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Double ionization of a helium–like atom or ion by a high energy photon

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Abstract

We review recent progress in calculations of the ratio of double to single photoionization of helium–like atoms and ions at high energies. The importance of relativistic effects is discussed, with particular emphasis on recent relativistic calculations of double ionization of helium–like atoms and ions by Compton scattering. These calculations, based on the sudden approximation or the impulse approximation, show that, for low Z elements, relativistic effects do not alter the high energy limit of the Compton double to single ratio R_C, even though they do affect single and double ionization Compton scattering separately.

I. INTRODUCTION

Double ionization of two electron atoms and ions by one photon is attracting significant attention both experimentally and theoretically. The motivation is the investigation of electron–electron correlation effects. The interest in the use of photons in ionizing two electron systems (particularly helium) is based on the observation that the interaction between electrons and the radiation field is described by a one–body operator. This means that, in the lowest order for a given ionization process, a photon interacts with only one electron. Hence the simultaneous ejection of two electrons is due to the electron–electron interaction between the two electrons, and therefore double ionization by a photon provides a clear probe of electron–electron correlation. In a two electron system no other correlations are present. For this simplest many–body system we may be able to develop more exact theoretical approaches and more detailed experimental methods. We wish to understand eventually all possible correlation effects in this relatively simple system, with the purpose of better understanding of these effects in more complex systems.

The photoionization processes are photoeffect (incident photon absorbed) and Compton scattering (incident photon scattered). Photoeffect dominates the total cross section at lower photon energies and its relative contribution in photon atom interaction decreases as photon energy increases. For a He–like system, at energies for which the incoming photon momentum k is comparable to the average momentum of the bound electrons p_{av} (of a two electron atom or ion in its ground state), photoeffect and Compton scattering are comparable. For higher energies Compton scattering dominates. In the case of helium Compton scattering dominates above, roughly, 6.5 keV.

The simplest observable, for both experimental and theoretical treatment of the correlation effects, has been the ratio between probabilities for double and single photoionization. This ratio is nonzero because of electron–electron correlation; its value depends on incident photon energy. Also, the value depends on the process under consideration. Therefore we talk about the Compton double to single ionization ratio R_C and the photoeffect ionization ratio R_P. The energy dependence of these observables, and particularly their high energy limits, are subjects of extensive studies. In the last several years significant progress has been achieved [1]. However, the relatively large disagreements in theoretical predictions as well as in experimental data indicate that we still do not understand all relevant aspects of the processes which are involved in determining the ratio for the double to single ionization for both Compton scattering and photoeffect.

As we will see, much understanding of these processes can be obtained within the shake–

off approximation, *i.e.* a sudden approximation. By shake–off approximation we mean the assumption that the final state electron–electron interaction can be neglected, and that one electron scatters the high energy photon and accepts almost all momentum transfer [2].

II. PHOTOEFFECT RATIO R_P

Until recently it was believed that the energy dependence of R_P for helium was well understood over the whole nonrelativistic energy region. The theoretical predictions were consistent with measurements showing that R_P increases with energy, reaching a maximum (R_P=0.05) at photon energies of $\omega = 150 - 250$ eV, and then decreases to the constant shake–off value $R_P = 0.017$ for energies above roughly 3 keV.

New measurements [3], performed using cold target recoil spectroscopy, have found the peak of R_P to be lower than previously assumed, by about 25 %. These new data are consistent with calculations [4] relating the photoeffect ratio to that for fast charged particle impact.

