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Ionization cross section of noble gas atoms by electron impact

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The paper presents an analysis of data on the cross sections for ionization by electron impact of noble gas atoms such as hydrogen, helium, neon, argon, krypton and xenon. For the selected sets of experimental and theoretical data an analytical formula is proposed, based on separate accounting for the knockout of electrons from the outer and inner shells, and the corresponding approximation coefficients are selected. By single-term and two-term analytical dependences, approximation errors and coefficients of the ionization cross sections of noble gas atoms were obtained. The obtained semi-empirical formula reproduces the values of the ionization cross sections in a wide range of energies with an accuracy of several percent. Energy dependence of the ionization cross section for an electron collision with a noble gas atoms were calculated and compared with available experimental data. The analysis of the approximation coefficients makes it possible to reduce the influence of errors in the initial experimental data and significantly increase the accuracy of estimating the ionization cross sections.

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

The bibliography on cross sections of electron-atomic collisions has thousands of works, and probably an exhaustive review and selection of data is contained in the works [1–6]. However, it should be borne in mind that a critical analysis of the results of experimental data in the review work is very difficult due to the fact that the errors given in the original works of the order of 1–3% differ from each other sometimes by 50%. Therefore, in the review work, only a comparative analysis of the results obtained is really possible, which shows that at best, the relative errors of measuring cross sections are of the order of 5–10%, and more often 20–50%, sometimes reaching 100%.

The most convenient form of presenting experimental and computational-theoretical data is the selection of analytical approximations for them. We began a critical analysis and assessment of the cross sections for electron scattering by atoms of noble gases and vapors of some metals in a wide range of energies in [16–20], where approximations were proposed for the cross sections of elastic and inelastic collisions of electrons with rare gas atoms. Ionization by electron impact from the ground state of the atom is the most common method for the formation and maintenance of a gas-discharge plasma. From a large number of experimental and calculated data on the cross sections for ionization of atoms by electron impact, we have chosen by comparative analysis such data that allowed us to significantly expand the range of applicability of the proposed analytical dependences.

2 Approximation of the ionization cross section

The formulation of the problem of finding an analytical approximation of the ionization cross section of an atom by an electron impact is based on the use of known analytical estimates, the results of experimental measurements and numerical quantum mechanical calculations. In 1912, Thomson proposed the dependence of the ionization cross-section on the electron energy of the following form [21]:

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

which is obtained for the case of a stationary valence electron at the energy of the incident electron $\varepsilon > I$. Here, *e* is elementary charge, *I* is the first ionization potential, a_0 is Bohr radius, *Ry* is ionization energy of a hydrogen atom. Formula (1) gives a linear increase in the ionization cross section at a small excess of the collision energy over the ionization potential and reaches the maximum value $\sigma_{\max}=\pi e^4/4I^2=\pi a_0^2(Ry/I)^2$ at the energy of the incident electron $\varepsilon=2I$, Ry=13.6 eV, $\sigma_0=\pi e^4=\pi a_0^2=0.876\text{Å}^2$.

The first experiments on measuring the dependence of the ionization cross section on the energy showed that, in full accordance with Thomson formula (1), the initial part of the curve is described fairly well by a linear function up to energies of the incident electron $\varepsilon<2I$. But the maximum ionization cross section for all inert gases lies in the energy range $\varepsilon\in(3I,7I)$. For the first time, a semi-empirical formula for approximating the initial part of the dependence of the ionization cross section on the energy of the incident electron was proposed by Compton and van Voorhees in 1925 [22]:

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

Wannier proposed a power-law dependence with an exponent of 1.127 to approximate the initial part:

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

which takes into account the interaction of the incident and bound electrons [23, 24].

