

A SEMI-EMPIRICAL FORMULA FOR CALCULATION OF ABSOLUTE CROSS SECTIONS FOR IONIZATION AND EXCITATION OF ATOMS BY ELECTRONS

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Synopsis

A simple analytical expression for the absolute ionization and (optically allowed) excitation cross sections, as a function of the electron energy, is "derived". In this expression there are two parameters. The first one is proportional to the optical oscillator strength and the second one is dependent on the shape of the differential cross section (generalized oscillator strength), for scattering of energetic electrons as a function of their (small) momentum loss. The analytical expression is exact for large energies of the impinging electron. For small energies of this electron, the expression gives good agreement with experiment. Ionization cross sections are calculated for H, He, Ne, Ar, Kr, Xe, H₂, N₂ and O₂. Excitation cross sections are calculated for H and He.

Theory. In the following we restrict ourselves to (optically allowed) excitation and ionization of atoms. The first part of the theory is based on a paper of Miller and Platzman¹). We briefly summarize those points of their paper which are important in the present investigation. For high energies E_1 of an incident electron, the differential cross section for a collision in which this electron is scattered with momentum loss $\hbar K$ is given, to the accuracy of the first Born approximation, by:

$$\sigma_n(K) dK = \frac{8\pi a_0^2 R^2}{E_1 U_n} f_n(K) \frac{dK}{K} \quad (1)$$

where a_0 is the radius of the first Bohr orbit of hydrogen, R is the Rydberg energy (binding energy of the electron in the first Bohr orbit of hydrogen), U_n is the excitation energy of (final) state n (in the case of excitation) and $f_n(K)$ is the generalized oscillator strength defined by Bethe (see ref. 1). The total excitation cross section is¹):

$$Q_n = \frac{4\pi a_0^2 R^2}{E_1 U_n} \int_{K=K_{\min}}^{K_{\max}} f_n(K) d \ln(K^2 a_0^2) \quad (2)$$

where K_{min} and K_{max} are given in ref. 1. Following Miller and Platzman, we represent eq. (2) in fig. 1 by a graph of $f_n(K)$ against $\ln(K^2 a_0^2)$. For each excitation or ionization process, such a graph gives all information on the differential and total cross sections (for all values of E_1 great enough to ensure validity of the Born approximation). For $Ka_0 \ll 1$, the ordinate is a constant and equal to the optical oscillator strength f_n . The total cross

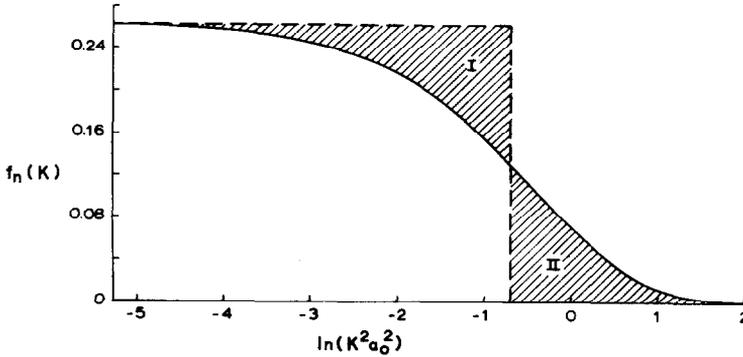


Fig. 1. Solid curve: generalized oscillator strength for the 2^1P state of He according to Miller and Platzman. Dashed curve: explained in the text. Area I = area II.

section Q_n (divided by $4\pi a_0^2 R^2 / E_1 U_n$) is the area under the solid curve between the limits given by K_{min} and K_{max} . This area is equal to the area of the rectangle (dashed curve in fig. 1) if area I = area II. The rectangle is bounded by ordinates f_n and 0, and abscissa $\ln(U_n^2 / 4E_1 R)$ and $\ln(c_n U_n^2 / 4R)$, where the constant c_n depends on the shape of the solid curve given in fig. 1. Thus c_n is defined by the expression $\ln(c_n U_n^2 / 4R)$ for the right hand side abscissa of the rectangle, so that this abscissa satisfies the condition I = II. Consequently eq. (2) may be written as:

$$Q_n = \frac{4\pi a_0^2 R^2}{E_1 U_n} f_n \ln c_n E_1 = \frac{A_n}{E_1} \ln c_n E_1 \tag{3}$$

where

$$A_n = 4\pi a_0^2 R^2 f_n / U_n. \tag{4}$$

For ionization of atoms, the expression corresponding to eq. (3) is:

