Ionization of Two-Electron Systems by Compton Scattering of a Photon

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Photoionization of two-electron systems is studied. The nonrelativistic impulse approximation is applied to treat the Compton ionization contributions. For asymptotically high energies this treatment formally reduces to the generalized shake theory and predicts a limit for the ratio of double to single ionization different from that for photoeffect. At finite energies the ratio for He, which includes both Compton and photoeffect contributions, is marginally in agreement with experimental results but it disagrees with previous theoretical calculations.

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In double ionization (γ, $2e$) of two-electron systems by a single photon the coupling between the electrons and the radiation field is described by a one-body operator. Hence the simultaneous ejection of two electrons is due to the electron-electron interaction and can be used to investigate electron-electron correlations. Double photoionization has been studied extensively for several decades and, until recently, it was reasonably assumed to be due to the photoabsorption mechanism (due to the relatively low photon energies available in typical experimental systems [1,2]). However, with the use of synchrotron facilities these investigations have recently been extended to higher photon energies, so far as high as 12 keV [3,4]. It has long been known that the relative contribution of Compton processes in the photon-atom interaction increases with energy [5], and it has recently been pointed out that realistic theoretical analysis of double ionization has to include the Compton contribution at such high photon energies [6]. Specifically, the single ionization of helium is dominated by Compton scattering at photon energies above 6 keV. It should also be noted that until recently experimental techniques did not discriminate between the photoelectric and Compton contributions and therefore, at least in principle, both effects should be considered at even low energies if high accuracy is to be attained. However, new techniques are being developed which distinguish between these mechanisms, and these have already been applied to study single ionization [7].

Andersson and Burgdörfer [8] estimated the Compton contribution to double ionization by convoluting the single ionization cross section, as a function of energy ω lost by the scattered photon, with the ratio $R(\omega) = \sigma^{++}/\sigma^{+}$. For $R(\omega)$ they used photoeffect data for final states in the P sector, and for higher angular momenta they used the value 0.73%. More recently, Hino, Bergstrom, and Macek [9] have applied lowest order many-body perturbation theory to the "A^2" term of the Compton scattering contribution, obtaining results which disagreed with the estimate of Andersson and Burgdörfer. Both methods gave results marginally consistent with experiment. Results of Hino, Bergstrom, and Macek are consistent with suggestions [10] that, in the high energy limit, the ratio between double ionization and single ionization cross sections should be the same for photoeffect and Compton scattering.

In this work we calculate the Compton effect contribution to double photoionization. Our results are based on the well-established impulse approximation (IA) often used in single Compton scattering at these high energies [11], together with the use of highly correlated ground state wave functions. The IA for two electron ejection by Compton scattering derived here is further analyzed for the case of infinite incident photon energy, where a relatively simple expression for the ratio of double to total ionization is obtained. This result may also be derived using the generalized shake theory of Åberg [12].

We start with the expression for the cross section for the process in which two electrons are ejected from a two-electron system by the scattering of a single photon. In the nonrelativistic approximation the cross section may be obtained, at the high photon energies, by dropping the $\mathbf{p}\cdot\mathbf{A}$ term and simply taking the lowest order matrix element of the A^2 interaction term, which gives

$$\frac{d^2\sigma_C^{++}}{d\omega_f d\Omega_f} = \left(\frac{d\sigma}{d\Omega_f}\right)_0 \left(\frac{\omega_f}{\omega_i}\right) \sum_f |\langle f|\, e^{i\mathbf{k}\cdot\mathbf{r}_1} + e^{i\mathbf{k}\cdot\mathbf{r}_2}\, |i\rangle|^2\, \delta(E_f - E_i - \omega). \quad (1)$$

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where $(d\sigma/d\Omega_f)_0$ is the Thomson cross section and $\omega = \omega_i - \omega_f$, $\mathbf{k} = \mathbf{k}_i - \mathbf{k}_f$ are, respectively, the energy and momentum transfers from the scattered photon to outgoing particles. The states $|i\rangle$ and $|f\rangle$ are the wave functions for the initial (bound) and final (continuum) states of two electrons, with energies E_i and E_f. These states are eigenstates of the Hamiltonian

$$H = -\frac{1}{2m}\nabla_1^2 - \frac{1}{2m}\nabla_1^2 - \frac{Ze^2}{r_1} - \frac{Ze^2}{r_2} + \frac{e^2}{r_{12}}, \qquad (2)$$

where Z represents the charge of the atomic nucleus and $r_{12} = |\mathbf{r}_1 - \mathbf{r}_2|$, $\mathbf{r}_1, \mathbf{r}_2$ being the electron coordinates.

