Tracing quantum trajectories of electrons in interaction with arbitrary-shape laser pulses

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Abstract. In this paper we present a theoretical and numerical tool for tracing quantum trajectories of electrons in interaction with arbitrary-shape laser pulses. The method calculates quantum trajectories using saddle-point analysis within the strong field approximation. The method is very general, thus it is applicable to arbitrarily shaped finite laser pulses. Example results and comparison with the classical trajectories are presented.

1. Introduction

High-order harmonic generation (HHG) is the phenomenon produced as a result of the interaction of atomic (molecular) systems with strong laser pulses. A suitable model that is simple enough but very successful in explaining the main processes involved in HHG is the well-known three step model [1]. Under the influence of an intense (> $10^{13}$ W/cm$^2$) laser field an electron first tunnels through the Coulomb barrier, becomes a free electron travelling in the laser field, then recombines with the parent system and emits a photon of high energy. This energy equals the kinetic energy gained by the electron during its excursion guided by the laser field plus the atomic/molecular ionization potential ($I_p$). Due to the interference between coherently emitted photons, only odd order harmonics of the fundamental radiation are obtained. Since the first and third steps in the three step model, namely the tunnel ionization and recombination are typical quantum phenomena, but the excursion of the electron in electromagnetic field is fairly a classical process, a reasonable approximation in the theoretical treatment of the HHG process is the strong-field approximation (SFA) [2]. This model relies on the basic assumption that the incoming laser field is much stronger than the atom’s Coulomb potential, thus during its travel the electron is driven solely by the electromagnetic field.

A great deal of information and phenomenological insight regarding the mechanisms leading to HHG can be obtained by examining the trajectories of electrons contributing to harmonic photon emission. Remarkable papers were published on this topic [3, 4, 5, 6] using linearly and elliptically polarized laser radiation, or even bi-circular fields. In all these cases the authors report results obtained from analytical calculations and numerical simulations assuming that the radiation propagates as a plain wave and its electric field component has an expression that can be handled analytically. Moreover, the envelope of the pulse is not included, constant amplitude is assumed for all optical cycles.

In experiments and practical applications, however, atomic systems (dilute gases) are irradiated with very short (several tens of femtoseconds), intense, low frequency (for example a
$T_i: Sa$ laser operates at $\approx 800$ nm central wavelength) laser pulses which are usually tailored to a complex shape and spatial-spectral profile in order to meet several requirements. Analytical calculations completely fail when one wants to trace electron trajectories realized in such realistic fields, and finding the suitable numerical method is not a trivial task. The purpose of the present paper is to present such a numerical tool and first results obtained for quantum trajectories of electrons in arbitrary laser fields, and to discuss and interpret them in comparison with the classically obtained trajectories.

2. Model and method

2.1. Strong-field approximation and saddle-point equations

The harmonic spectrum is obtained in the framework of the SFA as the Fourier transform of the dipole moment:

$$\vec{A}(t) = i \int_0^t dt' \int d^3\vec{p} \cdot \tilde{A}^*(\vec{p} - \vec{A}(t)) \cdot \exp(-i S(\vec{p}, t, t')) \cdot \vec{E}(t') \cdot \tilde{d}(\vec{p} - \vec{A}(t)),$$

where in the complex phase term ($\exp(-i S(\vec{p}, t, t'))$) the quantity $S(\vec{p}, t, t')$ is the quasiclassical action:

$$S(\vec{p}, t, t') = \int_{t'}^t dt'' \left( \frac{|\vec{p} - \vec{A}(t'')|^2}{2} + I_p \right),$$

with $\vec{p} = \vec{v}(t) + \vec{A}(t)$ the canonical momentum, $\vec{A}(t)$ is the vector potential of the field $-\partial \vec{A}(t)/\partial t = \vec{E}(t)$, where $\vec{E}(t)$ is the electric component of the electromagnetic field.

