SHAKE-OFF FOR CONDUCTIVITY ELECTRONS IN METALS CAUSED BY NUCLEAR DECAY

A.YA. DZYUBLIK, V.YU. SPIVAK

Institute for Nuclear Research, Nat. Acad. Sci. of Ukraine
(47, Nauky Ave., Kyiv 03028, Ukraine; e-mail: dzyublik@ukr.net)

UDC 539

We have analyzed the emission of conduction electrons from metals caused by the sudden alteration of a nuclear charge as a consequence of any nuclear decay. The refraction of the electron wave at the crystal surface is taken into account. It is shown that the energy distribution of ejected shake-off electrons contains a peak at an energy of the order of 1 eV. The calculated energy spectrum of shake-off electrons is consistent with experimental data.

1. Introduction

As far as in 1946, Migdal [1] and Feinberg [2] indicated that the alteration of a nuclear charge after the nuclear decay is apprehended by atomic electrons as a sudden perturbation of the Coulomb field, in which they move. As a result, any atomic electrons can leave their bound states in the atom and fly away. They are called shake-off electrons. Since then different aspects of the shake-off process have been studied both theoretically and experimentally [3–13] (see also reviews [14–17]). The probability of the electron transition from the K or L level to ones of the continuous spectrum, caused by the β decay of a nucleus, has been calculated in [1–4]. In those papers, the wave function $\psi_i(\mathbf{r})$ of the initial K state describes the motion of an electron in the Coulomb field of the point charge Ze, and the final wave function $\psi_f(\mathbf{r})$ describes the motion of the shake-off electron in the Coulomb field of the charge $Z'e$, where $Z' = Z + \Delta Z$. For the β^- decay, the electron capture, and the α decay, $\Delta Z = +1, -1$, and -2, respectively. The transition probability equals the squared modulus of the overlapping integral $< \psi_f(\mathbf{r})|\psi_i(\mathbf{r}) >$ multiplied by the density of final electron states in the continuous spectrum. Note that time-dependent perturbation theory provides the same shake-off probability if the alteration of the Coulomb field can be treated as a small perturbation of the initial field Ze^2/r, i.e. if $Z \gg 1$. Below we use just such an approximation.

It have been shown in [8] that the energy distribution of shake-off electrons contains the peak at the kinetic energy $E \approx 1$ eV which is followed by a monotonous lowering of the intensity with increase in E (see Figs. 1–3). Although Levinger [3] predicted this tail in the spectrum of shake-off electrons, he did not mention the appearance of the low-energy peak.

Here, we will concentrate on the energy and angular dependences of emitted shake-off electrons and analyze the role of the crystal environment in the shake-off process. Evaluations (see, e.g., [18]) show that the shake-off probability rapidly grows with lowering the transition energy of the electron from the initial bound state to the continuous spectrum. Therefore, the main contribution to the measured spectra of shake-off electrons is provided

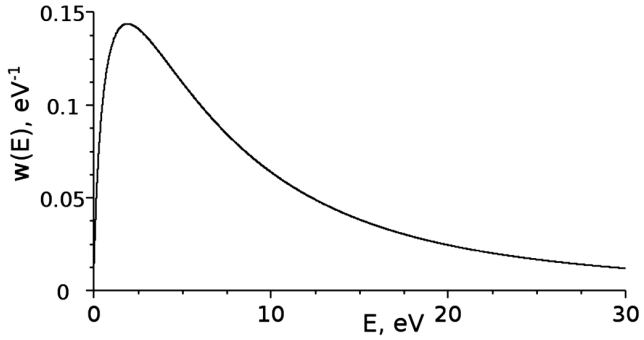

Fig. 1. Energy distribution of shake-off electrons integrated over angles for the screening radius $r_0 = 1$ Å

