

Calculation of the contribution of the quasifree mechanism to the two-electron 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, Hebrew University, Jerusalem 91904, Israel*

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

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A retardation correction to the high-energy limit of the dipole photoionization cross section and to the ratio of the double-to-single electron photoionization cross sections is calculated. This correction comes from the *quasifree* mechanism of the two-electron photoionization and is determined by the probability to find both ionizing electrons at the same point in the initial state. The calculations are performed for H^- , the helium atom and the heliumlike ions in the ground and some excited states using high-precision wave functions. The nuclear charge dependence of this correction for heliumlike ions in the ground and four lowest 1S excited states is calculated. The possibility to detect this correction experimentally is discussed.

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I. INTRODUCTION

The aim of this paper is to present results of the calculations of the lowest-order retardation corrections to the two-electron photoionization cross section of a number of two-electron objects, namely, H^- , the helium atom and the heliumlike ions at high but nonrelativistic photon energies ω . Both ground and excited states of the latter objects will be studied.

Until recently most of the publications dealing with two-electron photoionization consider this process in the dipole approximation. Thus they are neglecting completely the incoming photon momentum and therefore neglecting the effects of retardation of the 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 comes from the so-called shake-off mechanism [1–3]. According to this mechanism one of the electrons, namely, the one 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 $\epsilon_1 \approx \omega \gg I^{++}$, where I^{++} is the two-electron ionization potential, while the energy of the second electron ϵ_2 , is of the order of I^{++} , $\epsilon_2 \approx I^{++}$. The shake-off 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 in this ω region the ratio $R(\omega) = \sigma^{++}(\omega) / [\sigma^+(\omega) + \sigma^{+*}(\omega)]$ is ω independent. For He the corresponding value is $R_d \approx 0.0165$ [3], where the index d emphasizes that this value of R is calculated in the dipole approximation. The most recent value is $R_d \approx 0.01645$ (see Ref. [4] and discussion therein). The ω dependence of the cross section $\sigma^{++}(\omega)$ and the value of R at $\omega \gg I^{++}$, in fact starting from 1 keV for He, that follows from the shake-off approach, seems to be in good agreement with the experimental data

[5]. It is essential to have in mind that while $\sigma^{++}(\omega)$ rapidly decreases with ω growth, the cross section of ionization of two electrons in Compton scattering $\sigma_C^{++}(\omega)$ is almost ω independent. For He, $\sigma^{++}(\omega)$ and $\sigma_C^{++}(\omega)$ become equal already at $\omega = 6$ keV. However, the contribution of $\sigma^{++}(\omega)$ and $\sigma_C^{++}(\omega)$ can be separated even purely experimentally (see Ref. [5] and references therein).

In the framework of the shake-off mechanism the cross sections $\sigma^{++}(\omega)$ and $\sigma^+(\omega)$ are expressed via the integrals over the initial-state wave function $\bar{\Psi}_i(\mathbf{r}_1, \mathbf{0})$ where one electron is at the nucleus (at $|\mathbf{r}_2| \approx 0$, which corresponds to high photoelectron linear momentum) while the other is at the position $|\mathbf{r}_1|$ somewhere inside the atom. We shall use the notation $\bar{\Psi}(\mathbf{r}_1, \mathbf{r}_2) = \Psi(\mathbf{r}_{12}, \mathbf{s}_{12})$, where \mathbf{r}_1 , \mathbf{r}_2 denote the electron coordinates relative to the nucleus, and \mathbf{r}_{12} and \mathbf{s}_{12} denote the Jacobi vectors, \mathbf{r}_{12} connecting the electrons, and \mathbf{s}_{12} connecting the center-of-mass of the electrons and the nucleus.

The shake-off approach seems to be so well established that other alternative mechanisms are usually not discussed at all [6]. However it was demonstrated already in 1975 that there exists the so-called quasifree (QF) mechanism [7,8] that 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 [7,8] that the QF mechanism leads to corrections of the order of $\omega/c^2 \ll 1$ at nonrelativistic photon energies. It was shown [9] that at $\omega/c^2 \geq 1$ the QF mechanism becomes absolutely dominant. In this frequency domain the ratio $R(\omega)$ is again ω independent,

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. The values of \mathcal{I} for H^- , the helium atom, and the helium isoelectronic sequence ground and excited n^1S states. The last line gives extrapolated values given by the values of the parameter a_n in the fits of the Z dependence of the form $a_n + b_n/Z + c_n/Z^2$, $n=1, \dots, 5$. The numbers in brackets are the errors in the last digits, obtained by setting different numbers of small- Z weights in the fits to a small value, and keeping the remaining weights equal to 1.

