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THE DEPENDENCE OF RADIATIVE EMISSION FROM ATOMS SPUTTERED FROM SOLID SURFACE BY ION BOMBARDMENT ON THE GEOMETRY OF EXPERIMENT*

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The visible light emitted by Zn atoms sputtered from mono- and polycrystalline targets by He^+ ions of energy 8 keV was analyzed. The intensities of Zn I triplet lines (468 nm, 472 nm and 481 nm) were measured as a function of the incident angle of the primary beam. The experimental results were compared with theoretical calculations. The model used in calculations relates the mechanism of sputtering, excitation of atoms and radiationless relaxation. The value of the radiationless relaxation coefficient for which the experimental results best conform to the theoretical predictions has been determined.

1. Introduction

Initial investigations concerned with the influence of the incidence angle of the primary ions on photon emission from atoms sputtered from the target were performed for mono- and polycrystalline copper targets bombarded with Ne^+ and Ar^+ ions of energy between 10 and 60 keV [1, 2]. The authors of these papers found an empirical dependence relating the intensities of Cu I lines to the incidence angle of primary ions. They noticed a strong minimum for 45° when the face (100) of the Cu single crystal was turned around [100] axis. The presence of the minimum was explained as a consequence of the channeling effect. The paper of Kerkow [3] was concerned with the dependence of photon emission from sputtered atoms on the incidence angle of the primary alkaline ions of energy 2–10 keV. The aim of his work was to find the geometrical shape of the aurora that is formed by the sputtered atoms shining above the target. The present authors have reported, in previous papers [4–6], strong radiative emission of the sputtered Zn atoms. This research was continued to obtain the dependence of intensities of Zn I lines on the incidence angle of primary ions. The aim is to obtain data on the influence of the geometrical conditions on the excitation and radiationless relaxation of the excited atoms.

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2. *Apparatus and experimental technique*

The apparatus used in this work was described in earlier papers [5, 6]. The general experimental set up remained unchanged. The beam density was about 70–80 microamperes per cm^2 and the pressure of the restgases in the collision chamber was maintained around 10^{-5} Tr. The target was mounted on a goniometric cradle that enables one to change the incidence angle in the range 0–90°. The radiation was observed along an axis perpendicular to the direction of the primary beam. The surface of the target was slightly inclined towards the optical axis by 2°–3°. In this way it was possible to observe light generated just above the surface. The geometry of scattering and observation is shown in Fig. 1.

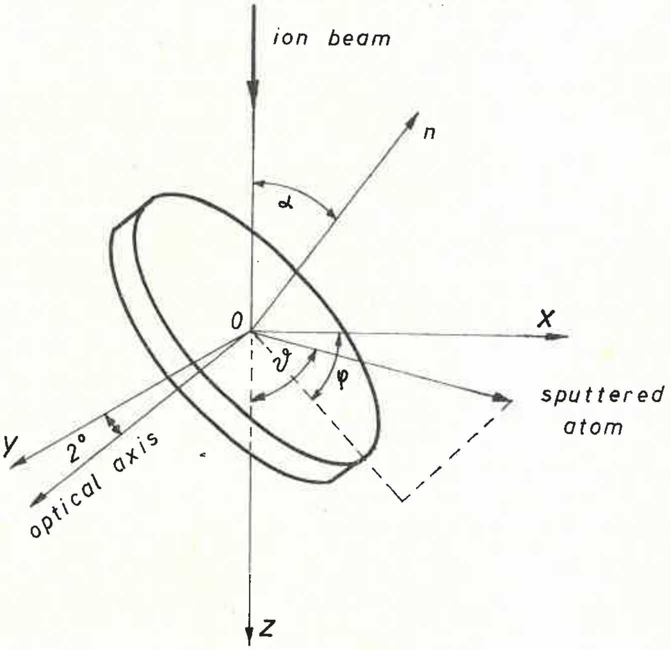

Fig. 1. Geometry of scattering and observation

Before measurements began the target was etched and chemically polished. The construction of the goniometric cradle provided for the heating of the target during bombardment in order to prevent deposition of restgas molecules and especially hydrocarbonic contaminations from the diffusion oil pumps. Zinc targets were heated to 200°C.

