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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 II)

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## Part II

# Energy spectra and angle distribution of ions, backsattered by singlecristal surface with submonolayer coverage

by

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## I Introduction

In this lecture I want to discuss the method, which we preposed for diagnostic of atomic adsorption states forming submonolayer coverage onto the surface [1]

This method based on analisis backscattered ion measured energy and angle distribution gave the possibility to found the adatom location relatively to the surface atoms and some others adsorbtion caracteristics

Let us consider the linear chain on the singlecristall surface Here,(Fig.1):

$M$ - is the mass of surface atoms
$d$ - is the interatomic distance in the chain
$m_1$ - is the mass of incident ion
$E_0$ - is the initial energy of incident ion beam
$\theta_1$ - is the polar angle of the first scattering
$\theta$ - is the polar angle of the second scattering
$m_2$ - is the mass of adatom
$a$ - is the distance between adatom and it's nearist neighbour surface atom.
$x$ and $y$ are it's two coordinates, $\Psi_0 = \text{arctg}\, y/x$
$\Psi$ is the total escaping angle, counting off the surface

Then, we assume the coincidence between ion beam axis and wide singlecristall axis direction. So, the incident ion either backscattered by surface atom or fall into the matter In additional, we assume the next unequality for the masses:

$$M > m_1 > m_2 \qquad (1)$$

The first part of (1) is necessary for existance of the ion single backscattering by surface atom. The second part of (1) means, that ion single backscattering by adatom is impossible. The thing is that the ion-adatom single backscattering peaks don't contein any information about adsorbate atom location, because their position in a spectrum depends only on the mass ratio. Moreover, as it will be clear later, this singlescattering peak position in the spectra is close to that one, which gives the necessary diagnostic information So, singlescattering peaks „ion-adatom" must be excluded for a best resolution the diagnostic peaks in the spectra.

The real diagnostic information about adsorbate atoms location relatively the surface atoms one can obtain by the double scattering peaks in the spectra. This double scattering conteins the first scattering „ion-surface atom" and the second scattering - „ion-adatom". The double typical trajectory is shown in Fig 1. Here p-is the impact parameter corresponding to $\theta$ scattering angle in the laborabory system. In our case $\nu = m_1/m_2 > 1$ and angle $\theta$ can't be greater then $\theta_0$ [1], where $\theta_0$ is limiting scattering angle:

$$\theta_0 = \arcsin \nu^{-1} \qquad (2)$$

E in the Fig1 is the final energy of double scattered ion. The final escaping angle is:

$$\Psi = \Psi_0 + \arcsin p/a + \theta \qquad (3)$$

The E and $\Psi$ quantites are the main measured parametres.

## II The analitical estimations.

For qualitive investigation the properities of this double scattering spectra we shall do some simple estimation: Really, it is clear that the first scattering on considered trajectory is violent enough as $\theta_1 \geq \pi$ and for $E_0 \ll keV$ $p \ll a$. So, one can assume the surface atom as a new point-like divergent and nearly isotropical beam sourse. In such approximation the double scattering cross-section reduces to a single scattering one, but calculated for nonparallel, divergent incident beam [3]. This single scattering cross-section reduces to:

$$\sigma(\theta, a) = \left\{\left[p^{-1} \cdot \sin\theta \cdot (1 - p^2/a^2)^{1/2} + a^{-1}\cos\theta\right]\left[a^{-1} + (1 - p^2/a^2)^{1/2}\frac{d\theta}{dp}\right]\right\}^{-1} \cdot 2\pi \qquad (4)$$

It should be noted, that if a in eq.(4) tends to infinity the $\sigma$ tends to the wellknown cross-section for parallel beam:

$$\lim_{a \to \infty} \sigma(\theta, a) = 2\pi \frac{p}{\sin\theta} \frac{dp}{d\theta} \qquad (5)$$

As the intensity of backscattered particles is always proportional to cross-section $\sigma$ the sharp peaks in the spectra appear, as it is seen from eq.(4), when $\sigma \to \infty$, i.e:

$$\frac{dp}{d\theta} = -\sqrt{a^2 - p^2} \qquad (6)$$

Eq (6) defines the main condition for existance of the singular, rainbow type peak in a spectra conteining desired diagnostic information.

