Effect of the ground-state correlations on the helium double photoionization and ionization with excitation

Anatoli S. Kheifets[1,*] and Igor Bray[2,†]
[1]*Institute of Advanced Studies, Research School of Physical Sciences, The Australian National University, Canberra, Australian Capital Territory 0200, Australia*
[2]*Electronic Structure of Materials Centre, The Flinders University of South Australia, G.P.O. Box 2100, Adelaide 5001, Australia*
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We calculate the helium double photoionization and ionization with excitation cross sections using the convergent close-coupling formalism. We investigate the effect of the ground-state correlations by employing various highly correlated ground-state wave functions: 6-, 10-, and 14-parameter Hylleraas and 7-, 15-, and 18-term multiconfiguration Hartree Fock. As a test of the quality of our calculation, we compare the double to single photoionization cross-section ratio calculated in the three different gauges: length, velocity, and acceleration. We also calculate the partial photoionization cross sections up to $n \leqslant 6$ ion excited states and compare them with the latest experimental data. Using the 14-parameter Hylleraas ground state, we obtain almost identical results for the three gauges from the double ionization threshold to 1 keV, and have good agreement with the recent measurements. [S1050-2947(98)02204-5]

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I. INTRODUCTION

The single photoionization with excitation and double photoionization of helium is consistently attracting a great deal of attention. This is true both experimentally, see Refs. [1–4], for example, and theoretically, see [5–12]. Various theoretical methods have been tried to describe these fundamental processes. In particular, it has been demonstrated that the convergent close-coupling (CCC) method can be applied successfully to the helium double photoionization problem [8]. The method obtained the fraction of the double photoionization events, consistent with the most recent experiments, in a wide photon energy range.

In the CCC formalism we consider double photoionization as a two-stage process. Single ionization is followed by electron-impact ionization of the resultant He^+ ion. The one-electron states of the He^+ ion are described by a Laguerre square-integrable basis with excitation of the positive-energy pseudostates corresponding to ionization of the He^+ ion, and therefore the double ionized channels for photon-impact ionization of helium. The CCC method has been tested for the e-He^+ scattering system [15], and has yielded quantitative agreement with the measurements of electron-impact total ionization cross section, suggesting that the method should obtain accurate double photoionization cross sections for helium.

In this paper we concentrate on the numerical aspects of the photoionization of helium calculations. The primary limitation on the accuracy of our approach is the description of the two-electron correlations in the helium atom ground state. This problem is addressed by employing highly correlated helium ground-state wave functions. Previously [8] we employed a seven-term MCHF expansion comprising s, p, and d orbitals. The double-to-single photoionization ratio was calculated in three different gauges of the electromagnetic interaction: length, velocity, and acceleration. The velocity and acceleration forms were within 5% of each other, but the length form was much too large and not presented. It was argued that the calculation in the length form enhanced large distances where the variational ground-state wave function was inaccurate. However, there was another line of argument [5] suggesting that the erroneous large distance behavior may be due to the positive energy pseudostates. Indeed, these states fall off exponentially at large distances whereas the true Coulomb waves have an infinite tail.

We demonstrate that by employing an increasingly accurate ground-state wave function and using the same set of positive energy pseudostates, we can make all three forms of the calculation convergent to within a few percent. We test two different types of ground-state wave functions: the multiconfiguration Hartree-Fock (MCHF) expansion [16] and the explicitly correlated Hylleraas expansion [17].

The paper is organized in the following way. In Sec. II we give details of the ground-state calculations. In Sec. III we briefly outline the CCC formalism. In Sec. IV A we present results of the double photoionization cross-section calculations. Results of ionization with excitation calculations are given in Sec. IV B

II. HELIUM-ATOM GROUND STATE

In our previous work on helium double photoionization [8] we employed a seven-term MCHF expansion comprising s, p, and d orbitals. This gave the ground-state energy of 2.90181 a.u. recovering 95.4% of the correlational energy. Presently, we start with essentially the same seven-term MCHF expansion but with a slightly different order of calculating the correlational orbitals. It gives us almost the same ground-state energy of 2.902 12 a.u., but allows us to expand

*Electronic address: ask107@ample2e.anu.edu.au
†Electronic address: I.Bray@flinders.edu.au

TABLE I. Parameters of various helium atom ground states.

