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Consistency test of double ionization of helium by photons and charged particles

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We have performed a consistency test of recent data for the ratios of cross sections of double to single ionization of helium by photons and by fast charged particles. It is based on a relationship that connects the dipole and nondipole components of cross sections by charged particle impact to photoionization and Compton scattering, respectively. We find the high energy ratio R_Z for charged particles to be a sensitive probe of the photoionization data near threshold. Using recent experimental data for photoionization, the asymptotic ratio R_Z is found to range from 0.240% to 0.295%, as compared with direct experimental measurement of 0.26%. [S1050-2947(96)08807-5]

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I. INTRODUCTION

Many-electron transitions as exemplified in double ionization of helium continue to challenge our understanding of the role of dynamic electron-electron interaction. In photoionization, double ionization of He occurs due to electron-electron correlation in the initial and final states [1]. The ratio, R_{PE}, of cross sections of double to single photoionization of He is a quantity most often studied. It is generally accepted that this ratio will approach a limit in the high energy limit. Both experiment and theory seem to establish the constant to be about $R_{PE} = 1.6 - 1.7\,\%$ [1]. In contrast the asymptotic ratio, R_Z, for light charged particles is smaller by about a factor of 6.4.

Not as clear is the ratio in the energy range from threshold at 79 eV to the maximum $\sim$ 200 eV. Recent experiments using synchrotron radiation have revealed considerable discrepancies in this region [2–6]. Theoretical calculations [7–13] also disagree among each other, making an assessment of the situation difficult. Differences are observed at the maximum and also very near threshold which are masked by a steep rise in the ratio. Final state correlation effects are presumably greater here since the two electrons slowly recede from each other. Understanding of these effects requires one to first sort out possible inconsistencies in the data.

In this paper, we perform a check of the mutual consistency of data for double ionization by photons against that by charged particles. We use the relationship proposed recently that separates the dipole and nondipole components of cross sections by charged particle impact [14]. In this relationship the ratio of double to single ionization cross sections by charged particles is expressed as a convolution in terms of photoionization and Compton scattering. The ratio for charged particles has also been investigated both experimentally and theoretically over a wide energy range [15–18]. Unlike photoionization, high velocity ratios for light charged particles such as protons and electrons are believed to be less ambiguous and converge to a value near 0.26% [1]. Checking against this well established value will give us an independent way to analyze the consistency of the available experimental and theoretical data. We find that the ratio of charged particles to be a sensitive probe for the ratios of photonization from threshold to the maximum.

II. THEORY

The relationship between ionization by photons and charged particles has been described in detail elsewhere [19,20,14]. In the following we present a brief summary [14]. The relationship is based on the recognition that both dipole and nondipole transitions are allowed for charged particles. The dipole term is usually large at small energy transfers. The nondipole terms become important for increasing energy transfers. Although in principle the dipole part will eventually dominate over the nondipole part as $\ln v^2$, this logarithmic dominance is too slow to be realized in practice for finite v. Both the dipole and nondipole terms, therefore, need to be included.

The double ionization cross section differential in energy transfer ϵ, $d\sigma_Z^{++}(\epsilon)/d\epsilon$, by charged particles may be expressed in terms of partial wave components L as

$$\frac{d\sigma_Z^{++}(\epsilon)}{d\epsilon}=\sum_L \frac{d\sigma_{Z,L}^{++}}{d\epsilon}=\frac{d\sigma_{Z,1}^{++}}{d\epsilon}+\sum_{L\neq 1}\frac{d\sigma_{Z,L}^{++}}{d\epsilon}. \qquad (1)$$

It is recognized from (1) that the first term ($L=1$) corresponds to the dipole (D) component and the second term ($\Sigma_{L\neq 1}$) to nondipole (ND) components. Setting

$$\frac{d\sigma_{Z,D}^{++}(\epsilon)}{d\epsilon}\equiv\frac{d\sigma_{Z,1}^{++}}{d\epsilon},\quad \frac{d\sigma_{Z,ND}^{++}(\epsilon)}{d\epsilon}\equiv\sum_{L\neq 1}\frac{d\sigma_{Z,L}^{++}}{d\epsilon},$$

