

## STUDY OF $e_0$ -ELECTRON YIELDS FROM THE SURFACE OF THIN FILMS AFTER $\beta$ -PARTICLE IRRADIATION FROM $^{152}\text{Eu}$ , $^{154}\text{Eu}$ , AND $^{226}\text{Ra}$ DECAYS

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We study the yields of  $e_0$ -electrons emitted from the surface of thin films after the bombardment with  $\beta$ -particles from the radioactive sources with  $^{152}\text{Eu}$ ,  $^{154}\text{Eu}$ , and  $^{226}\text{Ra}$  by the method of  $(e\gamma)$ -coincidences. The yields of  $e_0$ -electrons are found to be inversely proportional to the velocity of incident  $\beta$ -particles. This means that the ionization of target atoms is proportional to the duration of the perturbation induced by  $\beta$ -particles passing near the atoms.

occurs in deeper atomic layers, the charge in the near-surface layer appears only due to the ionization of its atoms by electrons which were emitted in the radioactive decay and passed through this layer. The charge created in this case is less than that in the case of decays in the near-surface layer. The ionization of atoms of the near-surface layer by X-rays and  $\gamma$ -rays can be neglected at all.

### 1. Introduction

The emission of near-zero energy electrons ( $e_0$ -electrons) is the electron shaking-off from the target surface due to the sudden appearance of an electric charge near it [1, 2]. They make up a "zero-energy peak" in the electron spectrum of radioactive sources [3]. The width of this peak has an order of 1 eV, and its maximum approximately corresponds to 0.5 eV. The intensity of emitted electrons decreases with increase in the energy so fast that it can be neglected as the energy approaches approximately 20 eV [4]. If the charge appears outside the near-surface layer (several layers near the surface), its appearance does not result in the  $e_0$ -electron emission.

The electric charge appears due to the different reasons depending on the type of radioactive decay. For instance, in the processes of electron capture and  $\gamma$ -ray internal conversion, the charge appears mainly due to the creation of vacancies in one of the inner atomic shells and the subsequent cascade of Auger electrons due to their filling, which results in the high-degree atom ionization. In the  $\beta^-$ -decay, the charge appears due to the self-ionization of an atom through the increase of the nucleus charge by  $+1e$  after the emission of a  $\beta^-$ -particle, shaking the electron shell, and the direct collisions of the  $\beta^-$ -particle with electrons of the atom shell. All this takes place if the decaying nucleus is located in the near-surface layer. If the radioactive decay

In the  $\alpha$ -decay, the electric charge in the near-surface layer is created due to the atom ionization during the passage of an  $\alpha$ -particle through this layer. There is no basic difference between the situations where the  $\alpha$ -particle crosses the near-surface layer moving from the source interior or moving into the layer during the target bombardment, if the angle of incidence is the same in both cases. During the passage of  $\beta^-$ -particles through the near-surface layer, the atom ionization is realized in the same way but with a much less probability, if the  $\beta^-$ -decay occurred outside the near-surface layer i.e. in the regime of the bombardment of the near-surface layer of the target or source. For the both types of decay, the  $e_0$ -electron yield can be represented as the product of three factors:

$$Y_{e_0} \sim PWN_e. \quad (1)$$

Here,  $P$  is the atom ionization probability in the near-surface layer by a charged particle in a single collision, which defines the probability for a charge  $\Delta Ze$  to appear in the near-surface layer,  $W$  is the probability of the  $e_0$ -electron creation under the action of the perturbation  $\Delta Ze^2/r$  formed by the charge created in the near-surface layer,  $N_e$  is the number of conduction electrons located at the distance of  $r$  and capable to come out in vacuum. The yields of  $e_0$ -electrons during  $\alpha$ - and  $\beta^-$ -decays are different due to the different ionization probabilities  $P$  of the target or source. We assume the  $e_0$ -electron yield  $Y_{e_0}$  to be the number of  $e_0$ -electrons emitted from the

surface per one radioactive decay or per one charged particle bombarding the target.

