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The persistent correlation effects in double ionization of helium–like atoms or ions which are probed with high energy photons

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Abstract

We analyze the correlation effects in two electron atoms and ions which can be probed in double ionization with high energy photons. We consider total cross sections and differential cross sections for double ionization by photoabsorption and Compton scattering and discuss the shake and non–shake mechanisms which may contribute to these observables. The total cross section for photoabsorption (at high energies) is determined by two mechanisms, the shake–off mechanism and a quasi–equal–energy sharing mechanism, while for Compton scattering it is determined by the shake–off mechanism. In general the shake–off mechanism probes different distances in the two processes, so that the shake–off ratios are different. Analyzing differential cross sections, we are able to distinguish the shake from the non–shake contributions in photoabsorption, or the variation of shake–off result with distance probed in Compton scattering. We discuss the utilization of the double ionization

Compton profile (as distinguished from the ordinary Compton profile) for studying the correlation effects in Helium.

I. INTRODUCTION

Our present understanding of complex systems in various fields of physics is to a high degree based on independent particle approximation. But as one wants and needs to go beyond this simple approximation the study of the correlation effects among the various constituents of a system becomes very important. In atomic physics, unlike some other fields, the constituent particles and quite exact interactions among them are well known, based on quantum electrodynamics. Yet, our understanding of correlation effects even for the simplest many body atomic systems is far from satisfactory. In this paper we focus our attention on our understanding of one of the simplest correlation effects in one of the simplest atomic systems: high energy double photoionization of two electron (helium–like) atoms and ions.

The dominant photoionization processes, generally resulting in ejection of one electron, 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 the total photon atom interaction decreases as photon energy increases. For a He–like system, at energies for which the incoming photon momentum k becomes 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.

Double ionization of two electron atoms and ions by one photon is now attracting significant attention, both experimentally and theoretically. 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, 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 hope to develop more exact theoretical descriptions and perform more complete experiments. Ultimately we wish to understand all correlation effects in this relatively simple system, with the purpose of thereby better understanding such effects in more complex systems. However, as a first step, further simplification in the study of this system may be obtained by using high energy photons. It has been shown, for both Compton scattering and photoeffect, that using high energy photons can provide conditions in which final state interactions (interactions between the two electrons subsequent to the photon–atom interaction) may be neglected; the processes reflect only the correlation effects in the initial state. This situation arises when one electron takes almost all energy transfered to the atomic system and is rapidly ejected, while the other electron just "shakes off" in the readjustment of the residual ion. The situation is particularly interesting for its simplicity in description and, since it generally dominates the total cross sections for the processes, it is reasonably accessible to experiment. We will here discuss the correlation effects which persist in photoionization with high energy photons.

The simplest observable to discuss, for both experimental and theoretical treatments of the correlation effects in these processes, has been the ratio R 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 and the inclusiveness of its observation. Therefore we talk, for total cross sections, about the Compton double to single ionization ratio R_C and the corresponding photoeffect ionization ratio R_P. The energy dependence of these observables, and particularly their high energy limits, are presently subjects of extensive studies. In the last several years significant progress has been achieved [1]. However, the relatively large disagreements between theoretical predictions, as well as among experimental data, indicate

that we still do not entirely understand, for either process, all aspects relevant in determining the ratio R.

Much understanding of these processes, and their ratios R, can be obtained within the shake–off approximation, *i.e.* a sudden approximation. Present experimental results using high energy photons can be understood within the shake–off picture (in fact some measured high energy photoabsorption ratios R_P implicitly assume a shake–off picture [2]). By shake–off approximation we mean the assumption that the final state electron–electron interaction can be neglected, since one electron accepts almost all energy transfer from the incident photon and moves away much faster than the other [3], which is generally ejected at nonrelativistic velocities consistent with the momentum distribution in the initial bound state. With such an assumption, we consider only a part (perhaps dominant) of the full kinematically allowed region and, therefore, study only (aspects of) the correlation effects in the initial atomic or ionic system.

