Contribution of Compton Scattering to the Double Ionization of Helium

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Double ionization of helium by single-photon impact is analyzed for photon energies from 4 to 12 keV. Many-body perturbation theory is applied to obtain the Compton scattering contribution to the total cross section for double ionization. Compton scattering dominates photoionization above 6 keV. Despite the differences between the two processes, the ratio of double to single ionization by Compton scattering appears to approach the asymptotic value given by photoionization calculations. These ratios are in agreement with recent experiments.

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Helium is a particularly useful paradigm for investigating electron-electron interactions because it is the simplest, abundant atomic system where such correlations are significant. Processes involving photons are pure probes of correlations in the sense that photons interact with individual electrons; transitions involving multiple electrons require interactions between the electrons. The ratio R of double to single ionization represents a key parameter characterizing such correlations. Tunable, high intensity radiation from synchrotron sources has recently been applied to experiments measuring this ratio by photon impact in the near threshold [1–3], the intermediate energy [4,5], and the asymptotic regions [6] of the double photoionization process.

Early theoretical investigations of helium successfully demonstrated that the high-energy dependence of the double ionization cross section is the same as that for single ionization yielding a constant asymptotic ratio [7–10]. Carter and Kelly [11] applied the many-body perturbation theory (MBPT) to investigate the ratio of double to single ionization in the near threshold regime. The agreement between their results and experiments at low energies recently prompted Ishihara, Hino, and McGuire [12] to extend this approach to higher energies. While the agreement between theory and experiment has been quite reasonable for photon energies from near threshold to 12 keV, Samson, Greene, and Bartlett [13] have recently noted the importance of the Compton scattering process to the interpretation of these measurements. Indeed, experiments performed thus far do not discriminate between photoionization and Compton scattering.

Here the ratio of double to single ionization for helium by photon impact reads

$$R = \frac{\sigma_{\rm cs}^{2+} + \sigma_{\rm ph}^{2+}}{\sigma_{\rm cs}^{+} + \sigma_{\rm ph}^{+}}, \tag{1}$$

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. The inability to distinguish between the two processes becomes problematic at high photon energies (for helium at 6 keV), where Compton scattering cross sections may dominate photoionization cross sections, so an accurate theoretical treatment of the Compton scattering contribution to double ionization is essential to interpret available experimental data. Recently, Andersson and Burgdörfer [14] have made a simple estimate of R based on photoionization data. In this Letter, we report the first *ab initio* calculations of the double ionization of helium due to Compton scattering using the lowest-order MBPT and discuss its contribution to the overall ratio R. The present theoretical framework is similar to that given in [15].

There exist substantial differences between Compton scattering and photoionization. (1) The most probable energies for electrons ejected by Compton scattering, equivalent to the energy transfer ω of photons to an atom, are significantly smaller than for photoionized electrons [13]. The energies of electrons ejected by photoionization are nearly equal to incident photon energies concerned here. However, the electron energies by Compton scattering distribute around 100 eV at the incident photon energy of about 8 keV. Electrons with energy of 1 keV by Compton scattering do not become likely until the incident photon energies reach 16 keV. MBPT has been successfully applied to photoionization for this wide range of ejected-electron energies [11,12]. (2) In photoionization, the nonrelativistic dipole approximation to the total cross section is known to hold to relatively high photon energy region due to a cancellation effect of relativistic and retardation effects [16]. In contrast with that, the momentum transfer $\mathbf{k}$ of photons to an atom is relatively large in Compton scattering and the dipole approximation is not valid. As a result, the scattered photon distribution is predominantly toward the backward angular region and more terms of the multipole expansion of the photon fields should be taken into consideration.

