

HIGH FREQUENCY LIMITS OF THE TWO-ELECTRON PHOTOIONIZATION CROSS SECTIONS

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Several cross sections of two-electron processes at high but nonrelativistic photon energies ω are considered, which are expressed solely via the initial state wave function of the ionized two-electron object. The new high precision and locally correct nonvariational wave functions describing the ground and several lowest excited states of H^- , He and helium-like ions are used in calculations of different cross sections in the pure dipole approximation and with account of first order corrections in ω/c^2 , and a number of the cross sections' ratios. The dependencies of all these quantities on the nuclear charge Z and the principal quantum number n (for $1 < n < 5$) of the initial state excitation are studied.

1. Introduction

The cross sections of two-electron photoionization and ionization with excitation of two-electron atoms at high photon energies ω can be expressed via the initial state wave functions. The simplest objects for studying these processes are He, helium-like ions and H^- . For these, the corresponding cross sections are expressed via the Coulomb three-body wave function, namely that of two electrons in the field of a nucleus. Usually the variational wave functions are considered most accurate. They are determined from the requirement to reproduce accurately the energy of the considered state. Unfortunately, a wave function that is adequate for reproducing the energy could be not good enough for other characteristics, such as the cross sections. The latter are determined by essentially different space regions in the wave function than the energy. This is why we decided to perform calculations of the high energy limits of the two-electron ionization and ionization with excitation cross sections using recently obtained wave functions¹ that are locally correct, having the best precision at any given point of space.

This paper presents the results of new calculations of the high- ω photoionization cross sections with elimination of one or both electrons from the following two-electron atomic systems: He atom, helium-like two-electron ions and the H^- ion. For the He and helium-like ions we also study a number of excited states that can be considered roughly as monopole, i.e. s -excitations of one of the electrons. In the one-electron approach these states can be described as $1s$, ns configurations. We concentrate on high photon frequencies ω , $\omega \gg I^{++}$, where I^{++} is the two-electron ionization potential. The aim of this paper is to study the dependence of different cross sections and their ratios on the nuclear charge Z and the principal quantum number n of the target excitation.

The processes of two-electron ionization and ionization with excitation are of interest because their theoretical and experimental investigation yields information on the corresponding initial state wave function of the atomic system. The system of two electrons in the field of a nucleus seems to be quite simple. However, despite the progress in computational power it is still an unsolved problem

to find accurate enough continuous spectrum wave functions for this system: such functions can be obtained only for its ground and low excited states.

For two-electron systems in the high but non-relativistic frequency^a limit, the cross section of the two-electron ionization $\sigma^{++}(\omega)$, the ionization with excitation $\sigma^{+*}(\omega)$, and the one-electron ionization $\sigma^+(\omega)$ are entirely determined by the initial state wave function (see e.g. Ref. 2) that can be calculated accurately enough. This is why we concentrate here on the high frequency limits. It is essential that the corrections to these cross sections that are first order in ω/c^2 , where c is the speed of light, are also expressed via the initial state wave function³ of the considered two-electron object.

When using very accurate and locally correct wave function,¹ one can expect to obtain more accurate values for these cross sections.

The first publications on two-electron ionization appeared at the end of the sixties.⁴⁻⁶ Afterwards, there was an almost 20-year-long intermission in studies of this subject. Recently, however, there has been a burst of activity in this field (see e.g. Ref. 7). It was stimulated by experimental developments for measuring $\sigma^{++}(\omega)$, in particular at high energy. Having in mind that several sources of high intensity and high frequency continuous spectrum electromagnetic radiation have recently been or are being constructed, a growing interest in more detailed studies of two-electron processes can be anticipated. Not only the ground but also excited states of two-electron systems, and not only neutral systems but also positive and negative ions, will attract the attention of researchers in this domain of atomic physics. This is why we concentrate on H^- , the He atom and the helium-like ions in their ground and excited states.

