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SPRING COLLEGE IN CONDENSED MATTER
ON
"THE INTERACTION OF ATOMS & MOLECULES WITH SOLID SURFACES"
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ION BACKSCATTERING FROM CLEAN AND LOW COVERAGE ADSORBATE SURFACE (Part I)

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Part I

Energy spectra of light atoms and ions backscattered by clean polycristalline surface

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I Introduction

In present lecture I'll tell about some models, which we have proposed for description the energy spectra of backscattered light atoms and ions as H, D, He in Kev energy range [1,2,3]

It is well known, that without understanding the behaviors of these spectra one couldn't solve such important problems as thermonuclear reactor first wall state, creation quantitative methods, using backscattered ions for diagnostics surface composition and structure and some others

Firstly, let us consider the main features of experimentally measured spectra, Fig 1.

In this figures $\mathcal{E}$ is the relative energy of backscattered particles, $\mathcal{E} = E/E_0$, E_0 - is intitial energy of incident ion beam, α - is the angle of incidence and Ψ is the escaping angle. both are counted off from the surface θ is the polar scattering angle counted off from beam direction

R and R^+ are twice differented over energy $\mathcal{E}$ and solid angle Ω reflection coefficients for neutrals and ions respectively. From these spectra the next conclusions may be done:

The first one: all these spectra both for ions and neutrs contein a wide cupola-like maxima at relative energy $\mathcal{E}_{max}$. The values $\mathcal{E}_{max}$ tends to the right, that is to the large $\mathcal{E}$ region, when initial energy E_0 or θ decreose. The values $\mathcal{E}_{max}$ chanages in the range $0.1 \leq \mathcal{E}_{max} \leq 0.8$ So, the most probable energy loss of backscattered particles with relative energy $\mathcal{E}_{max}$ is about several Kev. As the main stopping cause, for a light particles are the inelastic losses, the Lss formula

$$\frac{dE}{dl} = K\sqrt{E} \qquad (1)$$

is appropriable

Inelastic energy losses by (1), in Kev energy range are about $10 \div 20$ eV/Å. Thus, the particles escaped from the solid with energy $\mathcal{E} = \mathcal{E}_{max}$, scattered along the trajectories with

length about thousand angstroms. As the result - the considered scattering is significantly plural one

The second one: the half-width of these spectra at fixed value E_0 is changed non-monotonically when agle θ increases and reaches a maximum at middle θ values.

The third one: the H and D spectra don't contein any pecularities besides cupola maximum. In contrast, the He spectra conteins a so-colled surface peaks - an additional maxima in the hight energy region $\varepsilon \approx \varepsilon_I$, where ε_I - is the energy of singlt He scattering by separate surface atom.

It is interesting to note, that in ion He spectra the surface peaks exist for any values of initial energy E_0, but in neutral spectra these peaks appear only for initial energies which do not exceed some critical value E_c. For example, in the nicol target case, the $E_c \approx 5$ keV Apparently, ion and neutral surface peak formation mechanizms, are quite different ones.

These all experimentally established features are the set of problem for energy spectra theory

II Cupola-like distributions

1. Single scattering models.

Reasonable discribtion of the cupola-like spectra as it was mentiod above, must be done by multiple scattering theory based on Boltzmann Kinetic equation. Using this method a lot of impotent results was obteined for glansing incidence spectra. Unfortunately, the methods for Boltzmann equation analitical solution for large angles are unknown up to now. Then, it is necessary to find another way for discribtion of these cupola-like spectra, measured at large insident angles

The well known simplest alternative apporoximation has been given by single scattering model developed by Mac Cracken and Freeman in 1969 for high initial energy $E_0 > 100$ KeV.

Here the reflection coefficient for potential function $U(r) = A/r^2$ and inelastic energy losses only, is:

$$R_I = \frac{d^2R}{d\Omega\, d\varepsilon} \simeq \left[\sqrt{\varepsilon}\,(\sqrt{\varepsilon}\sin\psi + \sin\alpha)^2\right]^{-1} \qquad (2)$$

