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## LOW-ENERGY SPECTRUM OF ELECTRONS EMITTED AT IRRADIATION OF Au BY $\alpha$ -PARTICLES OF $^{238}\text{Pu}$

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By the timing ( $\alpha$ e)-coincidence method, we study the low-energy spectrum of electrons arising from the bombardment of an Au target by  $\alpha$ -particles of the  $^{238}\text{Pu}$  source. The ionization of atoms by charged particles is considered as the shake off of electrons into the continuous spectra at their sudden perturbation by passing particles. The experimental energy distribution of emitted electrons well agrees with the theoretical one, which confirms the validity of the consideration of the ionization as a result of the shake off process.

### 1. Introduction

If a particle with charge  $Ze$  flies with velocity  $V$  near an electron of the atom that is positioned on the surface of a target, then this particle transfers the energy  $\frac{Ze^2}{r}$  to the electron at the time moment, when the distance  $r$  between them is minimal. If the energy is transferred rapidly, almost instantly, then this electron has a probability to transit from a bound state to the continuous spectrum, by leaving the atom in the ionized state. The suddenness means that the time-of-flight of a particle through the atom,  $\tau \cong \frac{r}{V}$ , is much less than the duration of the transition of the atom from the neutral state  $i$  to an excited one  $f$ , so that  $\tau \ll 2\pi\omega_{fi}^{-1}$ , where  $\omega_{fi}$  is the transition frequency. Such a process can be considered as the shake effect. If the conditions of suddenness do not hold, or the shake did not happen due to the probabilistic character of this process, then the electron returns this energy to the particle backward at the termination of the perturbation, and the atom remains in the ground state.

It is known from quantum mechanics that if the perturbation of a system depends explicitly on time and oc-

curs suddenly, then the system can be forced to transit from the initial ground state to an excited ionized (with a hole) state, which is accompanied by the shake off of an electron into the continuous spectrum. In this case, the wave function of the initial state  $\psi_i^{(0)}(q)$  “has no time” to vary for this time [1, 2], and all other electrons remain on their places. In the case where a charged particle flies through an atom, the entire perturbation energy,  $\frac{Ze^2}{r}$ , which is transferred to the electron, is completely spent on overcoming the binding energy  $\varphi$  and on the acquisition of the kinetic energy  $E = \frac{Ze^2}{r} - \varphi$ . Hereafter,  $\varphi$  includes, in addition to the binding energy, the work function (we note that all relations will be written with the use of the absolute values of energy).

In most cases, the shake effect is considered under a sudden perturbation arising spontaneously in a system, which is in the state of rest before and after the perturbation. But under the bombardment of a target by a charged particle, the perturbation arises as a result of the short-range interaction of the  $\alpha$ -particle with an atomic electron, when they are maximally close to each other, and then disappears. In the first case, a change of the interaction Hamiltonian has the form  $H_0 \rightarrow H_0 + \Delta H$  and induces the shake with a perturbation of “the switch-on type”. In the second case, a change of the Hamiltonian is  $H_0 \rightarrow H_0 + \Delta H \rightarrow H_0$  and induces the shake with a perturbation of “the scattering type” [3, 4].

We note that, in our case, the electron in the final state of a system is in the continuous spectrum, and the interaction happens for a finite time interval  $\tau$ . We also assume that the probability of the excitation of an electron by a charged particle passing through an atom with radius  $r_a$  is the same over the cross-section of the atom. Then, by the theory of perturbations of the first

order for timing transitions [1, 2], the probability of the shake off of an electron into the continuous spectrum with its kinetic energy in the interval from 0 to  $E$  is given by the semiempiric formula [5, 6]

$$W(E) = \frac{c}{V} \left( \frac{Ze^2}{r_a} \right)^2 \left| \int \psi_i^{(0)} \psi_f^* dq \right|^2 bF(E). \quad (1)$$