Until recently most of the publications dealing with two-electron photoionization considered this process in the dipole approximation. Thus they are neglecting completely the incoming photon momentum and therefore neglecting the effects of retardation of electromagnetic field. In the frame of the dipole approximation it was demonstrated that the main contribution to the two-electron photoionization cross section $\sigma^{++}(\omega)$ at high ω , $I^{++} \ll \omega \ll c^2$,

comes from the so-called shakeoff mechanism.⁴⁻⁶ According to this mechanism one of the electrons, namely that absorbing the incoming photon, leaves the ionized atom very fast. This instantly changes the field that acts upon the second electron and causes its elimination from the atom. The fast electron carries away almost all photon energy $\epsilon_1 \approx \omega \gg I^{++}$, while the energy of the second electron, ϵ_2 , is of the order of I^{++} , $\epsilon_2 \approx I^{++}$. The shakeoff mechanism predicted that the ω -dependencies of $\sigma^{++}(\omega)$ and the one-electron photoionization cross section $\sigma^+(\omega)$ are the same, namely $\sigma^+(\omega) \sim \sigma^{++}(\omega) \sim \omega^{-7/2}$ at high ω , $\omega \gg I^{++}$. The same is also the ω -dependence of the cross section of ionization with excitation, $\sigma^{+*}(\omega)$. This is why this ω region the ratios of the cross sections are ω -independent.

For He, the ω dependence of the cross section $\sigma^{++}(\omega)$ and its value at $\omega \gg I^{++}$, in fact starting from 1 keV, which follows from the shakeoff approach, seems to be in good agreement with the experimental data.⁷ In the shakeoff framework the cross section $\sigma^{++}(\omega)$, $\sigma^{+*}(\omega)$ and $\sigma^+(\omega)$ are expressed via the integrals over initial state wave function $\Psi(\mathbf{r}_1, \mathbf{0})$, where one electron is at the nucleus, which corresponds to high photoelectron linear momentum, while the other is at the position \mathbf{r}_1 , somewhere inside the atom.

The shakeoff approach seems to be so well established that other alternative mechanisms are usually not discussed at all.⁸ However, it was demonstrated already in 1975 that there exists the so-called quasifree (QF) mechanism,^{2,9} which is becoming more and more important with ω growth. The main idea that forms the foundation of this mechanism is the following: two free electrons can absorb a single photon, in contrast to one electron. As a result of photon absorption the electrons acquire almost equal energies, $\epsilon_1 \approx \epsilon_2 \approx \omega/2$, and move in approximately opposite directions.

It was demonstrated long ago^{2,9} that the QF mechanism leads to corrections of the order of $\omega/c^2 \ll 1$ at high but nonrelativistic photon energies.

In this ω region, if ω/c^2 corrections are neglected, the ω dependencies of $\sigma^{++}(\omega)$, $\sigma^{+*}(\omega)$ and $\sigma^+(\omega)$ are the same, and it is convenient to study the

^aThe atomic system of the units is used in this paper: $m_e = e = \hbar = 1$, where m_e is the electron mass, e its charge and \hbar the Planck constant.

ω -independent ratios

$$R_d = \frac{\sigma^{++}(\omega)}{\sigma^+(\omega) + \sigma^{+*}(\omega)} \Big|_{\omega \rightarrow \infty}, \quad (1)$$

$$R^* = \frac{\sigma^{+*}(\omega)}{\sigma^+(\omega)} \Big|_{\omega \rightarrow \infty}, \quad (2)$$

$$R_0 = \frac{\sigma^{++}(\omega)}{\sigma^+(\omega)} \Big|_{\omega \rightarrow \infty} = R_d(1 - R^*), \quad (3)$$

$$\begin{aligned} R_1 &= \frac{\sigma^+(\omega) + \sigma^{+*}(\omega) + \sigma^{++}(\omega)}{\sigma^+(\omega)} \Big|_{\omega \rightarrow \infty} \\ &= (1 + R_d)(1 + R^*). \end{aligned} \quad (4)$$

Qualitatively, one can expect that the ratios become smaller and smaller with increase of Z , since the mean interelectron radius measured in units of the $1s$ electron orbit increases, so that relative role of interelectron interaction that leads to double ionization decreases when Z grows. On pure qualitative grounds one can expect also that R^* and R_0 will increase while n is growing, since the main mechanism leading to ionization with excitation is shakeup.²

We will calculate here also the corrections to R_d of the order of ω/c^2 . The formulas for this contribution expressing them via $\Psi(\mathbf{r}_1, \mathbf{r}_1)$ were derived recently.^{10,11} The investigation of the QF mechanism contribution is of interest and timely since now high intensity photon beams with $\omega \approx 100$ keV ($\omega/c^2 \approx 1/5$) are available.