A valid method to describe quantum trajectories within the SFA and dipole approximation is to perform saddle-point analysis of the quantum-mechanical transition amplitude [7]. The most probable trajectory of an electron that ionizes at time $t$, travels in the laser field, then recombines at time $t'$ occurs under the condition of stationary phase. The stationary phase condition leads to the following saddle-point equations:

$$\nabla_{\vec{p}} S(\vec{p}, t, t') = 0 \Rightarrow \frac{\vec{\alpha}(t) - \vec{\alpha}(t')}{{t'} - t} = \vec{p}_s$$

$$\frac{\partial}{\partial t} S(\vec{p}, t, t') = 0 \Rightarrow |\vec{p}_s - \vec{A}(t')|^2 = 2I_p$$

$$\frac{\partial}{\partial t'} S(\vec{p}, t, t') = 0 \Rightarrow |\vec{p}_s - \vec{A}(t')|^2 = 2(n\omega - I_p),$$

with $\vec{\alpha}(t) = \int_t \vec{A}(t')dt$ and $\vec{p}_s$ the stationary canonical momentum which is a constant of motion.

The first saddle-point equation (3) expresses the return condition for the electron, contains the assumption that only those electrons contribute to the harmonic spectrum which return and recombine to the nucleus. The second (4) and third (5) equations represent the energy conservation at the moment of ionization and recombination, respectively. We need to solve the above nonlinear system of equations for variables $\vec{p}_s, t$ and $t'$. Since $I_p < 0$ it results immediately that all the quantities $\vec{p}_s, t, t'$ will be complex. Rearranging the equations and writing them for the real and imaginary parts, the system of equation becomes a set of four equations with four unknowns: $t_{re}, t_{im}, t'_{re}, t'_{im}$.

It can be assigned a physical meaning to the complex quantities $\vec{p}_s, t, t'$. The fact that they become complex is a natural consequence of including in the model the pure quantum phenomenon of tunneling. The imaginary part of $t$ “birth” time can be interpreted as tunneling time, while the imaginary part of $t'$ recombination time is related to the probability of the event. One expects that the real parts of the complex electron trajectories obtained after solving the saddle-point equations to be close to those trajectories obtained by pure classical calculations (see 2.2).
2.2. Numerical procedure

The starting point in the proposed numerical method for obtaining the electron’s complex trajectories is the calculation of classical electron trajectories. This is a fair starting point, since it roughly indicates the range where the more realistic quantum trajectories have to be looked for. Classical electron trajectories are obtained by solving Newton’s equation of motion with the return condition for the electron included. The resultant expression for the classical trajectory is [8]:

\[ \vec{r}(t') = \vec{r}_0 + \vec{\alpha}(t') - \vec{\alpha}(t) - \vec{A}(t)(t' - t) \]  

(6)

In the first step we obtained, starting from the electric field given in numerical form, the quantities involved in the calculations, namely \( \vec{A}(t) \) and \( \vec{\alpha}(t) \). The next step is to solve the system of nonlinear equations for \( t_{re}, t_{im}, t'_{re}, t'_{im} \) and for a given harmonic order, the initial guess being the solution of the classical equation of motion. For this purpose we used appropriate subroutines from the NAG numerical package [9] for solving nonlinear systems of equations.

Our purpose was to work out a general trajectory-finding method for arbitrary laser fields, that is, a field which can be handled numerically. A basic requirement is to be able to define and calculate the quantities involved (\( \vec{E}(t), \vec{A}(t) \) and \( \vec{\alpha}(t) \)) at a complex time. This was performed through a power series expansion starting from \( \vec{E}(t) \) as a function of the real variable \( t \).

3. Results and discussion

Intense research is going on both on theoretical/modeling and on experimental side to obtain single attosecond bursts [10, 11]. For this purpose laser pulses of different complex shapes have to be produced. One particular choice within the so-called polarization gating technique was recently proposed [12]. The results for quantum electron trajectories presented here are obtained by using the laser fields and parameters defined in [12], which induce a polarization gating in the leading edge of the pulse.