During bombardment of the polycrystalline Zn target and single crystal (1000) face by He$^+$ ion beam of energy 8 keV the line spectra of He I and Zn I in the range 430 nm – 650 nm were observed. These spectra are from primary ions neutralized over the surface and target atoms sputtered during ionic bombardment. A typical microphotogram of the spectrum for the incidence angle 60° is presented in the Fig. 2.

Detailed analysis of the registered spectra was performed in paper [6] and therefore we shall not repeat it here.

The dependence of relative intensities of radiation emitted by sputtered atoms on the incidence angle of primary ions was determined by a photographic method for the strongest Zn I triplet lines with wavelengths 468.0 nm, 472.2 nm and 481.1 nm. The incidence angle varied from 6° to 66° for a polycrystalline target, and from 18° to 63° for a single crystal.

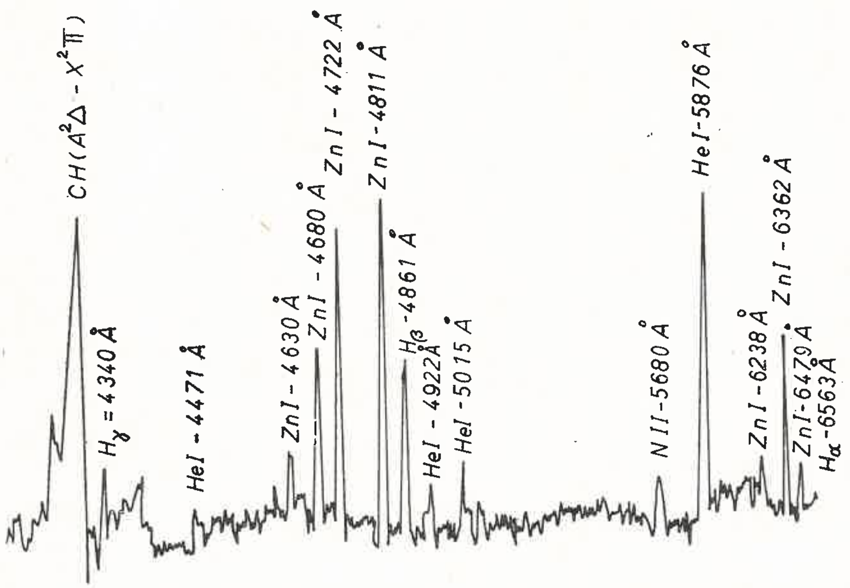

Fig. 2. Typical spectrum induced in $He^+ \to Zn$ (single crystal) impacts

3. The model of the phenomenon

To describe the angular dependence of the radiation emitted by sputtered atoms on the incidence angle of primary ions one must assume some model relating the mechanism of sputtering, excitation and radiationless relaxation of the excited atoms. All these factors strictly depend on the structure of the target and the nature of the projectile and its energy. Therefore, the proposal of the authors are confined to impacts of He^+ ions of energy 8 keV with a Zn target, however these results can be generalized for other cases.

3.1. The sputtering of zinc target

The majority of contemporary theories are based on the assumption, that the sputtering of atoms from the target is due to the cascade of the quasiellastic binary collisions between the primary ions and the target atoms. The classification of the collisions between atoms of atomic numbers Z_1 and Z_2 has already been done by Bohr [7], who introduced for this purpose such parameters as

— the screening radius $a = a_0(Z_1^{2/3}+Z_2^{2/3})^{-1/2}$, where $a_0 = 0.53 \times 10^{-8}$ cm;

— the distance of closest approach $b = Z_1Z_2e^2/\frac{1}{2}mv^2$, where m denotes the reduced mass of the colliding atomic system, and v is the relative velocity of the atoms.

In the case taken into consideration (He^+-Zn; $E_0 = 8$ keV) the ratio $b/a = 0.68$ indicates that a strongly screened Coulomb interaction occurs. The interaction potential is given by

$$V = (Z_1 Z_2 e^2/r) \exp(-r/a). \tag{1}$$

The differential cross-section for scattering of the particles that interact with themselves according formula (1) was numerically calculated for various values of the ratio b/a by Everhart et al. [8]. To obtain better agreement between theoretical and experimental data one must assume some limitations. According to Bulgakov [9] the main participants in sputtering are atoms lying near the surface. These atoms are ejected directly above the surface during ionic bombardment or leave the surface after a few collisions with neighbouring atoms in the lattice. This approximation seems to be correct especially for describing photon emission due to the sputtered atoms.