The dependence of R_d and R^* on n was studied recently.¹² In our paper we will calculate R_d and R^* using the recently obtained very accurate initial state wave function,¹ calculated by the correlation function hyperspherical harmonic method (CFHHM). In this method, the wave function Ψ is decomposed as

$$\Psi = e^f \phi, \quad (5)$$

where f is the correlation function describing the singularities of Ψ , and ϕ is a smooth remainder which can be expanded in a fast converging hyperspherical harmonic expansion. The function f depends on interparticle distances, which is necessary and sufficient to take into account analytically the two- and three-body Coulomb singularities (cusps) in the wave function, i.e. it satisfies the Kato cusp conditions¹³ exactly. The function Ψ is obtained by a direct solution of the three-body Schrödinger equation, which guarantees local correctness of Ψ because

the convergence of Ψ across the configurations space is uniform.

2. Details of Calculations

To obtain the expressions for the ratios R_d and R^* , the rather general approach² was used. Keeping in mind that for high but nonrelativistic ω the main contribution to $\sigma^{++}(\omega)$ comes from the strongly asymmetric energy sharing between the outgoing electrons of which one is fast while the other is slow, the expression for $\sigma^{++}(\omega)$ is simplified considerably, and is given by the formula

$$\sigma^{++}(\omega) = \frac{32\sqrt{2}Z^2\pi^2}{3c\omega^{7/2}} \left(\langle \delta(\mathbf{r}) \rangle - \sum_{\nu} I_{\nu 0} \right), \quad (6)$$

where $\langle \delta(\mathbf{r}) \rangle$ is the expectation value of $\delta(\mathbf{r})$ in the initial state and

$$I_{\nu 0} = 4\pi \left| \int_0^{\infty} \Psi(0, s) R_{\nu 0}(qs) s^2 ds \right|^2. \quad (7)$$

Here $q = 2Zm_{\alpha}/(1 + m_{\alpha})$, Z is the charge of the nucleus, $R_{\nu l}$ are the two-particle bound state Coulomb radial function, ν is their single-particle principal quantum number and c is the speed of light. In (7) Ψ depends on Jacobi coordinates \mathbf{r} and \mathbf{s} , where \mathbf{r} connects the nucleus and one electron, and \mathbf{s} connects the center of mass of these two particles with the other electron. Thus $\Psi(0, s)$ represents the three-body wave function at the coalescence, or cusp, region, in which CFHHM is *in principle* precise.

The quantity measured in experiments is²

$$\sigma^+(\omega) + \sigma^{+*}(\omega) = \frac{32\sqrt{2}Z^2\pi^2}{3c\omega^{7/2}} \sum_{\nu} I_{\nu 0}, \quad (8)$$

so that

$$R_d = \frac{\langle \delta(\mathbf{r}) \rangle - \sum_{\nu} I_{\nu 0}}{\sum_{\nu} I_{\nu 0}} \quad (9)$$

and

$$R^* = \frac{\sum_{\nu} I_{\nu 0} - I_{10}}{I_{10}} = \frac{\sum_{\nu \geq 2} I_{\nu 0}}{I_{10}}. \quad (10)$$

The ratio $R(\omega)$ of the double-to-single electron photoionization cross section for small values of ω/c^2 can be presented in the form^{9,11}

$$R(\omega) \approx R_d + \frac{8\sqrt{2}}{5Z^2} \mathcal{I} \frac{\omega}{c^2}, \quad (11)$$

where

$$\mathcal{I} = \frac{1}{N} \int d\mathbf{r}_1 |\Psi(\mathbf{r}_1, \mathbf{r}_1)|^2 = \frac{1}{N} \langle \delta(\mathbf{r}_{12}) \rangle \quad (12)$$

and

$$N = \sum_{\nu=1}^{\infty} 4\pi \left| \int_0^{\infty} \Psi(0, s) R_{\nu 0}(qs) s^2 ds \right|^2. \quad (13)$$

It is seen that R_d , R^* , R_0 and R_1 are determined by $\Psi(0, \mathbf{r})$, i.e. by the wave function with one electron at nucleus. \mathcal{I} is determined according to Eqs. (11) and (12) by both $\Psi(0, \mathbf{r})$ and $\Psi(\mathbf{r}, \mathbf{r})$, the latter defining the probability of finding two electrons at the same point anywhere in the atom.

