# Two-electron ejection from helium by Compton scattering

P. M. Bergstrom, Jr., Ken-ichi Hino,* and Joseph H. Macek
*Department of Physics and Astronomy, University of Tennessee, Knoxville, Tennessee 37996*
*and Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831*
(Received 26 August 1994; revised manuscript received 18 November 1994)

Double ionization of the ground state of helium by Compton scattering is investigated using many-body perturbation theory. Representative calculations of cross sections differential in the scattered photon energy and angle are given to illustrate the nature of the process. Total cross sections for single ionization and double ionization of helium and the ratio of these cross sections are presented for energies from 2.5 to 20 keV. Current experimental data for the ratio due to both Compton scattering and photoionization are in reasonable agreement with the corresponding calculations presented here. However, comparisons of the present results with other calculations reveal substantial discrepancies. Possible reasons for these differences at finite energies are proposed. Our results for double ionization by Compton scattering have not reached an asymptotic limit.

PACS number(s): 32.80.Cy, 32.80.Fb, 31.15.−p

## I. INTRODUCTION

One area in the study of atomic structure that has been of interest for many years is the investigation of the effects of the interactions (or correlations) between electrons. Perhaps no single correlated system has been the subject of as much investigation as the helium atom. Several types of projectiles have been used to study this system. Charged particles suffer from the fact that they interact with all of the electrons in the atom. Photons are superior probes in this respect because one photon interacts with mainly one electron. In principle, Compton scattering of photons from bound electrons can serve as a valuable probe of atomic structure [1]. The main reason for this is that the measured cross sections are approximately proportional to the momentum distribution of the scattering charge.

Coupling this relationship to the availability of high photon fluxes from synchrotron sources at energies where it is expected to hold seems to make inevitable the investigation of correlations in helium by Compton scattering. Such investigations have recently been reported [2]. However, the motivation and chronology leading to these experiments differ entirely from the reasons just discussed.

In fact, the use of synchrotron sources in photon interactions with helium has generally been motivated by the desire to understand correlation effects in the photoeffect, a problem that has been investigated on and off, using other tools, for nearly 30 years [3]. The new synchrotron based investigations have made possible observation of the ejected electrons [4] and precision measurements of the double to single ionization ratio [5–7] at energies near the photoeffect threshold. The extension of these studies to much higher photon energies, where photoeffect calculations predict the double to single ionization ratio to be a constant (approximately 1.7%), seemed to be a logical new direction.

Experiments measuring this ratio in the several keV range have been complicated by the fact that such investigations do not distinguish between the photoeffect and Compton scattering. Compton scattering is the dominant ionizing process at high photon energies. Recently, Samson, Greene, and Bartlett [8] noted that the agreement at the highest energies between experimental data and theory was curious because the theoretical predictions were for the photoeffect and the experiments were mainly measuring Compton scattering. A contemporaneous effort to estimate this scattering contribution to the double to single ionization ratio, at the highest energies measured by experiments, found substantially lower values than those predicted for the photoeffect, but still marginal agreement with experiment [9]. The agreement of this estimate with the measured result is not particularly surprising because experimental uncertainties are rather large and because the estimate was based on photoeffect data, also in agreement with experiment.

While such estimates of the Compton contribution represent a useful start in furthering the understanding of the experimental measurements of the double to single ionization ratio, a more systematic study of double ionization by Compton scattering is desirable for the following reasons. A first-principles treatment of double ionization by Compton scattering would dispense with the *ad hoc* assumptions of prior estimates. More importantly, a more complete treatment of correlations in Compton scattering could be used as a starting point for investigations that sensitively probe the effects of these interactions on the momentum distribution of bound electrons. Although only rigorous within the impulse approximation [10] to single ionization by Compton scattering, the relationship between the measured cross section and this

*On leave from Department of Applied Physics and Chemistry, University of Electro-Communications, Chofu, Tokyo 182, Japan.

1050-2947/95/51(4)/3044(9)/$06.00  

distribution (or Compton profile [11]) should be a valuable interpretive tool. A calculation that goes beyond the impulse approximation should provide guidance on the applicability of the impulse approximation to two-electron processes and on the importance of the inclusion of final state correlations.

We recently evaluated the nonrelativistic $A^2$ matrix element for the double ionization of helium by Compton scattering, using many-body perturbation theory, for incident photon energies from 4 to 12 keV [12]. These results were in agreement with the available experimental data. Our purpose here is to present the framework used in these calculations, to extend these results over a broader range of incident photon energies, and to more thoroughly investigate this process. In the next section we present the formalism used in our calculations and discuss the validity of our approach. In Sec. III we give additional numerical results, extending our calculation to higher as well as to lower energies. We examine in more detail structure observed in our previous results. Subsequent to our earlier work, several other authors have calculated double ionization by Compton scattering within the $A^2$ approximation, obtaining results substantially different from ours [13,14]. We discuss possible reasons for these discrepancies. In Sec. IV we present our conclusions.

## II. FORMALISM

An exact calculation of ionization by Compton scattering should start with the relativistic $S$ matrix element for the process. Evaluation of this amplitude for single ionization of an atom by Compton scattering is a substantial task and has only recently been accomplished with precision [15]. The prospects of extending this formalism to the double ionization problem, particularly at the high photon energies of experimental interest, seem rather remote. For high incident and scattered photon energies, the validity of using the $A^2$ term of the nonrelativistic interaction Hamiltonian [16,17] and approximations to it [10,11] have long been assumed. A recent systematic study, using the $S$ matrix code [18], confirmed the validity of these approximate methods used to calculate single ionization by Compton scattering for high photon energies.

