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Analytical approximation of cross sections of collisions of electrons with atoms of inert gases

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The paper presents an analysis of data on the cross sections of elastic and inelastic collisions of electrons with noble gas atoms. The transport (diffusion) cross section, the excitation and ionization cross sections are considered. The bibliography on the cross sections of electron-atomic collisions includes many thousands of works. But the critical analysis of the results of experimental data in the review work is very difficult due to the fact that the necessary initial data can only be available to the authors of the work. The errors of the order of 1-3% given in the original works are contrasted with each other, sometimes differing by 50%. Comparisons of the electron cross sections sets in noble gases was made. For the selected sets of experimental and theoretical data, optimal analytical formulas are found and approximation coefficients are selected for them. The obtained semi-empirical formulas allow us to reproduce the cross-section values for them in a wide range of collision energies from 0.001 to 10000 eV with an accuracy of several percent.

Key words: electron atomic collisions, transport cross section, excitation cross section, ionization cross section, approximation of cross sections, noble gases.
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Introduction

Electron-atom cross section data are important in a large variety of applications and fields (see for instance [1-3]). In a low-temperature plasma, electrons acquire energy from electromagnetic fields and expend it in collisions with atoms. For numerical simulation of various phenomena in a gas-discharge plasma by the particle method, in the hydrodynamic approximation, or based on the solution of the kinetic Boltzmann equation, it is necessary to know the cross sections of electron-atomic collisions. Since the degree of ionization of low-temperature plasma is low, the concentration of atoms is much higher than the concentration of electrons and ions. Therefore, the diffusion and drift of electrons in a gas are mainly determined by elastic collisions of electrons with atoms, since their frequency is two to three orders of magnitude higher than the frequency of inelastic collisions. But the energy characteristics of a gas discharge are determined by the inelastic processes of ionization, excitation, etc. It is not our intention to give are view of the experimental and theoretical work on elastic and total cross sections, with which we compare our results. For that we refer to the review articles [4-6].

Here we will limit ourselves to the case of noble gases and consider the most significant characteristics of electron-atomic collisions in the modeling of gas-discharge plasma problems. These are the transport cross section, which determines the rate of momentum loss and the rate of electron drift, the total excitation cross section, which determines the energy costs for the excitation of atoms, and the ionization cross section from the ground state, which determines the frequency of the appearance of new electrons during ionization and their energy spectrum [7-10].

The bibliography on the cross sections of electron-atomic collisions includes many thousands of works, and probably an exhaustive review and selection of data is contained in [1-6]. But it should be borne in mind that the critical analysis of the results of experimental data in the review work is very difficult due to the fact that the necessary initial data can only be available to the authors of the work. The errors of the order of 1-3% given in the original works are surprisingly contrasted with each other, sometimes differing by 50%. Therefore, in the review work, only a comparative analysis of the results obtained is really possible, which shows that in the best case, the relative errors of measuring cross sections are on the order of 5-10%, and more often 20-50%. Comparisons of the electron scattering cross sections sets in noble gases was made in [11 – 13].

The most convenient form of presentation of experimental and computational-theoretical data is the selection of analytical approximations for them. In [14], such approximations was created for the cross sections of collisions of electrons with atoms of inert gases, elastic and inelastic. The total cross section for argon is selected for a range of approximately starting from 10-20 eV. The total cross sections are radically different in the region of lower energies from the transport (diffusion) cross sections, which are of the greatest interest for applied problems of gas discharge physics. The difference in the region of low energies (<10-20 eV) is about 100 times, in the region of high energies (> 10-20 eV), the total cross section exceeds the transport one by about two times.

Although the elastic collision of an electron with an atom is determined by a simple Coulomb interaction, its result is rather complex. The angular distribution of electrons, which determines the differential cross section, is usually highly anisotropic, and the angular distribution of scattered electrons often has sharp peaks. In addition, the elastic scattering cross section usually strongly depends on the collision energy as well. Even in the case of noble monatomic gases, the cross section for elastic collisions can have a nonmonotonic dependence on the collision energy (the Ramsauer effect for heavy gases – argon, krypton, and xenon). But for practical purposes in the numerical simulation of applied problems in the physics of a gas discharge, it is possible to reduce all this variety of elastic scattering characteristics to one – the transport scattering cross section, which depends on the collision energy [1 – 4].

