The R-matrix with time-dependence approach for ultrafast dynamics

Contact: rmorgan12@qub.ac.uk

R. Morgan, O. Hassouneh, H. F. Rey, J. Wragg, A. C. Brown and H. W. van der Hart
Centre for Theoretical Atomic, Molecular and Optical Physics
Queen's University Belfast,
Belfast, BT7 1NN

1 Introduction

The dawn of the attosecond age, and the corresponding strides made in laser technology, have facilitated the experimental investigation of light-stimulated electron dynamics on their own characteristic time-scale [1]. The subsequent need for theory to interpret these results has largely been met by simple, approximate methods which can reproduce the experimental results without necessarily elucidating the complex underlying physics. There are very few methods capable of modeling the laser-atom system – including the important electron correlation effects – which are both accurate and computationally feasible. Here we describe the R-matrix with time-dependence (RMT) approach, which combines state-of-the-art computational techniques with well established atomic physics codes to describe general, multielectron atoms in short, intense laser fields.

2 R- matrix with time-dependence (RMT) approach

The RMT method [3, 4] is the latest in a long line of computational techniques for modeling light-matter interaction developed at Queen's University Belfast. It is an *ab initio* method for solving directly and accurately the time-dependent Schrödinger equation.

R-matrix theory separates configuration space into two distinct regions. In the inner region, all electrons are close to the nucleus. Thus the interactions between them are strong and non-local as the electron-exchange is important. The multielectron wavefunction in this region is constructed from configuration-interaction basis functions and the multielectron laser-atom Hamiltonian matrix elements are calculated explicitly. In the outer region, one ejected electron has become detatched from the residual ion. Thus exchange effects involving this electron are negligible and the electron moves under the action of the laser field and the long range multipole potential of the residual ion. Hence, a one-electron, multichannel problem is solved using a finite difference scheme. In both regions the TDSE is integrated directly, using high-order explicit time propagator methods.

The matching of a finite-difference single-electron wavefunction representation with a multielectron basis set representation at the boundary between regions is the major development of the RMT method. The improved efficiency and accuracy of the code will allow the study of long wavelength, high intensity regimes, bringing calculations in line with laser parameters of real experiments.

3 High-order harmonic generation from Ne^+

High harmonic generation (HHG) is one of the fundamental processes of ultrafast physics and is generally understood in terms of the famous, quasiclassical, three-step model [5]. An electron is ionised and then driven in a strong-laser field before recombining with the parent ion, releasing its energy in the form of a high harmonic photon. This mechanism is the driver behind the latest generation of ultrashort light sources [6].

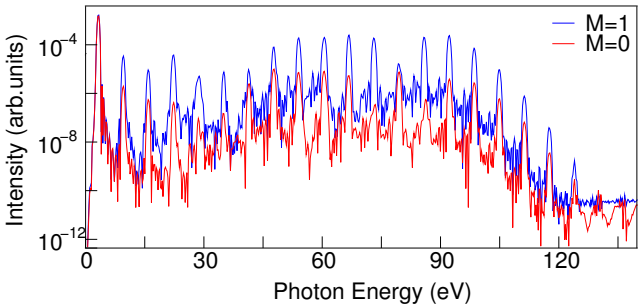

Figure 1: The HHG spectrum of Ne^+ in a 390nm, 10^{15} Wcm^{-2} laser pulse for both $M = 1$ (blue) and $M = 0$ (red) initial alignments.

We have investigated HHG from Ne^+ in 390nm, $10^{15}Wcm^{-2}$ laser pulses. Specifically, we have studied the effect of the initial magnetic alignment of the ion [7]. Figure 1 shows an increase in the HHG yield for $M = 1$ of about a factor 26 compared to $M = 0$. When the ion is initially in the $M = 0$ state, there are no emission channels coupling an ejected, $m_l = 0$ electron with the ground state of the Ne^{2+} ion, meaning the dominant contribution to HHG comes from channels connected to an excited threshold. For $M = 1$ these emission channels are open, and we can associate the observed increase in the HHG yield with a corresponding increase in the contribution from the ground state threshold. Thus, an apparently small change in the atomic structure of the target leads to a significant change in both the observed yield, and the precise mechanism of HHG.