Some attempts have been made to apply the simple formula (2) in the lower energy region. The comparison between formula (2) and experimental measured spectra represented in the fig. 1a It is clear, that this formula could describe only the high energy part of a spectra being larger, with the larger scattering angle θ. Really, R_I is monotonically decreasing function, but the experimental spectra always conteins a maximum. The first step to improve this model is to take into account the incident beam attenuation in the solid The beam attenuation functions P describes the probability of passing the length L in solid, without scattering at an angle, greater then Δ, both before and after the violent collision at angle θ, is:

$$P(\Delta) = \exp\left[-2\pi \int_0^L \int_\Delta^\pi n\sigma(E,\theta_1)\, d\Omega_1 dl\right] = \exp\left[-d\left(\frac{1}{\sqrt{2}}-1\right)\left(\frac{1}{\sin\Delta/2}-1\right)\right] \qquad (3)$$

where $d = 0.227\,(Z_1^{2/3}+Z_2^{2/3})(\pi\xi_e)^{-1}(E_B/E_0)^{1/2}$ for $U(r) = A/r^2$ potential function. Here Z_1 and Z_2 are the atomic numbers of the insident ion and target atom respectively, n - is the scattering centers concentration in the solid, $\xi_e = 1 \div 2$ is the LSS parameter and E_B is the Bohr energy. This modification of single scattering model, gives the next expression for reflection coefficient:

$$\bar{R}_I \sim \exp\left[-d\left(\frac{1}{\sqrt{2}}-1\right)\left(\frac{1}{\sin\Delta/2}-1\right)\right] \Big/ \sqrt{\varepsilon}\,(\sqrt{\varepsilon}\sin\psi + \sin\alpha)^2 \qquad (4)$$

$\bar{R}_I$ - function has a maximum at energy $\varepsilon = \varepsilon_{max}$, which is determined by equation

$$d\bar{R}_I / d\varepsilon = 0 \qquad (5)$$

It's solution may be written as:

$$\begin{cases} \varepsilon_{max} = a^2/4 , & \text{for } \alpha \to 0 \\ \varepsilon_{max} = a^2 , & \text{for } \psi \to 0 \end{cases} \qquad (6)$$

with $a(\Delta, z_1, z_2) = d\,(1/(\sin\Delta/2) - 1)$

In both cases $\varepsilon_{max} \sim E_0^{-1/2}$ in qualitative agreement with experiment. The formula (4) coensidence with experimentally measured spectra is shown in the next figures (Fig 2a, 2b, 2c) One can see, that the half-width of energy spectra $\Delta\varepsilon_{1/2}$ from ex. (4) agrees well with the data for large θ angles, satisfactory for middle angles and not well for small ones. The only free parameter Δ fits ε_{max} with experimental value

2. The meaning of the parameter Δ

In the frame of McCracken model one assumes, that all incident particles from parallel beam reaches the center of violent collision at the depth x and all particles, scattered at angle θ leaves the solid. Moreover, the initial and outgoing trajectories both are the strait lines

The beam attenuation function introduction means, that there is yet one collision on the path L and this additional collision at angle θ_1 divides the particles beam in the media into two parts:
- the particles scattered at angle θ_1 smaller than Δ, remain in parallel beam as unscattered ones;
- the particles scattered at angle θ_1 greater than Δ, leaves this beam without any probability for scattering at angle θ with energy ε

Therefore, the beam attenuation model is only the rough appoximation for plurual scattering theory. Here, the real angle and energy distribution for multiple scattered particles in the media are substituted by step-type function $P(\Delta)$, with Δ as any measure of plurality

Formally, this moudel come to McCracken's one if $\Delta = \pi$
The comporison between experimental data and calculation by

formula (4) has shown, that Δ values always are great enough:

$$\Delta = \chi\,\theta \qquad (7)$$

where θ is scattering angle and $\chi \leq 1$.
In ref. [1] we assumed that parameter Δ could be identifited with limiting angles for the family of double scattering trajectories at the given angle θ and given final energy ε.

Therefore, the validity of impirical formula (7) has been proved. For parameter χ the value $\chi \approx 0.5$ has been obtein.