Photoionization cross sections are obtained by a first-order (lowest-order) evaluation of the $\mathbf{p}\cdot\mathbf{A}$ term of the photon-electron interaction. The nonrelativistic amplitude for Compton scattering from bound electrons may be obtained by evaluating the diagrams of Fig. 1(a). The first diagram (seagull term) corresponds to a first-order evaluation of the $\mathbf{A}^2$ term of the photon-electron

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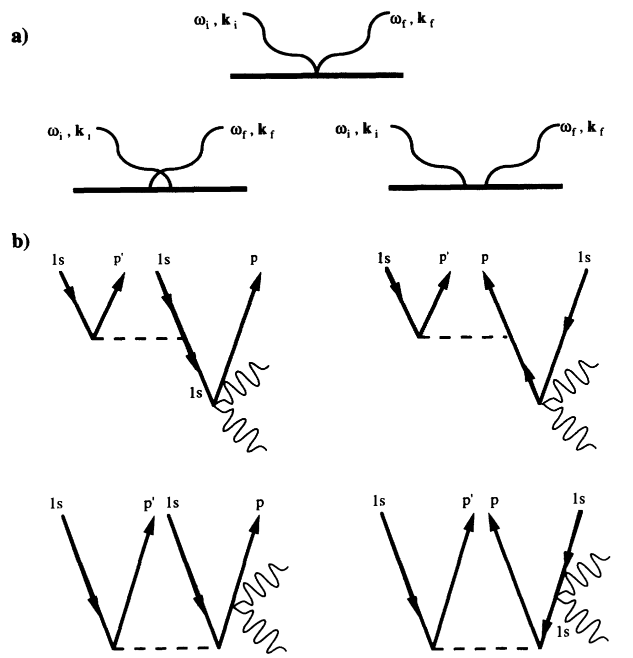

FIG. 1. (a) Diagrams representing the nonrelativistic amplitude for Compton scattering from bound electrons. (b) The lowest-order MBPT diagrams for double ionization of helium by Compton scattering using the seagull term of (a). The electron-electron interaction is shown by a dotted line. It is understood 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.

interaction. In the Coulomb gauge, it accounts for the peak region of the scattered photon spectrum, the width reflecting the Compton profile of the scatterer. The remaining diagrams (pole terms) represent the evaluation of the $\mathbf{p}\cdot\mathbf{A}$ term in second order. These terms give resonant behavior in outer atomic subshells and divergent behavior for soft scattered photon energies in all shells.

TABLE I. Cross sections (cm^2) of the single and double ionizations for Compton scattering, σ_{cs}^{+} and $\sigma_{\mathrm{cs}}^{2+}$, and for photoionization, σ_{ph}^{+} and $\sigma_{\mathrm{ph}}^{2+}$, vs the incident photon energy ω (keV). The data of photoionization are taken from [15]. The numbers in square brackets represent powers of 10.

ω	σ_{cs}^{+}	$\sigma_{\mathrm{cs}}^{2+}$	σ_{ph}^{+}	$\sigma_{\mathrm{ph}}^{2+}$
4.0	5.64[-25]	2.47[-27]	4.03[-24]	6.98[-26]
6.0	8.33[-25]	8.43[-27]	1.03[-24]	1.74[-26]
8.0	9.81[-25]	1.46[-26]	3.92[-25]	6.53[-27]
10.0	1.06[-24]	1.74[-26]	1.85[-25]	3.02[-27]
12.0	1.09[-24]	1.82[-26]	9.96[-26]	1.62[-27]

Here we consider contributions only from the seagull term, that is the $\mathbf{A}^2$ approximation to Compton scattering. The validity of this approximation in describing the Compton peak for scattering from helium was discussed in [17]. We note, however, that it is not possible to uniquely define a total cross section for Compton scattering as the scattered photon spectrum is infrared divergent. Rather one must add the Compton scattering amplitude to the radiative corrections to photoionization. This procedure yields a finite correction to the photoionization cross section [18,19]. The resulting corrections are expected to be small for the cases considered here. According to a recent analysis in the case of hydrogen [20], the total ionization cross section may be accurately approximated by adding the photoionization cross section (without radiative corrections) and the Compton scattering cross section obtained within the $\mathbf{A}^2$ approximation.

