The ratio of double to single ionization of helium: the relationship of photon and bare charged particle impact ionization

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Consider the photoionization of the He atom in its ground, $1s^2\,{}^1S$, state. The cross section for each ionization channel shall be termed $\sigma^j_{\gamma,i}(h\nu)$, where i=1 or 2, the charge state of the final ion, j designates the rest of the quantum numbers defining the ionization channel, and $h\nu$ is the photon energy. The ratio of double to single ionization of He by photons is thus given by

$$R_\gamma(h\nu) = \frac{\sum_j \sigma^j_{\gamma,2}(h\nu)}{\sum_j \sigma^j_{\gamma,1}(h\nu)} . \tag{1}$$

The channels in the sum for single ionization are all of the $n\ell\epsilon\ell'\,{}^1P$ final states, i.e., the He^+ ion left in its ground state or any discrete excited state. For double ionization the possible final channels are the $\epsilon\ell\epsilon'\ell'\,{}^1P$ states subject to the conservation of energy, $h\nu = \epsilon+\epsilon'+I_2$, where I_2 is the double ionization potential; actually then, the sum over channels in the denominator of Eq.(1) is really an integral over the possible energy sharings of the two ejected electrons plus a sum over the possible angular momenta of the individual electrons. It must be realized, however, that the designations of various states referred to signify only the configuration with the largest coefficient in a CI expansion. In fact, without CI, neither the double ionization process, nor the ionization plus excitation, would be possible since the transition operator for photoionization (in any of its forms) is a sum of single-particle operators; under the action of such an operator, two electrons changing state are not possible. The major contributions to the double ionization cross section arise from $1s\epsilon s\,{}^1S$ and $(n,\epsilon)s\epsilon's\,{}^1S$ mixing with the dominant $1s^2\,{}^1S$ configuration of the initial state, and $1s\epsilon p\,{}^1P$ mixing with the dominant $\epsilon s\epsilon'p\,{}^1P$ configuration of the final state.

For later use, it is of interest to cast the ratio in terms of differential oscillator strength, i.e.,

$$\sigma^j_{\gamma,i} \equiv 4\pi^2\alpha a_0^2 \frac{df^j_i(\Delta E)}{d(\Delta E)} \tag{2}$$

where α is the fine structure constant, a_0 is the Bohr radius, and ΔE (=$h\nu$) is the energy transferred to the atom. In terms of the oscillator strength, then, the double to single photoionization ratio [Eq. (1)] can be written as

$$R_\gamma(h\nu) = \frac{\sum_j df_2^j(\Delta E=h\nu)/d(\Delta E)}{\sum_j df_1^j(\Delta E=h\nu)/d(\Delta E)} . \qquad (3)$$

Now consider the single and double ionization of He by fast bare charged particles of charge z, mass M and velocity v. Define the reduced incident energy $T = \frac{1}{2}mv^2$, where m is the electron mass. The cross section for each ionization channel, differential in the energy transfer ΔE, will be written as $d\sigma^{j,T}_{i,z,M}(\Delta E)/d(\Delta E)$, where i and j are as in the photoionization case. The ratio of double to single ionization by charged particles, as a function of energy transfer ΔE is, thus given by

$$R^T_{z,M}(\Delta E) = \frac{\sum_j d\sigma^{j,T}_{2,z,M}(\Delta E)/d(\Delta E)}{\sum_j d\sigma^{j,T}_{1,z,M}(\Delta E)/d(\Delta E)} . \qquad (4)$$

The sums in Eq.(4) are over all channels, just as in Eq.(1). It is important to note that for charged particle impact, charge transfer processes can also produce singly and doubly charged target ions. The cross sections dealt with here are only for the ionization process, however.