Z	n	1	2	3	4	5
1		0.0169				
2		0.0597	0.0067	0.0019	0.0008	0.0004
3		0.0786	0.0144	0.0043	0.0018	0.0009
4		0.0890	0.0199	0.0061	0.0026	0.0013
5		0.0956	0.0238	0.0073	0.0031	0.0016
6		0.1002	0.0266	0.0082	0.0035	0.0018
7		0.1035	0.0287	0.0089	0.0038	0.0019
8		0.1061	0.0304	0.0094	0.0040	0.0021
9		0.1081	0.0317	0.0099	0.0042	0.0021
10		0.1097	0.0328	0.0102	0.0043	0.0022
15		0.1147	0.0363	0.0113	0.0048	0.0025
$\mathcal{I}_n^{\text{as}}$		0.1249(1)	0.0438(1)	0.01363(2)	0.00580(1)	0.00297(1)

$R(\omega) \rightarrow R_{\text{rel}}$, but considerably larger than the value $R_d \approx 0.0165$ obtained in the dipole approximation.

The results in Refs. [7–9] were calculated in the first order of the interelectron interaction. It is clear, however, that numerically such an approach is not accurate enough. Indeed, in the first order the value $R_d^{(1)}$ is 0.0235, which is considerably larger than the nonperturbative value. Qualitatively, it is clear that the contribution of the QF mechanism depends upon the initial-state wave function with two ionizing electrons at small interelectron distances $r_{12} \approx 1/p \rightarrow 0$, $\Psi(\mathbf{r}_1, \mathbf{r}_1) = \Psi(\mathbf{0}, \mathbf{s}_{12})$, with p being the photoelectron linear momentum that increases unlimitedly with ω growth.

In this paper we will concentrate on calculating the first order in the ω/c^2 correction to the ratio R_d using the best available initial-state two-electron wave functions [10–13]. The formulas for this contribution expressing them via $\Psi(\mathbf{r}_1, \mathbf{r}_1)$ were derived recently [14,15]. Numerical results were also obtained in those works, and we shall compare our results with previous ones in order to study how sensitive is

the ω/c^2 correction to the details of $\Psi(\mathbf{r}_1, \mathbf{r}_1)$. Recently the derivations of the corrections of the order ω/c^2 to R_d given in Refs. [7,14] and Ref. [15] were repeated in Ref. [16] confirming the previous results. The investigation of the QF mechanism contribution is of interest and timely since now high intensity photon beams with $\omega \geq 100$ keV ($\omega/c^2 \approx 1/5$) are available.

II. DETAILS OF CALCULATIONS

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 following form [7,15,17]:

$$R(\omega) \approx R_d + \frac{8\sqrt{2}}{5Z^2} \frac{\omega}{c^2}, \quad (1)$$

where

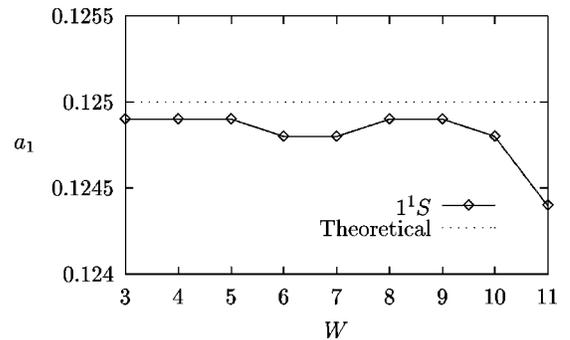


FIG. 2. Dependence of the asymptotic value of \mathcal{I} , i.e., the coefficient a_1 of the fits of Fig. 1, on W .