$$Q_i = \frac{4\pi a_0^2 R}{E_1} M_i^2 \ln c_i E_1 = \frac{A_i}{E_1} \ln c_i E_1 \tag{5}$$

where

$$M_i^2 = \int_{U_i}^{\infty} \frac{R}{E} \frac{df}{dE} dE \tag{6}$$

df/dE being the differential oscillator strength (for energy transfer in the region between E and $E + dE$) and U_i the ionization energy. The relation between M_i^2 and the optical oscillator strength for transitions to the continuum is:

$$M_i^2 = \alpha' f_i R / U_i, \quad \text{where} \quad \alpha' = \int_{U_i}^{\infty} \frac{U_i}{E} \frac{df}{dE} dE \bigg/ \int_{U_i}^{\infty} \frac{df}{dE} dE. \quad (7)$$

Thus $\alpha' < 1$ for all atoms and molecules.

Our purpose is, to calculate ionization and excitation cross sections for large E_1 and for small E_1 . For small E_1 , eqs. (3) and (5) are no longer valid because the Born approximation is not valid. Still we can use a graph such as given in fig. 1. The ordinate no longer represents the generalized oscillator strength, but still gives the differential cross section for momentum changes $\hbar K$. It is to be expected that the shape of the curve will change. In eqs. (3) and (5) this means that $A_{i,n}$ and $c_{i,n}$ become energy dependent. At the threshold ($U_{i,n} = E_1$) only one momentum change is possible, $K_{\min} = K_{\max}$ and the two abscissa in fig. 1 are the same. Thus $\ln c(E_1) E_1 = 0$ and $c(E_1) E_1 = 1$ at threshold. Now we try (as working hypothesis):

$$c(E_1) E_1 = 1 + (E_1 - U_{i,n}) c_{i,n} \quad (8)$$

This is the simplest energy dependence resulting both in a correct high energy behaviour and threshold value of $c(E_1)$. The function $A(E_1)$ is a constant (eq. 4) for large E_1 and is expected to decrease for small E_1 . It is not known how fast this decrease will be. We try (as working hypothesis) the following energy dependence of $A(E_1)$ (see also the discussion):

$$A(E_1) = \frac{(E_1 - U_{i,n})}{E_1} A_{i,n} F \quad (9)$$

where

$$F = 1 + (1 - cU_{i,n})(0.025 + 1.6/cU_{i,n})(1 - U_{i,n}/E_1)(U_{i,n}/E_1)^{\frac{1}{2}}$$

For $E_1 \gg U$ is $F = 1$ and for small E_1 , F is only considerably different from 1 for $cU \gg 1$ or $cU \ll 1$. The numbers 0.025 and 1.6 are chosen in order to obtain a good agreement between theory and (corrected) experiment for H, H₂ and Kr. Thus:

$$Q_{i,n} = AF \frac{(E_1 - U_{i,n})}{E_1^2} \ln [1 + c(E_1 - U_{i,n})], \quad (10)$$

where A and c are the constants $A_{i,n}$ and $c_{i,n}$, and the expression F is given above. This semi-empirical formula gives essentially correct cross sections for large E_1 (e.g. $E_1 > 500$ eV). For small E_1 , the agreement between calculated (eq. 10) and experimentally determined cross sections is very

good as is shown in the following section. Equation (10) contains no single free parameter because A is determined by the oscillator strengths (eqs. 4 and 6) and c is determined by the dependence of $f_n(K)$ on K for large E_1 .

Comparison with experiment. To calculate cross sections with eq. (10), it is necessary to know the values of A and c . A number of these values are determined by Schram *e.a.*²⁾. They measured absolute ionization cross sections for fast electrons (0.6–20 keV) incident on He, Ne, Ar, Kr, Xe, H₂, D₂, N₂, and O₂. Their values of M_i^2 are in good agreement²⁾ with values of M_i^2 derived from photo-ionization cross sections, proton ionization cross sections and with theoretically calculated values of M_i^2 . For excitation and ionization of atomic hydrogen, values of the oscillator strengths are given by Allen³⁾. For excitation¹⁾ of He,¹⁾ values of f_n have been measured⁴⁾ ⁵⁾ and calculated (see table 2 of ref. 5). To determine the value of M_i^2 for ionization of atomic hydrogen, we made use of the following sum rule for the oscillator strengths (see for instance ref. 2 where this sum rule is used in the same manner):

