PHYSICAL REVIEW A VOLUME 50, NUMBER 4 OCTOBER 1994

Excitation-ionization and double ionization of helium by Compton scattering

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(Received 21 June 1994)

We investigate the behavior of the cross section for inelastic photon scattering in the limit of high but nonrelativistic photon energies $\omega\to\infty$ at helium leading to two-electron transitions. Our calculation employs fully correlated wave functions for the helium ground state and different forms for approximate two-electron final states. We determine the $\omega\to\infty$ limit for the ratio of double to single ionization. We find the high-energy behavior to differ significantly from the one for photoabsorption. Our results confirm our previous estimate [Andersson and Burgdörfer, Phys. Rev. Lett. **71**, 50 (1993)] but disagree with recent predictions by Hino *et al.* [Phys. Rev. Lett. **72**, 1620 (1994)].

PACS number(s): 32.80.Fb, 32.80.Cy

Simultaneous excitation and ionization as well as double ionization of helium (and other two-electron systems) by photon impact provide a sensitive test for electron-electron correlation. The coupling between the electrons and the radiation field is a one-body operator. The simultaneous ejection or excitation 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 in the late 1960s it was recognized that in the nonrelativistic limit of high photon energies ($E\to\infty$) the ratio of double to single ionization $R_{\rm ph}=\sigma_{\rm ph}^{2+}/\sigma_{\rm ph}^{+}$ by photoabsorption was a very 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 $R_{\rm ph}(E\to\infty)=1.6\%$ 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 [5–8] and stimulated renewed theoretical interest in this problem [9–12]. The theoretically accepted value is now $R_{\rm ph}=1.67\%$ [1,2,13,14].

The experimental determination of $R_{\rm ph}$ faces, however, a fundamental difficulty: For sufficiently high photon energies $E=\hbar\omega\gtrsim 6$ keV, where this asymptotic value should be reached, the photoabsorption cross section becomes small compared to the cross section for inelastic photon scattering (Compton scattering), which also causes two-electron transitions [15]. Since Compton scattering and photoabsorption lead to different final states, they are, in principle, distinguishable. Their experimental separation requires, however, determination of the energy distribution of electrons or of the recoil ion. Such experiments are currently underway [16,17]. Beyond photon energies of ≈ 6 keV Compton scattering dominates the production of singly ionized and doubly ionized helium. Corresponding cross sections have only very recently become the focus of theoretical attention [14,18] and the high-energy value of the ratio $R_C=\sigma_C^{2+}/\sigma_C^{+}$ for Compton scattering is not yet well established. We present in the following, calculations for two-electron transitions by Compton scattering namely simultaneous excitation and ionization of the two electrons as well as double ionization by helium. We also determine the nonrelativistic limit for R_C to which our numerical results converge.

Compton scattering differs from photoabsorption in several important aspects: (i) the relevant interaction operator is A^2 rather than $\mathbf{p}\cdot\mathbf{A}$ for high but nonrelativistic photon energies $\omega\gg\epsilon_i$ (ϵ_i, binding energy of helium). (ii) In high-energy Compton scattering large values of the momentum transfer Q dominate so that dipole-forbidden transitions are important, while total cross sections for photoexcitation and ionization are dominated by the dipole terms. As has been shown recently [19] Compton scattering is closely related to "hard" charged-particle collisions in the binary encounter limit while photoabsorption is related to soft charged-particle collisions in the Bethe-Born limit. One therefore expects the behavior of the excitation-ionization cross sections as well as R_C to be different from the corresponding ratio for photoabsorption.

