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High energy limit of photon-atom interactions: example of double ionization of He by photoabsorption

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Abstract

An important class of high energy atomic processes can be understood from the theory of asymptotic Fourier transforms. The asymptotic behavior is determined from the singularities of initial and final state many-body wave functions and interactions. These in turn follow from the singularities of the interaction between particles. Final results are gauge and form independent, but dependence on the quality of initial and final wave functions varies with gauge and form. We illustrate these ideas in a discussion of the double ionization of helium in photoabsorption, relating the dominant absorption mechanisms to electron-nucleus and electron-electron coalescences.

I. INTRODUCTION

In this paper we discuss the high energy limit of double ionization of helium in photoabsorption as an example of the insights which can be achieved in the study of the high energy limit of photon atom interactions.

In general we can identify the dominant high energy processes which result when a photon is incident on a composite structure of charged particles, such as an atom, and how they differ from the corresponding processes (scattering, pair production, and photon splitting) of photons incident on free charges. Additional processes, such as excitation and photoionization of the structure, also occur. Appropriate description depends on the detail of observation; implicit sum rules relate exclusive and more inclusive characterizations of processes. We may identify the aspects of composite structure which are relevant to the description of these high energy situations. We may summarize what is known about the high energy limit of photoelectric effect, elastic (Rayleigh and Delbrück) and inelastic (Raman and Compton) scattering, and pair production (bound and free). We may discuss the relationship of high energy processes in which single or double ionization is specified. We may discuss the modifications and further features which result if the composite structure is in an excited state, rather than a ground state. We can note the further considerations if the composite structure is not isolated, but in an environment, such as a plasma, a strong field, or a lattice.

The study of atomic processes at high energies is of both fundamental and practical interest. Basic mechanisms are often revealed and isolated in their most transparent ways at high energies. We think here particularly of the study of multi-particle correlated interactions. This provides insight for the development of approaches to the description of these mechanisms, which can then be utilized more generally, as in the example which we discuss below. For example, at high energy there are situations in which one can neglect final state interactions, and this leads to the use of high energies in studying composite particles (as in photoeffect, Rayleigh and Compton scattering, and double ionization). High energy studies are also now much more firmly motivated, since the processes are now observable and theoretical views are subject to experimental test. We now have both modern high energy and intensity synchrotron sources and sources of fast highly charged ions.

Here we focus on a limited, but fundamental, aspect of these issues. We study the double ionization of the ground state of He in photoabsorption at high photon energies ω (but still

$\omega \ll m$), to lowest order in the electron–photon interaction Hamiltonian, including the first order retardation term in that interaction for the kinematics in which it gives a dominant contribution to the total ionization cross section. At these energies the portions of the matrix element which dominate the spectrum are asymptotic regions of single (rather than double) Fourier transforms (FT) in a single large momentum, of a function with coordinate-space singularities (i.e. points at which the function is not differentiable). These singularities originate in the Coulombic (electron–nucleus and electron–electron) interaction potential energy; in the matrix element they enter through wave functions (initial and final) and/or the electron–photon interaction, depending on the form used. We may use the properties of the asymptotic behavior of a Fourier transform (FT) of such singular functions in order to study the dominant contributions to the spectrum in double photoionization of He.

The matrix element for photoionization is an integral over a product of initial state wave function, final state wave function and electron–photon interaction operator. These wave functions, being solutions of the Schrödinger equation with Coulombic interactions, have singularities at the singularities of the potential. These singularities are located at the points where the particles coalesce and we will use the term coalescence points for the locations of the singularities. These are electron–nucleus (e–N) coalescences, when one electron is at the nucleus, and an electron–electron (e–e) coalescence when the two electrons coincide. There is also a triple coalescence point when all three particles coincide. The properties of wave functions in the near vicinity of these singularities which, for bound states are well understood [1,2], can be extracted from the Schrödinger equation. They are known as coalescence properties and in the case of s-states are often called Kato cusp conditions. We will here also discuss these general properties for continuum states, as well as related properties for high energy electrons.