In review [5] on the emission of secondary electrons, it is noted that the yield of secondary electrons emitted during the passage of a charged particle through the target is proportional to its charge squared and inversely proportional to its velocity:  $Y_{e_0} \sim Z_p^2 v_p^{-1}$ . In [6,7], the same dependence for  $\alpha$ -particles is presented. We suggest that  $e_0$ -electrons are emitted during the bombardment of the target by  $\alpha$ -particles due to sequential perturbations. At first, an  $\alpha$ -particle passing by the atom induces the sudden perturbation of atomic electrons, which causes the atom ionization. Then the charge created due to these sequential ionizations suddenly appears in the near-surface layer, and finally the electrons located near the surface are shaken-off in vacuum. It is obvious that the  $e_0$ -electron emission under the influence of  $\beta$ -particles is the same as the emission upon the  $\alpha$ -decay. The aim of the present work is the investigation of the yield of  $e_0$ -electrons upon the bombardment of thin films by  $\beta$ -particles emitted in the decay of radioactive nuclei.

## 2. Experimental Setup and Procedure

To define the dependences of the  $e_0$ -electron yield  $Y_{e_0}$  on the energy of  $\beta$ -particles, we chose the separate branches of  $\beta$ -decay with the different boundary energies of  $\beta$ -spectra. For each branch of  $\beta$ -decay, we calculate the averaged energy  $\bar{E}_\beta$  of the  $\beta$ -spectrum (along with the averaged velocity  $\bar{v}_\beta$  of  $\beta$ -particles), which is attributed to the yield  $Y_{e_0}$  measured for the given branch. The branches of  $\beta$ -decay are identified by  $\gamma$ -rays which are in the cascade with these  $\beta$ -transitions, through the measurements of  $(e_0\gamma)$ -coincidences.

The scheme of the experimental setup is presented in Fig. 1. To detect electrons, we use the assembly of two microchannel plates (MCP) in the form of a chevron, in front of which we arrange an aperture with  $\varnothing 26$  mm. The radioactive source is placed in the immediate vicinity of the detector. The angle between the planes of the source and detector is obtuse to prevent the incidence of direct rays on the MCP. This provides the decrease the MCP load from fast  $e_f$ -electrons and  $\alpha$ -particles, whereas  $e_0$ -electrons are extracted on its surface due to a positive voltage of 240 V supplied to it. To detect  $\gamma$ -quanta, we arrange an HPGe-detector outside the vacuum chamber at a distance of 5 cm from the source.

We use three sources for the measurements. Two sources made of  $^{152}\text{Eu}$  and  $^{154}\text{Eu}$  are the active spots  $\varnothing 12$  mm deposited on the stainless steel substrates  $\varnothing 20$  mm and 1 mm in thickness. To prevent the influence

