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Two-Flux Model of Charged-Particle Transport in a Condensed Material under Multiple Scattering: Average Energy Losses and Range of a Beam of Monoenergetic Electrons with Energies of 0.1 keV–1.0 MeV

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Abstract—The results of using a two-flux model of charged-particle transport in a substance are presented to describe the average energy of a monoenergetic electron beam passed through a film target with known composition and a given thickness. Formulas describing the distribution of the average energy of the electron beam over the target depth and the energy dependence of the electron-beam range for electrons with an energy of 0.1 keV–1.0 MeV are obtained. The results of calculating the electron ranges for a wide range of materials, namely, from Be to Au, are given. The particle ranges calculated using the formulas are compared with the experimental results of measuring the depth of their penetration into the target.

Keywords: elastic and inelastic multiple scattering of charged particles in condensed materials, most probable and average losses of the energy of a charged-particle beam, transport cross section and transport length of a monoenergetic electron beam in a sample, actual electron range in a material

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INTRODUCTION

The two-flux model of the multiple scattering of charged particles was proposed in [1, 2] to take into account the influence of the distribution of atomic electrons of the sample under study on the transport of a directed flux of charged particles in a condensed material. The results obtained within the framework of this model make it possible to calculate the spectra of energy losses of particles, i.e., their distributions over ranges and angles in film and solid targets. In this case, the performed results were in good agreement with the experimental results [3–5].

At the same time, for a series of important practical applications, in addition to the most probable values of parameters characterizing a moving flux of particles in the volume of the sample under study, information on their average values is required, for example, information on the average particle energy losses; this information is also required when calculating the ranges of a charged-particle beam in a sample material or the matrix correction for the stopping capability of a material during quantitative X-ray spectral microanalysis.

In this paper, we present the results of using the two-flux model to describe the average losses of a monoenergetic electron beam passed through a thin target with known composition and a given thickness. We obtained a universal formula for calculating the electron ranges R_e for wide range of electron-beam energies, namely, from 0.1 keV to 1.0 MeV. We present the results of verifying the obtained formulas by comparing the model calculations with the data of known experimental measurements of average electron losses in film targets. We present the results of calculating the electron ranges for a wide range of materials, namely, from beryllium ($_4$Be) to gold ($_{79}$Au). We compare the calculated values of R_e with those obtained using existing and widely used formulas of the Kanaya–Okayama diffusion model [6] and the power approximation of Fitting experimental measurements [7] and with many results of experimental measurements of R_e [8]. We show the obvious advantages of our new approach used to describe the average values and good achieved correspondence between our calculations and experimentally measured data on R_e.

MAIN CONCEPTS OF THE MODEL

The main concepts of the applied two-flux model concerning transport processes of the directed flux of fast electrons in a material and their energy dissipation as a result of inelastic scattering reduce to the following:

(i) Beam electrons moving in a material experience elastic and inelastic scattering events. To describe the average influence of these processes on the spatial distribution of primary electrons, in this model, we used a universal parameter – the transport length L_{tr} of the electron beam. It characterizes the range of primary electrons, after which there is no directed motion of the particle beam in the sample; i.e., all directions become equally probable for particles. In a material with the density of atoms n_0, the transport length L_{tr} of the electron beam with the energy E_0 is defined by the transport cross section σ_{tr} of electrons in accordance with the formula:

$$L_{tr} = 1/(n_0\sigma_{tr}),$$

where the transport cross section $\sigma_{tr} = \int(1-\cos\theta)\,d\sigma$ is the cross section for primary electron scattering in the material averaged over all possible angular (θ) deviations.

If the influence of elastic and inelastic scattering channels on the process of electron—material interaction is taken into account, then we have

$$\sigma_{tr} = \sigma_{tr}^{el} + \sigma_{tr}^{inel},$$

where σ_{tr}^{el} and σ_{tr}^{inel} are the transport lengths of electrons along the elastic and inelastic scattering channels, respectively. When calculating σ_{tr}^{el} and σ_{tr}^{inel}, formulas for the transport cross sections given in [9, 10] are used in the model.