$$\sum_{n=2}^{\infty} f_n R/U_n + M_i^2 = \langle r^2 \rangle / 3$$

where \mathbf{r} is the position vector of the atomic electron (in units a_0) and $\langle r^2 \rangle$ is the expectation value of r^2 . From the known values of f_n and $\langle r^2 \rangle$ it follows that $M_i^2 = 0.284$. To determine the values of c for excitation and ionization of atomic hydrogen, we made use of the known values of f_n , M_i^2 and of the absolute cross sections Q (for $E_1 = 1000$ eV) calculated with the first Born approximation⁶⁾. Thus the accuracy of these values of c depends on the accuracy of the Born cross sections for $E_1 = 1000$ eV. In table I, the obtained values of $f_{n,i}$, M_i^2 , A and c are listed.

With eq. (10) and with the values of A and c given in table I, we calculated several ionization and excitation functions. In table II, the calculated cross sections for ionization, and for excitation to the $2p$ -level, of atomic hydrogen are listed. This table also contains the cross sections obtained from different experiments (Fite *e.a.*⁷⁾, Rothe *e.a.*⁸⁾, and Boyd *e.a.*⁹⁾), and cross sections calculated with the first Born approximation⁶⁾.

From table II, it follows that the agreement between the present calculated values of Q and experiment is much better than the agreement between the Born cross sections and experiment. The ionization cross sections of He, Ne, Ar and H₂ are given in fig. 2. The solid curves are calculated with eq. (10) and the dashed curves represent the measurements of Tate and Smith¹⁰⁾ (H₂), Smith¹¹⁾ (He) and Bleakney¹²⁾ (Ne and Ar). From fig. 2 it follows that the agreement between the present calculations and experiment is very good except for Ne. This discrepancy for Ne will be explained in the discussion. Table III contains cross sections, calculated with

TABLE I

Values of $f_{n,i}$, M_i^2 , A and c						
Process	$f_{n,i}$	M_i^2	$A \cdot 10^{16}$ ($\text{cm}^2 \text{eV}$)	c (eV^{-1})	reference(s)	
H \rightarrow H ⁺	0.4359	0.284	13.6	5.62	3	6
H \rightarrow H 2p	0.4162		26.7	0.103	3	6
He \rightarrow He ⁺		0.489	23.4	0.108	2	
He \rightarrow He 2 ¹ P	0.276		8.5		4	5
He \rightarrow He 3 ¹ P	0.074		2.09		4	5
He \rightarrow He 4 ¹ P	0.03		0.82		4	5
Ne \rightarrow Ne ⁺		1.87	89.5	0.0319	2	
Ar \rightarrow Ar ⁺		4.50	215	0.049	2	
Kr \rightarrow Kr ⁺		7.51	360	0.037	2	
Xe \rightarrow Xe ⁺		11.75	561	0.035	2	
H ₂ \rightarrow H ₂ ⁺		0.721	34.5	0.281	2	

TABLE II

Electron energy (eV)	Cross sections Q in units 10^{-16}cm^2					
	Ionization of H			Excitation (2p) of H		
	Born appr.	Experi- ment ⁷⁾⁸⁾⁹⁾	Present (eq. 10)	Born appr.	Experi- ment ⁷⁾	Present (eq. 10)
20	0.54	0.30	0.30	1.15	0.59	0.46
30	0.92	0.48	0.47	1.13	0.658	0.65
40	1.05	0.61	0.61	1.04	0.675	0.69
45	—	0.64	0.65	—	0.675	0.70
50	1.01	0.66	0.68	0.95	0.675	0.69
75	—	0.66	0.68	0.78	—	0.62
100	0.74	0.60	0.62	0.64	0.53	0.55
150	0.56	0.49	0.50	0.51	0.45	0.45
200	0.445	0.41	0.41	0.41	0.395	0.38
300	0.325	0.31	0.31	0.30	—	0.29
400	—	—	0.24	0.25	—	0.24
700	—	—	0.15	0.176	—	0.16
1000	0.117	—	0.117	0.123	—	0.123

eq. (10), for ionization of Kr and Xe. This table also contains experimental cross sections measured by Asundi and Kurepa¹³⁾.