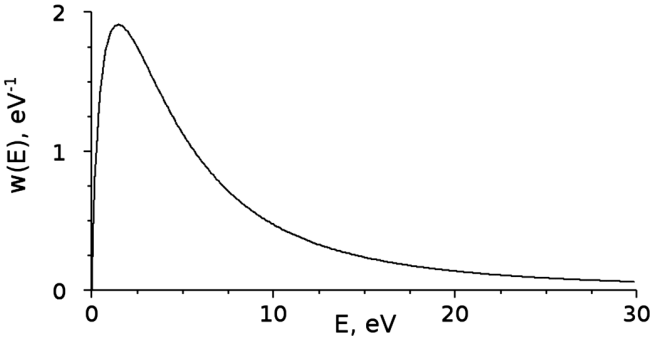

Fig. 2. Energy distribution of shake-off electrons integrated over angles for the screening radius $r_0 = 2$ Å

by electrons which are located on the upper-lying levels prior to the nuclear decay. In other words, the experimentalists "see" mainly the electrons which escape from valent levels. Recent shake-off experiments [8–13] have been carried out with metal radioactive sources. There the valence electrons become free carriers of charge, by moving in the conduction band. We consider here the shake-off process just for the conduction (valence) electrons of a metal.

It is also worth to mention work [6], in which an additional peak was discovered in the spectrum of conversion electrons, whose shape is a mirror image of the shake-off electron spectrum. In this case, the nuclear energy is shared among conversion and shake-off electrons. This effect has been explained in [19] by describing the whole process as a two-step decay of the coupled "nucleus + atomic electrons" system.

2. Sudden Perturbation

The conduction (valence) electrons of a metal, moving in a periodic crystal potential $U(\mathbf{r})$, are described by the Bloch wave functions. We use the oversimplified model of free electrons confined in the potential well (see, e.g., [20, 21])

$$U(\mathbf{r}) = \begin{cases} -U_0 & \text{inside the crystal,} \\ 0 & \text{outside it,} \end{cases} \tag{1}$$

where $U_0 > 0$.

The perturbation of this potential suddenly arises at the time moment $t = 0$, when the nucleus abruptly changes its charge, $Z \to Z' = Z + \Delta Z$. The charge excess ΔZe is immediately screened by valence electrons which are redistributed among levels of the conduction band near the Fermi level. The rate of such screening process is much higher than that of the electron transition to any state of the continuous spectrum with energy $E>0$, since the transition energies of an electron within the conduction band are much less than the transition energy of a shake-off electron.

Thus, the perturbation arising at the time moment $t = 0$ is

$$V(\mathbf{r}, t) = v_c(\mathbf{r})\theta(t), \tag{2}$$

where $v_c(\mathbf{r})$ is a screened Coulomb interaction and

$$\theta(t) = \begin{cases} 1, & t > 0, \\ 0, & t < 0. \end{cases} \tag{3}$$

We approximate $v_c(\mathbf{r})$ by the function

$$v_c(\mathbf{r}) = -\frac{\Delta Ze^2}{r} \exp\left(-\frac{r}{r_0}\right), \tag{4}$$

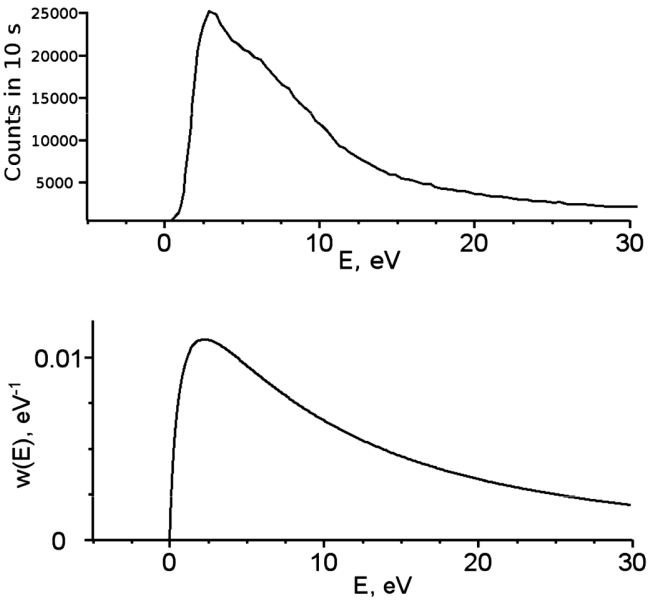

Fig. 3. Energy distribution integrated over the angle θ from 0 to $\pi/2$ for shake-off electrons ejected from a Co crystal. The results of calculations are compared with the data in [8]