(ii) The model assumes the presence of two fluxes of primary beam electrons in the sample volume. It is known that a large portion of electrons in an atom with the atomic number Z are located at distances on the order of $a_B Z^{-1/3}$ (a_B is the Bohr radius and is 0.529 Å) from the nucleus. Numerical calculation shows that half the total electric charge of the atom is inside the sphere with a radius of $r_{0.5} = 1.33a_B \times Z^{-1/3}$ [11]. In $_{13}$Al, $r_{0.5} = 0.299$ Å for an atomic radius of $r_{at} = 1.43$ Å [12]; in $_{29}$Cu, $r_{0.5} = 0.229$ Å for $r_{at} = 1.28$ Å [12]; in $_{47}$Ag, $r_{0.5} = 0.195$ Å for $r_{at} = 1.44$ Å [12]; and, in $_{79}$Au, $r_{0.5} = 0.164$ Å for $r_{at} = 1.44$ Å [12]. Such a nonuniform charge distribution in an atom for a directed flux of charged particles, the dimensions of each of which can be neglected and the scattering of each of which at the atom is determined by the impact parameter, must lead inevitably to separation of the flux into two groups of primary particles. Therefore, in thin films whose thickness x is much smaller than the transport range L_{tr}, the inelastic one-particle interaction of a fast charged particle occurs with a part of half the total atomic charge that is located beyond the region with the radius $r_{0.5}$ with a larger probability. This leads to the formation of two groups of primary particles: those that experienced inelastic scattering only at $Z/2$ external atomic electrons (the second group) and the flux of particles that lose their energy with the participation of all Z atomic electrons (the first group), as shown in Fig. 1. It is seen that as the layer thickness increases and, accordingly, the particle range in the material increases (because of the large-angle elastic scattering of particles), the degree of participation of atomic electrons of the screening region increases in the total energy losses, and the fraction of particles losing their energy with the participation of only external atomic electrons decreases. After the particles travel a distance exceeding L_{tr}, only particles of the first group remain, their energy losses are due to all Z atomic electrons. The possibilities and the effectiveness of such an approach for describing the energy spectra of the beam of fast electrons moving in the material were shown in [1–5]. The results obtained within the framework of this model (solution of the one-dimensional transport equation [3] and the formulas for the most probable energy losses [4] and the full widths of the energy spectra at half maxima (FWHMs) [3, 5]) make it possible to calculate the spectra of energy losses of particles passed through a film model with good correspondence with the experimental results, as shown in Fig. 2.

DISTRIBUTION OF THE AVERAGE ENERGY OF AN ELECTRON BEAM OVER THE DEPTH OF A SOLID TARGET

Using first principles, the author of [4] obtained a formula describing the dependence of the squared, most probable energy E_p^2 of a beam of electrons having the energy E_0, which passed through a thick film target and experienced an average number n of inelastic interactions in it:

$$E_0^2 - E_p^2 = nJ^2 \ln\left(\frac{n}{e}\right), \quad (1)$$

where J is the average excitation energy of target atomic electrons and e is the base of natural logarithms.

In this case, the average number n of inelastic interactions in (1) is calculated from the following rather simple relation

$$nJ^2 = 4\pi q^4 n_0 Z x, \quad (2)$$

in which the right-hand side, i.e., $4\pi q^4 n_0 Z x$, is the so-called "Bohr dispersion" for particles with an electric charge that is equal to unity, x is the film thickness, q is the elementary charge, n_0 is the number of atoms in the unit material volume, and Z is the average atomic number of the material.

