Excitation Ionization and Double Ionization of Helium by High-Energy Photon Impact

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The ratio of double to single ionization by photoabsorption is calculated for photon impact energies in the range 2–18 keV. Employing the acceleration form of the dipole operator and representing the electron-electron correlation in the final state by a Coulomb distortion factor, $R=\sigma^{++}/\sigma^{+}$ approaches an asymptotic value of 1.66% and reaches a value of about 2.5% at 2 keV photon energy. Comparison with experimental data shows good agreement when contributions due to Compton scattering are taken into account.

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Double ionization of helium (and other two-electron systems) by photoabsorption provides a sensitive test for electron-electron correlation. The coupling between the electrons and the radiation field ($\propto \mathbf{p}\cdot\mathbf{A}$) is a one-body operator. The simultaneous ejection of two electrons is therefore mediated through the electron-electron interaction in the initial state ("ground-state correlation") and/or in the final state of two electrons in the Coulomb continuum ("final-state correlation").

As early as the late 1960s it was recognized that, in the (nonrelativistic) limit of high photon energies ($E\rightarrow\infty$), the ratio of double to single ionization $R=\sigma^{++}/\sigma^{+}$ by photoabsorption provides a sensitive measure for electron correlation for which accurate calculations became possible. Byron and Joachain [1], Åberg [2], Brown and Gould [3], and Amusia *et al.* [4] obtained values ranging from $R(E\rightarrow\infty)=1.6\%$ to 2.3%. Only very recently with the advent of synchrotron light sources have measurements of this ratio for photon energies in the keV region become available [5–7] and they have stimulated renewed theoretical interest in this problem. At issue is not only the correct nonrelativistic asymptotic value (which, strictly speaking, cannot be realized because of the onset of relativistic effects) but also the behavior at large but finite energies.

Remarkably enough, large discrepancies exist among calculations for this fundamental quantity. Brown and Gould [3] calculated the dipole matrix element in the velocity gauge, using an independent-electron final-state wave function. They obtained $R\approx 2.5\%$ at 2 keV and an asymptotic value of 2%. The MBPT calculation of Ishihara, Hino, and McGuire [8], resulted in $R=1.6\%$ at $E=2.8$ keV, in perfect agreement with, at that time, the only available data point [5] above Carlson's measurement [9] at 625 eV, but is in relatively poor agreement with the data and the MBPT calculation of Carter and Kelly [10] in the threshold region. The authors attributed this discrepancy to differences in basis sets and to the omission of higher-order diagrams included by Carter and Kelly.

In the present Letter we present calculations of $R(E)$ for photoabsorption in the 2–18 keV photon-energy range. We employ a 20-parameter Hylleraas-type wave function [11] for ground-state He which satisfies the cusp condition at the origin to a good degree of approximation in order to accurately represent ground-state correlation. Furthermore, we implement for the first time a correlated distorted final-state wave function. The direct treatment of the three-body Coulomb continuum is avoided by considering instead excitation ionization and employing sum rules [1,2]. The final state with one continuum electron of momentum $\mathbf{k}$ ($k=\sqrt{2\epsilon}$) and one bound electron in a $He^{+}(nlm)$ state is here represented by the wave function

$$\Psi_f^{(-)}(\mathbf{r}_1,\mathbf{r}_2)=(1/\sqrt{2})[\Phi_{nlm}(\mathbf{r}_1)\Phi_{\mathbf{k}}^{(-)}(\mathbf{r}_2) \times D^{(-)}(\mathbf{k}_{12},\mathbf{r}_{12})+\mathbf{r}_1\leftrightarrow\mathbf{r}_2], \quad (1)$$

where Φ_{nlm} and $\Phi_{\mathbf{k}}^{(-)}$ are bound and continuum states of He^{+}, respectively, and

$$D^{(-)}(\mathbf{k}_{12},\mathbf{r}_{12})=e^{-\pi\alpha/2}\Gamma(1-i\alpha) \times {}_1F_1[i\alpha,1,-i(k_{12}r_{12}+\mathbf{k}_{12}\cdot\mathbf{r}_{12})] \quad (2)$$