3 Double scattering model

Significantly better results for discribtion of cupola-like spectra peculiarities, one can obtein in double scattering model, taking into account the beam attenuation over all the path in medva. This model is the next approximation to the real plurual scattering

Let a particle, after passing the length l_1 in the solid, be scattered at the angles (θ_1, φ_1) on the path dl_1 and then, after passing the length l_2, scattered at angles (θ_2, φ_2) on the path dl_2, Fig. The probabilities of these scattering may be written as:

$$\begin{cases} d^2R_1 = n\,P_{1,1}\,\sigma(\varepsilon_1, \theta_1)\,P_{1,2}\,d\Omega_1 dl_1 \\ d^2R_2 = n\,P_{2,1}\,\sigma(\varepsilon_2\,\theta_2)\,P_{2,2}\,d\Omega_2\,dl_2 \end{cases} \qquad (8)$$

where

$$\sigma(\theta, \varepsilon) = \frac{4.1\,e^2\,a_{T.\Phi}\,z_1 \cdot z_2\,(1+\nu)}{32\pi\,\varepsilon\,E_0\,\sin^3\theta/2} \qquad (9)$$

is the differential cross-section for $U(r) = A/r^2$ potential function, $a_{T.\Phi}$ - is the Thomas-Fermi radius, ν - is the mass ratio of insident ion and solid atoms masses.

$$\varepsilon_1 = (1 - l_1/l_0)^2, \quad \varepsilon_2 = (1 - (l_1+l_2)/l_0)^2 \qquad (10)$$

are the particle relative energies before the first and the second collisions respectively, l_0 - is total range for the insident particle in solid, $P_{i,\kappa}$ - are the beam attenuation function both before

and after each collision.

If these two scatterings are independent events, for the double scattering probability one could obtein:

$$d^4R = d^2R_1 \cdot d^2R_2 \qquad (11)$$

To calculate the total probability of double scattering it is necessary to integrate eq. (11) over all trajectories with given final values ε, θ; and $\varphi = 0$ These trajectorits some surface Φ formed, which equation is:

$$\cos\varphi_1 = [l_2\cos\theta_1\sin\alpha - (1-\sqrt{\varepsilon})l_0\sin\psi + (l_1+l_2)\sin\psi + l_1\sin\alpha] \times [l_2\cos\alpha\sin\theta_1]^{-1} \qquad (12)$$

As the result, we obtein:

$$R_{\bar{I}\bar{I}} = \frac{n^2}{\sin\theta} \iiint_{\Phi} \sigma_1\,\sigma_2\,P_{1,1}\,P_{1,2}\,P_{2,1}\,P_{2,2}\,\mathcal{J}\sin\theta_1\sin\theta_2\,dl_1\,d\theta_1\,d\varphi_1 \qquad (13)$$

Here $\mathcal{J}$ - is the transition Jacobian from $\varphi_1, \varphi_2, \theta_2$ to $\varepsilon, \psi, \theta$ variables. The function under integral is very comlicate, so this integral may be taken only by numerical computer calculation. However, the result of numerical calculation may be approximated by simple formula

$$R_{\bar{I}\bar{I}} \sim \frac{\exp\left[-d\left(\frac{1}{\sqrt{\varepsilon}}-1\right)\left(\frac{1}{\sin^3\theta/8}-1\right)\right]\cdot(1-\sqrt{\varepsilon})}{\sqrt{\varepsilon}\,(\sqrt{\varepsilon}\sin\psi + \sin\alpha)^3} \qquad (14)$$

The equation $dR_{\bar{I}\bar{I}}/d\varepsilon = 0$ has a solution $\varepsilon_{max}(E_0, \theta) < 1$ for all values of E_0 and θ

The result of comparison of the calculations by formula (14) with a wide set of experimental data are represented in the Fig. 4 and Fig. 5. From these figures it is clear, that double scattering approximation gives a reasonable agreement with experiment for cupola-like spectra

III Theory of surface peaks in neutral spectra

Formula (15) describing the main cupola-like part of the energy spectra, was obteined with the consideration of inelastic energy losses alone. But for the high energy part of the spectra $\varepsilon \to 1$ the quantitives of these two types of losses is comparable