In accordance with the approach used in [4], the logarithm in formula (1) is due to the contribution of the statistical probability, as applied to the discrete and multiply repeated process of charged-particle energy

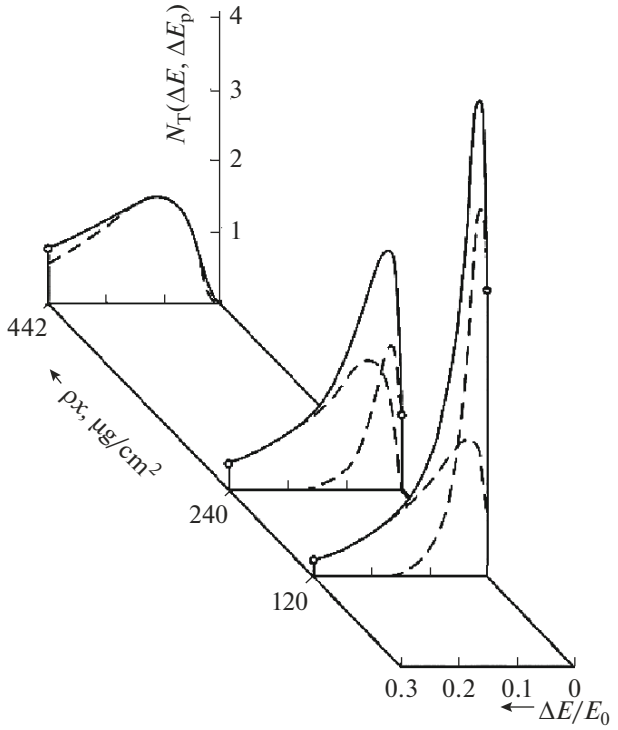

Fig. 1. Distribution of the energy losses of an electron beam with an energy of $E_0 = 18$ keV after multiple inelastic scattering in Au films with different mass thicknesses: the dashed line corresponds to the calculated contributions of the energy losses of two groups of primary electrons; and the solid line, to the experiment in [14]. The mass transport length ρL_{tr} of electrons with the same energy in gold is 174 μg/cm^2.

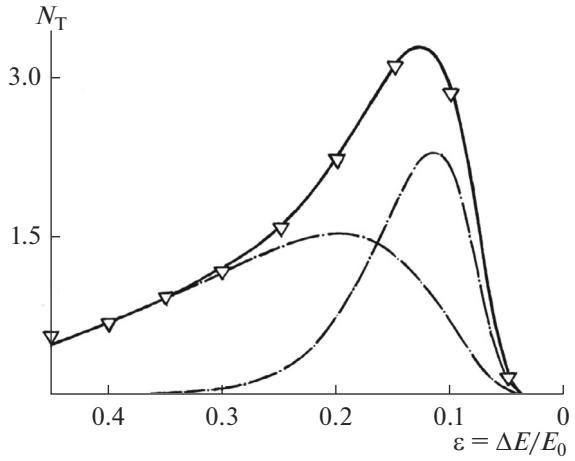

Fig. 2. Spectrum of energy losses of an electron beam with $E_0 = 18$ keV passed through an Al film with a mass thickness of 300 μg/cm^2 ($\rho L_{tr} = 443$ μg/cm^2): the dashed line corresponds to the calculated contributions of the energy losses of two groups of primary electrons; and the solid line, to the calculated total distribution (the experiment in [14]).

losses upon inelastic scattering. On the other hand, the quantity n can be represented as the ratio of the probable single energy loss $\varepsilon = n\varepsilon_{min}$ of primary electrons to the minimum probable energy loss ε_{min}. And in this case, for electrons that passed through a layer with the thickness x, $\ln(\varepsilon/\varepsilon_{min}e)$ is the result of averaging over the probable energy losses for the case of multiple scattering. As x increases, the contribution of the logarithmic term increases. Obviously, extending the range of energy-loss averaging toward the maximally possible single energy losses $\varepsilon_{max} = (n_{max}\varepsilon_{min})$, we can obtain an expression for the squared average energy E_m^2 as a function of the target thickness:

$$E_0^2 - E_m^2 = 4\pi q^4 n_0 Zx\ln(\varepsilon_{max}/\varepsilon_{min}e). \quad (3)$$