In the case of photoabsorption, assuming the validity of shake–off approximation (only initial state correlation), this means that we are restricting our study of such correlation effects in the initial state to the region when one electron is close to the nucleus (required if it is to be ejected with high energy). Even if considering more differential observables, not a total cross section, we will see later that in photoeffect the shake–off assumption restricts us to correlation effects in which one electron is near the nucleus. This means that, assuming shake–off, double ionization differential cross sections (in the fast ejected electron angle) are also simply proportional to single ionization differential cross sections, with R_P (0.017 in the case of Helium) being the constant of proportionality for differential as well as for total cross sections. However, it is now being more widely appreciated that there seems to be another, non–shake, mechanism for high energy photoabsorption which also contributes significantly to the total as well as to the differential double ionization cross section [4]. The study of this "quasi–equal–energy sharing mechanism", if experimentally feasible, would reveal the correlation when two electrons, in the initial state, are close to each other, not necessarily close to the nucleus. (Like shake–off this quasi–equal–energy sharing mechanism does not

involve final state correlations.)

In the case of Compton scattering, unlike the double photoabsorption, the shake–off mechanism does seem to be the only dominant mechanism for the double ionization total cross section at high energies. As one considers more differential observables other, non–shake, mechanisms may contribute or dominate. However, even when the shake–off assumption remains valid, the behavior of the ratio R_C for differential cross sections is more complex then for R_P (a constant). Here, even assuming shake–off, in studying differential cross sections we are still studying various regions of the initial state wave function. Therefore corresponding differential cross sections for double ionization and single ionization are not simply connected by a single shake–off ratio (as is the case for photoabsorption assuming shake–off). This will be discussed later, considering singly differential cross section with respect to energy transfer in scattering and obtaining, for high incident energy, a ratio R_C which is a function of the energy transfer.

Here we mention two other atomic processes, related to the photoionization processes, which also probe correlation effects at high energies. These processes, which have also attracted considerable attention both experimentally and theoretically, are (1) the double ionization of a helium–like atom by charged particles and (2) double–electron capture in heavy (highly–charged) ion–atom collisions. For high incident ion velocities the double to single ionization ratio (of total cross sections) for charged particle ionization is related to the photoabsorption ratios R_P at small photon energies [5,6]. For high energy transfers ϵ the ratio of differential cross sections (with respect to energy transfer) for ionization by charged particles is related to double photoionization processes (photoabsorption and Compton scattering) for high energy transfers [7,8]:

$$R_Z(\epsilon) = f_{Z,d}(\epsilon)R_P + (1 - f_{Z,d}(\epsilon))R_{C,nd}(\epsilon). \tag{1}$$

Here, $R_Z(\epsilon)$ is the ratio of double to single ionization differential cross sections for ionization by a charged particle, $R_{C,nd}$ is the ratio of non–dipole cross sections for Compton scattering and $f_{Z,d}(\epsilon)$ is the dipole fraction of the differential cross section for single ionization by a

charged particle.

Another interesting related process is double–electron capture in heavy (highly–charged) ion–atom collisions, in which a single high energy photon is emitted. In the target frame this primarily involves two quasi free electrons of similar momenta, which at relativistic velocities can be stopped with the emission of one photon. The correlation effects when electrons are close to each other may be important (these effects cause a non–shake contribution to the double to single photoabsorption ratio). For low Z ions the theoretical predictions for this process are reported to be in agreement with experimental data [9]. However, for highly charged ions a surprisingly enhanced cross section for double electron capture is found [10].

II. STUDY OF CORRELATION EFFECTS BY DOUBLE IONIZATION IN PHOTOEFFECT

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. However, there are new developments both in the high energy region (our concern here) and at low energies.

New measurements [11], performed using cold target recoil spectroscopy, have found the low energy peak of R_P to be smaller than previously believed, by about 25 %. These new data are in agreement with with some newer calculations [12,13] and they are also consistent with calculations [6] relating the photoeffect ratio to that for fast charged particle impact.

