Multielectron atoms in ultra-short light fields: a time-dependent R-matrix approach

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Introduction

The recent generation of extreme ultraviolet (XUV) attosecond light pulses [1] represents an exciting new era for ultra-fast science. Since the time structure of these light pulses is comparable to the time scale of atomic inner shell processes, these pulses have opened up the possibility for the measurement of time-resolved electron dynamics in atoms [2]. These ultra-short light fields have also recently enabled the stroboscopic study of single ionization events in Ar [3].

At present, the most advanced theoretical approaches for the description of atoms irradiated by intense light fields are approaches dedicated to two-active-electron systems [4]. However, very few theoretical approaches are currently available to treat complex multielectron atoms, such as Ne and Ar, irradiated by intense ultra-short light fields. Although the R-matrix Floquet (RMF) approach [5] has enjoyed much success at providing multiphoton ionization rates [6], it remains a time-independent approach. As a result it is applicable only to laser pulses involving many cycles of the field. In order to describe atoms irradiated by ultra-short pulses, the full time-dependent Schrödinger equation (TDSE) describing the laser-atom interaction must be solved.

So far, most time-dependent numerical simulations for multielectron atoms have used the single-active-electron (SAE) approximation, in which the electrons are assumed to be effectively independent and only one electron that ionizes is assumed to be 'active'. However, recent experiments have shown that the response of multielectron atoms to ultra-short pulses consists of a coherent response of many electrons [2]. In order to describe such a complex response to ultra-short pulses, an approach is needed which can accurately describe both multielectron atomic structure and the multielectron response to the light field. Based on this demand, we recently introduced an *ab initio* time-dependent approach that employs R-matrix basis functions in a box to investigate multiphoton ionization of complex atoms [7]. The approach has been shown to give accurate results for multiphoton ionization of Ar irradiated by a 390 nm laser pulse [7]. This approach, which consists of an R-matrix inner region only, represented the initial phase of our development of a time-dependent R-matrix (TDRM) theory which was first proposed by Burke and Burke for a 1D problem [8].

In this report we will describe the full 3D TDRM theory. This method can be used to accurately describe the interaction of intense ultra-short light fields with arbitrary multielectron atoms and atomic ions. As a means of demonstrating the capability of an R-matrix-based time-dependent approach, we will discuss recent extensions made to a modified inner region part of our full TDRM method [7], which allows us to investigate momentum distributions of electrons ejected from Ne due to interactions with two ultra-short light pulses.

The time-dependent R-matrix method

The TDRM theory starts from the non-relativistic TDSE describing the interaction of the light field with a general multielectron atom or ion. Throughout this method we assume that the light field is linearly polarized and spatially homogeneous. The time-dependent wave function can be propagated in time by solving the following system of linear equations at each time step t_q:

$$(H_{q+1/2} - E)\Psi_{q+1} = -(H_{q+1/2} + E)\Psi_q, \quad (1)$$

where Ψ_q is the wave function at step q, $H_{q+1/2}$ is the time-dependent Hamiltonian at the midpoint of t_q and t_{q+1}, which is described in the length gauge throughout, and $E = 2i\Delta t^{-1}$, where Δt is the size of the time-step. Equation (1) is an inhomogeneous equation where the right-hand-side can be calculated if the wave function at time $t = t_q$ is known. The solution of this equation then yields the wave function at time $t = t_{q+1}$. Hence this equation enables the solution to be propagated forward in time.

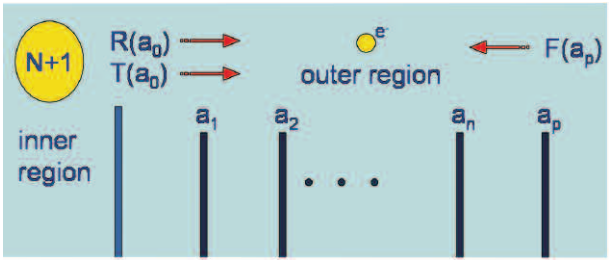

Figure 1. Partitioning of configuration space in TDRM theory.

In our TDRM method the solution of equation (1) is accomplished by partitioning configuration space into an inner and outer region as is shown in figure 1. The conditions used to define the boundary $r = a_0$ between the inner and outer regions are the same as in RMF theory. That is, in the inner region electron exchange and electron-electron correlation effects between the ejected electron and the remaining N electrons are important, while in the outer region electron exchange and correlation effects between the ejected electron and the remaining N electrons

are negligible, and hence the ejected electron moves in the local long-range multipole potential of the residual N-electron atom or atomic ion together with the laser potential. We note that unlike the partitioning scheme in RMF theory, we omit the asymptotic region since we make the outer region large enough so that the ejected electron wavefunction vanishes by the outer boundary $r = a_p$.

In the inner region $r \le a_0$, an R-matrix basis expansion of the wave function describing the $(N+1)$-electron complex is adopted and we use the R-matrix II suite of codes that have recently been adapted to include B-spline basis sets to set up the linear equations given by equation (1). The solution of equation (1) at each time step enables the R-matrix to be calculated on the boundary $r = a_0$ of this region and also enables the calculation of an inhomogeneous T-vector which is due to the to the right-hand-side of equation (1). The T-vector is needed in order to describe the flow of wave function into and out of the inner and outer regions.