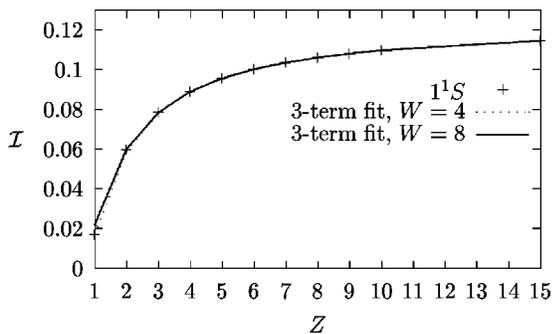
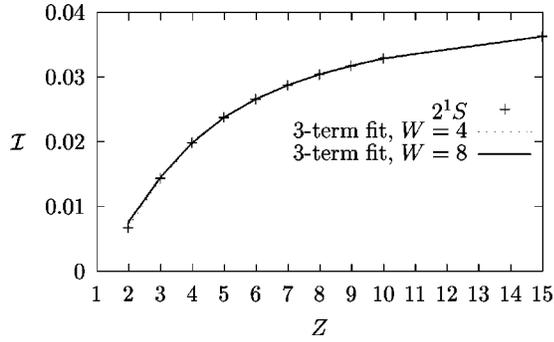


FIG. 1. Dependence of \mathcal{I} (dimensionless) and two of its fits of the form $a_n + b_n/Z + c_n/Z^2$, $n=1$, on Z for the 1^1S state of the helium isoelectronic sequence. W is the number of large- Z weights set to 1, while the other weights are small.


 FIG. 3. As in Fig. 1, but for the first excited state ($n=2$).

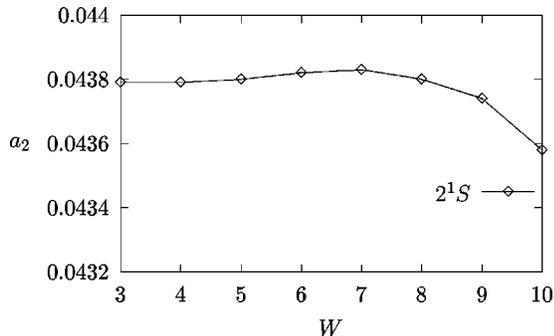
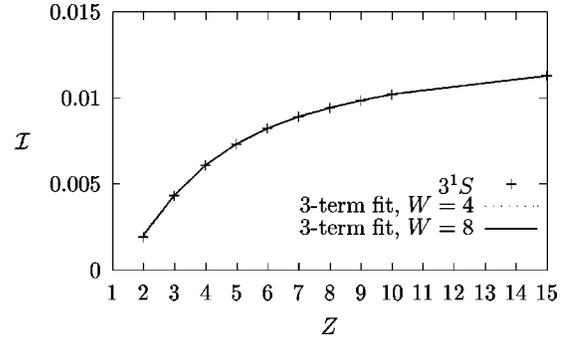
$$\begin{aligned} \mathcal{I} &= \frac{1}{N} \int d\mathbf{r}_1 |\bar{\Psi}_i(\mathbf{r}_1, \mathbf{r}_1)|^2 = \frac{1}{N} \int ds_{12} |\Psi_i(\mathbf{0}, \mathbf{s}_{12})|^2 \\ &= \frac{1}{N} \langle \delta(\mathbf{r}_{12}) \rangle \end{aligned} \quad (2)$$

and

$$\begin{aligned} N &= \sum_{\nu=1}^{\infty} \left| \int d\mathbf{r}_1 \bar{\Psi}_i(\mathbf{r}_1, \mathbf{0}) \psi_{\nu 00}(\mathbf{r}_1) \right|^2 \\ &= \sum_{\nu=1}^{\infty} \left| \int ds_{23} \Psi_i(\mathbf{0}, \mathbf{s}_{23}) \psi_{\nu 00}(\mathbf{s}_{23}) \right|^2 \\ &= \sum_{\nu=1}^{\infty} 4\pi \left| \int_0^{\infty} \Psi(0, s_{23}) R_{\nu 0}(q s_{23}) s_{23}^2 ds_{23} \right|^2 \end{aligned} \quad (3)$$

Here $q = 2Zm_{\alpha}/(1+m_{\alpha})$, m_{α} and Z are the mass and the charge of the nucleus, $\psi_{\nu lm}(\mathbf{r})$ are the one electron wave functions in the field of the nucleus, and $R_{\nu l}$ are the corresponding radial functions; only $l=m=0$ is used. In this paper $\bar{\Psi}_i(\mathbf{r}_1, \mathbf{0}) = \Psi_i(\mathbf{0}, \mathbf{s}_{23})$ is the H^- , He, or the heliumlike ion's three-body wave function in the ground or excited (except H^-) 1^1S states, and \mathbf{s}_{23} is the Jacobi vector connecting the center-of-mass of the nucleus (index 3) and one electron (index 2) with the other electron (index 1).