The shake–off value of the ratio R_P for high energy photoeffect is given in nonrelativistic calculations [14–17] as

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

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. (2) has usually been derived in the nonrelativistic dipole approximation without retardation. It reflects the fact that, in the dipole approximation, the dominant mechanism of photoabsorption is the one in which the nucleus takes almost all additional momentum required by energy and momentum conservation and that, at high energy, almost all energy transfer is to one electron (the shake–off situation). [Note that one of the electrons in Eq. (2) is removed from near the nucleus, $\mathbf{r}_2 = 0$.] In the dipole approximation a two–electron system, in the absence of the nuclear field, cannot absorb a photon.

Most theoretical analysis of photoeffect uses the dipole approximation. However, the shake–off result Eq. (2) is not changed, presuming the dominance of the shake–off mechanism, if retardation effects (*i.e.* higher multipole effects) are included, given that for low Z two–electron systems retardation has little effect on the 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 [18] 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, assuming the continued validity of 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 most often a shaken–off slow nonrelativistic electron, irregardless of the energies in the first step. 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. (2) [19]. 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. As shown by Surić, Pisk and Pratt [20], this correlation can be described by a photoeffect effective shake–off charge ($Z_{eff} \sim Z - 0.53$). This effective charge is determined from the monopole term of the electron-electron interaction in the ground state of the helium–like atom (the first term in the multipole expansion of the electron–electron interaction, *i.e.* $1/|\mathbf{r}_1 - \mathbf{r}_2|$ term of the exact Hamiltonian of a helium–like system is replaced by $1/r_>$ where $r_>$ is the larger of r_1 and r_2) [21].

At high energies, where the shake–off mechanism has been generally assumed to be valid, it now appears that another mechanism may contribute significantly to the double to single photoabsorption ratio. Recently, Drukarev [4] has called attention to this "quasi–equal–energy sharing" mechanism [22–24] 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 by about 12 keV. The prediction awaits experimental verification. In this, alternative, mechanism a large momentum is transfered to another electron (instead of to the nucleus as in the shake–off mechanism) and, therefore, both electrons leave the ion with similar large energies. (This would be forbidden in dipole approximation). This leads to a mechanism for double ionization which is not available for single ionization. Amusia *et al* [22] and Drukarev [4] performed an analysis, initially within nonrelativistic quantum mechanics, which did not assume dipole approximation, therefore allowing two electrons to absorb a photon even in the absence of the atomic nucleus. A pair of photoelectrons produced through this alternative mechanism share the photon energy nearly equally (and so we may call this mechanism an "quasi–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 substantial transfered momentum.

As noted by Surić, Pisk and Pratt [2], in the experimental situation in which photoeffect events are distinguished from Compton events by measuring the recoil of the residual ion [25,26], the events produced through the quasi–equal–energy sharing mechanism are not counted in photoeffect events, but with Compton events. These experiments count as photo-

effect events only those produced in the shake–off regime with large momentum transfered to the ion (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 quasi–equal–energy sharing mechanism, involving small momentum transfer to the nucleus. The largest photon energy used in experiments which distinguish photoeffect from Compton scattering was 7 keV, by Spielberger *et al* [26]. The linear term in the photoeffect ratio would contribute approximately 5% at these energies, according to Drukarev. However, as explained above, even increasing the accuracy of their experimental data (which is about 10%) they would not detect this rise in the ratio. The lower energy photoabsorption experiments, which are more precise, and which do not distinguish between photoeffect and Compton scattering (photoeffect however dominates the total cross sections) report the photoeffect double to single ratio to approach a constant above roughly 3 keV. At these energies the contribution of the quasi–equal–energy sharing mechanism (which would be included in some non–shake calculations such as MBPT) would be around 2.5 %, which is of the order of the Compton scattering contribution. At such a level of experimental accuracy both of these effects need to be included in comparison with theory.

Other, more accurate calculations should be able to reproduce this linear rise in the photoabsorption ratio R_P providing that the initial state used in the calculations describes correctly the correlation between the electrons when they are close to each other [27]. Many-body perturbation theory calculations should reproduce this behavior; the approach of Amusia *et al* is an example.

