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ENERGY DISTRIBUTION OF ELECTRONS IN THE "ZERO-ENERGY PEAK" INDUCED BY A RADIOACTIVE DECAY OR A TARGET BOMBARDMENT WITH CHARGED PARTICLES

The energy distribution of near-zero electrons (e₀-electrons) emitting from the surface of radioactive sources or from the surface bombardment with α - or β -particles is studied. The integrated spectrum N(E) of e₀-electrons with the energy $E = (0 \div 24)$ eV is determined from the measurements of the delay curve by applying a retarding potential between the source (or the target) and the detector of e₀-electrons. The calculated distribution of e₀-electrons is shown to be in good agreement with the theoretical one obtained in the framework of the shakeoff model, i.e. when the perturbation by an electric charge arising near the surface and it shakes off weakly bound electrons from the surface.

Keywords: reflection, passing through, near zero electrons (e₀-electrons), shakeoff effect, microchannel plates (MCP)

1. Introduction

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While studying radiation that is emitted in a radioactive decay, a zero-energy peak is always observed in the electron spectrum, which reflects the distribution of electrons with the maximum intensity at the energy $E \leq 1$ eV and a halfwidth of 1–2 eV. The intensity of this distribution falls down so rapidly that it can be neglected at energies higher than 15–20 eV. Those electrons are called near-zero ones. We designate them as e₀-electrons, in contrast to fast electrons, which are designated as e_f-electrons.

For the first time, the zero-energy peak was found in work [1], when analyzing the low-energy part of the electron spectrum emitted by ²³⁹Pu. For its registration, an ordinary β -spectrometer with a Geiger– Müller counter was used, although it is efficient for the registration of only electrons with energies of tens kiloelectronvolts and higher. Therefore, for the spectrum to be registered, electrons were preliminarily accelerated to the energy E. It turned out that the peak is also located at the energy E, i.e. the own energy of the observed electrons was close to zero. The authors of work [1] explained the appearance of this peak by the field emission. However, in work [2], in addition to a β -spectrometer, an electrostatic analyzer was applied, which allowed the experimenters to exclude the emergence of the field emission. Nevertheless, the zero-energy peak was always present in all spectra of investigated radioactive sources. The authors called it "zero-energy peak" and explained its emergence as a result of the secondary Auger electron emission, frozen primary or secondary electrons generated in the source or substrate by α - or β -particles, or γ -quanta.

However, the most detailed analysis of the zeroenergy peak was carried out in work [3] with the help of a specially constructed electrostatic spectrometer with high resolution. The intensity and the shape of a spectral distribution were found to depend on the temperature, thermal treatment, vacuum, and other factors. A conclusion was drawn that the origin of

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the zero-energy peak is associated with the state of the radioactive source surface. It was suggested to call this phenomenon as the radiation-induced electron emission (RIEE) and to determine it as follows: "RIEE is a genuine secondary emission of adsorbates invoked by radioactive radiation". Electrons are emitted from the surface of the source rather than from its bulk. At the same time, the properties of the surface are governed by properties of the electron system located on the surface of the adsorbed film. Therefore, e_0 -electrons arise at the source surface; however, the reason for the emission emergence and how it occurs are not contained in the definition of RIEE.

In work [4], under ultrahigh vacuum conditions, the specimen surface was cleaned using inert gases, and the zero-energy peak was simultaneously measured. The peak intensity was shown to decrease with increase in the time duration of the surface cleaning procedure, maybe owing to a reduction in the number of e_0 -electrons on the surface. Our work is not aimed at explaining the reasons for why e_0 -electrons emerge on the surface. The main purpose of our researches is to explain the origin of the e_0 -electron emission into vacuum at the radioactive decay or when charged particles pass through a target.

Our model consists in that the emission of e_0 electrons from the surface of a radioactive source is stimulated by the sudden emergence of an electric charge near the surface. This charge appears immediately at the time moment of a radioactive decay or when a charged particle crosses the surface. The emission of e_0 -electrons from the surface is a partial case of the shakeoff effect. The latter has been studied the most comprehensively in the case of β -decay, when an electron is shaken off from the atomic shell, and, as a result, the charge of the nucleus changes by +1 in a jump-like manner. In a series of works [5-9], we analyzed the yield Y of e_0 -electrons into vacuum and its dependence on the kind of radioactive decay, radioactive source thickness, magnitude of charge that arises in the course of the radioactive decay, and velocity of charged particles that cross the target surface. In this work, our efforts will be focused on the determination of the e_0 -electron distribution over energies and the binding energy of e_0 electrons, E_n , before they are emitted from the source or target surface.