For solution of the eq.(6) it is necessary to know the real form of function $p(\theta)$ determined by potential function $U(r)$ selection. In KeV energy range the Thomas-Fermi-Firsov potential is most appropriable [3]. For the analitical estimations it's inverse square approximation may be used:

$$U(r) = A/r^2 \qquad (7)$$

Then, the impact parameter is:

$$p = R_0 (\pi - \chi)(2\pi\chi - \chi^2)^{-1} \qquad (8)$$

where $\chi = \chi(\theta)$ is the scattering angle in the center mass system, $R_0 = (A/E_0)^{1/2}$ - is the minimum value for the distance of the clousets approach. The angles $\chi$ and $\theta$ are connected by expression:

$$\chi = \theta + (-1)^n \arcsin(\nu \sin\theta) \qquad (9)$$

In the studied case $\nu > 1$ one value $\theta$ corresponds to two values $\chi$:

$$\begin{cases} \chi_1 = \theta + \arcsin(\nu \sin\theta) \\ \chi_2 = \pi + \theta - \arcsin(\nu \sin\theta) \end{cases} \qquad (10)$$

Then $p(\theta)$ function becomes two valuedness, Fig 2. The disered solution - $\theta^*$ angle one can find on the upper branch of this curve only. By substitution the eq (10) into the eq (6) we obtein the next transident equation:

$$\pi (2\pi\chi_1 - \chi_1^2)^{3/2} \left[1 + \nu \cos\theta \left(1 - (\nu \sin\theta)^2\right)^{-1}\right] = \frac{\sqrt{a^2 - p^2}}{R_0} \qquad (11)$$

Let us denote by $f_1(\nu,\theta)$ the left part of eq (11) and by $f_2(E_0,\theta)$ it's right part. The graphical solution of transiden eq. (11) is shown in the Fig. 3. From this Fig. one can see, that eq (11) has the real roots only for the initial energy greater then some critical energy $E_t$. If $\nu \gtrsim 1$, the simple formula for $E_t$ is obtained.

$$E_t = 4A\pi^{-2}a^{-2}(\nu_1+1)^2\left[\nu_1\cos\theta_1 + (1-\nu_1^2\sin^2\theta_1)^{-1/2}\right]^{-2} \qquad (12)$$

Here $\nu_1 = m_1/M$, $\theta_1 = \psi + \frac{\pi}{2} - \theta^*$. In the other cases the $E_t$ values can be found only by numerical calculation, which have shown the following:

1) The values of $E_t$ are strongly dependented on $\nu_1$, $\nu$ and $a$ values. Then, for instance, in the case $x = d/2$, $\psi_0 = 45°$ for O- adatom upon Au surface chein [100] $E_t = 50$ eV for $Ne^+$ ions bombardement, $E_t = 2.6$ keV for $Ar^+$ and $E_t \approx 700$ keV for $Xe^+$.
2) If $E_0 \gg E_t$, the $\theta^*$ value is very close to the limiting angle $\theta_0$. Then, for some practical astimation we can assume that $\theta^* \approx \theta_0$. So, $\theta^*$ is approximately independent on $U(r)$ function form.

There are two possibilities for X and Y coordinats estimation. The first of them based on the measurements of greatist value $\psi_1^*$ determined the singular peak position in angle distribution and the $E_t$ value. By $E_t$ one can obtain a-value and $\psi_1^*$ gives the value of $\psi_0$ angle by: $\psi_0 = \psi_1^* - \theta_0 - \operatorname{arctg} P(\theta_0)/a$. Then

$$\begin{cases} X = a\cos\psi_0 \\ y = a\sin\psi_0 \end{cases} \qquad (13)$$

It is not clear at present if real possibility for $E_t$ measurements with necessary accurance exists or not.

