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Empirical method evaluation of charge – Changing cross sections

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ABSTRACT

A method based on experimental data, theoretical models, and the empirical estimation of experimental parameters in the equilibrium charge distribution of ions is proposed to evaluate charge changing cross sections. This method makes it possible to obtain cross sections for loss and capture of one or several electrons in gaseous and solid targets with the inclusion of the influence of excited states of ions and target atoms.

1. Introduction

Processes of ion—atom collisions during the passage of ions through materials and the change in the energy and ion charges related to this are intensively studied in various fields of the physics. Information about the ion charge and energy distributions is important in the acceleration technology for the achievement of the required intensity of the beam of ions with the required charge [1], in studies of energy losses [2], in the case of ion reflection from surfaces [3], in the radiative study of materials for a more precise determination of depth concentrations of defects [4], for the solution of problems of the radiation resistance of materials under irradiation conditions, and for a more accurate determination of ion ranges in radiation medicine. The importance of these studies implies the necessity to improve methods for calculating cross sections for inelastic ion—atom collisions in order to more accurately determine the ion distributions over the charge and energy.

The first model describing the charge-changing phenomena and based on the classical electrodynamics was developed by R. H. Fowler [5], who compared the balance between the electron capture and loss by α-particles with the thermodynamic equilibrium between He^{2+} and He^{+} ions in the electron gas with given temperature and density. N. Bohr interpreted changes in the ion charge and charge fluctuations of the particle losing and capturing electrons during its passage through matter in his underlying paper [6]. Using the classical description of the motion of ions and the statistical regularities of the electron distribution in atoms, N. Bohr obtained qualitative estimates of charge-changing cross sections for ions passed through light and heavy gases [6]. The improvements of this model [7–9] made it possible to qualitatively interpret the electron capture and loss by multiply charged ions and describe charge-changing cross sections for fission fragments in gases.

The development of the acceleration technology in the middle of the last century contributed to the accumulation of a significant level of experimental data on charge distributions of ions in gaseous and solid targets [10–18], which stimulated the appearance of a series of theoretical methods for the quantum–mechanical description of charge-changing processes. However, there is still no holistic picture of the description of the charge distribution of ions with given nuclear charges and target compositions up to now. Unfortunately, even in the simplest case, where the material is either hydrogen or helium, the theory does not describe experimental data in the region of the energy loss maximum within the limits of the measurement accuracy.

All elastic and inelastic cross sections for particle—matter interaction can be divided into three categories, namely, those experimentally measured with a definite accuracy, results of calculations using various theoretical models, and evaluated or recommended data for estimations. The range of existing experimental and theoretical data is limited by the accuracy of measuring devices and by approximations of applied models and does not include the variety of versions of projectile and target parameters. The evaluated data serve as a "superstructure" to the "basis" consisting of experimental and theoretical results and are used to determine the most reliable values of the used quantities. The evaluated values are required to conform between the existing experimental and theoretical results, interpolate them to the range of parameters where the experimental and theoretical data are few, extend the energy range, and generalize the results to the case of an arbitrary "ion—target atom" pair.

The evaluated data for neutron—nucleus interaction cross sections were already used for 50 years or more and were regularly refined and renewed [19]. Energy losses of ions with the equilibrium charge distribution are another example of evaluated data. Main approximations required to obtain elastic and inelastic energy losses on the basis of existing experimental and theoretical results were formulated [20] and

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used in the SRIM program [21]. There is no evaluated data for cross sections for interaction of ions with the change in their charges.

Ions passing through the material can change their charges.

$X(E,Z,q) + A(Z_t) \rightarrow X(E',Z,q')$, (1)

where E and E' are the energies, q and q' are the ion charges before and after the collision, Z is the ion nuclear charge, and Z_t is the nuclear charge of the target atom. If the target is assumed to be the homogeneous material whose properties remain unchanged during the interaction with the ion, then the ion charge distribution is independent of the target orientation with respect to the incident beam. In the first approximation, the target can be assumed to be thin, and the change in the ion energy can be neglected ($E'=E$). In this case, the dependence of the relative number of ions with the charge q' and the energy E after the passage through a target with the thickness $\times$ must be described. The main characteristics of this process are the charge-changing cross sections $\sigma_{q,q'}(E,Z,Z_t)$ ($q' \neq q$), which are independent of the target thickness, the ion and target-atom nuclear masses, but depend on ion velocity V. Therefore, in inelastic ion—atom collisions, the ion energy E is usually expressed in keV/nucleon or MeV /nucleon units. The cross section $\sigma_{q,q+k}(E,Z,Z_t)$ is related to the process of loss of one ($k = 1$) or several ($k \geq 2$) electrons; and the cross section $\sigma_{q,q-k}(E,Z,Z_t)$, to that of capture of one ($k = 1$) or several ($k \geq 2$) electrons.