According to Amusia *et al* [22] and Drukarev [4], different regions of the spectrum of outgoing electrons (differential cross section for double ionization photoabsorption with respect to an electron energy) are dominated by different photoabsorption mechanisms. These authors distinguish three different regions in the spectrum. (1) In the region where one electron accepts almost all photon energy (the other electron has energy of the order of binding energy E_B) the dominant mechanism is shake–off as previously discussed. One may use criteria in terms of electron velocities (which is intuitively connected with the shake–off pic-

ture), requiring that one electron has a much smaller velocity then the other. This criteria and the criteria of Amusia *et al* [22] and Drukarev [4] are equivalent. (2) As the energy of the other electron increases (both electrons have energies much bigger then E_B but the energy sharing is still highly asymmetric, $|E_1 - E_2|/\omega \sim 1$ [4]) the influence of the final state interaction becomes more important and eventually dominates. Here for example velocities are comparable. In the total cross section, the contribution of this region, for high energy photons, is negligible compared to the shake–off contribution. (3) As the energy sharing becomes nearly equal, the quasi-equal-energy sharing mechanism becomes important. For large incident photon energies, in the region of the spectrum defined by

$$\frac{|E_1 - E_2|}{\omega} \leq (\frac{\omega}{m})^{\frac{1}{2}}, \tag{3}$$

the quasi-equal-energy sharing mechanism highly dominates over the other two mechanisms. At high photon energies this mechanism, unlike the final state interaction mechanism, gives a significant contribution to the total double ionization cross section. According to Drukarev and Karpeshin [23], who calculated the effect of this mechanism on the photoeffect double to single ratio at relativistic energies, this mechanism dominates the double ionization at relativistic energies. At ultrarelativistic energies it saturates and the ratio approaches the constant $R_P = 0.59/Z^2$ (while both single and double cross section decrease as ω^{-1}), much different from the shake–off constant ($R_P^{s.o.} = 0.093/Z^2$ if using the same model).

This non–shake mechanism of high energy photoabsorption can be observed by detecting electrons which, for nonrelativistic energies, share energy nearly equally and have nearly opposite directions of their momenta. Following the analysis of Amusia *et al* and Drukarev, we may write the unpolarized differential cross section for double ionization through the quasi–equal–energy sharing mechanism in terms of relative electron momenta $\mathbf{p}$ [27], keeping only the dominant term in $|\mathbf{k}| = \omega$

$$\frac{d\sigma^{++}_{e.s.m}}{d\Omega} \sim \frac{(1 - \cos^2\vartheta)\cos^2\vartheta}{p^7}\frac{\omega}{m}, \tag{4}$$

where ϑ is the angle between electron relative momentum $\mathbf{p}$ and photon momentum $\mathbf{k}$. This term is of electric quadrupole character, as one could expect, since the mechanism is

forbidden in dipole approximation. [If the photon interaction with electron spin is included the magnetic dipole term will also contribute. This term has been calculated and discussed by Amusia *et al* [22]. For nearly exact energy sharing this term is dominant (for exactly equal energy sharing the quadrupole term Eq. (4) is exactly zero). However, the contribution of this term to the total double ionization cross section is small compared to the contribution of the electric quadrupole term [22].] Since (at nonrelativistic energies) electrons are ejected with nearly equal and opposite momenta (small momentum transfer to the nucleus) ϑ is nearly equal to the angle between an electron momentum and the direction of photon momentum. We may therefore compare Eq. (4) with the corresponding expression for double ionization through the shake–off mechanism, keeping also only the dominant term in $|\mathbf{k}| = \omega$, which in this case means neglecting retardation and keeping only the dipole term

$$\frac{d\sigma^{++}_{s.o.}}{d\Omega_1} \sim \frac{(1-\cos^2\vartheta_1)}{p_1^7}, \tag{5}$$

where label 1 denotes the fast electron.