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 $x=\varepsilon/I$. In our works [17-20] for atoms of noble gases, hydrogen and some metals, a formula is proposed for the cross section of ionization of an atom by electron impact with three approximation coefficients α, β, γ:

$$\sigma_{ionization}(x)=\frac{\alpha\Delta x}{(1+\beta\Delta x)^{\gamma}}, \quad (2)$$

где $x=\varepsilon/I$, $\Delta x=x-1$, $x > I$. For $\alpha=4\pi a_0^2(Ry/I)^2$, $\beta=1$, $\gamma=2$ it coincides with Thomson's formula (1). The errors of approximation of experimental data by analytical dependence (2) for noble gases and vapors of some metals are in the range of 3-7%, which corresponds in order of magnitude to the errors of the experiments themselves. The maximum cross-section value according to (2) is achieved at $\Delta x=1/(\beta(\gamma-1))$.

To approximate the functional dependences of the cross sections on the collision energy, the sum of the series is often used, and the number of terms of the series used can be quite large. At a sufficiently high electron energy, ionization of the atom due to the knocking out of electrons from the inner shells of the atom can play a significant role. To take this factor into account, Lotz in [25-27] analyzed the experimental and theoretical data existing at that time and, within the framework of the shell model of the atom, proposed a formula that has the form

$$\sigma_{ionization}(\varepsilon)= \\ =\sum\nolimits_1^n a_i q_i(1-b_i\exp(-c_i\Delta x_i))\ln(x_i)/x_i, \quad (3)$$

where q_1 is a number of electrons on the outer shell, q_i is the number of electrons on the next i-th inner shell, I_1 – first ionization potential of an atom, I_i is an ionization energy from the i-th inner shell of the atom, $x_i=\varepsilon/I_i$, $\Delta x_i=x_i-1$, $x_i > 1$, a_i, b_i, c_i – fitting parameters. When using two or three terms of series (3), he obtained the coefficients of approximation of experimental data with an accuracy of 10-20%.

In this work, an attempt is made to take into account the ionization of an atom due to the knocking out of electrons from the inner shells on the basis of our earlier proposed formula (2). A logical way to take into account this ionization channel is to use a two-term approximation, in which the first term corresponds to ionization from the outer shell, and the second term describes ionization from the second shell

$$\sigma_{ionization}(\varepsilon)=\mathrm{q}_1\frac{\alpha_1\Delta x_1}{(1+\beta_1\Delta x_1)^{\gamma_1}}+\mathrm{q}_2\frac{\alpha_2\Delta x_2}{(1+\beta_2\Delta x_2)^{\gamma_2}}, \quad (4)$$

where $x_1=\varepsilon/I_1, \Delta x_1=x_1-1,\ x_1 > 1$, and $x_2=\varepsilon/E_2, \Delta x_2=x_2-1,\ x_2 > 1$.

If we put all the fitting coefficients to be the same for both shells, then in the two-term approximation we obtain a formula with three fitting coefficients α_0, β_0, γ_0:

$$\sigma_{ionization}(\varepsilon) = q_1 \frac{\alpha_0 \Delta x_1}{(1+\beta_0 \Delta x_1)^{\gamma_0}} + q_2 \frac{\alpha_0 \Delta x_2}{(1+\beta_0 \Delta x_2)^{\gamma_0}} \qquad (5)$$

To determine the coefficients α_0, β_0, γ_0, the problem of minimizing the root-mean-square deviation of the cross sections from their experimental values was solved by the standard method of coordinate descent:

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

where $\sigma_{\exp}(x_i)$ – experimental values in points x_i : $i=1,...,N$, and $\sigma_{fit}(x_i)$ – cross-section values calculated by approximating functions. Minimizing the relative error instead of minimizing the absolute value of the error makes it possible to more correctly take into account the statistical weight of the cross sections at low and high electron energies, when the absolute values of the cross sections are small. However, the choice of optimization based on relative rather than absolute error leads to a greater error in determining the position of the maximum value of the ionization cross section. And in experiments it is the maximum value of the ionization cross section that is most accurately measured.