However, first of all, let us consider, in more details, the e_0 -electron emission as a result of the sudden emergence of an electric charge near the surface, i.e. the shakeoff effect. The model is based on the theoretical ideas set in works [10,11] and follows from the solution of a non-stationary Schrödinger equation obtained in the first order of perturbation theory.

2. Emission of e_0 -Electrons as a Shake-Off Effect

The yield of e_0 -electrons in the energy range from 0 to E, which are shaken off into vacuum as a result of the sudden emergence of an electric charge in their vicinity at the time moment, when a charged particle crosses the target surface, is determined by the formula

$$\Upsilon(E) = \pi \frac{c}{v_p} \left(\frac{\Delta Z e^2}{x}\right)^2 \left| \int \psi_f^* \psi_i^{(0)} dq \right|^2 \times b \int_0^E \frac{\sqrt{E} dE}{\left(E + E_n\right)^2} = B \int_0^E \frac{\sqrt{E} dE}{\left(E + E_n\right)^2} = BF(E). \quad (1)$$

Here, c is the light velocity; v_p the velocity of a charged particle; ΔZe a charge that suddenly arises, as the perturbation is transferred to an e_0 -electron; x is the average distance between two neighboring weakly coupled electrons on the surface, which can be shaken off into vacuum; $\left|\int \psi_f^* \psi_i^{(0)} dq\right|$ is the matrix element for the transition of the system from the initial neutral state i into the finite state f with a vacancy instead of the escaped electron; q are the coordinates of wave functions; $\psi(q)$ is the coordinate parts of wave functions in the case of stationary motion, i.e. $\Psi(q,t) = \psi(q) \exp\left(-iE\frac{t}{\hbar}\right), \ b = \frac{\sqrt{2m^3}V_e}{\pi^2\hbar^3}$ is the coefficient in the expression $\nu = b\sqrt{E}$ for the e₀-electron level density in the continuous spectrum; $V_{\rm e}$ is the volume occupied by an e_0 -electron in the continuous spectrum, m is the e₀-electron mass, and F(E) is the integrated distribution of e_0 -electrons in the continuous spectrum over their energies in the interval from 0 to E. The latter is defined as the integral of the differential distribution of e_0 -electrons,

$$F(E) = \int_{0}^{E} \frac{\sqrt{E}dE}{\left(E + E_n\right)^2} = \frac{1}{\sqrt{E_n}} \operatorname{arctg} \sqrt{\frac{E}{E_n}} - \frac{\sqrt{E}}{E + E_n},$$

$$F(0) = 0.$$
 (2)

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Here, E_n is the binding energy of an electron on the surface, from which it is shaken off. Only this parameter must determine the distribution of e₀-electrons over the energy.

When writing down formula (1), we proceeded from the main formula that determines the probability of the shakeoff effect [12],

$$dW\left(E\right) = \left(\frac{\Delta Z e^2}{r}\right)^2 \left|\psi_f^* \psi_i^{(0)} dq\right|^2 b \frac{\sqrt{E} dE}{\left(E + E_n\right)^2}.$$

When crossing the target surface, the charged particle interacts with an e₀-electron located at a distance r from it and transfers the perturbation $\frac{\Delta Z e^2}{r}$ to this e₀-electron.

We consider a perturbation to be sudden if the relation $\tau \ll \omega_{fi}^{-1}$ is satisfied. Here, ω_{fi} is the transition frequency, and τ is the perturbation transfer duration. In this case, the wave function Ψ_i^0 has no time to change and continues to remain in the previous state as $\Psi_i^0(q,t) = \psi_i^0(q)$, and the total energy of the obtained perturbation is spent by the electron only on overcoming the binding energy E_n and acquiring the kinetic energy E, so that

$$\frac{\Delta Z e^2}{r} = E + E_n,\tag{3}$$

where E = 0 at $r_{\text{max}} = \frac{\Delta Z e^2}{E_n}$. Electrons on the surface can get various amounts

Electrons on the surface can get various amounts of energy. However, owing to the condition $\tau \ll \omega_{fi}^{-1}$ and the uncertainty relation that looks like $\Delta E \tau = \hbar$, we obtain that $\frac{\Delta E}{E} \gg 1$ at the shakeoff time moment, so that their energies become indistinguishable, and the distribution of e₀-electrons over the energy is governed only by the function F(E). Therefore, the distance, from which an electron was emitted, can be determined only after the energy of e₀-electrons has been measured.