Eq. (1) may be rewritten as

$$\frac{d\sigma_Z^{++}(\epsilon)}{d\epsilon}=\frac{d\sigma_{Z,D}^{++}(\epsilon)}{d\epsilon}+\frac{d\sigma_{Z,ND}^{++}(\epsilon)}{d\epsilon}. \qquad (2)$$

We now define the dipole and nondipole ratios denoted by $R_{Z,D}$ and $R_{Z,ND}$, respectively, as

$$R_{Z,D}(\epsilon)=\frac{d\sigma_{Z,D}^{++}(\epsilon)}{d\epsilon}\Bigg/\frac{d\sigma_{Z,D}^{+}(\epsilon)}{d\epsilon}, \tag{3}$$

$$R_{Z,ND}(\epsilon)=\frac{d\sigma_{Z,ND}^{++}(\epsilon)}{d\epsilon}\Bigg/\frac{d\sigma_{Z,ND}^{+}(\epsilon)}{d\epsilon}, \tag{4}$$

where $d\sigma_{Z,D}^{+}(\epsilon)/d\epsilon$ and $d\sigma_{Z,ND}^{+}(\epsilon)/d\epsilon$ are the corresponding dipole and nondipole components of single ionization cross section by charged particles. In terms of the dipole and nondipole ratios (3) and (4), Eq. (2) may be expressed as

$$\frac{d\sigma_{Z}^{++}(\epsilon)}{d\epsilon}=R_{Z,D}(\epsilon)\frac{d\sigma_{Z,D}^{+}(\epsilon)}{d\epsilon}+R_{Z,ND}(\epsilon)\frac{d\sigma_{Z,ND}^{+}(\epsilon)}{d\epsilon}. \tag{5}$$

From (5) it is now straightforward to obtain the ratio R_Z of double to single ionization by charged particles

$$R_Z=\sigma_Z^{++}/\sigma_Z^{+}=\int_{I_2}^{\infty}\frac{d\sigma_Z^{++}}{d\epsilon}d\epsilon\Bigg/\int_{I_1}^{\infty}\frac{d\sigma_Z^{+}}{d\epsilon}d\epsilon$$
$$=\int_{I_2}^{\infty}R_{Z,D}(\epsilon)\rho_{Z,D}^{+}(\epsilon)d\epsilon+\int_{I_2}^{\infty}R_{Z,ND}(\epsilon)\rho_{Z,ND}^{+}(\epsilon)d\epsilon, \tag{6}$$

where $\rho_{Z,D}^{+}(\epsilon)$ and $\rho_{Z,ND}^{+}(\epsilon)$ are, respectively, the dipole and nondipole normalized energy distributions (i.e., normalized energy differential cross sections) for single ionization by charged particles given by

$$\rho_{Z,D}^{+}(\epsilon)=\frac{d\sigma_{Z,D}^{+}}{d\epsilon}\Bigg/\int_{I_1}^{\infty}\frac{d\sigma_Z^{+}}{d\epsilon}d\epsilon, \tag{7}$$

$$\rho_{Z,ND}^{+}(\epsilon)=\frac{d\sigma_{Z,ND}^{+}}{d\epsilon}\Bigg/\int_{I_1}^{\infty}\frac{d\sigma_Z^{+}}{d\epsilon}d\epsilon. \tag{8}$$

The first and second ionization potentials of helium are I_1 (24.6 eV) and I_2 (79 eV), respectively.