The critical analysis of electron cross sections for total scattering by noble gases over a large energy range was started in [14 – 15]. From a large number of experimental and calculated data on cross sections, we selected the data that, based on the performed analysis, were recommended in these works with minor additions from later works. This made it possible to significantly expand the range of applicability of the selected analytical dependences in comparison with those given in [14 – 15].

One of the factors leading to significant errors in determining the characteristics of diffusion and drift of electrons in gases are impurities in the working gas. It is well known that even small impurities can significantly change the drift characteristics. For example, the question of the influence of small fractions of hydrogen and nitrogen impurities on the electron drift in neon was studied in [17]. A numerical model developed for the study of barrier discharges in helium and dry air impurities is presented in paper [18]. This model was used to investigation of the influence of air traces on the evolution of the dielectric barrier discharge in helium. The level of dry air was in the range from 0 to 1500 ppm (parts per million), which corresponds to the most commonly encountered range in atmospheric pressure discharge experiments. This results clearly show that the discharge evolution is highly affected by the concentration level of impurities in the mixture. It was observed that air traces assist the discharge ignition at very low concentration levels (~55 ppm).

Just like elastic collisions, the excitation of atomic levels by electron impact is one of the main processes that determine the characteristics of a gas discharge. The appearance of excited atoms due to stepwise ionization can lead to a significant increase in the ionization frequency, metastable atoms can play a significant role in the formation of a gas discharge, sometimes the transfer of resonant radiation is the main mechanism of energy transfer, and sometimes super elastic collisions play an important role. But for the purposes of mathematical modeling of processes in a gas-discharge plasma, it is almost always sufficient to take into account only the total excitation cross section of atomic levels without loss of accuracy. Therefore, here we restrict ourselves to approximating the total cross section for excitation from the ground level.

Electron impact ionization from the ground state of an atom is perhaps the most common way to form and maintain a gas-discharge plasma. With a large excess of the electron energy above the ionization threshold, both experimental methods and theory provide good accuracy. But for low energies, there are practically no experimental data, and the accuracy of theoretical calculations is also low. When selecting the data for approximating the cross sections, we limited ourselves to considering the cross sections of ionization of noble gas atoms by an electron strike from the ground state, which is sufficient for the purposes of applied problems modeling in gas-discharge plasma.

Ionization by electron impact from the ground state of the atom is, perhaps, the most frequent method for the formation and maintenance of a gas-discharge plasma. With a large excess of the electron energy over the ionization threshold, both experimental methods and theory give good accuracy. However, there are practically no experimental data for low energies, and the accuracy of theoretical calculations is also low. When choosing the data for approximating the cross sections, we limited ourselves to considering the cross sections for ionization of noble gas atoms by electron impact from the ground state, which is sufficient for the purposes of modeling applied problems of gas-discharge plasma.

Analytical expression for the transport section in elastic collisions

Let us first consider the problem of approximating the transport cross section in elastic collisions of an electron with an atom. The natural scale of the collision energy can be the ionization potential of an atom. Depending on the elastic collision cross section from the energy, three characteristic sections can be distinguished: collisions with low energy<10 eV, collisions with medium energy, and collisions with high energy >300 eV. An approximation of the dependence of the transport cross sections for elastic collisions of electrons with atoms of noble gases on the collision energy will be sought in the form of a sum of a series of terms of the form: $(A+B\varepsilon^{C})/(1+D\varepsilon^{E})$. The first ionization potential I can serve as the natural scale of energy in the collision of an electron with an atom, therefore, it is convenient to go over to the dimensionless energy *x = ε / I*. Accordingly, we will approximate the dependence of the cross section $\sigma(x)$ on the collision energy as the sum of the series:

$$\sigma_{elastic}(x)=\sum_{i}\frac{A_i+B_i x^{C_i}}{1+D_i x^{E_i}}. \qquad (1)$$