The differential cross section of double ionization within the $\mathbf{A}^2$ approximation is given by the expression

$$\frac{d^2\sigma_{\mathrm{cs}}^{2+}}{d\omega d\Omega} = \left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{Th}} \left(\frac{\omega_f}{\omega_i}\right) \int d\mathbf{p}_1 d\mathbf{p}_2 \left| \langle \Psi_f | \sum_{j=1,2} e^{i\mathbf{k}\cdot\mathbf{r}_j} | \Psi_i \rangle \right|^2 \delta(E_f - E_i - \omega), \tag{2}$$

where $\left(\frac{d\sigma}{d\Omega}\right)_{\mathrm{Th}}$ is the classical cross section by Thompson scattering with Ω being the solid angle of the scattered photon. $\Psi_{i(f)}$, $E_{i(f)}$, and $\omega_{i(f)}$ stand for the initial (final) wave function, electronic energy, and photon energy, respectively. $\mathbf{p}_1$ and $\mathbf{p}_2$ are momenta of two ejected electrons. It should be understood that Ψ_i and Ψ_f are antisymmetrized many-body wave functions.

Applying the lowest-order MBPT and the $\mathbf{A}^2$ approximation to the double ionization by Compton scattering, we obtain the four MBPT diagrams shown in Fig. 1(b). These diagrams look similar to those used to calculate photoionization [15] besides one additional diagram (the lower right one in this figure). This diagram represents the ground-state correlation mediated by a hole propagation from the electron-electron interaction to the photon-electron interaction.

We concentrate here on the contribution of Compton scattering to the ratio R of double to single ionization in helium at energies where this process is important and where measurements of R have been reported. We defer a more detailed discussion on our calculations of the double Compton scattering process to a subsequent paper. In Table I we tabulate our results for cross sections of Compton scattering and photoionization at energies from 4 to 12 keV. In Fig. 2 we show our results for R as well as the individual ratios due to Compton scattering and photoionization. We also give the existing experimental data [6,21] and the theoretical estimate [14] for comparison. As expected, Compton scattering dominates photoionization above 6 keV, reflecting the importance of this process to the total ionization cross section at these

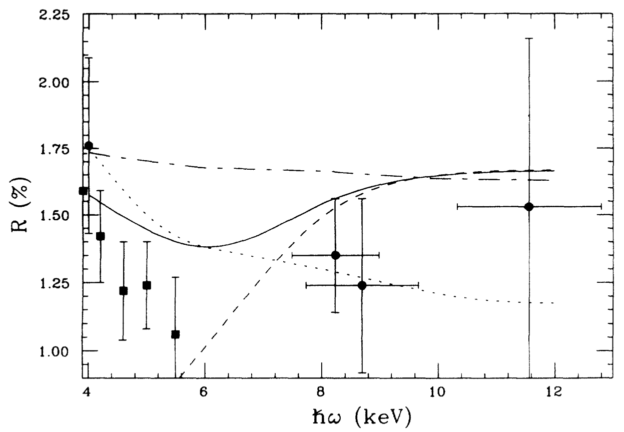

FIG. 2. Results for the ratio of double to single ionization of helium by single-photon impact. The solid line gives the present overall ratios incorporating both contributions of Compton scattering (given separately by the dashed line) and photoionization (given separately by the chain dashed line) processes. These data are interpolated for eye guide, obtained by applying a cubic-spline interpolation scheme to all of calculated results for total cross sections given in Table I. The dotted curve is the estimate by Andersson and Burgdörfer [14]. The full circles are the experimental data from [6] and the squares are the experimental data from [21].