Table 1 shows the characteristics of noble gas atoms from [26-27] and experimental data on the measurement of ionization cross sections from [28-30]: the symbol and number of the atom, the static polarizability coefficient of the atom K_0, the first ionization potential I1 and the ionization potentials of the atom I_2, I_3 from the next two (second and third) inner shells, the number of electrons q_1, q_2 and q_3 on the outer and next two inner shells, as well as the energy range for which the experimental values of the ionization cross sections were obtained, their number, our estimate of the experimental data error, and a reference to the source of the experimental data.

Note that the energy of the first ionization of an atom due to the knocking out of electrons from the inner shells of an atom is usually slightly higher than the energy of the second ionization potential, i.e. the ionization potential of a singly ionized atom. Binding energies have been taken from paper [27]. A helium atom has two electrons forming one shell (q_1=2, q_2=0), therefore, for helium, formula (5) is a one-term approximation.

Table 1 – Characteristics of atoms and experimental results on measuring the ionization cross sections of noble gases

Atom characteristics								Experiment Data			
Symbol, Number	**K_0,a^3_0**	**I_1, eV**	**I_2, eV**	**I_3, eV**	**q_1**	**q_2**	**q_3**	**$\varepsilon_1 \div \varepsilon_N$, eV**	**N**	**Δ, %**	**Ref.**
H, 1	4.5	13.6	-	-	1	-	-	15- 4000	10	2%	Shah, 1987
He, 2	1.383	24.587	-	-	2	-	-	30 – 4000	21	2-3%	Heer, 1977
Ne, 10	2.68	21.564	48.5	869.5	6	2	2	30 – 4000	21	7%	Heer, 1979
Ar, 18	11.08	15.759	29.2	247.7	6	2	6	20 – 4000	22	6%	Heer, 1979
Kr, 36	16.74	13.996	27.5	93.7	6	2	10	20 – 4000	22	7%	Heer, 1979
Xe, 54	27.06	12.127	23.4	68.1	6	2	10	15 – 4000	23	11%	Heer, 1979

3 Results

The results of approximation of the ionization cross sections of noble gas atoms by electron impact by analytical dependencies (2) and (5) with three fitting coefficients are shown in Table 2. For comparison, there are also given the parameters of approximation of the ionization cross section of the hydrogen atom in the one-term approximation according to formula (2). In addition to the values of the approximation coefficients for these two approximations, the relative root-mean-square approximation errors for the single-term Δ_1 and two-term Δ_2 approximations are also given.

Table 2 – Coefficients and errors of approximation of the ionization cross sections of noble gas atoms by single-term and two-term analytical dependences.

	α, Å^2	α_0, Å^2	β	β_0	γ	γ_0	Δ_1, %	Δ_2, %
H	0.827	-	0.351	-	1.91	-	2.0%	-
He	0.365	-	0.287	-	-	1.92	2.8%	-
Ne	0.383	0.0675	0.152	0.204	1.92	1.92	7.0%	7.3%
Ar	2.92	0.452	0.285	0.321	1.86	1.92	2.8%	5.5%
Kr	3.51	0.523	0.269	0.270	1.80	1.92	2.9%	7.4%
Xe	4.30	0.588	0.259	0.230	1.76	1.92	6.2%	11%

The results of the search for the minimum of the approximation error for the two-term approximation showed that for all gases the exponent is practically the same and its value is approximately equal to 1.92. Therefore, the parameter γ was fixed and set equal to 1.92, and the error was optimized for only two parameters.

Figures 1 – 5 show the plots of the ionization cross sections for all considered inert gases: helium, neon, argon, krypton and xenon, respectively. On each plot, the experimental data are marked with circles, the two-term analytical approximations are solid curves, the single-term ones are dashed lines, and the cross-section components corresponding to ionization from different shells in the two-term approximation (5) are dash-dotted lines. The splitting of the cross sections into ionization from the first and second shells shown in the plots for neon, argon, krypton, and xenon clearly shows the physical validity of the two-term approximation.

