Differential Double Ionization of He by Compton Photons and Charged Particles at Large Energy Transfers

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Double ionization of helium differential in energy transfer, ϵ, is studied for both high-energy Compton photons and charged particles. The ratios of double to single ionization, $R_C(\epsilon)$ for Compton scattering, and $R_Z(\epsilon)$ for charged particles, are found to display an unexpected behavior: For large ϵ up to the two-body binary encounter (BE) limit, ϵ_{BE}, we find $R_C(\epsilon) = R_Z(\epsilon) \simeq 0.86\%$ in good agreement with recent experimental data. For even larger $\epsilon > \epsilon_{\mathrm{BE}}$, $R_C(\epsilon)$ and $R_Z(\epsilon)$ are modified by an "exchange shakeoff" mechanism. At $\epsilon \gg \epsilon_{\mathrm{BE}} \gg 1$, both differential (unlike integrated) ratios are predicted to approach the photoionization limit. [S0031-9007(96)00861-7]

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Two-electron transitions in atoms by impact of photons and charged particles continue to attract considerable interest [1–10], primarily because it can serve as a sensitive probe of many-body and correlation effects in atoms. The simplest two-electron system is helium where correlation is important, but only partially understood.

The need to understand the similarities and differences between impact by photons and by charged particles is underscored by recent experimental and theoretical activities studying two-electron processes such as double ionization of He. New synchrotron radiation sources have extended the photon energy range such that Compton scattering, i.e., inelastic scattering of photons, can dominate over photoionization [3–9]. At the same time, advances in experiments have been reported in which double ionization by fast charged particles is measured at large energy transfers, ϵ, either directly [10] or indirectly [11–13]. At large ϵ Compton scattering and charged particles probe the correlated two-electron initial state over the whole coordinate space, thus providing new and complementary information to photoionization that is sensitive to the localized region near the nucleus. The high-energy ratio of double to single ionization, R, for photoionization is measured to be $\sim$1.7%, in agreement with most theories [2,14,15]. Theoretical predictions for the corresponding ratio for Compton scattering, integrated over all energy and momentum transfers, range from 0.8% [6,16] to 1.6% [5,17]. More recently, a data point at 58 keV has been reported at 0.84% [9]. Concurrently, the ratio by proton impact has been measured to be $\sim$0.8% at an energy transfer of 10 keV [10].

In this Letter, we present the first comparative theoretical study for double ionization of He by high-energy Compton scattering and by fast charged particles differential in energy transfer ϵ. Our goal is to explore the similarities between the two processes at large ϵ and show that the ratio $R_C(\epsilon)$ for Compton scattering is closely related to the ratio $R_Z(\epsilon)$ for charged particles. For large ϵ up to the binary encounter limit ϵ_{BE}, $R_C(\epsilon)$ and $R_Z(\epsilon)$ become identical and approach the same limit $\sim$0.86%. More remarkably, we find that for energy transfers beyond the binary encounter limit, the ratios Compton scattering $R_C(\epsilon)$ or charge particles $R_Z(\epsilon)$ change dramatically and both approach the photoionization limit. While completely negligible for the total cross section, this observation provides new insights into the interconnection between different mechanisms for double ionization.

To first order the triply differential cross section for Compton scattering [16,18] in terms of the ejected electron energies E_1, E_2, and momentum transfer Q is (in atomic units hereafter, unless noted otherwise)

$$\frac{d\sigma_C^{++}}{dE_1 dE_2 dQ^2} = \frac{\pi \alpha^2 k_1 k_2}{2\omega^2}[1 + \cos^2\theta] F_{fi}^{++}(Q), \quad (1)$$

where

$$F_{fi}^{++}(Q) = \int d\Omega_1\, d\Omega_2 |\langle f| \sum_{j=1,2} \exp(i\mathbf{Q}\cdot\mathbf{r}_j)\, |i\rangle|^2 \quad (2)$$

is the angle-integrated inelastic form factor, α the fine structure constant, $k_{1,2}$ and $\Omega_{1,2}$ the magnitude and direction of momenta of the ejected electrons,respectively, and ω and θ the energy and the scattering angle of the photon, respectively. The form factor $F_{fi}^{++}(Q)$ in Eq. (2) depends on the initial and final states of He $|i\rangle$ and $|f\rangle$. It also depends implicitly on the energy transfer $\epsilon = E_1 + E_2 - E_i$ where E_i is the energy of the initial state.