Our treatment of the ejection of both electrons bound in helium by Compton scattering starts from the nonrelativistic $A^2$ amplitude. The cross section doubly differential in scattered photon energy and angle is then written [19]

$$\frac{d^2\sigma^{2+}_{\rm CS}}{d\omega_f d\Omega_f} = \left(\frac{d\sigma}{d\Omega_f}\right)_{\rm Th}\left(\frac{\omega_f}{\omega_i}\right) \times \int d\mathbf{p}_1 d\mathbf{p}_2 \left|\left\langle \Psi_F \left| \sum_{j=1,2} e^{i\mathbf{k}\cdot\mathbf{r}_j} \right| \Psi_I \right\rangle\right|^2 \times \delta(E_F - E_I - \omega), \qquad (1)$$

where $\left(\frac{d\sigma}{d\Omega_f}\right)_{\rm Th}$ is the classical (or Thomson) scattering cross section with $\Omega_f$ being the solid angle of the scattered photon. $\Psi_{I(F)}$ and $E_{I(F)}$ represent the many-body wave functions and energies of the initial (final) atomic states. $\omega_{i(f)}$ are the energies of the incident (scattered) photons and $\mathbf{p}_1$ and $\mathbf{p}_2$ are the momenta of the ejected electrons. $\mathbf{k}$ $(\omega)$ is the momentum (energy) transferred to the atom during the process. In Eq. (1), the energies of the two ejected electrons $\epsilon_{p_1}$ and $\epsilon_{p_2}$ satisfy energy conservation

$$\epsilon_{p_1} + \epsilon_{p_2} = \omega + E_I.$$

As discussed above, choosing Eq. (1) as a starting point represents a considerable simplification of the task from that of a fully relativistic or $S$ matrix calculation. One may ask what has been neglected in using this approximation. Clearly, the approach neglects relativistic effects in the initial and final states. These effects should be fairly small in the helium atom. Additionally, contributions corresponding to a second-order evaluation of the $\mathbf{p}\cdot\mathbf{A}$ terms of the nonrelativistic interaction Hamiltonian are not included. These terms diverge at zero scattered photon energies in all subshells (the infrared divergence) and at scattered photon energies corresponding to fluorescence lines in outer shell cross sections. The latter divergences do not occur in scattering from the ground state of helium as all electrons are in the $K$ shell (such divergences are readily removed in any case by including the width of the state). The resolution of the infrared divergence problem in this context has been discussed elsewhere [18], where it was verified that accurate total photon-atom interaction cross sections may be obtained by adding photoeffect calculations and the $A^2$ approximation to the Compton scattering cross section.

At the high photon energies of interest here the cross section for single ionization of helium by Compton scattering is adequately described by this nonrelativistic $A^2$ approximation. In Fig. 1 the expected agreement of the $A^2$ approximation with the results obtained using the $S$ matrix code of Surić *et al.* [15] for single ionization by

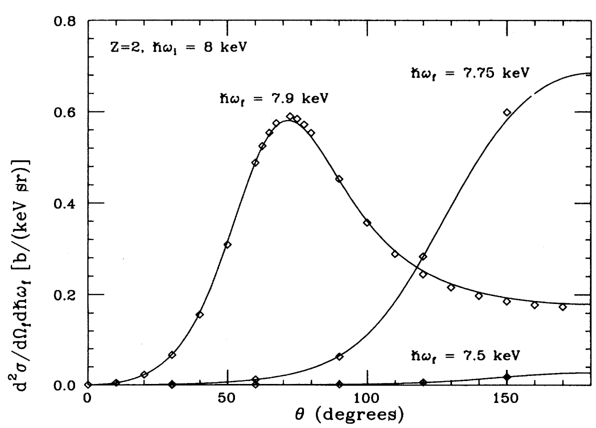


FIG. 1. Results obtained within the nonrelativistic $A^2$ approximation (solid curve) [20] and from the relativistic $S$ matrix (diamonds) [15,18] for the cross section doubly differential in scattered photon energy and angle for single ionization of helium by Compton scattering of 8-keV photons.

Compton scattering is demonstrated at photon energies typical of those considered in this report. Figure 1 also demonstrates the low energies available to the electrons ejected by Compton scattering. For 8-keV incident photons the energy transferred to the atom, equal to the difference between the incident and scattered photon energies, is likely to be less than several hundred eV.

Ideally one would evaluate Eq. (1) using exact wave functions for the correlated initial and final atomic states. While such wave functions may be obtained using the variational method for the ground state and for some excited states of helium [21], no such wave functions are known for the continuum three-body final state for arbitrary energy. One possible approach is to use the fully correlated ground state wave functions with an approximate wave function for the final state. Attempts to evaluate the double photoionization cross section using a correlated helium initial state and uncorrelated or approximate correlated final states have been successful in predicting the ratio of double to single ionization (using the velocity and acceleration form of the dipole operator) in the asymptotic region. However, this method has also produced results that vary widely, depending on the form of the dipole operator, at low ejected electron energies [22,23] such as those available in the present study.

Here we include the electron-electron interaction in the initial and final states consistently, using many-body perturbation theory (MBPT). We choose to apply this framework to the current problem of the double ionization of helium by Compton scattering for the following reasons. (i) We have demonstrated in Fig. 1 that, for the case of single ionization, the scattered photon carries off most of the energy of the incident photon and that the ejected electron is likely to have little kinetic energy. We expect this observation to hold as well for the case of double ionization (i.e., the final state energy shared between the two ejected electrons should be small relative to the incident photon energy). Accordingly one must take into account the wave function of the doubly ionized state in the low energy regime. In studies of the double photoionization of helium, calculations performed using the MBPT method agreed with experimental results over a wide range of energies [24,25]. (ii) It is known that the cross section for double photoionization within MBPT formalism remains almost invariant under the change of the form of dipole operator [25]. This invariance is a necessary (although not sufficient) criterion for selecting a valid theoretical framework. We are unaware of any other theoretical framework for double photoionization which preserves this invariance (even approximately). We subsequently show in detail that the "generalized" length $L$ and velocity $V$ forms of the $A^2$ Compton scattering amplitude for double ionization are identical in MBPT. (iii) Since the basis functions of MBPT are constructed as products of a radial wave function and an associated angular function, the scattering amplitude can be cast in the form of a partial wave expansion. This is quite convenient in numerical calculations. These facts indicate that the MBPT might be able to be successfully extended to double ionization by Compton scattering.

The present theoretical framework is similar to that used by Hino *et al.* in their study of the double photoionization of helium [25]. The transition matrix element of Eq. (1) is expanded in powers of the electron-electron correlation interaction $v$. The basis set $\{\phi_n\}$ of the expansion with orbital energies $\epsilon_n$ is constructed using the $V^{N-1}$ potential defined in Eq. (9) of Ref. [25].