where r_0 represents the screening length. Broadly speaking, the real Coulomb field of a nucleus screened by the surrounding electrons can have a much more complicated dependence on the radial coordinate r. It is described in terms of the dielectric function which has the so-called Kohn singularities (see, e.g., [20, 21]). But all this is beyond the scope of our paper.

Hereafter, we put the origin of the coordinate frame x, y, z into the radioactive nucleus and direct the axis z perpendicularly to the crystal surface having the coordinate z_s. The crystal slab occupies the region $z_s - D < z < z_s$, where D is the crystal thickness. The shake-off electrons fly into the region $z > z_s$.

The Hamiltonian for such an electron can be written down as

$$\hat{H}(\mathbf{r}, t) = \hat{H}_0(\mathbf{r}) + V(\mathbf{r}, t), \tag{5}$$

where the unperturbed Hamiltonian

$$\hat{H}_0 = -\frac{\hbar^2}{2m}\Delta + U(\mathbf{r}), \tag{6}$$

and m is the electron mass. The eigenfunctions and eigenvalues of the operator $\hat{H}_0$ are determined by the equation

$$\hat{H}_0\varphi_n(\mathbf{r}) = E_n\varphi_n(\mathbf{r}) \tag{7}$$

with the transition frequencies

$$\omega_{nm} = (E_n - E_m)/\hbar. \tag{8}$$

At the initial time moment $t = 0$, let the electron be in the state $\varphi_0(\mathbf{r})$. Then the probability of finding the electron in any final state n at $t \to \infty$ is given by the well-known formula of the time-dependent perturbation theory (see, e.g., [22]):

$$P_n(\infty) = \left| \frac{\langle n|v_c(\mathbf{r})|0\rangle}{\hbar\omega_{n0}} \right|^2. \tag{9}$$

3. Electron Waves

The conduction electrons moving in the potential well (1) are described by the wave functions

$$\psi_{\mathbf{q}}(\mathbf{r}) = \frac{1}{\sqrt{V}} e^{i\mathbf{qr}}, \tag{10}$$

where V is the crystal volume. The corresponding energy is

$$\varepsilon_{\mathbf{q}} = -U_0 + \hbar^2 q^2/2m. \tag{11}$$

The depth of the potential well equals

$$U_0 = \varepsilon_{\mathrm{F}} + A, \tag{12}$$

where $\varepsilon_{\mathrm{F}} = \hbar^2 q_{\mathrm{F}}^2/2m$ is the Fermi energy, and A is the work function. The radius of the Fermi sphere equals

$$q_{\mathrm{F}} = \left(3\pi^2 N/V\right)^{1/3}, \tag{13}$$

where N is the number of free electrons in the volume V given by

$$N = \nu(V/v_0) \tag{14}$$

with v_0 standing for the volume per one atom in the crystal and ν for the valency of atoms.

The average number of electrons on the level $\varepsilon_{\mathbf{q}}$ with a definite spin projection is determined by the Fermi distribution

$$\bar{n}(\mathbf{q}) = \left[\exp\left(\frac{\varepsilon_{\mathbf{q}} - \varepsilon_{\mathrm{F}}}{k_{\mathrm{B}}T}\right) + 1\right]^{-1}. \tag{15}$$