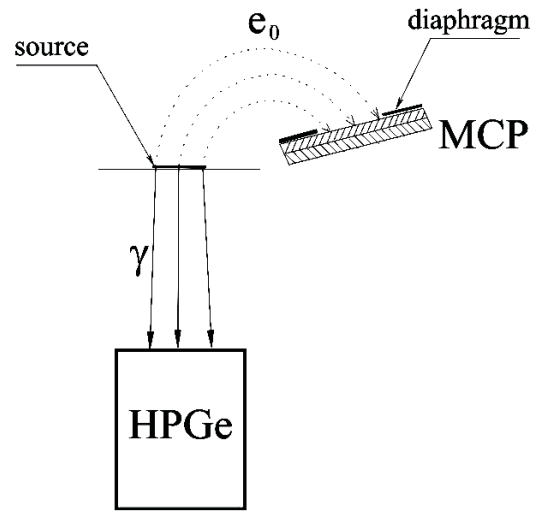


Fig. 1. Scheme of experimental setup

of the activity from the near-surface layer, each source is coated by an organic film having a thickness of  $2 \mu\text{m}$  with the transparent aluminum layer deposited on it. Thus, the near-surface layer of the source consists of inactive atoms of aluminum. The third source contains  $^{226}\text{Ra}$  from a spectrometric collection OSAI with an active spot  $\varnothing 12$  mm deposited on a stainless steel substrate with  $\varnothing 24$  mm and a thickness of 2 mm. The  $^{226}\text{Ra}$  source is coated by a protective film of  $\text{TiO}_2$  with a thickness of  $0.2 \mu\text{m}$  to keep the gas emanation and, hence, the whole following radioactive series [8]. In this case, the near-surface layer consists of inactive molecules of  $\text{TiO}_2$ .

The measurements are carried out by the following sequence. We measure the simple  $\gamma$ -spectra and  $\gamma$ -spectra in coincidences with  $e_0$ -electrons. The last spectra are derived as the difference spectrum of  $(e_0 + e_f)\gamma$ -coincidences and  $e_f\gamma$ -coincidences. The spectrum of  $e_f\gamma$ -coincidences is measured when a cutoff voltage of +280 V was supplied to the source. In this case,  $e_0$ -electrons do not reach the MCP. The separate peaks  $N_\gamma$  of the  $\gamma$ -spectrum allow us to identify the certain branches of  $\beta$ -decay that are in the cascade with these  $\gamma$ -transitions. In this case, the peaks of  $N_{e_0\gamma}$ -coincidences define the intensity of the emission of  $e_0$ -electrons. Then the detection probability of  $e_0$ -electrons per one detected  $\gamma$ -quantum is defined as