3.2. Remarks on the excitation of the sputtered atoms

According to Kerkow [3] the excitation of the atoms sputtered under bombardment by ions having energy between a few keV and a few tenth keV takes places during collisions between ions and atoms lying on the surface, but only those atoms that move outside the target contribute to the radiation. However, we must consider that non-zero radiation intensity is obtained for a normally directed ion beam. This means that part of the atoms primarily scattered towards the interior of the target changes direction after collisions with neighbouring atoms in the lattice and then can leave the surface in an excited state.

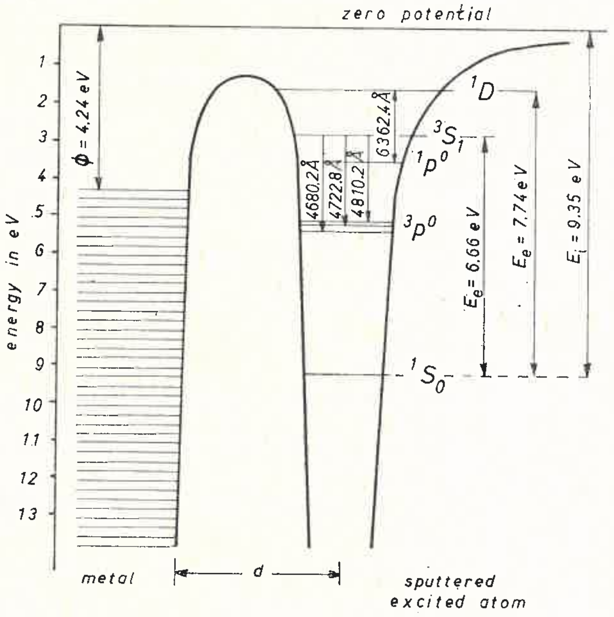

Fig. 3. System of energy levels for atom and solid (zinc) separated by small distance

The contribution to radiation due to the deeper atomic layers decreases rather rapidly. In regard to this simplified picture we do not assume the evident dependence of the probability of excitation on the incidence angle of the primary ion beam.

3.3. The radiationless relaxation of the excited atomic states

The measurements were performed for the spectral lines that have a common upper level with the excitation energy of 6.66 eV. Mutual position of the electron energy bands in metallic zinc and the atomic levels of a sputtered Zn atom located at a distance "*d*" in the front of the metallic target is shown in Fig. 3. The picture indicates the probability of a tunnel resonance transition of the electron from the excited atomic level to the empty conductivity band in the metal. The mechanism of this transition is described in the paper of Van der Weg and Rol [10]. The probability of the sputtered atom escaping in excited state outside the surface without radiationless relaxation is given by the formula

$$P = \exp(-A/av_{\perp}), \tag{2}$$

where A and a are constants depending on the system atom-solid and $v_{\perp}$ is the component of the velocity of the sputtered atom perpendicular to the surface.

The ratio A/a can be considered as a coefficient of radiationless relaxation.

4. Calculations

Assuming the suppositions mentioned above we show the basic steps of calculation that give the relative dependence of the radiation of the sputtered atoms on the incidence angle α of the primary ion beam. The geometry of experiment is shown in Fig. 1. The z-axis coincides with the direction of bombardment and the x-axis lies on the incidence plane of the beam. The number of particles sputtered in the direction denoted by angles ϑ and φ is

$$dN(\vartheta, \varphi) = N_0 \sigma(\vartheta)\, d\Omega \tag{3}$$

where N_0 is the number of atoms in the surface layer participating in the collisions; $\sigma(\vartheta)$ is the differential cross-section for sputtering the atom from the target in the direction denoted by angle ϑ. The numerical values of function $\sigma(\vartheta)$ were calculated in paper [8].