The othe way for X and Y determination is based on two singular angls measurements $\psi_1^*$ and $\psi_2^*$. The $\psi_2^*$ angle has the same sence as $\psi_1^*$, but for case of the scattering by the next neighbour atom in the chain. Than:

$$\begin{cases} X = \dfrac{d\cos(\psi_1^*-\theta_0)\sin(\psi_2^*-\theta_0) + p\cos(\psi_2^*-\theta_0) - p\cos(\psi_1^*-\theta_0)}{\cos(\psi_1^* - \psi_2^*)} \\ y = \dfrac{d\sin(\psi_1^*-\theta_0)\sin(\psi_2^*-\theta_0) + p\sin(\psi_2^*-\theta_0) - p\sin(\psi_1^*-\theta_0)}{\cos(\psi_1^* - \psi_2^*)} \\ p \equiv p(\theta_0) \end{cases} \qquad (14)$$

## III The computer simulation

These all analitical estimations have shown the singular peaks existance only in principle. It is necessary to understand how this peaks may be separated on the background of other features in the spectra. For this aim the computer simulation have been performed.

We consider the results of calculation of the energy spectra and the angle distribution of $Ar^+$ and $K^+$ ions backscattered by Au (100) singlecristall surface with submonolayer coverage by O-atoms. This adsorption coverage has been chosen to be less, than half of the monolayer. The oxygen atom has been located in the middle between Au [100] chain atoms. The calculation has been performed by the chain model with Vaida potential function:

$$U(r) = \frac{A_1}{r^2} + \frac{A_2}{r} + A_3 \qquad (15)$$

where $A_1$, $A_2$, $A_3$ are the impirical constants.

The $Ar^+$ and $K^+$ have been taken as incident ions to investigate the charge state formation prousses influence on these spectra form. The thing is that Ar and K atoms with equal masses and therefore the same scattering lows, neutralized in quate different ways. This strong differense is caused by different location of it's atomic energy levels relatively to the Fermi level in metals. The K energy level lies higher then Fermi one, then the resonance ionization processes are much probable. As the result the ionization degree $\eta^+$ for potassium is about 100% and $\eta^+$ is a weakly dependent function both on final energy $E$ and real form of scattering trajectory. The calculated reflection coefficient for K coinsides practically with it's ion spectra, Fig. 4.

In contrast, the Ar energy level lies lower then Fermi one. So, the tunneling and Auger neutralisation processes are much probable. In this case $\eta^+$ at $E_0 \approx$ keV is about several per cents and $\eta^+$ is strongly dependent function both on $E$ and the type of trajectory.

Therefore, the $Ar^+$ ions spectra calculation can not be performed without the knowledge of $\eta^+$ function for each scattering trajectory.

## 1. The charge state of particles, scattered by single-cristal surface

Let us consider the model for calculation of the ionization degree for particles scattered by single crystall serface. We have proposed this model some years ago.

The charge state formation at scattering by polycrystals have been explained by the model of competiting electron loss and capture processes. In case of single crystal it is convenient to introduce electron capture and loss rates $W_c(R)$ and $W_l(R)$ in interaction with a single surface atom.

The differential equation describing the ionization degree after scattering on i-th atom $\eta_i^+$ is given by:

$$\frac{d\eta^+}{dt} = -\eta_i^+ W_c^i(R) + [1-\eta_i^+(R)]\,W_l^i(R) \qquad (16)$$

The solution of eq. (16) is as follows:

$$\eta_i^+(R_{0,i}) = \exp\left[-\frac{2}{v_i}\cdot\int_{R_{0,i}}^{\infty}\frac{(W_c^i+W_l^i)\,R\,dR}{\sqrt{R^2-R_{0,i}^2}}\right]\times\left\{\eta_{i-1}^+ + \frac{2}{v_i}\cdot\int_{R_{0,i}}^{\infty} W_l^i \times\right.$$

$$\left.\times\exp\left[\int_{R_{0,i}}^{R}\frac{(W_c^i+W_l^i)\,R_1\,dR_1}{\sqrt{R_1^2-R_{0,i}^2}}\right]\cdot\frac{R\,dR}{\sqrt{R^2-R_{0,i}^2}}\right\} \qquad (17)$$

Here $v_i$ is the velosity of the particle, scattered by i-th atom; $R_{0,i}$ is the distance of closest approach to it, $\eta_{i-1}^+$ is the ionisation degree after interaction with (i-1) th atom along the trajectory.