Our discussion is general and does not depend on the choice of the form (length, velocity, acceleration etc.) of the photoionization matrix element. It is based on a general theorem of Fourier transforms, which states that a slow asymptotic decrease for large p, such as $1/p^n$ in a $1/p$ asymptotic power expansion, of the Fourier transform of a well localized function, comes only from singularities of that function. Namely, according to the Fourier transform theorem [3], a function $f(x, y, z)$, infinitely differentiable at all points (x, y, z), which decreases for large $r = (x^2 + y^2 + z^2)^{1/2}$ faster than any power of r, has a Fourier transform $F(p_x, p_y, p_z)$ which decreases faster than any power of $p = (p_x^2 + p_y^2 + p_z^2)^{1/2}$ for large p. Functions which appear in our photoionization matrix elements are well localized (due to the fact that the bound state is localized) and they are differentiable everywhere except at coalescence points, where they are singular [1], *i.e.* non–differentiable. The asymptotic form of the FT of such a function, for singularities resulting from point Coulomb potentials, is expandable in powers of 1/p. It can be obtained from expansion of the function in powers of r around the singular points, since for such potentials solutions of the Schrödinger equation can be expanded in powers of r in the vicinity of the singular points.

II. DETERMINATION OF HIGH ENERGY BEHAVIOR FROM THE SINGULARITIES IN INTERACTIONS BETWEEN PARTICLES

The matrix element for high energy double ionization is in general a double Fourier transform of functions of two coordinates in two large momenta. These momenta are for example $\mathbf{p}_1$, $\mathbf{p}_2$ of the two ejected electrons, or combinations of these two momenta, such as relative momentum $\mathbf{p} = (\mathbf{p}_1 - \mathbf{p}_2)/2$ of the two electrons and the total momentum $\mathbf{P} = \mathbf{p}_1 + \mathbf{p}_2$. The corresponding coordinates could be the coordinates of each electron, or their relative and center-of-mass coordinates. These always have singularities, at least in the case of the triple coalescence. However in certain kinematic situations (as for example when one of the outgoing electron momenta is small) the matrix element involves only a single Fourier transform of one coordinate (with a singularity) in one large momentum. At large energies at least one momenta (and we call it p) has to be large. This means that at large energies we are always in some asymptotic FT region (except in the unusual situation that the energy corresponds to a bound state in the continuum). The FT theorem tells us that a 1/p expansion (since our interactions are point Coulombic) of such Fourier transforms in their asymptotic region is connected with coalescences in the corresponding coordinate. Whether only one double coalescence or both double coalescences (i.e. the triple coalescence) contribute depends on the kinematics of the region, but in any such kinematical region we will have the dominant parts of the matrix element expressible in terms of powers of 1/p. The largest leading powers in 1/p are obtained in kinematics where all momenta are large (*i.e* the matrix element is a double Fourier transform in two large momenta), and in this case the triple coalescence determines this leading power in 1/p. However, there are kinematical regions where only one double coalescence contributes, and in this case the leading powers in 1/p are smaller than those determined by the triple coalescence. These are the regions where matrix elements can be written as a Fourier transform in just one large momentum associated with a coalescence. The kinematics are such that the second Fourier transform is not in an asymptotic region. The single large momentum may be the momentum $\mathbf{p}_1$ of one electron while the momentum $\mathbf{p}_2$ of the other electron is small (this region is known as the shake–off region), but it can also be the relative momentum $\mathbf{p}$ of the two electrons, while the total momentum $\mathbf{P}$ is small (quasi–free region), or the large momentum can be relative momentum $\mathbf{p}$ but with electron momenta nearly perpendicular (final–state–interaction region). It could also be some other combination.

Although in each of these kinematic regions the leading power of the matrix element in 1/p is determined by one Fourier transform and one coalescence point, this does not mean that the leading power is of the same order in each regions. [It is $1/p^3$ in the shake-off and quasi-free regions, $1/p^6$ in the final-state-interaction region. In fact in all of these examples the leading power is of lower order than that given by the triple coalescence ($1/p^7$)]. The power depends on the origin of the oscillating term of the final state wave function. This oscillating term may come from a plane wave (distorted), as in the SO and QF regions, or it may come from a product of plane wave and some spherical wave (as in the final-state-interaction region), or perhaps from some other combination of plane waves and spherical waves which may appear in the final state wave function. In other kinematical regions the matrix element is the asymptotic form of the FT in two large momenta. The leading power is determined by the triple coalescence (the two double coalescences coincide), which results in a higher power of 1/p. In all kinematical regions there will be a contribution from the triple coalescence.