$$R_\gamma = \frac{N_{e_0\gamma}}{N_\gamma},$$

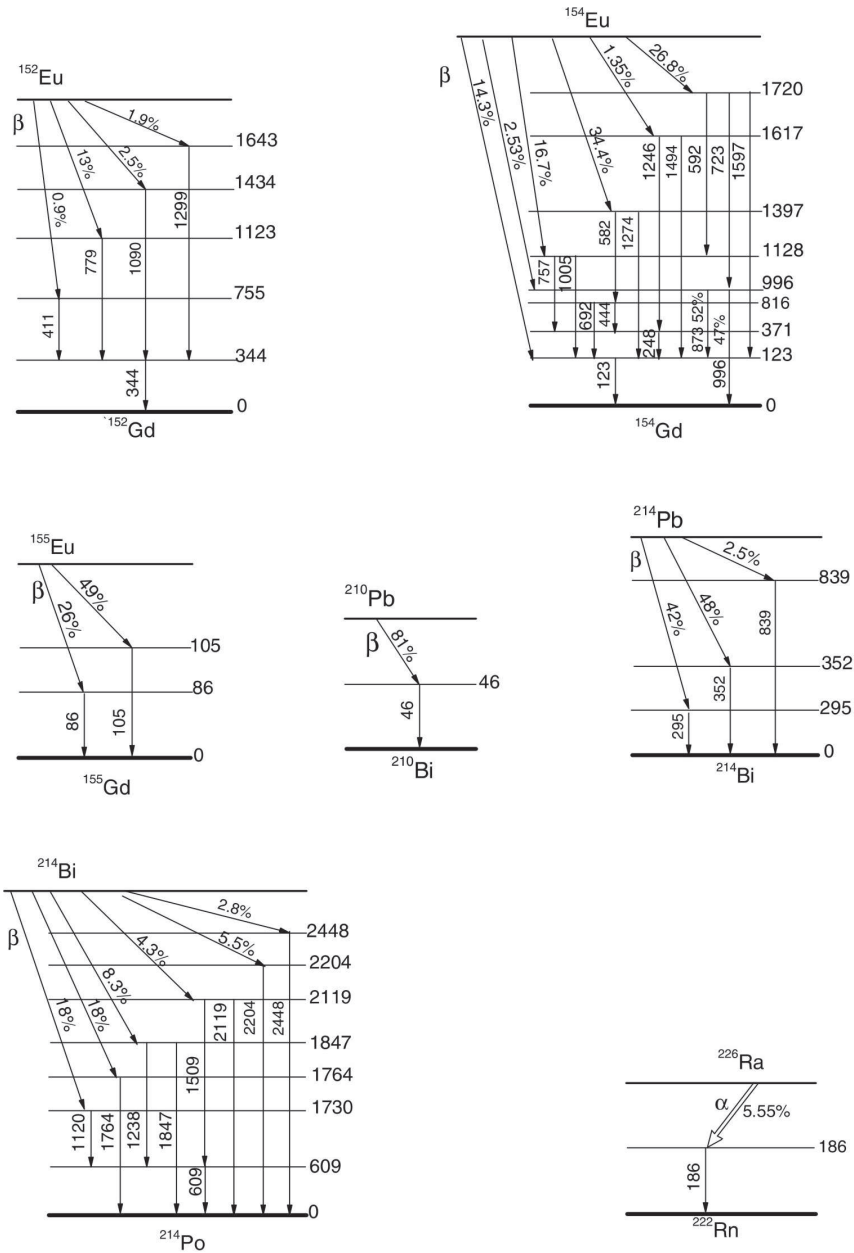


Fig. 2. Simplified fragments of the decay schemes for the nuclei, whose  $\beta$ -spectra are investigated in this paper

where  $N_{e_0\gamma} = N_{(e_0+e_f)\gamma} - N_{e_f\gamma}$  is the intensity of the  $\gamma$ -spectrum peak for coincidences with  $e_0$ -electrons measured during the same time as  $N_\gamma$ . The quantity  $R_\gamma$  directly defines the  $R_\beta$ -detection probability of  $e_0$ -electrons per one event of  $\beta^-$ -decay of the given type or through simple relations that follow from the nucleus

from the relation  $Y_\beta \approx \frac{R_\beta}{\varepsilon_0\Omega}$ , where  $\varepsilon_0$  is the efficiency of the detection of  $e_0$ -electrons by the MCP, and  $\Omega$  is the part of those  $e_0$ -electrons, which get to the MCP, relative to all the electrons emitted from the target surface. Since  $Y_\beta \sim R_\beta$ , we use hereinafter the values of  $R_\beta$  as the  $e_0$ -electron yields expressed in some arbitrary units, especially because they are directly measured in experiments. The estimate of  $\varepsilon_0\Omega$  will be given below.

The yield  $Y_\beta$  for the given branch can be determined

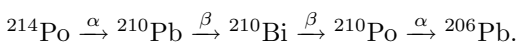
### 3. Experimental Results

In Fig. 2, we represent the simplified fragments of the decay schemes of the isotopes, whose  $\beta^-$ -transitions are studied in the present work.

Let us consider the  $\beta^-$ -transitions of the  $^{152}\text{Eu}$  decay. All the excited states of a daughter nucleus, which were created after various  $\beta^-$ -transitions, de-excite through the  $\gamma$ -transitions into the 344-keV first excited state. The transition  $344 \rightarrow 0$  (to the ground state) is weakly converted. Therefore, we can neglect the creation of  $e_0$ -electrons through the internal conversion  $\gamma_{344}$  keV. Then  $R_\gamma = R_\beta$ , where  $\gamma$  denotes the transition which de-excites the excited state created after  $\beta^-$ -decay. For  $R_{\gamma_{344}}$ , we can calculate the energy of  $\beta^-$ -particles as the sum of contributions of each branch to the creation of the 344-keV excited state.