Here the constants A_i, B_i values are like the cross section, have the dimension of the area, and the rest are dimensionless quantities. The value of the cross section for the collision of an electron with zero energy σ_0 is determined by the equality $\sigma_0 = \sum_i A_i$, which is found by solving the corresponding quantum mechanical problem. The parameters of (1) were obtained by a fitting procedure, minimizing the following summation by a program based on the method of coordinate descent

$$\Delta^2=\frac{1}{N}\sum_{i=1}^{N}\left[\frac{\sigma_{fit}(x_i)-\sigma_{\exp}(x_i)}{\sigma_{\exp}(x_i)}\right]^2, \qquad (2)$$

where $\sigma_{\exp}(x_i)$ are the measured values and $\sigma_{fit}(x_i)$ are the calculated cross sections in points $x_i : i=1, \ldots, N$. Minimizing (2), instead of minimizing the simple deviation, has the advantage of giving the correct statistical weight to cross sections at low and high impact electron energy. The same fitting procedure was used to evaluate excitation and ionization cross sections.

Even when using only two terms in formula (1), a satisfactory solution to the problem of minimizing the approximation error is obtained (2% – the root – mean-square relative error for helium and neon, 6-9% – for argon, krypton and xenon). When using three terms, the accuracy of the fit can be increased by two to three times, but since the errors in the input data are 10-20%, this makes no sense.

The fitting coefficients of electron transport cross sections in inert gases are given in Table 1. The collision energy must be expressed in dimensionless units *x = ε / I*, and the cross section is obtained in units $Å^2 = 10^{-16}cm^2 = 10^{-20}m^2$.

Let us note an important point to which attention should be paid when analyzing experimental data for the transport section. In inelastic collisions, the momentum of the incident electron also changes and, accordingly, inelastic collisions contribute to the deceleration of the electron flux as they drift in the gas. And since the excitation and ionization cross sections for noble gases can exceed the elastic collision cross section at energies of the order of 2-4 ionization potentials, the contribution of inelastic collisions to the transport cross section for energetic electrons can be the main one. To play elastic collisions in the Monte Carlo procedure, it is necessary to know the cross section of elastic collisions, since collisions with excitation and ionization are played separately. Therefore, one must take into account the difference in type possible collision: 'elastic', 'momentum', 'excitation', 'ionization', where momentum is the sum of the elastic and inelastic cross sections (useful for solving the Boltzmann equation in the 2-term approximation).

Table 1 – Values of parameters for approximation the transport cross sections for elastic collisions of electrons with noble gas atoms

Gas, *I, eV*	A_1 *Å^2*	B_1 *Å^2*	C_1	D_1	E_1	A_2 *Å^2*	B_2 *Å^2*	C_2	D_2	E_2	*Δ,* %
He, 24.584	0	7.19	1	3.67	2.79	5.16	6.09	0.41	15.0	1.91	1.7
Ne, 21.564	0	38.7	1	267.	1.64	0.31	2.99	0.50	0.20	1.93	1.7
Ar, 15.759	0	24.1	1	1.03	2.83	7.76	-65.5	0.455	1961	1.37	8.0
Kr, 13.996	0.17	115.	1.82	9.52	3.58	40.5	-101.	0.28	1275	1.40	6.1
Xe, 12.127	-3.1	182	1.53	10.1	2.8	136.	-143.	0.169	1453	1.37	9.0

Approximation of the excitation cross section

Excitation of atomic levels in many cases is the main channel of energy losses for electrons in the plasma of a gas discharge, and their correct consideration is very important. In noble gases, the first levels are located rather high, and for the excitation cross section near the excitation threshold E$_1$, a linear approximation of the dependence of the cross section on energy is sometimes used:

$$\sigma_{excitation}(\varepsilon) = C_{ex}(\varepsilon - E_1), \varepsilon > E_1 \qquad (3)$$

To approximate the excitation cross section in a wider energy range, we choose the formula

$$\sigma_{excitation}(\varepsilon) = \frac{A(\varepsilon / E_1 - 1)}{(\varepsilon / E_1 + B)^C}, \qquad (4)$$

where E_1 is the energy of excitation and *A, B, C* – adjustable parameters.

The coefficients of this approximation are given in Table 2, the collision energy should be expressed in eV, and the cross section is obtained in units of Å^2. There is also given the root-mean-square relative error, which for the considered gases is on the order of 2-6%. In addition, the table contains the constant of linear approximation of the initial section obtained from formula (4) $C_{ex} = A / (E_1(1+B)^C)$.

