INVESTIGATION OF THE ANGULAR DISTRIBUTION OF ELECTRONS EMITTED FROM THE SURFACE OF RADIOACTIVE SOURCES

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By the method of timing (eγ)-, (eα)-, and (eX)-coincidences, we study the angular distributions (ADs) of electrons e_0 with near-zero-energy and fast electrons e_f for different types of radioactive decay. It is established that the AD of e_0-electrons is strongly prolate forwards, whereas it has the cosine form for e_f. The character of the AD of e_0-electrons is affected by the passage of the surface barrier at different angles. The performed studies confirm our ideas of the emission of e_0-electrons as the shake-off of free electrons from the surface with radioactive sources due to the sudden appearance of an electric charge near the surface.

Introduction

On the radioactive decay, electrons with a very small energy are emitted from the source surface. These electrons form a peak in the spectrum named the "zero-energy peak" in the literature [1]. The peak maximum is located at an energy of 0.5 — 1 eV, and its half-width is about 1 eV. Its intensity rapidly decreases with increase in energy, and it can be omitted at an energy of ~20 eV [2]. We call the electrons forming the "zero-energy peak" as near-zero-energy electrons and denote as e_0 unlike the other electrons of the spectrum. The latter are called fast electrons and denoted by e_f.

On the basis of available experimental data and the studies performed by us [3], we assume the following mechanism of the creation of e_0-electrons. If an electric charge suddenly appears near the source surface, it can induce the transtition of a free valent electron from the surface in a continuous spectrum state like that this occurs, for example, upon β-decay (the shake-off effect). Moreover, the closer the charge appears to the surface, the greater the probability of the shake-off of an e_0-electron. On the contrary, if a charge appears inside the source at a distance of 5–6 atomic layers from the surface, the shake-off is practically absent. In this case, the distribution of e_0-electrons over energy is not changed, and the yield Y is the mean number of electrons which escape due to the appearance of charges in atoms located at various places of the near-surface layer. The probability of a shake-off is proportional to the square of a charge appeared in an atom, and the energy distribution of emitted e_0-electrons is approximately identical for various types of radioactive decay. To better comprehend the process of shake-off, we investigated the angular distribution of e_0- and e_f-electrons emitted from the surface of radioactive sources upon a radioactive decay.

1. Conditions of the Execution of Experiments

The angular distributions of the near-zero-energy electrons and fast electrons were determined in the measurements of the timing spectra of the coincidences of α-particles with electrons or the coincidences of γ, X_k-rays with electrons emitted from the surface of a radioactive source. As a source of α-particles, we took ^{238}Pu or ^{226}Ra from the spectrometric collection OSAI. Each source is made of stainless steel plate 24 mm in diameter and 2 mm in thickness, on which the radioactive spot 12 mm in diameter is applied and firmly fixed at the center [4]. The ^{226}Ra source is covered with a thin protective TiO_2 film 2 μm in thickness which allows one to preserve a gas emanation and, respectively, the whole sequential radioactive series, being in equilibrium with it. For the registration of α-particles, a surface—barrier detector on the basis of n-Si, whose total sizes were ⌀24×4 mm^2 upon the registration surface Ø12mm, was used [5].

The radioactive ^{152}Eu source was used in studying the angular distributions of e_0 and e_f emitted from the surface after the electron capture and internal conversion. It was a radioactive spot ⌀10 mm applied by electrolysis on a support made of stainless steel 20 mm in diameter. By our estimations, the thickness of the ^{152}Eu source was approximately two near-surface layers (10—12 atomic layers). Thus, our studies were carried out with real sources used in nuclear spectroscopy (sources have a "dirty" surface, and measurements are executed at the standard vacuum $p \sim 2 \cdot 10^{-6}$ mm Hg).

The reason for the sudden appearance of a large charge in a near-surface layer upon the decay of ^{152}Eu is the cascade filling of a vacancy on one of the deep shells of an atom with Auger-electrons. Such a vacancy is formed upon electron capture or internal conversion of γ-rays. ^{152}Eu undergoes also β^{-}-decay. But, in this case, a small charge is created, and we neglect this process here. Upon the α-decay of ^{226}Ra and ^{238}Pu, a reason for the appearance of a great charge in the near-surface layer is the ionization of several atoms.

