Energy distribution and degree of ionization of atoms scattered in bombardment of a surface by molecules

I. S. Bitenskiĭ, Ya. S. Gilenko, and E. S. Parilis

Electronics Institute, Uzbek Academy of Sciences

(Submitted 31 July 1987)
Zh. Eksp. Teor. Fiz. **94**, 66–75 (March 1988)

A general theory is developed of the broadening of the energy distribution of atoms scattered by a surface in impact dissociation of fast molecules caused by mutual collisions of the atoms of the molecules. The formation of the energy spectrum of scattered atoms of homo- and heteroatomic molecules consisting of light and heavy atoms is described. The influence of additional mutual collisions of the partners on the degree of ionization of the scattered atoms is analyzed. The calculation results are compared with the experimental data.

1. INTRODUCTION

Research into scattering of fast molecular ions by solid surfaces is attracting interest of late. Experiments[1–3] have shown that a fraction of the molecules are scattered without dissociation, even though the energy loss exceeds significantly the dissociation energy. In Refs. 4 and 5 is proposed a mechanism of nondissociative scattering of fast molecules, based on successive collisions of the molecule atoms with the surface atoms. Computer-modeling calculations[4–7] confirm this mechanism.

Experiments[2,8–12] show that the energy distributions of the ions produced by molecule dissociation are broader than the distributions of the ions scattered in bombardment by atomic ions having the same velocity. The authors of Refs. 2 and 13 attributed this broadening to an electronic transition produced in the molecule on scattering from the surface, leading to a subsequent mutual repulsion of the molecule nuclei, and hence to a change of their kinetic energy. It is possible that such a mechanism works at low bombarding energies or small glancing angles, for in this case the maximum energy that can be transferred from one atom to another is considerably lower than their energy after the scattering. Yet experiment reveals a spectrum broadening comparable with the initial energy. For example, measurements[9] of the energy spectra of nitrogen, oxygen, and carbon ions scattered by a surface bombarded by molecular ions yielded ions with approximately 15 keV energy, whereas the initial energy of the atom in the molecule was $E_0 = 10$ keV.

In Ref. 4 we have advanced the hypothesis that this broadening is the result of "shoving" of the scattered atom by another atom of the molecule and a redistribution of their kinetic energies, and obtained some estimates, while in Ref. 14 it was shown that it is possible to transfer to the atom in this manner an appreciable fraction of the initial kinetic energy of the molecule, independently of the change of its electronic structure. The authors of Ref. 9 also interpret their results in terms of collisions between atoms.

We present here a general theory of the collisional broadening mechanism. We describe the evolution of the energy distribution of the scattered atoms following impact dissociation of fast molecules on a surface of a solid in general form, in the case of spatial scattering with an arbitrary ratio of the atom masses in the molecule and in the solid. The influence of additional collision of the partners on the degree of ionization of the scattered atoms is analyzed.

2. FORMATION OF ENERGY SPECTRUM OF SCATTERED ATOMS

In the general case, the energy and angular distribution of the scattered atoms is determined by the doubly differential cross section

$$\sigma(E_0, E, \theta) = \iint\limits_{\Phi(E_0,E,\theta)} f(E_0, E, \theta_1, \theta)\,\sigma_2(\theta_2, E_1)\,d\theta_1\,d\theta_2, \qquad (1)$$

where θ is the total scattering angle; $f(\theta_1, E, E_0, \theta)$ is the probability of scattering of the atom by the solid at an angle θ_1; θ_2 and $\sigma_2(\theta_2, E_1)$ are the angle and cross section for the atom scattering by collision with its partner; E_0 and E are the initial and final energies of the particle. The intergration is over all possible scattering trajectories with given final values of E and θ allowed by the kinematic relations. It is also required that close mutual approach of the molecule atoms be possible after one of them is scattered in the solid. This calls for close values of the emission times and points. The function $f(\theta_1, E, E_0, \theta)$ and the trajectory family $\Phi(E_0, E, \theta)$ are different for molecules consisting of heavy or light atoms. In the first case it is possible to use the approximation of single or double scattering by atoms of one or two surface layers of a polycrystal.[15] The probability $f(\theta_1, E, E_0, \theta)$ of scattering through an angle θ is determined by the cross section $\sigma_1(\theta_l)$ for scattering of the molecule atom by an atom of the solid, or by the cross section for double scattering, and $\Phi(E_0, E, \theta)$ is one trajectory of scattering through an angle θ or a family of twofold- scattering trajectories.