is a Coulomb distortion factor describing the electron-electron interaction. $\mathbf{r}_{12}$ denotes the interelectronic position vector, $\mathbf{k}_{12}=\mathbf{k}/2$ is the relative momentum of the electron-electron system, and $\alpha=1/k$. This final-state wave function is the bound-free analog to the two-electron continuum wave function used by Brauner, Briggs, and Klar [12] in their treatment of $(e,2e)$ processes, and by Maulbetsch and Briggs [13] in a study of the asymmetry parameter for double photoionization of He close to threshold. For two electrons well separated in phase space, the distorted wave [Eq. (1)] is expected to be accurate.

We show numerically that the nonrelativistic asymptotic limit for photoabsorption $R(E\rightarrow\infty)$ is, indeed, independent of final-state correlation, as recently found by Dalgarno and Sadeghpour [14]. Furthermore, we determine the energy dependence of R and extract the leading term of an E^{-1} expansion which is strongly influenced by final-state correlations. Finally, in order to compare with the high-energy experimental data, which do not discrim-

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inate against inelastic photon scattering, we include contributions from Compton scattering, which sets in at ~ 3 keV.

The cross section for ionization of one electron into a continuum state of energy ϵ and angular momentum l and excitation of the second electron to a $He^+(nl')$ state is proportional to the oscillator strength

$$df(\epsilon l,nl')/d\epsilon = \tfrac{2}{3} E |\langle \epsilon l,nl'|\mathbf{d}|i\rangle|^2 , \quad (3)$$

where $\mathbf{d}$ is the dipole operator. In (3), $|\epsilon l,nl'\rangle$ denotes the distorted final state [Eq. (1)] decomposed into partial waves of the continuum and bound-state electrons and coupled to total angular momentum $L_{\rm tot}=1$, and $|i\rangle$ is the Hylleraas-type correlated ground-state ^1S wave function. The continuum state is normalized on the energy scale. Following Dalgarno and Sadeghpour [14], we use $\mathbf{d}$ in the acceleration gauge in which the wave function is probed near the origin and which is least sensitive to errors in the final state at high energies. Simultaneously performed calculations of the oscillator strengths in the velocity gauge show good agreement [15] since the ground-state wave function used satisfies the cusp condition to a good degree of approximation.

The single-ionization cross section is proportional to the sum of $df(\epsilon l,nl')/d\epsilon$ over all bound states of He^+,

$$\sigma^+ \propto \sum_n \sum_{l,l'} df(\epsilon l,nl')/d\epsilon , \quad (4)$$

with $l=l'\pm 1$ due to dipole selection rules, while the cross section for single *plus* double ionization is proportional to the quantity

$$\sigma \propto \sum_{l,l'} \left[\sum_n \frac{df(\epsilon l,nl')}{d\epsilon} + \int_0^{E-E_B} d\epsilon' \frac{df(\epsilon l,\epsilon' l')}{d\epsilon d\epsilon'} \right] , \quad (5)$$

where E_B is the total binding energy of He. The ratio R is, in terms of (4) and (5),

$$R=(\sigma-\sigma^+)/\sigma^+ . \quad (6)$$

The calculation of σ is facilitated in the asymptotic limit ($E\rightarrow\infty$) by the closure property of the He^+ eigenfunctions, which permits obtaining σ without reference to the two-electron continuum states [14,16] of He, provided that the upper limit of the integral in (5) tends to infinity. For finite E this is not strictly correct since energy conservation $\epsilon+\epsilon'=E-E_B$ must be satisfied and the upper limit of the integral in Eq. (5) is finite. However, the error introduced by extending the upper limit to infinity is small for E sufficiently large. We have verified, using independent-electron final states, that this approximation introduces an error in σ of the order of 10^{-4} at $E=2$ keV, and that the error decreases further as E increases. Note, however, that at lower energies the closure approximation breaks down, leading to an overestimate of double ionization and of R.