It may be shown that, within the first Born approximation, the dipole and nondipole ratios for charged particles are related to the ratios for photoionization and Compton scattering by [7,21,14]

$$R_{Z,D}(\epsilon)=R_{PE}(\epsilon),\quad R_{Z,ND}(\epsilon)\simeq R_C(\epsilon), \tag{9}$$

where R_{PE} and R_C are the ratios of double to single ionization for photoionization and Compton scattering, respectively. Therefore, the ratio of double to single ionization (6) for charged particles may be expressed in terms of R_{PE} and R_C as

$$R_Z\simeq\int_{I_2}^{\infty}R_{PE}(\epsilon)\rho_{Z,D}^{+}(\epsilon)d\epsilon+\int_{I_2}^{\infty}R_C(\epsilon)\rho_{Z,ND}^{+}(\epsilon)d\epsilon. \tag{10}$$

Thus, R_Z is given by an integral [Eq. (10)] which weighs separately the dipole photoionization ratio R_{PE} by the energy distribution of dipole transitions and the Compton ratio R_C by the energy distribution of all other multipole transitions.

One key feature of the relationship (10) is that we may obtain predictions for the ratios by charged particles from known experimental or theoretical ratios by photons with relative ease. This approach does not rely on an explicit description of two-electron transitions. The latter are taken into account through experimental (or theoretical) data for photoionization. All that is required is a calculation of the dipole and nondipole energy distributions for single ionization by charged particles. This calculation is much less difficult than double ionization by charged particles. This method, therefore, allows for a detailed consistency check of completely different sets of experimental or theoretical data with a minimum of theoretical assumptions.

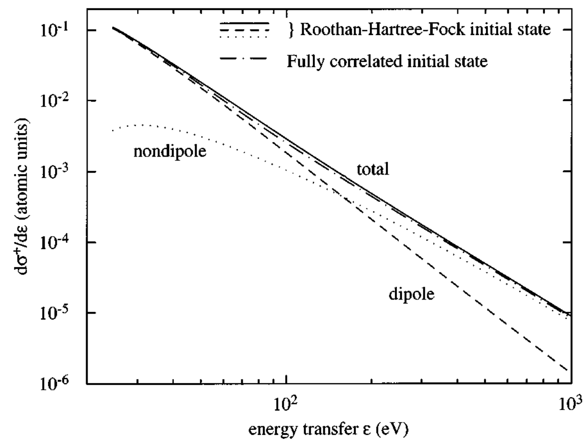

FIG. 1. (a) Single ionization cross section (energy distribution) as a function of energy transfer by 20 MeV proton + He using two initial-state wave functions (see text). Roothan-Hartree-Fock initial state: total (dipole + nondipole)—solid line; dipole term ($l=1$)—dashed line; nondipole terms ($\Sigma_{l\neq 1}$)—dotted line. Fully correlated configuration-interaction initial state: total—dash-dotted line. The threshold for single ionization is at 24.6 eV.

III. RESULTS AND DISCUSSION

The energy distributions for single ionization of He by 20 MeV protons are shown in Fig. 1. They are calculated in the first Born approximation [22,14] by partial wave expansion. A five-term Roothan–Hartree-Fock wave function [23] is used for the initial state of He. The final state is a Coulomb wave. To check the effect of electron correlation in the initial state on single ionization, we performed another calculation using fully correlated initial state. Thirty-five configuration-interaction terms up to $5g$ of the ^1S configuration are included in the state, reproducing the ground state energy to a relative accuracy better than 4×10^{-4}. The calculation is also shown in Fig. 1. The results from using the Roothan–Hartree-Fock wave function and the correlated wave function are within 5% of each other, showing that the single ionization cross section (unlike double ionization) is insensitive to electron correlation in the initial state at high energies.

The energy distributions show several universal features found in single ionization by fast charged particles. The small energy transfer region is dominated by dipole transitions, while nondipole transitions become important for in-

TABLE I. The dipole and nondipole terms of single ionization cross section of He by 20 MeV protons. The sum of the two components yields the total cross section. The number in brackets indicates the exponent.