In the case of light atoms, $f(\theta_1, E, E_0, \theta)$ describes the probability of multiple scattering, and Φ is the aggregate of multiple-scattering trajectories corresponding to the given E_0, E, and θ. The length of these trajectories can reach thousands of Angstroms.

When the surface is bombarded by molecules consisting of both heavy and light atoms, such as ArH molecules, the scattering of each atom should be described in its own approximation.

3. SCATTERING OF HEAVY ATOMS

Scattering of heavy atoms by a surface is well described in first-order approximation by a single collision with the surface atom, followed by collisions of molecule atoms with one another. The problem of broadening of an atom spectrum in collision of a fast molecule with an individual atom was solved by us in Ref. 16. It was shown there that mutual collisions of the molecule atoms cause a considerable broadening of the energy distribution of the atom scattered through an angle θ. The distribution extends to the high-energy part to values of E that exceed not only the single-scattering energy $E_1(\theta)$, but also the initial energy E_0 of the molecule per atom of mass m_1. The equation relating the scattering angle with the particle energy is

$$\cos\theta=[1-ux(1+\nu x)]\varepsilon^{-1/2}, \tag{2}$$

where $\varepsilon = E/E_0$, $u = 2/[(1+\mu)(1+\nu)]$, and

$$x=\frac{u(1-\nu u)\pm[u^2+\nu u(\nu u-2)(\varepsilon-1)]^{1/2}}{\nu u(\nu u-2)}. \tag{3}$$

Here $\mu = m_1/M$, $\nu = m_1/m_2$ (m_2 and M are the mass of the second atom of the molecule and of the solid-body molecule, respectively). Both signs should be used in Eq. (3) for $\nu > \nu^*$ and the minus sign for $\nu \leqslant \nu^*$. Then

$$\nu^*=\{[(3+\mu)^2+8\mu(1+\mu)]^{1/2}-(3+\mu)\}/4\mu.$$

For particular cases,

$$\begin{aligned}&\cos\theta=\varepsilon^{1/2}-\varepsilon^{-1/2}\pm(2/\varepsilon-1)^{1/2} \quad \text{for } \mu\to 0,\ \nu=1,\\ &\cos\theta=(\varepsilon-u^2+1)/(2\varepsilon^{1/2}) \quad \text{for } \nu\to 0.\end{aligned} \tag{4}$$

A plot of $\varepsilon(\theta)$ for $\mu \ll 1$ and different ν is shown in Fig. 1. If $\nu \geqslant \nu^*$, the maximum of ε is

$$\varepsilon_m=1+u/[\nu(2-\nu u)] \tag{5}$$

and is reached at $\theta = \theta_m$:

$$\theta_m=\arccos\left\{\left[1+\frac{u(1-\nu u)}{\nu(2-\nu u)^2}\right]\Big/\varepsilon^{1/2}\right\}. \tag{6}$$

If $\nu < \nu^*$, then $\theta_m = 0$, and $\varepsilon_m = [u(1-\nu)+1]^2$. For $\nu \to 0$ and $\mu \to 0$ at $\theta_m = 0$, in particular, we have $\varepsilon_m = 9$, while for $\mu \to 0$ and $\nu = 1$, which corresponds to a homonuclear molecule incident on a very heavy target, a value $\varepsilon_m = 2$ is reached at $\theta = 45°$, i.e., one of the atoms of the molecule receives its total kinetic energy.

It was shown in Ref. 16 that the largest contribution is made by scattering in the incidence plane, and the expression

$$\sigma(\theta,E)=\frac{2^{1/2}\sigma_1(\theta_{10}')\sigma_2(\theta_{20})\sin\theta_{10}'\sin\theta_{20}}{\pi l^2 v_0 u(1+\nu x)v_0'\sin\theta\cos(\theta_{10}'/2)} \tag{7}$$

was obtained, where the primed quantities pertain to c. m. s. of the incident atom and the atom of the solid, v_0 is the molecule velocity, l is the interatomic distance, and θ_{10}' and θ_{20} are the scattering angles in the incidence plane in the first and second collisions.