It is seen that the expression for the squared average energy in the solid target differs from the relation for the squared, most probable energy only in the form of its logarithmic term, much as the Bethe formula differs from the Landau formula. If the fact that ε_{min} for fast electrons is defined as $J^2/2E_0$ [4] and, as is known, the parameter J is ~ Z [13] is taken into account, then $\varepsilon_{min} \sim Z^2/E_0$. It is natural to assume that ε_{max} also depends on Z analogously, i.e., $\varepsilon_{max} \sim Z^2E_0$. Therefore, the logarithmic term for the average energies is defined only by the dependence on the initial electron energy E_0, and, with a certain approximation, expression (3) can be represented as

$$E_0^2 - E_m^2 = 4\pi q^4 n_0 Zx\ln(E_0^2/C_m^2), \quad (4)$$

where the universal constant C_m turns out to be almost independent of the target material and is $C_m \approx 790$ eV.

Thus, unlike the most probable energy, the logarithm for the average energy is defined by the primary energy E_0 and the universal parameter C_m. Therefore, the difference between the squared primary energy and average energy of the electron beam depends on the target thickness linearly. This is confirmed well when comparing the calculations of E_m^2, carried out in accordance with (4) and the experimental results of measuring E_m^2 in classical paper [14] for a series of materials that are shown in Fig. 3.

We note three undoubted advantages of the obtained relation, which describes the sample-depth distribution of the average beam energy and makes it possible to calculate, in a convenient form, the distributions of the average energy losses for quantitative electron-probe methods for studying materials and products based on them. First, unlike the model of continuous energy losses (the continuous slowing-down approximation), the statistical probability of the discrete process of multiple electron scattering in a material is taken into account completely. Second, the average energy E_m and its derivative dE_m/dx depend on