In order to apply formula (1) describing the yield of e₀-electrons which can be simultaneously shaken off into vacuum in the energy interval from 0 to E, it is necessary to take into account that those electrons occupy an area on the surface equal to that of the ring, $|\pi r^2 - \pi r_{\max}^2|$, and their number equals $\frac{1}{x^2} |\pi r^2 - \pi r_{\max}^2|$. Such a record of the ring area with the indicated order of its terms (that is why the usage of the absolute value operation is needed) is associated with the necessity to put in agreement its terms – just in that order! – with the integration

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limits of e_0 -electrons from 0 to E in the formula for F(E). Moreover, πr_{\max}^2 is multiplied afterward by F(E) = 0 and is excluded from the further consideration. At $r > r_{\text{max}}$, the energy transferred to the electron is lower than E_n , and the shakeoff cannot occur without violating the conservation law of energy. On the other hand, if e_0 -electrons are arranged within a circle of the radius r, which corresponds to energies higher than E, the shakeoff is not observed, because there are no such levels in the interval from 0 to E in the continuous spectrum of the final state. At last, taking into account that the probability for the system to be perturbed by a particle that passes by depends on the velocity v_p of this particle as $\frac{c}{v_p}$, the first multipliers in formula (1), which determine the probability of system perturbation by a charged particle that passes by, look like $\pi \frac{c}{v_p} \left(\frac{\Delta Z e^2}{x}\right)^2$.

In contrast to the case where the perturbation is suddenly created by a charged particle that passes by, which induces a shakeoff event of the "perturbationby-scattering" type [13], the source presented by a motionless charge that suddenly emerges at the distance h from the surface emitting e_0 -electrons induces a shakeoff event of the "perturbation-by-switchingon" type. In the latter case, it is worth excluding the multiplier $\frac{c}{v_p}$ from formula (1) and making changes in formula (3), so that we have

$$\frac{\Delta z e^2}{\sqrt{h^2 + r^2}} = E + E_n$$

where $\sqrt{h^2 + r_{\max}^2} = \frac{\Delta z e^2}{E_n}$ at E = 0. The shakeoff phenomenon can be described as con-

The shakeoff phenomenon can be described as consisting of two stages. At the first stage, the system is suddenly perturbed. The probability of perturbation is described by the multiplier $\pi \left(\frac{\Delta Z e^2}{x}\right)^2$ or $\pi \frac{c}{v_p} \left(\frac{\Delta Z e^2}{x}\right)^2$. The other terms in formula (1) are related to the second stage. They describe the probability for the system to transit from the initial state into the final one and the distribution of e₀-electrons in the energy spectrum. This stage does not depend on the perturbation origin at the the first stage, but cannot exist without the latter.

Below, we consider the experimental distribution of e_0 -electrons over the energy in the integrated spectrum, compare it with the theoretical description on

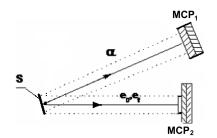


Fig. 1. Experimental setup: source (S), α -particle detector (MCP₁), electron detector (MCP₂)

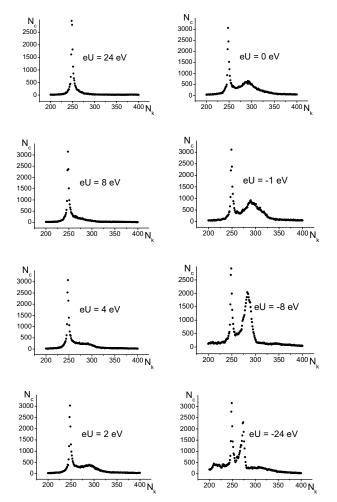


Fig. 2. Fragments in the spectra of $N_{e\alpha}$ -coincidences as a function of the delay potential, which illustrate the formation of the zero-energy peak. N_k is the analyzer channel number

the basis of the formulas presented above, and obtain a confirmation of the interpretation of the e_0 -electron emission as a shakeoff effect.