For reasonably high incident energy of the charged particle, the first Born approximation is applicable [1]. Furthermore, for small energy transfer, the Born cross section can be expanded in T, the reduced incident energy, as [1]

$$\frac{d\sigma^{j,T}_{i,z,M}(\Delta E)}{d(\Delta E)} = \frac{4\pi a_0^2 z^2}{T/R}\left[\left(\frac{R}{\Delta E}\right)\frac{df_i^j(\Delta E)}{d(\Delta E)}\ln\left(\frac{4T}{R}\right)+B_i^j(\Delta E)\right]+O\left(\frac{R^2}{T^2}\right) \qquad (5)$$

where R = 13.6 eV and $B_i^j(\Delta E)$ is a function of target properties only. When $\Delta E/T \ll 1$, retaining only the first two terms of Eq.(5) is an excellent approximation. In such a case, the ratio of Eq.(4) is independent of z and M, the charge and mass of the incident projectile, and can be written as

$$R^T(\Delta E) = \frac{\sum_j [(R/\Delta E)df_2^j(\Delta E)/d(\Delta E)\ln(4T/R)+B_2^j(\Delta E)]}{\sum_j [(R/\Delta E)df_1^j(\Delta E)/d(\Delta E)\ln(4T/R)+B_1^j(\Delta E)]} \qquad (6a)$$

$$= R_\gamma(\Delta E)\,\frac{1+\sum_j B_2^j(\Delta E)/\sum_j[(R/\Delta E)df_2^j(\Delta E)/d(\Delta E)\ln(4T/R)]}{1+\sum_j B_1^j(\Delta E)/\sum_j[(R/\Delta E)df_1^j(\Delta E)/d(\Delta E)\ln(4T/R)]} \tag{6b}$$

$$= R_\gamma(\Delta E)F(\Delta E,T)\ . \tag{6c}$$

Now the function $F(\Delta E,T)$ is of order unity, but the details are target-dependent. In the limit of such high T that $\ln(4T/R)$ becomes large enough to allow the second term in the numerator and the denominator of Eq.(6b) to be neglected, i.e., $F(\Delta E,T)\rightarrow 1$, one obtains the simple relation

$$R^T(\Delta E) = R_\gamma(\Delta E) \tag{7}$$

independent of T. In this limit then, the ratio of double ionization to single charged particle impact ionization at a particular energy loss, ΔE, is independent of the incident particle velocity (energy), mass or charge and equal to the ratio for photon impact at photon energy $h\nu = \Delta E$. It is important to emphasize that the charged particle ratio that is related to the photoionization is for a **fixed** energy transfer, ΔE, for both the single and double ionization process. This is **not** the same thing as a ratio of electrons of a given energy resulting from the single and double ionization by charged particle impact.

Up to this point, we have considered only non-relativistic kinematics. It is of interest, however, to inquire as to what modifications are introduced if the incident charged particle is relativistic. In the relativistic range, with T replaced by its definition ($\frac{1}{2}mv^2$) and introducing $\beta=v/c$, Eq.(5) becomes [1]

$$\frac{d\sigma^{j,T}_{i,z,M}(\Delta E)}{d(\Delta E)} = \frac{8\pi a_0^2 z^2}{mv^2/R}\left[\left(\frac{R}{\Delta E}\right)\frac{df_i^j(\Delta E)}{d(\Delta E)}\left\{\ln\left(\frac{2mv^2}{R}\right)-\ln(1-\beta^2)-\beta^2\right\}+B_i^j(\Delta E)\right]+O\left(\frac{1}{v^4}\right) \tag{8}$$

where the essential difference from Eq.(5) is the addition of the two β-dependent terms; thus, the ratio of Eq.(6b) is modified to

$$R_T(\Delta E) = R_\gamma(\Delta E)\,\frac{1+\sum_j B_2^j(\Delta E)/\sum_j[(R/\Delta E)df_2^j(\Delta E)/d(\Delta E)\{\ln(2mv^2/R)-\ln(1-\beta^2)-\beta^2\}]}{1+\sum_j B_1^j(\Delta E)/\sum_j[(R/\Delta E)df_1^j(\Delta E)/d(\Delta E)\{\ln(2mv^2/R)-\ln(1-\beta^2)-\beta^2\}]} \tag{9}$$

where the only change is seen to be the replacement of $\ln(4T/R)$ by $\ln(2mv^2/R)-\ln(1-\beta^2)-\beta^2$.

Thus, relativistic incident charged particles do not alter the non-relativistic results is any significant way; at high enough incident energy, Eq.(7) still holds.

For large energy transfer, where ΔE/T is no longer small compared to unity, the Bethe-Born expansion no longer valid, but the Born approximation is still applicable. In such a case, the dipole part of the interaction no longer dominates; in fact it contributes only a relatively small amount to the cross section. Thus many final states other than the optically allowed states discussed above are allowed and important correlations, configurations which mix with the main configuration, differ in detail from the photon case. Thus, while one would expect effects of the same order of magnitude, there is no direct relationship between the photon case and the charged particle case when ΔE/T is not small. However, recent experimental data [2], shown in Fig. 1, seems to suggest that even in the large energy transfer range, the ratio of double to single ionization of He is substantially the same as the photon case.