Energy transfer (eV)	Cross section (a.u.)	
	Dipole term	Nondipole terms
24.6	1.06 [-1]	3.79 [-3]
26.6	8.79 [-2]	4.31 [-3]
30.6	6.08 [-2]	4.51 [-3]
38.6	3.19 [-2]	4.07 [-3]
54.6	1.16 [-2]	2.76 [-3]
86.6	2.85 [-3]	1.34 [-3]
150.6	5.12 [-4]	4.85 [-4]
278.6	7.39 [-5]	1.38 [-4]
534.6	9.57 [-6]	3.26 [-5]

creasing ϵ and ultimately dominate at large ϵ. The distribution falls off rapidly as a function of energy transfer. The total cross section integrated over all ϵ is clearly determined by a small interval of energy transfer just above the ionization threshold. As a result, the total cross section can be accurately obtained by including a few partial waves, typically between 0 and 2. However, the differential cross section for larger energy transfers requires many partial waves to converge. We find seven partial waves to be adequate for energy transfers up to 1 keV. Since the convolution (10) starts upward from the second ionization threshold of 79 eV, a substantial part of the cross section where dipole transitions is dominant is effectively cut off. At the double ionization threshold the nondipole terms already comprise 30% of the cross section. At a higher energy transfer, the dipole and nondipole cross sections cross each other. The crossing point ϵ_c should move to higher energy transfers as the projectile speed is increased, reflecting the logarithmic dominance of the dipole term over the nondipole terms. However, the energy dependence of the crossing point is very slow. For instance, when the proton energy is doubled from 20 MeV to 40 MeV, ϵ_c shifts up by only 4 eV. It appears that for protons of energies greater than 10 MeV, no experimental data are available from which dipole and nondipole terms may be extracted and compared with theory. We list in Table I the numerical values of the energy distributions which may be important when convolutions based on different theoretical input for ρ are compared.

In Fig. 2 we show the results for the ratio of double to single ionization of He by protons from the convolution of the energy distributions of single ionization with ratios by photonization and Compton scattering. In addition to the existing data set available prior to 1995 [14], we have used four sets of new photonization R_{PE} data: data by Levin *et al.* [4], by Dörner *et al.* [5], and two sets by Samson *et al.* [6]. We use the same Compton ratio R_C which is unchanged from the previous calculation [24,25]. Since then, one experimental data point that unambiguously identifies Compton scattering became available which agreed very well with the calculation in Ref. [25]. The differences seen in Fig. 2 are exclusively due to the changes in R_{PE} entering the dipole component.

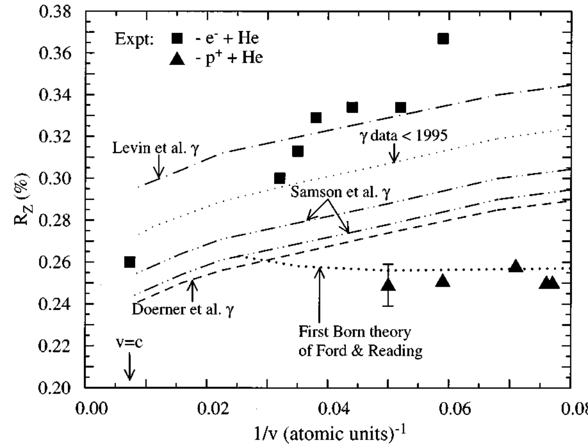

FIG. 2. Ratio, R_Z, of double to single ionization of He by protons as a function of the inverse impact speed $1/v$. Starting from the top, the theory curves represent results using the following: photoionization (γ) data by Levin *et al.* [4] (dash-dot line); γ data before 1995 [14] (thin dotted line); γ data sets of Samson *et al.* [6] (dash-dot-dot line) taken at different facilities, ALS (upper one) and BNLS (lower one); γ data by Dörner *et al.* [5] and theoretical values of Tang and Shimamura [13] (dashed line); direct first Born calculation by Ford and Reading including partial waves s, p, and d [26] (thick dotted line). Symbols: Experimental data for electrons (squares) and protons (triangles) from Refs. [15,16]. The arrow marks the speed of light c.