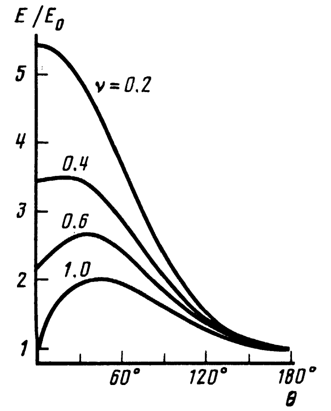

FIG. 1. Relative energy E/E_0 vs the scattering angle θ for different mass ratios ν of the molecule atoms.

Since the energy E is close to E_0 in the high-energy part of the spectrum, it follows that $\theta_{20} \ll 1$ and we obtain from (7) the approximate equation

$$\sigma(E,\theta)=\sigma_{max}\left(\frac{\pi U_2}{2l^2}\right)^2\frac{\sigma_1(\theta_1)E_1E_0\sin^2\theta}{\sigma_1(\theta)E_c^2(E-E_1)^2}, \tag{8}$$

where $\sigma_{\max}$ is the scattering cross section corresponding to the maximum impact parameter (equal to l) of the second collision

$$\begin{aligned}&E_c=[E_0+E_1-2(E_0E_1)^{1/2}\cos\theta_1]/2,\\ &\theta_1\approx\theta+(E-E_1)/[2(E_0E_1)^{1/2}\operatorname{tg}(\theta/2)].\end{aligned}$$

In the calculation of σ_2 we assumed an inverse-square approximation of the Firsov potential $U(r) = R_2/r^2$.

It is known that the energy spectrum of heavy atoms scattered by a polycrystal surface does not reduce to a single-scattering peak, but is broadened towards higher energies, or even contains in this region an additional maximum attributed to double collisions with the atoms of the solid. The two-atom scattering model, in which the solid is represented by a diatomic molecule, was developed in Ref. 15. Calculation of multiple scattering heavy atoms by a diatomic molecule[17] is also of independent interest for the physics of atom-molecule collisions. This calculation was used to describe the broadening of the energy spectrum of atoms in collision of a molecule with a molecule.[16] The result of the latter calculation can be used to describe the broadening of the energy spectrum of the scattered heavy atoms when a surface is bombarded by molecules, with allowance in the spectrum for double collisions of one of the atoms of the molecule with the atoms of the solid.

As shown in Ref. 16, in double scattering it is necessary to take into account the lag of the second atom of the molecule relative to the atom that undergoes double scattering in the solid. The requirement that the instants of time be close limits the first scattering angle and hence the orientation of the dumbbell molecule that simulates the solid. As a result, when (1) is integrated the region Φ is narrowed down, and not the entire double-scattering spectrum contributes to the broadened spectrum. With allowance for the double scattering, the cross section (1) is described by the expression derived in Ref. 16.

Experiments[9] permit a comparison of the calculated spectrum broadening with the data obtained by bombarding various metals with nitrogen molecules. Figure 2 shows the dependence of $\sigma/\sigma_{\max}$ on the relative value E/E_1 for different E_0 and for a scattering angle $\theta = 40°$. The figure shows that the derived Eq. (8) describes satisfactorily both the form of the high-energy part of the spectrum and the dependence of the cross section on the initial energy. The mass ratio for the considered combinations of bombarding ions and targets is $\mu \ll 1$. In this case $E_1 \approx E_0$, therefore, the dependence of the high-energy part of the spectrum for different energies is practically independent of the target material. It should be noted that the agreement was obtained without any fitting parameter whatever, at one and the same constant of the potential $U_2 = 33$ eV.

In Ref. 9 they measured also the ion energy spectrum for bombardment of Au by N_3^+ molecular ions. The pres-

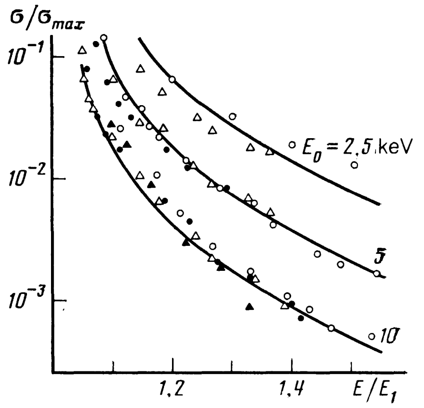

FIG. 2. High-energy sections of spectra in bombardment of different metal targets by N_2^+ ions. Experimental data[9]: ○ — Cu, ● — Rh, Δ — Au, ▲ — Ti. Solid curves—calculated using Eq. (9).