Wave-function type	Energy Total (a.u)	Energy Correlation (%)	σ^{2+}/σ^{+} ($\times 100\%$)
MCHF			
$\{2s,3p,4d\}$	2.90212	96.19	1.84
$\{4s,4p,5d,5f,5g\}$	2.90314	98.62	1.80
$\{4s,4p,5d,6f,6g,6h\}$	2.90326	98.90	1.80
Hylleraas			
6-term [a]	2.90324	98.85	1.83
10-term [b]	2.90360	99.72	1.72
14-term [c]	2.90370	99.95	1.74
''Near exact'' [d]	2.90372	100	
Experiment			1.72 [e]

[a]Green *et al.* [19].
[b]Chandrasekhar *et al.* [20].
[c]Chandrasekhar and Herzog [18].
[d]Frankowski and Pekeris [29].
[e]Spielberger *et al.* [22].

the basis to 15 terms and then subsequently to 18 terms. The orbitals included in the basis are shown in Table I, where we indicate the largest principal quantum number included for a given orbital momentum. Thus the seven-term $\{2s,3p,4d\}$ expansion has the form

$$\Phi_7 = C_{1s}|1s^2\rangle + C_{2s}|2s^2\rangle + C_{3s}|3s^2\rangle + C_{2p}|2p^2\rangle + C_{3p}|3p^2\rangle + C_{3d}|3d^2\rangle + C_{4d}|4d^2\rangle. \quad (1)$$

The Hylleraas-type wave functions are a power series of the three parameters $u=r_{12}$, $s=r_1+r_2$, and $t=r_1-r_2$. The biggest 14-term expansion used in the present work has the form [18]

$$\Psi_{14} = Ne^{-zs}(1 + a_1 u + a_2 t^2 + a_3 s + a_4 s^2 + a_5 u^2 + a_6 su + a_7 t^2 u + a_8 u^3 + a_9 t^2 u^2 + a_{10} s t^2 + a_{11} s^3 + a_{12} t^2 u^4 + a_{13} u^4). \quad (2)$$

The six-term expansion [19] and ten-term expansion [20] are truncated at a_5 and a_9, respectively.

In addition to the ground-state energies, we show in Table I the asymptotic ratio of the double to single photoionization cross sections σ^{2+}/σ^{+} in the high photon energy limit. This ratio can be calculated solely from the ground-state wave function [21] and gives an important indication of the accuracy of the given ground state. Experimentally this ratio was measured by Spielberger *et al.* [22], who separated the Compton scattering from the photoionization and reported the value of 1.72%.

As is seen from the table, the Hylleraas-type wave functions give better ground-state energies as well as the σ^{2+}/σ^{+} ratios as compared to the MCHF ground states with a comparable number of terms. This makes them better candidates for accurate double photoionization calculations. However, because of the explicit dependence on the interelectron separation r_{12}, these require a more elaborate implementation.

As was shown by Green *et al.* [19], a Hylleraas-type wave function can be expanded over the Legendre polynomials depending on the angle between the coordinates of the two electrons as

$$\Psi_n = \sum_L \Phi_n^L P_L(\theta_{12}). \quad (3)$$

The radial part Φ_n^L can be expressed in terms of the Slater functions

$$F_l(r_1,r_2) = \frac{r_<^l}{r_>^{l+1}}, \quad (4)$$

where $r_<$ and $r_>$ are the lesser and the greater of r_1 and r_2, respectively. For the six-term Hylleraas wave function this expansion has the form

$$\Phi_6^0 = Ne^{-z(r_1+r_2)}\left\{1 + a_1\left[(r_1^2+r_2^2)F_0 - \frac{2}{3}r_1 r_2 F_1\right] + a_2(r_1-r_2)^2\right\}, \quad (5)$$

$$\Phi_6^{L\neq 0} = Ne^{-z(r_1+r_2)} a_1 r_1 r_2 \left[\frac{F_{L+1}}{2L+3} - \frac{F_{L-1}}{2L-1}\right]. \quad (6)$$

Analogous expressions can be easily derived for Ψ_{10} and Ψ_{14}.