The electron in the final state has the energy $E > 0$ and the wave vectors $\mathbf{K}$ inside the crystal and $\mathbf{k}$ outside it. This electron is described by the wave function (see also [23])

$$\psi_{\mathbf{k}}(\mathbf{r}) = \begin{cases} e^{i\mathbf{K}_{||}\mathbf{r}}\left(e^{iK_z z} + re^{2iK_z z_s}e^{-iK_z z}\right), & z \le z_s, \\ te^{i(K_z - k_z)z_s}e^{i\mathbf{kr}}, & z \ge z_s, \end{cases} \tag{16}$$

where $\mathbf{K}_{||}$ is the component of the vector $\mathbf{K}$ along the plane (x, y), while r and t represent the amplitudes of reflected and transmitted waves, respectively:

$$r = \frac{K_z - k_z}{K_z - k_z}, \quad t = \frac{2K_z}{K_z + k_z}. \tag{17}$$

Matching two waves (19) at the boundary $z = z_s$, one has the equality $\mathbf{K}_{||} = \mathbf{k}_{||}$ which is nothing but the refraction law

$$K \sin\theta_0 = k \sin\theta, \tag{18}$$

where θ_0 denotes the angle between $\mathbf{K}$ and the axis z, and θ is the angle between $\mathbf{k}$ and the same axis.

The energy of the shake-off electron

$$E = \frac{\hbar^2 k^2}{2m} = \frac{\hbar^2 K^2}{2m} - U_0. \tag{19}$$

Therefore, the normal component of the wave vector inside the crystal is

$$K_z = \sqrt{2m(E\cos^2\theta + U_0)}/\hbar. \tag{20}$$

This helps us to rewrite the transmission coefficient from the crystal to the vacuum as

$$T(E) = \frac{4K_z k_z}{(K_z + k_z)^2} \tag{21}$$

or, in the form more comfortable for estimations, as

$$T(E) = \frac{4(1 + U_0/E\cos^2\theta)^{1/2}}{[1 + (1 + U_0/E\cos^2\theta)^{1/2}]^2}. \tag{22}$$

The effective potential has the imaginary part U_0'' which determines the attenuation of a wave inside the crystal. As a consequence,

$$K_z = K_z^{'} + i\mu/2. \tag{23}$$

Here, the attenuation coefficient

$$\mu = \sigma_{in}/v_0 \cos\theta_0, \tag{24}$$

and σ_{in} is the inelastic scattering cross section of electrons by one ion of the crystal. According to (17), the intensity of the reflected wave $\sim e^{-iK_z z}$ about the radioactive ion will be reduced by a factor of $e^{-2\mu z_s/\cos\theta_0}$. This enables us to keep only the direct wave $e^{i\mathbf{Kr}}$ in $\psi_f(\mathbf{r})$, when calculating the matrix element V_{fi}.

4. Shake-off Probability

Simple calculations provide us with the transition probability $P_{\mathbf{q}\to\mathbf{k}}$ of the electron from the initial state (10) into the final one (16):

$$P_{\mathbf{q}\to\mathbf{k}} = \frac{1}{V}\frac{1}{(E-\varepsilon_{\mathbf{q}})^2}\left[\frac{4\pi\Delta Ze^2r_0^2}{1+(\mathbf{K}-\mathbf{q})^2r_0^2}\right]^2. \tag{25}$$

Here, the screening length is estimated with the aid of the expression [20,21]

$$r_0^2 = \frac{\varepsilon_{\mathrm{F}}}{6\pi e^2 n_0}, \tag{26}$$

where n_0 is the concentration of the conduction electrons.

The flux density of shake-off electrons near the radioactive ion through the plane $z = \mathrm{const} \simeq 0$ equals

$$j_0 = \hbar K_z/m, \tag{27}$$

whereas the flux density outside the crystal through the plane $z = \mathrm{const} > z_s$ is

$$j_{\mathrm{out}} = e^{-\mu z_s/\cos\theta_0}|t|^2\hbar k_z/m. \tag{28}$$

The flux of electrons outside the crystal is reduced as compared with the flux near the origin of the shake-off electrons by a factor of j_{out}/j_0. Hence, the probability of finding a shake-off electron in the outgoing beam at $z > z_s$ will be

$$P^{\mathrm{out}}_{\mathbf{q}\to\mathbf{k}} = (j_{\mathrm{out}}/j_0)P_{\mathbf{q}\to\mathbf{k}} \tag{29}$$

or

$$P^{\mathrm{out}}_{\mathbf{q}\to\mathbf{k}} = e^{-\mu z_s/\cos\theta_0}T(E)P_{\mathbf{q}\to\mathbf{k}}. \tag{30}$$