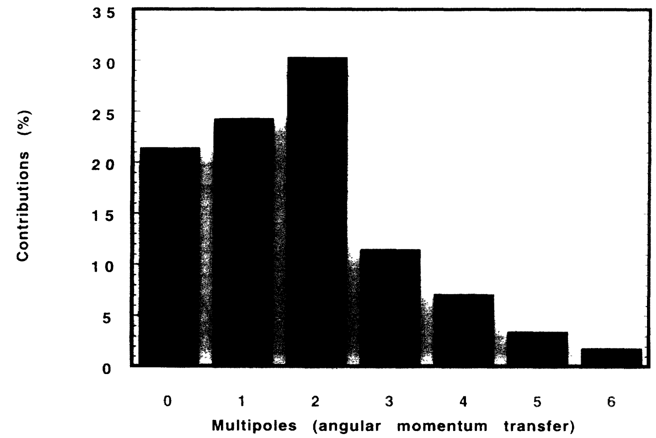

FIG. 3. Multipole contributions to the total cross section for double ionization of helium by Compton scattering at the photon energy of 12 keV.

energies. We note that our overall ratio has a shallow dip structure around 6 keV, which corresponds to a transitional change of the dominant ionizing process from photoionization to Compton scattering. The experimental data of Bartlett *et al.* [21] also show a decrease of R at these energies. This decrease is more pronounced than our calculations show, possibly reflecting the difference in efficiency of their apparatus to collecting residual ions by Compton scattering versus those by photoionization [22]. Furthermore, the Compton scattering ratio appears to be approaching an asymptotic value of 1.67% at high energy, which is almost the same as the asymptote predicted for photoionization. This is surprising due to the intrinsic differences between the two processes discussed above.

In Fig. 3 we present the relative contributions of the multipole series, for given angular momentum transfer, to the total cross section of double ionization by Compton scattering at 12 keV. It is easily understood that a complete multipole expansion is essential for an adequate description of this process. We note the substantial contribution of the monopole term, which corresponds to the case of no angular momentum transfer from the photon to the system; the angular momentum is fully carried away by the scattered photon. The need for additional multipoles becomes more important in Compton scattering as the incident photon energy is increased. The estimate of Andersson and Burgdörfer [14], predicting somewhat smaller ratios of double to single ionization, differs in detail from our more exact calculations. Their prediction is based mostly on the existing photoionization ratios for describing transitions to the final dipole electronic states and additionally on the shakeoff contribution to the photoionization ratio for estimating higher angular momentum transitions. Their estimate of the monopole term of the cross section is weighted by photoionization-excitation calculations [23]. Their multipole dependence differs from ours, being shifted to higher angular momenta. While our results for the total ratio are in agreement with experiment, more precise measurements with a monochromatic beam at high energy are needed to distinguish between the different theoretical predictions and to deepen our understanding of double ionization by Compton scattering.

In summary, we have applied the many-body perturbation theory to the bound-electron Compton scattering process in order to obtain its contribution to the ratio of double to single ionization of helium by single-photon impact in the energy region from 4 to 12 keV. We have confirmed that Compton scattering is the dominant ionizing process above 6 keV and have also demonstrated the essential difference between Compton scattering and photoionization, particularly the need for higher multipoles in Compton scattering. Our ratios by Compton scattering approach an asymptotic value similar to those by photoionization despite this difference between the two processes. Our results differ from earlier estimates, however, these differences are not yet distinguishable by experiment.

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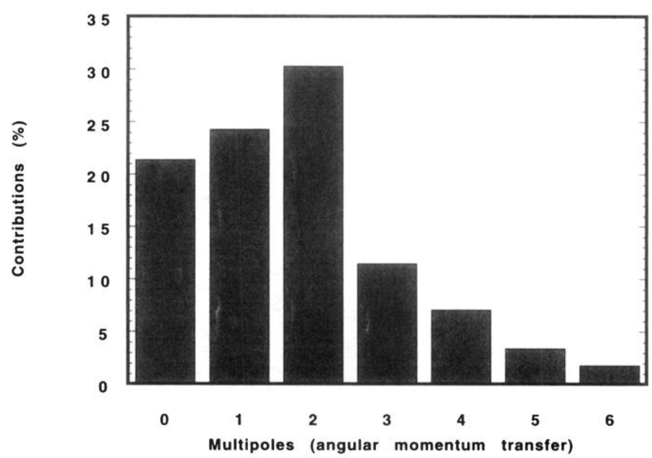

FIG. 3. Multipole contributions to the total cross section for double ionization of helium by Compton scattering at the photon energy of 12 keV.