The shake–off ratio is given in nonrelativistic calculations [5–8] as

$$R_P = 1 - \frac{\sum_B \left| \int \Phi_B^*(\mathbf{r}_1) \Psi_i(\mathbf{r}_1, 0) d^3\mathrm{r}_1 \right|^2}{\int \left| \Psi_i(\mathbf{r}_1, 0) \right|^2 d^3\mathrm{r}_1}. \tag{1}$$

Here $\Psi_i(\mathbf{r_1}, \mathbf{r_2})$ represents the initial state wave function, $\Phi_B(\mathbf{r}_1)$ is a bound state hydrogen–like electron wave function (in the potential of charge Z), and the summation is over all bound states. The shake off formula Eq. (1) has usually been derived in the nonrelativistic dipole approximation without retardation. It reflects the fact that, in the dipole approximation, the photon can be absorbed only if nucleus takes additional momentum (note that one of the electrons in Eq. (1) is removed from near the nucleus, $\mathbf{r}_2 = 0$). Namely, in the dipole approximation, a two–electron system, in the absence of nuclear field, cannot absorb a photon. Most theoretical analysis uses the dipole approximation. However, the shake–off result Eq. (1) is not changed if retardation effects (*i.e.* higher multipole effects) are included, given that for low Z two–electron systems retardation has little effect on ground state wave function Ψ_i and hydrogenic bound states Φ_B. In explicit calculations, using wave functions consistent with shake–off assumptions, Kornberg and Miraglia [9] have shown that, even when retardation alters the cross sections for single and double photoionization by significant factors, the same factor enters both processes, and so the ratio R_P is unchanged.

We may likewise expect that relativistic effects do not alter the photoeffect ratio R_P, based on the shake–off argument. The shake–off process, when applied to double photoionization, can be viewed as a two–step process. In the first step one electron is suddenly removed, corresponding to the single ionization process. Here we may allow very large, even relativistic, energies and therefore relativistic effects can be important. The second step is shake–off of the other electron which is (at least for small Z) most often a slow, nonrelativistic electron. Calculating the ratio between double and single photoionization cross sections, under the assumption of the validity of the sudden approximation, the contributions of the first step (the single ionization cross section), including relativistic as well as retardation effects, cancel out completely, leaving R_P as in Eq. (1).

However, at high energies, where the shake–off mechanism was assumed to be valid, Drukarev [10,11] has called attention to an additional mechanism [12–14] for double photoeffect, which would first manifest itself as a linear rise of the ratio with energy, causing a roughly 10% correction to the shake–off limit for R_P at about 12 keV. This prediction awaits experimental verification. The point is that, while a photon cannot be absorbed by one free electron in the absence of the atomic nucleus (and momentum must be transfered to the atomic nucleus), a photon <u>can</u> be absorbed by <u>two</u> free electrons (though not in the dipole approximation). This leads to a mechanism for double ionization which is not available for single ionization. The analysis was performed within nonrelativistic quantum mechanics without assuming dipole approximation, therefore allowing two electrons to absorb the photon even in the absence of the atomic nucleus [15].

A pair of photoelectrons produced through this alternative mechanism share the photon energy nearly equally (and we may call this mechanism an "equal energy sharing mechanism"). The residual ion is left with small momentum. This is different from the case when photoelectrons are produced through the shake–off mechanism, in which one electron takes almost all the photon energy and the residual ion takes almost all transfered momentum. Note that in the experimental situation where photoeffect events are distinguished from Compton events by measuring the recoil of the residual ion [16,17], the events produced through the equal energy sharing mechanism are not counted in photoeffect events, but in Compton events. These experiments count as photoeffect events only those produced in the shake–off regime (one electron takes almost all available energy while the other shakes–off), and therefore such experiments would not observe the term in R_P caused by the equal energy sharing mechanism.

Even though the shake–off ratio may therefore not be the high energy limit of R_P, it is a physically well–defined quantity which we may measure. This observable, which we may call the shake-off ratio $R_P^{s.o.}$, gives us information about the initial state electron–electron correlation when one of the electrons is near the nucleus [18,19].

Likewise, if the equal energy sharing mechanism is of the estimated magnitude, it may be within the scope of today's experimental methods. Its measurement should provide information on electron–electron correlation when the electrons are close to each other. This mechanism may also be important in double–electron capture processes in heavy (highly–charged) ion–atom collisions when a single high energy photon is emitted. For low Z ions the theoretical predictions are reported to be in good agreement with experimental data [20]. However, for highly charged ions a surprisingly enhanced cross section for double electron capture is found [21].