For describtion the features of surface peak in neutral spectra, we use the following conception: in the region $\varepsilon \to 1$ the scattering is really quasisingle one. Angle θ is equal the summ:

$$\theta = \theta_1 + \theta_2(l) \qquad (15)$$

where θ_1 is the large violent collision angle, and θ_2 is the small angle for most probable multiple scattering on path l, which is proportional to l. The probability for scattering at angle $\theta_2(l)$ on the small path l is approximatelly equal to 1. Formula for the dependence $\theta_2(l)$ was obteined by Fyrsov in the theory of multiple scattering in matter

$$\theta_2(l) = cl/E_0, \quad \text{where} \quad c = \frac{0.8\pi^2 \cdot n z_1 z_2 I_0 a_0^2}{(z_1^{2/3} + z_2^{2/3})^{1/2}} \qquad (16),$$

a_0 - is the Bohr radius, $I_0 = 27,2\,eV$.
The probability of quasisingle scattering at angle θ may be approximated by:

$$R_I \sim \frac{n\sigma(\varepsilon, \theta_1)}{d\varepsilon/dl} \qquad (17)$$

The surface peak appears, when $d\varepsilon/dl = 0$ The formula for $\varepsilon(l)$ including the inelastic energy loss in the violent collision at angle $\theta_1 = \theta - \theta_2(l)$ is:

$$\varepsilon(l) = 1 - \frac{2l}{l_0} - 4\nu\sin^2\frac{\theta - \theta_2(l)}{2} \qquad (18)$$

In formula (18) we have neglected the terms, proportional to small values: $\nu^2, (l/l_0)^2, \nu \cdot l/l_0$. From formula (18) it is seen, that with the increasing of the path l the inelastic energy losses increase, but elastic ones decrease, by decreasing the angle of violent collision. So, the dependence $\varepsilon(l)$ may have a maximum at some value $l = l_m$, which is the solution of equation $d\varepsilon/dl = 0$ with ex. (16) and ex. (18) one can obtaine:

$$l_m = \frac{E_0}{C}\left(\theta - \arcsin\frac{K\sqrt{E_0}}{2\gamma C}\right) \qquad (19)$$

where $K = 8\pi e^2 a_0 Z_1 Z_2 n \xi_c (Z_1^{1/3} + Z_2^{1/3})^{-3/2} \cdot E_B^{-1/2}$ - is the coefficient in LSS formula. The function $\varepsilon(\ell)$ changes really by nonmonotonic way and has the maximum if $l_m \geq 0$. The final condition is realised only if initial energy is no greater than some critical energy E_c, where:

$$E_c = \left(\frac{2\gamma C \sin\theta}{K}\right)^2 \qquad (20)$$

It is clear that the energy E_c is the critical energy for surface peak existanee. The dependences $\varepsilon(\ell)$ by formula (18) are shown in the Fig 6. In the Fig 6a, where E_0 is smaller then E_s, $\varepsilon(\ell)$ has a maximum and the surface peak exists. In the Fig 6b, where $E_0 > E_c$ there are no maxima and no surface peak.

So, the surface peaks in neutral spectra are the rainbow type peculurity caused by concentration of many trajecbories with different combinations of elastic and inelastic losses in narrow energy spectra range. The numerical values of critical energy may be easy calculated by ex. (21)

Then, for the case $He^+ \to Ni$ $\theta = 135°$, $E_c = 5.85$ kev and the experimantally found value is $E_s \approx 5$ kev. For $H^+ \to Ni$ $\theta = 135°$, $E_c = 355$ eV. For $D^+ \to Ni$, $\theta = 135°$, $E_s = 2.80$ kev. The final results explain the surface peaks absense in neutral H и D mesured spectra, where initial energy $E_0 \geq 3$ kev.

IV Theory of surface peaks in ion spectra.

Ion spectra for light backscattered particles also may be calculatied by double scattering model.

$$R^+ = \frac{n^2}{\sin\theta} \iiint_{\Phi} \sigma_1 \sigma_2 P_{1,1} P_{1,2} P_{2,1} P_{2,2} J Q^+_{(E,E_0,\gamma,\sin\theta_1,\sin\theta_2,d\ell,d\theta,d\gamma)} \qquad (21)$$

where $Q^+(E, E_0, \gamma)$ - is the ionization probability. So, for R^+ calculation it is necessary to now the Q^+ probability. Definision of Q^+ is the separate serios theoretical problem connected with investigation the electron exchange prousses between outgoing particle and the surface, hasn't exact solution up to now. The main difficulty caused by nonapplicability of quantum-mecanic perturbation theory for the particles having velosities, smaller then Fermi ones.