The scheme of the experiment is presented in Fig. 1. On a shaft in the vacuum chamber, a radioactive source is attached. The source can be rotated by an angle θ in the interval from $-90°$ до $+90°$. The angle θ is formed by the normal to the source surface and the electron registration direction. The registration of electrons is realized by a detector, being the assembly of two microchannel plates (MCP) in the form of a chevron. It is positioned at a distance of 4 cm from the source. For $\theta = 0$, the plane of the input window of MCP is parallel to the source plane. Before MCP, a brass diaphragm ⌀10 mm is positioned, which restricts the passage of the electron beam through it. In some cases, we used also another diaphragm with a rim of the same diameter and 6 mm in height put on its window to eliminate the registration of scattered electrons. The registration of γ-and X_k-rays was carried out with an X-ray HPGe-detector positioned on the outer side of the chamber at a distance of 6 cm from the source. With the purpose to decrease the absorption of γ-rays in the chamber wall, we have mounted a Be-window ⌀30 mm and 50 μm in thickness. A turn of the source does not practically change the efficiency of the registration of γ-rays by an HPGe-detector.

To register α-particles, we have mounted an α-detector at a distance of 5 cm from the source center. It is toughly connected with the source so that their mutual position was invariable upon a change in θ. The angle ϑ between the direction of the registration of α-particles and the normal to the source lay in the plane including the source rotation axis and its normal and was 35^0. That is, the angles θ and ϑ were in two mutually perpendicular planes. For small rotation angles θ, the α-detector moves above MCP. That is, the plane of its input window is positioned farther from the source than the input window of MCP. We verified that the α-detector does not disturb the angular distribution of e_0-electrons which are registered by MCP (we will discuss

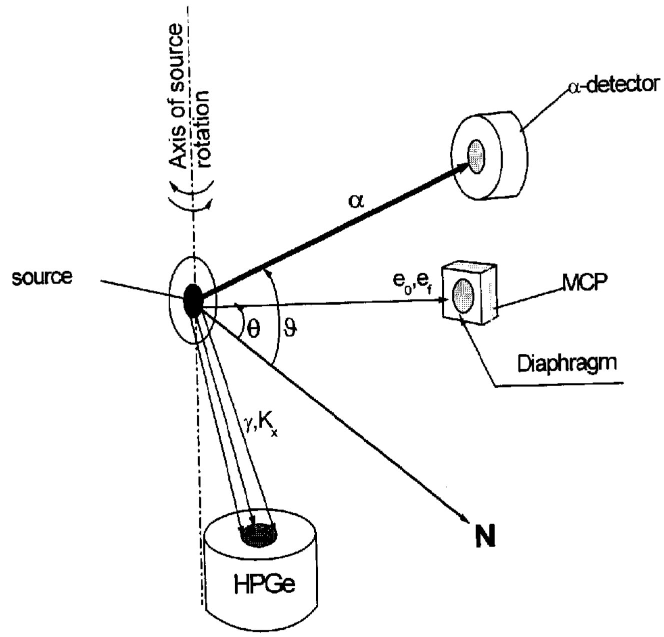

Fig. 1. Scheme of the experiment

this point below). Thus, upon a rotation of the source, MCP registered the electrons escaped at various angles to its surface in the coincidences with γ-and X_k-rays or with α-particles.

2. Execution of the Measurements with a ^{152}Eu Source

To determine the angular distribution of electrons emitted from the surface of the radioactive ^{152}Eu source, we measured the probability of the registration (eγ)-coincidences as a function of the electron escape angle θ relative to the normal to the source, $R_{e\gamma}(\theta)$. To this end, we measured two timing spectra of (eγ)-coincidences for each source rotation angle θ. In one of them, the MCP surface and the source were at the ground potential $U = 0$. In this case, we observed two peaks in the timing spectra of coincidences which are referred to ($e_0\gamma$)- and ($e_f\gamma$)-coincidences. The symbol ($e_0\gamma$) stands for the coincidences of e_0-electrons with γ- and X_k-rays. Upon the registration of another timing spectrum of (eγ)-coincidences, we supplied the cutoff voltage $U = +24$ V on the source. In this case, e_0-electrons cannot reach MCP and are not registered, and only one peak of fast electrons is observed in the spectrum of (eγ)-coincidences. This allows us to easily determine the intensity of the peak of ($e_f\gamma$)-coincidences, $N_{e_f\gamma}(\theta)$ and

