

Nuclear charge dependence of the two-electron high-frequency photoionization cross section

R. Krivec,¹ M. Ya. Amusia,^{2,3} and V. B. Mandelzweig²

¹*Department of Theoretical Physics, J. Stefan Institute, P.O. Box 3000, 1001 Ljubljana, Slovenia*

²*Racah Institute of Physics, The Hebrew University, Jerusalem 91904, Israel*

³*A. F. Ioffe Physical-Technical Institute, St. Petersburg, 194021, Russia*

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Using high-precision wave functions describing the ground and excited states of the H^- ion, He atom, and heliumlike ions, the cross sections of single- and double-electron photoionization are calculated. The dependence of the ratio R of the double- and single-ionization cross sections on the nuclear charge Z and the principal quantum number of excitation n is studied and compared with the predictions of the high- Z expansion and the high- n shake-off mechanisms.

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I. INTRODUCTION

This paper presents the results of calculations of the photoionization cross sections with elimination of one or both electrons from the following two-electron atomic systems: the H^- ion, the He atom, and the heliumlike two-electron ions. For the He atom and the heliumlike 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 the ratios of different photoionization cross sections on the nuclear charge Z and the principal quantum number n of the target excitation.

For two-electron systems in the high but nonrelativistic frequency¹ 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., [1]). When using very accurate and locally correct [2–5] initial-state wave functions one can expect to obtain precise values for these cross sections. By studying the cross-section dependence on Z and n one can see how the variation of the mean-interelectron distance (measured in units of the $1s$ orbit radius) affects the probability of the two-electron ionization or ionization with excitation processes.

The 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. Accurate wave functions can be obtained only for its ground and low-

excited states. Therefore it is of particular interest to study the high-frequency cross sections, which are expressible solely via bound-state wave functions that are reliably established. It is also essential to note [1] that the ω dependence of the cross sections at high ω is known and depends not on the initial-state wave function but on the angular momentum of the eliminated electrons. Therefore a given initial-state wave function is able to describe the single and double ionization and ionization with excitation cross-sections in a broad photon frequency region $\omega \gg I^{++}$.

The first publications on two-electron ionization appeared at the end of the 1960s [6–8]. Afterwards, there was an almost 20-year-long intermission in studies of this subject. A kind of an exception was the publication of the paper [9], where the Z dependence of the ratio of double-to-single photoionization cross section was obtained. Then, in the nineties, there happened a burst of activity in this domain (see [10–14], and references therein). The intense activity in this field is summarized and a number of new results are presented in a recent book [15]. This development was, and is still, stimulated by the progress in experimental possibilities of measuring the double-charged ions yield and $\sigma^{++}(\omega)$, in particular at high energies. The ratio of the yields of He^{2+} and He^+ was measured in a broad photon frequency region, up to 20 keV [16].

It appeared that it is possible purely experimentally to separate the contributions to two-electron ionization and ionization with excitation caused by either photon absorption or by scattering (Compton effect) [17]. This is of great importance for investigations at high ω since the cross sections of the latter processes, $\sigma_C^{++}(\omega)$, $\sigma_C^{+*}(\omega)$, and $\sigma_C^+(\omega)$, are almost independent of ω while the cross sections of photoionization are rapidly decreasing (as $\omega^{-7/2}$) with ω . For the He atom these cross sections become equal at approximately 6 keV [18]. Having in mind that several sources of high-intensity and high-frequency continuous spectrum electromagnetic radiation have been, or are being constructed recently, a growing interest for 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 attention of researchers in this domain of atomic physics. This is why we concentrate on H^- , He atom, and heli-

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

TABLE I. Values of the ratio $I_{\nu 0}/\langle\delta(\mathbf{r})\rangle$ values for small ν for the n^1S states of the helium atom.