Table 2 – Values of parameters for the excitation cross sections approximation of noble gas atoms

Gas, *E$_1$, eV*	ε_{min} - ε_{max}	A, *Å^2*	B	C	*Δ,* *%*	ε_{m}, *eV*	$\sigma(\varepsilon_{\text{m}})$ *Å^2*	C_{ex}, *(4)* *Å^2/eV*	C_{ex}, [27] *Å^2/eV*
He, 19.8	*30-4000*	0.99	0.63	1.75	5.9	63	0.21	0.021	0.046
Ne, 16.619	*30-4000*	1.50	1.98	1.85	1.9	75	0.17	0.012	0.015
Ar, 11.50	*20-4000*	6.48	1.83	1.81	3.8	52	0.80	0.086	0.070
Kr, 9.915	*20-4000*	8.95	2.09	1.82	2.8	47	1.01	0.116	-
Xe, 8.315	*20-4000*	15.8	3.08	1.87	3.8	47	1.28	0.137	-

Approximation of the ionization cross section

Thomson in 1912 proposed the dependence of the ionization cross section on the electron energy of the following form:

$$\sigma_{ionization}(\varepsilon) = \frac{\pi e^4}{\varepsilon}\left(\frac{1}{I} - \frac{1}{\varepsilon}\right) \equiv \equiv 4\pi a_0^2 \frac{I_H^2(\varepsilon - I)}{I\varepsilon^2}, \qquad (5)$$

which is obtained for the case of a stationary valence electron at the energy of the incident electron $\varepsilon > I$. It gives a linear increase in the ionization cross section with a small excess of the collision energy over the ionization potential and reaches the maximum value $\sigma_{\max} = \pi e^4 / 4I^2$ at the energy of the incident electron $\varepsilon = 2I$. A more precise expression for the ionization cross section, which takes into account the spherically symmetric motion of the valence electron in the Coulomb field of the atomic residue, has the form:

$$\sigma_{ionization}(\varepsilon)=\frac{\pi e^4}{\varepsilon}\left(\frac{5}{3I}-\frac{1}{\varepsilon}-\frac{2I}{3\varepsilon^2}\right). \quad (6)$$

In this case, the maximum value $\sigma_{\max}\approx\pi e^4/2I^2$ at the energy of the incident electron $\varepsilon=1.85I$. The first experiments on measuring the dependence of the ionization cross section showed that the initial section of the curve is well described by a linear function up to the energies of the incident electron $\varepsilon<2I$, and the maximum ionization cross section for inert gases lies in the energy range $\varepsilon\in(3I,5I)$. For the first time, a semi-empirical formula for approximating the initial section of the dependence of the ionization cross section on the energy of the incident electron was proposed by Compton and Van Voorhees in 1925 [21]

$$\sigma_{ionization}(\varepsilon)=C_i(\varepsilon-I),\quad I<\varepsilon<2I. \quad (7)$$

Wannier proposed a power dependence with the exponent equal to 1.127 to approximate the initial section:

$$\sigma_{ionization}(\varepsilon)=C_i(\varepsilon-I)^{1.127},\quad \varepsilon>I. \quad (8)$$

This dependence takes into account the interaction of incident and bound electrons [22].

Lotz in [23, 24] analyzed the experimental and theoretical data available at that time and proposed a formula based on the Bethe-Born approximation, which has the form

$$\sigma_{ionization}(x)=[A\ln x+\sum_{k=1}^{N}B_k(\Delta x/x)^k]/xI^2, \quad (9)$$
$$x=\varepsilon/I, \Delta x=x-1, x>1$$

Here the energy is normalized to the ionization potential: $x=\varepsilon/I$, $\Delta x=x-1$, $x>1$, A, B_k, – fitting constants. The Lotz formula (9) takes into account the universal dependence of the cross section on the ionization potential and is consistent with the asymptotic behavior of the Bethe formula [25] $\sigma_{ionization}(\varepsilon)=(A\ln\varepsilon+B)/\varepsilon I$.