In the outer region $a_0 \le r \le a_p$, a set of coupled differential equations describing the motion of the scattered electron in the presence of the light field is solved at each time-step by sub-dividing this region into p sub-regions and propagating the R-matrix and T-vector across them from $r = a_0$ to $r = a_p$ as is shown in figure 1. The R-matrix and T-vector at $r = a_p$ can then be used to propagate the wave function (denoted **F** in figure 1) backwards across the p-sub-regions. This propagated wave function then provides the starting point for the calculation at the next time-step.

Results and discussion

The properties of electrons ejected from Ne by a sequence of attosecond pulses in the presence of a few-cycle near-infrared pulse have recently received significant attention [9]. The experimental results show noticeable interference structures arising from the wavepackets generated by the different ultra-short pulses and it is with these results in mind that we verify the accuracy and capabilities of the inner region part of our full TDRM approach.

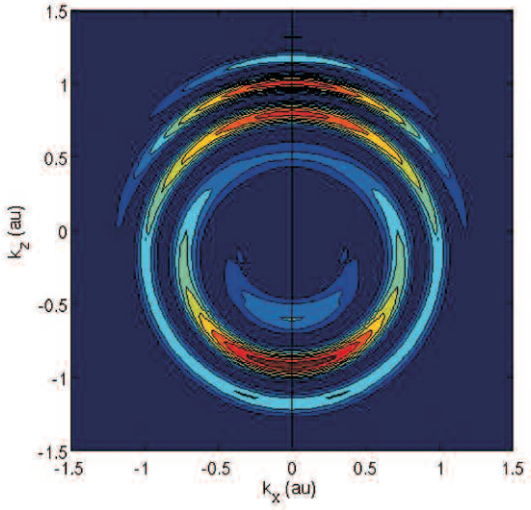

Figure 2. Momentum distributions in the $k_x k_z$-plane for ejection of a 2p electron from Ne irradiated by 390 nm laser light and a sequence of two time delayed ultra-short pulses.

We use a similar R-matrix basis set developed for single-photon ionization of Ne [10]. Although the results reported here include only the ground state of Ne^+ as an ionic 'target' state, results have been obtained with the inclusion of the first excited state of Ne^+ as an additional target state. Details on all the orbital functions can be found in ref. [10]. We use a box size of 500 au with a basis set containing 500 B-splines for each available angular momentum of the ejected-electron. Total angular momenta up to $L = 20$ are included in the calculations. We also include an absorbing potential to prevent unphysical reflections of the wave function from the boundary of the inner region.

The light field in the present calculations consists of a superposition of two fields, linearly polarized in the z-direction: a short 390 nm laser pulse and an ultra-short light pulse. The 390 nm laser pulse has a 3-cylce $\sin^2$ turn-on of the electric field, 2-cycles at constant peak amplitude, followed by a 3-cycle $\sin^2$ turn-off. The ultra-short pulse is made up of combination of four odd harmonics of 780 nm radiation and a Gaussian time envelope. We use a sequence of two ultra-short pulses, the first one occurring at an extremum of the electric field for the 390 nm pulse and the second one occurring half a 390 nm-cycle later. The field in the second ultra-short pulse has the opposite sign to the field in the first ultra-short pulse. The peak intensity of the 390 nm laser pulse is 3.6×10^{13} W/cm^2, while the peak of the ultra-short light pulse corresponds to an intensity of 2.0×10^{12} W/cm^2.

Figure 2 shows the ejected-electron distribution obtained using the R-matrix inner region (RMI) approach. The distribution can be explained by using a simple semi-classical picture in which it is assumed that electrons are only emitted at the peak of an ultra-short pulse. The overall 'ring' structure of the distribution is due to a phase difference gained by the electrons emitted during the first ultra-short pulse with respect to electrons emitted during the second ultra-short pulse, and is a function of the time delay, ΔT between the two ultra-short pulses. The presence of the 390 nm laser field generates a second phase difference between the two pulses. Once a wavepacket is generated, its centre moves under the influence of the 390 nm laser field. At the time that the second wavepacket is generated, the centre of the first wavepacket is stationary with a displacement of $2E_0/\omega^2$ au, (where E_0 is the peak amplitude of the 390 nm electric field and ω is the frequency) which leads to an additional phase factor that accounts for the interference patterns within the 'ring' structures. The sign difference between the two pulses leads to an additional phase difference of π which results in the overall asymmetry of the distribution.

Conclusions

In conclusion, we have presented in this report a new time-dependent R-matrix theory for ultra-fast atomic processes. The new approach is designed to be capable of describing the full multielectron response of complex atoms, such as Ne and Ar, to intense, ultra-short light fields. We have demonstrated the accuracy and capability of a modified inner region part of the approach by determining ejected-electron momentum distributions of Ne irradiated by two

ultra-short pulses in the presence of a 390 nm laser field. It is expected that the full TDRM approach described here, which includes an outer region also, may provide more flexibility than the RMI approach in the investigation of ultra-fast intense field processes. For example, it should be possible to propagate the wave function to much larger radial distances due to the presence of an outer region, which may be needed for the ejection of highly energetic fast electrons. We therefore intend to employ the present full TDRM approach to the study of such ultra-fast processes in the near future.

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