3. Researches of e_0 -Electron Distribution over the Energy

The emission of e_0 -electrons that are responsible for the appearance of the zero-energy peak was observed by measuring the delay curve N(eU). In this case, the retarding potential U was applied between the source (target) surface and the detector of electrons, so that the potential allowed only electrons with the energies E > eU to pass. An example of such measurements is the research of ²²⁶Ra decay, for which the experimental setup is shown in Fig. 1. A ²²⁶Ra source S from the set of standard spectrometric alpha-particle sources (OSAI) was covered with a $0.2-\mu m$ film to preserve the emanation of α -particles that penetrated through the source surface and induced the sudden appearance of a charge near weakly bound electrons at the surface, which resulted in their shakeoff into vacuum. Those e_0 -electrons were registered by detector MCP₂ consisted of two microchannel plates (MCPs) combined with each other in the form of chevron. In addition, fast e_0 -electrons also arrive at MCP₂, whereas α -particles emitted from the target are registered by detector MCP_1 . The source and detectors are arranged in a vacuum chamber. The measurements were carried out at a pressure of 5×10^{-6} mm Hg. The retarding potential U was created between the target and MCP₂. The delay curve N(E) was measured in the (e α)-coincidence regime, which allowed the N(E)components for e_0 - and e_f -electrons to be resolved in the time spectrum of coincidences. The spectra were registered on a multichannel pulse analyzer.

One may trace the zero-energy peak formation by analyzing Fig. 2, where the fragments of the $(e_f \alpha)$ and $(e_0 \alpha)$ -coincidence spectra are shown for various retarding potentials U. At eU = +24 eV, only the peak of fast $(e_f \alpha)$ -coincidences is observed in the spectrum. As eU decreases (eU = +8, +4, and+2 eV), the growth of the zero-energy peak is observed, which should have stopped at eU = 0 eV, when all e_0 -electrons in the given effective solid angle $\Omega_{\rm ef}$ reach detector MCP₂. However, the peak continues to grow even after the delay curve potential becomes negative, eU < 0. This occurs owing to the drawing of e_0 -electrons from other regions beyond the solid angle. It is visible from the consideration of three other spectra shown in Fig. 2, in which the e_0 peak intensity grows firstly and then falls down, with the peak shifting toward the e_f -one. However, we are interested only in the retarding potential interval

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from +24 to 0 eV. In the whole range of the retarding potential from eU = +24 eV to eU = -24 eV, the peak of fast-electron coincidences practically does not change. Therefore, generally speaking, for the delay curve to be analyzed further, it is not necessary that the measurements of $(e_0\alpha)$ -coincidences should be carried out separately. Instead, the ordinary delay curve $N(e_0 + e_f)$ plotted without coincidences can be used, which considerably simplifies the measurement procedure. At the center of Fig. 3, the circles demonstrate the values of delay curve in a simple spectrum of $(e_0 + e_f)\alpha$ -coincidences, and the squares show the corresponding values, when the contribution of $(e_0\alpha)$ -

coincidences was subtracted. The dependence of the integrated spectrum of e_0 electrons on the energy, $N_f(E)$, can be obtained from the following relation for the delay curve:

$$N_{f}(E) = \int_{0}^{E_{\max}} \frac{dN}{dE}(E) dE - \int_{E}^{E_{\max}} \frac{dN}{dE}(E) dE =$$
$$= \int_{0}^{E} \frac{dN}{dE}(E) dE = N(0) - N(E).$$
(4)

Then, for the equation

$$N_{f}\left(E\right) = AF\left(E\right),$$

where

$$A = \varepsilon \Omega_{\rm ef} n_{\alpha} B,\tag{5}$$

 ε is the efficiency of e₀-electron registration by detector MCP₂, $\Omega_{\rm ef}$ is the effective solid angle of registration, n_{α} is the number of registered α -particles by detector MCP₁ while measuring $N_f(E)$, and the quantities B and F(E) are defined in formulas (1) and (2), respectively. To fit the values of A and E_n over all measured points, the least-squares method was used.