The problem of analytical fits for the ionization cross sections is discussed in [26], and different approaches are considered on the sample case of neon. The feasibility of the standard (9) formula is investigated, and a number of other analytical expressions is suggested, approximating ionization cross sections in the wide range of energies. The factors influencing the accuracy of the fits and the physical meaning of the parameters obtained are discussed. The formula (9) with N= 3 gives A=2767 Å^{2}eV^2, B_1 = 2196, B_2 =3124, B_3 = 1840.

We made the attempt to approximate the dependence of the ionization cross section on energy by two dependences that have the following form:

$$\sigma_{ionization}(x)=\frac{A(x-1)}{(x+B)^C}, \quad (10a)$$

$$\sigma_{ionization}(x)=\frac{A(x-1)^{1.127}}{(x+B)^C} \quad (10b)$$

where $x=\varepsilon/I$, A, B, C approximation constants. The dependence (10b) corresponds to the approximation of the initial curve part proposed by Wannier [22]

The search for the optimal parameters for approximating of the experimental data by formulas (10a) and (10b) showed that the errors of approximations (10a) and (10b) have the same order, which does not allow us to determine how valid the proposed Wannier dependence (8). At one time, this issue caused a very lively discussion [27, 28], but the accuracy of the experimental data did not allow either to confirm or reject the approximation of the initial part of the curve of the ionization cross section versus energy proposed by Wannier. The approximation coefficients (10a) are given in Table 3, the collision energy should be expressed in eV, and the cross section in Å^2.

Table 3 – Values of parameters for approximation the ionization cross sections of noble gas atoms

Gas, I,eV	$\varepsilon_{\min}$ - $\varepsilon_{\max}$, *eV*	A, *Å²*	B	C	*Δ, %*	ε_{m}, *eV*	$\sigma(\varepsilon_{\mathrm{m}})$ *Å²*	*C_i, (10a) Å²/eV*	*C_i, [27] Å²/eV*
He, 24.587	*30-4000*	3.95	2.48	1.91	2.8	119	0.34	0.015	0.013
Ne, 21.564	*30-4000*	20.11	6.34	2.00	6.3	180	0.68	0.017	0.016
Ar, 15.759	*20-4000*	30.10	2.51	1.86	2.7	80	2.83	0.185	0.20
Kr, 13.996	*20-4000*	37.39	2.72	1.80	2.9	79	3.80	0.251	-
Xe, 12.127	*15-4000*	46.38	2.86	1.76	6.2	74	4.99	0.355	-

The approximation coefficients according to the formula (10b) give a close error value, so they are not given in the table. The root-mean-square relative error is also given there, which for the considered gases is about 2-6%. In addition, the same table shows the constant of the linear approximation of the initial section $C_{ion} = \mathrm{A}/(I(1+D)^{C})$ obtained from formula (4).

Discussion and conclusions

During selecting data for approximating the energy dependence of the electron-atomic collision cross sections, we used various analytical formulas, which contain both original experimental data and data from various databases [3-6, 11-17].

The fitting curves and the experimental data are shown in Figs. 1- 5 for He, Ne, Ar, Kr, Xe respectively. On each graph, the experimental values of the cross sections are represented by markers, and the solid curves represent the fits found, as well as the root-mean-square values of the approximation errors. For He and Ne from BOLSIG+, for Ar and Xe from Puech database (www.lxcat.net), for Kr from SIGLO database (www.lxcat.net). The data of the excitation and ionization cross sections for He are taken from [15], for Ne, Ar, Kr and Xe from [16].

Helium. The data for elastic collision cross sections are taken from BOLSIG+ . This data are from the compilation of A.V. Phelps (ftp://jila.colorado.edu/collision_data/, momentum transfer – from Crompton et al at low energy, Hayashi at high energies).

Neon. The data for elastic collision cross sections are taken from BOLSIG+. Same as in siglo.sec except extrapolation of allowed cross sections to 1 kV using log (energy)/energy scaling.

Argon. There is a large amount of data on argon due to its popularity and cheapness. The data for elastic collision cross sections are taken from Puech database.

Krypton. The data for elastic collision cross sections are taken from SIGLO database. The values of the experimental data for the transport cross section given in this database for Krypton at high energy have the same asymptotes as inelastic collisions, which prevail at high energies. Therefore, when approximating the transport cross-section in elastic collisions, the last 3 points out of 50 were not taken into account when selecting the fit coefficients.