In the  $\beta^-$ -decay of  $^{154}\text{Eu}$ , all the excited states of the daughter nucleus considered in this paper also decay to the first excited state. But its energy is 123 keV, and the transition to the ground state is highly converted and creates  $e_0$ -electrons. Hence,  $R_\gamma = R_\beta + R_{IC123}$  in this case. The additive nature of such probabilities was verified by us more than once. After a  $\beta^-$ -decay,  $e_0$ -electrons are emitted, and the system comes to the equilibrium. Then the internal conversion occurs, and  $e_0$ -electrons are emitted again. Both these emissions are detected as one. The 996-keV level is the exception, as it is de-excited through not only the transition  $\gamma_{873}$  keV to the first excited state, but also through  $\gamma_{996}$  keV to the ground state of  $^{154}\text{Gd}$ . This enable us to calculate  $R_{IC123} = R_{\gamma_{873}} - R_{\gamma_{996}}$  and, in turn,  $R_\beta$  for all studied branches. For  $R_{\gamma_{123}}$ , the averaged energy of  $\beta^-$ -particles is calculated as the sum of contributions of each branch to the creation of the 123-keV excited state. In addition, in the  $^{154}\text{Eu}$  source, there is the impurity of  $^{155}\text{Eu}$ , for the  $\beta^-$ -branch of which  $R_\gamma = R_\beta$ , because the  $^{155}\text{Gd}$  levels are de-excited through the direct  $\gamma$ -transitions to the ground state.

The  $^{226}\text{Ra}$  source is more complicated. The chain of its decay has the following form:



In the  $^{226}\text{Ra}$  source, the  $\beta^-$ -transitions accompany the decay of  $^{214}\text{Pb}$ ,  $^{214}\text{Bi}$ , and  $^{210}\text{Pb}$ . In the decay of  $^{210}\text{Pb}$  and  $^{214}\text{Pb}$ , we consider the direct  $\gamma$ -transitions to the ground state. Therefore,  $R_\gamma = R_\beta$ . In the decay of  $^{214}\text{Bi}$ , in addition to the direct  $\gamma$ -transitions, the cascade ones

to the underlying levels are considered as well. But all of them have sufficiently high energy, and the effect of internal conversion can be neglected, i.e. we have also  $R_\gamma = R_\beta$  for them. For the 609-keV first excited state, the averaged energy of  $\beta^-$ -particles for  $R_{\gamma_{609}}$  can be calculated, as it was done for  $R_{\gamma_{344}}$  of  $^{152}\text{Eu}$ .

The characteristics of the considered  $\beta^-$ -transitions and the obtained values of  $R_\beta$  are represented in the table. It contains the energies of excited states after the  $\beta^-$ -decay of  $E_{lev}$ , limiting energies  $E_\beta^{lim}$  of  $\beta^-$ -particles, their averaged energies  $\bar{E}_\beta$ , the averaged velocities  $\bar{v}_\beta$  of  $\beta^-$ -particles, and the values of  $R_\beta$  calculated as the weighted-mean of several values. The energies of  $\gamma$ -rays, which are used to determine  $R_\beta$ , are represented in the last column.

Next we dwell on making corrections for the values of  $R_\beta$  marked in the table with two asterisks (\*\*). These corrections are required as  $\beta^-$ -particles with energies  $\bar{E}_\beta < 50$  keV are appreciably absorbed during the passage through the organic film with a thickness of  $2 \mu\text{m}$  upon the  $^{152}\text{Eu}$  and  $^{154}\text{Eu}$  decays or through the  $\text{TiO}_2$  film with a thickness of  $0.2 \mu\text{m}$  upon the decay of  $^{226}\text{Ra}$ . The corrections have to be made for the measured values of  $R_\beta$ , since we determine the number of  $\beta^-$ -particles by the number of detected  $\gamma$ -quanta, which practically do not loss their energy in the films. These corrections result in the increase of  $R_\beta$  from 6 to 24 % depending on the energy. The exception to the rule is the correction for  $R_\beta$  upon the  $^{210}\text{Pb}$  decay. In this case,  $\beta^-$ -particles are absorbed in the  $\text{TiO}_2$  layer so intensively that the values of  $R_\beta$  obtained experimentally have to be increased by a factor of 4.2.