Two first factors describe the probability of the collision of a particle with the shaken electron and the transfer of a perturbation energy to the electron. At  $V = c$ , where  $c$  is the light velocity in vacuum, the probability of the shake off is minimal. In the matrix element, the wave functions of the initial and final states arise from stationary states of the type  $\Psi = \psi(q)e^{-i\frac{E}{\hbar}t}$ , where the wave function  $\psi(q)$  depends only on the coordinates  $q$ ;  $\psi_i^{(0)}(q)$  describes the initial state of the system with the Hamiltonian  $H_0$ , whereas  $\psi_f^*(q)$  describes the final state with the Hamiltonian  $H_0 + \Delta H$ . In the ground or normal state of the system  $\psi_f$ , the kinetic energy of the electron is minimal,  $E=0$ , i.e.,  $\Psi_f = \psi_f(q)$ . The probability of the induced transition  $i \rightarrow f$  does not depend on the energy transferred to the shaken electron, if  $\frac{Ze^2}{r} > \frac{Ze^2}{r_{\max}} = \varphi$ , because, due to the small duration of a perturbation, all distances  $r < r_{\max}$  become undetermined by virtue of the uncertainty relation. The function  $F(E)$  is the integral distribution of electrons, which are shaken into the continuous spectrum with the kinetic energy in the interval from 0 to  $E$ , has probabilistic character, and looks as

$$F(E) = \left[ \frac{1}{\sqrt{\varphi}} \arctg \sqrt{\frac{E}{\varphi} - \frac{\sqrt{E}}{E + \varphi}} \right], \quad F(0) = 0. \quad (2)$$

This formula follows from the differential spectrum of shaken electrons, which is resulted, in turn, from the consideration of the phase volume of the continuous spectrum [5]. The density of levels of electrons of the continuous spectrum reads

$$\frac{dv}{dE} = b\sqrt{E}, \quad (3)$$

where  $b = \frac{\sqrt{2}m^{3/2}V_e}{\pi^2\hbar^3}$ ,  $m$  is the electron mass, and  $V_e$  is the volume occupied by one electron in the phase volume of the continuous spectrum. Then the differential distribution of electrons, which are shaken in vacuum, takes the form

$$\frac{dW}{dE} = D \frac{b\sqrt{E}}{(E + \varphi)^2},$$

where

$$D = \frac{c}{V} \left( \frac{Ze^2}{r_a} \right)^2 \left| \int \psi_i^{(0)} \psi_f^* dq \right|^2. \quad (4)$$

This relation is well supported by the measurements of the spectra of shaken electrons with near-zero energy. Integrating (4) over the energy from 0 to  $E$ , we arrive at formula (2).

The goal of the present work is to determine the energy distribution  $F(E)$  of electrons shaken into the continuous spectrum at the flight of  $\alpha$ -particles from  $^{238}\text{Pu}$  through a target made of gold. Earlier, we studied such a distribution for targets made of aluminum and copper [6, 7]. Those experiments revealed a good agreement with the theory. However, the atoms of those targets have comparatively small numbers of electrons in shells, whereas their number in Au is 79. By comparing the experimental distribution of electrons with the theoretical one  $F(E)$ , we will verify whether the ionization of Au atoms by  $\alpha$ -particles can be explained by the shake effect.

## 2. Experimental

We study the energy distribution of ionization electrons after the passage of  $\alpha$ -particles through the substance by the method of timing ( $\alpha$ )-coincidence with the supply of a retarding voltage  $U$  in the electron registration channel. We measured the coincidence-count rate  $N$  as a function of the voltage  $U$  retarding electrons. The time coincidence spectra were registered with a multichannel analyzer ORTEC-NORLAND.

The measurements of the energy spectrum of electrons were carried out in two geometries shown in Fig. 1. In the transmission experiment (Fig. 1,a),  $\alpha$ -particles, which are emitted by source  $S$ , pass through target  $T$ , which is a transparent layer of Au sprayed on a Dacron film, and are registered by detector  $\text{MCP}_1$  composed of two microchannel plates in the form of a chevron. In order to eliminate the influence of the electrons that are emitted from the target and fall on detector  $\text{MCP}_1$ , it was screened by an Al-foil 10  $\mu\text{m}$  in thickness. The electrons  $e$  that are emitted from the target at its bombardment by  $\alpha$ -particles are registered by another detector  $\text{MCP}_2$  of the same type. In the experiment on reflection (Fig. 1,b), source  $S$  and detector  $\text{MCP}_1$  of  $\alpha$ -particles change places, by turning by 180 degrees around the central point of the target. In the experiment on transmission, the electrons are emitted from the target along the direction of motion of  $\alpha$ -particles, whereas the electrons are moving oppositely to  $\alpha$ -particles in the experiment on reflection. In both cases, the electrons go out of the same target surface.