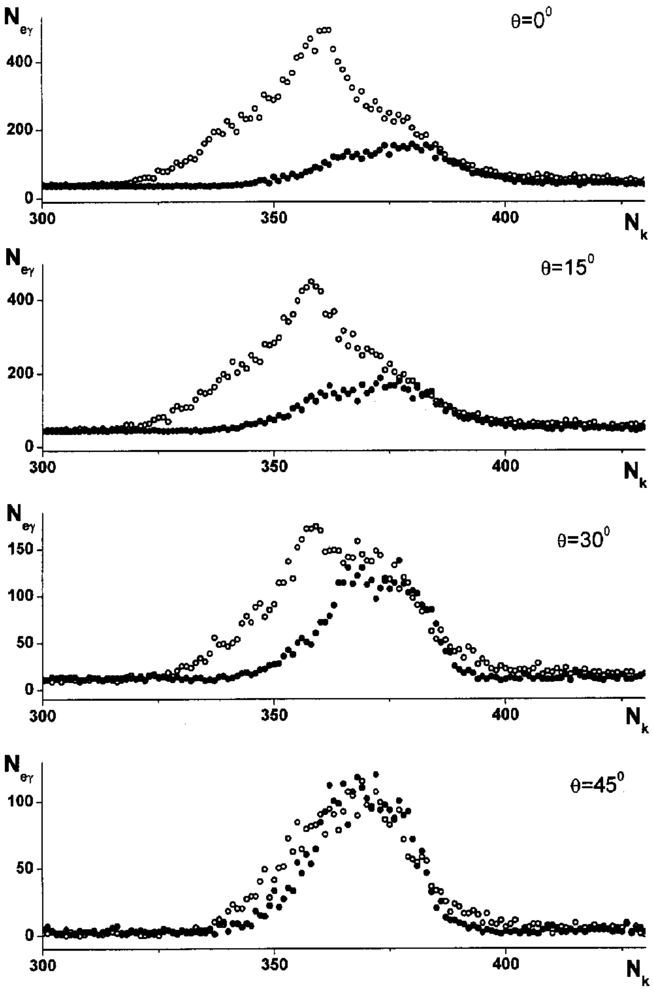

Fig. 2. Timing spectra of (eγ)-coincidences of electrons emitted from the source surface at various angles θ in the decay of ^{152}Eu. Open and filled circles correspond, respectively, to the spectrum derived at $U = 0$ and $U = +24$ V

then to find the intensity of the peak of ($e_0\gamma$)-coincidences, $N_{e_0\gamma}(\theta)$, from the first spectrum. The probabilities of the registration of coincidences for e_0- and e_f-electrons per one registered X_k- or γ-quantum can be defined as

$$R_{e_0\gamma} = \frac{N_{e_0\gamma}}{N_{\Sigma\gamma}} \quad \text{and} \quad R_{e_f\gamma} = \frac{N_{e_f\gamma}}{N_{\Sigma\gamma}},$$

where $N_{\Sigma\gamma}$ is the total intensity of the peaks of X_k-rays and γ in the simple spectrum which are registered for the same time interval as that for the spectra of coincidences. Performing the measurements at various rotation angles of the source, we can determine the probabilities of the registration of coincidences as a function of the escape angle of electrons from the source surface, $R_{e_0\gamma}(\theta)$ and