During the experiments with  $^{226}\text{Ra}$ , we also observe  $e_0$ -electrons upon the  $\alpha$ -decay to the 186-keV level of  $^{222}\text{Rn}$ . It is established that  $R_\alpha = R_{\gamma_{186}} = 0.23 \pm 0.03$ . We estimated  $\varepsilon_0 \Omega \approx 0.066$ , since the  $e_0$ -electron yield for this  $\alpha$ -transition was estimated by us as  $Y_\alpha = 3.5e_0$  per  $\alpha$ -decay. Then the  $e_0$ -electron yields for the  $\beta^-$ -decay represented in the table are within the range  $(0.05 \div 0.35)e_0$  per one  $\beta^-$ -decay of the given type.

### 4. Discussion

The dependences of the detection probability for  $e_0$ -electrons per the detected  $\beta^-$ -decay event on the velocity of  $\beta^-$ -particles of the investigated isotopes are represented in Fig. 3. The points of the dependence  $R_\beta(\bar{v})$  have different statistical accuracies, as different  $\beta^-$ -decay branches and, in turn,  $\gamma$ -transitions have different intensities. The dashed line is the dependence  $R_\beta \sim \bar{v}_\beta^{-1}$ . Having compared the position of experimental points

relative to the lines, we can conclude that the probability of the atom ionization by  $\beta$ -particle passing near it is described by this dependence rather well.

For the  $\alpha$ -decay, we observed the same dependence of the  $e_0$ -electron yield on the velocity of  $\alpha$ -particles, i.e.

**Characteristics of the studied  $\beta$ -transitions, probabilities of detection of  $e_0$ -electron induced by them, and the energies of the used  $\gamma$ -transitions**

$E_{lev}$	$E_{\beta}^{lim}, \text{keV}$	$\bar{E}_{\beta}, \text{keV}$	$\bar{V}_{\beta}, 10^{10} \text{ cm/s}$	$R_{\beta}$	$E_{\gamma}, \text{keV}$	
Source $^{152}\text{Eu} \xrightarrow{\beta^-} E_{lev} \text{ } ^{152}\text{Gd}$						
1643	176	48	1.22	0.0135(13)**	1299	
1434	385	114	1.73	0.0090(5)**	1090	
1123	696	221	2.15	0.0071(2)	779	
—	—	319	2.36	0.0067(1)	344	
755	1064	367	2.44	0.0066(6)	411	
Source $^{154}\text{Eu} \xrightarrow{\beta^-} E_{lev} \text{ } ^{154}\text{Gd}$						
105*	141	38	1.10	0.0208(36)**	105	
86*	160	44	1.17	0.0208(28)**	86	
1720	258	72	1.45	0.0179(7)**	592,723,1597	
1617	361	105	1.68	0.0142(15)**	1246,1494	
1397	581	180	2.02	0.0116(7)	444,582,692,1274	
—	—	265	2.26	0.0111(10)	123	
1128	850	280	2.29	0.0103(7)	757,1005	
Source $^{226}\text{Ra} \text{ } ^{210}\text{Pb} \xrightarrow{\beta^-} E_{lev} \text{ } ^{210}\text{Bi}$						
46	17	11**	0.60**	0.023(5)**	46	
Source $^{214}\text{Pb} \xrightarrow{\beta^-} E_{lev} \text{ } ^{214}\text{Bi}$						
839	185	50	1.24	0.0090(39)**	839	
352	672	207	2.11	0.0049(8)	352	
295	729	228	2.17	0.0048(8)	295	
Source $^{214}\text{Bi} \xrightarrow{\beta^-} E_{lev} \text{ } ^{214}\text{Po}$						
2448	822	261	2.25	0.0049(13)	2448	
2204	1066	352		0.0046(10)		2204
2119	1151	385		0.0043(11)		
1847	1423	492	2.58	0.0035(8)	1238; 1847	
—	—	516		0.0045(8)		609
1764	1506	525	2.61	0.0040(9)	1764	
1730	1540	538		0.0038(9)		1120
1543	1727	614	2.67	0.0038(9)	934	
1378	1892	681	2.71	0.0036(8)	768; 1378	