3. Result of Calculations

The values of the ratio R_d for the ground and excited states n^1S , $n = 1, \dots, 5$, of the helium isoelectron sequence from $Z = 1, \dots, 10$ are presented in Table 1. The ratios R_d for $n = 1, 2$ calculated in Ref. 12 agree with ours except at $Z = 3$ and $Z = 6$ for $n = 1$ and at $Z = 4$ and $Z = 8$ for $n = 2$, where the rounded values differ by 1×10^{-5} . This indicates that careful assessment of the quality of the three-body wave function input is needed.

We have used 200 single-particle states to calculate the cross section ratios, so that the error of summation over states is negligible and we do not have to resort to estimating the remainder as in Ref. 12. The carefully checked values maintain small differences from Ref. 12, giving the value $R = 0.01644$, which agrees with Ref. 12. The values of the ratios R^* and R_0 for n^1S , $n = 1, \dots, 5$, and $Z = 1, \dots, 10$ are presented in Tables 2 and 3, respectively. Particularly interesting is the behavior of R^* . While at $n = 1$ R^* decreases for $Z > 4$ slightly faster than $\sim Z^{-2}$, the situation is completely different for $n \geq 2$: there R^* decreases very slow with increasing Z . As expected, R^* increases with n . However, the rapidity of this increase when going from $n = 1$ to $n = 2$ is really impressive and unexpected. Indeed, the ratio $\eta = R_{(n+1)}^*/R_{(n)}^*$ really jumps for $n = 1$, from 327 for $Z = 2$ to 6440 for $Z = 10$! Subsequently, already for $n = 2$, this factor becomes almost Z -independent and slowly decreases with increasing n — from ≈ 3.5 for $n = 2$ to ≈ 2 for $n = 4$. From the behavior of R_0 it is seen that the excitation of one of the He or helium-like ions' electrons increases

the relative probability of double ionization, but by far not so strongly as that of ionization with excitation. Note that for $n \geq 3$, both R^* and R_0 depend on Z and n weakly.

In Refs. 9 and 12 the leading term of the Z -dependence of R_d was estimated and shown to be $1/Z^2$. We use here a three-term fit in inverse powers of Z . Our results are presented in Table 4. They are good from $Z = 2-3$ for $n = 1, 2$ and from $Z = 4-5$ for $n = 3-5$.

It is essential to note that the Z -dependence of R_d changes considerably with n growth: already for $n = 2$ it decreases much slower than $\sim Z^{-2}$.

We have calculated here also the parameter \mathcal{I} using Eq. (12) for H^- , He and helium-like ions. For all but H^- objects, \mathcal{I} for the four lowest 1S excited states was also calculated. The results are presented

Table 1. R_d values for states n^1S .

n	1	2	3	4	5
1	0.01602				
2	0.01644	0.00903	0.00369	0.00169	0.00088
3	0.00855	0.01204	0.00830	0.00546	0.00360
4	0.00508	0.00994	0.00849	0.00677	0.00530
5	0.00334	0.00768	0.00728	0.00643	0.00553
6	0.00235	0.00595	0.00599	0.00561	0.00512
7	0.00175	0.00469	0.00491	0.00479	0.00453
8	0.00135	0.00377	0.00406	0.00406	0.00395
9	0.00107	0.00309	0.00339	0.00346	0.00344
10	0.00087	0.00258	0.00287	0.00297	0.00299

Table 2. R^* values for states n^1S .

n	1	2	3	4	5
1	0.66554				
2	0.05838	19.105	72.411	179.97	361.09
3	0.02053	12.752	46.274	112.86	223.85
4	0.01037	10.951	39.015	94.475	186.71
5	0.00623	10.128	35.710	86.084	169.70
6	0.00415	9.6613	33.825	81.327	160.05
7	0.00297	9.3621	32.629	78.271	153.84
8	0.00222	9.1543	31.788	76.140	149.53
9	0.00173	9.0011	31.167	74.568	146.34
10	0.00138	8.8842	30.690	73.360	143.90

Table 3. R_0 values for states n^1S .