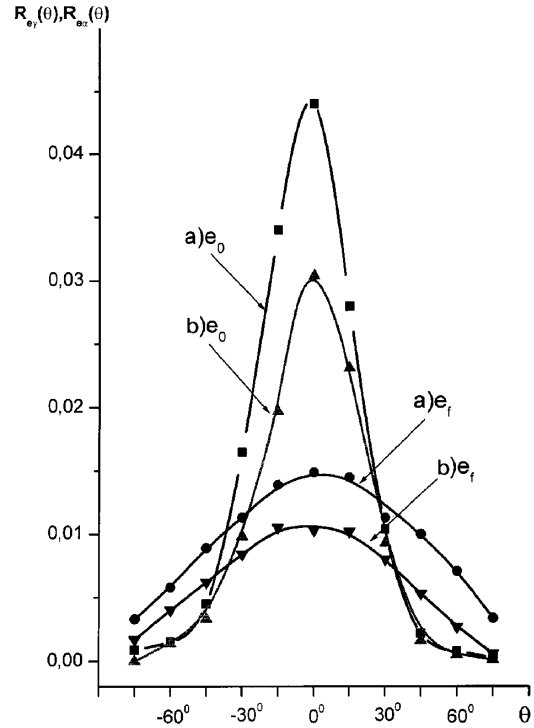

Fig. 3. Angular distributions of e_0- and e_f-electrons emitted from the source surface in the decays of ^{152}Eu (*a*) and ^{238}Pu (*b*)

$R_{e_f\gamma}(\theta)$. The intensity of electrons emitted from the source surface at various angles $dY/d\Omega(\theta)$ is proportional to the registration probability $R_{e\gamma}(\theta)$. Therefore, $R_{e\gamma}(\theta)$ can be considered as the angular distribution of electrons emitted from the source surface which is measured in some relative units.

As an example, Fig. 2 shows the timing spectra of (eγ)-coincidences derived in one of the series of measurements for electrons leaving the source at certain angles to its normal. From these spectra, we can determine the intensities of the peaks of fast electrons and e_0-electrons. The scale division of the analyzer channel was taken 0.8 ns, and the exposure time was 20 min for each spectrum. The intensities of peaks were determined by their areas in the timing (eγ)-spectra or in the simple γ-spectrum. As a result of the measurements, we established the dependences $R_{e_0\gamma}(\theta)$ and $R_{e_f\gamma}(\theta)$ presented in Fig. 3. As seen, the angular distribution $R_{e_0\gamma}(\theta)$ for e_0-electrons [curve $a)e_0$] has a clearly pronounced direction forwards.

Now consider the dependence $R_{e_f\gamma}(\theta)$ for fast electrons. Curve $a)e_f$ turns out to be elevated above the θ axis. This is explained by the presence of hard electrons emitted from the thin source isotropically, and their intensity does not depend on the escape angle θ (β^--particles, conversion electrons, and hard Auger-electrons). The remaining part of the distribution of electrons has an approximately cosine character like that observed in the works studying the angular distribution of truly secondary electrons [6].

3. Execution of Measurements with α-Sources

The initial scheme of the experiment with α-sources was somewhat different from that presented in Fig. 1. The distance between the source and MCP was 7 cm, rather than 4 cm. Therefore, an α-detector was positioned closer to the source, that MCP. It turned out that this strongly disturbs the angular distribution of e_0-electrons, whereas the angular distribution of fast e_f-electrons was not disturbed in this case if, of course, the α-detector does not shield the source from MCP. We convinced ourselves in this, by measuring $R_{e\gamma}(\theta)$ with a ^{152}Eu source in the presence of the α-detector, when the source rotates together with the α-detector by an angle θ, and without the α-detector. Upon the approach of the α-detector to MCP (at small angles θ), the intensity of the peak of $N_{e_0\gamma}(\theta)$ sharply decreases. But it increased without the α-detector. In both cases, the behavior of the peak of $N_{e_f\gamma}(\theta)$ remained the same, namely the increase in the count rate with decrease in an angle θ. Obviously, the movement of the α-detector is associated by the creation of some small fields which strongly change the trajectories of e_0-electrons upon the approach of the α-detector to MCP. Therefore, we have to be very careful as to the mounting any diaphragms between the source and the detector of e_0-electrons. After the approach of MCP to the source (see Fig. 1) and the movement of the α-detector beyond MCP, the distribution $R_{e_0\gamma}(\theta)$ became independent of the position of the α-detector (generally, of its presence or absence), and we can begin to measure $R_{e_0\alpha}(\theta)$ and $R_{e_f\alpha}(\theta)$, being the probabilities of the registration of e_0 and e_f-electrons by MCP per one α-particle registered by the detector at a certain escape angle of electrons. They are determined analogously to $R_{e_0\gamma}(\theta)$ and $R_{e_f\gamma}(\theta)$ in the decay of ^{152}Eu, but we measured the count rate N_α of the detector of α-particles rather than $N_{\Sigma\gamma}$.