Let consider the model for charge state formation, which we proposed some yeurs ago. The basic prinsiples of this model are:

The first one: charge state of scattered atoms is formed in the competition of some electron capture and loss processes

The second one: this processes occur outside the surface only, if there is no effective bound state for atomic electron into the media and they occur inside the media too in opposite case

The scheme of these processes shown in the Fig 7. The capture processes are Auger or tunneling electron transitions from occupied part of metal conduction band to the ion. The back-electron transition to occupate conduction band is forbiden by Pauly-Fermi principle. Then, the loss processes are electron transition from the atom to the free part of conduction band. This transition needs energy transfer from nuclear subsistem to electronic one, which is similar to ionization processes during atom collision in gase phase. The capture and loss rates have been taken in form:

$$W_c = \begin{cases} A & , \ s < 0 \\ A f(s) & , \ s > 0 \end{cases} \qquad (22)$$

$$W_e = \begin{cases} B(E) & , \ s < 0 \\ B(E) f(s) & , \ s > 0 \end{cases}$$

Here s is the distance from the surface along the normal direction, $f(s)$ is some monotonically decreasing function: $\lim_{s \to \infty} f(s) = 0$ and $f(0) = 1$. In (22) formulak A is constant $A \approx 10^{15} \div 10^{16}\, s^{-1}$ and $B(E)$ is function, determined by expression $B(E) = n \sigma_e \cdot v$, where σ_e is eross-section of ionization in ion-atom collisions estimated by Firsov in form:

$$\sigma_e = \pi a_1^2 \left[(E/E_t)^{1/2} - 1\right]^2 \qquad (23)$$

$a_1 = 1.32\, a_0 Z_2^{-1/2} (Z_1^{1/6} + Z_2^{1/6})$, E_t is the threshold ionization energy.

The ionization probability is the solution of equation:

$$v_\perp \frac{dQ^+}{ds} = -[W_c + W_e] Q^+ + W_e \qquad (24)$$

with the initial condition $Q^+(s=0) = Q_1^+$

$v_\perp$ - is normal to the surface component of backscattered particle velosity. Q_1^+ is the ionization probability for particle on the surface $s=0$ The Q^+ from (24) is

$$Q^+ = \frac{B}{A} + \left(Q_1^+ - \frac{B}{A}\right) \exp(-v_0/v_\perp) \qquad (25),$$

where $v_0 = \int_0^\infty A(s)\,ds$ - is the model parameter

For our surface peak problem two different cases is necessary to distinguish. If atom hasn't boundary states for electron into the media, it may come to the surface only as ion and then $Q_1^+ = 1$ If these boundary states exist, the capture and loss processes take place in media too and Q_1^+ is the solution of analogous to (24) equation. But in this equation the monotonic stopping it is necessary to take into account. It's solution is:

$$Q_1^+ = \frac{B(\varepsilon)}{A} + (1 - l_i/l_0)^{2h}\left(1 - \frac{B(\varepsilon)}{A}\right) = Q_e^+ + \Delta Q^+ \qquad (26)$$

Here $h \approx 10^2$ l_i - is the particle path in solid after the last violent collision proposed as completly ionizing one

Equation (26) describes the charge equilibrum establishment into the solid. The first term Q_e^+ is the equilibrum charge and the second term ΔQ^+ is unequilibrum additave The dependence $Q_1^+(\varepsilon)$ in the single scattering approximation is shown at the Fig. 8. The sharp increas the value of Q_1^+ at $\varepsilon \to 1$, caused by ΔQ^+. As the parameter $h \approx 10^2$, then $\Delta Q_1^+ \approx 0$ for all trajectories with violent collision in solid Only for surface scattering, where $l_i \approx 0$, $\Delta Q^+ \approx 1$.

This sharp increas of the value Q^+ at $\varepsilon \to 1$ is the main cause for surface peak existance in ion spectra

The surface peaks exist in He ion spectra and do not exist in H and D spectra, because He and H has quate different ionization potentials $J_0(He) \approx 24.5$ eV, $J_0(H) \approx 13.6$ eV. As the result He can exist inside the metall as neutral atom, but H u D can't.

By substitution ex. (25) with Q_1^+ by ex. (27) into eq. (21), one can calculate the ion spectra. The result for He^+ shown in the Fig. 1d. It coincides well with experimental data.

So, the considered simple models are useful to describe all main peculiarities of backscattered light particle spectra.