Z	n	1	2	3	4	5
1		0.02669				
2		0.01740	0.18157	0.2708	0.3057	0.3182
3		0.00873	0.16560	0.3924	0.6217	0.8105
4		0.00513	0.11883	0.3396	0.6465	0.9947
5		0.00336	0.08541	0.2671	0.5596	0.9445
6		0.00236	0.06339	0.2086	0.4623	0.8249
7		0.00175	0.04860	0.1651	0.3794	0.7022
8		0.00135	0.03833	0.1331	0.3135	0.5951
9		0.00107	0.03094	0.1091	0.2618	0.5062
10		0.00087	0.02548	0.0909	0.2211	0.4336

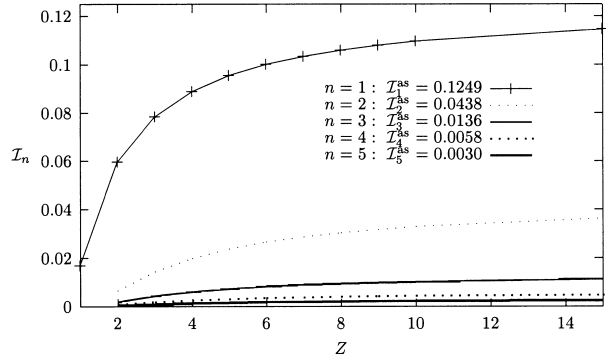
 Table 4. Coefficients of the fit $AZ^{-2} + BZ^{-3} + CZ^{-4}$ for states n^1S , corresponding to the last four weights at large Z set equal to 1 and the other weights set to the small value of 0.01, stressing the behavior at large Z .

n	A	B	C
1	0.090	-0.021	-0.053
2	0.327	-0.709	0.227
3	0.410	-1.330	1.034
4	0.473	-1.960	2.026
5	0.522	-2.524	2.995

in Fig. 1. The curves permit an extrapolation to asymptotically high values of Z : $Z \rightarrow \infty$. In this limit the pure hydrogenic model must be valid, and the interelectron interaction can be treated in the first order. For the ground state of the target ion in the frame of this model, one obtains $\mathcal{I} = 1/8$. This value coincides excellently with the result of our extrapolation of the calculated curve to $Z \rightarrow \infty$. We performed the extrapolation in the following way. Because of the smooth dependence of the parameter \mathcal{I} upon Z for different n , the values \mathcal{I}_n were approximated by the formula

$$\mathcal{I}_n = a_n + \frac{b_n}{Z} + \frac{c_n}{Z^2}. \quad (14)$$

The asymptotic values $\mathcal{I}_n^{\text{as}} = a_n$ are also given in Fig. 1. Thus, using our numerical results we managed to get values that may be derived purely analytically. It is of interest to note that the QF


 Fig. 1. The ratio \mathcal{I}_n for helium isoelectronic sequence. $\mathcal{I}_n^{\text{as}}$ are the extrapolated values for $Z \rightarrow \infty$.

contribution to the R values is almost the same for He and H^- : indeed, the value of \mathcal{I}/Z^2 is 0.01149 in He and 0.0169 for H^- .

Using our numerical values for He, the following expression for $R(\omega)$ can be given:

$$R(\omega) \approx R_d \left(1 + 0.58 \frac{\omega}{c^2} \right). \quad (15)$$

It is seen that at $\omega = 100$ keV the QF correction is about 10%. This is the required accuracy of the experiment in order to observe the QF contribution.

4. Discussion and Conclusion

Summing up, in this work the cross section ratios R_d , R^* and R_0 have been calculated for the n^1S states, $n = 1, 2, \dots, 5$, of the helium isoelectronic sequence for $Z = 1, \dots, 10$ using very accurate and locally correct nonvariational wave functions. They incorporate all three cusp conditions.