The four diagrams contributing in lowest order MBPT to double ionization by Compton scattering are shown in Fig. 2. In these diagrams it is understood that the hole-hole interactions are incorporated to all orders to give the ground state energy of helium

$$E_I = 2\epsilon_{1s} - \langle \phi_{1s}\phi_{1s}|v|\phi_{1s}\phi_{1s}\rangle$$

rather than the sum of the $1s$ orbital energies. While one may not attach physical meaning to the individual diagrams, they have acquired names that bring to mind different physical mechanisms. The diagram labeled TS1 represents the so-called two-step-1 amplitude. This amplitude and the ground state correlation mediated by a hole propagation (GSCh) amplitude result from expanding the final state wave function in the electron-electron interaction. The amplitudes labeled SO (shake-off) and GSCp (ground state correlation mediated by a particle propagation) [26] are associated with the initial state wave function. The meaning of these diagrams as initial state or as final state correlations is fixed. However, the actual contributions of the amplitudes are dependent on the form of the interaction operator.

The transition amplitudes for these four diagrams are expressed as

$$A^{\rm SO} = -\frac{\langle \phi_{\mathbf{p}_2}\phi_{1s}|v|\phi_{1s}\phi_{1s}\rangle\langle\phi_{\mathbf{p}_1}|V_{\rm ext}|\phi_{1s}\rangle}{\epsilon_{1s}-\epsilon_{\mathbf{p}_1}+\omega}, \qquad (2)$$

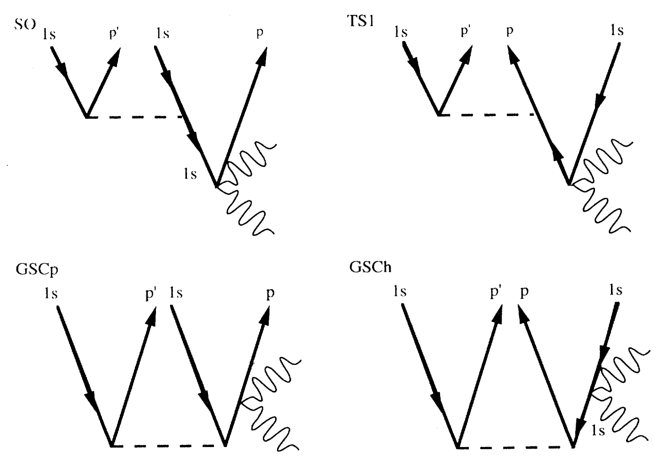


FIG. 2. Lowest-order MBPT diagrams for double ionization of helium by Compton scattering using the nonrelativistic $A^2$ approximation. The electron-electron interaction is shown by a dashed line. It is understood that the hole-hole interactions are incorporated to all orders to give the correct Hartree-Fock energy for the ground state of helium. Exchange diagrams are also included in the present calculations. The individual amplitudes are called shake-off (SO), two-step-1 (TS1), ground state correlation particle (GSCp) and ground state correlation hole (GSCh).

$$A^{\mathrm{GSCp}} = \sum_{\rho \neq 1s} \frac{\langle \phi_{\mathbf{p}_1} | V_{\mathrm{ext}} | \phi_\rho \rangle \langle \phi_\rho \phi_{\mathbf{p}_2} | v | \phi_{1s} \phi_{1s} \rangle}{E_I - \epsilon_\rho + \epsilon_{\mathbf{p}_2}}, \qquad (3)$$

$$A^{\mathrm{GSCh}} = -\frac{\langle \phi_{1s} | V_{\mathrm{ext}} | \phi_{1s} \rangle \langle \phi_{\mathbf{p}_1} \phi_{\mathbf{p}_2} | v | \phi_{1s} \phi_{1s} \rangle}{E_I - \epsilon_{\mathbf{p}_1} - \epsilon_{\mathbf{p}_2}}, \qquad (4)$$

$$A^{\mathrm{TS1}} = \sum_{\rho \neq 1s} \frac{\langle \phi_{\mathbf{p}_1} \phi_{\mathbf{p}_2} | v | \phi_\rho \phi_{1s} \rangle \langle \phi_\rho | V_{\mathrm{ext}} | \phi_{1s} \rangle}{\epsilon_{1s} - \epsilon_\rho + \omega + i\eta}. \qquad (5)$$

The one-body operator $V_{\mathrm{ext}}$ representing the $A^2$ interaction of the applied electromagnetic field with an electron is

$$V_{\mathrm{ext}} = e^{i\mathbf{k}\cdot\mathbf{r}}. \qquad (6)$$

It is worthwhile to investigate the dependence of the transition matrix element on the form of the interaction operator in Compton scattering. To this end we first define an alternative interaction operator $V'_{\mathrm{ext}}$ [27] by

$$V'_{\mathrm{ext}} = [V_{\mathrm{ext}}, H_0]\,, \qquad (7)$$

where $H_0$ is the Hamiltonian of the target atom. If the wave functions $\psi_I$ and $\psi_F$ are exact eigenfunctions of $H_0$ (and assuming energy conservation), the following relationship holds:

$$\langle \psi_F | V'_{\mathrm{ext}} | \psi_I \rangle = -\omega \langle \psi_F | V_{\mathrm{ext}} | \psi_I \rangle. \qquad (8)$$

The squared magnitude of the matrix element $\langle \psi_F | V_{\mathrm{ext}} | \psi_I \rangle$ is proportional to the generalized oscillator strength (GOS). One may also write

$$\langle \psi_F | V'_{\mathrm{ext}} | \psi_I \rangle = \frac{i\mathbf{k}}{2} \cdot \int d\mathbf{r} e^{i\mathbf{k}\cdot\mathbf{r}} \left[ \psi_F^* \left( \nabla \psi_I \right) - \left( \nabla \psi_F^* \right) \psi_I \right]. \qquad (9)$$

These different forms of the operator have been used by Kim and Inokuti [27] in calculations of the GOS for helium. Appealing to the behavior of these forms at very low momentum transfer (i.e., $\mathbf{k} \to \mathbf{0}$), the two expressions become the matrix elements of the length and velocity forms of the dipole operator. In this context, the matrix element of $V_{\mathrm{ext}}$ may be called the "generalized length form" and that of $V'_{\mathrm{ext}}$ may be called the "generalized velocity form" of the GOS, although not too much significance should be placed on these names. The identity of the two forms can be used as a test of the exactness of wave functions and of the accuracy of calculations.