The final ionization degree for a particle escaping the surface after scattering along the given trajectory is calculated by consequent application of formula (17). It is important, that $\eta^+$ calculated by these manner depends not only E and $\varphi$, but also on the given set of values $\{R_{0,i}\}$ which is different for different trajectories.

Thus, $\eta^+$ will display some orientational effects. Such orientational dependenses we have calculated in our early paper.

But the real calculation by eq. (17) are too complicate even for computer simulations. For simplicity we separate

all collisions along the trajectory on neutralizing and ionizing ones in the following way:

1) The collision is considered to be a neutralizing one if $E_{in}(R_{0,i}) < I_0 - e\varphi$, where $E_{in}(R_{0,i})$ - is inelastic energy transfer per projectile atom. The value of $E_{in}$ may be estimated by Firsov formula:

$$E_{in}(R_{0,i}) = \frac{n_1}{n_1+n_2}\,\frac{b\,v_i}{(1+a_1 R_{0,i})^3} \qquad (18)$$

Here:

$b = 0.3\,Z_2\,(\sqrt{Z_1}+\sqrt{Z_2})\,(Z_1^{1/6}+Z_2^{1/6})$

$a_1 = 0.76\,\sqrt{Z_2}\,a_0^{-1}\,(Z_1^{1/6}+Z_2^{1/6})^{-1}$, $a_0$ - is the Bohr radius

$Z_1, n_1$ - is atomic number and the number of outershell electrons for projectile atom

$Z_2, n_2$ - is the same numbers for surface atom or adsorbate one

$I_0$ - is the projectile ionization potential

$e\varphi$ - is the surface work function.

For the neutralizing collisions $W_c \gg W_l$ is considered. Thus, taking $W_l = 0$ in eq. (17), we obtain:

$$\eta_i^+(R_{0,i}) = \eta_{i-1}^+\cdot P_i(R_{0,i}) \qquad (19)$$

where

$$P(R_{0,i}) = \exp\left[-\frac{2}{v_i}\int_{R_{0,i}}^{\infty}\frac{W_c\,R\,dR}{\sqrt{R^2-R_{0,i}^2}}\right] = \exp[-f_c(R_{0,i})] \; -$$

is the probability that the neutralization on the i th atom has not been occured.

If the ground energy level of backscattered particle lies below the bottom of conduction band, $W_c(R)$ is the Auger neutralization rate. This interatomic neutralization rates have been obtained by Kishinevsky and Parilis in form:

$$W_c = \begin{cases} B, & \text{for } R \le R_a \\ B, \exp[-\beta(R-R_a)], & \text{for } R > R_a \end{cases} \qquad (20)$$

Here $B$ is the Auger neutralization rate in quasimolecula formed by scattering ion and surface or adsorbate atom.

$$B = b(n_2-1)\cdot n_2 \sim b\cdot n_2^2 \qquad (21)$$

where $b \lesssim 10^{14}s^{-1}$ - is the value of neutralization rate for one pair of atomic electrons and $n_2(n_2-1)$ is the total number of such electron pairs in atomic outer shell. $R_a$ is approximately the sum of radii of colliding atoms and $\beta \approx 1\div 2\,Å^{-1}$ is given by the low of assimptotic electron density decreasing. So, the the Auger transition rates may be found for each collision

Then, by substitution eq. (20) to eq. (19) we obtain:

$$f_i(R_{0,i}) = \frac{2B_i}{v_i}\begin{cases}\sqrt{R_a^2-R_{0,i}^2} + \displaystyle\int_{R_a}^{\infty}\frac{\exp[-\beta(R-R_a)]R\,dR}{\sqrt{R^2-R_{0,i}^2}}, & \text{for } R_{0,i}\le R_a \\ \exp(\beta R_a)\,R_{0,i}\,K_1(\beta R_{0,i}), & \text{for } R_{0,i}>R_a\end{cases} \qquad (22)$$

Here $K_1$ - is McDonald function.

