Time-dependent study of Ar in intense few cycle pulse laser fields

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Introduction

One of the major challenges in the understanding of ionisation processes in intense laser fields lies in accurately describing the structure and electron correlation effects of an atom in a laser field in a fully non-perturbative manner. One of the methods that can provide a means to accurately describe electron and photon scattering by complex atoms and ions is the R-matrix approach [1]. In this approach, configuration space is separated into two regions, an inner region in which all interactions are described in full detail, and an outer region, in which only long-range interactions are included.

Several years ago, the R-matrix approach was combined with the Floquet Ansatz [2] to form the R-matrix Floquet (RMF) approach. This approach can describe the atomic structure in sufficient detail to provide accurate *ab-initio* multiphoton ionisation rates for a general atom. It has been used extensively over the last decade to investigate the properties of a range of atoms and ions subjected to intense laser fields. Recently, it has been adapted to use B-spline basis set techniques [3,4]. The results obtained by this approach are highly accurate [5]: a comparison with time-dependent calculations obtained agreement within 10% for (at least) nine-photon ionisation.

Although the RMF approach has enjoyed much success, it remains a time-independent approach. As a result it is applicable only to laser pulses involving many cycles of the field. Another disadvantage of the RMF method is the limited intensity range over which the calculations can be carried out due to the large Floquet expansions required to obtain convergence for high intensities (>10^{14} W/cm^2). For example, in the case of argon subjected to 390 nm light, the maximum intensity for which reliable RMF calculations [6] can be performed is about 2×10^{14} W/cm^2. In this report we describe a time-dependent method that does not suffer from either of the above drawbacks and can therefore be used to describe laser-atom interactions for higher laser intensities and shorter pulses than the RMF technique.

The time-dependent method

The time-dependent method combines the existing R-matrix II suite of codes [7] with B-spline basis sets and represents the first step towards a time-dependent R-matrix (TDRM) theory. This theory was first envisaged a decade ago and applied to a 1-D model potential [8]. For the present calculations, we solve the time-dependent Schrödinger equation in a box using an R-matrix-based inner region basis set. We describe the laser field using the length form of the dipole operator as this choice of gauge emphasizes low-energy excitations and is therefore more suitable for finite basis calculations. The disadvantage of using the length gauge is that many additional angular momenta need to be included in the basis. In the present calculations a maximum angular momentum, $L_{max} = 20$ is needed for the highest intensity studied (3.0×10^{14} W/cm^2).

The time-dependent wavefunction is propagated in time by solving the following system of linear equations at each time step:

$$(1+iH_{n+1/2}\Delta t/2)\Psi_{n+1} = (1-iH_{n+1/2}\Delta t/2)\Psi_n, \quad (1)$$

where Ψ_n is the wavefunction at step n, Δt is the time step and $H_{n+1/2}$ is the Hamiltonian at the midpoint of t_n and t_{n+1}. We typically use a set of 80 B-splines of order $k > L_{max}$ within a box of radius 100 a.u., and include an absorbing potential near the boundary to prevent spurious reflections from that boundary.

In this report we have chosen to present calculations of ionisation rates of Ar for laser intensities in the range 10^{13}-10^{14} W/cm^2 at a wavelength of 390 nm. The availability of reliable *ab initio* results for this wavelength and intensity range [6] allow us to test the accuracy of the approach and to demonstrate the capability of extending the previous RMF calculations to higher intensities. To describe Ar, we have used the R-matrix basis developed [9] for single photon ionization of Ar, using only the ground $3s^23p^5\ ^2P^o$ state of Ar^+ as a target state. We find that the inclusion of the first excited state of Ar^+ as a target state makes a difference of less than 2% to the rates. The description of Ar includes all $3s^23p^5\sum l$ channels up to $L = L_{max}$ and details of all the orbital functions used are given by Burke and Taylor [9].

The laser pulse shape used is a 3-cycle $\sin^2$ turn-on of the electric field, followed by n cycles of an oscillating electric field with constant amplitude, followed again by a 3-cycle $\sin^2$ turn-off. We obtain the ionisation rate by investigating the dependence of the survival probability as a function of n, and also, we separately investigate the decrease in the norm of the wavefunction during the pulse.

Results

Figure 1 shows how the norm of the total wavefunction and the population of the field-free ground state wavefunction vary as a function of time. A constant intensity of 0.75×10^{14} W/cm^2 is maintained for 10 optical cycles so that a well-defined ionisation rate can be obtained. It can be seen that during the constant-intensity period, the wavefunction norm decreases exponentially, with an additional oscillation due to the influence of an excited state. The effect of this state is also seen as the oscillation of the maximum population in the ground state as a function of time due to the resonant transfer of population between the ground and excited state.

In order to illustrate the temporal nature of the laser-atom interaction, figure 2 shows the probability density of the $3s^23p^5\sum s\ ^1P$ continuum channel at different stages of the laser pulse.

