The effect of dynamic electron correlation in attosecond pulse generation

Contact mccann@qub.ac.uk

G. A. McKenna and J. F. McCann
Dept. Applied Mathematics and Theoretical Physics, Queen's University Belfast, Belfast BT7 1NN, UK

R. Nepstad
Department of Physics and Technology, University of Bergen, N-5007 Bergen, Norway

D. Dundas
Atomistic Simulation Centre, School of Mathematics and Physics, Queen's University Belfast, Belfast BT7 1NN, UK

Introduction

The development of sources of coherent ultrashort (attosecond timescale) extreme-ultraviolet light is a topic of great interest at present. The aim is to provide a tool that can illuminate, initiate and control ultrafast electronic transitions. While many technical challenges remain in designing an operational facility for such a source, several important fundamental technology breakthroughs have been made.

In particular, the basic generation process of attosecond bursts in the extreme ultraviolet is well developed. Schemes using the conversion of high-intensity infrared laser light through high-order harmonic generation, have been very successfully implemented: delivering harmonic pulses with frequencies in excess of the 300th harmonic [1,2]. These techniques have been refined since, based on developments in pulse shaping of the intense pump laser. In the UK, such schemes are the basis of experimental work being carried out at centres in Belfast (QUB) and London (ICSTM), as well as at RAL (CLF).

Current interest is focusing on how these attosecond bursts might be tailored and optimized. Of course, the efficiency of these processes depends upon the nature of the active medium, as well as the phase, frequency and amplitude profile of the fundamental laser. In this regard, the electronic structure and the dynamic response of the medium is a fundamental component of the device. It follows that, one would assume that electron-electron correlation might play a critical role in the optical response. In the photoionization of atoms by x-rays, for example, this is known to be the case. The effect of correlation, and indeed collective excitation, has been very intensively studied over the decades. However, this level of understanding is certainly not yet achieved for intense-field dynamic hyperpolarizability.

In this report, we investigate dynamic electron correlation in atoms in intense laser fields, and the effect this has on attosecond pulse generation. The ideal system for such a study is the structurally-simplest correlated atom: helium. The pump laser is chosen to be the frequency-doubled Ti:Sapphire laser (λ=390 nm) operating at intensities up to 0.5 PW/cm^2. High-harmonic generation yields are calculated and the effect of the pump phase is studied.

Our conclusions, from these limited simulations, are that (for these conditions) the spectrum is dominated by a low-frequency collective mode together with a high-frequency single-electron response due to the nuclear singularity, both of which dominate electron correlation effects. In the emission, we observe a secondary maximum below the classical cut-off frequency. The imprint of the carrier-envelope phase, for the time-integrated spectral density appears at frequencies above the high-frequency drop-off, consistent with previous studies in the infrared.

Time-dependent density-functional theory (TDDFT)

TDDFT has been applied extensively to the study of atomic and molecular systems driven by external laser pulses [3]. Indeed TDDFT provides one of the most detailed, practical and feasible ab initio approaches for tackling many-body problems. Such effects are, in principle, included exactly through an exchange-correlation functional; in practice the form of this functional is unknown and at best it can only be approximated. Our implementation of TDDFT follows the formalism of Dundas [4].

In the simulation of helium, the two-electron wavefunction is approximated by a one-particle Kohn-Sham orbital, so that the electron density is given by: $n(r,t)=2\left|\Psi_{KS}\right|^2$, where t is the time. The corresponding Kohm-Sham integro-partial-differential equation takes the form:

$$H_{KS}\psi_{KS} = i\hbar(\partial/\partial t)\psi_{KS}$$

The effective Hamiltonian is a combination of the single-particle kinetic energy (T), potential energy due to the nucleus (V_{ion}) the laser field (V_{laser}) the Hartree potential, and an exchange correlation correction (V_{xc}), respectively:

$$H_{KS} = T + V_{ion} + V_{laser} + \int dr' \left| r - r' \right|^{-1} n(r',t) + V_{xc}$$

For a linearly-polarized pump laser, we can exploit the cylindrical symmetry of the system and hence reduce the problem to two spatial dimensions. Then, in terms of the axial coordinate (z), the dipole moment takes the form:

$$d(t) = -2\left\langle \psi_{KS} \middle| z \middle| \psi_{KS} \right\rangle,$$

and the dipole acceleration is calculated using the expression:

$$a(t) = -2\left\langle \psi_{KS} \middle| \left[H_{KS}, \left[H_{KS}, z \right] \right] \middle| \psi_{KS} \right\rangle .$$

The angle brackets denote the two-dimensional spatial integration. Given an expression for the acceleration as a function of time, the spectral emission is derived from Larmor's formula. Taking the Fourier transform gives the single-atom spectral density response for harmonic generation.

A crucial element in our model is the functional form of the exchange-correlation potential. While many sophisticated approximations to this potential have been developed, the simplest is the adiabatic local density approximation in the exchange-only limit (xLDA). While this approximate functional is simple to implement, it does suffer from the drawback of containing long-range self-interaction errors: the asymptotic form of the potential is exponential instead of Coulombic. The anomalous long-range form of the self-interaction potential means that the spectrum of single-particle highly-excited states are incorrect.