¿From these expressions we see that in the quasi–equal–energy sharing region, unlike in the shake–off region, the electrons, which are opposite in momentum, are leaving the ion mostly in directions between the direction of photon polarization and the direction of the photon beam; the maximum in Eq. (4) is at $\vartheta_1 = \pi/4$. The maximum in Eq. (5) appears at $\vartheta_1 = \pi/2$. If retardation effects were included this maximum would shift toward forward angles. Consequences of higher order retardation corrections in Eq. (4) have not been examined.

III. STUDY OF CORRELATION EFFECTS FOR DOUBLE IONIZATION IN COMPTON SCATTERING

It is only recently that calculations of the Compton scattering ionization ratio R_C [28–35] have become available. For finite energies (up to 20 keV) these calculations, performed for He, disagree significantly. However in the high energy limit all these theoretical results

appear to be in agreement, as discussed by Surić, Pisk and Pratt [20]. We quote here our recent description [2] of these Compton calculations.

Surić *et al* [32,3], using the generalized shake–off model or impulse approximation (IA), have derived a high energy limit formula, analogous to Eq. (2) for photoeffect, for the double to total ionization ratio for Compton scattering:

$$R_C = 1 - \sum_B \int d^3\mathrm{r}_1 \left| \int \Phi_B^*(\mathbf{r}_2)\Psi_i(\mathbf{r}_1,\mathbf{r}_2) d^3\mathrm{r}_2 \right|^2 . \tag{6}$$

Here, as in Eq. (2), $\Psi_i(\mathbf{r}_1,\mathbf{r}_2)$ 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. Using a highly correlated initial state wave function Surić *et al* obtain $R_C = 0.008$ (0.8%) for He. At finite energies their results with IA are approaching the limiting shake–off value from below. The shake–off ratio for Compton scattering, given by Eq. (6), differs from the photoeffect shake–off ratio, given by Eq. (2), because different regions of the initial state contribute to these two processes, even though in both cases a high energy incident photon is assumed. The photoeffect shake–off ratio will be determined exclusively by the region where one of the electrons is at the nucleus (or, in the relativistic case, at small–Compton wavelength–distances), as required for the ejection of a single electron when the photon is absorbed. 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.

Andersson and Burgdörfer [29] had 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 a 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. Originally [30] they thought that their results reached an asymptotic limit of around $R_C = 1.7\%$ somewhat above 10 keV. But in their later work [31], considering energies up to 20 keV, they found that their ratio was beginning to decrease and that their component matrix

elements had not at all reached an asymptotic limit. They concluded their results were consistent with the high energy limits of Andersson and Burgdörfer [29] and Surić *et al* [32].

Kornberg and Miraglia [35] have 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. (6) ($R_C = 0.008$ for Helium). The L-form results, like those of Andersson and Burgdörfer (who used the same form of the A^2 operator, but more correlated initial state and final states) give an asymptotic limit which 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 apparent contrast to the above results, in a calculation using the lowest order MBPT based on hydrogenic wave functions and introducing an effective nuclear charge, Amusia and Mikhailov [33,34] interpreted their result as giving $R_C = 0.0168$ for Helium at high energy. However, Surić, Pisk and Pratt [20] afterwards showed that the choice of effective charge for the calculation was unjustified. If bare charge is used in the calculation, as is in fact more appropriate, the result would be consistent with other high energy predictions for R_C.

The experimental data which now exist for Compton scattering are suggestive but not yet entirely 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* [36] and by Spielberger *et al* [26], than are the other predictions. At higher energies, 15-20 keV, the measurements of Levin *et al* [36] seem to be in quite good agreement with the predictions of Bergstrom, Hino and Macek [31], which are higher then the results of Andersson and Burgdörfer by as much as 50%. Finally, recent measurement by Spielberger *et al* [37] 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* [38] performed with 57 keV photons is less conclusive. They found $R_C = (1.25 \pm 0.3)\%$.

As one considers higher energy regions, it is clear relativistic effects must be examined. Recently, Surić *et al* have extended their theoretical analysis of double ionization by Compton

scattering, based on sudden approximation [3], to relativistic regions. They have demonstrated explicitly 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 [19]).