On the front surface of detector  $\text{MCP}_2$  of electrons, we fixed two grids 1 and 2. On internal grid 2, we supplied

retarding potential  $U$  which varied during the measurement. External grid  $1$  is grounded in order to conserve the effective solid angle, at which the electrons are registered under the variable retarding potential. The entire system was in a vacuum chamber at a pressure of  $5 \times 10^{-6}$  mmHg. As a source of  $\alpha$ -particles, we took  $^{238}\text{Pu}$  from the OSAI collection. The energy of  $\alpha$ -particles was 5.5 MeV. As a target, we used a transparent layer of Au sprayed on a Dacron film.

In Fig. 2, we presented fragments of the time spectra measured in the geometries on transmission and reflection at various values of retarding potential  $U$ . The upper curves are the timing ( $\alpha e$ )-coincidence spectra in the experiments on transmission, and the lower curves correspond to the experiments on reflection. As is seen, at  $U = 0$  V on grid 2, each of the distributions contains two peaks: on the left – the peak of electrons with a near-zero energy  $e_0$ , and, on the right – the peak related to fast electrons  $e_f$ . The first peak is a result of the interaction of surface electrons with the immovable charge, which arises suddenly near the target surface after the passage of an  $\alpha$ -particle through the surface (the effect of shake off of  $e_0$ -electrons from the surface [8]). The maximum of the energy distribution of  $e_0$ -electrons is about 1 eV. Its amplitude decreases rapidly, so that it can be neglected at energies more than 24 eV. The angular distribution of  $e_0$ -electrons is elongated along the normal to the target surface in both experiments on transmission and reflection. The description of the output of  $e_0$ -electrons emitted at the radioactive decay (as the shake off effect) is given in [8, 9, 10] in detail.

The appearance of the peak of fast electrons  $e_f$  is related to two processes, which are revealed differently in the experiments on transmission and reflection. In the experiment on transmission, an  $\alpha$ -particle moving through the target interacts with bound electrons of surface atoms, which causes the shake of electrons into the continuous spectrum. We denote these ionization electrons by  $e_f^i$ . Their angular distribution is directed forward along the motion direction of an  $\alpha$ -particle, i.e., in the direction of the normal to the target surface. Such electrons are not observed in the experiment on reflection, as we found earlier in work [9]. However, it is worth noting that the shake of electrons occurs in both cases (transmission and reflection) at the passage of an  $\alpha$ -particle through an atom located inside the target. In this case, the electron transits in a higher unfilled shell of the atom, by creating a vacancy on the previous place. While these vacancies are filled, we observe the output of fast Auger-electrons. We denote them by  $e_f^A$ . The directions of the emitted Auger-electrons do not depend