III. CHARACTERIZING THE KINEMATIC REGIONS IN CROSS SECTIONS

Due to the generality of our approach we are able to address from a unified viewpoint a number of issues which have appeared and been discussed separately in previous treatments of double and single photoionization processes. These issues include characterization of the kinematic regions of the matrix element and cross sections (differential and integrated):

1) The dominant contributions to the total cross section for double ionization of He. These contributions are usually called the shake–off (SO) (where one electron takes almost all photon energy and has high momentum while the other shakes–off with low momentum) and the quasi–free (QF) (electrons have nearly equal and nearly opposite momenta, *i.e.* the relative momentum is large while the total momentum is small) contribution. The shake–off contribution has been considered extensively and often considered the only dominant contribution to the total cross section for double ionization of the ground state of He. It was obtained using V-form [4]and A-form [5] in coordinate space or V-form in a momentum space approach [6]. All these approaches agree providing the cusp condition at the nucleus is satisfied by the initial state wave function, as required by the Schrödinger–equation. The derivations of the shake–off contribution rely on the assumption that one electron is fast and the other slow, and therefore the whole contribution (at high energies) comes from the point at the nucleus. In our asymptotic FT approach this contribution is understood to represent a single FT in the large momentum $\mathbf{p}_1$ (of the fast electron) associated with the singular point $\mathbf{r}_1 = 0$ which, according to FT theory, gives the dominant asymptotic behavior for large momenta. Naturally, in the case of high energy single ionization (including ionization both with and without excitation) the same singularity is necessarily associated with the single large electron momentum, and so the same general discussion follows.

In the case of double ionization the point at the nucleus is not the only singular point of a two electron wave function which can be associated with a large momentum. The other point is at the electron–electron coalescence. In the case of the ground state of He (s–state and spin singlet) this singularity has the same behavior as the electron–nucleus singularity. In the case of double ionization there is a kinematic region (large relative momentum $\mathbf{p}$ of the two electrons and small total momentum $\mathbf{P}$) for which the matrix element is a single FT in the large momentum $\mathbf{p}$ associated with the coalescence at the relative coordinate $\mathbf{r} = 0$. The contribution to the cross section from this singularity has the same momentum dependence as the contribution from the electron–nucleus coalescence. It is, however, forbidden in dipole approximation, and it only appears in the first retardation correction. Our analysis confirms that the contribution from this coalescence is the quasi–free contribution predicted in [7] and [8,9]. We discuss this contribution in V–form, A–form, and in momentum space in order to compare with the existing analysis of the shake–off contribution. We discuss the energy dependence of the SO and QF contributions to the total cross section and the consequences of this energy dependence for the ratio of double to single ionization by photoabsorption. Our analysis confirm the result [8,9] that for high but nonrelativistic energies the double to single ionization ratio for photoeffect from the ground state of He is a linearly rising function in which the linear term (due to the first retardation correction) is a consequence of the existence of the cusp in the e–e coalescence of the ground state of He.

2) The dominant contributions to the spectrum. We may identify three regions in the spectrum, and we may identify the leading contribution to the matrix element in each. Two of these leading contributions (SO and QF) which are associated with specific kinematics, as already discussed, are determined by a corresponding singularity of the interaction potential energy. In a suitable form (A or V) they are independent of the final state interaction between electrons. The third such contribution to the spectrum (the final–state–interaction contribution, dominant in the intermediate energy region between the SO and the QF regions) is also determined by a coalescence point (e-N singularity). However, in all three forms (A,V,L) it requires the e–e interaction in the final state, described by the spherical

waves of the relative e–e motion, which means that relatively large (but not too large) distances between the two electrons are involved. Again, there is specific kinematics (the two electrons leaving in nearly perpendicular directions) for which the matrix element is a single FT in the large momentum $\mathbf{P}$ associated with the e–N coalescence. This third contribution (final–state–interaction) can be obtained in a simple way using V-form, neglecting the interaction of the electrons with the nucleus in the final state. Thus in the contributions we have discussed, and with our choice of form, we have neglected either e–e or e–N interactions in the final state. Can additional contributions of the same order appear if we simultaneously include the interaction of all three particles in the final state? To get some sense of the possibilities we have used C3 functions for the final state; in this case we find no additional contributions. However, we do not know whether the exact final state wave function may in fact contain other terms (similar to the one described by the e–e spherical wave in C3) which would give such contributions. Our analysis does show that all known dominant contributions to all points of the spectrum, in addition to the SO and QF contributions which dominate the total cross section, are also determined by singularities at the two singular points of the wave functions.