III. COMPTON RATIO R_C

There are now several calculations of the Compton scattering ionization ratio R_C [22–29]. For finite energies (up to 20 keV) these calculations, performed for He, disagree significantly. However in the high energy limit these theoretical results appear to be in agreement, as discussed by Surić, Pisk and Pratt [18].

Andersson and Burgdörfer [23] performed Compton calculations for helium using highly correlated initial state wave functions and approximately correlated or uncorrelated final state wave functions. Their various calculations all predicted the ratio decreasing toward the same constant ratio of $R_C = 0.8\%$. Hino, Bergstrom and Macek performed lowest order MBPT calculations for He with a basis set constructed using the V^{N-1} potential. In their first paper [24] they asserted that their results reached an asymptotic limit of around $R_C = 1.7\%$ somewhat above 10 keV. However in their later work [25], performed with energies up to 20 keV, they found that their results for the ratio were decreasing and had not reached an asymptotic limit. They concluded their results were consistent with the high energy limits of Andersson and Burgdörfer [23] and Surić et al. [26]. Surić et al. [26,2], using generalized shake–off model or impulse approximation (IA) have derived a high energy limit formula for the double to total ionization ratio for Compton scattering

$$R_C = 1 - \sum_B \int d^3\mathrm{r} \left| \int \Phi^*_B(\mathbf{r}_1)\Psi_i(\mathbf{r},\mathbf{r}_1) d^3\mathrm{r}_1 \right|^2 . \qquad (2)$$

Here $\Psi_i(\mathbf{r},\mathbf{r}_1)$ represents the initial state wave function, $\Phi_B(\mathbf{r}_1)$ is a bound state hydrogen like electron wave function (in the potential of charge Z), and the summation is over all bound states. Using highly correlated initial state wave function they obtain $R_C = 0.008$ (0.8%) for He. At finite energies their results with IA approach the limiting shake–off value from below. The shake–off ratio for Compton scattering, given by Eq.(2), differs from the photoeffect shake–off ratio, given by Eq.(1), because different regions of the initial state contribute to these two processes. Namely, when the incoming photon energy goes to infinity the photoeffect shake–off ratio will be determined exclusively by the region where one of the electrons is at the nucleus. By contrast, the Compton process can occur on free electrons, and all regions contribute in proportion to the probability amplitude that an electron can be found there.

Using the lowest order MBPT based on hydrogenic wave functions and introducing an effective nuclear charge, Amusia and Mikhailov [27,28] obtained $R_C = 0.0168$ for helium. Subsequently, Surić, Pisk and Pratt [18] showed that this introduction of effective charge in the calculation was unjustified. If bare charge is used in the calculation, as is more appropriate, the result would be consistent with other high energy predictions for R_C.

Kornberg and Miraglia [29] studied the energy dependence of R_C using L and V form of the A^2 interaction operator, correlated initial state and uncorrelated final electron states. Their results are form dependent for finite energies, but, for both forms, approach the limit predicted by Eq. (2) ($R_C = 0.008$ for helium). The L-form results support the results of Andersson and Burgdörfer (who used the same form of the A^2 operator, but more correlated initial state and final states) that the asymptotic limit is approached from above. However, the V-form results approach the asymptotic limit from below, as do the IA results of Surić *et al.*.

In the energy region between 6 keV and 20 keV, experimental data are not yet definitely conclusive. At energies between 6 keV and 11 keV the theoretical predictions of Andersson and Burgdörfer seem to be in better agreement with measurements, performed by Levin *et al.* [30] and by Spielberger *et al.* [17], than are the other predictions. At higher energies, 15-20 keV, the measurements of Levin *et al.* [30] seem to be in quite good agreement with the predictions of Bergstrom, Hino and Macek [25], which are higher then the results of Andersson and Burgdörfer as much as 50%. Recent measurement by Spielberger *et al.* [31] with 58 keV photons found $R_C = (0.84^{+.08}_{-.11})\%$ in good agreement with the theoretical predictions for the shake–off limit. Another measurement by Wehlitz *et al.* [32] performed with 57 keV photons is less conclusive. They found $R_C = (1.25 \pm 0.3)\%$.