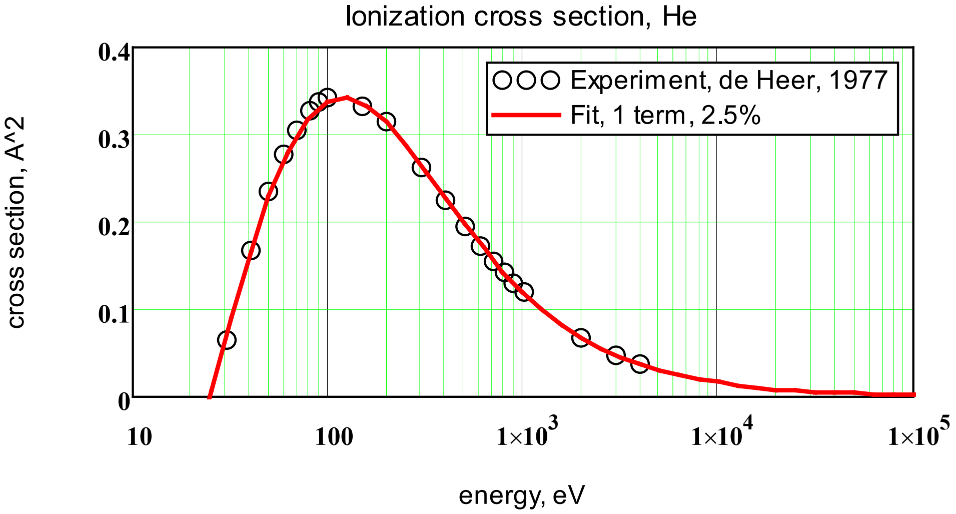

Figure 1 – Energy dependence of the ionization cross section for an electron collision with a helium atom: circles – experimental data, solid curve – approximation in the one-term approximation with a rms relative deviation of 2.5%

In a helium atom, the entire electronic system consists of two electrons, which form only one – the outer shell. The approximation of the cross section by two terms (4) with six fitting parameters and $I_1 = E_2 = 24.587\, eV$ leads to a very insignificant decrease in the relative error – from 2.8% to 2.1%. Obviously, such a decrease in the error when using a two-term approximation with six parameters is due to an increase in the number of fitting coefficients from three in formula (2) to six in formula (5). On this basis, we estimate the error of the experimental data [29] for helium at 2–3%. The maximum cross-section value according to (2) is achieved at $\Delta x = 1/(\beta(\gamma-1)) \approx 1/\beta$. Accordingly, for the found approximation of the cross section, its maximum is 0.34 Å^2 and is reached at an energy of 125 eV.