2. Theoretical concepts

2.1. Charge-changing equations for ions with the equilibrium charge distribution

As the layer thickness $\times$ increases, the electron capture and loss processes begin to gradually compensate each other, and the equilibrium charge distribution is established and is characterized by the equilibrium charge fractions $F_q(E,Z,Z_t)$. In this case, the interrelation between the charge fractions and the charge-changing cross sections is described by the system of homogeneous equations [10]:

$$\sum_{q' \neq q} F_{q'}(E,Z,Z_t)\sigma_{q',q}(E,Z,Z_t) - F_q(E,Z,Z_t)\sum_{q' \neq q}\sigma_{q,q'}(E,Z,Z_t) = 0 \quad (2)$$

with the normalization condition

$$\sum_q F_q(E,Z,Z_t) = 1 \quad (3)$$

The equilibrium ion charge distribution is characterized by the charge mean

$$\overline{q}(E,Z,Z_t) = \sum_q qF_q(E,Z,Z_t) \quad (4)$$

and the width parameter of the equilibrium charge distribution

$$[d(E,Z,Z_t)]^2 = \sum_q (q - \overline{q}(E,Z,Z_t))^2 F_q(E,Z,Z_t) \quad (5)$$

The homogeneity of system of equations (2) implies that, if all cross sections $\sigma_{q,q'}(E,Z,Z_t)$ are multiplied by the constant, then all charge fractions $F_q(E,Z,Z_t)$ and, accordingly, the parameters $\overline{q}(E,Z,Z_t)$ and $d(E,Z,Z_t)$ remain unchanged. The charge mean $\overline{q}(E,Z,Z_t)$ depends on the ratios $\sigma_{q,q+k}(E,Z,Z_t)/\sigma_{q,q-k}(E,Z,Z_t)$, and the charge fractions $F_{q''}(E,Z,Z_t)$, for which the condition $\sigma_{q'',q''+1}(E,Z,Z_t) \approx \sigma_{q'',q''-1}(E,Z,Z_t)$ is satisfied, make the main contribution to sum (4) [22].

Each of the equations in (2) is the balance equation and describes how many ions with the charge q appear and how many of them change their charges for $q' \neq q$ as a result of one inelastic collision of the ion with the target atom. System of differential equations (2) can be written in the matrix form $\widehat{A}F = 0$, where the symbol F means the vector of charge states $F_q(E,Z,Z_t)$ (-1 $\leq q \leq Z$), and the elements of the square matrix $\widehat{A}$ consist of the combinations of charge-changing cross sections.

If the matrix $\widehat{A}$ is known, then the equilibrium and nonequilibrium ion charge distributions can be calculated. Unlike the equilibrium distribution, the nonequilibrium one depends on the target thickness $\times$ and the ion distribution before the collision with the target. Thus, the problem of describing the equilibrium and nonequilibrium charge distributions of ions with the given initial distribution over q and E after their passage through a layer with the thickness $\times$ of the homogeneous target (1) reduces to the determination of elements of the matrix $\widehat{A}$ in the energy range under study.

The difficulty in the description of ion charge distribution consists in the variety of processes occurring in the broad ranges of energies E and ion charges q (-1 $\leq q \leq Z$). The dependences of the cross sections $\sigma_{q,q+1}(E,Z,Z_t)$ and $\sigma_{q,q-1}(E,Z,Z_t)$ on E are qualitatively different because of the absence of the similarity relations for these cross sections, and the ratio $\sigma_{q,q+1}(E,Z,Z_t)/\sigma_{q,q-1}(E,Z,Z_t)$ for given Z and Z_t depends on the ion energy E and charge q. As a consequence, the processes of one-electron loss and capture must be considered independently. Another difficulty is that inelastic collisions (1) can lead to the formation of excited states of the ion $X(E,Z,q')$ and the target atom. If the excited particles had no time to return to the ground state before the next collision, then the conditions for the ion—atom interaction and all cross sections $\sigma_{q,q\pm k}(E,Z,Z_t)$ ($k \geq 1$) are changed, which affects $F_q(E,Z,Z_t)$, $\overline{q}(E,Z,Z_t)$, and $d(E,Z,Z_t)$. This feature can lead to the difference between the ion charge distributions for gases and solid targets. In addition, the ion can lose or capture several electrons in one collision. The inclusion of the influence of such "multiple" processes with the participation of several electrons ($k \geq 2$) in one collision on the ion charge distribution requires an additional analysis. In the general case, to determine all elements of the matrix $\widehat{A}$ and describe the ion charge distribution in a broad energy range, the values of $Z \times (Z + 1)$ are required for the cross sections $\sigma_{q,q'}(E,Z,Z_t)$ in the case of $0 \leq q \leq Z$ and $0 \leq q' \leq Z$. As a rule, the change of the one of the parameters Z, Z_t, q or k even by unity leads to a noticeable change in the cross section $\sigma_{q,q\pm k}(E,Z,Z_t)$. Consequently, the problem of the description of the ion charge distribution can generally be solved only by taking additional approximations into account because of the variety of combinations of the parameters E, Z, Z_t, q, and k and because of their wide ranges.

2.2. Dependence of one-electron loss and capture cross sections on the ion energy

As the ion velocity increases, the one-electron loss cross section $\sigma_{q,q+1}(E,Z,Z_t)$ first increases because of the increase in the momentum transferred to the active electron and then decreases as $1/E$ [6] because of the decrease in the interaction time. The dependence $\sigma_{q,q+1}(E,Z,Z_t) \propto 1/E$ as $E \rightarrow \infty$ follows from the first order of perturbation theory and is independent of q, Z, and Z_t. In the case where, for the energy $E = E_q$, the ion velocity V and the velocity of valence electrons in the ion with the charge q coincide, the cross section $\sigma_{q,q+1}(E,Z,Z_t)$ reaches its maximum value $\sigma^{max}_{q,q+1}(E_q,Z,Z_t)$.

The cross section for one-electron capture by slow ions depends weakly on the energy $\sigma_{q,q-1}(E,Z,Z_t) \approx const$ for $E < 0.1$ MeV/nucleon. In this energy range where the ion velocity is close to the velocity of valence electrons of the target atoms, the system consisting of the incident ion and the target atom can be regarded as a molecule with common electrons in the field of two Coulomb centers.