4 Multiphoton ionisation of Ne^+ irradiated by an intense, two-colour laser pulse

A recent experiment investigated double ionisation of neon by a two-colour laser field [2]. We have used the RMT method to investigate the ionisation of Ne^+ to Ne^{2+} by a 2×10^{14}Wcm^{-2} light field with an IR (780nm) and XUV (34nm) component. The XUV pulse promotes an electron to an excited state, and the IR field 'strips off' the electron, ionising the target. The delay between the two pulses determines the final ionisation yield.

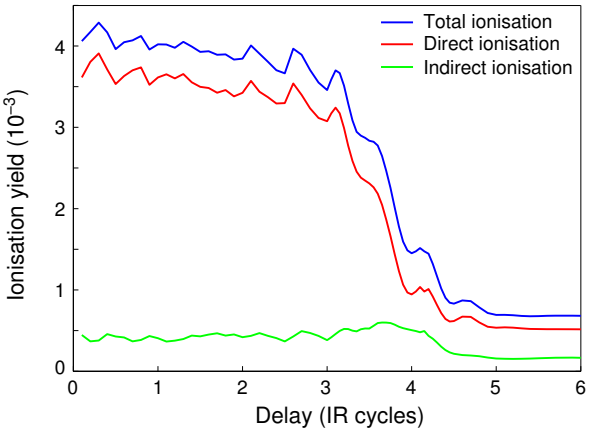

Figure 2: Ionisation yield from Ne^+ in a two-colour laser field as a function of delay (measured in IR cycles) between the two pulses.

The ionsation yields for Ne^+ show similar modulations as observed in experiments on Ne. By analysing the final wavefunction, we can associate these modulations with populations of different residual-ion states. Near the peak of the IR field, the modulations are primarily due to population left behind in $3p$ states. At longer delays, they arise from population trapping in intermediate states for the IR ionisation process.

Figure 2 shows two different ionisation pathways. The Ne^{2+} ion has three accessible thresholds. If population is trapped in a Rydberg state associated with an excited state of Ne^{2+}, ionsation may occur through autoionisation. Figure 2 shows that this, indirect contribution is not insignificant.

5 Electron dynamics in the carbon atom induced by spin-orbit interaction

We have investigated spin-orbit dynamics in the ground state of carbon. By applying RMT to calculate multiphoton ionisation of C by 390nm, 10^{14}Wcm^{-2} laser pulses for different initial magnetic quantum numbers, we can model these dynamics. In experiment, the atom is prepared in a superposition of the different J-levels of the ground state [8]. The spin-orbit induced time-evolution is then obtained by analysing the ejected electron momentum spectra as a function of time delay in a pump-probe scheme. Figure 3 shows that the experimental observations are in good agreement with the theoretical model.

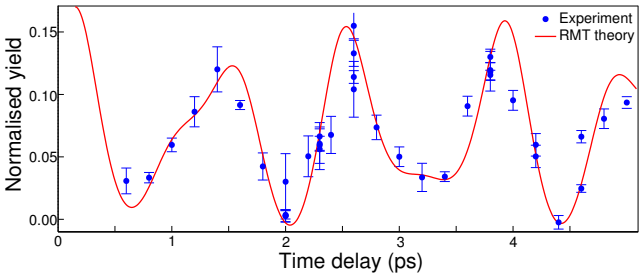

Figure 3: Normalised ionisation yield from carbon as a function of pump-probe time delay for experiment (blue dots) and RMT theory (red line).

6 Ongoing work

The RMT method is currently being extended to treat processes involving the emission of two electrons. This requires a large basis set to describe the doubly ionised states, and an efficient means of treating the interaction between the two, ejected electrons. We recently demonstrated a prototype code capable of describing double ionisation in helium [9]. The method is also being extended to address laser wavelengths in the near- to mid-IR regime. This is a massive challenge for theory, as electrons in such fields can absorb hundreds of photons, obtaining a very high angular momentum. Furthermore, ejected electrons can be driven far from the nucleus, necessitating a massively increased configuration space. Thus the calculations are immensely demanding.

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