So in general case there are few violent ionization collisions and some neutralization ones on the given trajectory of scattering ion, where $N$ - is the total number of collisions. Let us consider that in any ionization collision the ionization probability $Q_i^+=1$ and during any neutralization the nonneutralisation probability $P_i$ is given by eq. (19). The following simple cases are possible:

1) In low energy region $E_0 < 5$ keV and for small $\theta_i$ a violent collision may not occur anywhere on the trajectory, then:

$$\eta_1^+ = \eta_0^+ \exp\left[-\sum_{i=1}^{N} f_i(R_{0,i})\right] \qquad (23)$$

$\eta_0^+$ - is the ionization degree of incident beam.

2) In anothe case on the atoms with numbers $k, l, \ldots f$ the ionization occurs. The final ionization degree is independent both on $\eta_0^+$ and the history before the last violent collision in this case. Then:

$$\eta_2^+ = \exp\left[-\sum_{i=f+1}^{N} f_i(R_{0,i})\right] \qquad (24)$$

After these remarks, we can discuss the results of computer simulation of $Ar^+$ and $K^+$ backscattered ions spectra.

## The energy spectra and the angle distributions

In the Fig. 5. the energy spectra for total number of backscatted particles is shown. The spectra contein two well separated group of peaks. The hight energy group consist of single and double scattering peaks related to incident ion scattering on Au atoms only. The lowenergy group consist of the peaks of quasisingle and qusidouble scattering along the trajectories where oxigen atom is the one of scattering center, because during $K^+$ or $Ar^+$ collision with O-atom the great elastic energy loss take place. The intensity of low energy peaks sharp increase for these trajectories where the last violent scattering angle $\theta \approx \theta_0$. So, these peaks may be easy separate in the angle distribution. They correspond to singular peaks wich was in question for previosely analitical estimation.

In the next figure (Fig. 6) the angle distribution for intensities of low energy $K^+$ and $Ar^+$ scattering ions is shown. The calculation was performed for some values of height $y$. In the Fig. 6 the result for $y = 1.1$ Å is shown.

The $K^+$ distribution conteins four sharp peaks. The corresponding trajectories shown upper the Fig. 6. The $Ar^+$ distribution calculated with the charge state formation accounting at exactly the same conditions conteins only two peaks with the numbers II and IV. The first and the second peak disappear due to strong neutralization.

So, the singular peaks III and IV are the best separated ones in all cases. It's positions in angle distribution show the high sensitivity to the adsorbate atom coordinates. Our computer simulation show in additional:

1) If the accuraney of this peaks position measurements is not poorer then 1°, one can obtein the adatom coordinates with accuraney about 0.1 Å.

2) Let us again to see on the (14) formulae for adatom coordinates calculation. The computer simulation show that practically in all cases $\rho(\theta_0) \ll d$. Then, with the enough accuraney:

$$\begin{cases} x \approx d\cos(\psi_1^*-\theta_0)\sin(\psi_2^*-\theta_0)\sec(\psi_1^*-\psi_2^*) \\ y \approx d\sin(\psi_1^*-\theta_0)\sin(\psi_2^*-\theta_0)\sec(\psi_1^*-\psi_2^*) \end{cases} \qquad (25)$$

Here $\Psi_1^*$ and $\Psi_2^*$ is the measured peaks III and IV position in angle distribution, $\theta_0 = \arcsin m_2/m_1$.

It is very importent, that (25) formulae don't conteins any terms connected with the scattering dinamics, but only the simple cinematic's relations. So, the experimental data treatments may be done by very simple way, don't needed in computer simulation for all cases, as it take place in the others ion-scattering methods for surface diagnostic.

As the result, the two highest intensitive low energy peak position in angle distribution of scattered ions gives the real possibity to obtein adatom coordinates with great accurancy.