By expanding the Legendre polynomials in Eq. (4) over the spherical harmonics

$$P_L(\theta_{12}) = \frac{4\pi}{2L+1}\sum_{M=-L}^{L} Y_{LM}^*(\hat{\mathbf{n}}_1) Y_{LM}(\hat{\mathbf{n}}_2), \quad (7)$$

we separate out completely the coordinates $\mathbf{r}_1$ and $\mathbf{r}_2$ and thus reduce the problem to the independent particle model. So the CCC formalism developed in our previous work [8] is readily applicable.

III. CCC FORMALISM

The convergent close-coupling (CCC) method was introduced for the electron-hydrogen scattering by Bray and Stelbovics [23], and then extended to hydrogenic targets by Bray [24]. The CCC method has already been applied successfully to the e-He^+ scattering system [15].

In the present study we employ the CCC method to describe the electron-impact excitation and ionization of the He^+ ion occurring after the single photoionization process. The one-electron states of the He^+ ion are described by a Laguerre square-integrable basis with excitation of the positive-energy pseudostates corresponding to ionization of the He^+ ion, and therefore the double ionized channels for photon-impact ionization of helium.

We use the multichannel expansion for the final state wave function of the system He^+ ion plus ejected electron:

$$|\Psi_j^{(-)}(\boldsymbol{k}_b)\rangle = |j\boldsymbol{k}_b^{(-)}\rangle + \sum_i \int\!\!\!\!\!\!\diagup d^3k \frac{\langle \boldsymbol{k}^{(+)} i|T|j\boldsymbol{k}_b^{(-)}\rangle}{E-\varepsilon_k-\epsilon_i+i0}|i\boldsymbol{k}^{(+)}\rangle, \tag{8}$$

with boundary conditions corresponding to an outgoing wave in a given channel $|j\boldsymbol{k}_b^{(-)}\rangle$ and incoming waves in all other channels $|i\boldsymbol{k}^{(+)}\rangle$.

Here $E=k_b^2/2+\epsilon_j$ is the final-state energy. The channel wave function $|j\boldsymbol{k}_b^{(-)}\rangle$ is the product of a one-electron orbital $\overline{\phi_j}$, obtained by diagonalizing the He^+ Hamiltonian in a Laguerre basis, and a Coulomb $Z=1$ outgoing wave $\chi^{(-)}(\boldsymbol{k}_b)$ or bound state. The sum and integral sign in Eq. (8) indicates a sum over the hydrogen $(Z=1)$ bound states and an integral over the continuum $(\varepsilon_k=k^2/2)$. The half off-shell T matrix in Eq. (9) is the solution of the corresponding Lippmann-Schwinger integral equation [24]:

$$\begin{aligned}\langle \boldsymbol{k}^{(+)} i|T|j\boldsymbol{k}_b^{(-)}\rangle &= \langle \boldsymbol{k}^{(+)} i|V^S|j\boldsymbol{k}_b^{(-)}\rangle \\ &+ \sum_{i'} \int\!\!\!\!\!\!\diagup d^3k' \frac{\langle \boldsymbol{k}^{(+)} i|V^S|i'\boldsymbol{k}'^{(-)}\rangle\langle \boldsymbol{k}'^{(-)} i'|T|j\boldsymbol{k}_b^{(-)}\rangle}{E-\varepsilon_k-\epsilon_{i'}+i0}.\end{aligned} \tag{9}$$

This only needs to be calculated for the total spin $S=0$ and total orbital angular momentum $J=1$. The potential matrix elements of V^S may be found in Ref. [24].