A more detailed study of nonrelativistic Compton scattering (more then just the total cross section), at high energies, has recently been presented by Wang *et al* [39]. These authors report calculations of the ratio $R_C(\epsilon)$ between double and single ionization singly differential cross sections (with respect to energy transfer $\epsilon = \omega_i - \omega_f$; ω_i is incident photon energy and ω_f scattered photon energy) by Compton scattering. In their numerical calculations they used only the A^2 term from the complete photon–electron interaction Hamiltonian. (See our discussion below of the appropriateness of this assumption). These calculations show that, except for small energy transfers (where sudden approximation is not supposed to be good) this ratio $R_C(\epsilon)$ is constant, (equalling the shake–off constant R_C of total cross sections) up to the free Compton scattering edge (maximum energy transfer in Compton scattering from free electrons at rest, $\epsilon_{free}^{max} = \omega_i/[1 + m/(2\omega_i)]$). This is consistent with the usual Compton shake–off picture. For larger energy transfers (which contribute negligibly to the total cross section) the ratio changes. It first slightly decreases, then increases toward a maximum [their value of $R_C(\epsilon)$ at this peak is between 3.6 % and 4.2 % for incoming photon energies between 20 keV and 90 keV], and then decreases toward a value above the shake–off ratio (for 20 keV photons this value is around 2.6 %, according to Wang *et al*). They find that at relatively low incident photon energies (such as 20 keV) an additional term contributes significantly to the shake–off term even at the largest allowed energy transfers. They call this term an "exchange shake–off term". The authors argue that for larger incident photon energies the "exchange shake–off term" becomes small and, therefore, the ratio $R_C(\epsilon)$ should approach the photoeffect ratio R_P exactly, for large energy transfers. (The authors assume that the high energy photoeffect ratio is the shake–off ratio $R_P^{s.o.} = 1.66\%$). This behavior of the ratio $R_C(\epsilon)$ reflects the fact that large energy transfers, as for photoeffect, are only

possible when large momenta are transfered to another particle in the atomic system.

This (nonconstant) behavior of the ratio $R_C(\epsilon)$ with energy transfer ϵ can qualitatively be understood within the shake–off picture. The utility of a shake–off calculation of $R_C(\epsilon)$ for high energy transfers was analyzed by Surić *et al* [40], taking into account the complete electron–photon interaction Hamiltonian, *i. e.* in the nonrelativistic formulation including both A^2 and $\mathbf{p} \cdot \mathbf{A}$ interactions. The shake–off contribution to the differential ratio $R_C(\epsilon)$ is obtained if most energy transfer is taken by one of the two electrons. Then, necessarily, large momenta are transfered to the nucleus and not to another electron, which only shakes off. This can be experimentally distinguished by observing the recoil of the nucleus which, for large energy transfers, becomes large.

For large energy transfers the dominant part of the photon–bound electron interaction which determines Compton scattering becomes the $\mathbf{p} \cdot \mathbf{A}$ term, not the A^2 term considered by Wang *et al.* It can be shown, using the low energy theorem, that for high energy transfers (low outgoing photon energies) the Compton scattering matrix element becomes proportional to the photoabsorption matrix element. As earlier discussed, photoabsorption at these energies is determined by two mechanisms, the shake–off and quasi–equal–sharing mechanisms. Both of these mechanisms should influence the Compton ratio $R_C(\epsilon)$ when $\epsilon \to \omega_i$. However the shake–off ratio does not depend on the choice of interaction as long as the same region of the matrix element is being emphasized. Therefore, as discussed by Surić *et al*, for very large energy transfers ($\epsilon \to \omega_i$) the Compton ratio $R_C(\epsilon)$ can be understood in terms of these mechanisms. In the situation when the shake–off assumption is valid (we can experimentally distinguish such events) the Compton ratio $R_C(\epsilon)$ approaches the photoeffect shake–off ratio $R_P^{s,o}$, as suggested by Wang *et al.*