References

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2. E.S Parilis, V.H. Ferleger, Poverhnost, 1985, v.4. p. 21
3. E.S Parilis, V.H. Ferleger, Poverhnost, 1985, v.5 p. 17

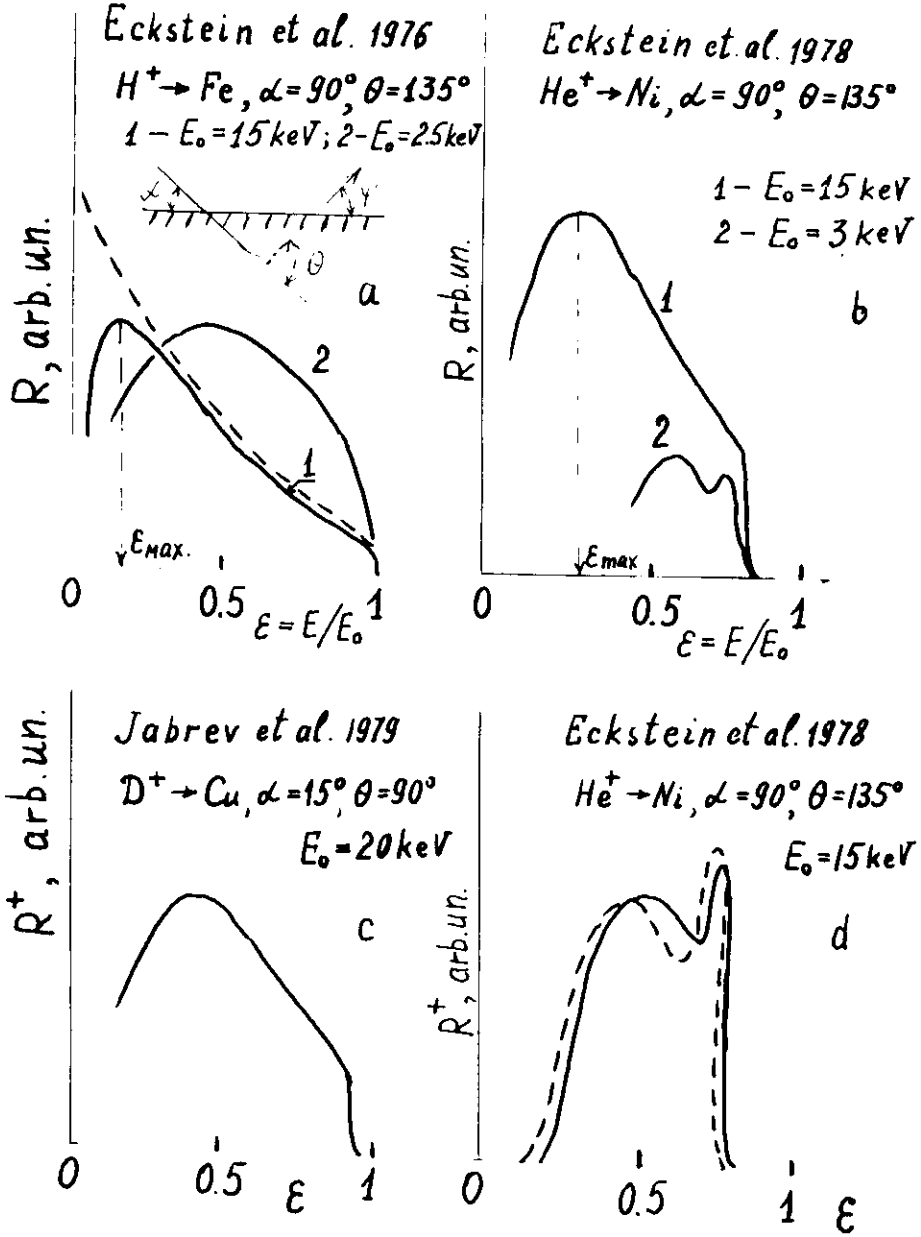

Fig. 1. The measured (solid lines) and calculated (dash lines) energy spectra.