The photoionization cross section, as a function of the photon energy ω, corresponding to a particular bound electron state j is given by [25]

$$\sigma_j(\omega) = \frac{4\pi^2}{\omega c}\sum_{m_j} \int d^3k_b |\langle \Psi_j^{(-)}(\boldsymbol{k}_b)|\mathcal{D}|\Psi_0\rangle|^2 \times \delta(\omega - E_f + E_0), \tag{10}$$

where $c\simeq 137$ is the speed of light in atomic units.

The dipole electromagnetic operator $\mathcal{D}$ can be written in one of the following forms commonly known as length, velocity, and acceleration [25]:

$$\mathcal{D}^r = \omega(z_1+z_2),$$

$$\mathcal{D}^\nabla = \nabla_{z_1}+\nabla_{z_2}, \tag{11}$$

$$\mathcal{D}^{\dot\nabla} = \frac{2}{\omega}\left(\frac{z_1}{r_1^3}+\frac{z_2}{r_2^3}\right).$$

The dipole matrix element with the CCC final-state wave function of Eq. (8) can be calculated as

$$\begin{aligned}\langle \Psi_j^{(-)}(\boldsymbol{k}_b)|\mathcal{D}|\Psi_0\rangle &= \langle j\boldsymbol{k}_b^{(-)})|\mathcal{D}|\Psi_0\rangle \\ &\times \sum_i \int d^3k \frac{\langle j\boldsymbol{k}_b^{(-)}|T|\boldsymbol{k}^{(+)} i\rangle\langle \boldsymbol{k}^{(+)} i|\mathcal{D}|\Psi_0\rangle}{E-k^2/2-\epsilon_i+i0}.\end{aligned} \tag{12}$$

After some angular momentum algebra the first-order dipole matrix element $\langle \boldsymbol{k}^{(+)} i|\mathcal{D}|\Psi_0\rangle$ breaks down into one-electron radial integrals and simple angular coefficients.

We separate the contribution from the final channels $|j\boldsymbol{k}_b^{(-)}\rangle$ into single and double ionization according to the energy of the ϵ_j, which is positive for the doubly ionized channels and negative for the singly ionized channels. We also ensure that the negative-energy state cross sections, which contribute to the ionization plus excitation cross sections, are multiplied by the projection of the state onto the true target discrete subspace [26]. This way we have a relatively clear separation between the discrete and continuous spectrum of the e-He^+ excitation.

IV. RESULTS

With the hope to improve our earlier results [8], and to obtain an accurate cross section for ionization with excitation to $n=6$ levels, we tested various ground-state wave functions with the same set of CCC parameters. The He^+ target space was expanded using $N_\ell=17$ each of s, p, d, and f states, i.e., a total of 68 states. The calculations have been performed at around 100 energy points suitably distributed

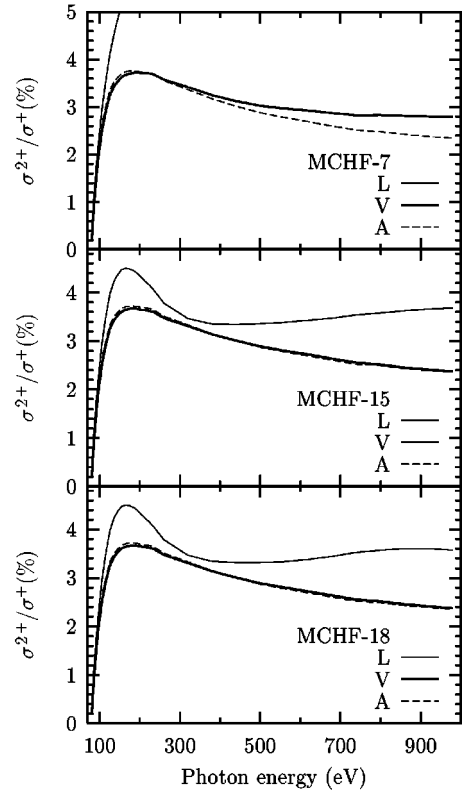

FIG. 1. Double to single photoionization cross-section ratio calculated with the indicated MCHF wave functions; see text.