The first order cross section for charged particles may be similarly expressed as [19]

$$\frac{d\sigma_Z^{++}}{dE_1 dE_2 dQ^2} = \frac{4\pi Z^2 k_1 k_2}{v^2} \frac{F_{fi}^{++}(Q)}{Q^4}, \quad (3)$$

where Z, v are the charge and speed of the charged particle. Except for the prefactor in Eq. (1) arising from summing of polarization vectors and the characteristic momentum transfer dependence of Coulomb scattering $1/Q^4$ in Eq. (3), the cross sections for Compton scattering Eq. (1) and for charged particles Eq. (3) are structurally similar. Specifically, the dependence on the initial and final states is in either case given by the atomic transition form factor $F_{fi}(Q)$.

For a given set of energy and momentum transfers ϵ, Q, the prefactors in Eqs. (1) and (3) will be the same for double as well as single ionization. As has been noted before [20], the ratios of double to single ionization $R_C(\epsilon, Q)$ and $R_Z(\epsilon, Q)$ should therefore be identical for given ϵ and Q, both proportional to $F_{fi}^{++}(Q)/F_{fi}^{+}(Q)$ where $F_{fi}^{+}(Q)$ is the form factor for single ionization. In the following, we will show that in the limiting case of large energy transfer, this identity holds also for the ratios singly differential in energy transfer. To this end we define the binary encounter energy $\epsilon_{\mathrm{BE}} = Q_{\mathrm{BE}}^2/2$ as the maximum energy transfer to an electron permitted in a two-body collision with a photon, ϵ_{BE}^C, and a charged particle, ϵ_{BE}^Z. The maximum momentum transfers, Q_{BE}^C for Compton scattering, and Q_{BE}^Z for a heavy charged particle, in these binary encounter limits are

$$Q_{\mathrm{BE}}^C = \frac{2\omega - \epsilon_{\mathrm{BE}}^C}{c} \approx \frac{2\omega}{c}, \qquad Q_{\mathrm{BE}}^Z \approx 2v. \quad (4)$$

In the energy interval $I_2 \ll \epsilon \lesssim \epsilon_{\mathrm{BE}}$, $F_{fi}^{++}(Q)$ peaks narrowly along the Bethe ridge $\epsilon \simeq Q^2/2$ [20]. I_2 denotes the second ionization potential. Near the Bethe ridge the prefactors in (1) and (3) vary only slightly. One may invoke a peaking approximation to obtain the cross sections as a function of energy transfer for Compton scattering $d\sigma_C^{++} \propto [1 + \cos^2\theta] \int dQ^2\, F_{fi}^{++}(Q)$, and similarly for charged particles $d\sigma_Z^{++} \propto Q^{-4} \int dQ^2\, F_{fi}^{++}(Q)$. The prefactors are again independent of final states, and by the same reasoning as above, the ratios $R_C(\epsilon)$ and $R_Z(\epsilon)$ are proportional to $\int dQ^2\, F_{fi}^{++}(Q)/\int dQ^2 F_{fi}^{+}(Q)$ and approximately equal, i.e.,

$$R_C(\epsilon) \simeq R_Z(\epsilon), \qquad I_2 \ll \epsilon \lesssim \epsilon_{\mathrm{BE}}. \quad (5)$$

This relation is a special case of the previously proposed relation [20] valid for large ϵ. However, it represents a simplification as only ϵ needs to be observed. We show numerically below that the above relation is fulfilled for $\epsilon \gtrsim 3$ keV.