In the calculations presented here we use the generalized length form of the $A^2$ operator exclusively. It is useful, however, to examine how the individual MBPT amplitudes vary depending on the form of this operator. As the $V^{N-1}$ potential used here is chosen to be a local potential [25], Eq. (7) can be recast as

$$V'_{\mathrm{ext}} = [V_{\mathrm{ext}}, h]\,. \qquad (10)$$

Here $h$ is the Hartree-Fock Hamiltonian. The eigenfunctions of $h$ have been adopted as our basis functions $\{\phi_n\}$. Defining

$$\Delta A^i \equiv A_L^i - \frac{A_V^i}{(-\omega)} \qquad (11)$$

as the difference between the generalized length form of each of the amplitudes $i$ given in Eqs. (2)–(5) and the corresponding generalized velocity form amplitude obtained by using Eq. (10), we obtain

$$\Delta A^{\mathrm{SO}} = -\frac{1}{\omega} \langle 1s | v_{p_2 1s} | 1s \rangle \langle p_1 | V_{\mathrm{ext}} | 1s \rangle, \qquad (12)$$

$$\Delta A^{\mathrm{GSCp}} = -\frac{1}{\omega} \langle p_1 | V_{\mathrm{ext}} v_{p_2 1s} | 1s \rangle - \Delta A^{\mathrm{SO}}, \qquad (13)$$

$$\Delta A^{\mathrm{GSCp}} = \frac{1}{\omega} \langle 1s | V_{\mathrm{ext}} | 1s \rangle \langle p_1 | v_{p,1s} | 1s \rangle, \qquad (14)$$

$$\Delta A^{\mathrm{TS1}} = -\frac{1}{\omega} \langle p_1 | V_{\mathrm{ext}} v_{p_2 1s} | 1s \rangle - \Delta A^{\mathrm{GSCh}}, \qquad (15)$$

where

$$v_{\mathbf{p}_2,1s}(r) = \int \phi_{\mathbf{p}_2}^* (r') \, v \left( |\mathbf{r} - \mathbf{r}'| \right) \phi_{1s} (r') \, d\mathbf{r}'. \qquad (16)$$

The difference between the forms of an individual amplitude is not zero. Therefore the cross sections corresponding to each amplitude depend in general on the form of the interaction operator. Provided that our lowest-order perturbation is a good approximation, the right-hand sides of Eqs. (12)–(15) should cancel when summed. In the present case they cancel exactly, as was the case for double photoionization [25]:

$$\sum_i \Delta A^i = \frac{1}{\omega} \langle \phi_{\mathbf{p}_1} | \left[ v_{\mathbf{p}_2,1s}, V_{\mathrm{ext}} \right] | \phi_{1s} \rangle = 0. \qquad (17)$$

This means that correlation must be included in the initial and final states consistently to get an invariant amplitude.

## III. RESULTS

In this section numerical results for single and double ionization of helium by Compton scattering are presented. Results are selected that help to elucidate the nature of the process, that provide accurate predictions of the cross sections for these processes in the energy range of experimental interest, and that provide ratios of double to single ionization for comparison with experiment. The calculations presented here are for incident photon energies ranging from 2.5 to 20 keV. The lower limit of this energy range has been selected for two reasons. One reason is that below this energy Compton scattering makes a negligible contribution to the total ionization cross section (compared to the photoeffect). Additionally, below this energy, the $\mathbf{p} \cdot \mathbf{A}$ terms that we have neglected become larger than the $A^2$ terms retained in our approximation. The higher end of the energy range was selected because at higher photon energies relativistic effects may have to be considered and because the calculations become increasingly cumbersome as the photon energy increases. In fact, the data for double ionization presented here at 16 and at 20 keV have been obtained using the full calculation for only the first nine partial waves of the $A^2$ operator and using the shake-off terms calculated here to estimate the convergence of the entire amplitude for higher angular momenta.

In Fig. 3, angle integrated cross sections for the double ionization of the ground state of helium by photons with energies 4, 12, and 20 keV are presented. This figure clearly demonstrates the low electron energies available to the ionized electrons (here the total energy available in the final state is shown). In the photoeffect, the ejected electrons share an amount of energy nearly equal to the incident photon energy. In Compton scattering, however, the scattered photon takes much of the energy of the incident photon. The most probable electron energies are below 100 eV with higher energies possible at higher incident photon energies. The vertical arrows represent the predictions of the energy transferred in a free Compton scattering process if the photon is backscattered. This is clearly a reasonable estimate of the maximum energy available to the ejected electrons. At zero energy transfer, the cross section for double ionization vanishes (as expected). This contrasts with the single ionization case where the cross section is finite at threshold [10].

The angular distribution of 12-keV photons inelastically scattered from helium are given in Fig. 4. Here the results for single ionization calculated within the $A^2$ and impulse approximations [11] and for double ionization (multiplied by 100 in order to be visible on this scale) are shown together with the incoherent scattering factor results of Hubbell *et al.* [28]. One effect of binding on the scattering process is that all of these curves vanish at small angles. This is well known and contrasts with the free Compton scattering (Klein-Nishina) angular distribution [29] that predicts equal probability for forward and backward scattering at low photon energies and forward peaking at higher energies [30]. The incoherent scattering factor is derived using a sum over all final states with fully correlated ground state wave functions [31]. It therefore represents a sum over all inelastic scattering processes (Raman scattering, single Compton ionization, ionization excitation by Compton scattering, and double ionization by Compton scattering) [32].