In Fig. 4, we give the timing spectra of (eα)-coincidences at some angles θ for the ^{238}Pu source which were derived in one of the series of measurements. The determination of $N_{e_0\alpha}(\theta)$ and $N_{e_f\alpha}(\theta)$ by these spectra was carried out analogously to that described above for $N_{e_0\gamma}(\theta)$ and $N_{e_f\gamma}(\theta)$. In Fig. 3,*b*, we present the angular distribution of electrons emitted from the surface of a

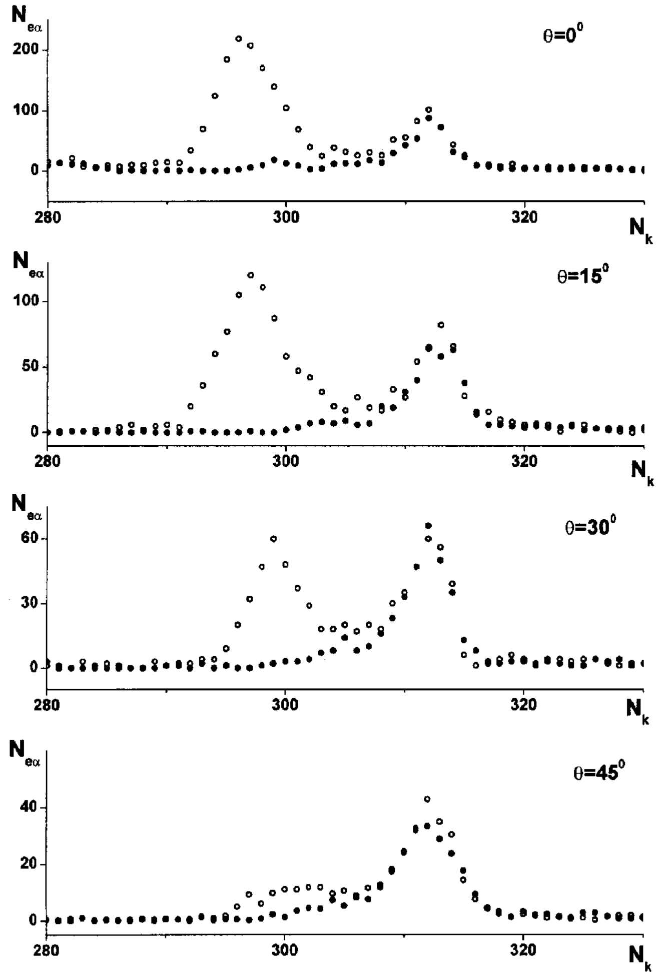

Fig. 4. Timing spectra of $(e\alpha)$-coincidences of electrons emitted from the source surface at various angles θ in the decay of ^{238}Pu. Open and filled circles correspond, respectively, to the spectrum derived at $U = 0$ and $U = +24$ V

radioactive ^{238}Pu source upon α-decay. The angular distribution of fast electrons $R_{e_f\alpha}(\theta)$ is close to cosθ like that observed in the decay of ^{152}Eu. The angular distribution of e_0-electrons from ^{238}Pu is practically the same as the distribution for ^{152}Eu. The former is sharply directed forwards and has the same half-width $\Delta_{1/2} \sim \pm 20°$. This confirms the identical nature of the emission of e_0-electrons after the electron capture, internal conversion of γ-rays and α-decay, which was indicated earlier upon the comparison of the energy spectra of e_0-electrons revealed in these phenomena [5].