Here, $\cos\theta_0$ may be expressed, according to (19), (20), in terms of the experimentally measurable angle θ:

$$\cos\theta_0 = \left(1 - \frac{E}{E+U_0}\sin^2\theta\right)^{1/2}. \tag{31}$$

The number of electrons emitted from the crystal into a unit energy interval ΔE and a unit solid angle $\Delta\Omega_{\mathbf{k}}$ after the decay of a single nucleus located at the distance z_s from the surface amounts to

$$\frac{\partial^2 N_e^{(1)}(E,\theta)}{\partial E\partial\Omega_{\mathbf{k}}} = \sum_{\mathbf{q}}\bar{n}(\mathbf{q})P^{\mathrm{out}}_{\mathbf{q}\to\mathbf{k}}\varrho(E), \tag{32}$$

where $\varrho(E)$ stands for the density of final states:

$$\varrho(E) = (2\pi)^{-3}(mk/\hbar^2). \tag{33}$$

We replace the summation in (33) over $\mathbf{q}$ by integration:

$$\sum_{\mathbf{q}} \longrightarrow \frac{V}{(2\pi)^3}\int d\mathbf{q}. \tag{34}$$

It is useful to introduce the spherical coordinates q, β, α of the vector $\mathbf{q}$ with β specifying the angle between the vectors $\mathbf{q}$ and $\mathbf{k}$. Then the integration over β and α is easily performed by using the identity

$$(\mathbf{K}-\mathbf{q})^2 = K^2 + q^2 - 2Kq\cos\beta, \tag{35}$$

giving us the distribution of shake-off electrons emitted from the crystal:

$$\frac{\partial^2 N_e^{(1)}(E,\theta)}{\partial E\partial\Omega_{\mathbf{k}}} = w(E,\theta)\exp\left\{-\mu(E)z_s/\cos\theta_0\right\}, \tag{36}$$

where $w(E,\theta)$ is the distribution of electrons, whose emission is ensured by the decay of nuclei lying on the surface ($z_s \approx 0$).

Introducing the notations

$$\tilde{K} = Kr_0, \quad \tilde{q} = qr_0, \quad \alpha = \hbar^2/2mr_0^2,$$

$$\tilde{n}(\tilde{q}) = \left[\exp\left\{-\frac{\alpha\tilde{q}^2 - \varepsilon_{\mathrm{F}}}{k_{\mathrm{B}}T}\right\} + 1\right]^{-1}, \tag{37}$$

we write down the result in the form

$$w(E,\theta) = T(E,\theta)\frac{1}{2\alpha}\sqrt{\frac{E}{\alpha}}\left(\frac{e^2}{r_0\alpha}\right)^2\frac{1}{\pi^3}\times$$

$$\times\int_0^{\tilde{q}_{\max}}\frac{\tilde{n}(\tilde{q})\tilde{q}^2d\tilde{q}}{[\tilde{K}^2-\tilde{q}^2]^2[1+2(\tilde{K}^2+\tilde{q}^2)+(\tilde{K}^2-\tilde{q}^2)^2]}. \tag{38}$$

Here, the upper limit of integration $\tilde{q}_{\max} = q_{\max}r_0$. It should satisfy the condition $q_{\max} - q_{\mathrm{F}} \gg k_{\mathrm{B}}T$. Then we take all the conduction electrons into account, since $\bar{n}(q_{\max}) \approx 0$. Moreover, $q_{\max}$ should be less than $\sqrt{2mU_0}/\hbar$ in order to avoid the divergence of the integrand.

