \theta_g$ . Considering the smooth dependence of the sputtering coefficient on the ion energy and the fact that  $\epsilon_{k0} \gg \Delta\epsilon_{k0}$ , the quantity  $\Delta\epsilon_{k0}$  in the definition of  $\gamma$  can be neglected.

Figure 1 plots the experimental values obtained in Ref. 12 and the values calculated according to formula (22) of  $Y$ , the coefficient of sputtering of a gold target by protons and deuterons as a function of energy. At low ion energies the experimental values of  $Y$  increase rapidly with  $\epsilon_{k0}$ , reaching their maximum value at  $\epsilon_{k0} = \epsilon_p$ . For example, for the Au/ $H^+$  system we have  $\epsilon_p \approx 5$  keV. The value of  $\epsilon_p$  agrees with the estimate of the ion velocity  $v \leq e^2 \hbar$  corresponding to the maximum of the average specific losses of the ion energy due to electron bremsstrahlung. In this energy range, the inelastic channels of electronic excitation in the target have a substantial influence on the bound-free transition amplitudes of the surface atoms.

For  $H^+$  ion energies  $\epsilon_{k0} \leq 10$  keV and the  $D^+$  ion energies  $\epsilon_{k0} \leq 7$  keV, the calculated values of  $Y$  according to the mechanism of quantum sputtering are in quantitative agreement with the experimental data on the sputtering of a gold target. The value of  $n$  in expression (24), is the same for both protons and deuterons:  $n=5/2$ . The values of  $\epsilon_{eff}$  are different:  $\epsilon_{eff}=4590$  eV for Au/ $H^+$  and  $\epsilon_{eff}=2794$  eV for Au/ $D^+$ .

At high energies,  $\epsilon_{k0} > 10$  keV for protons and  $\epsilon_{k0} > 7$  keV for deuterons, the surface mechanism gives smaller values of  $Y$ , and with increasing  $\epsilon_{k0}$  the discrepancy between the calculated and the experimental values of the sputtering increases. We think that in this region, collision cascades begin to develop as the ion energy increases, which creates a possible alternative channel of target sputtering.

Figure 2 plots calculated curves of the normalized dependence of the differential sputtering cross section  $\sigma(\epsilon_{k0})^{-1} \partial\sigma/\partial\epsilon_g$  on the energy  $\epsilon_g$ , which reflect the energy

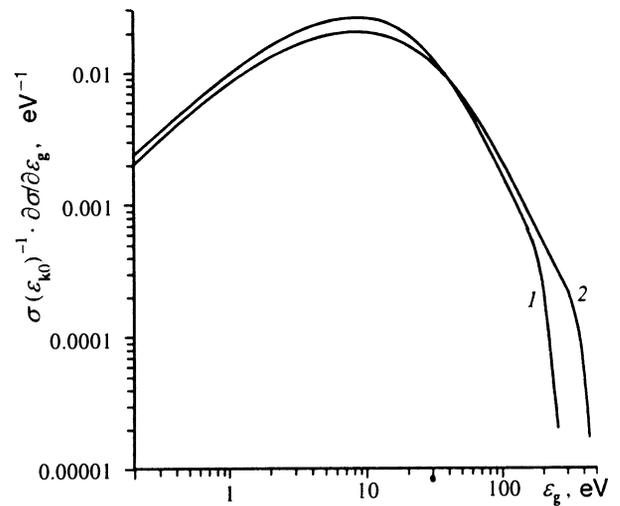


FIG. 3. Influence of the isotope effect on the normalized distributions  $\sigma(\epsilon_{k0})^{-1} \partial\sigma/\partial\epsilon_g$  as a function of  $\epsilon_g$ , the energy of the Au atoms in the continuum: 1) Au/ $H^+$ , 2) Au/ $D^+$ . Ion energy  $\epsilon_{k0}=10$  keV.

distribution of the gold atoms ejected from the surface for various energies of the  $H^+$  ions. With increasing  $\epsilon_{k0}$ , the peak of the distribution shifts toward higher energies and the contribution to the cross section from high-energy parts of the distribution grows. Similar features obtain for deuterons.