Xenon. The data for elastic collision cross sections are taken from Puech database. As in the case of Krypton, the values of the experimental data for the transport cross section given in this database for Xenon at high energy have the same asymptotes as inelastic collisions. Therefore, when approximating the transport cross-section in elastic collisions, the last 6 points out of 86 were not taken into account when selecting the approximation coefficients.

In our previous papers [29-32], which have been published since 2010 [29-32], linear approximations were used to calculate the excitation and ionization frequencies. Test calculations with the new data showed that the differences in the drift characteristics for the data domain corresponding to the physics of low-temperature plasma (gas discharge) are insignificant. But for pulsed discharges, where high-energy collisions and inelastic interaction are important, the differences can be significant.

This paper provides a brief overview of the available data on the cross sections of collisions of electrons with noble gas atoms, and analyzes them to select the most reliable ones. Formulas for the analytical approximation of the cross sections of elastic (transport) and inelastic (ground-state excitation and ionization) collisions of electrons with noble gas atoms are obtained for them, which have an error of the same order as the experimental and theoretical data available in the literature.

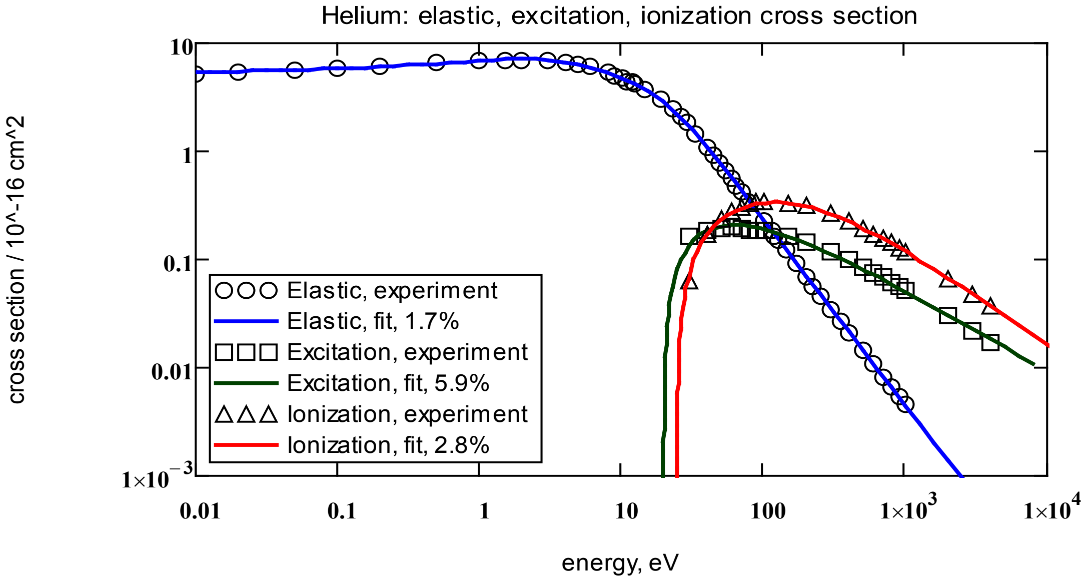

Figure 1 – Dependence of the cross sections of the collision of electrons with helium atoms on the energy.