The leading order of the Z -dependence of R_d for $Z \gg 1$ is very simple: $R \approx 0.094Z^{-2}$.⁹ This result was obtained in the first order in the interelectron interaction. However, an attempt to use this formula for the He atom, substituting Z by the Slater effective nuclear charge $Z_{\text{eff}}^{\text{He}} = 27/16 \approx 1.69$, fails. Indeed, instead of the very accurate value $R_d \approx 0.01644$ (see Ref. 14 and references therein), one obtains the value $R_d = 0.033$, which is by a factor of 2 too large. This discrepancy can be explained by the fact that the effective charges corresponding to double ionization and the ionization with excitation processes are different. One is the screening or Slater effective charge $Z_{\text{eff}}^{\text{SI}} = Z - 5/16$, and the other

is the interelectron interaction $V_{12} = 1/|\mathbf{r}_1 - \mathbf{r}_2|$ effective charge $z_{\text{eff}}^{\text{in}}$. This charge transforms $1/|\mathbf{r}_1 - \mathbf{r}_2|$ into $z_{\text{eff}}^{\text{in}}/|\mathbf{r}_1 - \mathbf{r}_2|$, thus permitting one to take into account the higher order corrections in V_{12} . With $z_{\text{eff}}^{\text{in}}$, the Z -dependence of R_d can be parametrized as $R \approx 0.094(z_{\text{eff}}^{\text{in}})^2/(Z_{\text{eff}}^{\text{Sl}})^2$. For the He atom, using the Slater charge $Z_{\text{eff}}^{\text{He}} = 27/16$ and the most reliable value $R \approx 0.01644$, one obtains the value $z_{\text{eff}}^{\text{in, He}} \approx 0.705$.

To illustrate the sensitivity of \mathcal{I} values to the quality of the initial state wave function, let us discuss the results of \mathcal{I} that were calculated in Ref. 10 using different wave functions. Using the Hartree–Fock wave function for He the result $\mathcal{I}_{\text{He}} = 0.11$ was obtained. With the Hylleraas six-parameter wave function the \mathcal{I}_{He} value 0.068 is obtained. Thus, \mathcal{I} is quite sensitive to the choice of the wave function. To obtain \mathcal{I} the interelectron interaction must be taken into account nonperturbatively. This is demonstrated by the large difference in \mathcal{I} values for finite Z and $Z = \infty$. The difference between \mathcal{I}_{He} and \mathcal{I}_{∞} is rapidly increasing the growth of n .

It is of interest to learn whether our results can be reproduced within the framework of the lowest order hydrogenic approximation,⁹ but with effective charges. As was demonstrated above, the ratio R_d has to include at least two effective charges, namely the screening or the Slater one, $Z_{\text{eff}}^{\text{He}} = Z - 5/16$, and the interelectron interaction one, $z_{\text{eff}}^{\text{in}}$. Having in mind that $\mathcal{I} \sim (z_{\text{eff}}^{\text{in}})^2$ and substituting Z in (6) by $Z_{\text{eff}}^{\text{Sl}} = Z - 5/16$, an effective value $\mathcal{I}_{\text{eff}} = (z_{\text{eff}}^{\text{in}} Z / Z_{\text{eff}}^{\text{He}})^2 / 8$ instead of $\mathcal{I} = 1/8 = 0.125$ is obtained.

Using simple expressions for the effective charges, the interelectron one, $z_{\text{eff}}^{\text{in}} = 1 - A/Z$, and the nuclear or Slater one, $Z_{\text{eff}}^{\text{He}} = Z(1 - 5/16Z)$, one can fit the \mathcal{I} data reasonably well. Of course, the value of A depends upon n . For He, in order to reproduce the $\mathcal{I}_{\text{He}} = 0.0597$ value precisely one has $A = 0.832$ for $n = 1$, which means that $z_{\text{eff}}^{\text{in}} = 0.584$. This differs considerably from the above-mentioned value $z_{\text{eff}}^{\text{in}} = 0.705$, which corresponds to $A = 0.59$.

It is seen that the results of calculations of the parameter $R(\omega)$ are very sensitive to the accuracy of the initial state wave function. The experimental observation of the corrections to $R(\omega)$ due to the QF mechanism would contribute considerably to deepening our understanding of the two-electron photoionization process.

For R_n with $n > 1$ the Z^{-2} dependence appears for higher Z only: the bigger the n , the higher the Z value starting from which the Z^{-2} dependence is in effect.

The approach presented in this paper can be applied also to triplet excited initial states of the considered two-electron objects. The three-particle wave functions employed in this paper can be used in calculations of high accuracy cross section for Compton and fast incoming particles two-electron ionization with excitation of the same objects. We are planning to concentrate on these problems in our future publications.

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