

Tracking individual electron trajectories in a high harmonic spectrum

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Received 6 December 2005; accepted 28 March 2006

Abstract

The harmonic spectrum generated by a few-cycle laser pulse propagating through a gas jet is calculated. Two complementary atomic response models are used, one based on the time-dependent Schrödinger equation and the other on quantum orbits in which only a single pair of electron trajectories are taken into account. The role played by individual electron trajectories in determining particular features of the spectrum are considered and phase-matching maps are used to help understand their structure. A method based on this connection is proposed for diagnosing the carrier-envelope phase and the pulse duration of the incident laser field.

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PACS: 32.80.Rm; 42.65.Ky; 42.65.Re

Keywords: High harmonic; Ultrafast; Attosecond

1. Introduction

High order harmonic generation (HHG) was first observed some 20 years ago [1], shortly after its key characteristics were predicted by Shore and Knight [2]. These authors realised that the continuum–continuum transitions responsible for above threshold ionisation (ATI) must lead to a non-linear response displaying a relatively high efficiency plateau in the harmonic spectrum.

Since that early work, the field has undergone rapid growth with many impressive observations and new theoretical understanding. For instance harmonic photons of >1 keV have recently been reported [3] and conversion efficiencies in the 10^{-4} region with micro-joule level XUV pulse energies have been demonstrated [4]. HHG has now been confirmed as a highly versatile source of XUV radiation and a number of important applications are now recorded [5,6]. Recently it was suggested, and confirmed,

that the time structure inherent to the HHG process leads to trains of sub-femtosecond (attosecond domain) light pulses. There is much current interest in this area since sources of, particularly isolated, attosecond pulses are being used and optimised for ultra-fast measurement, with a view to measuring electron motion in matter in real time.

To understand most directly the temporal structure of HHG, one can adopt the widely used strong field approximation theory, which is based on a simple semi-classical picture [7,8]. The approach is valid when the Keldysh parameter $\gamma = \sqrt{I_p/2U_p} \lesssim 1$ where I_p is the ionisation potential of the atom and $U_p = (eE)^2/4m_e\omega_0^2$ is the ponderomotive potential of the electric field of which ω_0 is the central frequency; for 800 nm radiation this implies that the intensity should exceed 100 TW cm^{-2} . The process of HHG in the strong field limit can be seen as a three-stage process: (1) tunnel ionisation of the most weakly bound electron near the local peak of the electric field; (2) acceleration of the electron in the time-varying laser field, leading to recollision with the atomic core about two thirds of a cycle later for electrons born at the correct phase; and (3) recombination of the continuum electron into the bound electronic state accompanied by the emission of HHG

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radiation [9]. The time structure of HHG emerges naturally within this picture, since the period of emission is inherently brief (typically a few hundred attoseconds). The formulation of this theory in terms of electron trajectories [10] provides a powerful insight into the quantum physics and is adopted here.

In this paper, we explore the explicit connection between the quantum trajectories of a single atom in a strong few-cycle laser field with the full spatio-spectral behaviour that emerges after propagation through the macroscopic medium. In this way it is possible to relate directly the observed spectrum to the inherent properties of the optical field. This investigation has several objectives aimed at improving the reliable production of isolated attosecond pulses in the laboratory. Firstly, the understanding of the spectral and spatial properties of the harmonic radiation will guide the experiments designed to generate higher brightness beams of harmonic radiation. Secondly, the need to employ few-cycle pulses to generate isolated attosecond XUV pulses inherently makes the process highly sensitive to the amplitude and carrier-envelope phase (CEP) of the pulse. Controlling these parameters is extremely important for stable performance of attosecond sources. It would therefore be logical if these parameters could be directly extracted from the harmonic spectrum.

Earlier work established the connection between the single atom dipole response and the fields generated after propagation, see e.g. [11–14]. In this paper, phase-matching maps are used to relate particular quantum paths in the single-atom response to features in the fully propagated HHG spectrum, in particular the radial distribution of the harmonic field. The extension here is the ability to link experimentally observable characteristics to specific atomic events. As these quantum paths are directly sensitive to CEP this report looks at how the HHG spectrum could be used for the diagnosis of few-cycle IR pulses.