Both $\Phi_{\mathbf{k}}^{(-)}$ and the distortion factor $D^{(-)}$ were expanded in partial waves and coupled to s and p waves. In each expansion, partial waves up to $l=4$ were retained until convergence was achieved. For the bound states, s and p states were included. We label the different partial-wave contributions $[(l_C,l_{12}),L,l_B]$, where l_C and l_{12} denote the partial waves of $\Phi_{\mathbf{k}}^{(-)}$ and $D^{(-)}$, respectively, L is the angular momentum resulting from coupling l_C and l_{12}, and l_B is the angular momentum of the bound electron.

The distortion factor $D^{(-)}$ describes the electron-electron interaction following the absorption of an energetic photon by one electron in the ground state. This process, often referred to as two-step one (TS1), amounts to an exchange of energy (in the monopole limit [17]) and of angular momentum of the fast electron on its "way out" with the residual electron, either bound or in a low-lying continuum state.

For a detailed analysis of the high-energy behavior of R for photoabsorption we plot our results in Fig. 1, according to an asymptotic high-energy expansion around $E^{-1}=0$

$$R(E)=R(\infty)+a_{-1}E^{-1}+a_{-2}E^{-2}+\cdots . \quad (7)$$

We have calculated R with and without the distortion factor $D^{(-)}$ in the final-state wave function in order to illustrate the influence of final-state correlation at finite, but large, E. Omitting $D^{(-)}$ and using an unscreened Coulomb wave for the continuum electron yield R to be essentially independent of energy down to 2 keV. Our calculation with both correlated and uncorrelated final-state wave functions converge to the same, universal, value of $R(\infty)=1.66\%$, in perfect agreement with the

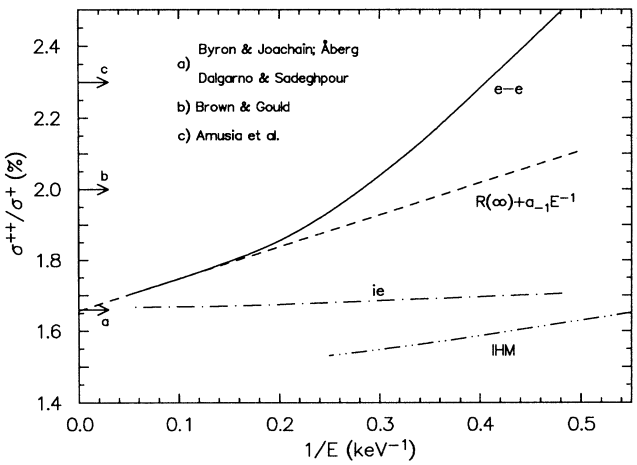

FIG. 1. $R=\sigma^{++}/\sigma^+$ as a function of E^{-1}. Present results: dash-dotted curve, independent-electron final state (ie); solid curve, final state including electron-electron distortion (e-e); dashed curve, linear extrapolation of the e-e curve correct to order E^{-1}. Dash-double-dotted curve, MBPT calculation by Ishihara, Hino, and McGuire [8]. Arrows represent asymptotic calculations of Refs. [1–4,14]. Note all calculations include only photoabsorption.

early calculations of Byron and Joachain [1] and Åberg [2], using the accurate 39-parameter Hylleraas-type initial state of Kinoshita [18], as well as the very recent analysis by Dalgarno and Sadeghpour (upon correction of a small numerical error [14]). This confirms the conclusion drawn by these authors that the exact value of $R(\infty)$ is independent of correlation in the final state and can be obtained by using an accurate representation of the ground-state wave function provided that a gauge is employed that emphasizes the contributions from small distances from the nucleus. Deviations from this value are due to the usage of a less accurate wave function and/or a different gauge.