The influence of the isotope effect on the normalized distribution functions  $\sigma(\epsilon_{k0})^{-1} \partial\sigma/\partial\epsilon_g$  as a function of the energy  $\epsilon_g$  is shown in Fig. 3. For  $D^+$ , high-energy particles make a greater contribution to sputtering than for  $H^+$  ions.

In conclusion, we have proposed here a new microscopic mechanism of sputtering of solid-state targets due to elastic collisions of the incident ions with the surface atoms, accompanied by a transition of the atomic nucleus from the ground state in the surface to a continuum state. In accounting for the interaction of the surface atoms only with incident ions in an initial state whose evolution is due to inelastic scattering processes in the target, we obtained an expression for the differential sputtering cross section.

We showed that for  $H^+$  ion energies  $\epsilon_{k0} \leq 10$  keV and the  $D^+$  ion energies  $\epsilon_{k0} \leq 7$  keV, the results obtained for sputtering via a surface quantum mechanism is in quantitative agreement with available experimental data on the sputtering of gold targets by protons and deuterons. At low ion energies  $\epsilon_{k0} \leq \epsilon_{eff}$  ( $\epsilon_{eff}=4590$  eV for Au/ $H^+$  and  $\epsilon_{eff}=2794$  eV for Au/ $D^+$ ), the inelastic ion scattering channels in the target have a substantial effect on the sputtering cross section, leading to smaller values than that determined only by the cross section of reaction (1) on the target surface.

In the high energy region of the  $H^+$  ions,  $\epsilon_{k0} > 10$  keV, and the  $D^+$  ions,  $\epsilon_{k0} > 7$  keV, the calculated surface sputtering cross sections become less than the experimental value, and with increasing  $\epsilon_{k0}$  this discrepancy increases. We think that only in this energy range do collision cascades, which have a much greater influence on the sputtering coefficient, begin to develop.

- <sup>1</sup>P. Sigmund, Nucl. Instrum. Meth. B **27**, 1 (1987).  
<sup>2</sup>D. Falcone, Usp. Fiz. Nauk **162**, 71 (1992).  
<sup>3</sup>E. S. Mashkova and V. A. Molchanov, Rad. Eff. **108**, 307 (1989).  
<sup>4</sup>A. I. Tolmachev, Nucl. Instrum. Meth. in Phys. Res. B **93**, 415 (1995).  
<sup>5</sup>R. Behrisch, G. Maderlechner, B. M. U. Scherzer, and M. T. Robinson, Appl. Phys. **18**, 391 (1979).  
<sup>6</sup>U. Littmark and S. Fedder, Nucl. Instrum. Meth. **194**, 607 (1982).  
<sup>7</sup>J. P. Biersack, Nucl. Instrum. Meth. B **27**, 21 (1987).  
<sup>8</sup>A. I. Agafonov and É. A. Manykin, Zh. Éksp. Teor. Fiz. **104**, 3656 (1993) [JETP **77**, 747 (1993)]; Phys. Lett. **181**, 413 (1993).  
<sup>9</sup>V. N. Ageev, O. P. Burmistrova, and Yu. A. Kuznetsov, Usp. Fiz. Nauk **158**, 389 (1989) [Sov. Phys. Usp. **32**, 588 (1989)].  
<sup>10</sup>T.-Y. Wu and T. Ohmura, *Quantum Theory of Scattering*, Prentice-Hall, Englewood Cliffs, N.J. (1962).  
<sup>11</sup>M. Atoji, Acta Cryst. **10**, 291 (1957).  
<sup>12</sup>H. L. Bay, J. Roth, and J. Bohdansky, J. Appl. Phys. **48**, 4722 (1977).

Translated by Paul F. Schippnick