In the range of fast collisions ($E \geq 0.3$ MeV/nucleon), the ion passing through the electron cloud of the target atom has time to interact, as a rule, only with one of the electrons. In this energy range, the cross section $\sigma_{q,q-1}(E,Z,Z_t)$ decreases rapidly with increasing E, and it can be represented in the form of the power function $\sigma_{q,q-1}(E,Z,Z_t) \propto 1/E^{\alpha}$ as $E \rightarrow \infty$ [23], where $\alpha > 0$ is the dimensionless parameter. In the Oppenheimer—Brinkman—Kramers (OBK) approximation [24,25], the calculations for collisions between protons and hydrogen atoms gave $\alpha = 5.5$.

In the case of the multielectron atom, α depends of the number of electrons in the target atom, i.e., on Z_t. It is related to the fact that the probability of capturing electrons from inner shells of the target atom increases with increasing ion velocity [1]. The average value of the parameter α is α = 4.8 for collisions between fast protons and multielectron atoms [23]. It is assumed in this approximation that the parameter α is independent of Z_t.

2.3. Loss and capture of several electrons by the ion

As a rule, when describing the ion charge distributions, it is assumed that the processes of loss and capture of one electron by the ion ($k = 1$) mainly contribute to the formation of the equilibrium charge distribution. This means that the probability of processes of loss and capture of several electrons by the ion in one collision ($k \geq 2$) can be regarded as a correction. This assumption means that the following relations are postulated:

$$\sigma_{q,q+1}(E,Z,Z_t) > \sigma_{q,q+2}(E,Z,Z_t) > \sigma_{q,q+3}(E,Z,Z_t) > \ldots > \sigma_{q,q+k} \quad (6)$$

$$\sigma_{q,q-1}(E,Z,Z_t) > \sigma_{q,q-2}(E,Z,Z_t) > \sigma_{q,q-3}(E,Z,Z_t) > \ldots > \sigma_{q,q-k}$$

Then system of equations (2) can be solved by the method of successive refinements. The solution $F_q^{(k=1)}(E,Z,Z_t)$ was first found taking only processes of one-electron loss and capture into account. In this approximation, the cross section matrix $\widehat{A}$ becomes quasidiagonal, and the parameters of the charge distribution take the values

$$\overline{q}^{(k=1)}(E,Z,Z_t) = \sum_q q F_q^{(k=1)}(E,Z,Z_t) \quad (7)$$

$$[d^{(k=1)}(E,Z,Z_t)]^2 = \sum_q (q - \overline{q}^{(k=1)}(E,Z,Z_t))^2 F_q^{(k=1)}(E,Z,Z_t) \quad (8)$$

The addition of the cross sections $\sigma_{q,q\pm2}(E,Z,Z_t)$ to the matrix $\widehat{A}$ leads to the refinement of the solution for the charge fractions $F_q^{(k=2)}(E,Z,Z_t)$ and the parameters of the charge distribution $\overline{q}^{(k=2)}(E,Z,Z_t)$ and $d^{(k=2)}(E,Z,Z_t)$. Then this procedure is used for all cross sections $\sigma_{q,q\pm k}(E,Z,Z_t)$ with $k \leq K_{max}$. Relation (6) implies that, as the parameter K_{max} increases, the solution of the system of equations $F_q^{(k=K_{max})}(E,Z,Z_t)$ converges to the exact solution with the inclusion of all charge-changing processes $F_q^{(k=K_{max})}(E,Z,Z_t) \rightarrow F_q(E,Z,Z_t)$ as $K_{max} \rightarrow \infty$. Approximation (6) means the possibility of using the parameter K_{max}, when solving system of equations (2) by the method of successive refinements with respect to the number of electrons participating in one inelastic collision between the ion and the one target atom. The quantity K_{max}, which determines the convergence of the solution of system of equations (2) to the exact solution, depends on the ion energy E, because the ratios for the processes of loss $\sigma_{q,q+k+1}(E,Z,Z_t)/\sigma_{q,q+k}(E,Z,Z_t)$ and capture $\sigma_{q,q-k-1}(E,Z,Z_t)/\sigma_{q,q-k}(E,Z,Z_t)$ of electrons by the ion are different [17] and depend on E.

3. Parameters of the equilibrium ion charge distribution

3.1. Ion charge means in gaseous and solid targets

The empirical method makes it possible to obtain parameters of the equilibrium charge distributions for an arbitrary "ion—target atom" pair in a broad energy range. This method is based on the assumption that parameters (4) and (5) are continuous and smooth functions of E, Z, and Z_t, whose forms are determined using the averaging of experimental data. The accuracy of the experimental distribution approximation is improved if the difference between the parameters of the gaseous and solid targets is taken into account [26]. The most precise expressions for the ion charge mean in the range of $0 \leq \overline{q}/Z \leq 1$ are expressions [27] obtained independently for gases

$$\overline{q}_{gas}(E,Z,Z_t)/Z = \frac{376x + x^6}{1428 + 1206x^{0.5} + 690x + x^6}$$

$$x = [Z_t^{0.03-0.17y}y]^{1+0.4/Z},\ y = Z^{-0.52}V/V_0 \quad (15)$$

and solid targets

$$\overline{q}_{sol}(E,Z,Z_t)/Z = \frac{8.29X + X^4}{0.06/X + 4 + 7.4X + X^4}$$

$$X = [1 - 0.26exp(-Z_t/11 - (Z_t - Z)^2/9][Y/(1 + 0.03Yln(Z_t))/1.54]^{1+1.83/Z}$$