ence of one more atom in the molecule should cause a large broadening of the spectrum because of the additional scattering by the third atom of the molecule. This broadening was in fact observed, but its dependence on E_1, the same as for N_2^+, and its relatively small value indicate that it is due only to a decrease of the maximum impact parameter in the molecule, which amounts to $l \cdot 3^{1/2}/2$ for a triangular configuration of the atoms. In fact, the experimental ratio $\sigma N_3^+/\sigma N_2^+ = 1{,}9$ is close to the value $(2/3^{1/2})^4 \approx 1{,}8$ obtained from Eq. (8). Copper was bombarded in Ref. 18 by ArH^+ molecules to simulate the scattering of slow protons by a solid surface. Our result shows that in scattering of heteronuclear molecules with atoms of greatly differing mass, the spectrum of the scattered light particles should be broadened much more strongly than for homonuclear molecules. For example, under the conditions of the experiment of Ref. 18, where $\theta = 40°$, the maximum energy, as follows from (4), is $E_m \approx 7E_1$.

It is also possible to consider the broadening of the argon peak for bombardment by ArH^+ molecules compared with bombardment by atomic Ar^+. It turns out here that the total kinetic energy of the proton can be transferred to the Ar atom, resulting in a corresponding lengthening of the tail of the energy spectrum of the Ar^+ ions. To attribute this lengthening to decay from the repulsive state[2] at an initial ArH^+ energy 20 keV in the experiment of Ref. 18, the energy required for such a repulsion would be about 100 eV.

4. SCATTERING OF LIGHT ATOMS

Scattering of light atoms by a solid surface at glancing angles is via multiple collisions. It is accompanied by a spread of the points of the emergence of the particles from the solid. The probability of collision of molecule atoms on emerging from a solid is $p \approx l^2/S$, where S is the area of the region of particle emergence. An expression for the scattering coefficient, with allowance for the coordinate of the atom emergence points, was obtained in Ref. 19. It yields $S \approx \alpha_0^5/\langle\theta_s^2\rangle^2$, where α_0 is the glancing angle and $\langle\theta_s^2\rangle$ is the mean squared scattering angle per unit path. Since $S \gg l^2$ in the case, the probability of particle collision after the emergence is $p \ll 1$. Thus, additional scattering of the molecule atoms by one another on emergence, such as considered in the preceding spectrum, has low probability. At the same time, the spectra of the scattered particles upon bombardment by atomic and molecular particles differ substantially.[20] In Ref. 20 the largest broadening of the spectrum was observed at the lowest energies, corresponding to long paths of the scattered particles inside the material. Clearly, the difference is due to interaction of the molecule atoms with one another as they pass through the material, and not when they are scattered at the surface. A rigorous analysis of such a process entails solution of kinetic equations and is a complicated problem. The influence of the collisions of molecule atoms with one another on their energy distribution after emerging from the solid was considered by us under the following approximations. The angle divergence of the molecule-atom trajectories is such that the path L^* along which the distance between them exceeds l and mutual collisions become unlikely, is equal to $L^* = [lL(E)/\theta]^{1/2}$, where $L(E)$ is the atom path corresponding to a final energy E in the solid. In this case $L(E) = L_0[1-(E/E_0)^{1/2}]$, where L_0 is the total path of an atom with initial energy E_0, and $L^* \ll L(E)$.

The substantial influence of the interaction of the molecule atoms in the initial section of the trajectory on the energy distribution of the particles is confirmed by the experiment of Ref. 21. Its authors investigated the energy spectra of protons passing through a carbon film bombarded by H_2^+ molecules of energy 12.5 keV/atom. Broadening of the energy distributions was observed already in passage through a film 15 Å thick, and the largest measured energy of the transmitted protons reached 13.5 keV. The calculations of Ref. 21 show that, as a result of multiple screening of the Coulomb scattering of the protons in dissociation of the H_2^+ molecule, the deviation angle is 5–8° already on a path of the order of 10–20 Å, and the transverse energy in the c. m. s. reaches 60–80 eV. The mutual scattering of the ions takes place thus on the initial part of the trajectory. It leads, in contrast to mutual collisions of heavy atoms on a surface (considered in the preceding section), to a change of the trajectories of the multiple scattering of the ion in the substance, and this change determines in the case of light ions their final energy.