5. Yield of Electrons

While the previous theoretical papers have dealt only with the emission of shake-off electrons from inner levels of isolated atoms and completely ignored the role of environment, we considered this process for conduction electrons in metals. Note that electrons from all levels of the conduction band take part in the shake-off process. Therefore, our final result (33) contains the sum over all the levels od the conduction band.

To illustrate our results, we calculated the energy distribution $w(E,\theta)$ of shake-off electrons ejected from a copper crystal. In this case, the volume of the elementary cell $v_0 = 1.2 \times 10^{-23}$ cm^3, the screening radius $r_0 = 0.55$ Å [20, 21], the Fermi energy $\varepsilon_{\rm F} = 7.0$ eV, and the work function $A = 4.4$ eV. Therefore, the depth of the potential well for Cu is $U_0 = 11.4$ eV. In order to understand how the energy distribution of shake-off electrons for the copper depends on the screening radius, we calculated $w(E)$ integrated over the angles, taking the screening radii $r_0 = 1$ and 2 Å, when $|\Delta Z| = 1$. These results are displayed in Figs. 1 and 2. We see that, while the shape of the curve $w(E)$ slightly depends on r_0, the integral yield of shake-off electrons very rapidly grows with increase in r_0. Namely, for $r_0 = 1$ Å, the integral yield is 1.8 electrons per one nuclear decay at the crystal surface, whereas it is equal to 15 electrons per one decay if $r_0 = 2$ Å.

We calculated also the energy distribution integrated over the angle θ from 0 to $\pi/2$ for shake-off electrons ejected from the cobalt crystal, where $r_0 = 0.55$ Å, $\varepsilon_{\rm F} = 7.36$ eV, $A = 5$ eV. These calculations are compared with data of [8] in Fig. 3. We see that they well correlate with each other.

The main peculiarity of such energy spectra of shake-off electrons is the existence of the peak at an energy E of the order of 1 eV. Its appearance can be explained as follows. At $E \to 0$, the density of final states $\rho(E) \sim \sqrt{E}$ also tends to zero. At the same time, the transition probability $P_{\mathbf{q}\to\mathbf{k}}$ has no singularity at the point $E = 0$. As a result, the energy distribution $w(E)$, being proportional to $\rho(E)$, vanishes at $E = 0$. On the other hand, the denominator of the integrand in (33) equals the product of two polynomials in $K^2 = 2m(E + U_0)/\hbar^2$. They provide a monotonous decrease of the electron distribution $w(E)$ with increase in the energy E. Thus, the function $w(E)$ rapidly grows from zero to a maximum value and then slowly falls down (long tail), when E varies from 0 to ∞.

The angular dependence of the integral electron yield $\int_0^\infty w(E,\theta)dE$ from a Cu crystal as a function of the angle θ is drawn in Fig. 4. This function goes to zero as θ approaches $\pi/2$, since the transmission coefficient of the electron wave from the crystal to vacuum is characterized by the same behavior.

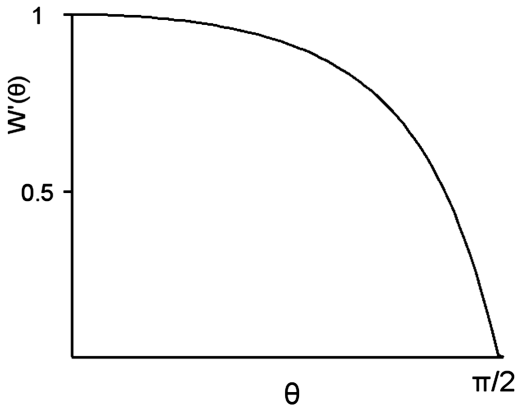

Fig. 4. Angular dependence of the electron yield from a Cu crystal

The results of calculations of the shake-off electron yield with regard for the attenuation of the electron wave inside a crystal will be presented elsewhere.