Under the initial geometry mentioned at the beginning of this section, we measured the angular distributions $R_{e_f\alpha}(\theta)$ for the ^{226}Ra source covered with

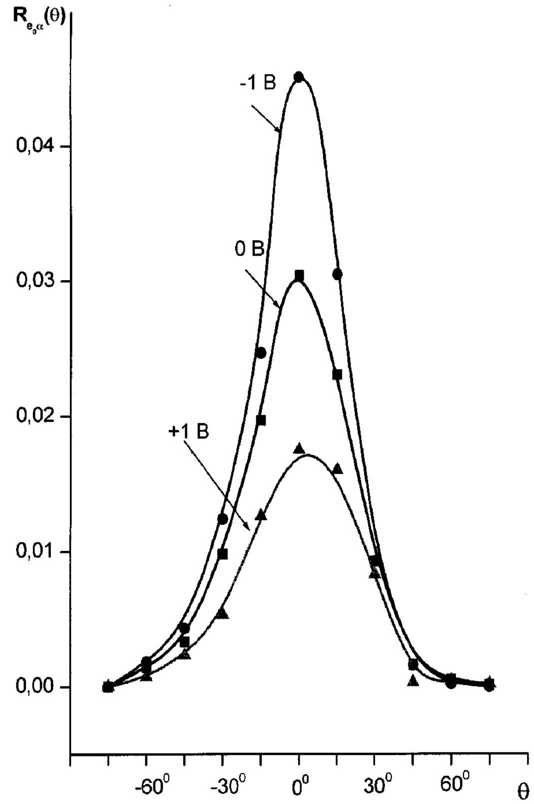

Fig. 5. Angular distributions of e_0-electrons emitted by ^{238}Pu upon the supply of the voltages of +1 V, 0, −1 V on the source

different foils such as: a) sprayed gold one 400 Å in thickness on a dacron 3.6-mg/cm^2 film, b) sprayed aluminum 20-μg/cm^2 foil on the same film, c) aluminum 10-μm foil, and d) very thin layer of Al sprayed on a dacron 180-μg/cm^2 film. All the distributions have forms similar to the до cosine one, and the maxima of the distribution at θ=0 decrease, for the first three mentioned coatings, sequentially in the limits of 20%. In case d) where α-particles did not lose practically their energy passing across the Al-film, this maximum was less by a factor of 1.5. We also estimated the energy composition of e_f-electrons by the delay curves. For example, for a group of α-particles emitted by ^{226}Ra with an energy of 4.8 MeV without coating and with a coating of the source with an Al-foil 10 μm in thickness, e_f-electrons with an energy more than 1 keV constituted, respectively, 2 and 6% of the total intensity of the registered e_f-electrons. For a group of 7.7-MeV α-particles, these numbers were, respectively, 14 and 16%. That is, the major amount of electrons emitted upon α-decay have energy < 1 keV.

In Fig. 5, we give three curves of the angular distribution of e_0-electrons emitted by ^{238}Pu which