$$Y = Z^{-0.543}V/V_0 \quad (16)$$

It should note that the formula (16) have been updated later [28]. The error in approximating the experimental values of $\overline{q}(E,Z,Z_t)$ by relations (15) and (16) depends on the energy E. In the energy range of $E < 0.1$ MeV/nucleon, this error is noticeably smaller for gases than for solid targets. In this approach, the dependence $\overline{q}(E,Z,Z_t)$is described by slowly varying analytical functions of Z and Z_t, which leads to the averaged dependence without considering the shell structure of the ion and target atom. The relations $\overline{q}_{gas}(E,Z,Z_t)$ and $\overline{q}_{sol}(E,Z,Z_t)$ in [27] are independent of the target density $\rho(Z_t)$, and, consequently, there is no gradual passage from the dense gas to the solid target for the ion charge mean $\overline{q}(E,Z,Z_t)$. This feature is analogous to the dependence of inelastic energy losses in MeV × cm^2/mg units, which depend on the aggregate target state rather than on its density [21]. The maximum of $\Delta q_{g-s} = \overline{q}_{sol}(E,Z,Z_t) - \overline{q}_{gas}(E,Z,Z_t)$ is in the energy range of $E = 0.07 - 1$ MeV/nucleon. The ion energy at which the maximum distinction of the charge mean Δq_{g-s} is reached increases with increasing Z. For ions with energies of $E > 10$ MeV/nucleon, the ion charge means in gases and solid targets coincide ($\Delta q_{g-s} \rightarrow 0$).

3.2. Width of the ion charge distribution

The error in the experimental parameter $d(E,Z,Z_t)$ is noticeably larger than $\overline{q}(E,Z,Z_t)$. This is explained by the increase in the role of charge fractions with small intensities and by that in their errors. To empirically describe the width parameters $d(E,Z,Z_t)$ in gaseous and solid targets, the authors of [29] proposed to use the ratio $\overline{q}/Z$ as an argument of the function $d(E,Z,Z_t)$. In this case, the domain of definition of the functions $d(\overline{q}/Z)$ is limited ($0 \leq \overline{q}/Z \leq 1$), which is convenient for its approximation. The dome-shaped dependence $d(\overline{q}/Z)$ is described by the function [29]:

$$d(\overline{q}/Z) = C\{1\text{–}\exp[-(\overline{q}/Z)^{\gamma}]\}\{1\text{–}\exp[-(1 - \overline{q}/Z)^{\beta}]\}, \quad (17)$$

where the dimensionless parameter C characterizes the "plateau" height in the intermediate energy range, and the dimensionless parameters γ and β characterize the decrease in d in the case of slow ($\overline{q}/Z \rightarrow 0$) and fast ($\overline{q}/Z \rightarrow 1$) collisions, respectively. The dependence on the energy E and the target-atom nuclear charge Z_t in (17) is taken into account in the equilibrium charge mean $\overline{q}(Z,Z_t,E)$. In the general case, it is possible that the result of approximating the experimental dependence by the function of three parameters $d(\gamma,\beta,C)$ does not give a unique solution in the case of the correlation between them. Such a correlation exists between γ and C and also between β and C. For the unambiguity of the determination of the parameters in (17), it was assumed that γ and β depend only on the aggregate state of the material and are independent of Z and Z_t. Restricting themselves by the linear dependence of C on Z and Z_t, from the experimental dependence $d(E,Z,Z_t)$, the authors of [29] found that, for gases,

$$\gamma = 0.40,\ \beta = 0.75,\ C = 3.01904\text{–}0.01821\ Z_t + 0.07817\ Z + 0.00132\ Z\ Z_t, \quad (18)$$

and, for the solid targets,

$$\gamma = 0.23,\ \beta = 0.32,\ C = 2.66939\text{–}0.0098\ Z_t + 0.05802\ Z + 0.00048\ Z\ Z_t. \quad (19)$$

When the parameters $\overline{q}(E,Z,Z_t)$ and $d(E,Z,Z_t)$ are described empir-

ically, the influence of effects related to the difference between the binding energies of electrons for different shells in the ion and the target atom, which can lead to oscillations of cross section values in the dependence of Z and Z_t, is neglected. Some materials can have a rather large range of densities under normal conditions, for example, the graphite density varies from 1.7 to 2.4 g/cm^3 as a function of its brand. This difference is not taken into account in the case of the empirical evaluation of the parameters $\overline{q}(E,Z,Z_t)$ and $d(E,Z,Z_t)$, because the target density $\rho(Z_t)$ is not contained in (15) – (17). As a result, the density effect is discretely taken into account in the empirical evaluation of the parameters $\overline{q}(E,Z,Z_t)$ and $d(E,Z,Z_t)$when passing from the gaseous to the solid target, namely, the target is either the gas or the solid material.

4. Empirical evaluation of charge-changing cross sections

4.1. One-electron loss and capture cross sections in gases

The evaluation of one-electron loss and capture cross sections in gases is based on experimental data and the approximation in which the cross section $\sigma_{q,q\pm1}(E,Z,Z_t)$ is assumed to be a continuous function of E, q, Z, and Z_t, and the dependence of the cross sections $\sigma_{q,q\pm1}(E,Z,Z_t)$ on the ion energy E has at most one maximum.