The action of the mutual scattering reduces in the first approximation to a change of the glancing angle α_0. We have then in Eq. (1) $f = P(\theta_1)R(\alpha, E)$, where $P(\theta_1)$ is the probability of scattering through an angle θ_1, $R(\alpha, E)$ is the backscattering coefficient of the theory of Ref. 22, and $\alpha = \theta - \alpha_0$. The scattering cross section is expressed in the form

$$\sigma(\theta, \varepsilon) = \iint P(\theta_1) R\left(\frac{\alpha}{\alpha_1+\alpha_0}, \frac{E}{E_2}\right) \frac{\partial(\theta_1, \theta_2)}{\partial(\alpha_1, E_2)} \sigma_2(\theta_2)\, d\alpha_1\, dE_2. \tag{9}$$

Here $P(\theta_1) = \delta_1/(\theta_1^2 + \delta_1^2)$, which is an approximation of the probability of deflection of the atom by an angle θ_l over a path L^* as a result of multiply screened Coulomb scattering.[21] The parameter $\delta_1 U_1 \pi n L^*/E_0$ depends on the initial (E_0) and final (E) energies of the scattered atom via L^* (n is the number of atoms per unit volume). The function R of Ref. 22 is of the form

$$R(\psi, \varepsilon) = \frac{3^{1/2}\psi}{2\pi^{3/2}\varepsilon^{1/2}} \frac{\exp\{-(\psi^2+\psi+1)/\varkappa s\}}{\varkappa s^2} \operatorname{Erf}\left[\left(\frac{3\psi}{\varkappa s}\right)^{1/2}\right], \tag{10}$$

where $\Psi = \alpha/\alpha_0$, $\varepsilon = E/E_0$, $s = 1-\varepsilon^{1/2}$, $\varkappa$ is a dimensionless parameter and Erf (x) is the probability integral. The integration region in (10) is determined with the aid of a

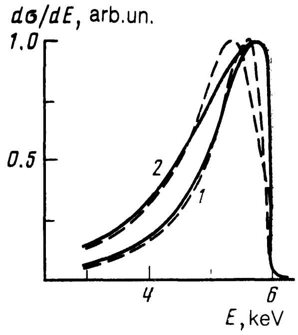

FIG. 3. Energy distribution of D^+ ions scattered by a polycrystalline Na surface bombarded by atomic D^+ ions (1) and by molecular D_2^+ ions (2) of energy 6 keV/atom. Solid curves—experiment,[20] dashed—calculations: 1—from Eq. (10) of Ref. 22, 2—from Eq. (9).

kinematic diagram similar to that given in Ref. 16. The integration variable α_1 is the angle of scattering of the molecule atom by the partner in the lab, and the region of integration with respect to energy is a combination of two joined segments, $[E, E_0(1-2\sin\alpha_1)]$ and $[E_0(1+2\sin\alpha_1), E_{2\,\max}]$. The lower integration limit is E, since $R(\psi,\varepsilon)=0$ for $\varepsilon\geqslant 1$. The upper integration limit $E_{2\,\max}$ is given by

$$E_{2\,max}=E_0(\cos\alpha_1+\sin\alpha_1\operatorname{ctg}\beta)^2, \tag{11}$$

where β is the root of the equation $\alpha_1=\sin\beta\tan\beta\tan[(\beta+\alpha_1)/2]$. Since the function $R(\psi,\varepsilon)$ decreases rapidly with increase of the scattering angle, it is the small angles that contribute to the integral in (9). At $\alpha_1\ll 1$ the energy $E_{2\,\max}$ can be approximated by the expression

$$E_{2\,max}=E_0[1+(2\alpha_1^2)^{1/3}]. \tag{12}$$

In the integration over the angle it must be recognized that the angle of scattering of the molecule atoms by one another is $\theta_2\geqslant\theta_{\min}$ where $\theta_{\min}=2U_2/E_0l^2\theta^2$ is the minimum scattering angle, determined by the finite distance l between the molecule atoms. As $\theta_{\min}\to 0$, the contribution of the small angles increases without limit, the cross section $\sigma_2(\theta_2)$ becomes σ-function-like, and the integration (9) yields a nonbroadened function $R(\alpha, E)$ corresponding to the scattering of atomic particles.