More extensive study, in a wide energy range, is clearly required in order to understand the energy dependence of R_C. There does seem to be agreement among the theoretical predictions that the high energy limit of R_C is given by Eq. (2), and this is supported by the measurement of Spielberger *et al.* [31]. However, more measurements in this and even higher energy regions are required. As stated by Spielberger *et al* [31], the experiments with photons above 100 keV up to '....150 keV are clearly within reach'. This is based on the facts that Compton scattering cross section slowly varies with energy, in this energy region, and on the availability of new–generation high energy synchrotron radiation facilities.

As one considers this higher energy region, it is clear relativistic effects must be examined. (All existing predictions are nonrelativistic). Recently, we have extended our theoretical analysis of double ionization by Compton scattering, based on sudden approximation [2], to relativistic regions. This approach should be appropriate for the relativistic treatment of double ionization Compton scattering from He-like systems. We have demonstrated that for low Z elements relativistic effects do not alter the high energy limit of the Compton double to single ratio R_C, although these effects do affect single and double ionization Compton scattering separately. (The shake–off argument that relativistic effects do not alter the ratio R_C is the same as for the shake–off for photoeffect).

Justification of the sudden approximation for double ionization by Compton scattering at high energies comes from the observation that, in single ionization Compton scattering at high incident photon energies, most outgoing electrons have large energy. Figure (1) illustrates that, in high energy Compton scattering, most electrons do have enough energy to apply the sudden approximation. We show the singly differential cross section (with respect to outgoing electron energy) in single ionization Compton scattering from helium, calculated using using an exact numerical approach or IA (which can not be distinguished on this figure for these energies). Results for incident photon energies of 25 keV and 150 keV are displayed. We see that most electrons have energies below the edge determined by the Compton energy for scattering from free electrons at rest. Between zero energy and this edge, the differential cross section is a slowly varying function of energy, and all these energies are, approximately, equally likely. If we assume that 3 keV outgoing electron energy is required to apply the sudden approximation (the shake–off ratio R_P in photoeffect, for helium, is approached at this energy) then we see that at 150 keV this approximation is valid for most of the Compton events.

Assuming the validity of sudden approximation we have concluded that relativistic effects are the same in the double and single ionization cross sections. Therefore, in treating double ionization cross sections by Compton scattering at high, relativistic energies it is appropriate to apply the shake–off factor to the single ionization cross sections. At these relativistic energies, and for low Z elements, the single ionization cross sections can be treated, within a high degree of accuracy, using impulse approximation. This is well established [33–36] for the treatment of cross sections up to the level of detail of doubly differential cross section (with respect to outgoing photon angles and energy) [37]. Therefore, in evaluating such differential cross sections, in this energy region we may use IA. Conversely, Surić *et al.* have verified that if one calculates double and single ionization singly differential cross sections the ratio is 0.008 (for He) for all, except very small angles, where both sudden approximation and IA are not valid.

An interesting application of shake–off related ideas in nonrelativistic Compton scattering has been presented by Wang *et al.* [38]. These authors present calculations of the ratio between double and single ionization singly differential cross sections (with respect to energy transfer) by Compton scattering. These calculations show that, except for small energy transfers (where sudden approximation is not supposed to be good) this ratio is constant (the shake–off constant R_C) up to the free Compton scattering edge. This is consistent with with the usual Compton shake–off picture. However, for larger energy transfers (which contribute negligibly to the total cross section) the ratio changes and rises toward the shake–off ratio for photoeffect R_P, reflecting the fact that large energy transfers, as for photoeffect, are only possible when large momenta are transfered to the atomic nucleus.

IV. CONCLUSION

We have reviewed recent progress in calculations of the ratio of double to single photoionization of helium–like atoms at high energies. These ratios, both for ionization through photoeffect and Compton scattering, are largely to be understood through the shake–off mechanism, which assumes that one electron is suddenly removed from a two–electron system allowing the other only to shake off, as the residual system relaxes. Retardation does not change the photoeffect shake–off ratio (retardation is already included in the Compton process). Relativistic effects do not significantly change the ratio for either process. Both retardation and relativity affect the separate cross sections for double ionization and single ionization processes significantly. In the case of photoeffect we have also called attention to the recently proposed additional double ionization mechanism, involving equal energy sharing, which modifies the shake–off prediction for R_P at high energy.