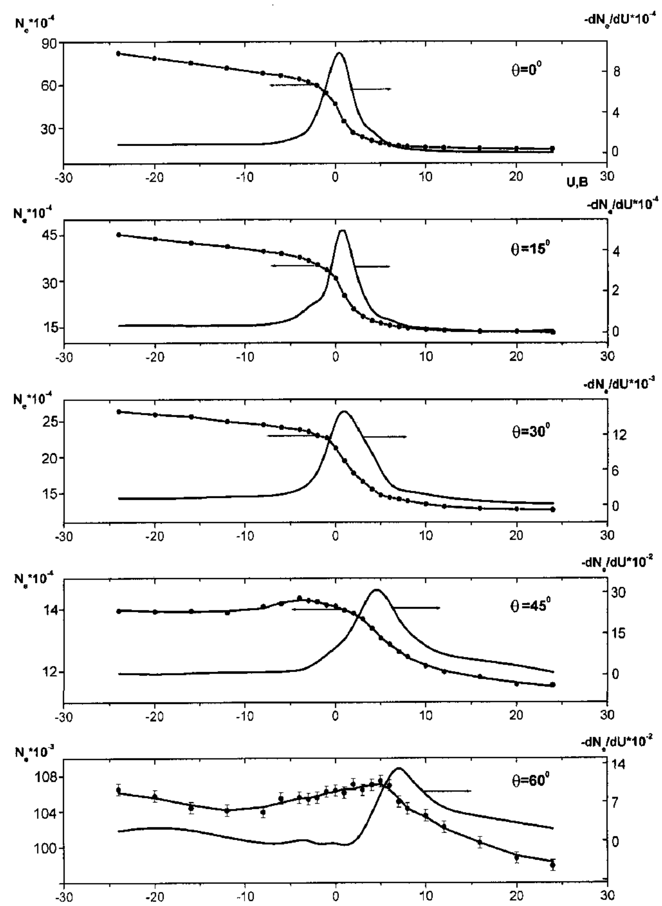

Fig. 6. Delay curves and their derivatives for electrons emitted from the ^{238}Pu source surface at various angles θ

correspond to the following voltages U supplied on the source: +1, 0, and −1 V. At U = −1 V, the emission intensity at the maximum of a curve increases approximately twice, and the angular distribution is narrowed. But, at U = +1 V, it decreases approximately twice, and the angular distribution is widened as compared to the distribution at U = 0. Such strong reaction of $R_{e_0\alpha}(\theta)$ on a small variation in the source potential can lead to a significant change in the distribution, if the work functions of the source surface and a detector are different.

We measured the delay curves $N_e(U)$ for electrons emitted from the ^{238}Pu source surface upon its rotation by various angles θ. The derivative Nd_e/dU gives an idea of the distribution of electrons over energy. In Fig. 6, we present the results of these studies. At $\theta = 0$, the distribution maximum of electrons is positioned near zero. But, with increase in the angle θ, the maximum of the energy distribution of e_0-electrons decreases and reaches $U \sim 7$ V at θ=60°. The reason for such a behavior is related to the necessity for e_0-electrons to overcome the surface barrier at an angle. If we remember that the emission of e_0-electrons decreases sharply with increase in their energy, we may also understand the sharp drop in the intensity of e_0-electrons with increase in the escape angle θ.

4. Discussion of the Results of Studies

In our opinion, the emission of near-zero-energy electrons is the shake-off of electrons from the source surface due to the sudden appearance of an electric charge near the surface [3]. The reason for the appearance of a large charge upon the internal conversion or electron capture was already explained above. Let us consider more thoroughly the reasons for the appearance of a large charge upon the α-decay. In the case of α-decay, the appearance of such a charge can be presented as the ionization of the core electrons of several atoms upon the passage of the near-surface source layer by an α-particle. According to work [7], upon the ionization of a core electron, it has a small energy (at most 5 eV) and cannot leave a source. However, this electron creates a vacancy in the filled zone. As the result of Auger-recombination, one of the electrons of the conduction zone fills in the created vacancy, whereas another electron of this zone escapes outside with energy $\delta - 2\varphi$, where δ is the binding energy of the filled zone, and φ is the electron work function. Thus, as the result of Auger-recombination, a charge equal to $+2e$ is created in the near-surface layer. By passing the near-surface layer, an α-particle can make several such ionization acts. Therefore, the total charge is approximately $+(6\div8)e$. In a simplified manner, we may imagine that this total charge is created at some single point of the near-surface layer. Just its sudden appearance in the near-surface layer leads to the shake-off of electrons from the source surface.

We note that, in a certain sense, the reason for the ionization of an atom upon α-decay is also a shake-off, i.e. a quantum-mechanical transition of an electron from the filled zone to the conduction zone under the action of a suddenly appeared excitation. In this case, the reason for the excitation is the charge of an α-particle during its approach to the electron shell of the atom, which lasts approximately 10^{-17}s, until the α-particle leaves the atom. This time of its flight satisfies the excitation suddenness criterion. The longer the α-particle is in the atom, the larger the transition probability. That is, the probability is inversely proportional to the square of its velocity.