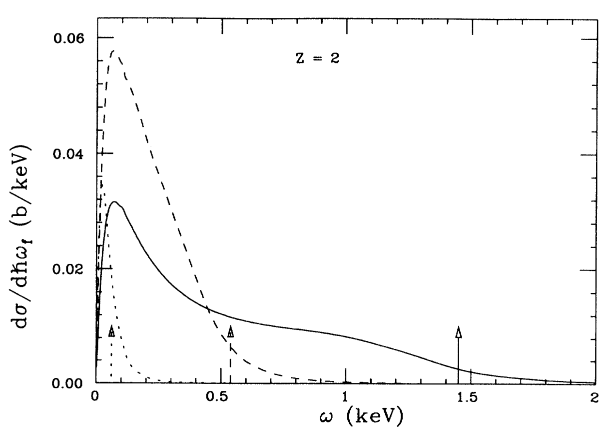


FIG. 3. Angle integrated cross sections for double ionization by Compton scattering shown as a function of the energy transfer. The results are given for incident photon energies of 4 keV (dotted curve), 12 keV (dashed curve), and 20 keV (solid curve). The arrows represent the energy transferred to a free electron in a Compton scattering event at maximum momentum transfer. These arrows are given for photon energies of (from left to right) 4, 12, and 20 keV.

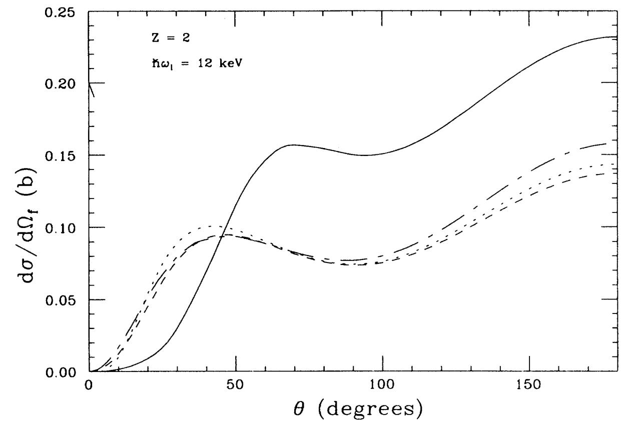


FIG. 4. Angular distribution of 12-keV photons inelastically scattered from the helium ground state. The curves are for the double ionization (solid curve, numbers are multiplied by 100) and for the single ionization (dashed curve) $A^2$ approximation calculations presented here, for single ionization within the impulse approximation (dotted curve) obtained using the Compton profiles of Biggs, Mendelsohn, and Mann [11], and for the incoherent scattering factor (chain dashed curve) [28,31].

The shape of the angular distribution of photons scattered in the double ionization process can be understood in terms of this "sum rule," i.e., roughly in terms of the differences between the incoherent scattering factor and single ionization $A^2$ approximation curves. Where there is a small difference between these curves, the amplitude for other inelastic photon scattering processes such as double ionization must be small. Where the difference is large, the amplitudes for these other processes can be large. Therefore the double ionization curve is small at small angles and increases to a maximum at back angles where this difference is largest. If all processes were calculated using the same wave functions, this sum rule would be a rigorous check of the results. Here this approach is not expected to be quantitatively accurate due to the use of different wave functions in the various approximations. For example, the impulse approximation for single ionization (calculated using Hartree-Fock Compton profiles) exceeds the incoherent scattering factor (obtained with variational wave functions) at some angles.

Total cross sections for photons interacting with the ground state of helium are presented in Fig. 5. The

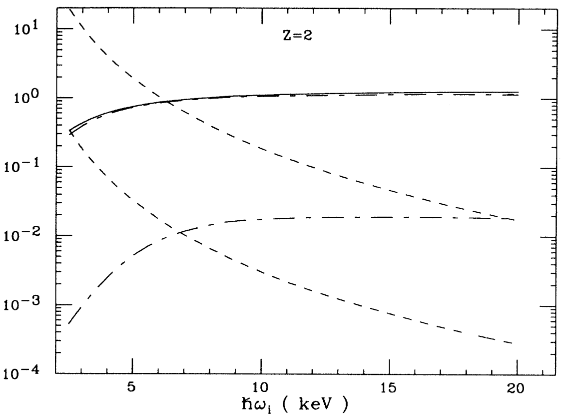


FIG. 5. Total cross sections for photon-atom interactions are given. Cross sections for the single (double) ionization of helium by Compton scattering, using the formalism presented here, are given by the upper (lower) chain dashed curve. Single (double) photoionization cross sections of Hino *et al.* [25]. are given by the upper (lower) dashed curves. The incoherent scattering factors of Hubbell *et al.* [28] are also given for reference (solid curve).

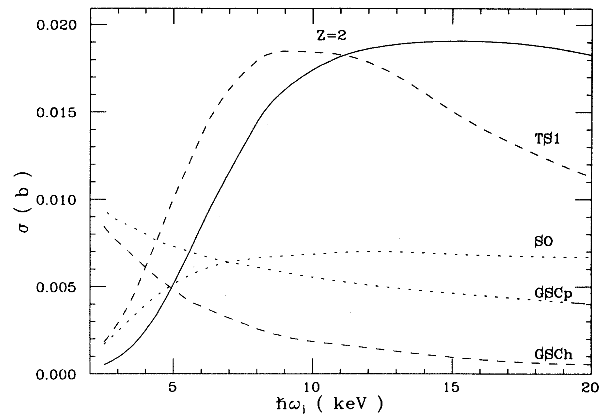


FIG. 6. Cross sections for double ionization of helium by Compton scattering within the many-body perturbation theory framework. The solid curve is the full MBPT cross section. The dashed curves are derived from the MBPT amplitudes corresponding to correlation in the initial state. The dotted curves are calculated from the MBPT amplitudes corresponding to correlation in the initial state.

data for single and double ionization by photoeffect are from the calculations of Hino *et al.* [25]. The energy where Compton scattering becomes the dominant ionizing mechanism is near 6 keV. Double ionization by Compton scattering becomes larger than double ionization by photoeffect at nearly the same energy. It is remarkable that no measurement of the total cross section for single ionization by Compton scattering had been reported until quite recently [33]. The results given in Fig. 5 should provide accurate cross sections for comparison with such measurements at low photon energies. At higher energies relativistic corrections are important and, at the very least, the Klein-Nishina cross section should be used rather than the Thomson cross section. The correction to the total incoherent scattering cross sections reported here, found using this substitution, is approximately 8% at 20 keV.