The maximum value of the one-electron loss cross section $\sigma^{max}_{q,q+1}(E_q, Z,Z_t)$ in gases is reached at an energy of $E_q = 50\ |\varepsilon_q|$ MeV/nucleon, where ε_q is the binding energy of the valence electron in the ion $X(E,Z,q)$ in atomic units. The results of calculating ε_q by means of the Hartree—Fock method [30] are known. In the energy range of $E \geq E_q$, the ratio of the cross sections

$$f_{los}(E) = \sigma_{q,q+1}(E,Z,Z_t)/\sigma^{max}_{q,q+1}(E_q,Z,Z_t) \leq 1 \quad (20)$$

is calculated in the first order of perturbation theory for proton and hydrogen atom collisions. We note that the function $f_{los}(E)$ is independent of q, Z, and Z_t. We then use the approximation in which the cross section $\sigma_{q,q+1}(E,Z,Z_t)$ increases in accordance with the power law with increasing slow-ion energy E. To describe the cross section $\sigma_{q,q+1}(E,Z,Z_t)$ in the entire range of energies E, we introduce the asymptotic functions

$\sigma^{A}_{q,q+1}(E,Z,Z_t) \propto E^{\lambda(q)}$ as $E \to 0$, (21)

$\sigma^{B}_{q,q+1}(E,Z,Z_t) = f_{los}(E)\sigma^{max}_{q,q+1}(E_q,Z,Z_t)$ for $E \geq E_q$, (22)

where the values of $\lambda(q)$ in Eq.(21) were obtained from experimental data [17].

To describe $\sigma_{q,q+1}(E,Z,Z_t)$ in the range of ion energies $E < E_q$, the following interpolation is used:

$$1/\sigma_{q,q+1}(E,Z,Z_t) = 1/\sigma^{A}_{q,q+1}(E,Z,Z_t) + 1/\sigma^{B}_{q,q+1}(E,Z,Z_t) \quad (23)$$

To describe the one-electron capture cross section $\sigma_{q,q+1}(E,Z,Z_t)$ in gases, two asymptotic functions are used:

$\sigma^{A}_{q,q-1}(E,Z,Z_t) = \sigma^{max}_{q,q-1}(Z,Z_t)$ as $E \to 0$, (24)

$\sigma^{B}_{q,q-1}(E,Z,Z_t) = \sigma^{\infty}_{q,q-1}(Z,Z_t)/E^{\alpha(Z_t)}$ for $E \geq 0.3$ MeV/nucleon. (25)

Then we assume that the parameter $\alpha(Z_t)$, which characterizes the energy dependence of the cross section for one-electron capture by the fast ion, is independent of Z. In the OBK approximations, the calculation [24,25] for multiply charged ions with $Z \geq 5$ give $\alpha(Z_t) \approx 4.5$ for $Z_t \leq 2$, $\alpha(Z_t) \approx 3.5$ for $Z_t = 7$, $\alpha(Z_t) \approx 3.0$ for $Z_t = 18$, and $\alpha(Z_t) \approx 2.85$ for $Z_t \geq 36$ [31]. To describe $\sigma_{q,q-1}(E,Z,Z_t)$ in the ion energy range of $E < 0.3$ MeV/nucleon, the following interpolation is used:

$$1/\sigma_{q,q-1}(E,Z,Z_t) = 1/\sigma^{A}_{q,q-1}(E,Z,Z_t) + 1/\sigma^{B}_{q,q-1}(E,Z,Z_t) \quad (26)$$

If the energy dependence of the cross sections $\sigma_{q,q\pm1}(E,Z,Z_t)$ is known, then it is possible to calculate the charge fractions $F_q(E,Z,Z_t)$, then the ion charge mean $\overline{q}(E,Z,Z_t)$ (4), and the parameter of the charge distribution width $d(E,Z,Z_t)$ (5) by means of the system of charge-changing equations for the equilibrium ion charge distribution (2). These parameters can differ from empirical values in gases (15) and (17). The following minimum difference between the calculated and empirical parameters is reached by varying the parameters of $\sigma^{max}_{q,q+1}(E_q, Z,Z_t)$ and $\sigma^{\infty}_{q,q-1}(Z,Z_t)$ for ions with an energy of $E \geq 0.05$ MeV and $\lambda(q)$ and those of$\sigma^{max}_{q,q-1}(Z,Z_t)$ for ions with $E < 0.05$ MeV:

$$\left|\overline{q}_{gas}(E,Z,Z_t) - \overline{q}(E,Z,Z_t)\right| \to 0 \quad (27)$$

$$\left|d(\overline{q}_{gas}/Z) - d(E,Z,Z_t)\right| \to 0 \quad (28)$$

When the parameters $\sigma^{max}_{q,q+1}(E_q,Z,Z_t)$ are varied we also assume that one-electron loss cross section decreases with increasing q and the relation $d\sigma^{max}_{q,q+1}(E_q,Z,Z_t)/dq < 0s$ is true. Then all cross sections $\sigma_{q,q\pm1}(E,Z,Z_t)$ are normalized to the experimental ones $\sigma^{exp}_{q,q\pm1}(E,Z,Z_t)$. In this procedure, the common normalization factor $C_{exp}(Z)$ is calculated; it is independent of E, q, and Z_t. In these calculations, among all existing experimental data, the cross sections $\sigma_{q,q+1}(E,Z,Z_t)$ at $E \approx E_q$ and the cross sections $\sigma_{q,q-1}(E,Z,Z_t)$ in the range of $E \approx 0.1 - 0.3$ MeV/nucleon are given top priority.

4.2. One-electron loss and capture cross sections in solid targets

In solid targets, the ion charge mean exceeds the corresponding value in gases [27]. The increase in the target density and the decrease in the time interval between collisions lead to the fact that the ion in the solid material has no time to return to the ground state from the excited one. The influence of the excited states of the ion leads to an increase in the probability of electron loss and to a decrease in the probability of electron capture in the next collision for solid targets compared with gases and to an increase in the ion charge mean.