Hence, in order to describe the distribution of e_0 electrons composing the zero-energy peak, it is necessary to measure the delay curve in the interval from 24 to 0 eV, then to calculate the integrated distribution of e_0 -electrons over energies, and to carry out a fitting procedure for the parameters A and E_n in the theoretical distribution (2) over all measu- red points.

In Fig. 3, the integrated energy spectra of e_0 electrons obtained from the delay curves in the cases

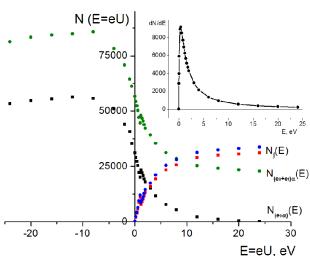


Fig. 3. Delay curves $N_{e\alpha}$ and the corresponding integrated spectra N_f . The inset illustrates the differential distribution of e₀-electrons over the energy (see other explanations in the text)

of $(e_0+e_f)\alpha$ - and $(e_0\alpha)$ -coincidences are shown. They agree well with one another and determine the binding energy as $E_n = 0.93 \pm 0.07$ or 0.94 ± 0.08 eV, respectively. Three years ago, we measured the delay curve for this ²²⁶Ra source twice. According to those data, the binding energy was $E_n = 0.83 \pm 0.06$ and 0.70 ± 0.04 eV, which can be regarded as a good agreement. On the basis of the value of binding energy E_n for e₀-electrons that are shaken off from the surface and the experimental results, we calculated their differential distribution over the energy by formula (2). The results of calculations are depicted in the inset in Fig. 3.

As was already mentioned above, the changes of the potential U induce variations of the solid angle. As a result, the energy spectrum of e_0 -electrons becomes distorted. The differential spectrum is especially sensitive to that. Really, the derivative of a delay curve should pass through zero at $E_{\rm e} = -0$ eV, but actually it does not. Even in the works, where the differential spectrum was measured directly on high-resolution spectrometers, the spectrum was strongly deformed in a vicinity of 0 eV owing to the appreciable influence of a device line in this region (the spectrum has negative values at E < 0 eV) [3]. The differential spectrum looks like a narrow peak with the intensity maximum at $E_{\text{max}} = \frac{1}{3}E_n$, i.e. in a vicinity of several tenths of electronvolts, where all distortions manifest themselves most strongly.

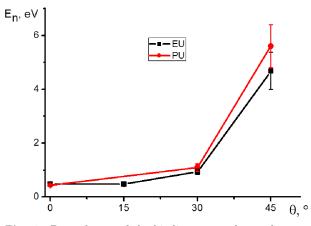


Fig. 4. Dependences of the binding energy for e₀-electrons on the exit angle θ with respect to the normal to the target surface for ¹⁵²Eu (squares) and ²³⁸Pu (circles) sources

The integrated spectrum undergoes smaller distortions than the differential one. First, the equality $N_{\ell}(E) = 0$ is always satisfied at E = 0, as it must be. Second, distortions more weakly affect the result at a low energy of e_0 -electrons, because the main values of $N_f(E)$ are obtained at higher energies. For instance, the value $E_n = (0.93 \pm 0.07)$ eV obtained by us in the main consideration, when the points with delay eU in the interval from 0 to 2 eV with an increment of 0.2 eV (see Fig. 3) were used, can be compared with E_n obtained, when the points are taken in the order 0, 1, 2 eV and so on, which lowers the first point down. The binding energy E_n changes at that only from 0.85 ± 0.09 to 0.98 ± 0.07 eV, i.e. the possible distortions at the beginning of the integrated spectrum weakly affect the final result for E_n .

During several years, when we studied the yields of e₀-electrons from various sources, we also often measured the corresponding delay curves. Now, this enables us to obtain the data for them concerning the binding energy of weakly bound electrons on the surface. The following E_n -values were obtained for the measured sources: 0.41 eV for ${}^{46}\text{Sc} - 0.41$, 0.60 eV for 60 Co, 0.62 eV for 152 Eu, 0.58 eV for 153 Gd, 0.74 eV for 154 Eu, 0.82 eV for 226 Ra, 0.79 eV for 233 U, 1.0 eV for ²³⁸Pu, and 0.86 eV for ²³⁹Pu. The first five sources are stable isotopes sputtered onto substrates and irradiated in a reactor according to the reaction $(n\gamma)$. The other sources are classed to the group of standard α -sources (OSAI); here, the radioactive isotopes were deposited directly onto substrates. Isotope ²²⁶Ra was an exception, which was already mentioned above.