The curve for single ionization by Compton scattering approaches a constant at high energy. At first glance it appears that the curve for double ionization by Compton scattering does also. In Fig. 6 we demonstrate that 20 keV is not a high enough energy to identify such a limit. Here the cross section for double ionization by Compton scattering is shown along with cross sections derived from the individual many-body amplitudes. The full double ionization cross section drops slightly from its peak value (although this drop is still within the expected numerical error of the extrapolation procedure that was described above). The dramatic decrease of the cross sections corresponding to specific final state correlation amplitudes, particularly TS1, demonstrates that the asymptotic limit has not been reached. One can extrapolate, using an *ad hoc* $1/\omega^3$ dependence for the TS1 amplitude, to determine the incident photon energy at which the TS1 cross section is less than 1% of its value at 20 keV to be of the order of 75 keV. At this photon energy, the average energy available to the ejected electrons is larger the electron energy at which the photoeffect double to single ionization ratio has nearly reached the asymptotic value.

In Fig. 7 the ratio $R_c(\hbar\omega_i) = \sigma^{++}(\hbar\omega_i)/\sigma^{+}(\hbar\omega_i)$ obtained from the cross sections for single and double ionization by Compton scattering are shown. We also show the recent results of Surić *et al.* [13] and of Andersson and Burgdörfer [14]. There are quite large quantitative differences between all of the calculations. As discussed above, the low electron energies available in the final state may mean that one needs to go beyond the lowest-order

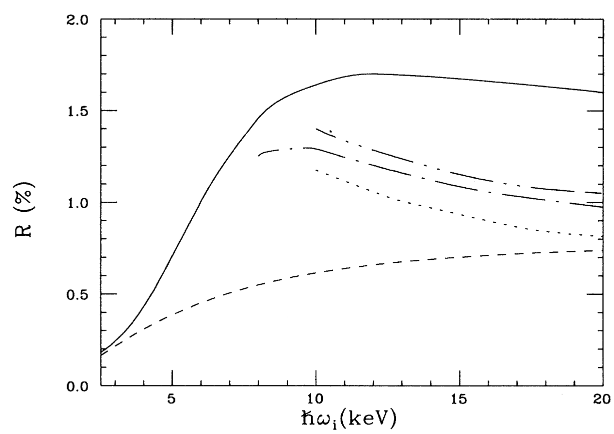


FIG. 7. Ratios of double to single ionization of helium by Compton scattering of a single photon calculated in the present work are given by the solid curve. The ratios of Surić *et al.* [13] are given by the dashed curve. Also shown are three curves from the work of Andersson and Burgdörfer [14]. The approximate final state wave functions used in these calculations are the uncorrelated product of two Coulomb waves (dots), the continuum distorted wave function (dash-dotted curve), and a Byron-Jochain-type function with the correlation coefficient as determined in [9] (dash-double-dotted curve).

MBPT approach used here. However, our methods retain final state correlations ignored in the impulse approximation used by Surić *et al.* The calculations of Burdörfer and Andersson [14] were obtained using two Coulomb waves for the final state wave function (no correlation) and with two different approximate correlated final state wave functions. The difference between the calculations using different uncorrelated final states and different approximations to correlated final states shows the need to include correlation consistently at finite energy.

The need for final state correlation at finite energy, when the generalized length form of the $A^2$ operator is used, is further demonstrated in Fig. 8. Here the angle integrated scattered photon spectrum obtained using the full MBPT amplitude is contrasted with the spectra obtained from using initial (GSCp + SO) and final state contributions (GSCh + TS1) separately. Integrating over the spectra shown here, one finds that the contribution from the final state correlations alone is slightly larger than the integrated result obtained by coherently summing all four amplitudes. The initial state correlation is approximately two-thirds of this full result. Clearly, at this energy (12 keV), all amplitudes are necessary to accurately describe the physics of the problem. For an incident photon energy of 20 keV, the relative importance of the final state correlations decreases. However, at this energy the magnitude of the contribution of these correlations is still fairly substantial, being approximately two-thirds of the full result.

There had been some speculation that the asymptotic Compton scattering and photoeffect ratios should be the same [8,34,35]. The results of Surić *et al.* [13] approach an asymptotic ratio of approximately 0.8%. These authors performed a direct calculation of the ratio, also within the impulse approximation, at infinite photon energy, finding a value of 0.797%. A similar value has been obtained by Andersson and Burgdörfer [14], although the energy at which the asymptotic value is reached is rather different. We cannot project an asymptotic value from our calculations, but find it rather unlikely to be the same as for the photoeffect. The reason is our expectation that the TS1 amplitude should become vanishingly small, as discussed above. It should be noted that the vanishing of the TS1 amplitude is a necessary, but not sufficient, condition for the impulse approximation to hold. Based on the agreement between the asymptotic results of Surić *et al.* [13] and of Andersson and Burgdörfer, it would appear that the asymptotic value of ratio of double to single ionization of helium by Compton scattering has been determined. We note that this asymptotic value has been obtained using the generalized length form of the $A^2$ operator and that this result has not been determined to be invariant under changes of the form of the $A^2$ operator.

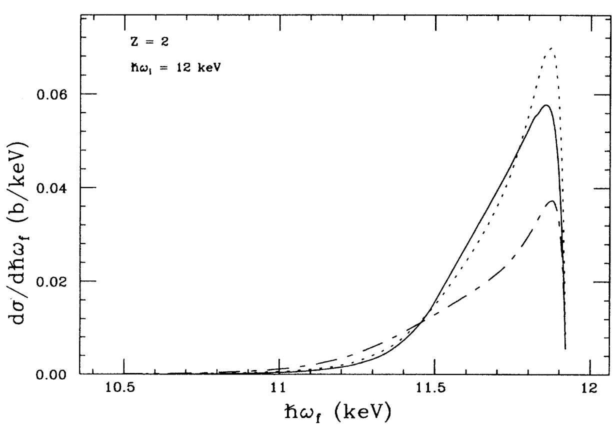


FIG. 8. Angle integrated cross sections for double ionization by Compton scattering of 12-keV photons from the ground state of helium are shown. The solid curve includes all amplitudes. The other curves are obtained from the final state (TS1+ GSCh) correlation based amplitudes (dotted curve) and from the two initial state (SO + GSCp) correlation amplitudes (chain dashed curve).