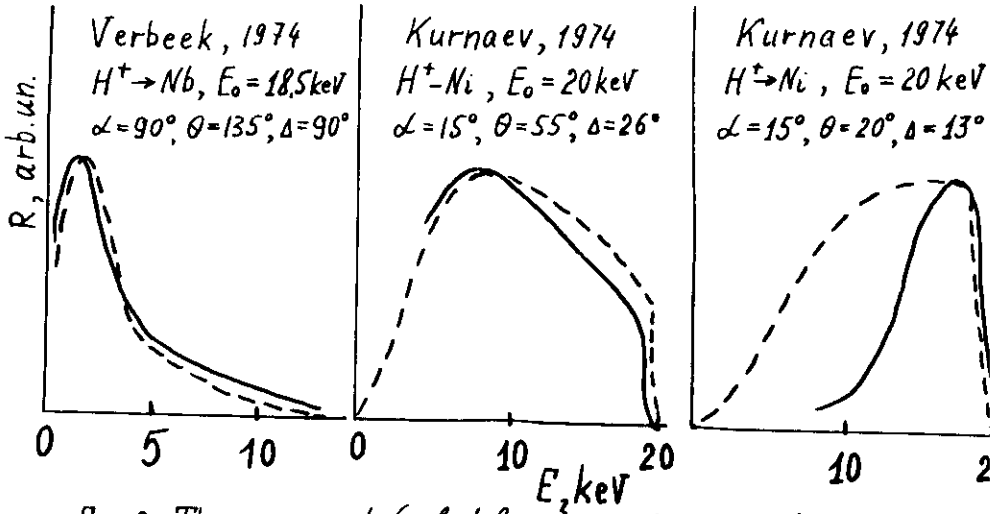

Fig. 2. The measured (solid lines) and calculated by formula (4) (dash lines) energy spectra.

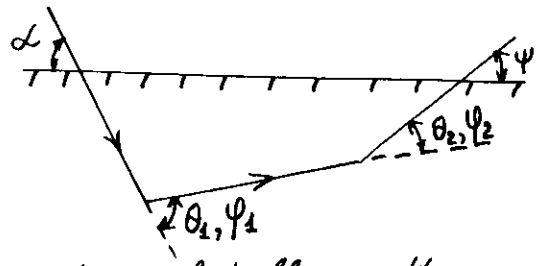

Fig. 3. The scheme of double sceattering.

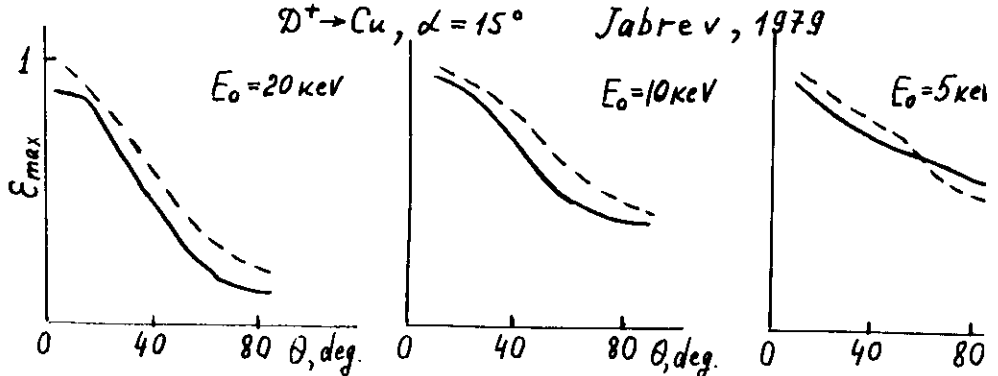

Fig. 4. The measured (solid lines) and calculated by formula (14) (dash lines) dependenses $\varepsilon_{max}(\theta)$.