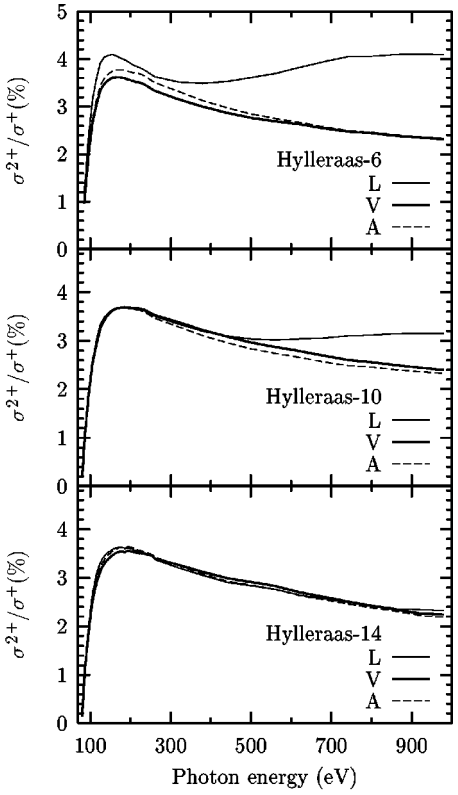

FIG. 2. Double-to-single photoionization cross-section ratio calculated with the indicated Hylleraas-type wave functions; see text.

over the presented photon energy range. Note that no averaging of the CCC results is undertaken, we rely on taking sufficiently large Laguerre bases to ensure that pseudoresonances are of sufficiently small magnitude [27]. In addition, for each energy we vary the Laguerre exponential fall-off so that the total energy bisects two of the pseudothresholds. This way we ensure that the integration rule, induced by the diagonalization of the target Hamiltonian, has the total energy E as one of the endpoints, see Ref. [28] for more detail. The variation of the fall-off factors gives us an immediate estimate of the accuracy of the results, within the $\ell \leqslant 3$ model, by simply observing any oscillations. We also performed smaller calculations with $\ell \leqslant 2$, which do not differ substantially from those presented, thereby confirming convergence with increasing target-space orbital angular momentum ℓ.

A. Double photoionization

The double-to-single photoionization ratio $R(\omega) = \sigma^{2+}/\sigma^{+}$, as a function of the photon energy ω, is presented in Figs. 1 and 2 for MCHF and Hylleraas ground states, respectively. The MCHF-7 results are essentially the same as those presented earlier [8]. On this plot we also show the length form calculation, which exceeds the scale of the picture and was not shown in our earlier work. As we increase the number of configurations in the MCHF expansion to 15, the velocity and acceleration forms become identical. The length form improves considerably, but is still substantially above the other two. Further enlargement of the MCHF basis to 18 configurations does not improve the previous result in any way. Thus we believe that the MCHF

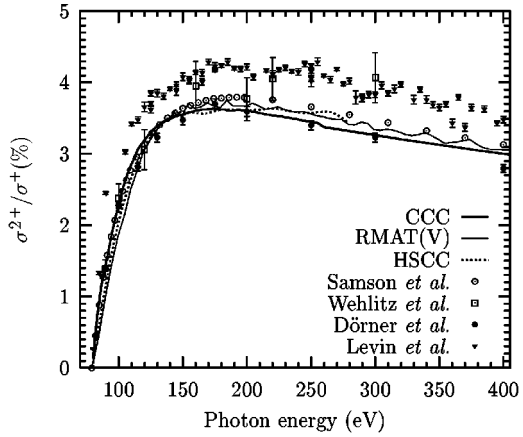

FIG. 3. Double to single photoionization cross-section ratios. The CCC results are using the Hylleraas-14 ground state with the length gauge; see Fig. 2. The velocity gauge eigenchannel R-matrix calculations [11] are denoted by RMAT(V). The hyperspherical close-coupling results of Tang and Shimamura [6] are denoted by HSCC. The experimental data are due to Samson *et al.* [1], Dörner *et al.* [4], Wehlitz *et al.* [2], and Levin *et al.* [3].

wave function cannot describe the two-electron correlation at large distances. At least it cannot be practically achieved with a reasonable number of configurations.