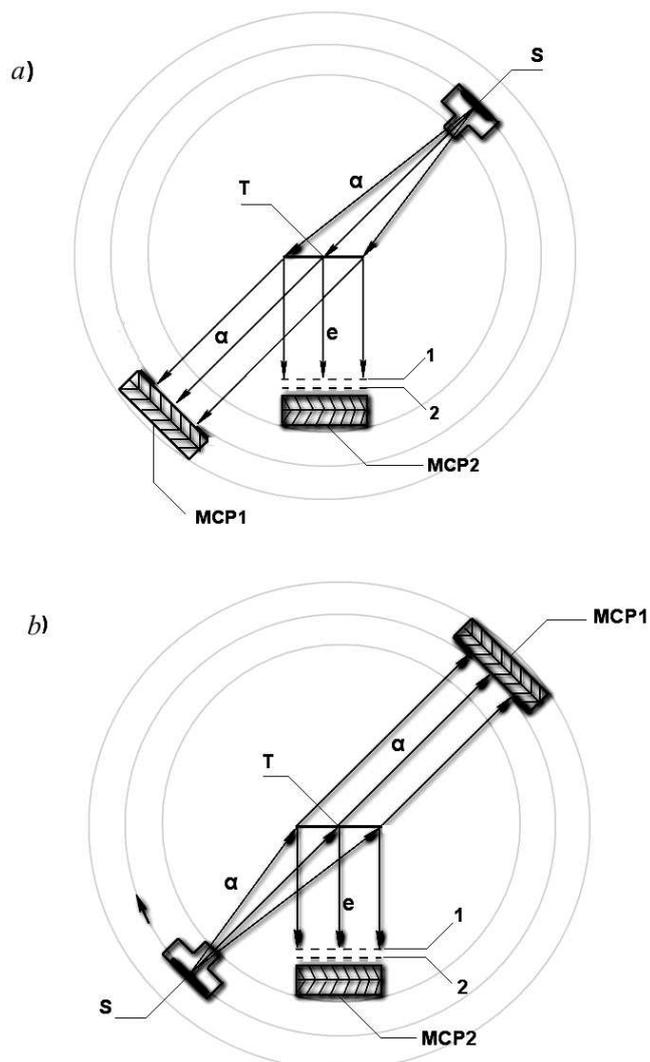


Fig. 1. Scheme of the experiment in the geometry on transmission (a) and on reflection (b).  $S$  –  $^{238}\text{Pu}$  source,  $T$  – target, MCP<sub>1</sub> – detector of  $\alpha$ -particles, MCP<sub>2</sub> – detector of electrons. External grid  $1$  is grounded; the internal grid  $2$  is under the retarding potential  $U$ . The distance from target  $T$  to source  $S$  and to MCP<sub>1</sub> is 40 mm and to MCP<sub>2</sub> is 25 mm

on the direction of motion of the  $\alpha$ -particle. Their distribution is isotropic [9].

Thus, the peaks of fast electrons  $e_f$  that are observed in the experiments on transmission and reflection differ essentially from each other by the nature. In the experiment on transmission, the appearance of the peak of fast electrons  $e_f$  is related to electrons  $e_f^i$  which appear at the ionization of atoms located on the surface and to Auger electrons  $e_f^A$ , so that  $e_f = e_f^i + e_f^A$ . In the experiment on reflection, only Auger