We remark that the values of Q_i given by Asundi and Kurepa are probably somewhat too high for $E_1 > 60 \text{eV}$. Relative measurements of Fox¹⁴⁾ indicate that the cross sections are lower for $E_1 > 60 \text{eV}$ (in these measurements, the ionization-functions reach a maximum at much lower electron energies: 45 eV for Kr and 35 eV for Xe), which is in agreement with the high energy measurements of Schram *e.a.*²⁾. For the same reason we prefer, for Ne and Ar, the measurements of Bleakney¹²⁾ above the measurements of Smith¹¹⁾ and Asundi *e.a.*¹³⁾. It is possible that the cross sections for ionization of H₂, measured by Tate and Smith¹⁰⁾ are somewhat too high for $E_1 > 60 \text{eV}$.

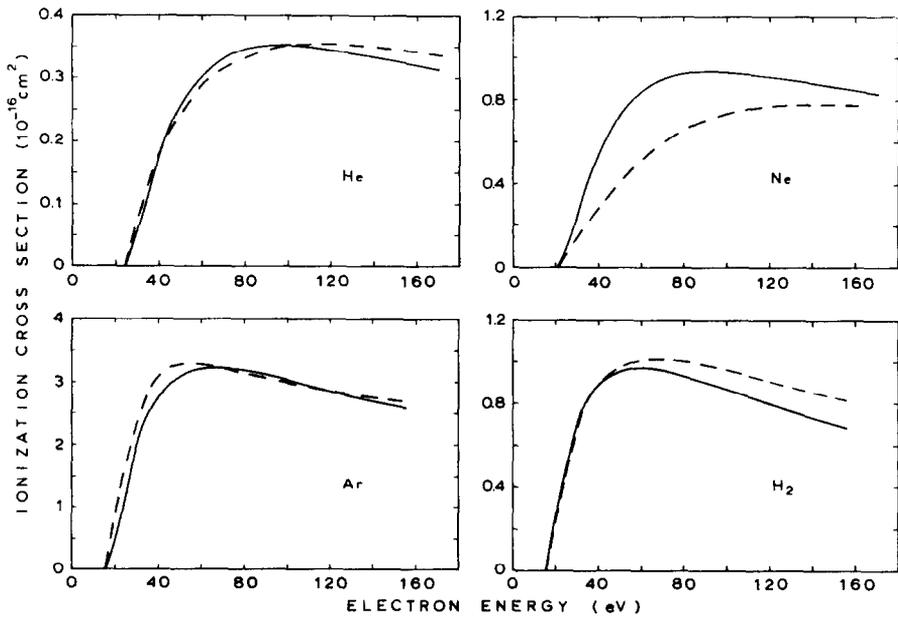


Fig. 2. Ionization cross sections of He, Ne, Ar and H_2 . Solid curves: present calculations; dashed curves: experiment¹¹⁾¹²⁾¹⁰⁾. The agreement between theory and experiment for H_2 is better than may be concluded from the figure (see text). The Ne curves show a discrepancy which is a basic one as is discussed at the end of this paper. Numerical values of cross sections for H, Kr and Xe are given in tables II and III.

TABLE III

Electron energy (eV)	Cross sections Q_t in units 10^{-16} cm^2			
	Kr		Xe	
	Experiment ¹³⁾	Present (eq. 10)	Experiment ¹³⁾	Present (eq. 10)
20	1.5	1.4	3.4	3.8
30	3.6	3.8	5.6	7.2
40	4.6	4.7	6.7	8.4
50	5.0	5.1	7.2	8.6
60	5.3	5.2	7.5	8.6
70	5.5	5.1	7.6	8.3
80	5.4	5.0	7.6	8.0
90	5.4	4.9	7.7	7.8
100	5.3	4.7	7.8	7.5
150	—	4.1	—	6.3
200	—	3.5	—	5.5
400	—	2.4	—	3.7
1000	—	1.3	—	2.0

For ionization of N_2 and O_2 , Schram *e.a.*²⁾ give values of M_i^2 equal to 3.85 and 4.75 and values of c (in eV^{-1}) equal to 0.070 and 0.053 respectively. From the values of M_i^2 it follows that A is 184 for N_2 and 228 for O_2 (in units $10^{-16} cm^2 eV$). With these values and eq. (10), the calculated cross sections Q_i , for $E_1 = 100 eV$, are 2.98 and $3.57 \times 10^{-16} cm^2$, while the measurements of Tate and Smith¹⁰⁾ give $Q_i = 2.91$ and $2.92 \times 10^{-16} cm^2$ respectively.