Applying the method of Eisenberger and Platzman [13], we may derive the doubly differential cross section in the IA for double Compton ionization, already known to be suitable for single ionization in the regime where photon momentum transfer is much larger than the expectation value of the bound electron momentum. The details of the derivation will be presented elsewhere. We obtain

$$\frac{d^2\sigma_C^{++}}{d\omega_f d\Omega_f} = \left(\frac{d\sigma}{d\Omega_f}\right)_0 \left(\frac{\omega_f}{\omega_i}\right) S^{++}(\omega, \mathbf{k}), \qquad (3)$$

where

$$S^{++}(\omega,\mathbf{k}) = \int d^3p\, \delta\left(\omega - \frac{k^2}{2m} - \frac{\mathbf{k}\cdot\mathbf{p}}{m}\right) \left[\int |\Psi_i(\mathbf{p},\mathbf{q}\,)|^2 d^3q - \sum_B \left|\int \Phi_B^*(\mathbf{q}\,)\Psi_i(\mathbf{p},\mathbf{q}\,)d^3q\right|^2\right]. \qquad (4)$$

In Eq. (4) $\Psi_i(\mathbf{p},\mathbf{q})$ is the initial two-electron wave function which is symmetric in momentum space, $\Phi_B(\mathbf{q}\,)$ is a single-electron bound wave function in momentum space (hydrogenlike in the potential of the charge Z), and the summation goes over all bound states. These wave functions are normalized to unity and can be obtained as Fourier transforms of the corresponding wave functions in coordinate space.

In deriving Eq. (4), in addition to assuming IA, we have also neglected bound-electron–continuum-electron correlations which appear in the second term of Eq. (4). Initial state correlation is explicitly included in Eq. (4), while continuum-electron–continuum-electron correlation is implicitly included at the level of IA. With increasing incoming photon energy these assumptions become more justified.

It should be noted that the first term of Eq. (4) represents the Compton profile of the ground state of the two-electron system. It accounts for the total ionization Compton cross section, of which the dominant contribution is single-electron ionization. Double ionization makes around a 1% contribution (as verified later) to this term. The second term represents single-electron ionization by Compton scattering with the other electron remaining bound.

Equation (3) along with Eq. (4) exhibits two important features. First, if the initial wave function represents noninteracting electrons in the field of charge Z, the cross section for double ionization is zero. This illustrates the necessity of the electron-electron interaction in the initial state. We note that recent numerical analysis [8] has shown that the ratio of the cross section for double photoabsorption to that for single photoabsorption is independent of the final state correlation at high photon energies, as had been discussed by Dalgarno and Sadeghpour [14].

The second feature is that, when we can neglect the average bound electron momentum in comparison with the momentum transfer $\mathbf{k}$, as we can for high energy photons, the total cross section takes the form of that for the "shake off" process, accompanying the ejection of the first electron by Compton scattering. In the limit of infinite incoming photon energy the free electron Compton regime is approached. We may neglect the term $\mathbf{k}\cdot\mathbf{p}/m$ in the δ function of Eq. (4), which is responsible for Doppler broadening of the scattered photon energy [13]. After integration of Eq. (3) we obtain the simple formula for the ratio of double to total ionization cross section

$$R_C = 1 - \sum_B \int d^3r \left|\int \Phi_B^*(\mathbf{r}_1)\Psi_i(\mathbf{r},\mathbf{r}_1)d^3r_1\right|^2. \qquad (5)$$

In this equation the eigenfunctions $\Psi_i(\mathbf{r},\mathbf{r}_1)$ and $\Phi_B(\mathbf{r})$ are the same as in Eq. (4) except they are now represented in coordinate space. Each term in the sum of Eq. (5) is the probability for ionization of one electron while the other remains in the bound state B. It is interesting to note that exactly the same expression as Eq. (5) can be obtained from Åberg's generalized shake probability [12] when applied to Compton scattering; the derivation will be presented elsewhere.