## IV Any additional results.

Let us consider an energy analizer rotating in the plane passing through the beam direction and serface normal. By the intensity measurements in choosing energy range at the different analizer position one can found the direction where singular peak is observed and then to fix the analizer in this direction. It is interesting to understand: what additional information about adsorption states one can obtein by such measurements?

1) It is well known, that the atom onto the surface may be located in limited numbers of adsorbtion states. This states may be distinguish by the values of coupling energy or, from the other side, by adatom location relatively to the surface atoms. Such conditions on the surface may be always choosen, when only one adstate is effective. Then, by vareing the surface conditions (the temperature for example) one can achive the precense of the second adstate. In this case of adsorbtion an additional singular peaks will be observe at any other directions. By the singular peak intensities measurement at this two directions one can investigate the migration process result in adatom transition from the first to the second adstate. For instance, one can measure the rate of this migration as the the temperature function.

2) Let us return again to the one fixed adsorption state. During ion beam bombardement, the number of adsorbate

particles $N(t)$ changes with the time $t$ by the low:

$$\frac{dN}{dt} = - N \sigma_{isd}\, j^+/e \qquad (26)$$

Here $j^+$ is the density of incident ion beam, $\sigma_{isd}$ is the ion-stimulated desorbtion cross-section. Let us the initial number of adatoms denote by $N(t=0) = N_0$, where $N_0$ is given in the part of monolayer. Then

$$\frac{N(\tau)}{N_0} = \exp\left[-j^+/e\,\sigma_{isd}\cdot\tau\right] = \frac{J(\tau)}{J(0)} \qquad (27),$$

where $\tau$ is any choosen time moment, $J(\tau)$ is the singular peak intensity at $t=\tau$ and $J_0$ is it's intensity at $t=0$. From (27) it is easy to obtain the $\sigma_{isd}$ for the given adstate.

3) Let us consider the case, when besides ion beam the flux of slowly adatoms full on the surface. In this case (Fig 7):

$$\frac{dN}{dt} = - N\sigma_{isd}\frac{j^+}{e} + \left(1 - N(t)\right)P_1\cdot q_0\cdot\sigma_0 \qquad (28)$$

Here $q_0$ - is atomic flux density, $P_1$ - is the stikking coefficient, $\sigma_0$ - is the area per adatom on the surface. During any time $N_e$ the equilibrum coveredge would be establish and $N(t) \to N_e$. The singular peak intensity will be reach the equilibrum value too, $J(t) \to J_e$. Then

$$N_e = N_0\frac{J_e}{J_0} = \frac{P_1 q_0 e}{\sigma_{isd}\, j^+ + P_1\cdot q_0\cdot e} \qquad (29)$$

If $\sigma_{isd}$ have been measured early, then by $J_e$ and $J_0$ measurements one can to found the $P_1$ - stikking coefficient for given adsorption state.

4) Let in the upper discussed case the additional desorbtion factor take place, for example the temperature or electron beam and $W_d$ is this desorbtion rate. Then early reached equilibrum would be brake up and any new equilibrium coveredge would be established. This transition may be described by the equation:

$$\frac{dN}{dt} = - N\left(\sigma_{isd}\cdot j^+/e + W_d\right) + (1-N)P_1 q_0\sigma_0 \qquad (30)$$

Then

$$\frac{J_e}{J_e'} = 1 + \frac{W_d\, e}{\sigma_{isd}\cdot j^+ + P_1\varphi_0 e\sigma_0} \qquad (31),$$

where $J_e'$ - is the new equilibrum value of singular peak intensity. From (31) the $W_d$-desorbtion rate may be found.

If additional desorbtion process is the electron stimulated desorbtion, then $W_d = \sigma_{esd}\, j^-/e$, $j^-$ - is the electron beam density. In this case:

$$\sigma_{esd} = \frac{j^+}{j^-}\,\frac{J_e - J_e'}{J_e'}\left(\sigma_{isd} + P\varphi_0 e\sigma_0\right) \qquad (32)$$

It should be emphasis, that $W_d$ and $\sigma_{esd}$ obtained by this way don't depended on charge state of desorbate particles, so $\sigma_{esd}$ is total cross-section. It is well known, that $\sigma_{esd}$ measurements for neutrals is very difficult for experiment. We hope, that this method may be useful for found $\sigma_{esd}$ value for neutrals, because the neutrals are the main part of desorbed particles.