ν	n				
	1	2	3	4	5
1	0.9296	0.0493	0.0136	0.0055	0.0028
2	0.0446	0.5346	0.0702	0.0231	0.0106
3	0.0055	0.3992	0.1669	0.0533	0.0236
4	0.0018	0.0035	0.7319	0.0019	0.0062
5	0.0008	0.0017	0.0131	0.7762	0.0593
6	0.0005	0.0009	0.0001	0.1379	0.4991
7	0.0003	0.0005	0.0001	0.0003	0.3868
8	0.0002	0.0003	0.0001	0.0000	0.0106

unlike ions in their ground and excited states. Because the ω dependencies of $\sigma^{++}(\omega)$, $\sigma^{+*}(\omega)$, and $\sigma^+(\omega)$ at high ω are the same it is convenient to study ω -independent ratios

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

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

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

and

$$R_1 = \frac{\sigma^+(\omega) + \sigma^{++}(\omega) + \sigma^{+*}(\omega)}{\sigma^+(\omega)} \Bigg|_{\omega \rightarrow \infty} = (1 + R)(1 + R^*). \quad (4)$$

Qualitatively, one can expect that the ratio R becomes smaller and smaller with the increase of Z , since the mean inter-electron radius measured in units of the $1s$ electron orbit increases, causing the relative role of the inter-electron

TABLE II. R values for the n^1S states in the helium isoelectronic sequence.

Z	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 III. As in Table II, but R^* .

Z	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	33.835	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

interaction that leads to double ionization to decrease when Z grows. On pure qualitative ground one can expect also that R^* will increase when n is growing, since the main mechanism leading to ionization with excitation is shake-off [1].

The leading order of the Z dependence of R 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 $R \approx 0.01644$ (see [19], and references therein) one obtains the value $R = 0.033$ that 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 $Z_{\text{eff}}^{\text{Sl}} = Z - 5/16$, 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 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 $z_{\text{eff}}^{\text{in,He}} \approx 0.705$.

The dependence of R on Z and n was studied recently [20]. In this paper we will calculate R and R^* using the

TABLE IV. As in Table II, but R_0 .

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 V. Coefficients of the fit $AZ^{-2} + BZ^{-3}$ for states n^1S . The first line for each n , 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, stresses the behavior at large Z ; the second line corresponds to all weights equal to 1.

n or Ref.	A	B
1	0.091	-0.037
	0.098	-0.078
[20]	0.09	-0.03
2	0.323	-0.652
	0.309	-0.568
[20]	0.32	-0.66
3	0.394	-1.072
	0.339	-0.708
4	0.442	-1.454
	0.340	-0.757
5	0.476	-1.776
	0.330	-0.765

recently obtained very accurate and locally correct [2–5] initial-state wave function, calculated by the correlation function hyperspherical harmonic method. 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 that can be expanded in a fast converging hyperspherical harmonic (HH) 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 [21] conditions exactly. Furthermore, Ψ is obtained by a direct solution of the three-body Schrödinger equation, which guarantees local

TABLE VI. As in Table V, but for the fit $AZ^{-2} + BZ^{-3} + CZ^{-4}$.

n	A	B	C
1	0.090	-0.021	-0.053
	0.090	-0.022	-0.052
2	0.327	-0.709	0.227
	0.338	-0.830	0.448
3	0.410	-1.330	1.034
	0.424	-1.483	1.325
4	0.473	-1.960	2.026
	0.475	-1.982	2.095
5	0.522	-2.524	2.995
	0.500	-2.315	2.651

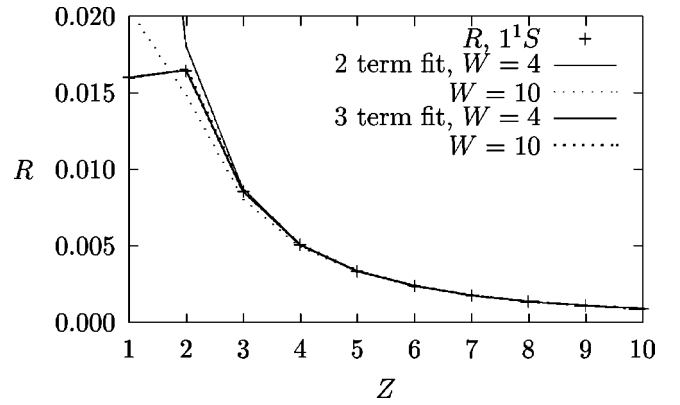


FIG. 1. Calculated R values (dimensionless) of the 1^1S state and their least squares fits of the forms $AZ^{-2} + BZ^{-3}$ (two-term fit) and $AZ^{-2} + BZ^{-3} + CZ^{-4}$ (three-term fit). W denotes the number of weights at large Z equal to 1; other weights are set to 0.01.

correctness [2–5] of Ψ because the convergence of Ψ across the configurations space is uniform,