It is well known, from the photoeffect, that the independence of results from the form of the interaction operator only applies when exact initial and final state wave functions are used. We have already discussed how, in calculations of the double photoeffect at finite energies, results depended strongly on the form of the interaction operator [22,23] when approximate wave functions were used to represent the final state. Asymptotically, even the energy dependence of the double photoionization cross section depends on the form of the operator. While the acceleration form of the dipole operator has been determined to be the most useful in calculating double photoeffect [36,37], no similar analysis has been performed for the $A^2$ operator. These observations do not apply to the work presented here, which has been demonstrated to be independent of the form of the interaction operator when the full amplitude is considered.

A direct experimental resolution of the differing predictions for the ratio of double to single ionization by Compton scattering of Fig. 7 is not likely in the near future. The reason is that the experiments currently being performed measure the time of flight of the residual ions without distinguishing between the different mechanisms that create them. One obtains instead the ratio of double to single ionization by all ionizing processes

$$R(\hbar\omega_i) = \frac{\sigma_{\rm CS}^{2+}(\hbar\omega_i) + \sigma_{\rm ph}^{2+}(\hbar\omega_i)}{\sigma_{\rm CS}^{+}(\hbar\omega_i) + \sigma_{\rm ph}^{+}(\hbar\omega_i)}, \qquad (18)$$

where $\sigma_{\rm CS}^{+}$ and $\sigma_{\rm CS}^{2+}$ are the single and double ionization cross sections by Compton scattering and $\sigma_{\rm ph}^{+}$ and $\sigma_{\rm ph}^{2+}$ are counterparts by photoionization. In Fig. 9 we show the ratios obtained in the experiments of Levin *et al.* [2] and of Bartlett *et al.* [38] with the theoretical predictions obtained by combining the theoretical predictions for Compton scattering from the present work and those of Surić *et al.* [13] with the MBPT predictions of the ionization due to photoeffect [25]. Also shown is the estimate of Andersson and Burgdörfer [9]. Due to the rather large experimental uncertainties, the measurements are not as yet able to distinguish between the various theoretical predictions. However, the high energy data [2] certainly suggest that the ratio at these energies, where Compton scattering is dominant, may be similar to the

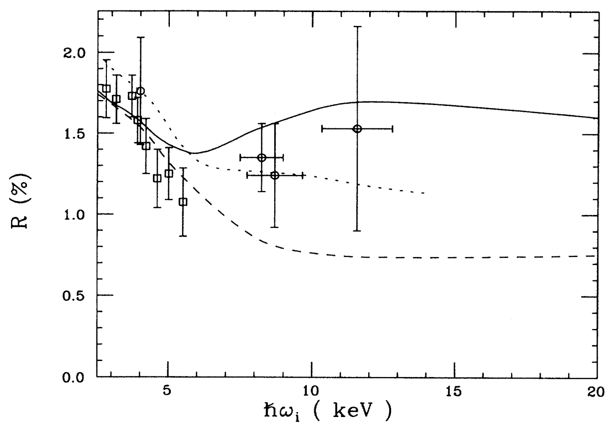


FIG. 9. Results for the ratio of double to single ionization of helium by single-photon impact. The solid line gives the present overall ratios incorporating contributions of both Compton scattering and photoionization The dotted curve is the estimate by Andersson and Burgdörfer [9]. The dashed curve is from the recent calculations of Surić *et al.* [13]. The circles are the experimental data from [2] and the squares are the experimental data from [38].

asymptotic photoeffect ratio. The dip in this ratio between 2.5 and 8 keV and the high energy range are both likely regions where future experiments should be able to distinguish between these theories.

## IV. CONCLUSIONS

Double ionization of helium by Compton scattering has been investigated. The nonrelativistic matrix element for the $A^2$ interaction operator has been expanded to lowest order, beyond the Hartree-Fock approximation, in the electron-electron interaction. While the generalized length form of the $A^2$ operator has been used in this work, the form invariance of this operator within our framework has been explicitly demonstrated. However, the amplitudes corresponding to the individual MBPT diagrams have been shown to be dependent on the form of the operator.

The incident photon energies considered here (from 2.5 to 20 keV) cover the energies of interest for this process, from the energies where the process becomes significant to energies beyond the range of experimental data. The energy transferred to the ejected electrons is rather small compared to the energy transferred in the photoeffect process at the same energy. These low electron energies suggest that inclusion of higher-order correlations may be desirable.

The ratio of double to single ionization by Compton scattering differs substantially from the recent results of Surić *et al.* [13] and of Andersson and Burgdörfer [14]. At the energies considered in this paper, and for the generalized length form of the $A^2$ operator, the neglect or approximate inclusion of final state correlations in their work and the approximate inclusion in our own work may be responsible for these discrepancies. The asymptotic value of this ratio, estimated by Surić *et al.*, differs from our high energy data. The reason for this is that our data have likely not reached an asymptotic limit.

The ratio of double to single-ionization by all single-photon processes has been compared with the results of Andersson and Burgdörfer [9] and with the results of Surić *et al.* [13]. All of these approaches yield different predictions. Unfortunately, experimental measurements are not yet sensitive enough to distinguish between the approaches. The resolution of these discrepancies will continue to present theorists and experimentalists with challenges in the immediate future. The longer term prospects are also good as Compton scattering can be a unique probe of the atomic charge density, including the electron-electron interaction.

## ACKNOWLEDGMENTS

Support of this research by the National Science Foundation under Grant No. PHY-9222489 is acknowledged. K.H. thanks the Japanese Ministry of Education for supporting his stay in the U.S. We thank the Joint Institute for Computational Science of the University of Tennessee for use of their Connection Machine CM-5 computer. The present numerical computations were also supported in part by the Institute of Physical and Chemical Research (RIKEN).

[1] J. W. M. Dumond, Phys. Rev. **33**, 643 (1929).