The ratio of total single and double ionization cross sections of He, integrated over energy transfer, ΔE, for incident bare charged particles can, however, be related to the photon ratio. This is because these total cross sections are dominated by the small energy transfer region where there is a relationship to the photon case, as has been shown above. The ratio can be written as

$$R^T_{z,M} = \frac{\sigma^T_{2,z,M}}{\sigma^T_{1,z,M}} = \frac{\sum_j \int [d\sigma^{j,T}_{2,z,M}(\Delta E)/d(\Delta E)]d(\Delta E)}{\sum_j \int [d\sigma^{j,T}_{1,z,m}(\Delta E)/d(\Delta E)]d(\Delta E)} . \tag{10}$$

Using Eq.(4) then, this can be rewritten as

$$R^T_{z,M} = \frac{\sum_j \int [R^T_{z,M}(\Delta E)d\sigma^{j,T}_{1,z,M}(\Delta E)/d(\Delta E)]d(\Delta E)}{\sum_j \int [d\sigma^{j,T}_{1,z,m}(\Delta E)/d(\Delta E)]d(\Delta E)} . \tag{11}$$

Thus, the total cross section ratio is a weighted average of the differential cross section ratio, the weighting factor being the differential cross section for single ionization over the total cross section for single ionization. Of crucial importance in Eq.(11) is the fact that although the lower limit of integration is ΔE = 24.58 eV, the ionization potential of He, the ratio $R^T_{z,M}(\Delta E)$ in the integral in the numerator of Eq.(11) is **zero** below the double ionization potential of He, ΔE = 79.98 eV. Then, since most of the total single ionization cross section comes from the small ΔE region, it is evident that the ratio of Eq.(11), the weighted average, will be much smaller than $\bar{R}_{z,M}(\Delta E)$ of Eq.(4) because of the heavy weighting of the ΔE region where $R^T_{z,M}(\Delta E)$ vanishes.

If the incident energy is great enough so that Eqs.(6) apply, Eq.(11) becomes

$$R^T_{z,M} = \frac{\sum_j \int [R_\gamma(\Delta E) F(\Delta E, T) d\sigma^{j,T}_{1,z,M}(\Delta E)/d(\Delta E)] d(\Delta E)}{\sum_j \int [d\sigma^{j,T}_{1,z,m}(\Delta E)/d(\Delta E)] d(\Delta E)} . \quad (12)$$

Note that, strictly speaking, the integrand in the numerator of Eq.(12) is only correct for small $\Delta E/T$. However, since the contribution of small $\Delta E/T$ dominates the integral, i.e., the total single ionization cross section is dominated by low energy electrons, this is a very minor approximation. Finally, at high enough incident energies so that Eq.(7) applies, this ratio becomes

$$R^T_{z,M} = \frac{\sum_j \int [R_\gamma(\Delta E) d\sigma^{j,T}_{1,z,M}(\Delta E)/d(\Delta E)] d(\Delta E)}{\sum_j \int [d\sigma^{j,T}_{1,z,m}(\Delta E)/d(\Delta E)] d(\Delta E)} \quad (13)$$

which depends only upon the photon ratio, $R_\gamma(\Delta E)$, and the shape of the differential single ionization cross section, i.e., the ratio of the differential single ionization cross section to the total single ionization cross section. This latter ratio approaches a limit for high enough T as can easily be demonstrated by the use of the Bethe-Born expansion. Thus, the ratio of total cross sections for double and single ionization, $R^T_{z,M}$ also approaches a limit for high enough T. That it approaches a limit is verified by the experimental data [3] presented in Fig. 2, which shows the ratio for a number of different charged particles over a broad range of energies. In addition, it is seen from Fig. 2 that the limit of the ratio of about 0.28 % is indeed much smaller than the asymptotic ratio for photoionization of 1.67 % [4], just as predicted above and previously [5].