The doubly differential Compton scattering cross section as a function of the energy transfer ϵ and momentum transfer Q plane for inelastic scattering from an arbitrary initial state $|i\rangle$ to an arbitrary final state $|f\rangle$ may be expressed [20–22] to first order in A^2 as

$$\left(\frac{d^2\sigma}{d\epsilon\, dQ^2}\right)_C=\frac{\pi r_0^2}{2k^2}\left[1+\left(1-\frac{Q^2}{2k^2}\right)^2\right]F_I(\epsilon,Q^2), \qquad (1)$$

with

$$F_I(\epsilon,Q^2)=\int d\Omega_e\sqrt{2(\epsilon+\epsilon_i)}\left|\left\langle f\left|\sum_{j=1}^{N}e^{i\mathbf{Q}\cdot\mathbf{r}_j}\right|i\right\rangle\right|^2 \qquad (2)$$

the inelastic transition form factor (proportional to the generalized oscillator strength [22]) for the N-electron atom integrated over all emission angles of the emitted electron and weighted with the density of continuum final states. In the case of multiple ionization, F_I is understood to include an

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integral over emission angles of all electrons and the proper density of states. As the atomic initial state in Eq. (2) we employ accurate highly correlated ground-state wave functions, namely Hylleraas-type wave functions with 20 [23] and 38 [24] parameters and the configuration-interaction (CI) wave function of Nesbet and Watson [25]. For final states which correspond to either double ionization or simultaneous excitation and ionization we use the following approximations: (a) an uncorrelated final state given by a product of two Coulomb functions

$$\psi_f^{(-)}(\mathbf{r}_1,\mathbf{r}_2)=\frac{1}{\sqrt{2}}[\phi_{nlm}(\mathbf{r}_1)\phi_{\mathbf{k}}^{(-)}(\mathbf{r}_2)+\mathbf{r}_1\leftrightarrow\mathbf{r}_2], \quad (3)$$

(b) an approximate correlated final state following the Brauner-Briggs-Klar [26] prescription

$$\psi_f^{(-)}(\mathbf{r}_1,\mathbf{r}_2)=\frac{1}{\sqrt{2}}[\phi_{nlm}(\mathbf{r}_1)\phi_{\mathbf{k}}^{(-)}(r_2)D^{(-)}(\mathbf{k}_{12},\mathbf{r}_{12}) + r_1\leftrightarrow r_2], \quad (4)$$

where ϕ_{nlm} and $\phi_k^{(-)}$ are bound and continuum states of He^+, respectively, and

$$D_c^{(-)}(\mathbf{k}_{12},\mathbf{r}_{12})=e^{-\pi\alpha/2}\Gamma(1-i\alpha) \times {}_1F_1(i\alpha,1,-i(k_{12}r_{12}+\mathbf{k}_{12}\cdot\mathbf{r}_{12})) \quad (5)$$

is the 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 $\alpha=1/k$. Corresponding expressions are used for two electrons in the continuum. It should be noted that Eq. (4) is a high-energy approximation and ceases to be accurate for small ejection velocities. We use the Coulomb distortion only for low L partial waves. (c) A Byron-Joachain type CI final state with a distortion factor of the form

$$D(E,\mathbf{r}_{12})=1+c(E)r_1r_2P_1(\cos\theta_{12}), \quad (6)$$

with an energy-dependent admixture coefficient $c(E)$, which has been found to reproduce photoionization data [27].

The dominant contribution to Compton scattering in Eq. (1) stems from the "Bethe ridge," i.e., from the region near the free-electron dispersion curve $\epsilon\sim Q^2/2$ [19]. This contrasts with photoabsorption where the optically allowed region corresponds to $Q\approx 0$ or $\epsilon\gg Q^2/2$. Translated into coordinate space this implies that the entire probability distribution of the electron contributes to Compton scattering while only the near-nuclear region contributes to high-energy photoabsorption, since in the latter case the high Q components of the initial-state wave function (and hence the nuclear recoil) are needed in order to balance energy and momentum. This difference manifests itself directly in the behavior of two-electron transitions.