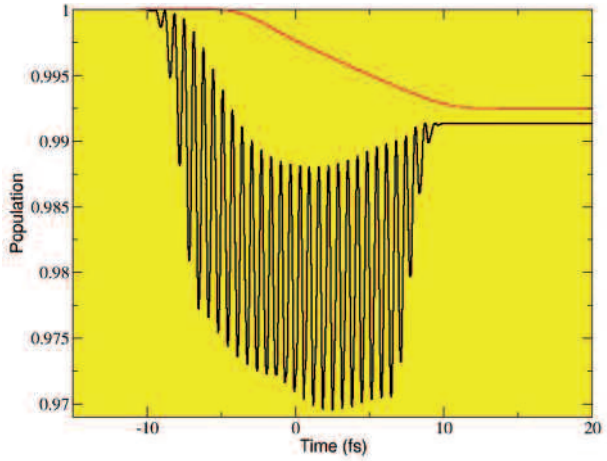

Figure 1. The norm of the Ar wavefunction (red) and the population of the field-free ground-state wavefunction (black) as a function of time. The laser pulse has a constant intensity of I = 0.75×10^{14} W/cm^2 lasting for 10 cycles.

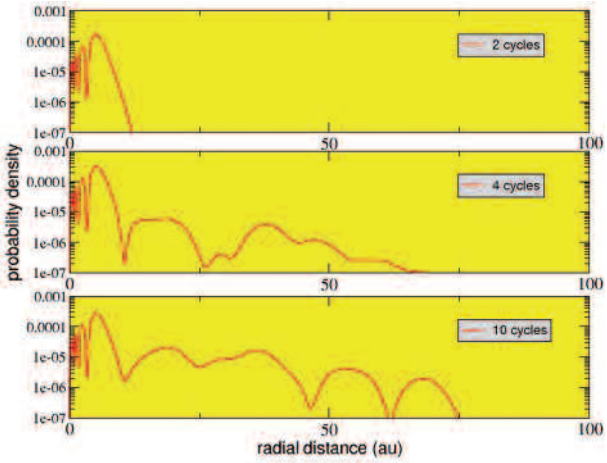

Figure 2. Probability density of the 3s^{2}3p^5∑s ^{1}P continuum channel at three different stages of the laser pulse.

The laser pulse has a constant intensity of 0.75×10^{14} W/cm^2 lasting for 10 optical cycles. It can be seen that the probability density of the channel flows outwards from the core region as a function of time as expected.

Figure 3 shows single ionisation rates for the ground state of Ar as a function of intensity between 0.2×10^{14} W/cm^2 and 3.0×10^{14} W/cm^2, obtained using the RMF [6] and the present time-dependent approaches. At these intensities, single ionisation of Ar requires absorption of at least six photons. The rates obtained by the time-dependent and RMF approaches are in excellent agreement typically well within 10%. However, the limited capability of the RMF technique is clearly evident, as it was difficult to extend the calculations above 2.0×10^{14} W/cm^2. Our new time-dependent approach, on the other hand, has been applied for intensities up to 3.0×10^{14} W/cm^2, with the main limitation on the calculations being the need to include more and more angular momenta for increasing intensities.

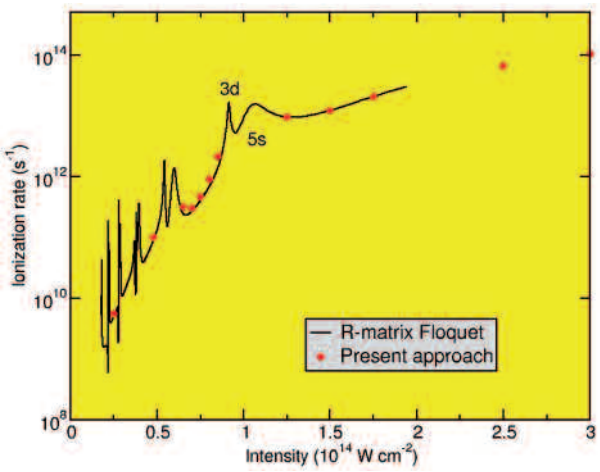

Figure 3. Comparison of ionisation rates for Ar subjected to 390 nm laser light as a function of intensity. Rates from the present approach are compared to those obtained using the RMF approach [6].

Conclusion

In conclusion, we have presented the results of a new non-perturbative time-dependent method that combines B-spline basis techniques with the new R-matrix II inner-region code [7]. We have investigated few-cycle multiphoton single ionisation of argon subjected to 390 nm laser light for various pulse durations and at intensities of up to 3.0×10^{14} W/cm^2 and have found our results to be typically within 10% of ionisation rates obtained by other reliable techniques. We have demonstrated the advantage of our time-dependent method by calculating ionisation rates at higher intensities than are possible using the RMF method, representing a significant advance in the description of complex atoms in intense short laser pulses.

References

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