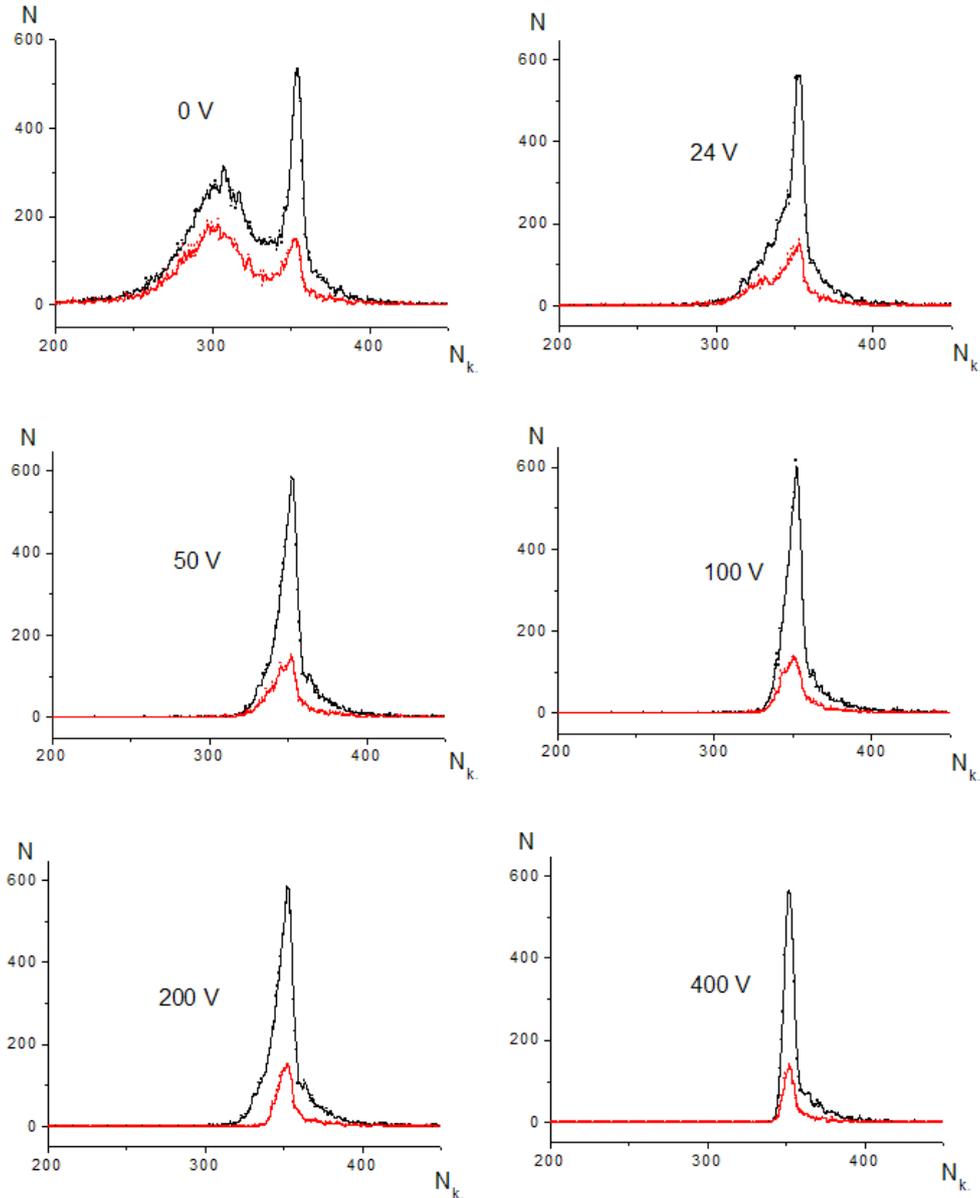


Fig. 2. Sections of the time spectra of  $(\alpha e)$ -coincidences measured in the geometries on transmission and reflection at various values of retarding potential  $U = 0, 24, 50, 100, 200,$  and  $400$  V

electrons contribute to the peak of fast electrons, i.e.,  $e_f = e_f^A$ . The number of vacancies created at the excitation of  $\alpha$ -particles in the near-surface layer of the target. Therefore, the numbers of electrons  $e_f^A$  appeared in the spectra after the filling of these vacancies are identical in the experiments on transmission and reflection. Hence, in order to determine the intensity of the peak of fast ionization electrons  $e_f^i$ , we must subtract the peak intensity of fast electrons in the spec-

trum on reflection from that in the spectrum on transmission.

It is seen from Fig. 2 that the intensity of the peak of fast electrons decreases gradually, as the retarding potential  $U$  increases, due to the cutting on the lower boundary of the integral spectrum. At  $U = 24$  V, the zero-energy peak practically disappears.

In the present work, we measured the time spectra of  $(e\alpha)$ -coincidences in the geometries on transmission and reflection in the interval 0–400 V at various values

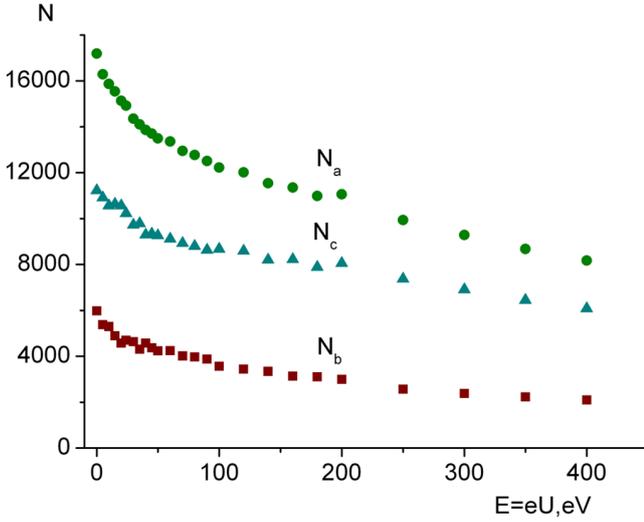


Fig. 3. Counting rate of timing  $(\alpha e)$ -coincidences versus the retarding potential at different geometries of the experiment:  $N_a$  – in the geometry on transmission;  $N_b$  – in the geometry on reflection;  $N_c$  – counting rate of  $(\alpha e)$ -coincidences only for fast electrons  $e_f^i$  arising at the ionization