The breakdown of the singly differential cross section $d\sigma^+(ns)/d\epsilon_f$ for excitation of the ns state and ejection of the second electron with energy ϵ_f into partial wave L is shown in Fig. 1 for $n=2$. While the cross section summed over all partial waves is remarkably flat for all energies up to the binary encounter (photon backscattering) limit,

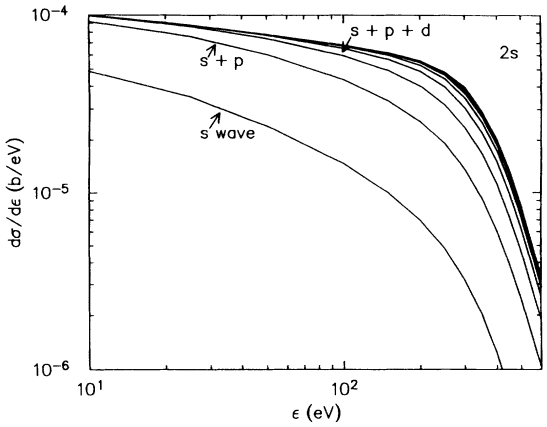

FIG. 1. Cumulative partial wave distribution of the singly differential cross section $d\sigma^+(n)/d\epsilon_f$ for simultaneous excitation of the $2s$ state in He^+ by Compton scattering as a function of the energy of the ionized electron, $\omega=10$ keV [uncorrelated final state, Eq. (3)].

$$0\leq\epsilon_f\lesssim 2\omega^2/(c^2+2\omega), \quad (7)$$

the partial-wave distribution changes dramatically. With increasing energy of the ejected electron the dipole-allowed transition ceases to be important and the cross section is primarily determined by high angular momentum components. Calculations for several different final nl states and different photon energies indicate that the partial-wave distribution is an approximately universal function of energy transfer ϵ (or ϵ_f), but is rather insensitive to the final state nl or the primary photon energy. The range of partial waves L can be estimated from a classical binary-encounter model for a collision between a photon and an electron at its mean radius from

$$L\simeq\sqrt{2\epsilon}\langle r\rangle. \quad (8)$$

Equation (8) reflects the fact that, unlike photoabsorption, the entire electron charge cloud contributes to Compton scattering rather than only the region near the nucleus.

The increasing dominance of high partial waves has a profound effect on the probability of two-electron processes. For excitation-ionization they can be measured by the ratio $R_C^+(nl)=\sigma_C^+(nl)/\sigma_C^+(1s)$ signifying the conditional probability for ionization and simultaneous excitation to the state nl relative to pure single ionization without excitation (Fig. 2). Broken down into different partial wave components of the emitted electron L, we find this ratio to decrease with increasing L. As this ratio measures the electron correlation (mostly) in the initial state, this result indicates that the role of electron-electron correlation is diminished at the outer fringe of the atom compared to the inner region where the low L contribution originates. Furthermore, the calculated average $\langle R_C^+(5s)\rangle_{L\neq 1}$ over all dipole-forbidden transitions weighted with the partial cross section for each L is found to be much smaller than the corresponding ratio for the P sector $\langle R_C^+(1s)\rangle_{L=1}$. Moreover, for large ejection energies near the binary-encounter maximum energy transfer [Eq. (7)] this ratio tends for $L=1$ to the exact value [13] for photoabsorption

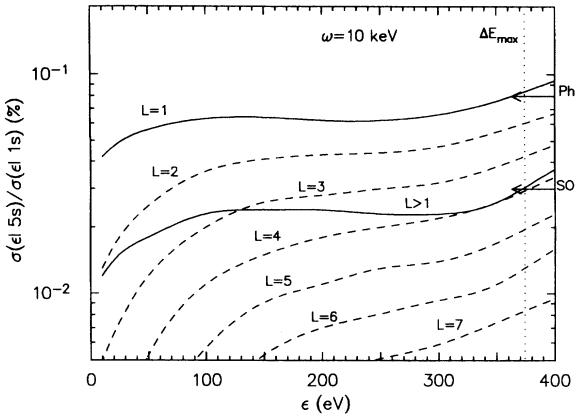

FIG. 2. Ratios of the cross sections for simultaneous ionization and excitation to $5s$ to pure ionization without excitation by Compton scattering as functions of the energy of the ejected electrons. The solid lines represent the $L=1$ and $L>1$ contributions. The dashed lines show the individual contributions from $L=2$–7. Arrows indicate the asymptotic ratios for photoabsorption (Ph) and hydrogenic shake-off (SO). The dotted lines indicate $\Delta\epsilon_{\max}$ given by Eq. (6).