It is seen from Eq. (1) that \mathcal{I} is determined by the initial state two-electron wave function $\bar{\Psi}_i(\mathbf{r}_1, \mathbf{r}_2)$ in two different space coordinate regions: at $r_k=0$, $k=1,2$, and at $|\mathbf{r}_1 - \mathbf{r}_2|=0$. As to R_d , it is determined only by the initial-state two-

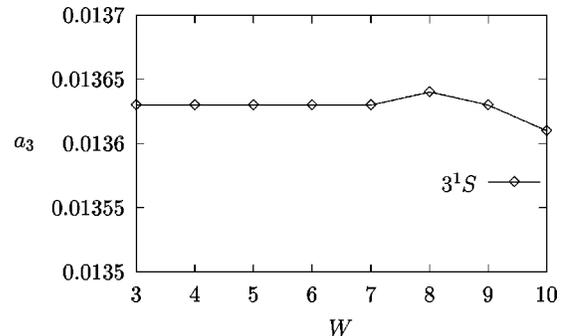

 FIG. 4. As in Fig. 2, but for the first excited state ($n=2$).

 FIG. 5. As in Fig. 1, but for the second excited state ($n=3$).

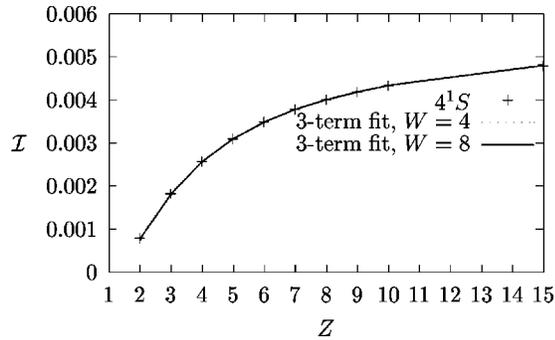
electron wave function with one coordinate being equal to zero, $\bar{\Psi}_i(\mathbf{r}_1, \mathbf{0})$. As a consequence of the nonanalyticity of the nuclear Coulomb potential $-Z/r$ under the change of sign $r \rightarrow -r$, the wave function $\bar{\Psi}_i(\mathbf{r}_1, \mathbf{r}_2)$ has singularities at $r_k \rightarrow 0$, $k=1,2$, called the Kato cusps [18]. A similar singularity exists in $\bar{\Psi}_i(\mathbf{r}_1, \mathbf{r}_2)$ when $|\mathbf{r}_1 - \mathbf{r}_2| \rightarrow 0$, which is a consequence of the nonanalyticity of the interelectron interaction $1/r_{12}$ under the change of sign $r_{12} \rightarrow -r_{12}$. The peculiarities that characterize the wave function $\bar{\Psi}_i(\mathbf{r}_1, \mathbf{r}_2)$ at $r_k=0$, $k=1,2$ and at $|\mathbf{r}_1 - \mathbf{r}_2|=0$, must be reproduced well by the wave function that we use in our calculations in order to give reliable values for R_d and \mathcal{I} .

In this paper, the parameter \mathcal{I} is calculated numerically using the recently computed very accurate and locally correct initial-state wave function described in Refs. [4,10–13] calculated by the correlation function hyperspherical harmonic method. In this method, the wave function Ψ is decomposed as

$$\Psi = e^f \phi, \quad (4)$$

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 conditions exactly. Furthermore, Ψ is obtained by a direct solution of the three-


 FIG. 6. As in Fig. 2, but for the second excited state ($n=3$).

FIG. 7. As in Fig. 1, but for the third excited state ($n=4$).