Equation (5) is very similar to that of Dalgarno and Sadeghpour [14] for photoeffect, which can be written as

$$R_{\mathrm{PE}} = 1 - \frac{\sum_B \left|\int \Phi_B^*(\mathbf{r}_1)\Psi_i(\mathbf{r}_1,0)d^3r_1\right|^2}{\int |\Psi_i(\mathbf{r}_1,0)|^2\, d^3r_1}. \qquad (6)$$

Formally one can get Eq. (6) putting the function $\delta(\mathbf{r})/\int |\Psi_i(\mathbf{r}_1,0)|^2\, d^3r_1$ under the first integral of Eq. (5). This demonstrates that different regions of the initial two-electron state contribute to these two processes. Namely, when the incoming photon energy goes to infinity photoeffect will be determined exclusively by the region where one of the electrons is at the nucleus. This is due to the fact that photoeffect can occur only on bound electrons. By contrast, the Compton process can occur on free electrons, and all regions contribute in proportion to the probability amplitude that an electron can be found there.

It is easy to verify that if the initial wave function can be factorized, i.e., if $\Psi_i(\mathbf{r},\mathbf{r}_1) = \psi_i(\mathbf{r})\psi_i(\mathbf{r}_1)$, Eq. (5) for Compton scattering and Eq. (6) for photoeffect become the same. For example, if the one-parameter ground state wave function of He with screened charge of $Z^* = 27/16$ is used, the ratio obtained using Eq. (5) is 0.72%, as it is using Eq. (6) [14].

In Table I we examine the partition of the Compton process for H^- and He into ionization, ionization excitation, and double ionization channels, using the high energy limit formula [Eq. (5)]. We use the 20-parameter Hylleraas-type wave function of Ref. [15]. We present excitation probabilities only for S and P states, as contributions of all higher orbital momenta are less than about 0.003% for He and 0.03% for H^-. However, these higher states are included in our calculations. For the double to total ionization ratio we obtain 0.695% for the negative hydrogen ion and 0.797% for helium. If this table is compared to the similar table of Dalgarno and Sadeghpour [14] we can notice that, unlike for photoeffect, in Compton scattering $l \neq 0$ excitation channels also contribute, although S state excitations are still dominant. We can see, for example, that in the case of H^-, ionization with excitation to the $2P$ state (with relative probability of 0.75%) is more likely then double ionization (0.69%). The asymptotic values obtained using the Byron and Joachain [16] 45-parameter and Hylleraas-type [17] 6-parameter helium ground state wave functions were very similar to those shown in Table I, being 0.802% and 0.812%, respectively.

In Fig. 1 we show our estimates of the ratios $R_C = \sigma_C^{++}/\sigma_C^{\rm total}$ and $R = (\sigma_{\rm PE}^{++} + \sigma_C^{++})/(\sigma_{\rm PE}^{+} + \sigma_C^{\rm total})$ for helium as a function of photon energy. The values for σ_C^{++} were obtained using the Byron and Joachain [16] 45-parameter helium ground state wave function in Eqs. (3) and (4). The values for $\sigma_C^{\rm total}$ were obtained employing only the first term of Eq. (4). For $\sigma_{\rm PE}^{++}$ and $\sigma_{\rm PE}^{+}$ we have used the photoeffect calculation of Hino *et al.* [18]. The asymptotic value of the ratio R_C, obtained using Eq. (5), is represented by the solid horizontal line. Our results show that a value within 10% of the asymptotic value was reached at about 17 keV. Our results reached the asymptotic value within 4% at about 30 keV and within 1.6% at about 50 keV. This means that high energies were required to approach the limit, as may be expected from electron energy considerations [6]. Namely, at these energies the ejected electrons do not have high enough energy to fully apply the shake mechanism, which would guarantee constant value.