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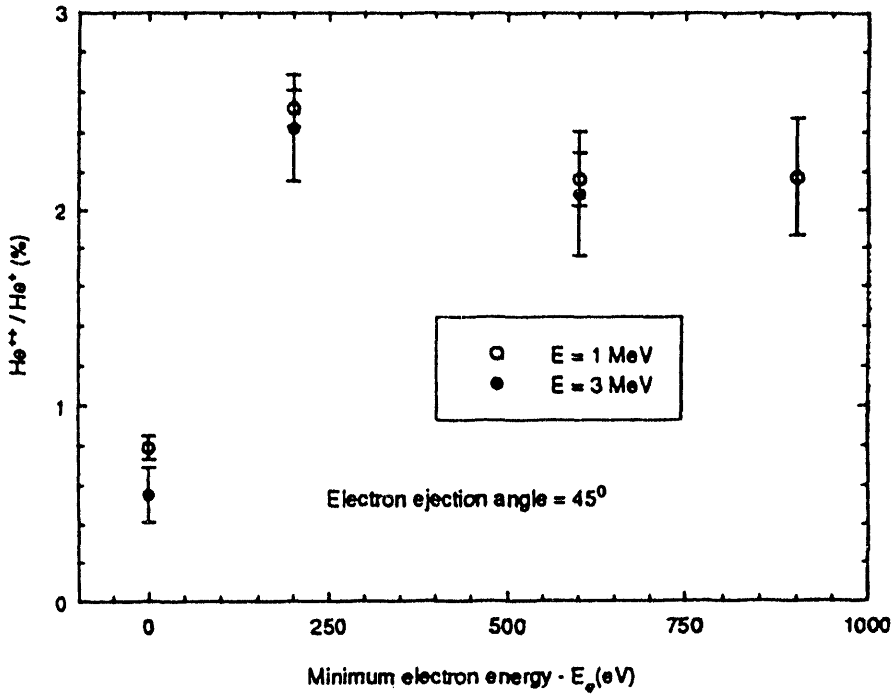

Figure 1. *Ratio of double to single ionization of He for events which generate electrons with energy exceeding a minimum value, for 1 and 3 Mev protons, from Ref. 2.*

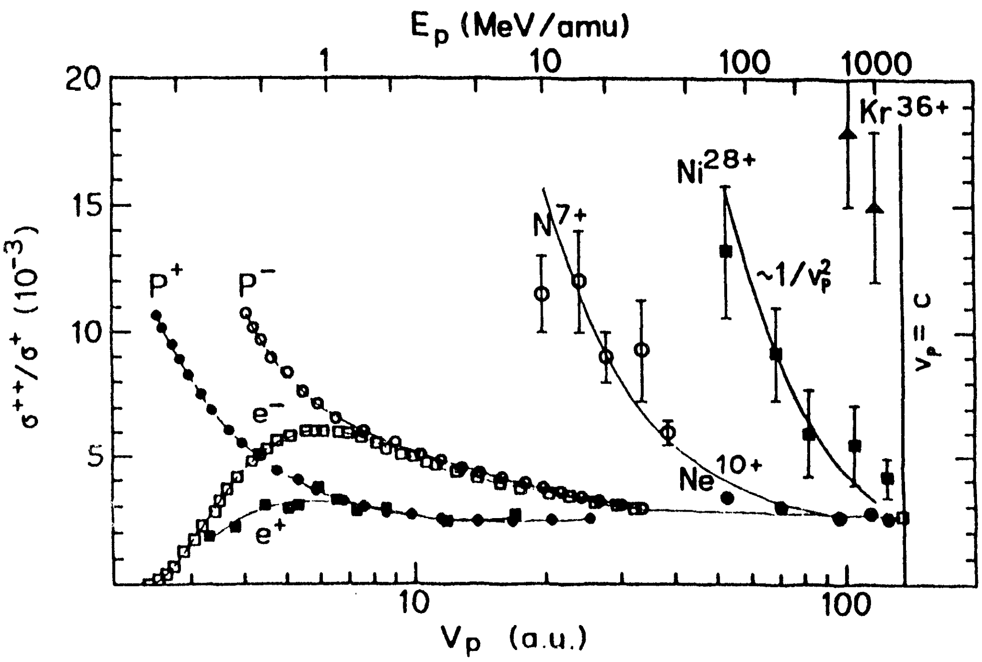

Figure 2. *Ratio of double to single ionization of He for different projectiles as a function of projectile velocity (lower scale) and energy (upper scale) from Ref. 3. The open square at the far right is for electron impact.*