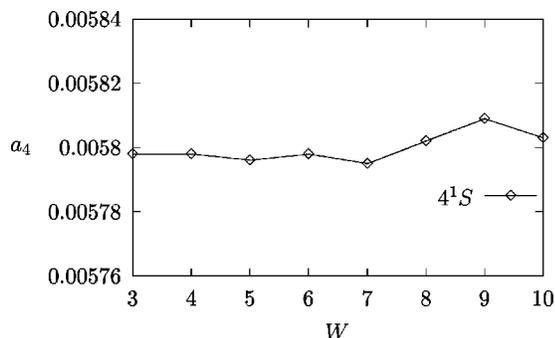
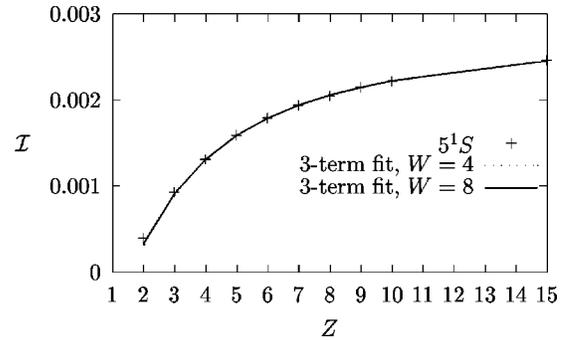
body Schrödinger equation, which guarantees local correctness of Ψ because the convergence of Ψ across the configuration space is uniform.

III. RESULTS OF CALCULATIONS

We have calculated the parameter \mathcal{I} using Eq. (2) for H^- , He, and heliumlike ions. For all systems but H^- , \mathcal{I} for the four lowest $1S$ excited states was also calculated. The results are presented in Table I and in Figs. 1–10. The calculated values are correct to at least one more digit than quoted in the table. The curves in Figs. 1, 3, 5, 7, and 9 permit an extrapolation to asymptotically high values of Z , $Z \rightarrow \infty$. In this limit the pure hydrogenic model must be valid. In its frame the interelectron interaction is treated in the first order, which is correct for $Z \rightarrow \infty$. For the ground state of the target ion in the frame of this model, one obtains $\mathcal{I}=1/8$.² 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

$$\mathcal{I}_n = a_n + \frac{b_n}{Z} + \frac{c_n}{Z^2}. \quad (5)$$

²In a recent paper [19] a value $\mathcal{I}=1/16$, which is smaller by a factor of 2, is calculated in the same approximation as Ref. [14]. We checked the results of Ref. [14] and confirm them here.

FIG. 8. As in Fig. 2, but for the third excited state ($n=4$).FIG. 9. As in Fig. 1, but for the fourth excited state ($n=5$).

Several fits were performed for each n , setting the weights at a number of small- Z points to a small value, and setting the remaining W weights to 1. As shown in Figs. 2, 4, 6, 8, and 10, the fits were remarkably stable against the variation of W from $W=3$ to $W=9$ or 10.

The asymptotic values $\mathcal{I}_n^{\text{as}} = a_n$ are also given in Table I. 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 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 in H^- .

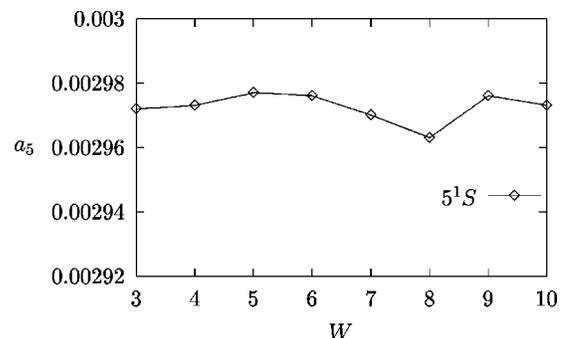
Using our numerical values for He, the following expression for $R(\omega)$ can be given:

$$R(\omega) \approx R_d \left(1 + 0.58 \frac{\omega}{c^2} \right). \quad (6)$$

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

IV. DISCUSSION AND CONCLUSIONS

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. [14] using different wave functions. Using the Hartree-Fock wave function for He, the results $\mathcal{I}_{\text{He}}=0.11$ was obtained. Using the Hylleraas three- and six-parameter wave functions, the \mathcal{I}_{He} values are 0.07 and 0.068, respectively. For the Kinoshita wave function the

FIG. 10. As in Fig. 2, but for the fourth excited state ($n=5$).

value is 0.055. The details about these wave functions can be found in Table 6 of Ref. [20]. It is seen that \mathcal{I} is quite sensitive to the choice of the wave function. One should have in mind that the interelectron interaction must be taken into account nonperturbatively. This is demonstrated by the large difference in \mathcal{I} values for physical Z and for $Z=\infty$. This difference between \mathcal{I}_{He} and \mathcal{I}_{∞} is rapidly increasing with the growth of n .