For the initial state entering Eqs. (1) and (3), we use a fully correlated configuration-interaction (CI) type wave function for the initial state. Since we are interested in the high-energy behavior of the cross section, we construct a CI wave function subject to the constraint that the cusp condition of the wave function at the origin $\partial\Psi_i(r_1, r_2, r_{12})/\partial r_2|_{r_2=0} = -2\Psi_i(r_1, 0, r_1)$ for He [15] is exactly satisfied. The cusp condition governs the behavior of the wave function near the nucleus where significant contributions to the high-energy cross section originate. We use a CI expansion in terms of Sturmian functions. In diagonalizing the Hamiltonian we keep the exponent in the Sturmians to be exactly 2 to preserve the cusp property. By including as many as 84 CI terms up to $n, l = 7, 6$ of the ^1S configuration in the initial state, we reproduce the ground state energy within this constraint to a relative accuracy of 4×10^{-4}. Since the high-energy behavior is expected to be weakly dependent on final-state correlations (FSC), we choose the uncorrelated limit for the final state that consists of an antisymmetrized product of two Coulomb waves

$$|f\rangle = \frac{1}{\sqrt{2}}[\psi^-(\mathbf{k}_1, \mathbf{r}_1)\psi^-(\mathbf{k}_2, \mathbf{r}_2) + \mathbf{r}_1 \leftrightarrow \mathbf{r}_2], \quad (6)$$

where $\psi^-(\mathbf{k}, \mathbf{r})$ is the incoming continuum states of He^{++}. The uncorrelated limit may also serve as a useful gauge for FSC at lower energies. Since the initial state is accurate, any discrepancy with experiment can be attributed to the lack of FSC.

The form factor Eq. (2) is analyzed in terms of angular momentum transfer L by expanding $\psi^-(\mathbf{k}_{1,2}, \mathbf{r}_{1,2})$ (6) into partial waves l_1 and l_2 and recouple them to give a total angular momentum L. The nonorthogonality between the S components ($L = 0$) of the initial and final states is removed by a Gram-Schmidt procedure.

The cross sections as a function of the angular momentum transfer L are shown in Fig. 1 for $\epsilon = 1$ and 10 keV. The distribution becomes broader for larger ϵ, as expected. At $\epsilon = 10$ keV, its half-width ΔL is about 20, and it decays slowly as L increases. Large $L_{\max} \sim 40$ are needed to obtain good convergence. For these ϵ's the dominant contribution does not come from the dipole term as in photoionization, but from nondipole terms. Mapped into coordinate space, the broad L distribution illustrates that at large ϵ (but less than ϵ_{BE}), Compton scattering and charged particles probe the entire electron cloud, while photoionization ($L = 1$ only) probes the inner region.

Although the angular momentum transfers of the two processes are strikingly similar (Fig. 1), the spectral distributions as shown in Fig. 2 are very different in magnitude and shape. Compton scattering is remarkably weakly dependent on ϵ up to ϵ_{BE} and is therefore dominated by large ϵ. The cross section for charges particles, on the other hand, is dominated by small ϵ and decreases quickly as $\sim\epsilon^{-2.2}$ until the binary encounter energy, at which point it drops sharply. For $\epsilon < \epsilon_{\mathrm{BE}}$, the Compton photon and the charged particle can deliver sufficient momentum Q to balance ϵ such that the process is localized on the Bethe ridge $\epsilon = Q^2/2$. For energies beyond the binary encounter limit $\epsilon > \epsilon_{\mathrm{BE}}$, the maximum momentum transfer Q_{BE}^C for Compton scattering is insufficient while the minimum momentum transfer $Q_{\min}^Z = \epsilon/v$ for charged particles exceeds the Q value required for the Bethe ridge. In other words,

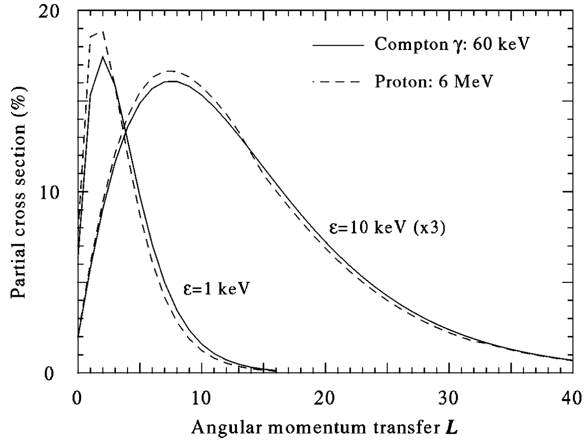

FIG. 1. Distribution of cross sections in terms of angular momentum transfer L for double ionization of He by 60 keV Compton scattering (solid line) and 6 MeV protons (dashed line) at two energy transfers $\epsilon = 1$ and 10 keV.