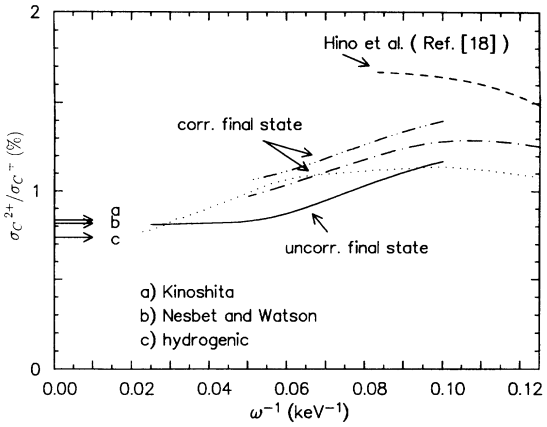

FIG. 3. Ratios of $R_C=\sigma_C^{2+}/\sigma_C^{+}$ of double to single ionization by Compton scattering as a function of the ionized photon energy. ——, uncorrelated final state, Eq. (3); – · –, correlated final state [Eq. (4)]; – · · –, correlated final state [Eq. (6)]; - - -, Hino, Bergstrom, and Macek [18]; · · ·, previous estimate [14]. Asymptotic ratios [Eq. (4)] with initial state, (a) Kinoshita [24], (b) Nesbet and Watson [25], and (c) hydrogenic.

(0.08%), while the weighted average for all dipole-forbidden processes lies in the vicinity of the hydrogenic shake value [13]

$$\langle R_C^+(5s)\rangle_{\text{shake}}=\left|\frac{\langle\phi_{5s}(Z=2)|\phi_{1s}(Z=1.68)\rangle}{\langle\phi_{1s}(Z=2)|\phi_{1s}(Z=1.68)\rangle}\right|^2=0.03\%. \tag{9}$$

These trends observed for several excitation-ionization processes ($n=2$ to 6) provided the justification for the approximation underlying our first calculation for double ionization by Compton scattering [14].

The ratio of double to single ionization by Compton scattering R_C as given by the present calculation together with the previous estimate for large energies is given in Fig. 3. The previous estimate is found to be close to the present three calculations using the approximations [Eqs. (3), (4), and (6)] as final states. The variation of R_C for different choices of final states gives an indication for the error introduced by the approximation of the final state at photon energies where the Compton electron is still comparably slow ($\lesssim 1$ keV). All calculations tend monotonically to a value $R_C\simeq 0.8$ as $1/E\to 0$, or equivalently $E\to\infty$, in agreement with the asymptotic limit discussed below. It should be noted that this limit refers to the nonrelativistic limit $E\to\infty$ whose physical significance should be viewed with caution. Nevertheless, the asymptotic value appears to be reached for $E\gtrsim 25$ keV, where the neglect of relativistic effects may not yet constitute a serious problem. We also show in Fig. 3 the recent calculation by Hino, Bergstrom, and Macek [18] for double ionization by Compton scattering employing a many-body perturbation theory approximation. Their values lie within the energy range shown systematically higher and, more importantly, according to the authors appear to converge to the asymptotic value $R_C(E\to\infty)=1.67\%$ for photoabsorption, in disagreement with our calculation as well as with a very recent calculation by Suric *et al.* [28] employing the impulse approximation. The latter calculation approaches this asymptotic value with increasing energy, however, from below rather than from above.