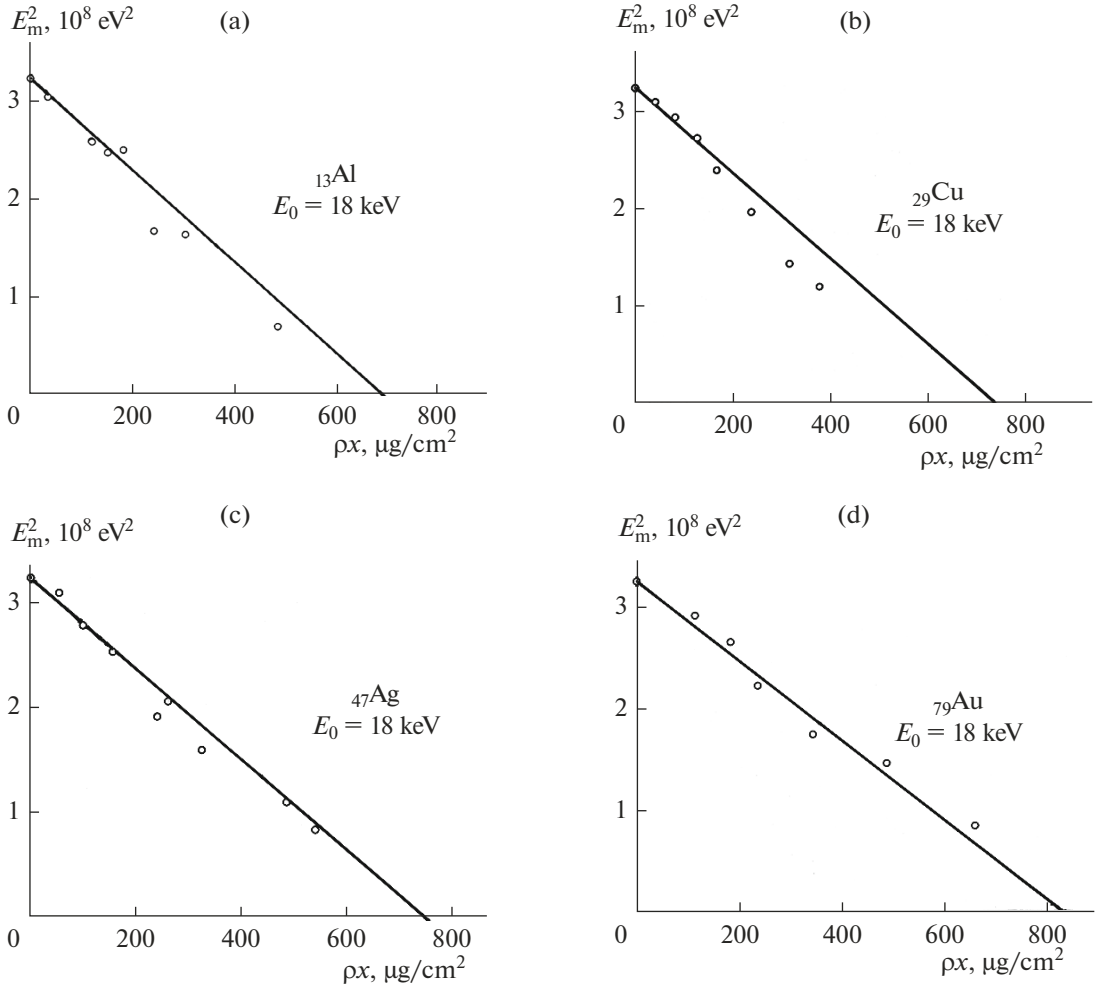

Fig. 3. Dependence of the squared average energy of passed electrons on the mass thickness of the film target for an electron beam with an energy of E_0 = 18 keV for different materials: (a) Al, (b) Cu, (c) Ag, and (d) Au.

the material layer thickness x rather than on the particle transport range s, which is convenient in many practical applications. Third, direct experimental verification of this formula, the results of which for Al, Cu, Ag, and Au are shown in Fig. 3, is admissible.

The use of the obtained formula (4) in a broader range of beam electron energies E_0 (from 0.1 keV to 1.0 MeV) is reached by introducing a relativistic correction for energies of $E_0 > 20$ keV to it and also by taking into account the dependence of the probability of the inelastic scattering of a primary electron beam with $E_0 < 3$ keV in a material on the ratio of its velocity to the average velocity of atomic electrons in accordance with a procedure that was proposed previously and used in [4]. As a result, we obtain

$$E_0^2 - E_m^2 = \begin{cases} \dfrac{4\pi q^4 n_0 Z x}{(1-\beta^2)^{1/2}} \left\{ \ln\left[\dfrac{E_0^2 \left(1-\beta^2\right)^{1/2}}{C_M^2} \right] \right\}, & \text{for } 2.62 \text{ keV} \le E_0 \le 1.0 \text{ MeV}, \\ 4\pi q^4 n_0 Z x F_M, & \text{for } E_0 \le 2.62 \text{ keV}, \end{cases} \quad (5)$$

where $F_M = 5.554\{1 - \exp[-0.1714(E_0/C_M)]\}$.

ELECTRON-BEAM RANGE. APPROBATION OF THE OBTAINED ANALYTICAL EXPRESSION

Formulas (4) and (5) can easily be used to find the most important parameter characterizing the electron—material interaction (the range R_e of electrons in the material) as a function of the distance from the surface at which the average kinetic energy of primary electrons becomes almost equal to the thermal energy, i.e., $E_m = 0$, which, as applied to expression (5), gives

$$R_e = \begin{cases} \dfrac{E_0^2(1-\beta^2)^{1/2}}{4\pi q^4 n_0 Z\left\{\ln\left[\dfrac{E_0^2(1-\beta^2)^{1/2}}{C_M^2}\right]\right\}}, & \text{for } 2.62 \text{ keV} \le E_0 \le 1.0 \text{ MeV}, \\ \dfrac{E_0^2}{4\pi q^4 n_0 Z F_M}, & \text{for } E_0 \le 2.62 \text{ keV}. \end{cases} \quad (6)$$