The shake–off picture is valid for those Compton events in which one electron takes most of the energy transfer. (Judging by the required energy for the shake–off assumption in photoabsorption to be valid, this energy transfer should be larger then approximately 3 keV [40], which would require a far higher incident photon energy). In the region between roughly 3 keV energy transfer and the free Compton scattering edge one bound electron takes most

energy transfer through the free Compton mechanism (no momentum transfer to another particle is necessary). The differential ratio in this region is nearly constant, corresponding to the total ratio R_C. We are probing some average correlation between electrons in the initial state, as in the case of studying the total ratio R_C. Note that, to obtain 3 keV electrons through this free Compton mechanism (which dominates the total cross section), incident photons should have about 30 keV. To have the shake–off picture valid for the total cross section for the double ionization by Compton scattering most electrons should have energy above 3 keV, which means that photon energies above 50-60 keV are required.

For higher energy transfers, momentum transfer (by the electron) to another particle is necessary. In the case of the shake–off mechanism this particle is the nucleus. In order to transfer large momenta to the nucleus an electron must approach it and as energy transfer increases, it must be closer, so that in double ionization we are probing the correlation between the two electrons when one electron is near the nucleus. For very large energy transfers we are probing the same region as in the case of photoeffect with shake–off. Therefore, in the shake–off region for large energy transfer ϵ, the differential ratio $R_C(\epsilon)$ approaches the photoeffect shake–off ratio $R_P^{s;o}$ as $\epsilon \to \omega_i$. Thus different regions of energy transfer give us information about different regions of the ground state, even when the shake–off approach remains valid. This example shows that even when the shake–off assumption remains valid, the behavior of $R_C(\epsilon)$ for differential cross sections is more complex, dependent on ϵ but not ω_i, than for R_P (a constant), independent of ω_i (which is the energy transfer ϵ in photoeffect). Study of $R_C(\epsilon)$ with energetic photons probes electron–electron correlations in various regions of the ground state wave function.

These ideas can be further extended in studying correlation effects, using double ionization doubly differential cross sections (with respect to outgoing photon energy and angles) at high momentum transfers. As is well known, ordinary Compton scattering became an important tool in the study of the momentum distribution of bound electrons, through utilization of the impulse approximation. This momentum distribution is experimentally extracted from the Compton profile, which includes, in the case of helium–like systems,

both single and double ionization (single ionization being dominant). However, we may define a double ionization Compton profile by specifying the state of ionization of the ion.

Using the impulse approximation formula obtained by Surić *et al* [32] the doubly differential cross section for double ionization Compton scattering from a two-electron atom is

$$\frac{d^2\sigma_C^{++}}{d\omega_f d\Omega_f} = \left(\frac{d\sigma}{d\Omega_f}\right)_0 \left(\frac{\omega_f}{\omega_i}\right) S^{++}(\omega, \mathbf{k}), \tag{7}$$

with the expression S^{++} given by

$$S^{++}(\omega, \mathbf{k}) = \int d^3\mathrm{p}\delta(\omega - \frac{k^2}{2m} - \frac{\mathbf{k}\cdot\mathbf{p}}{m}) \left[\int |\Psi_i(\mathbf{p}, \mathbf{q}\ )|^2 \, d^3\mathrm{q} - \sum_B \left|\int \Phi_B^*(\mathbf{q}\ )\Psi_i(\mathbf{p}, \mathbf{q}\ ) d^3\mathrm{q}\right|^2\right]. \tag{8}$$

Here $(d\sigma/d\Omega_f)_0$ is Thomson scattering cross section (for two electrons) and the meaning of other symbols is as in Eq. (6), except that the wave functions are represented in momentum space.