Let us now turn to the Hylleraas-type wave functions, see Fig. 2. The six-parameter Hylleraas wave function has an energy comparable to the MCHF-18 (see Table I). Interestingly enough, the double-to-single photoionization ratio is also similar for these two functions (cf. part 3 of Fig. 1 and part 1 of Fig. 2). Increasing the number of terms to 10 in the Hylleraas expansion improves the length form dramatically. Now it is consistent with the two other forms and deviates slightly only at large photon energies approaching 1 keV. A further step to Hylleraas-14 makes the length form very similar to the other two.

The results of the Hylleraas-14 length-gauge calculation together with other recent experimental data are shown in Fig. 3. In addition, comparison is given with those theories, which also have gone to some considerable effort to obtain a very accurate ground state. These include the hyperspherical close-coupling (HSCC) approach of Tang and Shimamura [6] (available below 300 eV), and the eigenchannel R-matrix (RMAT) results of Meyer, Greene, and Esry [11] (available below 500 eV). The former approach is attractive because the HSCC method is applied to both the ground and final state of the system. The HSCC results have been given in the length and acceleration gauges only, which are very similar. Though the velocity form has not been given, we expect it to be very similar to the other two. The RMAT approach is novel in that it used a finite element approach and also obtained a very accurate ground state. In this calculation the velocity and acceleration gauges are almost identical, but the length form begins to diverge a little at high energies.

Only statistical errors of the experiments have been given. Though the Samson *et al.* [1] data are somewhat above that of Dörner *et al.* [4], taking systematic errors into account suggests that the experiments are consistent with each other. In this case, perhaps the presented CCC results are the most accurate.

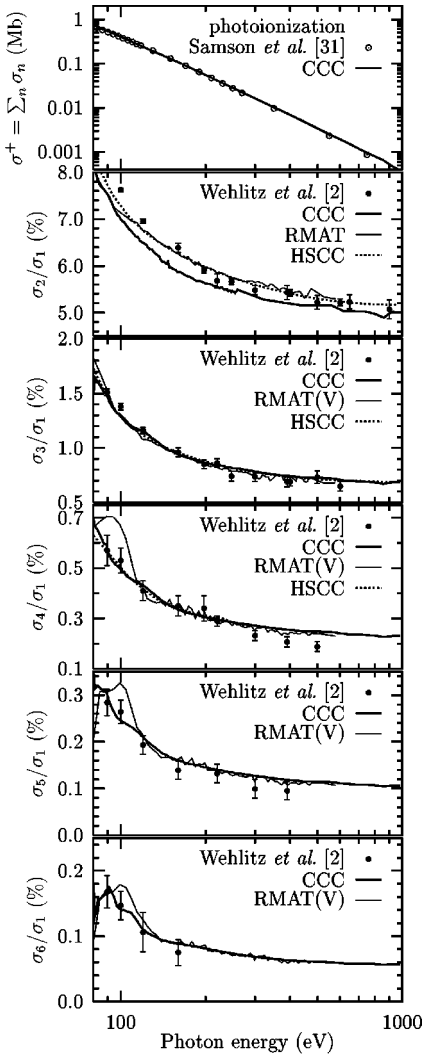

FIG. 4. Photoionization with excitation cross sections to various n ion states. The measurements are from the indicated references. The CCC theory uses the Hylleraas-14 ground state within the length gauge (other gauges are almost identical; see Fig. 2). The HSCC theory is due to Tang and Burgdörfer [10]. The RMAT(V) calculations are due to Meyer *et al.* [30].

We consider that further improvement of the present theory requires either a very substantial enhancement of the CCC basis or, more likely, a bigger Hylleraas expansion. Such expansions are known to produce extremely accurate helium atom ground states (see, for instance, Ref. [29]). However, their practical use requires substantial rewriting of the existing CCC code.