of retarding potential  $U$  as follows: in 5 V from 0 to 50 V, in 10 V from 50 to 100 V, in 20 V from 100 to 200 V, and in 50 V from 200 to 400 V. Totally, we have 25 points. The duration of measurement at each point was 2 h. In this case, detector MCP<sub>1</sub> registered about  $7.2 \times 10^5$   $\alpha$ -particles.

### 3. Results of Measurements

In Fig. 3, we present the energy dependences of the counting rates in the experiments on transmission  $N_a(E)$  and on reflection  $N_b(E)$ . Preliminarily, we removed the distribution of electrons with near-zero energy from these dependences at energies  $E < 24$  eV. The difference of the counting rates in the experiments on transmission and on reflection determines the counting rate for  $(\alpha e)$ -coincidences  $N_c(E)$  only for fast electrons  $e_f^i$  that arise at the ionization of atoms and fly out in vacuum. Thus, we can obtain the integral energy spectrum  $N_f(E)$  in the interval from 0 to  $E$  for ionization electrons which are shaken into the continuous spectrum. This allows us to compare the obtained experimental distribution with the theoretical one based on the consideration of the process of ionization as the shake off effect:

$$N_f(E) = N_c(eU = 0) - N_c(eU = E) = \sum_k m_k A_k F_k(E).$$

The index  $k$  is used to denote different subshells of Au, from which electrons are shaken off,  $m_k$  is the number of

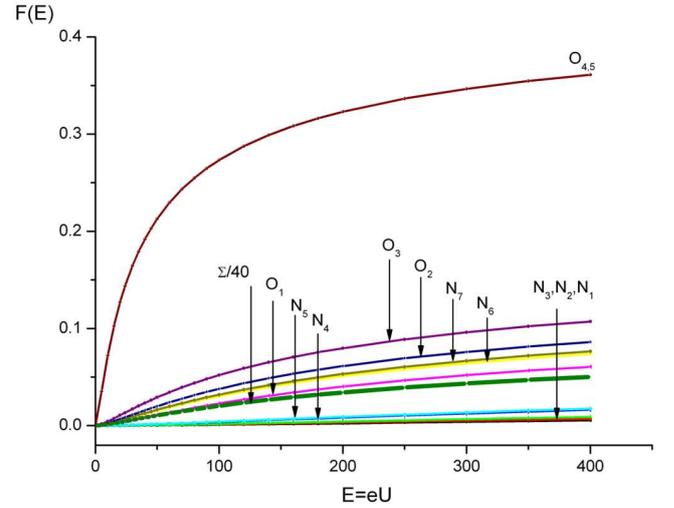


Fig. 4. Calculated integral energy distributions  $F_k(E)$  for electrons of various subshells of Au

electrons on the subshell, and  $A_k$  is a factor independent of the energy distribution of shaken electrons:

$$A_k = \eta \frac{c}{V} \left( \frac{Z_e^2}{r_a} \right)^2 \left| \int \psi_f^* \psi_i^{(0)} dq \right|^2 b N_\alpha.$$

Here,  $\eta$  depends on the conditions, under which the experiments are carried out, and is given as the product of the efficiency of registration of electrons  $\varepsilon$  on detector MCP<sub>2</sub>,  $\Omega_{\text{eff}}$ , and the transparency of the grids  $\delta$ , and  $N_\alpha$  is the number of registered  $\alpha$ -particles in the process of measurements at each point. The other factors were defined at the analysis of formula (1). The values of  $A_k$  differ from one another only by the values of matrix elements.