To calculate the one-electron loss and capture cross sections $\sigma^{sol}_{q,q\pm1}(E,Z,Z_t)$ in solid targets, an approximation in which the difference between the charge-changing cross sections in gases and solid targets reduces to the scale factor [32]:

$\sigma^{sol}_{q,q+1}(E,Z,Z_t) = \sigma^{gas}_{q,q+1}(E,Z,Z_t) \times C_{g-s}(E,Z,Z_t)$, (29)

$\sigma^{sol}_{q,q-1}(E,Z,Z_t) = \sigma^{gas}_{q,q-1}(E,Z,Z_t) / C_{g-s}(E,Z,Z_t)$ (30)

is used. It is assumed in this approximation that the coefficient$C_{g-s}(E, Z,Z_t)$, which takes into account the influence of the excited states of the ion on the cross sections $\sigma^{sol}_{q,q\pm1}(E,Z,Z_t)$ in solids, is independent of the ion charge q. The experimental relation $\overline{q}_{sol}(E,Z,Z_t) \geq \overline{q}_{gas}(E,Z,Z_t)$ implies that $C_{g-s}(E,Z,Z_t)$. When passing from the cross sections in gases to those in solid targets, the increase in the cross section $\sigma^{sol}_{q,q+1}(E,Z,Z_t)$ and the decrease in the cross section $\sigma^{sol}_{q,q-1}(E,Z,Z_t)$ lead to a decrease in the energy E", at which the relation $\sigma^{sol}_{q,q+1}(E'',Z,Z_t) \approx \sigma^{sol}_{q,q-1}(E'',Z,Z_t)$ is valid, and the ion with the charge q can capture or lose one of its electrons with the equal probability. The dependence of $C_{g-s}(E,Z,Z_t)$ on the energy E takes into account the change in the role of the ion and target-atom excited states in the charge-changing process. In the energy range of $E = 0.07 - 1$ MeV/nucleon, $C_{g-s}(E, Z, Z_t)$(and $\Delta q_{g-s} = \overline{q}_{sol}(E, Z, Z_t) - \overline{q}_{gas}(E,Z,Z_t)$) reaches the maximum value$C^{max}_{g-s}(Z,Z_t)$; in this case, as Z increases, the parameter $C^{max}_{g-s}(Z,Z_t)$ increases. For fast collisions, $\overline{q}_{sol}(E, Z,Z_t) \approx \overline{q}_{gas}(E,Z,Z_t)$, and the influence of the density effect on the ion charge-changing cross sections decreases, i.e., $C_{g-s}(E,Z,Z_t) \to 1$ as $E \to \infty$.

The dependence of relation (29) on the ion energy leads to the fact that the energy E^{sol}_q at which the maximum one-electron loss cross section $\sigma^{max}_{q,q+1}(E^{sol}_q, Z,Z_t)$ is reached differs from the energy E_q in gases. The relation $E^{sol}_q \leq E_q$ is explained by the impurity of ion excited states and by the decrease in the average binding energy of the valence electron for solid targets compared with gases [33].

For processes of electron loss and capture, the density effect is taken into account by the same coefficient $C_{g-s}(E,Z,Z_t)$ in (29) and (30). This is a simplification, because the influence of the excited states on the loss

and capture cross sections is different in the general case. However, the use of, for example, two coefficients differing in value for loss and capture cross sections leads to a strong correlation between them and violates the unambiguity in the calculation of $\sigma^{sol}_{q,q\pm1}(E,Z,Z_t)$.

4.3. Cross sections for loss and capture of several electrons

The existing experimental data [11,17] showed that the cross sections in the charge-changing matrix $\widehat{A}$ decrease as the distance between the matrix element and the diagonal of this matrix (6) increases. The processes of one-electron loss and capture contribute mainly to the ion charge distribution, and multiple processes, which are described by the cross sections $\sigma_{q,q\pm k}(E,Z,Z_t)$ $k \geq 2$, can be regarded as a correction. Consequently, when solving the system of the charge-changing equations, it is possible to use the method of successive refinements, including the cross sections$\sigma_{q,q\pm k}(E,Z,Z_t)$ with higher values of k in the charge-changing matrix $\widehat{A}$ at each step. As the number of electrons participating in this interaction (6) increases, a decrease in the interaction probability is related to the approximation of pair interactions. The ion passing through the electron shells of the target atom interacts with each of the electrons independently rather than with several electrons at the same time. This model is based on the hypothesis about the pre-equilibrium ion charge distribution in which the initial ion charge q_0 slightly differs from the equilibrium value $\overline{q}(E)$ for this ion velocity. The problem of the existence of the energy and ion-charge ranges where relations (6) can be violated remains unsolved. In this case, it is impossible to consider multiple processes in the form of the correction, and the method for solving the system of equations requires the improvement.

The approximation in which the ratios of the cross sections for the processes of loss and capture of several electrons are the same and are independent of the ion charge q [34] and the number of active electrons k

$$W^{gas}(E,Z,Z_t) = \frac{\sigma^{gas}_{q,q\pm(k+1)}(E,Z,Z_t)}{\sigma^{gas}_{q,q\pm k}(E,Z,Z_t)} < 1 \tag{31}$$

$$W^{sol}(E,Z,Z_t) = \frac{\sigma^{sol}_{q,q\pm(k+1)}(E,Z,Z_t)}{\sigma^{sol}_{q,q\pm k}(E,Z,Z_t)} < 1 \tag{32}$$

is used to describe the cross sections $\sigma_{q,q\pm k}(E,Z,Z_t)$ $(0 \leq q \pm k \leq Z, k \geq 2)$.