If one assumes that the ion beam density is constant then the number of the target atoms participating in impacts depends simply on the incidence angle

$$N_0 = N_0'/\cos\alpha.$$

The geometry of the collision implies that only those atoms sputtered according to formula (3) can be ejected above the surface for which the angle φ varies in the range $-\varphi_0, \varphi_0$ and the scattering angle ϑ varies in the range $\vartheta_1(\varphi), \vartheta_2$ where ϑ_1 — is the value of the scattering angle at which the atom slides over the surface, ϑ_2 — is maximal value of the scattering angle at which the energy transferred to the sputtered atom is higher than the binding energy in the lattice, φ_0 — is the angle determined by the formula $\vartheta_1(\varphi_0) = \vartheta_2$.

According to the assumptions mentioned above the probability of excitation is constant for the considered energy of primary ions and the spectral line induced during collision.

Finally, if one considers radiationless relaxation and the symmetry of the system, the number of the excited atoms sputtered above the surface and sharing in the radiation is expressed by the formula

$$N_{\mathrm{I}}^{*}(\alpha) \sim \cos^{-1}\alpha \int\limits_{0}^{\varphi_0}\int\limits_{\vartheta_1(\varphi,\alpha)}^{\vartheta_2} \sigma(\vartheta)\cdot\exp\left(-\frac{A}{av_{\perp}(\vartheta,\alpha)}\right)\sin\vartheta d\vartheta d\varphi. \tag{4}$$

Other atoms scattered inside the target can be drawn out to the surface if reflection on the neighbouring atoms occurs. The probability of this incident is determined by

$$W(\alpha) = 1-\exp\left(-\frac{\bar{d}}{\lambda\,\overline{\cos x(\alpha)}}\right), \tag{5}$$

where $\bar{d}$ denotes the mean interatomic distance, λ is the mean free path of atoms inside the target and $\overline{\cos x(\alpha)}$ is the mean value of the cosine of the scattering angle for atoms moving inside the target. The interaction between atoms colliding inside the target can be decribed by an elastic spheres model and therefore the mean free path λ and mean velocity $\bar{v}(\alpha)$ of the atoms after collision needed for equation (2) can be easily calculated.

A second group of sputtered atoms originated as a consequence of secondary collisions inside the target shares in the radiation as expressed by formula

$$N_{\mathrm{II}}^{*}(\alpha) \sim \cos^{-1}\alpha\Big[\int\limits_{0}^{\varphi_0}\int\limits_{0}^{\vartheta_1(\varphi,\alpha)} \sigma(\vartheta)\sin\vartheta d\vartheta d\varphi + \int\limits_{\varphi_0}^{\pi}\int\limits_{0}^{\vartheta_2}\sigma(\vartheta)\sin\vartheta d\vartheta d\varphi\Big]$$

$$\times W(\alpha)\exp\left(-\frac{A}{a\bar{v}_{\perp}(\alpha)}\right). \tag{6}$$

Joint photon emission is the sum of the intensities described by formulas (5) and (6)

$$I(\alpha) = \mathrm{const}\,(N_{\mathrm{I}}^{*}(\alpha)+N_{\mathrm{II}}^{*}(\alpha)). \tag{7}$$

5. *Results*

The measurements of the dependence of radiation emitted by sputtered atoms on the incidence angle of primary ions demonstrated no essential difference among single crystal and polycrystalline targets. The character of both functions is the same with the exception of the relative difference between maximal and minimal values of the functions within the range of the measurements. This difference is larger for the polycrystalline target. The function $I(\alpha)$ experimentally determined for Zn I 481.1 nm line in the collision $\mathrm{He}^{+} \rightarrow \mathrm{Zn}$ (polycrystalline target) is shown in Fig. 4. The results of numerical calculations according to formula (7) are shown by a smooth curve on the same picture.