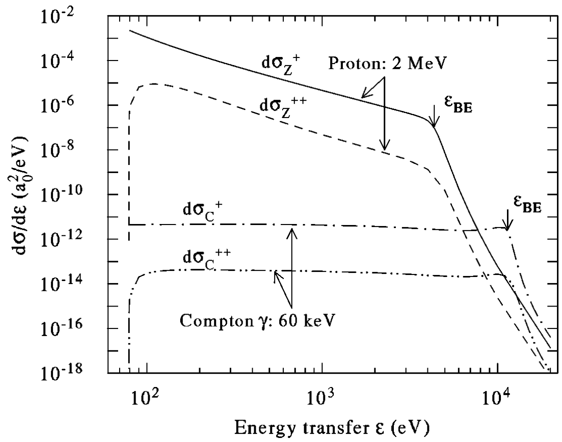

FIG. 2. Cross sections for single and double ionization of He as a function of energy transfer ϵ by 60 keV Compton scattering and 2 MeV protons. The binary encounter energies are marked by ϵ_{BE}.

for $\epsilon > \epsilon_{\mathrm{BE}}$ both processes "fall off" the Bethe ridge, however, in opposite directions.

The ratio of double to single ionization as a function of energy transfer is displayed in Fig. 3. The ratios $R_C(\epsilon)$ for different incident photon energies, ω, are almost identical to each other below binary encounter energy. They rise quickly from threshold and level off around 0.86% for $I_2 \ll \epsilon \leq \epsilon_{\mathrm{BE}}$. $R_Z(\epsilon)$ shows a maximum near 200 eV, and approaches the same value as Compton scattering, thus numerically proving Eq. (5). In comparison with recent experimental data [10,12], also shown in Fig. 3, our results are in good agreement with the experiment for $\epsilon \gtrsim 3$ keV. Our Compton limit is also in good agreement with other theoretical results [6,16] ~0.8% and with recent experimental data 0.84% [9] for the ϵ-integrated ratio which is dominated by large ϵ. Considerable differences exist between theory and experiment toward lower ϵ. Experimental data show a decrease from about 2% at 1 keV down to 1% at 3 keV and are about twice as high as our theory in this energy range. Two sources of possible discrepancies are present in our theory: the lack of final state correlation and higher order terms of the Born series. The second order Born term, in particular the Z^3 effect, is known to be important for the total cross section in the energy range studied here [21,22]. However, the total cross section is dominated by small energy transfers. We believe that the final state correlations are most likely the cause of discrepancy between theory and experiment for ϵ between 0.5 and 3 keV, consistent with experimental observations [10,12]. This could be tested experimentally by using negatively charged $(-Z)$ particles. Compton scattering has no Z^3 effect and would be even more suitable for such a test. The second-order term $(\mathbf{p} \cdot \mathbf{A})^2$ for Compton scattering is of the order of ϵ/ω relative to the first-order term (A^2) and is negligible at high photon energies, with the possible exception of extremely high energy tranfers ($\epsilon \sim \omega$; see below) where the cross section is, however, many orders of magnitude smaller than values near the binary ridge.

Figure 3 displays another remarkable and surprising feature: a small dip followed by a sharp rise of both $R_C(\epsilon)$ and $R_Z(\epsilon)$ at $\epsilon \gtrsim \epsilon_{\mathrm{BE}}$. The sharp rise is partially due to two-electron kinematics in double ionization. The rise in $R_C(\epsilon)$ could be experimentally tested provided sufficient photon flux is available since the cross section is relatively small. At $\epsilon > \epsilon_{\mathrm{BE}}$ the behavior of both $R_C(\epsilon)$ and $R_Z(\epsilon)$ is similar to the corresponding ratio for photoionization at much lower energies. These similarities are not accidental. As Compton and charged particle scattering move away from the Bethe ridge, they cease to exist as two-