II. DETAILS OF CALCULATIONS

To obtain the expressions for the ratios R and R^* , we use the rather general approach presented in Ref. [1]. 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,

$$\sigma^{++}(\omega) \approx \frac{32\sqrt{2}Z^2\pi^2}{3c\omega^{7/2}} \left\{ \int |\Psi(\mathbf{0}, \mathbf{s})|^2 ds - \sum_{\nu lm} \left| \int \Psi(\mathbf{0}, \mathbf{s}) \psi_{\nu lm}(s) ds \right|^2 \right\}, \quad (6)$$

where Ψ is the initial state three-body wave function and $\psi_{\nu lm}(s)$ is the unperturbed single-particle wave function of the second electron in the field of the nucleus, after the first electron has left the atom. Here 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

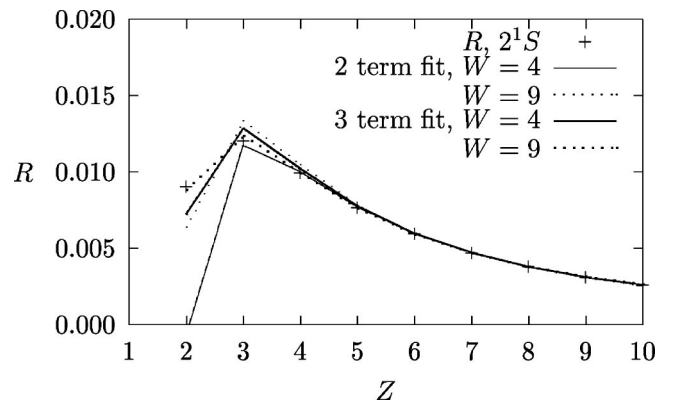
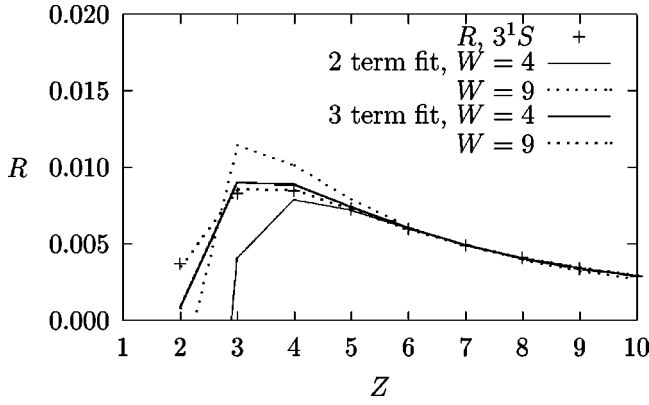


FIG. 2. As in Fig. 1, but for the 2^1S state.

FIG. 3. As in Fig. 1, but for the 3^1S state.

with the other electron. In the framework of the present approximation, we set $\mathbf{r}=\mathbf{0}$; then \mathbf{s} represents the distance of the second electron from the nucleus. Thus $\Psi(\mathbf{0},\mathbf{s})$ represents the three-body wave function at the coalescence, or cusp, region, in which, as one can see from the discussion in the Introduction and from references therein, local high accuracy of the wave function is especially difficult to obtain. Finally, ν is the single-particle principal quantum number, l and m are the angular momentum quantum numbers, and c is the speed of light.

As shown in [1] we have the following expressions for the total angular momentum $L=0$ and $\omega \rightarrow \infty$:

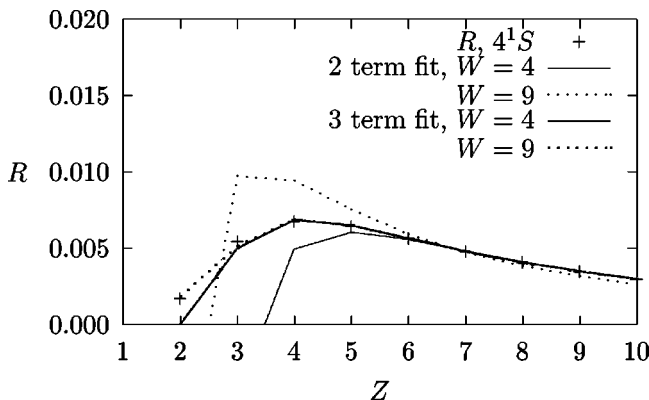
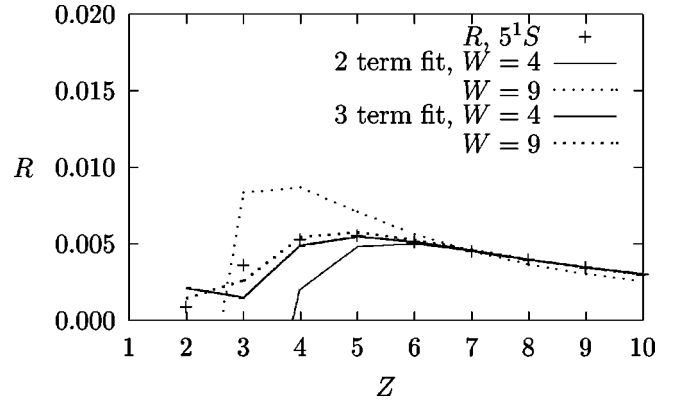
$$\sigma^{++}(\omega) = \frac{32\sqrt{2}Z^2\pi^2}{3c\omega^{7/2}} \left(\langle \delta(\mathbf{r}) \rangle - \sum_{\nu} I_{\nu 0} \right), \quad (7)$$

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

$$I_{\nu 0} = 4\pi \left| \int_0^{\infty} \Psi(0,s) R_{\nu 0}(qs) s^2 ds \right|^2, \quad (8)$$

where $q = 2Zm_{\alpha}/(1+m_{\alpha})$ and $R_{\nu l}$ are the two-particle bound state Coulomb radial functions (see Table I). The quantity measured in experiments [1] is

$$\sigma^+(\omega) + \sigma^{++}(\omega) = \frac{32\sqrt{2}Z^2\pi^2}{3c\omega^{7/2}} \sum_{\nu} I_{\nu 0} \quad (9)$$

FIG. 4. As in Fig. 1, but for the 4^1S state.FIG. 5. As in Fig. 1, but for the 5^1S state.

so that

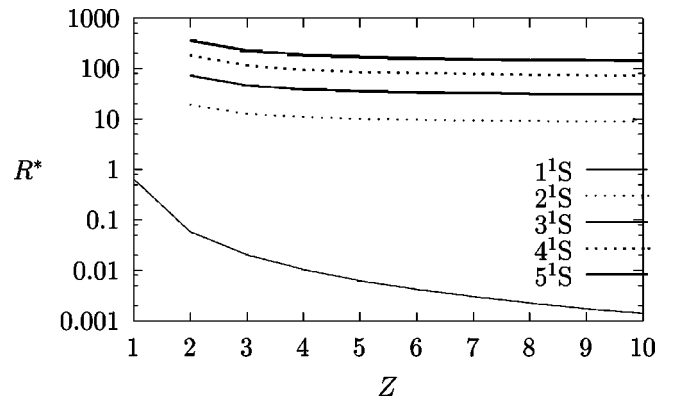
$$R = \frac{\langle \delta(\mathbf{r}) \rangle - \sum_{\nu} I_{\nu 0}}{\sum_{\nu} I_{\nu 0}} \quad (10)$$

and

$$R^* = \frac{\sigma^{+*}(\omega)}{\sigma^+(\omega)} = \frac{\sum_{\nu} I_{\nu 0} - I_{10}}{I_{10}} = \frac{\sum_{\nu \geq 2} I_{\nu 0}}{I_{10}}. \quad (11)$$

III. RESULTS OF CALCULATIONS

The values of ratio R for the ground and excited states n^1S , $n=1, \dots, 5$ of the helium isoelectron sequence from $Z=1$ to $Z=10$ are presented in Table II. Most results can be obtained with a small hyperspherical harmonic basis with $K_m=40$, for lower states even with $K_m=32$. Here K_m is the maximum global angular momentum used in the HH expansion. For H^- , we also calculated with K_m up to 64 and different parametrizations of the correlation function f in order to make sure that R is correct to the presented precision. The exception is He ($Z=2$) where the excited states, if Z is decreased from larger values, assume a true three-body char-

FIG. 6. Dependence of R^* (dimensionless) on Z for the n^1S states.

acter, become densely spaced and therefore not very differently extended in space, and can no longer be deduced by scaling in Z from large Z ; while $K_m=40$ is very good for the ground state, $K_m=100$ is necessary for the 2^1S-5^1S states. These results were taken from the recent work [5].