As was noted above, the numbers of shells and subshells are large for gold. But, for  $K$ -,  $L$ -, and  $M$ -shells possessing great binding energies, the values of  $F_k(E)$  in the region  $E \leq 400$  eV are small, as compared with their maximum value and with  $F_k(E)$  of the rest shells. In the time coincidence spectra (see Fig. 2), we can observe a prolongation of the drop in the line intensity on the right side, which is possibly related to the presence of electrons from those shells in the spectrum. Their contribution to the total line intensity does not exceed several per cent. Therefore, their influence on the energy distribution of shaken electrons in the region under consideration is else less, and they can be neglected. Other electrons of  $N$ - and  $O$ -subshells play also various roles in the energy distribution of shaken electrons. This is seen from Fig. 4, where we give the calculated values of  $F_k(E)$  for the binding energies of various subshells. The

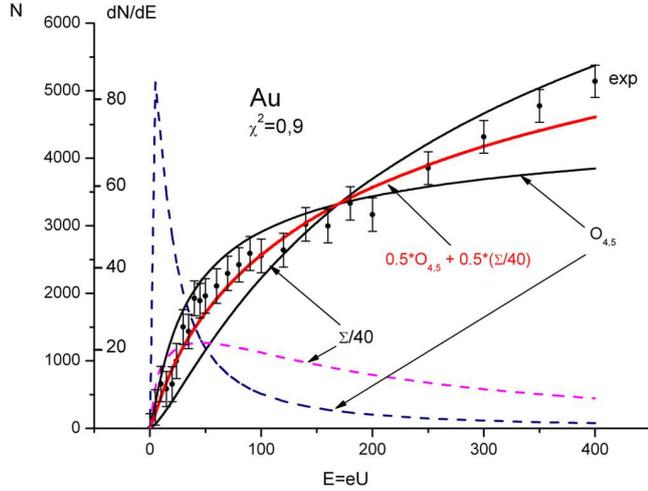


Fig. 5. Comparison of experimental values of  $N_f(E)$  with the distribution calculated for the electrons shaken only from  $O_{4,5}$  and  $\Sigma/40$  subshells. The calculated differential distributions of these electrons over energies are shown as well

distribution for  $O_{4,5}$ -subshells, whose binding energy is equal to 11.7 eV (in the calculation of  $F_k(E)$ , we used the known binding energies with regard for the surface ionization potential equal to 9.2 eV), demonstrates the significantly different behavior. Therefore,  $N_f(E)$  can be described as that composed mainly from only two distributions:  $10A_{O_{4,5}}F_{O_{4,5}}(E)$  and  $40B_{\Sigma/40}F_{\Sigma/40}(E)$ . Here, 10 and 40 are the numbers of electrons on subshells  $O_{4,5}$  and other shells of Au atom, respectively. On the bold curve (Fig. 4), the symbol  $\Sigma/40$  is referred to the resultant curve, which replaces all other curves  $F_k(E)$  related to other subshells under consideration. The distribution  $F_{\Sigma/40}(E) = \frac{1}{40} \sum_k m_k F_k(E)$ , and  $B_{\Sigma/40}$  corresponds to the averaged value of the square of the matrix element  $|M_{fi}|^2$  over this sum in the interval of energies of shaken electrons from 0 to 400 eV. Each of the values of  $A_{O_{4,5}}$  and  $B_{\Sigma/40}$  was determined by the fitting over all 25 points of the measurement by the least-squares method,  $\chi_{\min}^2$ . In Fig. 5, we show the calculated distributions and the experimental values of  $N_f(E)$  given by points на рисунку точками.

We represent the energy distribution of electrons as the sum of two terms (the term corresponding to  $O_{4,5}$ -subshell and the resulting contribution from all other shells):

$$N_f(E) = 10xA_{O_{4,5}}F_{O_{4,5}}(E) + 40(1-x)B_{\Sigma/40}F_{\Sigma/40}(E).$$

The value of  $x$  was also determined by the  $\chi_{\min}^2$  method:  $x = 0.5$ . In other words, a half of the electrons shaken off into vacuum at the passage of an  $\alpha$ -particle of  $^{238}\text{Pu}$

through the surface of Au appears from  $O_{4,5}$ -subshells. The result of the comparison of the theoretical and experimental distributions changes slightly, if we neglect the influence of the electrons present in  $N_{1-3}$ -subshells due to a small value of  $F_k(E)$  (i.e., if we use curve  $\Sigma/32$  instead of curve  $\Sigma/40$  shown in Fig. 4). They are close to each other, and the distribution of the electrons shaken off from  $O_{4,5}$ -subshells is practically invariable. The binding energy that is used for the resultant curve  $F_{\Sigma/40}(E)$  equals 140 eV, whereas it is 117 eV for  $F_{\Sigma/32}(E)$ .