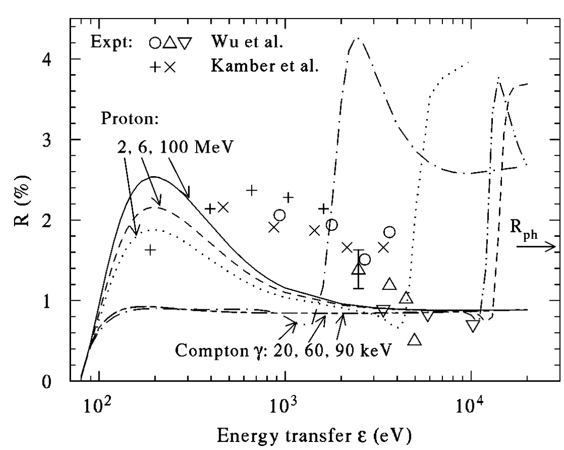

FIG. 3. The ratio, R, of double to single ionization of He as a function of energy transfer ϵ by Compton scattering (20 keV, — · — · —; 60 keV, — · · — · · —; 90 keV, – – – – –) and protons (2 MeV, · · · · ·; 6 MeV, - - -; 100 MeV, ——). Experimental data are from Wu *et al.* [10] (2 MeV, ○; 3 MeV, △; 6 MeV, ▽) and from Kamber *et al.* [12] (3 MeV, +; 6 MeV, ×). R_{ph} denotes the high-energy photoionization limit 1.66% [1].

body processes. Instead, they emerge as three-body processes with the recoil of the He nucleus required to bring the scattering processes back on the energy shell, just as for photoionization. The direct link to photoionization can be shown analytically in the limit of very large but nonrelativistic energy transfers $\epsilon_{\mathrm{BE}} \ll \epsilon \ll c^2$. In this limit, the cross section for emitting a fast electron with momentum $\mathbf{k}_1$ and a slow electron with $\mathbf{k}_2$ by Compton scattering is proportional to

$$\sigma^{++} \propto \left| \int d\mathbf{r}_2\, \psi^{-*}(\mathbf{k}_2, \mathbf{r}_2) \int d\mathbf{r}_1\, e^{i(\mathbf{Q}^C_{\mathrm{BE}} - \mathbf{k}_1)\cdot \mathbf{r}_1} |i\rangle + \int d\mathbf{r}_2\, \psi^{-*}(\mathbf{k}_2, \mathbf{r}_2) e^{i\mathbf{Q}^C_{\mathrm{BE}}\cdot \mathbf{r}_2} \int d\mathbf{r}_1\, e^{-i\mathbf{k}_1\cdot \mathbf{r}_1} |i\rangle \right|^2 \quad (7)$$

Since for $\epsilon \ll \epsilon_{\mathrm{BE}}$ we have $1 \ll Q^C_{\mathrm{BE}} \ll k_1$, the first term in Eq. (7) is equivalent to the sudden approximation limit of photoionization [14,15]. Note, however, that in addition to this direct shakeoff term, there is also an "exchange shakeoff" term. For the case of 20 keV photons shown in Fig. 3, $Q^C_{\mathrm{BE}} \approx 8$. For this momentum transfer, the exchange shakeoff term is important and causes the ratio to lie above the high energy limit of photoionization $\approx$1.66%. The exchange shakeoff effect is pronounced when $Q^C_{\mathrm{BE}} \sim k_2$. It should be noted that a similar exchange shakeoff contribution is expected in photoionization, if retardation is taken into account. In the limit $Q^C_{\mathrm{BE}} \to \infty$ the exchange shakeoff term becomes negligible. Consequently, for $\epsilon_{\mathrm{BE}} \ll \epsilon \lesssim \omega$, $R_C(\epsilon)$ approaches the high-energy transfer limit which is equal to the photoionization limit of 1.66%. The same limit also holds for charged-particle scattering. We emphasize that the contribution of the region $\epsilon > \epsilon_{\mathrm{BE}}$ to the total cross section is small and is not expected to affect the integrated ratios [3,6,16]. This region is, however, of strong conceptual interest as it provides a unified description for double ionization by all three processes.