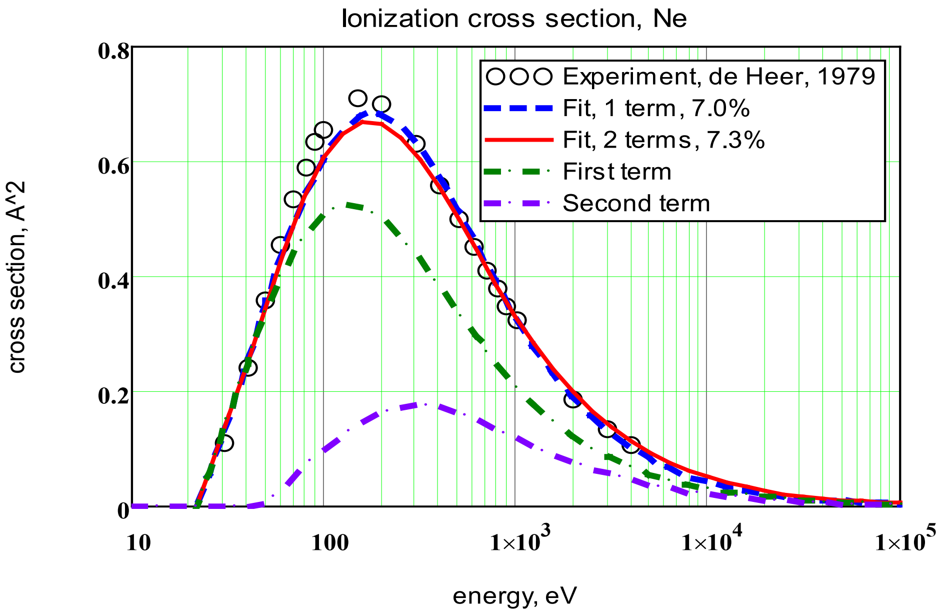

Figure 2 – Energy dependence of the ionization cross section for an electron collision with a neon atom: circles – experimental data, dashed and solid curves – approximation in the one-term and two-term approximations, dash-dotted curves – the first and second terms of the approximation

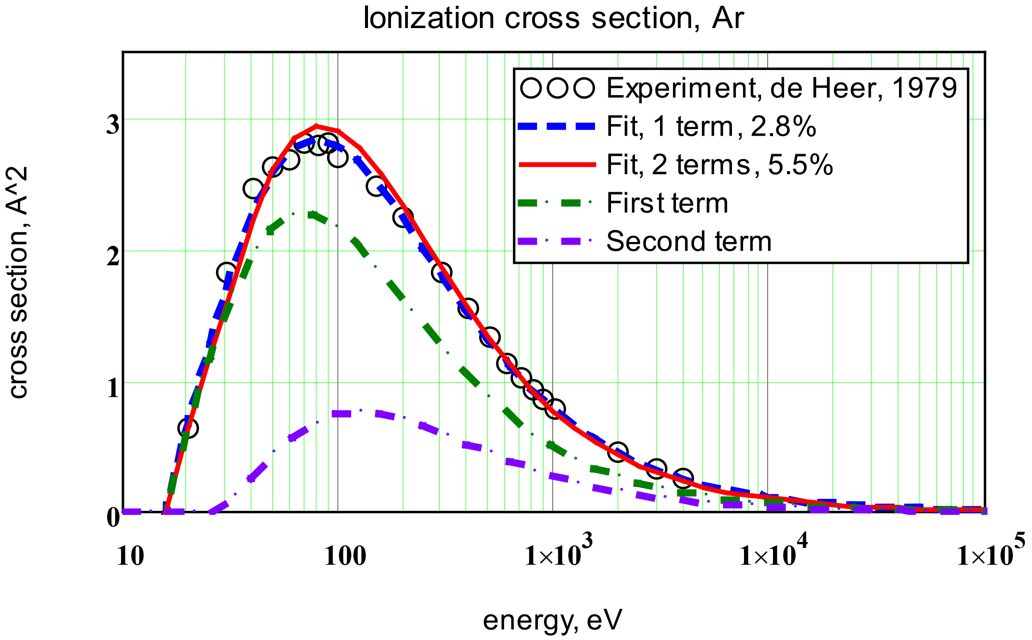

Figure 3 – Energy dependence of the ionization cross section for an electron collision with a argon atom

The neon atom has three shells – the outer shell has 6, the second and the inner one also have two electrons (10 in total). Ionization from the third shell is insignificant due to the large ionization potential – 869 eV. The approximation of the cross section in the two-term approximation leads to an insignificant increase in the relative error – from 7% to 7.3%. According to the found approximation of the cross section, its maximum is 0.67 Å^2 and is reached at an energy of 160 eV. We estimate the error of experimental data [30] for neon at 7%.

The argon atom has 6 and 2 electrons on the outer and next inner shells, respectively, and the remaining shells closest to the nucleus are clearly not taken into account in the two-term approximation. Argon has six electrons on the third shell with an ionization potential of 248 eV. When choosing the approximation coefficients, naturally, their contribution to ionization is taken into account through the contribution of the experimentally measured cross sections. We estimate the error of experimental data [30] for argon at 6%. According to the found approximation, the maximum value of the cross section is 2.9 Å^2 and is reached at an energy of 90 eV.