Now consider the theoretical foundations of the emission of near-zero-energy electrons from the surface of a radioactive source. They are presented, e.g., in Section 41 of monograph [8]. According to [8], the transition probability upon a sudden excitation is defined as

$$dW_{if} = \frac{\left|\int \psi_f^* \frac{\Delta Z e^2}{r} \psi_i^{(0)} dq\right|^2}{(E_{e_0} + \varphi)^2} d\nu,$$

where dW_{if} – shake-off probability for a free electron of the surface, ΔZe — total charge created by an α-particle upon the passage of the near-surface layer (or due to the cascade of Auger-processes under the electron capture or internal conversion), r — distance from the charge to an electron of the surface, E_{e_0} — energy of an e_0-electron, ψ_f^* — wave function of an e_0-electron in the continuous spectrum when it leaves the source surface, φ — electron work function, $d\nu$ — number of states of e_0-electrons in the energy interval from E_{e_0} to $E_{e_0} + dE$, and $\psi_i^{(0)}$— wave function of a free electron of the surface at the moment of the creation of a charge ΔZe.

The yield of e_0-electrons, Y, will be defined by the shake-off probability W_{if} of an electron from the surface and the number of valent electrons of the surface, N_e, which can participate in the shake-off. This number can be estimated by using the following consideration. The depth of the near-surface layer is 5—6 atomic layers [9]. Taking the distance to be equal to 5 atomic layers as the radius of action of a charge on the surface, we get an area occupied by 75 atoms. Let us assume that every atom has 2 valent electrons. In this case, the total number of electrons able to be shaken off from the surface is $N_e =$ 150. However, the shake-off probability depends on the distance between a charge and the shaken-off electron and will be different for different arrangements of atoms. The mean shake-off probability W can be estimated from the yield of e_0-electrons upon the shake-off, $Y = WN_e$.

By our estimates, 7 e_0-electrons per one α-particle leaving the surface are emitted from the ^{226}Ra source surface [10]. Then we can roughly estimate the mean probability of the shake-off from the source surface for an electron which is located in the zone of influence of a suddenly appearing charge created by an α-particle flying through the near-surface layer as $W \sim 0.05$. For the ^{154}Eu source of $\sim$1 μg/cm^2 in surface density, we get the yield Y=4e_0 per one act of internal conversion [11], which corresponds to $W \sim 0.03$. We recall that, contrary to the α-decay where the yield of e_0-electrons from thin sources is practically insensitive to a change in their thickness. Upon the electron capture and internal conversion, the yield rapidly decreases with increase in the source thickness, because the appearance of a great charge beyond the near-surface layer does not induce the emission of e_0-electrons, and $N_{\Sigma\gamma}$ increases due to a growth of the source activity.

As for the angular distribution of e_0-electrons which has a pronounced directivity sideways the normal to the source surface, it is a result of the overcoming of a surface barrier by e_0-electrons. The greater the escape angle, the higher should be the energy of an e_0-electron to overcome the surface barrier and the harder in energy will be the emission at great angles. But the emission intensity will be less, because electrons with small energy are shaken off much more than those with great energy. The performed studies confirm our ideas of the emission of e_0-electrons as the shake-off of free electrons from the source surface due to the sudden appearance of an electric charge near the surface.

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ДОСЛІДЖЕННЯ КУТОВОГО РОЗПОДІЛУ ЕЛЕКТРОНІВ, ЩО ВИПРОМІНЮЮТЬСЯ З ПОВЕРХНІ РАДІОАКТИВНИХ ДЖЕРЕЛ

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Р е з ю м е

Методом часових ($e\gamma$)-, ($e\alpha$)-, (eX)-збігів досліджено кутові розподіли (КР) електронів близьконульової енергії e_0 і швидких електронів e_f для різних типів радіоактивного розпаду. Встановлено, що КР e_0-електронів сильно витягнутий вперед, тоді як для e_f він косинусоїдальний. На характер КР e_0-електронів впливає проходження поверхневого бар'єра під різними кутами. Проведені дослідження підтверджують наші уявлення про емісію e_0-електронів як струс вільних електронів з поверхні радіоактивних джерел внаслідок раптової появи електричного заряду поблизу поверхні.