The parameter $W^{gas}(E,Z,Z_t)$ is calculated using the minimum for relation (28). The inclusion of multiple processes ($k > 1$) makes it possible to noticeably decrease the difference between the calculated parameter $d(E,Z,Z_t)$ and the empirical value of $d(\overline{q}_{gas}/Z)$ in (28). In this case, the ion charge mean $\overline{q}(E,Z,Z_t)$ changes slightly, because the cross sections $\sigma_{q,q\pm1}(E,Z,Z_t)$ are independent of $W^{gas}(E,Z,Z_t)$. Nevertheless, the addition of the cross sections $\sigma_{q,q\pm k}(E,Z,Z_t)$ for $2 \leq k \leq K_{max}$ to the charge-changing matrix $\widehat{A}$ leads to a small variation of the ion charge mean $\overline{q}(E,Z,Z_t)$ compared with the solution for $K_{max} = 1$ (7).

The evaluation of cross-sections takes place in several stages. At first, the trial values of the coefficients $C_{g-s}(E,Z,Z_t)\rightarrow1$ and $W^{sol}(E,Z,Z_t)\rightarrow0$ are set and cross sections $\sigma^{sol}_{q,q\pm k}(E,Z,Z_t)$ are calculated. Then, the charge fractions are determined using equations (2) and the mean charge (4) and width of the ion charge distribution (5) are calculated. Finally, the obtained parameters of the charge distribution of ions are compared with empirical values, and the calculations are repeated after variation of the coefficients $C_{g-s}(E,Z,Z_t)$ and $W^{sol}(E,Z,Z_t)$. Thus the values of the parameters $C_{g-s}(E,Z,Z_t)$ and $W^{sol}(E,Z,Z_t)$ are estimated using the minimum of the difference between the parameters of the equilibrium charge distribution $\overline{q}(E,Z,Z_t)$, $d(E,Z,Z_t)$ calculated with the cross sections $\sigma^{sol}_{q,q\pm k}(E,Z,Z_t)$ for $k \leq K_{max}$ and the empirical values of $\overline{q}_{sol}(E,Z,Z_t)$, $d(\overline{q}_{sol}/Z)$:

$$\left|\overline{q}_{sol}(E,Z,Z_t) - \overline{q}(E,Z,Z_t)\right| \rightarrow 0 \tag{33}$$

$$\left|d(\overline{q}_{sol}/Z) - d(E,Z,Z_t)\right| \rightarrow 0 \tag{34}$$

The use of the coefficient $W(E,Z,Z_t)$, which is common for cross sections of all multiple processes, is independent of the parameters q and k, and is the same for processes of loss and capture of electrons by ions, is a simplification related to the necessity of the unambiguity in the results of calculating $\sigma_{q,q\pm k}(E,Z,Z_t)$. The method for evaluating the charge-changing cross sections in solid targets uses only two variable parameters $C_{g-s}(E,Z,Z_t)$ and $W^{sol}(E,Z,Z_t)$ and two relations (33) и (34) to determine them. It should be noted that the weak correlation between the values of $C_{g-s}(E,Z,Z_t)$ and $W^{sol}(E,Z,Z_t)$ leads to an unambiguous solution.

5. Comparison with experimental data

To compare the evaluated and experimental data, the relations are used:

$$(C/E)_{\text{los}} = \sigma_{q,q+1}(E,Z,Z_t)/\sigma^{exp}_{q,q+1}(E,Z,Z_t) \tag{35}$$

$$(C/E)_{\text{cap}} = \sigma_{q,q-1}(E,Z,Z_t)/\sigma^{exp}_{q,q-1}(E,Z,Z_t) \tag{36}$$

where $\sigma^{exp}_{q,q\pm1}(E,Z,Z_t)$ are the cross sections for the loss and capture of one electron measured in gases. The number of such measured cross sections for electron loss and capture by fast ions is sufficient to cover a wide range of ion charges Z and energies E. The theoretical cross sections for the loss and capture of one electron by ions in gases are compared with some available experimental data (Table 1). When choosing the experimental data to be compared, priority was given to ions with energies $E > 0.03$ MeV/nucleon.

Figs. 1 and 2 show a satisfactory agreement between the theoretical and experimental cross sections. The difference in the C/E ratio within the factor of 2 ($0.5 \leq C/E \leq 2$) is 58% for the cross sections $\sigma_{q,q+1}(E,Z,Z_t)$ and 48% for the cross sections $\sigma_{q,q-1}(E,Z,Z_t)$. The difference in the C/E ratio within a factor of 10 ($0.1 \leq C/E \leq 10$) is 93% for the cross sections $\sigma_{q,q+1}(E,Z,Z_t)$ and 91% for the cross sections $\sigma_{q,q-1}(E,Z,Z_t)$. Large values of the C/E ratio can be associated with the need for further improvement of the model for evaluating the charge-changing cross sections and significant experimental errors (up to 15%) [17]. It is important that the number of estimated cross-sections in Figs. 1 and 2 with $C/E < 1$ is

Table 1
The number of experimental cross sections for the loss N^{los}_{exp} and capture N^{cap}_{exp} of one electron by ions with a nuclear charge Z in gases with an atomic charge Z_t in the energy range from E_{min} to E_{max}.