We are thankful to A.I. Feoktistov, V.T. Kupryashkin, V.M. Pugach, V.I. Sugakov, and V.N. Malnev for helpful discussions.

1. A. Migdal, J. Phys. (USSR) **4**, 449 (1941).
2. E.L. Feinberg, J. Phys. (USSR) **4**, 424 (1941).
3. J.S. Levinger, Phys. Rev. **90**, 11 (1953).
4. H. Primakoff and F.T. Porter, Phys. Rev. **89**, 930 (1953).
5. M.S. Freedman, F.T. Porter, F.J. Wagner, and P.P. Day, Phys. Rev. **108**, 836 (1957).
6. F.T. Porter, M.S. Freedman, and F. Wagner, jr., Phys. Rev. C **3**, 2246 (1971).
7. B.V. Bobykin, S.K. Lyubov, and Yu.A. Nevynni, Zh. Tekh. Fiz. **58**, 1524 (1988).
8. A. Kovalik, V.M. Gorozhankin, Ts. Vylov *et al.*, J. Electron Spectrosc. and Related Phenomena **95**, 1 (1998).
9. V.T. Kupryashkin, L.P. Sydorenko, A.I. Feoktistov, and I.P. Shapovalova, Izv. Russian Acad. Nauk (Ser. Phys.) **67**, 1467 (2003).
10. V.T. Kupryashkin, L.P. Sydorenko, A.I. Feoktistov, and I.P. Shapovalova, Izv. Russian Acad. Nauk (Ser. Phys.) **68**, 1208 (2004).
11. A.A. Valchuk, V.T. Kupryashkin, L.P. Sidorenko *et al.*, Ukr. J. Phys. **51**, 5 (2006).
12. V.T. Kupryashkin, L.P. Sydorenko, and A.I. Feoktistov, Ukr. J. Phys. **51**, 126 (2006).
13. A.I. Feoktistov, A.A. Valchuk, V.T. Kupryashkin, and L.P. Sidorenko (to appear).

14. A.M. Dychne and G.L. Yudin, Uspekhi Fiz. Nauk **121**, 157 (1977)

15. A.M. Dychne and G.L. Yudin, Uspekhi Fiz. Nauk **125**, 377 (1978)

16. I.S. Batkin and Yu.G. Smirnov, Particles and Nuclei **11**, 1421 (1980).

17. V.I. Matveev and E.S. Parilis, Uspekhi Fiz. Nauk **138**, 573 (1982)

18. A.B. Migdal, *Qualitative Methods in Quantum Theory* (Addison-Wesley, New York, 1977).

19. A.Ya. Dzyublik, in *Isomers and Quantum Nucleonics: Proceedings of the 7th AFOSR Workshop* (Dubna, June 26 – July 1, 2005), edited by S.A. Karamian, J.J. Carroll, and E.A. Cherepanov. (JINR, Dubna, 2006), pp.145-152.

20. Ch. Kittel, *Introduction to Solid State Physics* (Wiley, New York, 1995).

21. J. Ziman, *Principles of the Theory of Solids* (Cambridge University Press, Cambridge, 1979).

22. L.D. Landau and E.M. Lifshitz, *Quantum Mechanics* (Pergamon, New York, 1980).

23. V.I. Ignatovich, *Physics of Ultracold Neutrons* (Nauka, Moscow, 1986) (in Russian).

Received 11.07.07

СТРУС ЕЛЕКТРОНІВ ПРОВІДНОСТІ В МЕТАЛАХ, СПРИЧИНЕНИЙ ЯДЕРНИМ РОЗПАДОМ

О.Я. Дзюблик, В.Ю. Співак

Р е з ю м е

Проаналізовано вихід з металу електронів провідності, спричинений раптовою зміною заряду ядра внаслідок ядерного розпаду. Враховано рефракцію електронної хвилі на поверхні кристала. Показано, що енергетичний розподіл електронів струсу містить пік при енергії порядку 1 еВ. Енергетичний спектр електронів струсу відповідає експериментальним даним.