To study the possibility of using formulas (6) for the practical problems of electron-probe investigation methods, they were verified by comparing the quantities R_e calculated using these formulas and characterizing the energy dissipation process in the material with widely used results of the calculations of R_{K-O} of the diffusion model [6] and with many experimental measurements of R_F presented in the Fitting papers [7, 8]. As was shown in [8], the experimentally measured ranges of electrons with energies E_0 from 0.4 to 1000 keV in a material can be approximated by the following analytical expressions:

$$R_F = \frac{450}{\rho^{0.9}} E_0^{1.7} \quad \text{for an energy of } E_0 \ge 10 \text{ keV}$$

$$\text{and } R_F = \frac{900}{\rho^{0.8}} E_0^{1.3} \quad \text{for an energy of } E_0 < 10 \text{ keV},$$

where $[E_0]$ is in keV, $[\rho]$ is in g/cm^3, and $[R_F]$ is in Å.

The results of calculating parameter R_e in accordance with formula (6) and also R_F and R_{K-O} in different materials for beam electron energies of 1–50 keV are given in Table 1. It is seen that good agreement between the calculations of R_e and R_F is observed. The diffusion model makes the range R_{K-O} larger for $E_0 \ge 10$ keV and smaller for $E_0 < 1$ keV. In the entire announced range of 0.1 keV–1.0 MeV, for Be, Al, Cu, and Au, the calculated dependences of R_e on the energy E_0 ($R_e = f(E_0)$) are shown in Fig. 4 together with the results of experimental measurements of the electron ranges in these materials. It is seen that the formula obtained for R_e describes the experimental results well in the entire chosen energy range. Another important result following from the properties of this relation should be mentioned. Formula (6) also clarifies the possibility of using the power dependence on the initial energy of the form $\sim E_0^p$ to describe the electron range in the sample cross section and explains the practical impossibility of establishing a single value of this exponent p in a wide range of electron-beam energies E_0. Indeed, from the representation of the logarithmic term in formula (6) in the form $2\ln(E_0/C_m) = 2(E_0/C_m)^p$, it is easy to calculate p ($p = \{\ln[\ln(E_0/C_m)]\}/\ln(E_0/C_m)$) for each value of the applied energy E_0. So, for $E_0 = 10$, 30, and 50 keV, p is 0.37, 0.355, and 0.34, respectively. That is, the dependence of R_e on the energy E_0 in the expression $R_e \sim E_0^p$ can be represented as $E_0^{1.63}$ for 10 keV, $E_0^{1.645}$ for $E_0 = 30$ keV, and $E_0^{1.645}$ for 50 keV. For electrons with an energy of $E_0 = 100$ keV, R_e already is $\sim E_0^{1.67}$. Therefore, the rea-

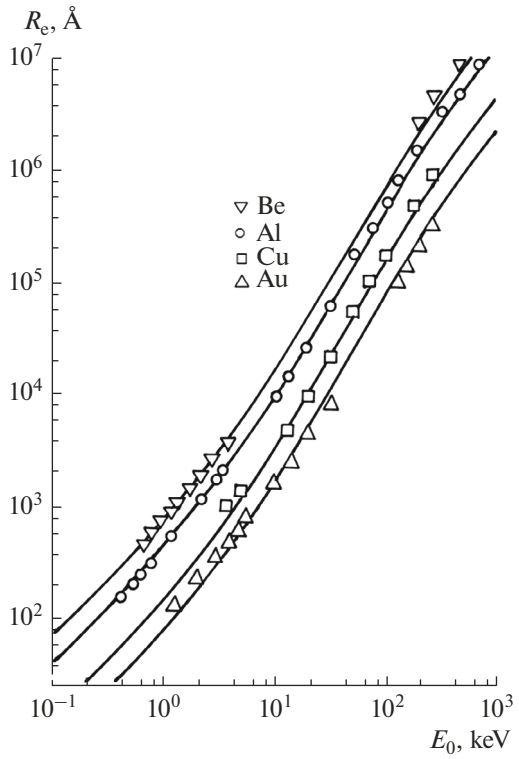

Fig. 4. Dependence of the electron-beam range on the primary electron energy in targets of a series of materials: the solid lines correspond to calculation using formula (6); and Δ, ○, □, and ▽ to the experimental results obtained from [8].