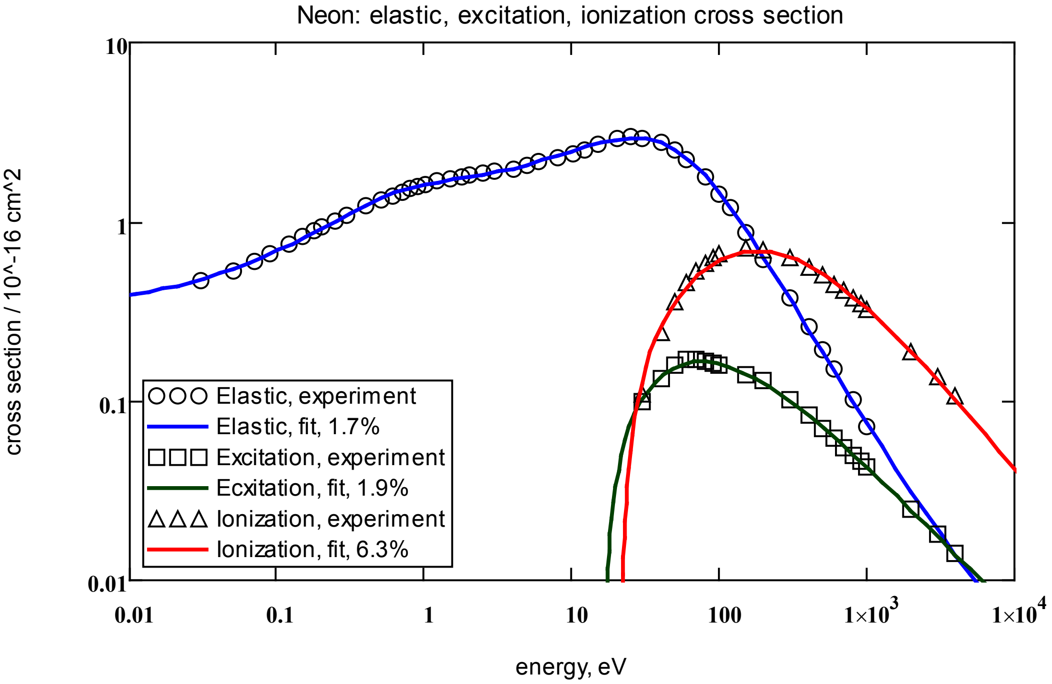

Figure 2 – Dependence of the cross sections of the collision of electrons with neon atoms on the energy.

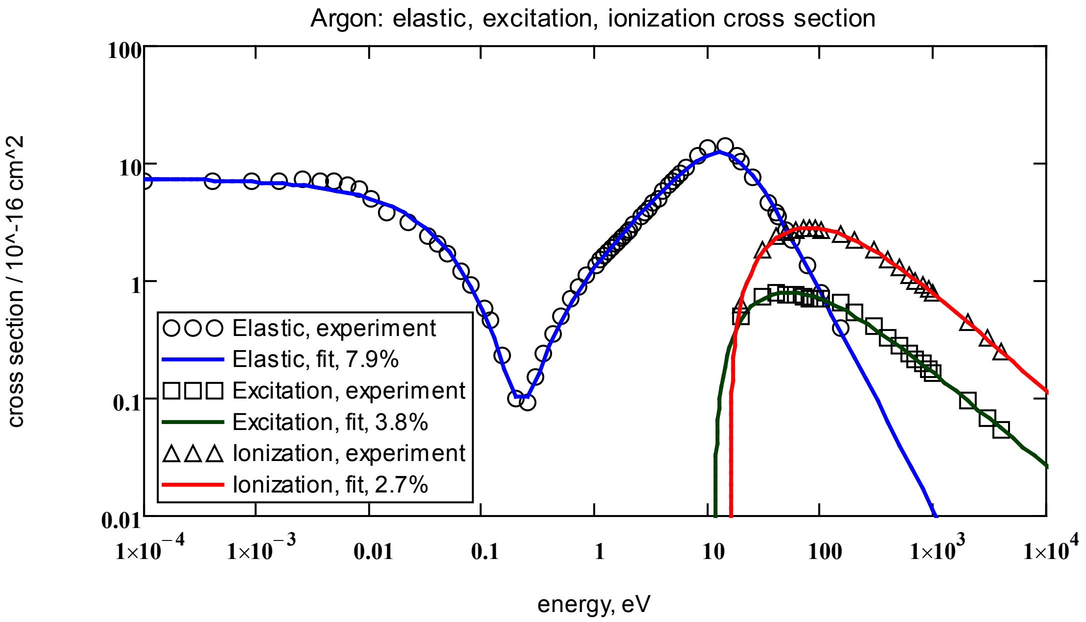

Figure 3 – Dependence of the cross sections of the collision of electrons with argon atoms on the energy.