3) Behavior of cross sections in other regions (the triple coalescence). Our approach also allows considerations of more differential cross sections for double ionization (i.e. differential both in energy and angles) at high energies. The discussion in points 1) and 2) is concerned with the regions in which the matrix element is a FT in one large momentum connected with a singularity. In kinematics in which both momenta are large the asymptotic FT is connected with the two singularities $\mathbf{r}_1 = 0$ and $\mathbf{r} = 0$, *i.e.* with the triple singularity. The leading power in an inverse momentum expansion (the two large momenta are now of the same order, say p) is $1/p^7$ in the case of the He ground state. This means that, generally, in any kinematics there is at least a contribution from the triple coalescence point (which, in the case of the ground state, gives a $1/p^7$ contribution). Unequal energy sharing (away from the SO region) with electrons moving in the same or opposite directions would be examples where matrix element is determined by the triple coalescence point. Recently,

the dipole matrix element for double ionization has been evaluated in the kinematics of electrons moving in the same direction with the same energies, in order to evaluate the inverse process, *i.e.* radiative double electron capture [10]. At high energies the result shows an energy dependence characteristic of triple coalescence, as follows from our approach.

IV. FURTHER ISSUES

4) The important properties of coalescence points and various forms of electron–photon interaction. While the analysis, in points 1) and 2), of the importance of the coalescence points does not depend on the choice of the form of the photoionization matrix element, in different forms the needed information about these coalescences is differently positioned among the interaction, the final state, and the initial state wave functions. We find that if the A-form is used the important information about the singularities is contained in the electron–photon interaction. Only the proper value of the initial state wave function at the coalescence points is needed to evaluate the leading contributions. No information on coalescences from the final state wave function is needed. This analysis agrees with that of [5,11], who discussed the SO region.

In the V form, but only when s–singlet states are considered, the coalescences which give the leading contribution to any region of the spectrum are simply the initial state coalescences. Here information on the slope of the wave function at the coalescence points is needed to evaluate the leading contributions. This information is contained in Schrödinger equation and is connected to the value of the wave function at coalescences. This connection is usually described in terms of (Kato) cusp conditions. No information on coalescences from the final state wave function is needed. This is known for the SO region [4,11]. However, it should be noted that, if non–s–states are considered, the final state wave function coalescences are important and must be treated correctly [12]

In the L form the needed information about the coalescence is contained in both initial and final state, even when ionization of the ground state is considered. Inconsistent treatment of coalescence points in initial and final states can lead to a spurious $\omega^{-5/2}$ dependence of the double ionization total cross section, rather than the correct $\omega^{-7/2}$ (in dipole approximation). This is an example of the general experience that, unless improvements are introduced consistently, they may not lead to improvements in prediction. According to Åberg the spurious contribution appears when using approximate initial state wave functions which do not satisfy the e–N Kato cusp condition, together with final state wave functions which do. This is correct neglecting e–e coalescence in both initial and final state (as Åberg does). More generally, if approximate wave functions are used, the coalescences in both initial and final states must be treated in the same way, to ensure cancelation of spurious terms. If the initial state wave function is accurately described at coalescence points, so the final state wave function must also be accurately described. For example, if the C3 function (which properly treats coalescence points in the final state) is used, the initial state wave function must satisfy Kato conditions at both coalescence points in order to cancel the spurious $\omega^{-5/2}$ contribution.

5) The treatment of states other than the ground state. Although we are mainly concerned with the photoionization of the He ground state our approach allows us to address problems in treating other states. In particular we wish to address the issue of how appropriate it is to neglect various final state interactions (as is often done at high energies), considering the various forms of the photoionization matrix element (L, V, A) which may be utilized. We know that in single electron problems [or in independent particle approximation (IPA)] the final state interaction can not be neglected in L form for any initial state. In the case of V-form the final state interaction can be neglected only for s–state ionization. In A–form the final state interaction can be neglected for any state.