**Figure 1.** Short and long electron trajectories which generate the 35th harmonic. Black lines stand for quantum trajectories, red lines for classical trajectories. Travel times in units of optical cycles are 0.46 and 0.93 for the quantum trajectories; 0.48 and 0.91 for the classical short and long trajectories respectively.

**Figure 2.** Electron trajectories near the cut-off which generate the 55th harmonic. Black lines stand for quantum trajectories, red lines for classical trajectories. Travel times in units of optical cycles are 0.62 and 0.79 for the quantum trajectories; 0.67 and 0.75 for the classical trajectories.
Figure 1. shows typical electron trajectories that generate plateau harmonics, while Figure 2. represents trajectories generating harmonics close to the cutoff. As general observations, valid for both cases we can affirm the followings: (i) Both graphs represent trajectories produced right after (almost) perfect linear polarization is lost. This is the explanation for the loop-like quantum trajectories instead of a straight line which would have occurred in case of linearly polarized resultant field. (ii) Although the travel times of classical and quantum trajectory pairs are not very different, their actual itineraries differ substantially. The explanation for this lies in the fundamental difference between the two models. When we include quantum tunneling effect in the model, the ionized electron appears in continuum with non-zero initial velocity, contrary to the zero initial velocity assumption of the classical calculations. (iii) Due to the tunneling effect the electron appears in the continuum at a non-zero $r_0$ position, but it returns almost exactly to zero (the assumed position of the nucleus). This realistic feature is not reproduced by the classical electron trajectories, they seem to recombine much farther from the nucleus then their birth position.

In the case of plateau harmonics (Figure 1.) the difference between short and long quantum trajectories is qualitative. Electrons appear in the continuum at different positions and with different initial velocities which predefine their whole excursion. As we approach the cutoff, differences between shorter and longer trajectories decrease both as birth time and initial velocity (Figure 2.). Exactly at the cutoff we would obtain one single family of trajectories with travel time $\approx 0.67$ optical cycles in the interval when the linear polarization gate is open. Since in classical calculations it is assumed that electrons appear in the continuum near the origin (nucleus) with zero initial velocity, classical trajectories do not show any qualitative change in their shape, except the duration of the travel times.

4. Conclusions
We have developed a novel general method for tracing quantum trajectories of electrons in interaction with strong low-frequency and arbitrary-shape laser fields. With this method we can obtain supplementary information concerning the conditions in which a pulse of the harmonic order of interest is generated. In particular, from the phase of the trajectory one can extract in which conditions phase matching is obtained.

As it was shown on the results, by including the quantum effect of tunnelling in the model, the obtained electron trajectories became significantly more realistic and accurate. The first results obtained with the help of this new instrument were presented here. More elaborated results reproducing different experimental conditions are left for further publications.

References
[1] Corkum P B 1993 Phys. Rev. Lett. 71 1994
[2] Lewenstein M et. al. 1994 Phys. Rev. A 49 2117
[3] Kopold R, Becker W and Kleber M 2000 Optics Communications 179 39
[4] Milošević D B 2000 J. Phys. B: At. Mol. Opt. Phys. 33 2479
[5] Milošević D B, Becker W and Kopold R 2000 Phys. Rev. A 61 063403
[6] Milošević D B, Paulus G G and Becker W 2005 Phys. Rev. A 71 061404(R)
[7] Salitri P et. al. 2001 Science 292 902
[8] Milošević D B, Paulus G G and Becker W 2003 Opt. Express 11 1418
[9] See the library documentation on www.nag.co.uk
[10] Tosa V, Yakovlev V S and Krausz F 2008 New Journal of Physics 10 025016
[11] Kienberger R et. al. 2004 Nature 427 817
[12] Altucci C, Esposito R, Tosa V and Velotta R 2008 Optics Letters 33 2943.