So, the diagnostic, based on the measurements of singular peaks position in angle distribution of backscattered ions, may give varied information about the adsorbtion states.

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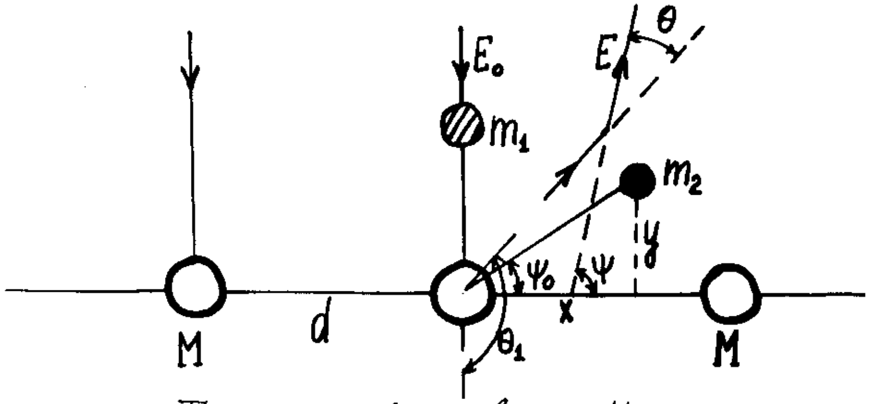
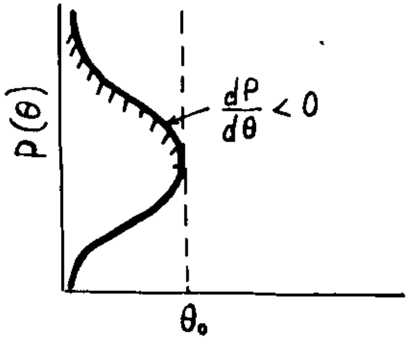


Fig. 1. The scheme of ion backscattering by singlecristal surface with submonolayer coverage.



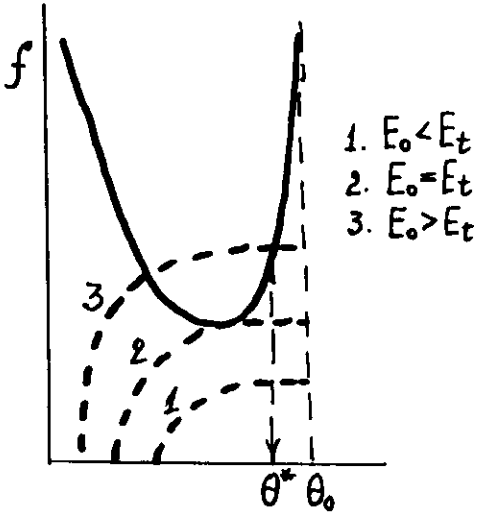


Fig. 2. The impact parameter versur the scattering.