Discussion. In this paper we presented a new semi-empirical formula to describe ionization and excitation of atoms. In previous investigations, several empirical formulae, to calculate ionization cross sections of atoms, have been proposed, for example by: de la Ripelle, by Elwert and by Drawin¹⁵⁾. Similar formulae are obtained from the "classical" two particle collision theories as developed by Thomson¹⁶⁾ and Gryzinski^{17) 18)}. Thomson's formula is not empirical and does not give a good agreement with experiment. Gryzinski's formula as given in ref. 17 is semi-empirical because he made use of an (unnecessary¹⁹⁾) approximation resulting in a better agreement with experiment. Gryzinski's formula as given in ref. 18 is semi-empirical because of a similar unnecessary approximation and because of the logarithmic factor in his formula. This factor is introduced in his theory by means of an incorrect¹⁹⁾ velocity distribution of the atomic electrons. To the arguments given in ref. 19 (to prove the incorrectness of this velocity-distribution)¹⁹⁾ we can add the following. Gryzinski assumes the velocity distribution to be proportional to $1/v^3$ for large v , v being the velocity of the atomic electron(s). In reality, this velocity distribution falls off much faster. For atomic hydrogen (for which atom the exact atomic wave functions are known) this velocity distribution is proportional to $1/v^6$ for the electron in the first Bohr orbit.

The "classical" two particle collision theories predict the ionization cross sections Q_i to be proportional to N/U_i^2 , where N is the number of electrons in the shell corresponding with the ionization, and U_i is the ionization energy. The previous empirical formulae are also based on the same idea, though Drawin introduced two free parameters. These empirical and semi-empirical formulae give reasonably good agreement with experiment^{15) 17) 18) 20)}. This is due to the fact that, if $Q_i U_i^2 / N$, where Q_i are the experimental cross sections, is plotted against E_1 / U_i for several atoms, most graphs are very much alike. Here we restrict ourselves to those atoms where mainly one shell of the atom is responsible for the ionization. But in some cases, for example for Ne, the agreement between such a universal (mean) curve and experiment (Ne) is very bad²⁰⁾. As pointed out by Seaton²¹⁾, this can be understood by considering the ionization as an induced radiative process.

The present eq. (10) is essentially different from previous formulae,

because we based our formula on the optical properties of an atom (optical oscillator strength) and on the shape of the differential cross section (generalized oscillator strength) as a function of K . For large E_1 , our formula gives the best agreement with experiment, because eq. (10) is exact for these E_1 . For small E_1 , the agreement between our formula and experiment is very good. The previous empirical formulae do not give a better agreement for small E_1 , though in some of these formulae free parameters are used (Drawin¹⁵).

Now we write eq. (10) as follows:

$$\frac{QU^2}{N} = \frac{AUF}{N} \frac{(x-1)}{x^2} \ln [1 + cU(x-1)] \quad (11)$$

where $x = E_1/U$. If a universal curve gives a correct description of the ionization process, then the values of $AU/N (= 4\pi a_0^2 R U_i M_i^2/N)$ and of cU will be nearly the same for different atoms. In table IV a number of values of AU/N and cU are listed. From table IV it follows that the values of

TABLE IV

Values of AU/N and cU		
Process	$10^{16} AU/N$ ($\text{cm}^2 \text{eV}^2$)	cU
H \rightarrow H ⁺	185	76.3
He \rightarrow He ⁺	289	2.65
Ne \rightarrow Ne ⁺	322	0.690
Ar \rightarrow Ar ⁺	563	0.770
Kr \rightarrow Kr ⁺	844	0.520
Xe \rightarrow Xe ⁺	1133	0.425
H ₂ \rightarrow H ₂ ⁺	267	4.33
H \rightarrow H (2p)	271	1.04
He \rightarrow He 2 ¹ P	91	

AU/N and cU , and thus the differential cross sections (for large E_1 and probably also for small E_1), are very different for several atoms. But for most atoms, the major part of these differences is compensated in the total cross sections, because if AU/N is large then cU is small. This is illustrated by the following example, where we denote QU^2/N by z . Then is:

$$z(\text{He} \rightarrow \text{He}^+, x = 4)/z(\text{H} \rightarrow \text{H}^+, x = 4) = 0.84$$

while the values of AU/N differ by a factor 1.56 and the values of cU by factor 29.

From table IV it also follows that the value of cU for ionization of atomic hydrogen is much greater than cU for excitation of atomic hydrogen to the 2p-level. This means that "large" momentum transfers are more probable for ionization than for excitation. This agrees with the fact that for excitation only one energy transfer U_n occurs while for ionization also energy transfers greater than U_i are possible.