The krypton atom has 6 and 2 electrons on the outer and next inner shells, respectively. As in argon, ionization of the shells closest to the nucleus affects the asymptotics of the dependence of the ionization cross section at high energies. In the two-term approximation, their influence is not explicitly taken into account. On the third shell, krypton has ten electrons with an ionization potential of 93.7 eV. Therefore, ionization from the third shell for krypton has a large effect on the ionization cross section near the maximum. In addition, after knocking out an electron from the third shell, autoionization of an electron from the outer shell is possible. To improve the accuracy for krypton and xenon, it is necessary to take into account the ionization from the third shell, i.e. use the three-term approximation [25]. However, when choosing the approximation coefficients and in the two-term approximation, their contribution to the ionization of the atom is naturally taken into account, since their contribution is present in the experimentally measured cross sections. We estimate the error of experimental data [30] for krypton at 7%. According to the found approximation of the cross section, its maximum is 3.9 Å^2 and is reached at an energy of 85 eV.

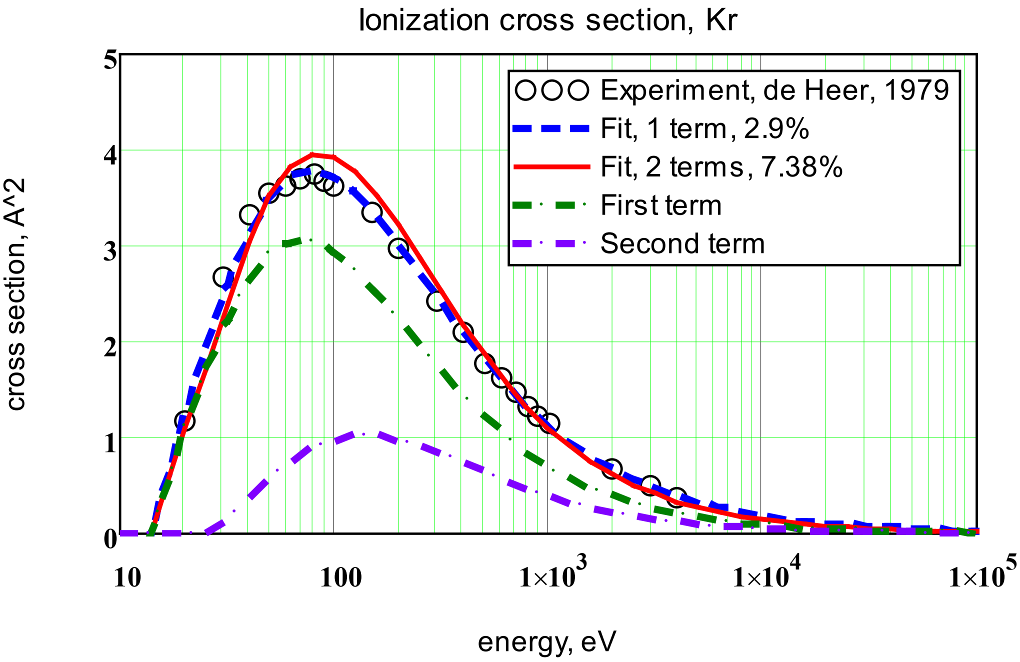

Figure 4 – Energy dependence of the ionization cross section for an electron collision with a krypton atom