The cross section $\sigma(\theta, E)$ in (9) was calculated for bombardment of a polycrystalline Na surface by D_2^+ ions of initial energy $E_0=6$ keV/atom at a glancing angle $\alpha_0=5°$ and a scattering angle $\theta=10°$. The parameter $\varkappa$ in (10) was chosen to make the function $R(\alpha, E)$ describe correctly the energy distribution of the scattered particles in bombardment by atomic ions. The calculation result is shown in Fig. 3. It can be seen that the low-energy part of the spectrum is satisfactorily described by expression (9). The reason for the deviation from experiment in the high-energy part is that the ions scattered with energy close to the initial one have a short path in the material and the change of energy of such particles can no longer be regarded as a result of a trajectory change and expressed in terms of the change of the effective glancing angle. In addition, the function $R(\alpha, E)$ (Ref. 22) itself differs in the high-energy part from the experimental dependence.

In the region $E>E_0$ the broadening of the spectrum can take place, just as for heavy particles, only as a result of a direct redistribution of the energy among the colliding partners. An estimate of the broadening of the spectrum in this region can be obtained from the equations of Secs. 2 and 3. It turns out to be small, but experimentally observable at $E>E_0$ (Fig. 3).

5. DEGREE OF IONIZATION OF SCATTERED PARTICLES

The interaction of the molecule atoms with one another when scattered by the surface of the solid can influence various inelastic processes. In particular, the degree of ionization of the scattered particles can be different, depending on whether the solid is bombarded by molecule or atomic ions. This should be particularly manifested if a collision between the molecule atoms occurs on the final section of the trajectory, for example when at large angles of heavy-molecule scattering by a solid surface.

The evolution of the charge state of a scattered particle in processes in which electrons are captured and lost was considered in Refs. 23 and 24. Electron loss occurs in a hard collision of an incident particle with a surface atom, when the inelastically transferred energy $\Delta\varepsilon_{\text{in}}$ exceeds the ionization energy J.

The following equation was obtained in Ref. 23 for scattering of an ion by surface atoms:

$$\eta_a{}^+(E)=\left\{\exp\left(-\frac{v_c}{v_{\perp i}}\right)+F(E_0,\theta)\left[1-\exp\left(-\frac{v_c}{v_{\perp i}}\right)\right]\right\}\times\exp\left(-\frac{v_c}{v_{\perp f}}\right), \tag{13}$$

where $v_{\perp i}=v_0\sin\alpha$ and $v_{\perp f}=v_1\sin(\theta-\alpha)$ are the components, normal to the surface, of the incident-ion velocity v_0 and the scattered-ion velocity v_i respectively; v_c is a parameter characterizing the electron capture, $\exp(-v_c/v_{\perp i})$ is the fraction of ions in the beam incident on the surface atom, and $F(E_0,\theta)$ is the ionization probability of an atom of energy E_0 scattered by a surface atom at an angle θ. The probability $F(E_0,\theta)$ was calculated from the equation[24]

$$F(E_0,\theta)=\Delta\varepsilon_{\text{in}}/J-1.$$

As shown in Sec. 2, in surface bombardment by molecules, deflection of a particle by an angle θ is the result of scattering of a molecule atom by a surface atom at an angle θ_1 and additional collision with the partner in the molecule. Since $\theta_1>\theta$, the collision of the molecule atom with the surface atom is harder than in atomic-ion scattering by the same angle θ. As a result, the probability of electron loss is increased. This should increase the degree of ionization η_M^+ of the scattered particles in bombardment by atomic ions, at equal initial energy E_0 per atom at equal scattering angle θ.

The degree of ionization η_M^+ is calculated by summing over different molecule-atom trajectories leading to scattering by an angle θ. As a result we get

$$\eta_M{}^+=\left\{\int_0^\pi\exp[-(v_c/v_{\perp f})\exp(-az(\gamma))]\sigma(E,\theta,\gamma)\times[B+(1-B)F_2(E_0,E,\gamma)]d\gamma\right\}[\sigma(E,\theta)]^{-1}, \tag{14}$$

where

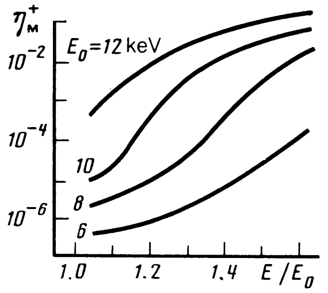

FIG. 4. Degree of ionization η_M^+ of particles scattered by a single-crystal Cu surface bombarded by N_2^+ molecules vs the energy of the scattered particles.