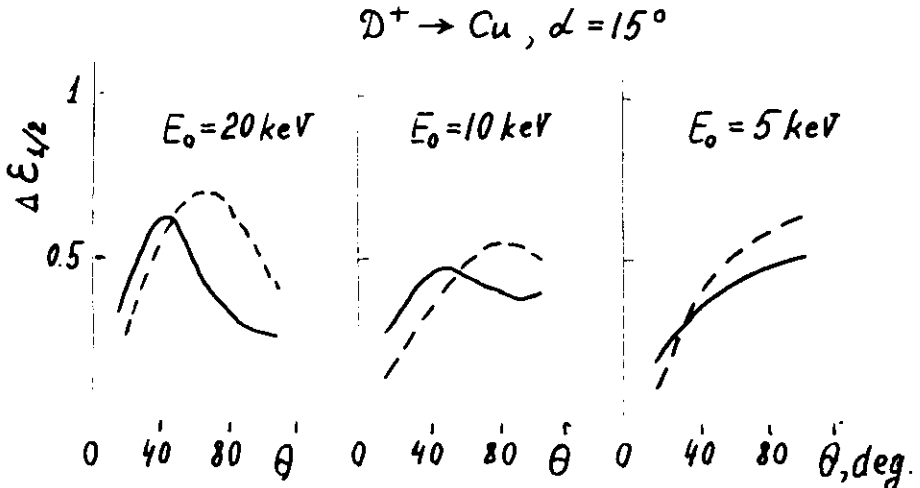

Fig. 5. The measured (solid lines) and calculated by formula (14) (dash lines) dependenses $\Delta E_{1/2}(\theta)$.

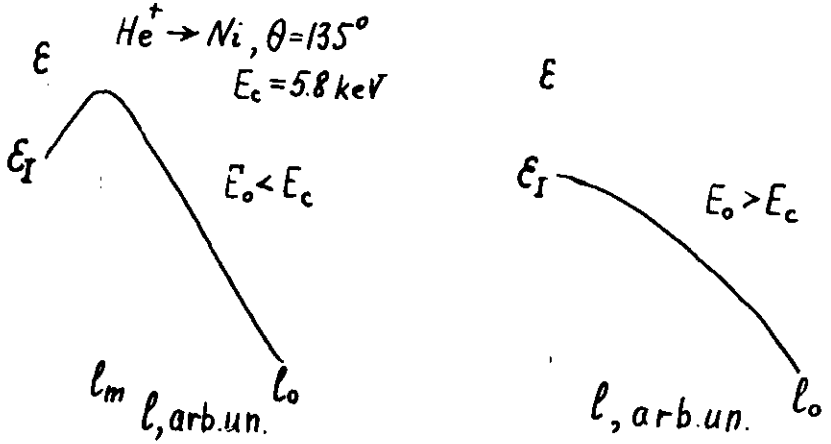

Fig. 6. The relative energy of backscattered particle versus the pass length in the solid.

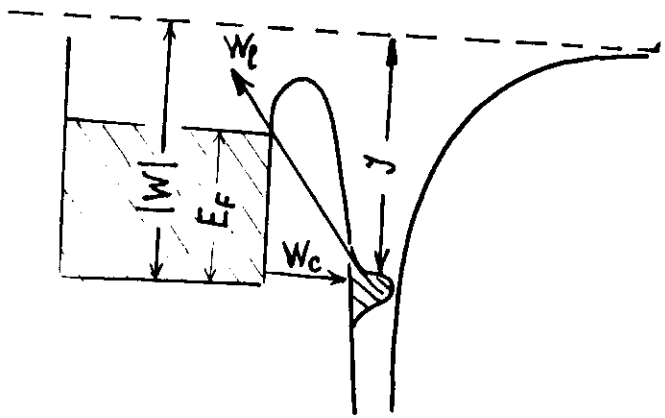

Fig. 7. The scheme of charge state formation processes.

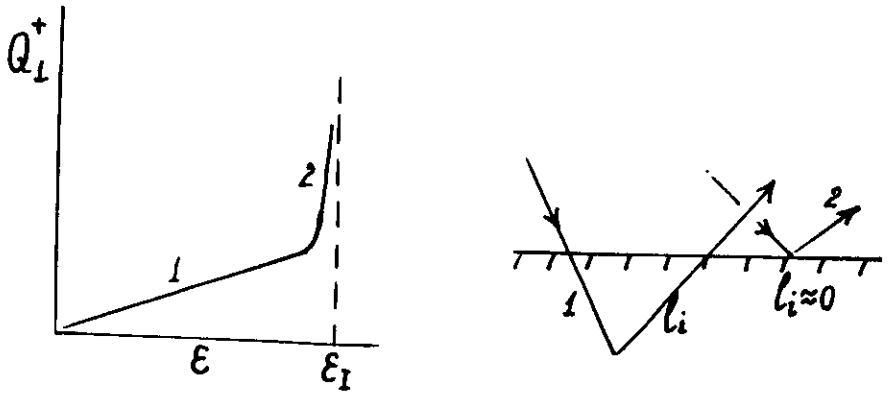

Fig. 8. The ionization probability versus energy ε; by numbers upon the curve the type of trajectory, giving the main contribution in this region of spectra is denoted. These typical trajectory are shown on the right of the figure.