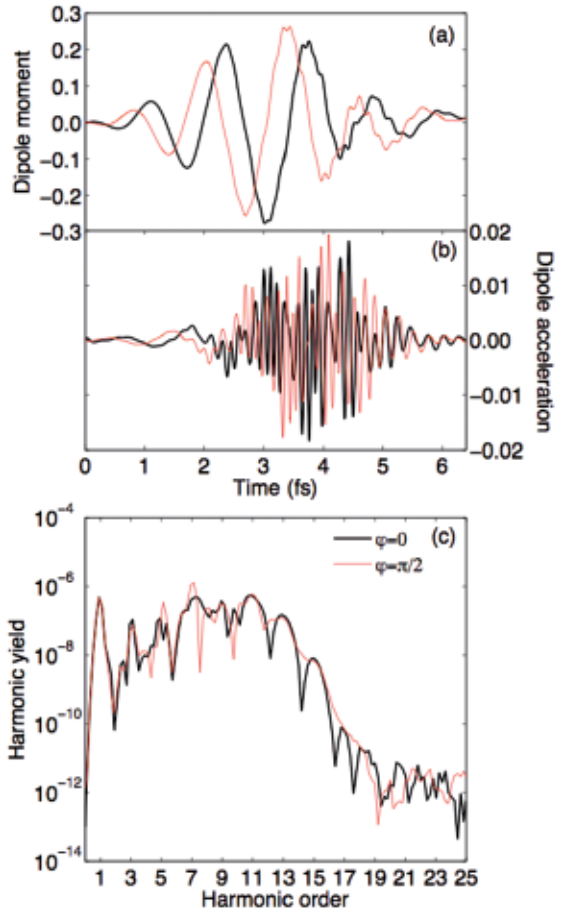

Figure 1. Theoretical results (TDDFT) for single-atom helium response. The pump laser is a five-cycle (6.4 fs) pulse at λ=390 nm with intensity of 5×10^{14} W/cm^2. The black (dark) line corresponds to a carrier-envelope phase 0. The red (light) line corresponds to a phase π/2. Figure (a) displays the dipole moment, (b) the dipole acceleration and (c) the spectral density. Atomic units are used for the dipole moments. The spectral density is in arbitrary units.

Nonetheless the short-range potential which is critical for the dipole acceleration, is well reproduced by the model exchange correlation.

Results

We consider a pulse with a sine-squared profile (envelope) with a five-cycle (6.4fs) duration. The pump laser wavelength is λ=390 nm and the intensity chosen for an illustration of the dynamics is 5×10^{14} W/cm^2. In Fig. 1(a) the dipole moment represents the centre-of-charge of the electron pair in the helium atom. The imprint of the amplitude and phase of the external field is clearly visible in this figure, corresponding to quasi-static polarization. This centre-of-mass motion is rather insensitive to internal forces, and hence dynamic correlation effects will not have a strong part to play. The carrier-envelope phase can be adjusted to provide a sub-femtosecond probe. In this case, we note the spectral emission corresponding to this motion (the fundamental mode) does not depend on the carrier phase. This can be seen in Fig 1(c) where the dominant fundamental peak is identical for the black (dark) and red (light) lines.

The high-frequency quiver motion superimposed on the low-frequency mode is more clearly visible in the acceleration results presented in Fig 1(b). These small-amplitude oscillations, occurring on timescales that are fractions of a femtosecond, give rise to the attosecond pulses. These bursts are localized in time at the peak of the electric field. A deeper analysis [5] reveals that the highest-frequency bursts are also localized in space since they are primarily created by the nuclear singularity (V_{ion}), where the restoring force is strongest, the electron density is highest, and the electron acceleration is largest. Thus the effective Hamiltonian has the character of single-electron (uncorrelated) form for the ultraviolet emission. We carried out equivalent calculations using the simple single-active electron (SAE) model to confirm this conjecture [5]. Our results, comparing TDDFT and SAE models are in very good quantitative agreement for the intensity of the high-frequency peaks, even though the models treat electron-electron correlation in quite different manners. Regarding the shape of the spectrum, in Fig. 1 (c), rather than a classical plateau in the high-frequency range, we observe a dip followed by a secondary maximum. This is a feature of the short pulse employed in the simulations.

Baltuska *et al.* [6] studied the dependence of high-harmonic generation on carrier-envelope phase. Their experiments with helium, using the fundamental Ti:Sapphire mode in the infrared λ = 800 nm, showed that, provided the phase was chosen to coincide with the peak of the envelope, the spectrum generated in the cut-off region loses the odd-order harmonics. These results were supported by theoretical simulations in the same letter (fully-dimensional, fully-correlated). We examined whether this holds true for our two models (TDDFT and SAE) at a higher pump frequency.

Examination of the results, Fig. 1 c, at frequencies above the 15th harmonic in the drop-off region, shows that this is reproduced by our TDDFT simulations for λ = 390 nm. In the drop-off region, the spectrum is continuous when the phase is π/2, and has a harmonic modulation when the phase is 0. Of course,

confirmation of this finding from a full-dimensional two-electron simulation and experimental measurements would be important for extensions to other atoms and molecules. Further work from our group will focus on using different atoms/ions/molecules to determine optimal targets for attosecond generation.

Acknowledgements

GAMcK is grateful to EPSRC for the financial support of this work through a PhD studentship (grant EP/C007611/1). We thank Tore Birkeland (Bergen) for his assistance and discussions on this project.

References

1. Z. Chang, A. Rundquist, H. Wang, M. M. Murnane and H. C. Kapteyn, *Phys. Rev. Lett*. **79**, 2967 (1997).
2. M. Schnürer *et al*., *Phys. Rev. Lett*. **80**, 3236 (1998).
3. X. Chu and S.-I. Chu, *Phys. Rev. A* **63**, 023411 (2001); T. M. Uhlmann *et al*., *Phys. Rev. A* **67**, 013413 (2003); F. Calvayrac *et al*., *Phys. Rep*. **337**, 493 (2000).
4. D. Dundas, *J. Phys. B: At. Mol. Opt. Phys*. **37**, 2883 (2004).
5. G. A. McKenna *et al*., *Phys. Rev. A* submitted (2009).
6. A. Baltuska *et al*., *Nature* **421**, 611 (2003).