As for the 79-th electron of  $P_1$ -shell of Au, its shake off from the target surface occurs similarly to electrons with near-zero energy. In other words, it is registered at the zero-energy peak, for which the distribution maximum is about 1 eV. So, it was excluded by us at the consideration of fast shaken electrons.

We gave earlier a good description of the energy distribution of ionization electrons for targets made of Al and Cu in the low-energy region, by basing on the representation of the process of ionization as the shake off effect [6, 7]. Here, such a description has been obtained also for the target made of Au, which has much more electrons on the subshells of atoms than those of Al and Cu.

#### 4. Conclusion

The good agreement of the integral experimental distribution of shaken electrons over energies with the calculated one  $F(E)$  indicates that the description of the differential distribution of electrons over energies, which is determined by formula (4) and is shown in Fig. 5 by dashed lines, should be correct as well irrespective of the type of a flying charged particle and the value of perturbation energy. The maximum of this energy distribution  $\frac{dN}{dE}$  should be positioned at  $E = \frac{1}{3}\varphi$ , and the half-width of the distribution should be close to  $1.9\varphi$ . In addition,  $F(E)$  depends only on the binding energy  $\varphi$  and tends to the saturation,  $F(E) \rightarrow \frac{\pi}{2\sqrt{\varphi}}$ , at high energies a sa function of  $\varphi$ .

The energy distribution of shaken electrons has the probabilistic character and, as the probability of the indexed transition  $i \rightarrow f$  (electron-hole), is independent of the transferred perturbation. It seems that the energy of shaken electrons itself is determined from the relation  $E = \frac{Ze^2}{r} - \varphi$ , where  $r < r_{\max}$ , and  $r_{\max}$  follows from the relation  $\frac{Ze^2}{r_{\max}} = \varphi$ . However, due to the Heisenberg uncertainty relation, the energy transfer to a shaken electron at the time moment of the interaction cannot be

determined exactly (the uncertainty of the transferred energy exceeds the energy itself by several times), because it is determined by the distance  $r$ , which becomes indeterminate at the time moment of the interaction. At the passage of a particle through the atom, all all distances  $r < r_{\max}$  become indeterminate, and the perturbation, which appears equiprobably in this case, causes the shake off. The dependence of the shake off on the electron energy appears only when the electron fills in one of the free states of the continuous spectrum, which is determined by the probability of this filling. As the final energy  $E$  of the electron becomes known, it becomes possible to evaluate the distance  $r$ , at which the particle flew near it. Otherwise, this will contradict the uncertainty relation, which follows from the effect of suddenness. Thus, the shake off effect at the ionization of an atom by a flying charged particle is one of the examples of the manifestation of the uncertainty relation in quantum mechanics.

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СПЕКТР ЕЛЕКТРОНІВ НИЗЬКОЕНЕРГЕТИЧНОЇ  
ОБЛАСТІ, ЩО ВИНИКАЄ ПРИ ОПРОМІНЕННІ  
Au  $\alpha$ -ЧАСТИНКАМИ  $^{238}\text{Pu}$

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Р е з ю м е

Методом часових ( $\alpha e$ )-збігів досліджено низькоенергетичний спектр електронів, що виникає внаслідок бомбардування атомів мішені Au  $\alpha$ -частинками  $^{238}\text{Pu}$ . Іонізація атомів при бомбардуванні їх зарядженими частинками розглядається як струс електронів у неперервний спектр при раптовому збуренні їх налітаючою частинкою. Порівняння експериментального розподілу вилітаючих електронів по енергії з розрахунковим показало гарне узгодження, що підтверджує правильність опису іонізації як результат ефекту струсу.