The ratios R for $n=1,2$ calculated in Ref. [20] 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. [19]. While the carefully checked values maintain small differences from Ref. [20], the recalculation of R for the ground state of He now gives the value $R=0.01644$ that agrees with Ref. [20], where the remainder is apparently estimated for $n>8$. Adding our own estimated remainder [19] would also make the values agree.

The values of the ratios R^* and R_0 for the ground and excited states n^1S , $n=1, \dots, 5$ of the helium isoelectronic sequence from $Z=1$ to $Z=10$ are presented in Tables III and IV, respectively. Particularly interesting is the behavior of R^* . While at $n=1$ the ratio R^* decreases for $Z>4$ slightly faster than Z^{-2} , the situation is completely different for $n \geq 2$: there the R^* values decrease very slowly with Z . As it was 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 $\eta_n=R_{n+1}^*/R_n^*$, where the index n denotes the state, really jumps for $n=1$, from 327 for $Z=2$ to 6440 for $Z=10$. With subsequent growth of n , already for $n=2$, η_n becomes almost Z independent and slowly decreasing with n —from $\eta_n \approx 3.5$ for $n=2$ to $\eta_n \approx 2$ for $n=4$. From the behavior of R_0 it is seen that the excitation of one of the electrons in helium or in heliumlike ions increases the relative probability of double ionization, but by far not so strongly as that of the ionization with excitation. Note that for $n \geq 3$ both R^* and R_0 depend weakly on Z and n .

In Refs. [9,20] the leading term of the Z dependence of R was estimated and shown to be $1/Z^2$. Our calculations show that for R with $n>1$, R_n , the Z^{-2} dependence appears for higher Z only: the bigger the n , the higher the Z value at which the Z^{-2} dependence starts.

We use here two- and three-term fits in inverse powers of Z . In addition in each case we use two different least square fits, one with all weights equal to one (which favors equally small and large Z), and another with a very small weight

(0.01) for $Z \leq 6$ and the weights equal to one for $Z \geq 7$ (that favors strongly a better description at large Z). Our results are displayed in Tables V and VI and in Figs. 1–5, where W denotes the number of weights at large Z that are equal to 1. One can see that the two-term fits are reasonable starting from $Z=3-4$ for all weights for $n=1,2$ and from $Z=6-7$ for $n=3-5$. The three-term fits are generally better than the two-term fits that, of course, is what one expects since three-parameter curves are always easier to fit to data than two-parameter curves. They are good even from $Z=2-3$ for $n=1,2$ and from $Z=4-5$ for $n=3-5$. The coefficients of inverse powers of Z in two- and three-term fits, however, are somewhat different, so the question whether the coefficient of the $1/Z^4$ term theoretically equals zero has a direct consequence for the value of the coefficients of the first and the second terms. Figure 6 gives the dependence of R^* on n and Z in logarithmic scale.

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

IV. CONCLUSION

The cross section ratios R , R^* , and R_0 have been calculated for the n^1S states, $n=1,2, \dots, 5$, of the helium isoelectronic sequence for $Z=1$ to $Z=10$. According to qualitative estimates, R for the ground state decreases very fast with Z . But already starting from R_2 , i.e., R at $n=2$, this ratio at first increases with Z and only after reaching a maximum starts to decrease. As to R^* , it dramatically increases with n for any Z . The Z dependence of R^* for $n=1$ and $n \geq 2$ proved to be qualitatively different: extremely rapid decrease for $n=1$ contrasts with very slow decrease at $n \geq 2$.

To calculate the cross sections and the corresponding ratios, in the H^- case it was necessary to employ a nonlinear correlation function f in Eq. (5), but for $Z \geq 2$ a simple linear f and a rather small value of $K_m=32$ (81 HH states) was sufficient for most states, except for the higher excited states where it was prudent to use $K_m=40$ (121 HH states) although some value $32 < K_m < 40$ could be sufficient, and for the higher excited states of He where K_m up to 100 was needed because of their close spacing and consequently different structure than for $Z>2$. In all cases the three cusp conditions were fulfilled exactly.

To achieve stability of the coefficient B of the term BZ^{-3} in the fit of the Z dependence one has to either (i) discard R values for small Z , or (ii) add the term CZ^{-4} to the fitting function, which may indicate an open theoretical problem.

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