In summary, the ratios of double to single ionization of He by Compton photons and charged particles are shown to undergo three stages at large energy transfers, ϵ: a constant region of 0.86% for $1 \ll \epsilon \lesssim \epsilon_{\mathrm{BE}}$, a rapidly changing region influenced by the exchange shakeoff mechanism near $\epsilon \gtrsim \epsilon_{\mathrm{BE}}$, and the photoionization limit at $\epsilon \gg \epsilon_{\mathrm{BE}}$ for the differential (not integrated) ratios. The emerging interconnection between photons and charged particles impact can serve as guidance to further differential studies of multiple ionization processes.

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[1] J.H. McGuire, N. Berrah, R.J. Bartlett, J.A.R. Samson, J.A. Tanis, C.L. Cocke, and A.S. Schlachter, J. Phys. B **28**, 913 (1995).
[2] A. Dalgarno and H. Sadgepour, Phys. Rev. A **46**, R3591 (1992).
[3] L.R. Andersson and J. Burgdörfer, Phys. Rev. Lett. **71**, 50 (1993).
[4] J.A.R. Samson, C.H. Greene, and R.J. Bartlett, Phys. Rev. Lett. **71**, 201 (1993).
[5] K. Hino, P. Bergstrom, and J.H. Macek, Phys. Rev. Lett. **72**, 1620 (1994).
[6] T. Suric, K. Pisk, B.A. Logan, and R.H. Pratt, Phys. Rev. Lett. **73**, 790 (1994).
[7] L. Spielberger, O. Jagutzki, R. Dörner, J. Ullrich, U. Meyer, V. Mergel, M. Unverzagt, M. Damrau, T. Vogt, I. Ali, Kh. Khayyat, D. Bahr, H.G. Schmidt, R. Frahm, and H. Schmidt-Böcking, Phys. Rev. Lett. **74**, 4615 (1995).
[8] J.C. Levin, G.B. Armen, and I.A. Sellin, Phys. Rev. Lett. **76**, 1220 (1996).
[9] L. Spielberger *et al.*, Phys. Rev. Lett. **76**, 4685 (1996).
[10] W. Wu, S. Datz, N.L. Jones, H.F. Krause, B. Rosner, K.D. Sorge, and C.R. Vane, Phys. Rev. Lett. **76**, 4324 (1996).
[11] R. Wehlitz *et al.*, Phys. Rev. A **53**, 3720 (1996).
[12] E.Y. Kamber, C.L. Cocke, S. Cheng, and S.L. Varghese, Phys. Rev. Lett. **60**, 2026 (1988).
[13] G. Schiwietz, G. Xiao, P.L. Grande, B. Skogvall, R. Köhrbrück, B. Sulik, K. Sommer, A. Schmoldt, U. Stettner, and A. Salin, Europhys. Lett. **27**, 341 (1994).
[14] F.W. Byron, Jr. and C.J. Joachain, Phys. Rev. **164**, 1 (1967).
[15] T. Åberg, Phys. Rev. A **2**, 1726 (1970).
[16] L.R. Andersson and J. Burgdörfer, Phys. Rev. A **50**, R2810 (1994).
[17] M. Amusia and A. Mikhailov, J. Phys. B **28**, 1723 (1995).
[18] W. Heitler, *The Quantum Theory of Radiation* (Dover, New York, 1984).
[19] M.R.C. McDowell and J.P. Coleman, *Introduction to the Theory of Ion-Atom Collisions* (North-Holland, Amsterdam, 1970).
[20] J. Burgdörfer, L.R. Andersson, J.H. McGuire, and T. Ishihara, Phys. Rev. A **50**, 349 (1994).
[21] J.F. Reading and A.L. Ford, Phys. Rev. Lett. **58**, 543 (1987).
[22] J. Ullrich, R. Moshammer, H. Berg, R. Mann, H. Tawara, R. Doerner, J. Euler, H. Schmidt-Boecking, S. Hagmann, C.L. Cocke, M. Unverzagt, S. Lencinas, and V. Mergel, Phys. Rev. Lett. **71**, 1697 (1993).