The data on the binding energies of electrons, E_n , are quoted as the averaged values over several measurements carried out at various times. Those values are presented without measurement errors, because other errors, besides statistical ones, can emerge owing to different conditions of measurements. The deviations from the quoted averaged values can reach 20–30%. Nevertheless, in all cases, e₀-electrons are emitted into vacuum from bound states with energies lower than 1 eV.

In work [14], we studied the angular distribution of e₀-electrons emitted from the source surface into vacuum. The research showed that the emission of e_0 electrons is directed strongly forward along the normal to the surface and quickly diminishes as the angle θ between the direction of their motion toward detector MCP_2 and the normal to the surface increases. In this work, the delay curves were measured for e_0 -electrons at various angles θ for ¹⁵²Eu and ²³⁸Pu sources in order to determine E_n . The ²³⁸Pu source was covered with an aluminum foil. The values of E_n obtained in those measurements are shown in Fig. 4. A substantial growth of the binding energy E_n with the exit angle θ is observed. Therefore, since the intensity maximum in the differential spectrum is located at the energy $E_{\text{max}} = \frac{1}{3}E_n$, and the line halfwidth is equal to about $2E_n$, all the lines drastically broaden out and shift to the right, toward higher energies. At $\theta = 60^{\circ}$, the shakeoff effect becomes practically unobservable.

While studying the yield of e_0 -electrons for the $^{152}\mathrm{Eu}$ source depending on the kind of radioactive decay [15], we also measured the corresponding delay curves. We measured the coincidences with γ -rays using a GX-40 γ -spectrometer. The delay curves for various decay types were registered in three spectra of electron coincidences: with $\gamma 344$ keV, when the β^{-} decay plays the role of a sudden perturbation source; with $\gamma 122$ keV, when a sudden perturbation arises after the electron capture; and with K_{α} -rays, when a sudden perturbation arises after the internal conversion of $\gamma 122$ keV. All three curves gave rise to the energy distribution of e_0 -electrons, which is described by formula (2) with the parameter $E_n = (1.3 \pm 0.6) \text{ eV}$ for $(e_0\gamma 344)$ -coincidences, 1.8 ± 0.3 eV for $(e_0\gamma 122)$ coincidences, and 1.3 ± 0.2 eV for (e_0K_α) -coincidences. The larger values of E_n that were observed in those measurements resulted from the fact that the registered e_0 -electrons were emitted at a certain angle with

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respect to the source surface. Since the same surface was engaged in all three cases, the E_n -values must be identical. We may adopt that all the three values obtained coincide within the measu- rement errors.

4. Conclusions

To summarize, our research has shown that the zeroenergy peak observed at the radioactive decay always contains the energy distribution of e_0 -electrons described by formula (2) as the shakeoff effect, i.e. the sudden perturbation by a charge that arises near the surface and shakes off weakly bound electrons on the surface. The binding energy of those electrons does not exceed 1 eV, and their concentration depends on the surface contamination, although it is impossible to exclude their presence on extremely clean surfaces as well [4].

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РОЗПОДІЛ ЕЛЕКТРОНІВ ЗА ЕНЕРГІЄЮ В "ПІКУ НУЛЬОВОЇ ЕНЕРГІЇ", ЩО ВИНИКАЄ ПРИ РАДІОАКТИВНОМУ РОЗПАДІ АБО ПРИ БОМБАРДУВАННІ МІШЕНІ ЗАРЯДЖЕНИМИ ЧАСТИНКАМИ

Резюме

Проведено дослідження розподілу за енергією е₀-електронів, що вилітають з поверхні джерел при радіоактивному розпаді та при бомбардуванні їх зарядженими α - і β частинками. З вимірів кривої затримки при подачі затримуючого потенціалу між джерелом (або мішенню) та детектором е₀-електронів визначався інтегральний спектр е₀електронів N(E) в інтервалі від 0 до 24 еВ. Показано, що отриманий розподіл е₀-електронів добре узгоджується з теоретичним, який випливає з опису його як ефекту струсу – це раптове збурення електричним зарядом, який виникає поблизу поверхні, що приводить до струшування слабкозв'язаних електронів з поверхні.