Fig. 3. The graphical solution of the angle equation

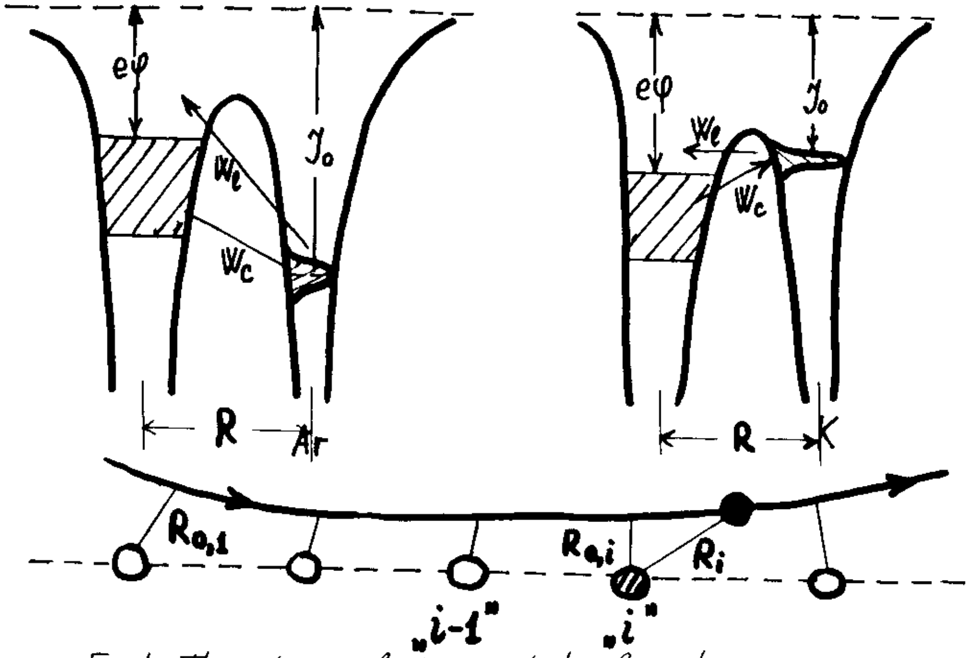
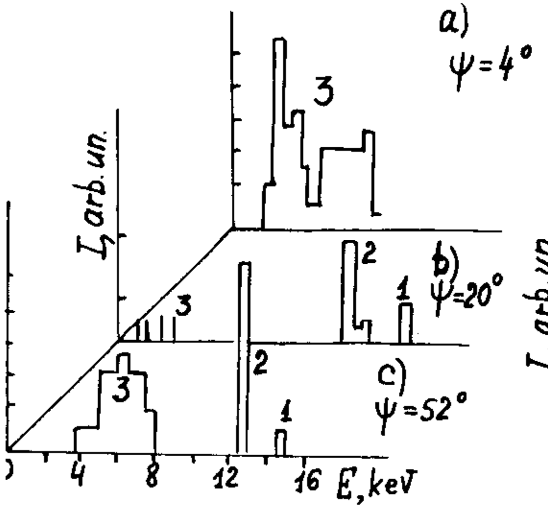


Fig. 4. The scheme of charge state formation processes for the particle scattered by singlecristal surface.



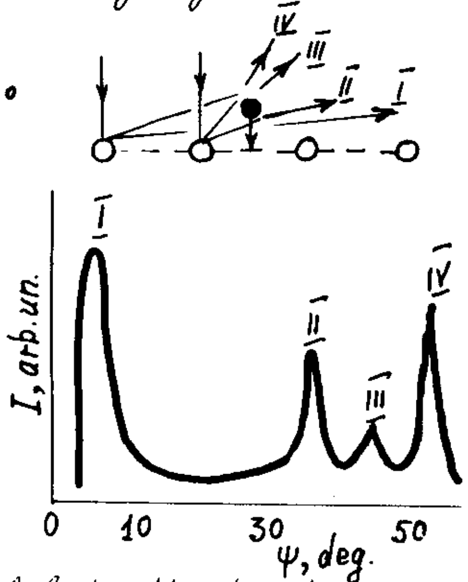


Fig. 5. The energy spectra of backscattered particles

Fig. 6. The angle distibution of backscattered ions.

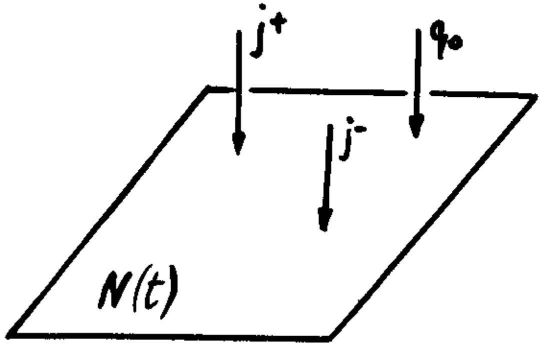


Fig. 7. The sheme of surface bombardement by ions, neutral and electron beams.