[2] J. C. Levin, D. W. Lindle, N. Keller, R. D. Miller, Y. Azuma, N. Berrah Mansour, H. G. Berry, and I. A. Sellin, Phys. Rev. Lett. **67**, 968 (1991); J. C. Levin, I. A. Sellin, B. M. Johnson, D. W. Lindle, R. D. Miller, N. Berrah, Y. Azuma, H. G. Berry, and D.-H. Lee, Phys. Rev. A **47**, R16 (1993).

[3] Thomas A. Carlson, Phys. Rev. **156**, 142 (1967).

[4] R. Wehlitz, F. Heiser, O. Hemmer, B. Langer, A. Menzel, and U. Becker, Phys. Rev. Lett. **67**, 3764 (1991).

[5] V. Schmidt, N. Sandner, and H. Kuntzemüller, Phys. Rev. A **13**, 1743 (1976); V. Schmidt, N. Sandner, H. Kuntzemüller, P. Dhez, F. Wuilleumier, and E. Källne, *ibid.* **13**, 1748 (1976).

[6] H. Kossman, V. Schmidt, and T. Andersen, Phys. Rev. Lett. **60**, 1266 (1988).

[7] Roger J. Bartlett, Peter J. Walsh, Z. X. He, Y. Chung, E.-M. Lee, and James A. R. Samson, Phys. Rev. A **46** 5574 (1992); James A. R. Samson, R. J. Bartlett, and Z. X. He, *ibid.* **46** 7277 (1992).

[8] James A. R. Samson, Chris H. Greene, and R. J. Bartlett, Phys. Rev. Lett. **71**, 201 (1993).

[9] Lars R. Andersson and Joachim Burgdörfer, Phys. Rev. Lett. **71**, 50 (1993); L. R. Andersson and J. Burgdörfer, in *The Physics of Electronic and Atomic Collisions, XVIII International Conference, Aarhus, 1993*, edited by Torkild Andersen, Bent Fastrup, Finn Folkmann, Helge Knudsen, and N. Andersen, AIP Conf. Proc. No. 295

(AIP, New York, 1994), p. 836.

[10] P. Eisenberger and P. M. Platzmann, Phys. Rev. A **2** 415, (1970).

[11] F. Biggs, L. B. Mendelsohn, and J. B. Mann, At. Data Nucl. Data Tables **16**, 201 (1975).

[12] Ken-ichi Hino, P. M. Bergstrom, Jr., and Joseph H. Macek, Phys. Rev. Lett. **72**, 1620 (1994).

[13] T. Surić, K. Pisk, B. A. Logan, and R. H. Pratt, Phys. Rev. Lett. **73**, 790 (1994).

[14] L. R. Andersson and J. Burgdörfer, Phys. Rev. A **50**, R2810 (1994).

[15] T. Surić, P. M. Bergstrom, Jr., K. Pisk, and R. H. Pratt, Phys. Rev. Lett. **67**, 189 (1991).

[16] F. Schnaidt, Ann. Phys. (Leipzig) **21**, 89 (1934).

[17] M. Schumacher, F. Smend, and I. Borchert, J. Phys. B **8**, 1428 (1975); Comput. Phys. Commun. **11**, 363 (1976).

[18] P. M. Bergstrom, Jr., T. Surić, K. Pisk, and R. H. Pratt, Phys. Rev. A **48**, 1134 (1993).

[19] Note that atomic units are used unless otherwise stated.

[20] The Compton scattering cross sections for single ionization reported here were obtained using an equation similar to Eq. (1) with Hartree-Fock initial and final states.

[21] G. W. F. Drake and Zong-Chao Yan, Phys. Rev. A **46**, 2378 (1992).

[22] F. W. Byron, Jr. and Charles J. Jochain, Phys. Rev. **164**, 1 (1967).

[23] M. A. Kornberg and J. E. Miraglia, Phys. Rev. A **48**, 3714 (1993).

[24] Steven L. Carter and Hugh P. Kelly, Phys. Rev. A **24**, 170 (1981).

[25] Ken-ichi Hino, Takeshi Ishihara, Futoshi Shimizu, Nobuyuki Toshima, and J. H. McGuire, Phys. Rev. A **48**, 1271 (1993).

[26] The difference between the GSCp and the GSCh diagrams is in the propagator from the electron-electron interaction to the interaction with photons. The former is mediated by a particle propagator, the latter by a hole propagator.

[27] Yong-Ki Kim and Mitio Inokuti, Phys. Rev. **175**, 176 (1968).

[28] J. H. Hubbell, Wm. J. Veigele, E. A. Briggs, R. T. Brown, D. T. Cromer, and R. J. Howerton, J. Phys. Chem. Ref. Data **4**, 471 (1975).

[29] O. Klein and Y. Nishina, Z. Phys. **52**, 853 (1929).

[30] See, for example, Robley D. Evans, *The Atomic Nucleus* (McGraw-Hill, New York, 1955).

[31] Yong-Ki Kim and Mitio Inokuti, Phys. Rev. **165**, 39 (1968).

[32] The total inelastic scattering cross sections in Ref. [28] are obtained by integrating their incoherent scattering factors and the relativistic free Compton (or Klein-Nishina) cross section. We are working in a nonrelativistic framework and have chosen to use their incoherent scattering factors and the Thomson cross section. The difference between these choices is apparent at high energies where the total Thomson scattering cross section approaches a constant and the Klein-Nishina cross section does not.

[33] J. A. R. Samson, Z. X. He, R. J. Bartlett, and M. Sagurton, Phys. Rev. Lett. **72**, 3329 (1994).

[34] M. Ya. Amusia, Argonne National Laboratory Report No. ANL/PHY-94/1, 1994 (unpublished).

[35] E. G. Drukarev and F. F. Karpeshin, J. Phys. B **9**, 399 (1976).

[36] A. Dalgarno and A. L. Stewart, Proc. Phys. Soc. London **76**, 49 (1960).

[37] A. Dalgarno and H. R. Sadeghpour, Phys. Rev. A **46**, R3591 (1992).

[38] R. J. Bartlett, M. Sagurton, J. A. R. Samson, and Z. X. He (unpublished).