Expressions (5), (6) and (7) show that the slope of the function $I(\alpha)$ is determined by the value of the coefficient A/a. This coefficient was fitted to obtain the best agreement

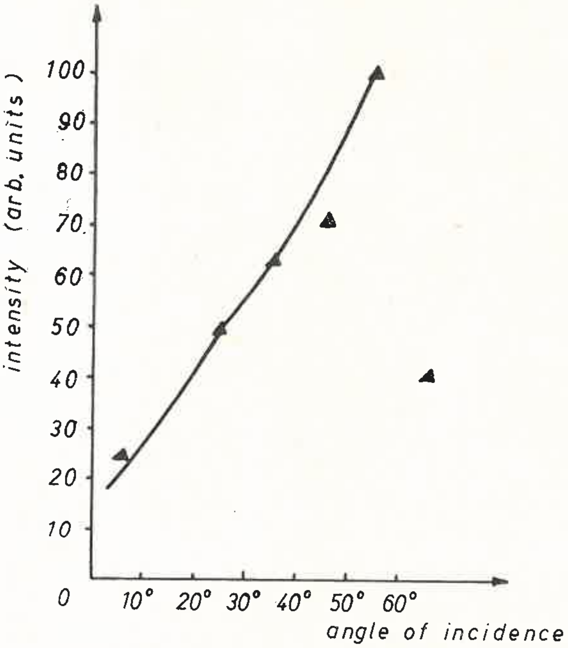

Fig. 4. Plot of the function $I(\alpha)$ for $He^+ \rightarrow Zn$ (polycrystalline target) impacts. Smooth curve — theory; triangles — experimental points

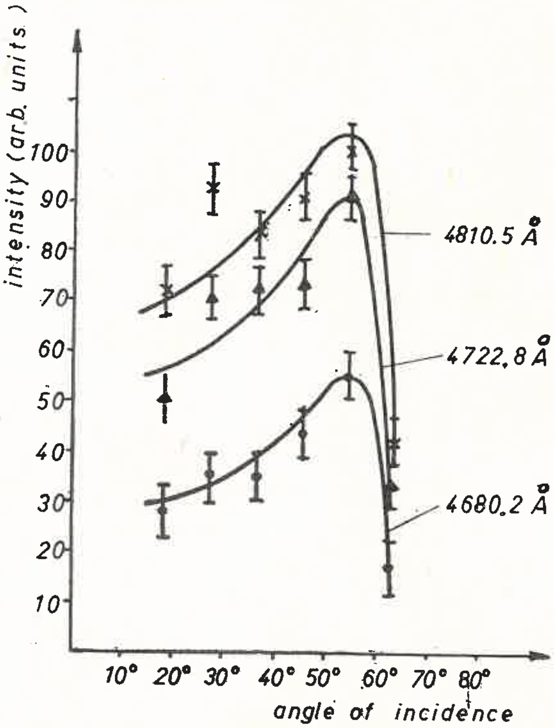

Fig. 5. Plot of the function $I(\alpha)$ for $He^+ \rightarrow Zn$ (single crystal) impacts. Smooth curve represents the mean experimental values for each component of Zn I triplet

with experimental points by the method of least squares. The best value of the coefficient of radiationless relaxation is $A/a = 8.0 \times 10^4$ cm/sec. For this value the standard deviation is $\sigma = 3.25$ arb. units.

The model described above does not take into consideration the strong fall of the sputtering ratio for large angles of incidence. This was observed by many authors and is the reason for the deviation between the last experimental point and the calculated value in Fig. 4.

Experimental results for three components of Zn I triplet emitted in $He^+ \rightarrow Zn$ (single crystal) are shown in Fig. 5.

6. *Conclusions*

The results indicate that the monotonical growth of the intensity of light from small angles up to nearly 55° can be explained by a simple model of sputtering and optical excitation based on the binary collision theory. The decadent part of the curve just above 55° is probably due to the lower value of the sputtering ratio for incidence angles above the critical value. This was noted by Wehner [11] as is due to the strong dependence of the sputtering ratio on the beam density. The relatively large experimental error that amounts about 10% is related to the photographical method used in this work. The value of the coefficient of radiationless relaxation, determined in the present paper, indicates a high probability that this process, which leads to a considerable fall of the slow component of excited atoms sputtered from the surface, occurs.

The slope of the curve is very sensitive to the value of the parameter A/a. This means that the fitting the theoretical curve by a proper value of A/a makes the precise determination of this coefficient possible.

Further efforts will be undertaken to prove precision by photomultiplier registration with narrow-band interference filters. The application of optical filters instead of a spectrograph gives in this case a better opportunity to observe the totality of the radiation above the surface. On the other hand, we look for further arguments in favour of the proposed model of excitation especially by investigation of polarization effects that were initiated in our previous paper [6].

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