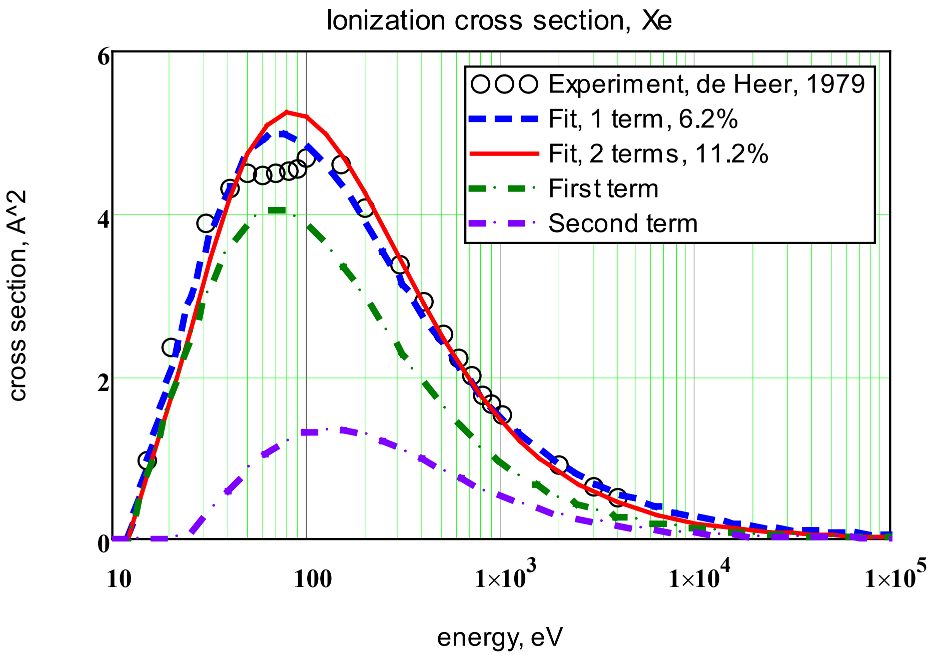

Figure 5 – Energy dependence of the ionization cross section for an electron collision with a xenon atom

The xenon atom has 6 and 2 electrons on the outer and next inner shells, respectively. On the third shell, xenon, like krypton, has ten electrons with an ionization potential of 68.1 eV. Therefore, ionization from the third shell for xenon has a large effect on the ionization cross section near the maximum. In addition, after knocking out an electron from the third shell, autoionization of an electron from the outer shell is possible. Ionization by knocking out electrons from deep shells strongly affects the asymptotics of the dependence of the cross section at high energies. The approximation of the cross section in the two-term approximation leads to an increase in the relative error from 6.2% to 11.2%; therefore, we estimate the error of the experimental data [30] for xenon at 11%. According to the found approximation of the cross section, its maximum is 5.2 Å^2 and is reached at an energy of 80 eV.

4 Conclusions

As follows from the data presented in the tables, the use of a two-term approximation purely formally leads to a significant increase (up to two times) in the error in approximating experimental data by analytical dependences. However, the addition of the second term is not accompanied by the addition of new adjustable constants – their number is still three. It can be assumed that a physically more adequate model, which takes into account the ionization of strongly bound electrons from the inner shells, gives more accurate data on real cross sections. The answer to this question can be obtained by analysis and comparison with various experimental data and the transition to consideration of other elements [31, 32]. In particular, for alkaline earth metals, which have only one electron on the outer shell, the effect of ionization from the inner shells will be greatest. Such a study is planned for the future.

The analysis of the obtained analytical dependences of the ionization cross sections of inert gas atoms by electron impact, together with the analysis of the corresponding experimental data, allows us to draw several important and interesting conclusions:

1. Since helium has only one electron shell, the variation of the coefficients of the first and second terms of the approximation for helium can apparently serve as an additional estimate of the experimental error. For helium an increase in the number of expansion terms from one to two does not lead to any noticeable increase in accuracy,

which indicates that the error is apparently caused not by the functional form of formula (2), but by errors in the experimental data.

2. In contrast to the one-term approximation (2), when using an approximation that takes into account ionization from the second shell, the value of the parameter γ can be chosen the same for all elements and equal to 1.92.

3. The division of the cross sections into ionization from the first and second shells shown in the graphs for neon, argon, krypton and xenon clearly indicates the physical validity of the two-term approximation. Therefore, the carried out mathematical processing of the experimental data makes it possible to achieve a significant increase in the accuracy in determining the cross sections by finding the analytical dependences obtained from the experimental data.

4. In addition, the analysis of the approximation parameters makes it possible to find their values for elements for which there are no experimental data.

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