Furthermore, we find that the leading-order correction as $E \rightarrow \infty$ is proportional to E^{-1} down to about 5 keV. Because of long-range Coulomb forces, terms proportional to $\ln E$ could be present in Eq. (7) as well. However, as is evident from Fig. 1, their contribution is small at high energies. Unlike $R(\infty)$, we find the asymptotic coefficient a_{-1} to be very sensitive to final-state correlation. For uncorrelated final states a_{-1} is very small ($a_{-1}=0.03$ keV). For correlated final states, both the angular-momentum exchange and energy exchange mediated by the TS1 contribute to a_{-1} ($a_{-1}=0.90$ keV). The angular momentum exchange in the exit channel leads to an admixture of the $(\epsilon s, np)$ final channels ([(0,0),0,1] in our notation) to the dominant channels [(1,0),1,0] representing an ϵp ionized electron and an ns "shakeup" electron. Further contributions to a_{-1} come from the [(0,1),1,0] term signifying the scattering of the s wave of the outgoing electron at the noncentral force field of the residual electron resulting in an asymptotic p wave. Contributions from combinations of higher angular momenta have little influence in the asymptotic region but become increasingly important at lower photon energies.

In order to compare our theoretical results with recent experimental data, which do not discriminate against inelastic photon scattering, ionizing Compton scattering must be taken into account [19,20]. This final channel will, in fact, dominate over photoabsorption for single ionization for photon energies above ~ 5 keV. The single-ionization cross section by Compton scattering σ_C^+ can be estimated either from the incoherent Compton cross section (which includes bound-state excitation) or from convoluting the Klein-Nishina differential cross section with the momentum distribution of ground-state helium and requiring an energy transfer greater than the first ionization potential. Deviations between the two provide a measure for the uncertainty (Fig. 2). The uncertainties are considerably larger for the estimate of double ionization by Compton scattering. σ_C^{++} was estimated similarly to σ_C^+ requiring, however, an energy transfer greater than the second ionization potential and convoluting the single-ionization cross section as a function of the energy E lost by the scattered photon by the ratio $R(E)=\sigma^{++}/\sigma^{+}$. For $R(E)$ we use for the final-state P sector the calculation of Carter and Kelly [10] at low energies and our present calculation at higher energies. For higher angular momenta we use the shakeoff value 0.73% [14]. The physical picture underlying this estimate relies on the fact the asymptotic regions for photoabsorption and ionization by inelastic photon scattering are different. For the photon energies considered here, the (nonrelativistic) asymptotic limit is reached for photoabsorption while they correspond to the near-threshold region for Compton scattering. The dominant values of $R(E)$ entering the convolution originate from E in the range from threshold to ~ 600 eV. Near threshold, $R(E)$ is expected to be strongly dependent on the final-state interaction, which is identical for the two processes, but only weakly dependent on the primary excitation mechanisms ejecting the fast electrons, which are different. With increasing E, Compton scattering accesses, unlike photoabsorption, increasingly higher angular momenta ($L_{\text{tot}} > 1$) in the final state, for which initial- and final-state correlations are expected to be less important. Furthermore, the convolution method may break down at much higher photon energies when the Compton scattered electron itself reaches "asymptotic" energies (~ 1 keV). In that regime the primary excitation mechanism rather than the final-state interaction plays the decisive role in determining $R(E)$.

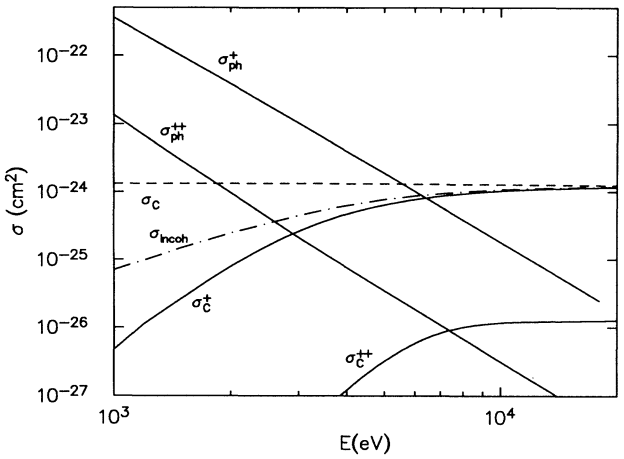

FIG. 2. Cross sections for ionization of He by photon impact. σ_{ph}^+ and σ_{ph}^{++}: present calculation for single and double ionization by photoabsorption. σ_C^+ and σ_C^{++}: single and double ionization by Compton scattering. σ_C and σ_{incoh} signify Compton scattering by two free electrons and incoherent Compton scattering by helium [21], respectively.