The structure of the double ionization Compton profile is similar to the structure of the Compton profile in ordinary Compton scattering [3] and the position of the peak is the same. As in ordinary Compton scattering, different regions of the Compton profile probe different regions of the initial bound two–electron state momentum distribution. However, in the ordinary Compton profile the correlation effects (we are primarily interested in correlation effects beyond the Hartree–Fock model) are relatively small (of the order of 1% or less for the helium–like atom which we discuss here). Contrary to this, in the double ionization Compton profile the correlation effects (beyond Hartree–Fock) are much more pronounced [3], as one would expect. Therefore, the study of the double ionization Compton profile can provide a more precise tool for the study of correlation than the ordinary Compton scattering. For simple systems, like Helium, these type of experiment seems to be within reach. Experimentally one may detect photons in coincidence with a doubly ionized ion. Detection of recoil ions produced in double ionization Compton scattering has already been successfully accomplished.

Some comments on the validity of impulse approximation for the calculation of the double ionization Compton profile are in order. The utility of impulse approximation in calculating the ordinary Compton profile is well established, even in circumstances which stretch the assumptions of its derivation [41–45]. In the case of the Helium atom the theoretical predictions have been successfully compared with experimental Compton profiles at photon energies around 15 keV [41] for backward angles (to assure large energy transfers). These energies, which in fact push the parameters assumed in its derivation, may not be sufficient for double ionization Compton profile and larger energies may be required.

This can be seen from the study of the triply differential cross section of the single ionization Compton scattering. It has been found by Z. Kaliman *et al* [46] that for the triply differential cross section (when the outgoing electron is also observed) the impulse approximation approach is less justified at these energies. We may view the double ionization Compton profile as a less averaged observable (as is the triply differential cross section in single ionization), and therefore larger momentum transfers, corresponding to the assumptions underlying impulse approximation, are in fact required for its validity in describing non–averaged observables. These requirements correspond to the region of validity of the shake–off description. As already discussed, the shake–off assumption is valid when the fast outgoing electron has energy above approximately 3 keV. In order to reach this region for backward angles at least 30 keV photons are required.

IV. CONCLUSIONS

We have analyzed the correlation effects in two electron atoms and ions which can be probed in double ionization with high energy photons. The analysis is made for the two dominant photoionization processes (photoabsorption and Compton scattering) in which, due to the correlation effects, simultaneous ejection of two electrons may occur. In the case of high energy photoabsorption two correlation effects in the initial state are responsible for the mechanisms, shake–off and the quasi–equal–energy sharing, which determine the total

cross section. The correlation effects in the initial state which lead to these mechanisms are: (1) monopole interaction (correlation between the electrons when one electron is close to the nucleus) for the shake–off mechanism (one electron takes all photon energy and the nucleus recoils with nearly equal but opposite momentum) and (2) correlation between the electrons when they are close to each other (nonrelativistic electrons share the photon energy nearly equally and they are ejected with nearly equal and opposite momenta while the nucleus recoils with relatively small momentum). In the case of Compton scattering the total cross section is dominated by the shake–off mechanism. However, unlike in photoeffect, this shake–off mechanism is a result of an averaged correlation between the two electrons at all positions in the initial state.

Analyzing differential observables (cross sections) we are able to distinguish the shake from the non–shake contributions in photoabsorption, or the variation of the shake–off result with distance probed in Compton scattering. In the case of double photoabsorption (for differential cross sections) the shake–off mechanism probes the same correlation effects as in the case of total cross section (one electron must approach close to the nucleus in order to accept almost all photon energy). Therefore, it provides no further information on correlation in the initial state. Due to the non–shake mechanisms (both quasi–equal–energy sharing and final state interaction), additional information on correlation effects can be obtained from the differential cross sections for high energy photoabsorption. In the case of high energy Compton scattering, unlike in photoabsorption, the study of differential cross sections, even in the shake region, can provide further information on correlation effects in the initial state. Particularly, as discussed, the double ionization Compton profile can be used for studying two–electron wave functions of helium–like atoms or ions in momentum space in a similar way as the ordinary Compton profile is used in studying momentum distribution of bound electrons within independent particle approximation. Varying the energy transfer varies the regions of the initial state electron (to be ejected fast) in which correlations with the subsequent shake–off electron are probed.

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