Table 1. Values of the ranges R_e, R_F, and R_{K-O} of the electron beam in a series of materials at a primary electron energy of E_0 1–50 keV

№/№	Element (atomic number)	Electron beam energy, keV	R_e, μm	R_F, μm	R_{K-O}, μm
1.	$_6$C	1	0.052	0.047	0.030
		5	0.38	0.38	0.44
		10	1.11	1.08	1.39
		20	3.50	3.51	4.41
		30	7.00	6.99	8.66
		40	11.54	11.40	14.00
		50	17.06	16.66	20.33
2.	$_{13}$Al	1	0.045	0.0405	0.028
		5	0.33	0.33	0.41
		10	0.96	0.92	1.31
		20	3.03	3.00	4.16
		30	6.06	5.97	8.17
		40	9.98	9.74	13.20
		50	14.75	14.23	19.07
3.	$_{22}$Ti	1	0.028	0.027	0.019
		5	0.21	0.22	0.275
		10	0.60	0.58	0.87
		20	1.90	1.89	2.77
		30	3.81	3.77	5.45
		40	6.27	6.14	8.78
		50	9.27	8.98	12.78
4.	$_{29}$Cu	1	0.014	0.016	0.010
		5	0.11	0.13	0.143
		10	0.30(7)	0.31(4)	0.46
		20	0.97	1.02	1.45
		30	1.93	2.03	2.84
		40	3.18	3.31	4.59
		50	4.70	4.84	6.66
5.	$_{47}$Ag	1	0.013	0.014	0.009
		5	0.09	0.11	0.135
		10	0.27	0.27	0.43
		20	0.86	0.88	1.36
		30	1.72	1.76	2.68
		40	2.84	2.87	4.33
		50	4.19	4.19	6.28
6.	$_{79}$Au	1	0.008	0.008	0.006
		5	0.06	0.07	0.085
		10	0.16	0.16	0.27
		20	0.51	0.51	0.85
		30	1.02	1.02	1.68
		40	1.68	1.66	2.71
		50	2.48	2.42	3.93