The present results for $R(E)$ corrected for Compton scattering are shown together with the experimental data of Levin and co-workers [5,6], Bartlett and Samson [7], and Carlson [9], and the MBPT calculation of Ishihara, Hino, and McGuire [8] in Fig. 3. Different energy regions can be distinguished. Below 3 keV, R is determined by photoabsorption; at intermediate energies ($\sim$3–8 keV) photoabsorption and Compton scattering are com-

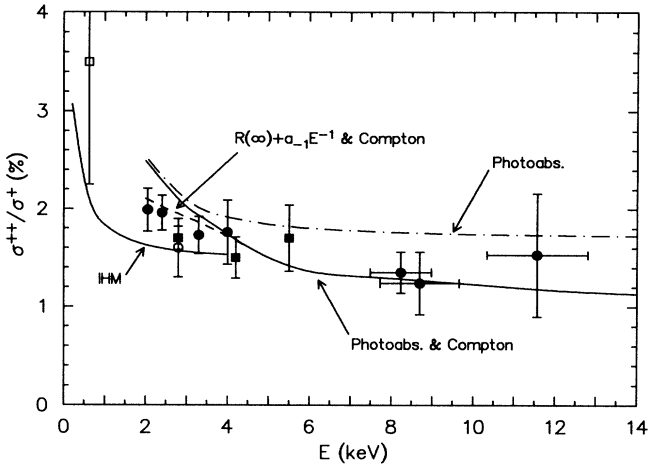

FIG. 3. $R=\sigma^{++}/\sigma^{+}$ vs photon impact energy. Experimental points: open circle, Levin *et al.* [5]; solid circles, Levin *et al.* [6]; solid squares, Bartlett and Samson [7]; open square, Carlson [9]. Solid curve labeled IHM, MBPT calculation by Ishihara, Hino, and McGuire [8]. Present calculation: dash-dotted curve, photoabsorption only; solid curve, photoabsorption and Compton scattering; dashed curve, the leading contribution in the E^{-1} expansion [Eq. (7)] for photoabsorption corrected for Compton scattering.

petitive, with Compton scattering contributing primarily to single ionization thereby lowering the apparent ratio R below the values predicted by photoabsorption. At even higher energies Compton scattering completely dominates. We find good agreement with the experiment for $E \geq 3$ keV.

At lower energies ($E \leq 3$ keV) we observe that the extrapolation of our asymptotic expansion to order E^{-1} (dashed curve in Figs. 1 and 3) which is governed by soft electron-electron collisions in the exit channel appears to improve the agreement with the data. At these energies, the result using the full final-state distortion factor is already strongly influenced by higher-order terms a_{-n} ($n \geq 2$) in the E^{-1} expansion, whose accuracy is less certain. One possible explanation currently under investigation is the overemphasis of close Coulomb collisions built into the Coulomb distortion factor for final-state correlations at intermediate energies.

In conclusion, we have calculated $R=\sigma^{++}/\sigma^{+}$ for photoionization of helium for photon energies in the 2–18 keV range using a final-state wave function with a Coulombic distortion factor representing the electron-electron interaction. We arrive at an E^{-1} high-energy behavior of R with the asymptotic limit $R(\infty)=1.66\%$ in agreement with Byron and Joachain [1], Åberg [2], and Dalgarno and Sadeghpour [14], and a finite energy correction $a_{-1}E^{-1}$ with $a_{-1}=0.90$ keV. Upon correction for Compton scattering our results are consistent with recent synchrotron light measurements.

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Note added.—While this manuscript was reviewed we learned about two other calculations [22,23] of double ionization by photoabsorption which employ the two-electron continuum version of the final-state wave function in Eq. (1).

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