It is of interest to learn whether the accurate results computed in this paper can be reproduced within the framework of the lowest-order hydrogenic approximation [7], but using effective charges. It is clear that the ratio R 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 $z_{\text{eff}}^{\text{in}}$, which takes into account the higher-order corrections in this interaction, $z_{\text{eff}}^{\text{in}}\approx 0.705$. This value of $z_{\text{eff}}^{\text{in}}$ had to be introduced into the hydrogenic approximation [7] in order to fit the correct value of $R_d=0.01645$. Having in mind that $\mathcal{I}\approx(z_{\text{eff}}^{\text{in}})^2$ and substituting Z in Eq. (1) 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 introduced. For He one has $\mathcal{I}_{\text{eff}}=0.0873$ that is considerably smaller than 0.125 and prominently bigger than the accurate value 0.0597. It means that the effective charge $z_{\text{eff}}^{\text{in}}$, which serves to reproduce the correct value of R_d , is not good enough to describe \mathcal{I} . This is quite understandable, since the interelectron distances essential in R_d and \mathcal{I} , as it

was noted above while discussing Eqs. (1) and (2), are quite different.

Note also that 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 data in Table I 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 to use $A=0.832$ for $n=1$, which means that $z_{\text{eff}}^{\text{in}}=0.584$. This differs considerably from the mentioned above 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 the deepening of our understanding of the two-electron photoionization process.

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- [1] T.A. Carlson, Phys. Rev. **156**, 142 (1967).
 [2] F.W. Byron and C.J. Joachain, Phys. Rev. **164**, 1 (1967).
 [3] T. Åberg, Phys. Rev. A **2**, 1726 (1970).
 [4] R. Krivec, M.Ya. Amusia, and V.B. Mandelzweig, Phys. Rev. A **62**, 064701 (2000).
 [5] J.H. McGuire *et al.*, J. Phys. B **28**, 913 (1995).
 [6] A. Dalgarno and H.R. Sadeghpour, Comments At. Mol. Phys. **30**, 143 (1994).
 [7] M.Ya. Amusia, E.G. Drukarev, V.G. Gorshkov, and M.P. Kazachkov, J. Phys. B **8**, 1248 (1975).
 [8] M. Ya. Amusia, *Atomic Photoeffect* (Plenum, New York, 1990).
 [9] E.G. Drukarev and F. Kapershin, J. Phys. B **9**, 913 (1976).
 [10] M. Haftel, R. Krivec, and V.B. Mandelzweig, J. Comp. Physiol. **123**, 149 (1996).
 [11] M. Haftel and V.B. Mandelzweig, Phys. Rev. A **42**, 6342 (1990).
 [12] R. Krivec, V.B. Mandelzweig, and K. Varga, Phys. Rev. A **61**, 062503 (2000).
 [13] R. Krivec, M. Ya. Amusia, and V. B. Mandelzweig, Phys. Rev. A (to be published).
 [14] E.G. Drukarev, Phys. Rev. A **51**, R2684 (1995).
 [15] E.G. Drukarev, Phys. Rev. A **52**, 3910 (1995).
 [16] M.A. Kornberg and J.E. Miraglia, Phys. Rev. A **60**, R1743 (1999).
 [17] R. H. Pratt and T. Surić (unpublished).
 [18] T. Kato, Commun. Pure Appl. Math. **10**, 151 (1957).
 [19] A.I. Mikhailov and I.A. Mikhailov, Zh. Èksp. Teor. Fiz. **114**, 1537 (1998) [JETP **87**, 833 (1998)].
 [20] H. A. Bethe and E. E. Salpeter, *Quantum Mechanics of One- and Two-electron Atoms* (Plenum, New York, 1977).