We remark that the use of differential cross sections is very important, both for a better understanding of the ionization and excitation process and to test the reliability of a theory. For example: to understand why Gryzinski's theory gives erroneous results¹⁹⁾ for large E_1 , it is necessary to look at the differential cross sections²²⁾.

For excitation of He, no accurate values of c are known. The absolute cross sections for excitation of He to the 3^1P and 4^1P levels are^{23) 24)} about $3 \times 10^{-18} \text{ cm}^2$ and $1.5 \times 10^{-18} \text{ cm}^2$ respectively, for $E_1 = 100 \text{ eV}$. With eq. (10) and with the values of A given in table I, we derived the corresponding values of c . These values (in units eV^{-1}) are about 0.08 (3^1P) and 0.18 (4^1P). The first value is quite reasonable because the value of c for ionization of He is 0.108. The second value is rather large, but this can be due to the fact that small errors in A and Q introduce large errors in c .

By eq. (9), we introduced a simple energy dependence of $A(E_1)$. We also tried another energy dependence given by:

$$A(E_1) = A_1 F \frac{(E_1 - U_{i,n})}{E_1} + A_2 F \quad (12)$$

where $A_1 + A_2 = A$. This gives a somewhat better threshold behaviour for small values of A_2 ($A_1 > 40 A_2$). We remark that eq. (10) does not give a correct (linear) threshold behaviour.

To calculate ionization cross sections, our semi-empirical formula can be applied for all atoms (molecules) in which mainly one shell is responsible for the ionization (e.g. for H, He and H_2). For Ne, Ar, Kr and Xe also autoionization (for instance after excitation of an innershell electron) and innershell ionization is possible. For some atoms (e.g. for Hg) such innershell ionization will be important, as is indicated by our calculations given in ref. 20. In these calculations, the possibility of autoionization is not included, though this may also be important for Hg. Thus for Hg and for Ne, Ar, Kr and Xe, a better description of the ionization process will perhaps be given (for moderate E_1 , e.g. $E_1 < 10U_i$) by:

$$Q_i = \sum_k A_k F_k \frac{(E_1 - U_k)}{E_1^2} \ln[1 + c_k(E_1 - U_k)] \quad (13)$$

where the index k corresponds with the different processes resulting in ionization of the atom. But the values of A_k and c_k are unknown, for which reason application of eq. (13) is much more difficult than application of eq. (10). Another and probably more reliable possibility to take account of autoionization and innershell ionization, is given in the final part of the discussion.

Many values of $A(M_i^2)$ and c used in this paper are given by Schram *e.a.*²⁾. They measured the gross ionization cross sections; i.e. also the contributions of multiply charged ions are included in their measurements. This means

that the values of M_i^2 given by them (and used by us) correspond with the gross ionization. For atoms such as Ne, Ar, Kr and Xe a correction of M_i^2 and A should be necessary and according to eq. (10) this will result in somewhat lower cross sections for single ionization; especially because in their measurements the double charged ions are counted twice. We note that in the experimental cross sections of Tate, Smith, Asundi and Kurepa^{10) 11) 13)}, the multiple ionizations are also included.

We remark that our method is based on the accurate knowledge of the values of $A(f_n$ or $M_i^2)$ and c (generalized oscillator strength). We mention the possibility that other (more complicated) trial functions as given in eqs. (8), (9) and (12) will give a somewhat better agreement with experiment. More refinements in the presented theory are not impossible. For instance: for ionization of atoms we assumed the cross sections Q_i to be proportional to M_i^2 , where M_i^2 is given by eq. (6). Physically it is more meaningful to take these ionization cross sections proportional to χ where:

$$\chi = \int_{U_i}^{E_1} \frac{R}{E} \frac{df}{dE} dE \quad (14)$$

For $E_1 \gg U_i$, χ is equal to M_i^2 , and for a value of E_1 just above U_i , χ is proportional to R/U_i times the differential oscillator strength for energy transfer U_i . The method of Seaton²¹⁾, which gives threshold ionization cross sections, is based on the same idea. Of course, such a refinement as discussed here will result in a complication of the calculations. From the photoionization cross sections of He, Ne and Ar²⁵⁾, it follows that χ/M_i^2 for Ne decreases much faster for small E_1 than χ/M_i^2 for He and Ar. Therefore the refinement "to take Q_i proportional to χ " will result in a much better agreement between the theoretical and experimental cross sections for Ne.

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