**R e m a r k.** Data are given in the ascending order of averaged  $\beta$ -particle velocity. \* — Values correspond to the decay of  $^{155}\text{Eu}$  which is present in the source as an impurity. \*\* — The values corrected for the absorption of  $\beta$ -particles from  $^{152}\text{Eu}$  and  $^{154}\text{Eu}$  or  $^{226}\text{Ra}$  by the aluminum-coated or  $\text{TiO}_2$  film, respectively. Hereinafter, the data enclosed in brackets are averaged.

proportional to  $v_{\alpha}^{-1}$  [6,7]. In this paper, we define more precisely the result obtained in [6] that the  $e_0$ -electron yield  $Y_{e_0}$  for the source with the inactive near-surface layer under the  $\alpha$ -decay of  $^{224}\text{Ra}$  is higher by a factor of  $(46 \pm 4)$  as compared to that under the  $\beta$ -decay of  $^{212}\text{Pb}$ . Both isotopes are belong to the same chain of the  $^{232}\text{U}$  decay and measured simultaneously ( $^{232}\text{U}$  is an impurity in the OSAI source  $^{233}\text{U} + ^{238}\text{Pu} + ^{239}\text{Pu}$ ). Having assumed that  $Y_{e_0} \sim Z_p^2 v_p^{-1}$ , where  $Z_p$  and  $v_p$  are, respectively, the charge and velocity of an emitted particle, we obtain the yield ratio to be equal to 41 for these two decays that is close to the value obtained

experimentally (the charge ratio is 4:1, the values of velocities differ by an order:  $1.62 \times 10^9$  and  $1.65 \times 10^{10}$  cm/s). Next we give another example from this work. The ratio of  $e_0$ -electron yields obtained experimentally for the  $\alpha$ -decay of  $^{226}\text{Ra}$  ( $v_{\alpha} = 1.46 \times 10^9$  cm/s) and  $\beta^-$ -decay of  $^{214}\text{Pb}$  with an end point energy of 672 keV ( $v_{\beta} = 2.11 \times 10^{10}$  cm/s, see the table) equals 52(5), while it equals 58 for the considered dependence. These values equal  $(10 \pm 3)$  and 16 for the  $\beta$ -decay of  $^{210}\text{Pb}$  with  $E_{\beta}^b = 17$  keV ( $\bar{v}_{\beta} = 6.0 \times 10^9$  cm/s). The agreement between these two values can be considered as satisfactory, since the correction for the absorption of  $\beta$ -particles in the

TiO<sub>2</sub> film is large in this case.

Returning to formula (1), we can conclude the following. Under the same experimental conditions, the particles with various charges passing through the near-surface layer create the equal charge of  $\Delta Ze$  in it, but the probability for this charge to be created depends on the probability of ionization of separate atoms of the target  $P(z_p, v_p)$ .