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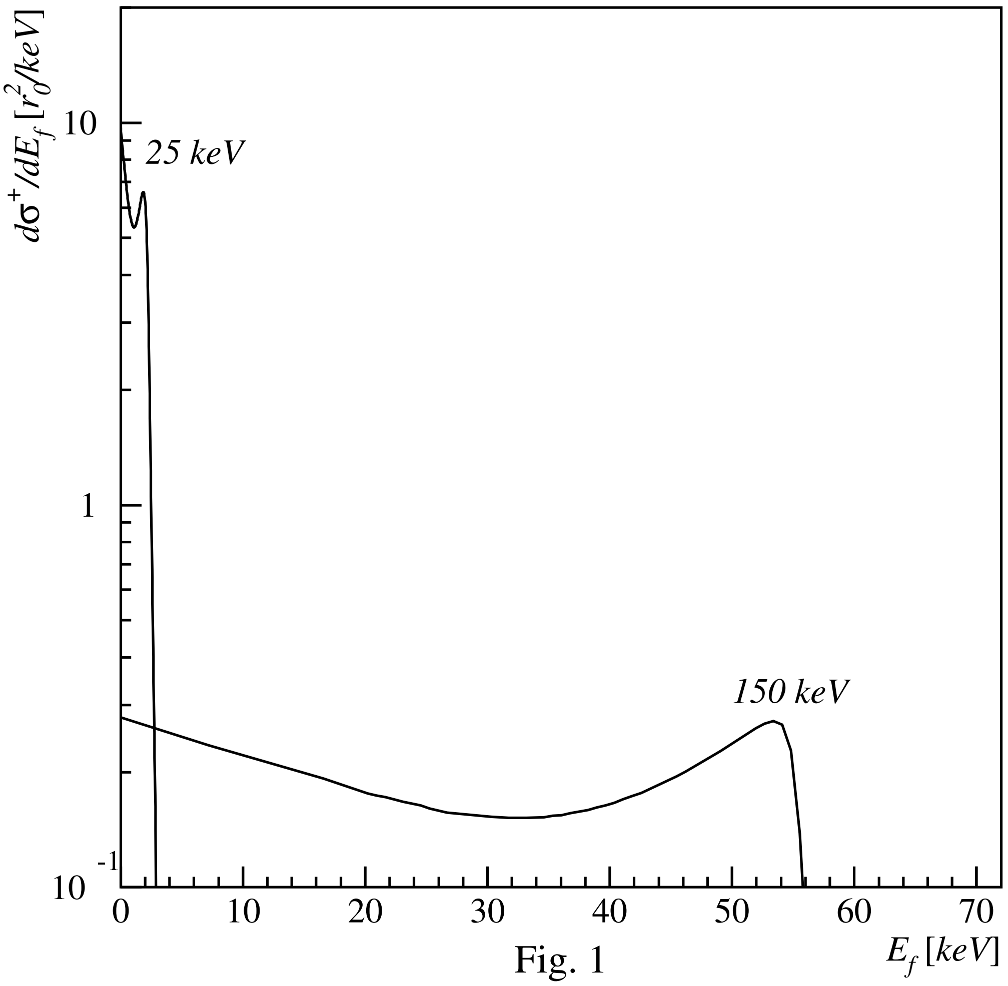

FIG. 1. Singly differential cross section (with respect to outgoing electron energy) in single ionization Compton scattering from helium. Results for incident photon energies of 25 keV and 150 keV are displayed. The maximum outgoing electron energy (free Compton scattering edge), in Compton scattering from a free electron at rest, is 2.2 keV (for 25 keV incident photons) and 55.5 keV (for 150 keV incident photons). Above these energies the singly differential cross sections decrease rapidly with increasing energy.