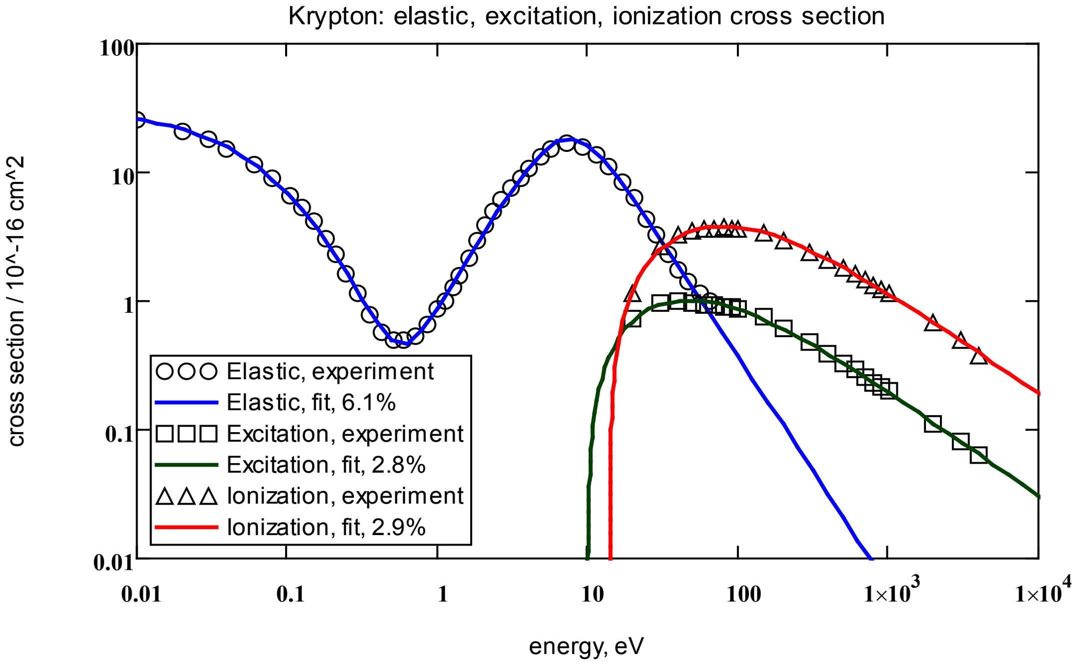

Figure 4 – Dependence of the cross sections of the collision of electrons with krypton atoms on the energy.

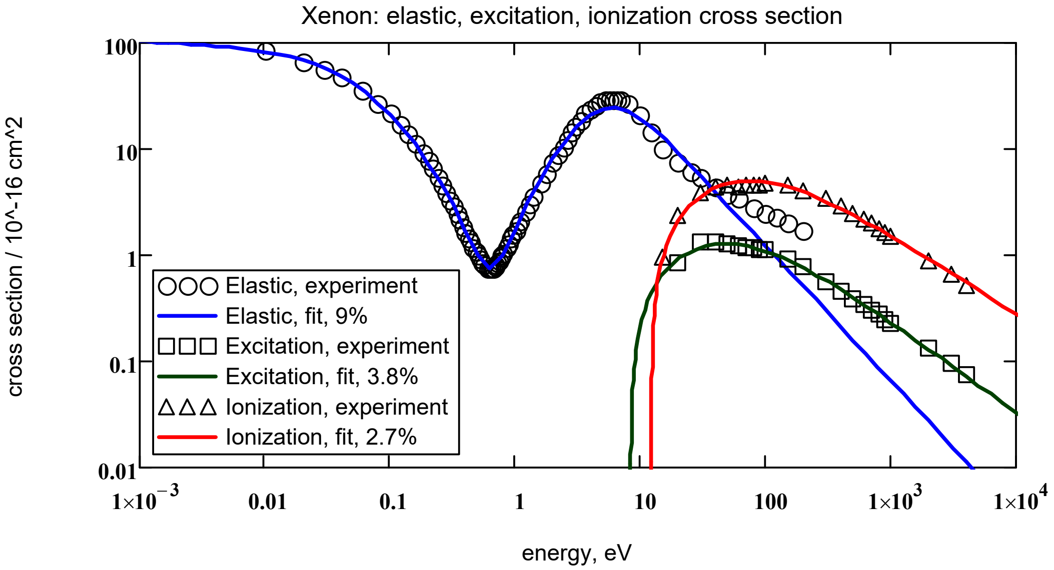

Figure 5 – Dependence of the cross sections of the collision of electrons with xenon atoms on the energy.

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