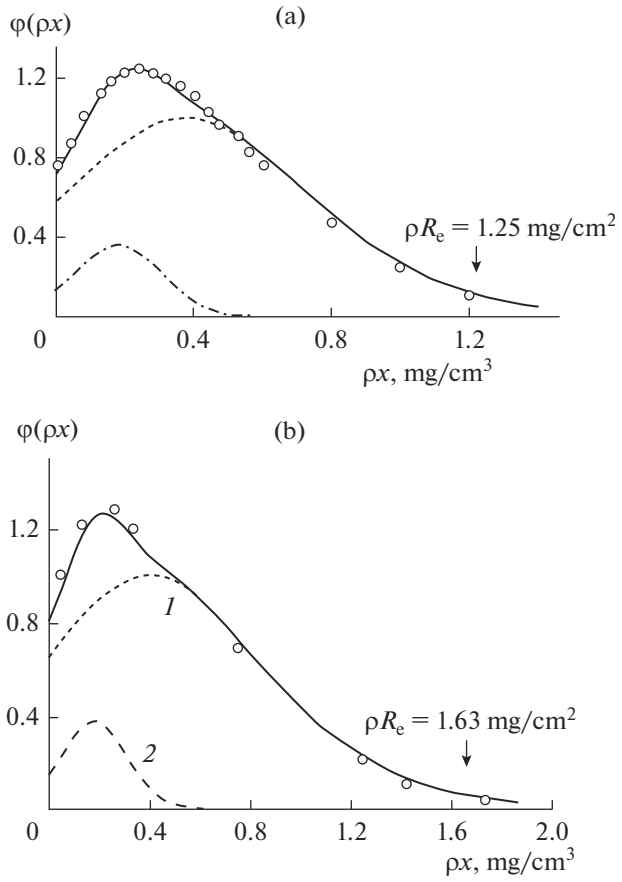

Fig. 5. Distribution of the energy losses of beam electrons in (a) Cu for $E_0 = 29$ keV and in (b) Ti for $E_0 = 25$ keV. The solid line corresponds to the calculation using the formulas in [5]; and o, to the experimental data obtained by means of the "labeled layer" method [17, 18].

son, for which researchers (starting from the Gruen paper [15] published in 1967 up to now) cannot empirically chose a single value of the exponent for R_e in the case of energies E_0 in the wide energy range 3—100 keV, becomes understandable. The actual dependence of R_e on E_0 is determined by the presence of the logarithmic function $\ln(E_0/C_m)$, which varies gradually with E_0, in the denominator of the formula for the stopping power of a material under electron bombardment.

CORRESPONDENCE OF THE PARAMETER R_e WITH THE PRACTICAL AND EXTRAPOLATED RANGES AND ALSO WITH THE DISTRIBUTION OF THE BEAM ELECTRON RANGES OVER THE TARGET DEPTH

The question, how is the range R_e defined by formula (6) related to the generally accepted, used definitions of the electron range in a material (extrapolated [16] and practical ranges R_F [8, 16]), is of undoubted interest and is important. As a rule, by the practical range, the thickness of a target in which ~99% of primary beam electrons are stopped is meant. It is seen from the results given in Table 1 that R_e and R_F are identical within several percent (1—5)%. To reveal the correspondence between R_e and the target-depth distribution of the electron ranges, Figure 5 shows the results of calculating such distributions for Ti and Cu obtained in accordance with formulas in [5] and the results of experimental measurements conducted in accordance with the "labeled layer" procedure for these materials [17, 18]. Good mutual correspondence between the calculation and the experiment in the figures shows that the distributions of the electron ranges in these two cases is very close to the real one. Therefore, the values of $R_e(\rho R_e)$ denoted by arrows in the figure can be identified as very close to the generally accepted definition, i.e., the extrapolated electron range [16]: as the particle penetration depth corresponding to extrapolation of the rectilinear portion of the curve $\varphi(\rho x)$ to its intersection with the abscissa axis. Thus, the obtained results make it possible to determine the place and importance of parameter R_e obtained taking into account only the average electron energy losses as an important estimation parameter, which characterizes the electron penetration depth in the sample under study and is easily calculated using formula (6). To find the electron penetration depth in the sample more exactly, it is necessary to use more complicated and more tedious calculations using formulas given in [5]. They describe the depth distribution of the electron ranges by taking into account the most probable beam electron energy losses. In this case, the contribution of the elastic scattering of charged particles and the influence of the processes of primary-electron backscattering on the transport of charged particles in a sample are also taken into account.

CONCLUSIONS

We have obtained a formula describing the distribution of the squared average energy of a monoenergetic electron beam over the depth of a solid target. It was established that the stopping power of the material depends on the material layer thickness rather than on the trajectory range of particles, which is convenient for use in practice in many applications. We have verified the obtained analytical expression by comparing the calculations with the existing results of experimental measurements of this quantity in Al, Cu, Ag, and Au. We have obtained a universal formula for calculating the electron ranges R_e in materials for electron-beam energies ranging from 0.1 keV to 1.0 MeV. We presented the results of calculating the electron ranges for a wide range of materials, namely, from beryllium to gold. The performed comparison of the obtained values of R_e with the results of calculations carried out in accordance with existing and widely used formulas of the diffusion model and the power approximation, and also with the results of experimental measure-

ments of R_e shows the obvious advantages of the new approach when describing the average values of the electron-beam energies in condensed materials.

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