In Fig. 1 we also compare our predictions for the ratio R with the experimental results of Levin *et al.* [2], Levin *et al.* [3], and Bartlett *et al.* [4]. The estimates of Andersson and Burgdörfer and of Hino, Bergstrom, and Macek are also shown. In the former work σ_C^{++} was not calculated directly but was derived convoluting the single ionization cross section with the photoeffect ratio $R_{\rm PE}$ as a function of photon energy transfer. Hino, Bergstrom, and Macek applied the lowest order many-body perturbation theory along with the A^2 approximation. (There are some concerns with this approximation at these energies, for which outgoing electrons have energies smaller than 1 keV. For photoeffect in this electron energy region and below, the lowest order approximation was not sufficient [18].)

All the predictions for R agree somewhat with experiment, even though they differ substantially from each other in the region where the Compton contribution is significant. Andersson and Burgdörfer's results seem to be further decreasing for higher energies. Very recent calculations of Burgdörfer, Andersson, McGuire, and Ishihara [19] for even higher energies, but for the momentum-

TABLE I. The relative percentages of ionization, ionization excitation, and double ionization accompanying Compton scattering, for the negative hydrogen ion and for helium at the high energy limit, obtained using Eq. (5). Here n represents principal and l orbital quantum number of the second electron when remaining bound. All numbers are in %. R_C represents the double ionization contribution.

	H^-		He	
n	$l=0$	$l=1$	$l=0$	$l=1$
1	79.682		96.005	
2	18.534	0.7536	2.4949	0.1567
3	0.0649	0.0881	0.2806	0.0303
4	0.0296	0.0321	0.0900	0.0114
5	0.0154	0.0152	0.0409	0.0055
6	0.0089	0.0084	0.0222	0.0031
≥ 7	0.0227	0.0204	0.0523	0.0076
R_C(%)	0.695		0.797	

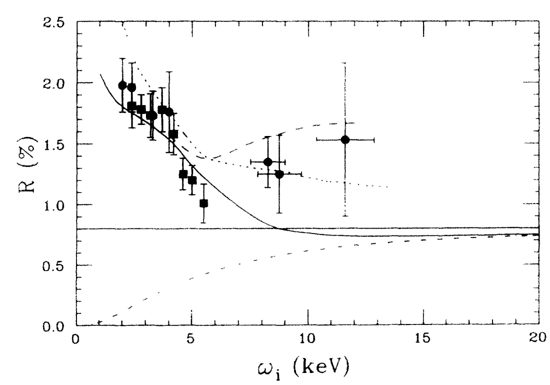

FIG. 1. Ratio of the double to single ionizaton cross section of helium as a function of incoming photon energy. The solid curve represents results for R obtained using our Compton data with photoeffect data of Hino *et al.* [18]. The horizontal solid line represents the asymptotic value of the ratio. The ratio of Compton ionization contributions is also shown (dot-dashed line). The dotted curve is the result of Andersson and Burgdörfer [8], and the dashed curve is the result of Hino, Bergstrom, and Macek [9]. The circles represent the experimental results of Refs. [2,3] and squares represent experimental results of Ref. [4].

differential ratio, suggest that the integrated ratio R would decrease toward a high energy limit similar to ours.

In conclusion, we have treated double ionization by Compton scattering using well-established nonrelativistic IA. We present expressions which can be used for any two-electron system in the ground state. We have demonstrated that in the high energy limit, where Compton scattering dominates over photoeffect, the ratio of double to single cross section approaches the limit which can also be predicted by generalized shake theory. However, the numerical value for this ratio is not the same as that obtained for photoeffect, due to differences in the regions of the initial two-electron state which contribute to these processes. In our calculations for helium we observed that high energies were required to approach that limit. In such an energy region relativistic effects should be considered. Clearly Compton contributions are important in any realistic theoretical analysis of photoionization and are particularly important at higher photon energies. Such calculations, along with more accurate experimental measurements, should lead to an important test of the strength of the electron-electron correlation for the ground state wave function of the two-electron system.

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