B. Ionization with excitation

We use the Hylleraas-14 length-gauge calculations to produce the single photoionization cross sections corresponding to various ion states $n=1,\ldots,6$. The results are shown in Fig. 4. As above, we compare with recent experiment and the HSCC calculations of Tang and Burgdörfer [10] and RMAT calculations of Meyer *et al.* [30]. Comparison of the HSCC results with other theories has been given in Ref. [10].

As a first check of our calculations, we find that our single photoionization cross section corresponding to the ion ground state $n=1$ is identical in the three gauges and is in very good agreement with the most recent data due to Samson *et al.* [31]. The photoionization cross-section ratio σ_n/σ_1 is presented for $n=2,\ldots,6$, in comparison with the recent measurement of Wehlitz *et al.* [2]. The convergence between the three gauges is even better than for the case of double ionization, so only the length form is presented. The agreement with the experimental data is generally good for all n, though suprisingly less so for $n=2$. Generally, our results are supported by the hyperspherical close-coupling calculations of Tang and Burgdörfer [10] (available for $n \leqslant 4$ only) and the eigenchannel calculations of Meyer *et al.* [30]. We find it particularly remarkable that the latter calculations are able to obtain such accuracy for the $n=6$ states, as these extend outside the R-matrix boundary. In the CCC theory we used large radii (200 a.u.) and basis sizes (17 states for each l) so as to ensure that up to $n=7$ states were well-described in the calulations. It is extremely encouraging to find two such diverse implementations of the close-coupling method yielding very similar results for the $n \leqslant 6$ states. Incidentally, a calculation with only eigenstates results in poor agreement with experiment and between the three gauges.

TABLE II. Asymptotic ratios σ_n/σ_1 for the helium single photoionization to various ion excited states at the limit of high photon energy. The calculation of Andersson and Burgdörfer [32] is at the photon energy of 2 keV. The results of Wehlitz *et al.* [2] are extrapolations of measurements.

n	MCHF-18	Hylleraas-14	Ref. [2]	Ref. [32]
2	4.782	4.727	4.89	4.80(13)
3	0.605	0.597	0.62	0.543(33)
4	0.200	0.199	0.21	0.118(37)
5	0.092	0.092	0.095	0.048(30)
6	0.050	0.050	0.052	

In Table II we present the asymptotic values of ratios σ_n/σ_1 in the limit of high photon energies. Similarly to the fraction of the double ionization events, these ratios can be calculated solely from the ground-state wave function and do not require CCC computations. Comparison is given with the values calculated with the best MCHF and Hylleraas wave functions used in the double photoionization calculations. We also present the experimental values extrapolated by Wehlitz *et al.* [2] and the calculation of Andersson and Burgdörfer [32] at the photon energy of 2 keV. The present data are consistent with the experiment of Wehlitz *et al.* [2] and compare well with the earlier calculation of Andersson and Burgdörfer [32] for $n \leqslant 3$.

V. CONCLUSIONS

We have demonstrated that improvement of the helium atom ground-state wave function allows accurate calculations of the helium double photoionization and ionization with excitation using the convergent close-coupling method. Agreement between the double-to-single photoionization cross-section ratio obtained with the electromagnetic interaction in the three gauges—length, velocity, and acceleration—is very good, with the difference among the three not exceeding 3% in a wide photon energy range from

the threshold up to 1 keV. The ionization with excitation (to $n \leq 6$ states) cross sections has also been calculated and found to be in good agreement with the recent measurement of Wehlitz *et al.* [2].

The primary strength of the present calculations over others is the good agreement between the three gauges over such a large energy range and the ability to obtain excitation cross sections up to $n \leq 6$ over the same energy range.

It is our view that here we have primarily demonstrated the numerical robustness of the CCC approach in calculating integrated cross sections. Given the claim that close-coupling theories should obtain, in the limit of infinite basis size, a step function in the energy distribution of the two-electron continuum [13], we are very keen to investigate in detail the energy distributions within the double-ionization channel; see [14] for example. The angular distributions will also be investigated. This work is currently underway.

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