The use of R_{PE} data by Levin *et al.* reveals that the ratio R_Z is enhanced rather than reduced when compared to the original results. It appears at first glance to be surprising since the new R_{PE} data are significantly lower than the original data near the maximum. Closer inspection shows that very near the threshold ($\sim$100 eV), this data set is higher than the original set. Since the energy distribution for single ionization decreases rapidly for increasing ϵ, the R_{PE} data around the threshold is more heavily weighted. In this case, higher R_{PE} data near the threshold more than offset the lowering due to smaller R_{PE} at the maximum. On the other hand, the data of Dörner *et al.* are in accord with the original set near threshold but even lower than the data of Levin *et al.* near the maximum. They result in an overall reduction of R_Z, but less than the reduction of R_{PE} near the maximum would suggest, for the reason just discussed. The theoretical data of Tang and Shimamura give values of R_Z (not shown in Fig. 2) very close to the experimental data of Dörner *et al.*, and are almost indistinguishable from the latter on the scale of Fig. 2. The two sets of data by Samson *et al.*, taken at different facilities and times [Advanced Light Source (ALS) and National Synchrotron Light Source (NSLS)], give results in between the other two experimental data sets. The difference reflects the fluctuations between the two data sets by Samson *et al.* near 150 eV [6].

The high-velocity ratio of R_Z at 400 MeV is found to be 0.295%, 0.240%, 0.254%, and 0.244%, respectively, using the four sets of new experimental data. They are to be compared with the value of 0.26% [15,16]. Assuming this accepted number to be accurate within less than 10%, it would indicate that the R_{PE} data of Levin *et al.* might be too high in the immediate region above threshold. It would also indicate that the data of Dörner *et al.* may represent the lower bound of the accepted value. The data of Samson *et al.* give results in between the other two data sets and are closer to the asymptotic value of R_Z. Recent theoretical photoionization data have also been used as input into calculating R_Z at 400 MeV. The data by Tang and Shimamura [13] yield 0.240%, and the data by Meyer and Greene [12] in the acceleration and velocity gauges give 0.223% and 0.254%, respectively. The energy range covered by the data of Proulx and Shakeshaft [10] is insufficient for input into the convolution.

We emphasize that the important characteristics of the ratio R_Z include not only the asymptotic value of R_Z, but also the way in which it is approached. As has been noted before [14], the dipole contribution [first term in Eq. (10)] is rather flat across the energy region. The downward trend as $1/v \rightarrow 0$ in our theoretical results is mostly due to the nondipole contribution [second term in Eq. (10)]. Within the validity of the first Born approximation, the R_Z values reconstructed from photoionization and Compton scattering data are correct only to the order Z^2. They should lie in between R_Z's for protons and electrons, thereby averaging out the sign dependence of higher order Z^3 contributions to charged particle impact. The experimental data in Fig. 2 show a considerable Z^3 effect even at $1/v = 0.05$ (10 MeV/u). At this energy, the nondipole contribution is comparable to the Z^3 effect. The latter is expected to be negligibly small for $1/v < 0.03$ ($>$ 20 MeV/u) [18], while the former will decrease very slowly as $1/\ln v$ [14].

There are several sources of uncertainty in our analysis. Apart from interpolation error, the accuracy in R_{PE} is limited by experimental error bars. Values of R_C are still being actively pursued both experimentally and theoretically. There is also some uncertainty in the calculation of $\rho_Z^+(\epsilon)$. Considering these uncertainties, we estimate the error to be $\sim$10–20 % for our overall results. However, we stress that while this error could shift the absolute values, it will not eliminate the observed difference when different sets of photoionization data are used as input. Also, possible uncertainties do not affect the the fact that our results lie in between the data for positively and negatively charged particles, as expected from a theory correct to order Z^2.

IV. SUMMARY

In summary, we have investigated the consistency of photoionization data against charged particle data. We find the latter to be very sensitive to the former in the near threshold region. We suggest that the photoionization data in this region should be further investigated experimentally and theoretically.

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