References	Z	Z_t	E_{min},MeV/nucleon	E_{max},MeV/nucleon	N^{los}_{exp}	N^{cap}_{exp}
[17]	7	2, 7, 10, 18, 36	0.035	0.752	165	163
[17]	10	2, 7, 10, 18, 36	0.035	0.522	106	115
[35]	18	1, 18	3.4	8.5	2	3
[36]	18	18	2.38	2.38	0	4
[35]	26	18	3.4	8.5	1	7
[37]	26	1	0.282	3.4	12	14
[38]	26	2, 7, 10, 18	7.14	7.14	0	4
[39,40]	35	1, 2, 18	0.076	0.316	36	36
[17]	36	2, 7, 36	0.035	0.035	4	6
[41]	42	1	0.060	0.161	0	37
[40,42]	53	1, 2, 8	0.039	0.197	66	132
[43]	53	2	0.100	0.250	0	18
[44]	53	7, 18	0.048	0.506	0	72
[45]	54	7	2.4	8.4	41	39

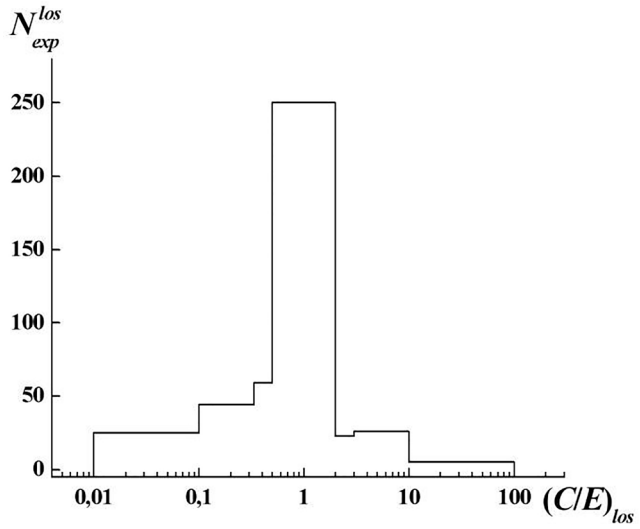

Fig. 1. Dependence of the number of experimental cross sections for electron loss, N^{exp}_{los}, on the ratio of the evaluated $\sigma_{q,q+1}(E,Z,Z_t)$ and experimental $\sigma^{exp}_{q,q+1}(E,Z,Z_t)$ cross sections $(C/E)_{los}$.

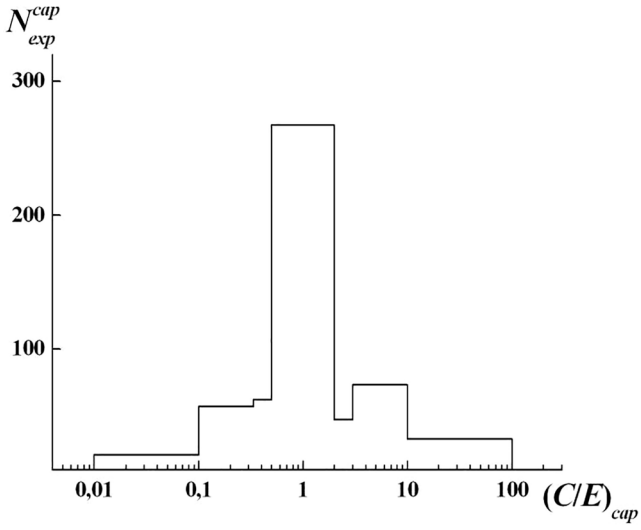

Fig. 2. Dependence of the number of experimental cross sections for electron capture by an ion, N^{exp}_{cap}, on the ratio of the evaluated $\sigma_{q,q-1}(E,Z,Z_t)$ and experimental $\sigma^{exp}_{q,q-1}(E,Z,Z_t)$ cross sections $(C/E)_{cap}$.

approximately equal to the number of cross-sections with $C/E > 1$.

6. Conclusions

The obtained estimates of the cross sections $\sigma_{q,q\pm k}(E,Z,Z_t)$ for gaseous and solid targets in the ranges of 1 keV/nucleon $\leq E \leq$ 50 MeV/nucleon, $5 \leq Z \leq 54$, $-1 \leq q \leq Z$, and $1 \leq k \leq 8$ can be used to describe charge distributions and inelastic energy losses of ions with the pre-equilibrium charge distribution. The boundaries of these ranges were explained by several reasons. The upper ion energy boundary ($E = 50$ MeV/nucleon) is due to the smallness of relativistic corrections in the dependences of the cross sections on the ion energy. For slow collisions ($E < 1$ keV/nucleon), effects related to the influence of thermal vibrations and target atom resonances (which are neglected in the used model) on the cross sections became noticeable. For light ions ($Z \leq 4$), the agreement between the empirical parameters $\overline{q}(E,Z,Z_t)$ and $d(\overline{q}/Z)$ and the experimental data became worse because of a small number of charge fractions of ions, and the charge-changing cross sections for these ions must be evaluated using other methods. The influence of multiple processes on the ion charge distribution was enhanced with increasing nuclear charge Z. The energy range where $W(E,Z,Z_t) \rightarrow 1$ appears for heavy multiply charged ions, and the processes of loss and capture of several ions in one collision cannot be considered as a correction. In these ranges of charges q and energies E, for ions with $Z > 54$, it is required to improve the description of the interaction of the ion with several electrons at the same time and generalize computing methods for solving the system of charge-changing equations without the approximation of the smallness of the parameter $W(E,Z,Z_t)$.

The importance of the proposed method is determined by new prospects, which appeared when using the charge-changing cross sections calculated in broad ranges of ion energies and charges. The obtained cross sections make it possible to take into account features of nonequilibrium processes in the case of the establishment of the charge distribution in surface target layers. The dependence of inelastic energy losses on the ion charge q and the modification of the charge distributions of ions with the change in their energies give a possibility of simulating the ion charge and energy distributions within the framework of the holistic model.

Declaration of Competing Interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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