2. Modelling – single atom

The non-linear polarisation response of the medium is calculated using two single atom models: an *ab initio* calculation of the time-dependent Schrödinger equation (TDSE) and a quantum orbit model. Using both approaches in parallel provides a more complete understanding of the physics involved. Solving the TDSE gives a fuller description of the system, while the quantum orbit approach, which extracts information about specific electron trajectories, offers greater physical insight.

Constraints on computational time requires the TDSE to be solved in only one-dimension, reducing the TDSE to

$$i \frac{\partial \Psi(x, t)}{\partial t} = \left(\frac{1}{2} [p_x + A(t)]^2 + V(x) \right) \Psi(x, t). \quad (1)$$

Atomic units ($\hbar = 1$, $e = 1$, $m_e = 1$, $a_0 = 1$) are used here and throughout. In our calculation the Coulomb potential of the atom is approximated by a one-dimensional soft-core potential $V(x) = -1/\sqrt{x^2 + a^2}$ where a is chosen such

that the ionisation potential, I_p , matches that for neon ($I_p = 0.79$ a.u.), which is the gas used in all the calculations in this report. The vector potential of the linearly polarised laser field is defined as

$$A(t) = (E_{\max}/\omega_0)h(t) \sin(\omega_0 t + \phi), \quad (2)$$

where $h(t)$ is the pulse envelope function, ω_0 the carrier frequency and ϕ the CEP of the field. The single atom dipole acceleration is given by $\ddot{D}(t) = \langle \Psi | -\partial_x V(x) + E(t) | \Psi \rangle$, where $E(t)$ is the electric-field of the laser.

The quantum orbit model is based on the work of Lewenstein et al. [9], and uses the saddle point method to select a finite number of orbits which can be summed up to approximate the entire integral, see e.g. [15,16]. This allows individual or groups of trajectories to be studied in isolation. The complex amplitude of the saddle point solution s for emission of a harmonic with frequency ω , with polarisation $\hat{\epsilon}$ (parallel to the laser field in this report), is given by,

$$x_s(\omega) = \frac{2\pi i}{\sqrt{\det S''(t, t')|_s}} \left[\frac{\pi}{\varepsilon + \frac{i}{2}(t_s - t'_s)} \right]^{\frac{3}{2}} \hat{\epsilon} \cdot \vec{d}^*[\vec{p}_s + \vec{A}(t_s)] a(t_s) \times \vec{E}(t'_s) \cdot \vec{d}[\vec{p}_s + \vec{A}(t'_s)] a(t'_s) e^{i(S(t_s, t'_s) + \omega t_s)}, \quad (3)$$

$$\det S''(t, t')|_s = \left(\frac{\partial^2 S}{\partial t \partial t'} \Big|_s \right)^2 - \frac{\partial^2 S}{\partial t^2} \Big|_s \frac{\partial^2 S}{\partial t'^2} \Big|_s \quad (4)$$

where t'_s and t_s are the ionisation and recollision times respectively,

$$p_s = -\frac{1}{t_s - t'_s} \int_{t'_s}^{t_s} A(\tau) d\tau \quad (5)$$

is the continuum state drift momentum, $a(t) = \exp(-\int_{-\infty}^t w(\tau) d\tau)$ is the ground state population at time t , $w(t)$ is the quasistatic ionisation rate calculated using ADK theory and ε is a small non-negative correction factor. $\vec{d}[\vec{p}] = \langle \vec{p} | \vec{r} | \psi_0 \rangle$ is the dipole matrix element for an electron to make a transition from the ground state to the Volkov state $|p\rangle$. The ground state is approximated as a hydrogenic s-orbital, giving,

$$\vec{d}[\vec{p}] = -\frac{i8\sqrt{2}(2I_p)^{5/4}}{\pi} \frac{\vec{p}}{(p^2 + 2I_p)^3}. \quad (6)$$

The action is given by

$$S(t, t') = -\int_{t'}^t d\tau \left(\frac{[\vec{p}(t, t') + \vec{A}(\tau)]^2}{2} + I_p \right). \quad (7)$$

Quantum orbits correspond to the paths followed by the majority of the electronic wavefunction responsible for HHG and so provide a useful insight into the process. Unless the contributions from individual orbits are required the solutions to the saddle point equations are summed up using the uniform approximation, see e.g. [15,16]. Thus the time-dependent atomic dipole acceleration is given by $\ddot{D}(t) = -\int \exp(i\omega t) \omega^2 X(\omega) d\omega$, where $X(\omega)$ is the summation of quantum orbit solutions, $x_s(\omega)$.

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