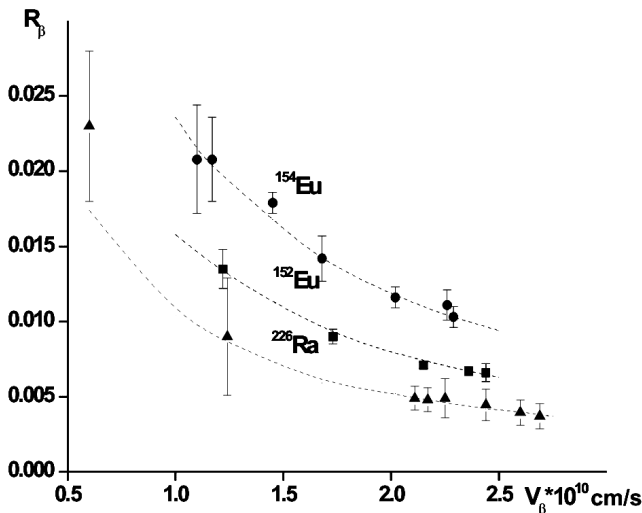


Fig. 3. Dependence of the detection probability  $R_\beta$  of  $e_0$ -electrons per one detected event of the  $\beta$ -decay on the averaged velocities of  $\beta$ -particles for various decay branches of the studied isotopes: circles —  $^{152}\text{Eu}$ , squares —  $^{154}\text{Eu}$ , and triangles —  $^{226}\text{Ra}$ . Dashed lines are the dependences of  $R_\beta$  on  $v_\beta^{-1}$ .

In [7], we studied the yield of  $e_0$ -electrons from the surface of various targets upon the bombardment of them by  $\alpha$ -particles emitted in the decay of  $^{226}\text{Ra}$ . The analysis of the obtained values of  $Y_{e_0}$  was carried out with the use of the quantum-mechanical ideas of a sudden atom excitation by an  $\alpha$ -particle passing near it [9]. In the approximation of the sudden perturbation, the condition  $a\omega_{nm}v^{-1} \leq 1$  has to be satisfied, where  $a$  is the atom radius,  $\omega_{nm}$  is the frequency of the transition from the state  $m$  to the state  $n$ . The velocity  $v_p \sim 10^9$  cm/s for  $\alpha$ -particles emitted in the radioactive decay, and the condition of suddenness is satisfied at the low transition energies  $\hbar\omega_{nm}$ . All the more, it is valid for the averaged velocity of  $\beta^-$ -particles. It is possible that the atom ionization under the  $\beta^-$ -decay has to be considered in the approximation of suddenness.

In monograph [9], the electron excitation in the approximation of suddenness was considered for both negative and positive particles. In [10], we established that the yields  $Y_{e_0}$  are close for the  $\beta^-$ -decay of  $^{59}\text{Fe}$

and  $\beta^+$ -decay of  $^{65}\text{Zn}$  which have the approximately equal averaged energies and velocities of  $\beta$ -particles. These active nuclei were in the Al substrate of a  $^{193}\text{Pt}$  source as impurities, and their yields were measured simultaneously. Thus, the perturbations caused by the particles with charges of  $\pm 1$  act on electrons in the same way, by creating the same charge under the ionization in the near-surface layer.

Thus, the near-zero electrons are easily observed, and therefore they can be used to investigate the processes of ionization.

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#### ДОСЛІДЖЕННЯ ВИХОДУ $e_0$ -ЕЛЕКТРОНІВ З ПОВЕРХНІ ТОНКИХ ПЛІВОК ПРИ БОМБАРДУВАННІ ЇХ $\beta$ -ЧАСТИНКАМИ ВІД РАДІОАКТИВНИХ ДЖЕРЕЛ $^{152}\text{Eu}$ , $^{154}\text{Eu}$ І $^{226}\text{Ra}$

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Резюме

Методом  $(e\gamma)$ -збігів проведено дослідження виходу  $e_0$ -електронів з поверхні тонких плівок при бомбардуванні їх  $\beta$ -частинками від радіоактивних джерел  $^{152}\text{Eu}$ ,  $^{154}\text{Eu}$  і  $^{226}\text{Ra}$ . Встановлено, що вихід  $e_0$ -електронів обернено пропорційний швидкості налітаючих  $\beta$ -частинок. Це означає, що ступінь іонізації атомів мішені пропорційний часу дії збурення, спричиненого  $\beta$ -частинками, що пролітають повз них.