$$B=\left[\frac{1}{2}\exp\left(-\frac{v_c}{v_{\perp i}}\right)+\left(1-\frac{1}{2}\exp\left(-\frac{v_c}{v_{\perp i}}\right)\right)F_1(E_0,E,\gamma)\right]$$
$$\times\exp\left[-\frac{v_c}{v_\perp(\gamma)}[\exp(-az(\gamma))-1]\right]$$

is the probability of the particle remaining charged prior to collision with its partner; $v_{\perp i}, v_\perp, v_{\perp f}$ are the components, normal to the surface of the respective of the atom in incidence on the surface, after scattering by a surface atom, and after scattering by its molecular partner; $z(\gamma)$ is the distance from the surface to the point of collisions between the molecule atoms; $a\approx 2$ Å^{-1}; F_1 and F_2 are the ionization probabilities in the first and second collision. The scattering cross section $\sigma(E, \theta_1\ \gamma)$ was calculated in Ref. 16.

Figure 4 shows the dependence of the degree of ionization $\eta_M^+(E)$ on the energy of an atomic ion N^+ scattered by the surface of a copper single crystal, calculated from Eq. (14) for different energies of bombardment by N_2^+ molecule ions. As seen from Fig. 4, molecular effects are manifested in the degree of ionization $\eta_M^+(E)$. Their gist is that $\eta_M^+(E)$, unlike $\eta_a^+(E)$, depends on the initial energy E_0. A dependence of $\eta_a^+(E)$ on E_0 was observed also in Ref. 25. It was shown in Ref. 23, however, to be due to the nonequilibrium character of electron capture and loss in scattering by the surface, and to be manifested by the so-called surface peaks in $\eta_a^+(E)$ when E is approximately equal to the single-scattering energy E_1. On the contrary, the dependence of $\eta_M^+(E)$ on E_0 manifests itself in the region $E>E_0$ and is due to additional collisions.

It was shown in Ref. 5 by analysis of experimental data[3] that the ionization degrees $\eta_M^+(\theta)$ and $\eta_a^+(\theta)$ of ions scattered in bombardment by molecular and atomic ions N_2^+ and N^+, respectively, are different at glancing angles $\alpha_0 \geqslant 5°$ and are equal at smaller angles. This behavior of the degree of ionization can be attributed to blocking effects in ion scattering by a single-crystal surface. At small glancing angles the ion scattering is the result of multiple collisions. Blocking makes impossible scattering from a surface atom at the large angles needed for the subsequent additional scattering by the partner. These collisions become possible only for glancing angles $\alpha_0 > \alpha_{cr}$ (Ref. 26). The critical glancing angle, calculated with an inverse-square potential $U(r) = U_0/r^2$, is given by

$$\alpha_{cr} = (\pi U_0/2E_0 d^2)^{1/3}, \qquad (15)$$

where d is the distance between the atoms on the surface. For $N^+ \to Cu$ scattering with initial energy $E_0 = 15$ keV, when the scattering plane coincides with the ⟨100⟩ direction on a ⟨100⟩ face, the critical angle is $\alpha_{cr} \approx 7.7°$, which is close to the observed value.

For $\alpha_0 > \alpha_{cr}$ and $\theta = 2\alpha$ the degree of ionization η_M^+ increases with θ, whereas $\eta_a^+(\theta)$ becomes practically independent of θ (Fig. 5). This difference also confirms the role played in the evolution of the charge state of atoms by additional ionization in collisions between the atoms. In fact, in accordance with (14) the degree of ionization increases with increase of the angle θ_1, which increases in turn with α_0, whereas η_a^+, according to Eq. (13), tends to saturate with increase of the velocity component normal to the surface, i.e., with increase of the emission angle $\alpha = \theta - \alpha_0$ (Fig. 5). Since α_{cr} depends on the distance between the atoms, the difference between the degrees of ionization of scattered particles in bombardment by molecular and atomic ions should be manifested at different glancing angles, depending on the azimuthal angle of the target rotation relative to the beam-incidence plane. It would be of interest to verify experimentally the existence of this azimuthal orientational effect in the degree of ionization of scattered atoms when a single-crystal surface is bombarded by molecules.

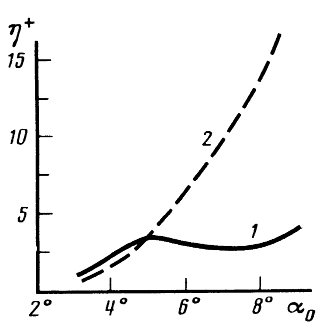

FIG. 5. Degree of ionization vs the glancing angle α_0 calculated from theory[5] and experiment[3]: 1 — η_a^+, 2 — η_M^+.