However, if one goes beyond the IPA picture, the situation changes. Recently deviations from the IPA predictions were observed for 2p–state single ionization of Ne [12]. This was explained as due to final state interaction (beyond IPA) between outgoing electron and the ion; there is also a correction to the initial state normalization due to initial state correlation interactions. It can be shown (the most convenient way is in the A–form), using our approach of expansion around the singular point (only the e-N singular point contributes in single ionization), that the important part of the final state interaction beyond IPA, which may persist (depending on the configuration) even in the asymptotic limit of 2p ionization, is the interaction of the outgoing continuum electron at the electron-nucleus coalescence.

Most of the discussion regarding various forms (including the discussion of the treatment of non–s states) can be presented using the example of single photoionization in independent particle approximation, which involve only one (electron–nucleus) coalescence. We may start with such an example, in order to explain our approach in calculating the asymptotic behavior of a Fourier transform of a function with singularities, using expansion around the singularity. We can analyze the A, V and L forms of the matrix element and for each identify the needed quality of approximations for the initial and final state wave functions to get the proper high energy result. This can be illustrated analytically in the special case of the point Coulomb potential, demonstrating how the same result is obtained in all these forms when singularities are treated properly in both initial and final states.

V. CONNECTIONS TO MOMENTUM SPACE APPROACHES

A discussion of the high energy limit of photoabsorption (and related processes), similar to that presented here in coordinate space, can be given in momentum space. The matrix element is an integral over initial and final wave functions and the photon–electron operator, now all represented in momentum space. Such an approach is illustrated by work in the SO region [6] and the QF region [7,9]. Using the standard form of the electron–photon interaction in Coulomb gauge (corresponding to V–form in coordinate space) the matrix element, in these regions, can be written in terms of the initial (bound) state wave function in momentum space, evaluated for large momenta appropriate to the region ($\mathbf{p}_1$ in the SO region or relative momentum $\mathbf{p}$ in the QF region). Using the Schrödinger equation in momentum space, we may evaluate the matrix element in an iterative procedure, obtaining the leading powers in inverse momenta from the interaction potential energy evaluated at these large momenta. For the SO region the relevant part of the interaction is the e-N interaction, while for the QF region the relevant portion is the e-e interaction. In this way the leading term in the power expansion in $1/p$ is obtained directly from the interaction potential energy, as it is in the coordinate representation when A–form is used. The final result is the same as the coordinate space result. The SO result of [6], obtained in momentum space, and the result of [5], obtained in coordinate space using A–form, are both expressed in terms of the value of the wave function at the coalescence.

VI. CONCLUSIONS

After giving an over-view of the issues and interests which arise in considering the dominant high energy processes which result when a photon is incident on a composite structure of charged particles, such as an atom, we have focused on a limited, but fundamental, aspect of these questions. We have discussed the double ionization of the ground state of He in photoabsorption at high photon energies. This is an example of the important class of high energy atomic processes which can be understood from the theory of asymptotic Fourier transforms. The asymptotic behavior is determined from the singularities of initial and final state many-body wave functions and interactions. These in turn follow from the singularities of the interaction between particles. In this case the dominant absorption mechanisms may be related to electron-nucleus and electron-electron coalescences.

At high energies the portions of the matrix element which dominate the spectrum of double photoionization are asymptotic regions of single (rather than double) Fourier transforms (FT) in a single large momentum, of a function with coordinate-space singularities (i.e. points at which the function is not differentiable). The dominant contributions to the total cross section for double ionization of He are usually called the shake–off (SO) (where one electron takes almost all photon energy and has high momentum while the other shakes–off with low momentum) and the quasi–free (QF) (electrons have nearly equal and nearly opposite momenta, *i.e.* the relative momentum is large while the total momentum is small) contribution. In the shake–off contribution one electron is fast and the other slow, so that the whole contribution (at high energies) comes from the point at the nucleus. This contribution is understood o represent a single FT in the large momentum $\mathbf{p}_1$ (of the fast electron) associated with the singular point $\mathbf{r}_1 = 0$ which, according to FT theory, gives the dominant asymptotic behavior for large momenta. In the quasi-free contribution there is a kinematic region (large relative momentum $\mathbf{p}$ of the two electrons and small total momentum $\mathbf{P}$) for which the matrix element is a single FT in the large momentum $\mathbf{p}$ associated with the coalescence at the relative coordinate $\mathbf{r} = 0$. The contribution to the cross section from this singularity has the same momentum dependence as the contribution from the electron–nucleus coalescence, but it is forbidden in dipole approximation and appears in retardation terms.

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