The nonrelativistic high-energy limit can be found with the help of a straightforward extension of the Kabir-Salpeter analysis [29] for photoabsorption to Compton scattering. The high-energy limit for simultaneous excitation and ionization to the state n (bound and continuum) is given for photoabsorption by

$$\sigma_{\text{ph}}^+(n)\propto\left|\int d^3r_2\phi_n^*(\mathbf{r}_2)\psi_i(0,\mathbf{r}_2)\right|^2. \tag{10}$$

Note that Eq. (10) explicitly incorporates the fact that photoabsorption probes the initial-state wave function ψ_i at the origin $r_1=0$ of the coordinate of the fast electron while projecting the exact initial-state wave function onto the final state of the "shaken up" electron. This picture is also consistent with the remarkable observation [30] and the theoretical prediction [31] that the electron spectra resulting from photoionization of Kr at 17 keV and from nuclear internal conversion closely mimic each other. Compton scattering, on the other hand, is a two-body scattering process between the photon and the electron which does not rely on the close proximity or recoil of the nucleus. The entire radial distribution of the Compton scattered ("fast") electron contributes. Accordingly, we remove the constraint $r_1=0$ and, instead, average [Eq. (10)] over all coordinates $\mathbf{r}_1$ of the Compton scattered electron to arrive at

$$\sigma_C^+(n)\propto\int d^3r_1\left|\int d^3\mathbf{r}_2\phi_n^*(\mathbf{r}_2)\psi_i(\mathbf{r}_1,\mathbf{r}_2)\right|^2. \tag{11}$$

We note that (11) can be rigorously derived from Eq. (1) using the limit $\omega\to\infty$, $Q\to\infty$. A derivation of Eq. (11) using a somewhat different approach has been independently given by Suric *et al.* [28]. The present argument intends to empha-

size the underlying physical picture. Following now the similar analysis for photoabsorption [1,2,13,14], we have

$$R_C = \frac{\sum\!\!\!\!\!\!\int \sigma_C^+(n) - \sum_n \sigma_C^+(n)}{\sum_n \sigma_C^+(n)}, \tag{12}$$

where $\sum\!\!\!\!\!\!\int$ stands for the sum over both bound and continuum states while Σ_n includes only bound states. Using the closure relation

$$\sum\!\!\!\!\!\!\int \phi_n(\mathbf{r})\phi_n^*(\mathbf{r}') = \delta(\mathbf{r}-\mathbf{r}'), \tag{13}$$

we arrive at

$$R_C(E\to\infty) = \frac{1-\sum_n \sigma_C^+(n)}{\sum_n \sigma_C^+(n)}. \tag{14}$$

This expression contains only the accurately known ground-state wave function for helium and hydrogenic states of the final state and can be evaluated to high accuracy. We find the values $R_C = 0.815\%$ using the CI wave function by Nesbet and Watson [25] and for the more accurate wave function of Kinoshita $R_C = 0.835\%$ [24], also shown in Fig. 3. Using the hydrogenic wave function with $Z = 1.688$ leads instead to the "shake" value $R_C = 0.73\%$ used previously [14]. We note that R_C displays some variation if less accurate initial-state wave functions are employed.

In conclusion, our numerical and analytical analysis indicates that the nonrelativistic ratio of double to single ionization by Compton scattering decreases monotonically for $E \gtrsim 10$ keV to an asymptotic value of 0.835%. Experiments in the region of photon energies $\gtrsim 25$ keV should yield values in the proximity of this asymptotic limit. Our result agrees with our previous estimate [14] and the very recent calculation by Suric *et al.* [28] but disagrees with calculations by Hino, Bergstrom, and Macek [18].

We gratefully acknowledge discussions with P. Bergstrom, C. Greene, K. Hino, R. Pratt, and T. Suric. This work is supported in part by the National Science Foundation and by the U.S. Department of Energy, Office of Basic Energy Sciences, Division of Chemical Sciences, under Contract No. DE-AC05-84OR21400 with Martin Marietta Energy Systems, Inc., and by the Swedish National Science Research Council (NFR).

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