[1] W. Eckstein, H. Verbeck, and S. Datz, Appl. Phys. Lett. **27**, 527 (1975).
[2] W. Heiland, U. Beitat, and E. Taglauer, Phys. Rev. **B19**, 1677 (1979).
[3] L. Balashova, A. I. Dodonov, O. B. Firsov, *et al.*, Rad. Eff. **77**, 67 (1983).
[4] I. S. Bitenskiĭ and E. S. Parilis, Zh. Tekh. Fiz. **51**, 1798 (1981) [Sov. Phys. Tech. Phys. **26**, 1042 (1981)].
[5] I. S. Bitensky and E. S. Parilis, Nucl. Instr. Meth. **B2**, 384 (1984).
[6] M. Jakas and D. Harrison, Surf. Sci. **149**, 500 (1985).
[7] V. I. Shul'ga, Zh. Tekh. Fiz. **55**, 2027 (1985) [Sov. Phys. Tech. Phys. **30**, 1189 (1985)].
[8] H. H. W. Feijen, Thesis, Groningen, 1975, p. 43.
[9] S. A. Evstingeev, S. N. Zvonkov, and V. M. Chicherov, Zh. Eksp. Teor. Fiz. **82**, 1096 (1982) [Sov. Phys. JETP **55**, 640 (1982)].
[10] A. I. Dodonov, Sh. N. Garin, E. S. Mashkova, and V. A. Molchanov, Surf. Sci. **140**, L244 (1984).
[11] B. Willerding, H. Steininger, K. J. Snowdon, and W. Heiland, Nucl. Instr. Meth. **B2**, 453 (1984).
[12] B. Willerding, W. Heiland, and K. J. Snowdon, Phys. Rev. Lett. **53**, 2031 (1984).
[13] V. Imke, K. J. Snowdon, and W. Heiland, Phys. Rev. **B34**, 41 (1986).
[14] I. S. Bitenskiĭ, Ya. S. Gilenko, and E. S. Parilis, Izv. AN SSSR Ser. Fiz. **49**, 1988 (1985).
[15] A. A. Mukhamedov, E. S. Parilis, and F. F. Umarov, *ibid.* **43**, 630 (1979).
[16] I. S. Bitenskiĭ, Ya. S. Gilenko, and E. S. Parilis, Zh. Tekh. Fiz. **57**, 1692 (1987) [Sov. Phys. Tech. Phys. **32**, No. 9 (1987)].
[17] A. A. Mukhamedov and E. S. Parilis, Pis'ma Zh. Eksp. Teor. Fiz. **7**, 1474 (1981) [Sov. J. Tech. Phys. Lett. **7**, 631 (1981)].
[18] V. P. Vlasov, S. A. Evstingeev, S. N. Zvonkov, *et al.*, Pis'ma Zh. Eksp.

Teor. Fiz. **35**, 508 (1982) [JETP Lett. **35**, 628 (1982)].

[19] V. S. Remizovich, M. I. Ryazanov, and I. S. Tilinin, Dokl. Akad. Nauk SSSR **251**, 848 (1980) [Sov. Phys. Doklady **25**, 272 (1980)].

[20] P. J. Scheider, W. Eckstein, and H. Verbeek, J. Nucl. Mater. 111–112, 795 (1982).

[21] W. H. Escovitz, T. R. Fox, and R. Levi-Setti, IEEE Trans. NS-26, 1147 (1979).

[22] V. S. Remizovich, M. I. Ryazanov, and I. S. Talinin, Zh. Eksp. Teor. Fiz. **79**, 448 (1980) [Sov. Phys. JETP **52**, 225 (1980)].

[23] E. S. Parilis and V. Kh. Ferleger, Poverknost, No. 7, pp. 13 and 21, (1980).

[24] S. L. Nizhnaya, E. S. Parilis, and V. Kh. Ferieger, *ibid.*, No. 4, 72 (1982).

[25] W. Eckstein, V. A. Molchanov, and H. Verbeek, Nucl. Instr. Meth. **149**, 599 (1978).

[26] Yu. V. Martynenko, Rad. Eff. **20**, 211 (1973).

Translated by J. G. Adashko