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Coupled-coherent state approach for high-order harmonic generation

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Abstract: We present the first ever computation of HHG spectra using the orbit-based Coupled-Coherent State (CCS) method, whose outcome exhibits a plateau and a cutoff. The CCS fully accounts for quantum interference and the binding potential.

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1. Introduction

High-order harmonic generation (HHG) is a phenomenon in which matter responds highly nonlinearly to an input near-infrared field. Both experimental and theoretical studies have shown that a typical HHG spectrum has a general character: It decreases rapidly for the first few harmonics, then exhibits a broad plateau, and finally ends up with a sharp cutoff. HHG can be well understood by means of the well-known semiclassical three-step model [1] involving ionization, acceleration, and recombination. At present, several theoretical approaches have been established to compute HHG spectra. The numerical solution of the time-dependent Schrödinger equation contains no physical approximation and is straightforward for one-dimensional, one-electron systems, but it is not applicable to a three-dimensional, complex system, as the numerical effort increases exponentially with the number of degrees of freedom. Furthermore, it lacks the clarity of an orbit-based picture. Semi-analytical approaches, such as strong field approximation (SFA), provide a transparent physical picture for the quantum interference in this phenomenon [2]. The SFA, however, is based on a series of assumptions which are not justified and over-simplify the problem, such as neglecting the influence of the binding potential in the electron propagation in the continuum, or the internal structure of the target. This poses serious difficulties if the structure of the system becomes important.

To overcome the above-mentioned problems, we need an orbit-based method with no simplification on the target/binding potential. The coupled-coherent state (CCS) method developed by Shalashilin and Child [4,5], which exploits the properties of trajectory-guided grids of Gaussian wave packets (i.e. coherent states) as a basis set for the coupled quantum equations, fully accounts for the residual binding potential and makes no simplifying assumption on the target. Furthermore, CCS has the following advantages 1) it can be extended to more degrees of freedom such as multi-electron atoms and molecules; 2) the initial state can be chosen randomly; 3) in the coupled equations some cancellations will appear which simplify the quantum mechanical coupling terms; 4) the motion of electrons can be guided by classical mechanics; 5) It accounts for quantum mechanical effects such as tunneling and quantum interference.

2. Theory

A coherent state (CS) of a quantum harmonic oscillator is an eigenstate of the annihilation operator $\hat{z}$ with eigenvalue z ,

$$\hat{z}\left|z\right\rangle = z\left|z\right\rangle \;\; , \left\langle z\right|\hat{z}^{+} = \left\langle z\right|z^{*} . \tag{1}$$

The quantum trajectories are determined by the Hamiltonian with quantum corrections where,

$$\frac{dz}{dt} = -\frac{i}{\hbar}\frac{\partial H_{ord}(z^{*},z)}{\partial z^{*}}, \;\; \frac{dz^{*}}{dt} = \frac{i}{\hbar}\frac{\partial H_{ord}(z^{*},z)}{\partial z} \tag{2}$$

$H_{ord}(z^{*},z) = \left\langle z\right|\hat{H}\left|z\right\rangle$ represents the diagonal elements of the Hamiltonian matrix and corresponds to the classical propagation.

The Schrödinger equation in the coupled-coherent state representation (CSR) is

$$\frac{d\left\langle z\middle|\Psi(t)\right\rangle}{dt} = \frac{1}{\pi}\int\left\langle z\middle|z^{'}\right\rangle\left[\frac{i}{\hbar}\frac{dS_{z}}{dt} - \frac{i}{\hbar}\delta^{2}H_{ord}(z^{*},z)\right]\left\langle z^{'}\middle|\Psi(t)\right\rangle d^{2}z^{'}, \tag{3}$$

where $\Psi(t)$ is an arbitrary wavefunction, $S_{z} = \int\left[\frac{i\hbar}{2}\left(z^{*}\frac{dz}{dt} - \frac{dz^{*}}{dt}z\right) - H_{ord}\left(z^{*},z\right)\right]dt$ is the classical action along

the trajectory, and $\delta^{2}H_{ord}(z^{*},z) = H_{ord}(z^{*},z^{'}) - H_{ord}(z^{*},z) - \dfrac{\partial H_{ord}(z^{*},z)}{\partial z}(z^{'}-z)$ couples different coherent states z and $z^{'}$.

Physically, this term allows for quantum interference to occur.

The HHG spectrum can be obtained by the Fourier transform of the dipole $D(t)$

$$S(\omega) \sim \left|\int\exp(-i\omega t)D(t)dt\right|^{2}, \tag{4}$$

where $D(t)$ is the expectation value of the dipole operator. We compute this expectation value in the CCS basis, and consider the length, velocity and acceleration forms of the dipole operator.

3. Results

The results that follow are for Hydrogen in a trapezoidal laser field. As a starting point, we use a 1-dimensional target. We assume the initial electronic wave packet to be localized at the electron quiver distance instead of the origin. This simplifies the problem considerably, as the ionization step is removed and one is able to focus solely on the recombination step, and leads to clean spectra with well-defined odd harmonics. In this case, the HHG cutoff is located at $I_{p} + 2U_{p}$ instead of the standard cutoff energy of $I_{p} + 3.17U_{p}$ associated with a wave packet starting from the core. Similar assumptions have been employed by van de Sand and Rost [4]. Therein, it is also stated that the plateau is a quantum interference effect. In order to verify this statement, we calculated the dipole moment in the acceleration form (left panels in Fig. 1 and Fig. 2) and the corresponding high-order harmonic spectrum (right panels in Fig. 1 and Fig. 2) by considering both the diagonal and off-diagonal parts and diagonal part only of the dipole expectation value in the CCS basis. Our computations show that, if the off-diagonal part is included, there are high-frequency oscillations in the acceleration, which come from the quantum interference

between different trajectories and lead to the HHG plateau. This result agrees with the main conclusion in Ref. [4].
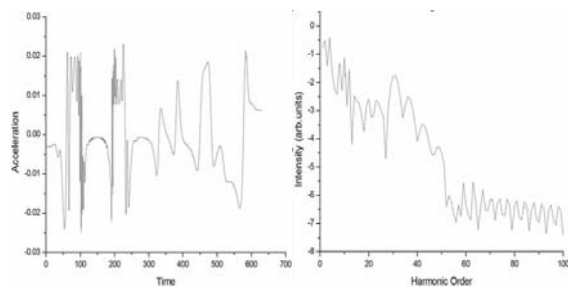

Fig. 1 Dipole acceleration and corresponding HHG spectrum (left and right panels, respectively) computed in the CCS basis for Hydrogen in a laser field of strength 0.05 a.u. and frequency 0.05 a.u., including the diagonal and off diagonal terms in the CCS basis.
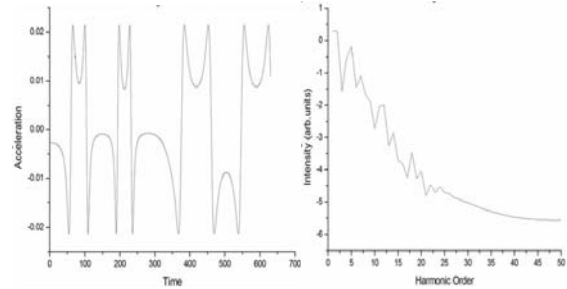
Fig. 2 Dipole acceleration and HHG spectra for the same parameters as in the previous figure, but considering only the diagonal terms in the CCs basis.

In Fig. 2, we go beyond a one-dimensional model and compute HHG spectra for an initial three-dimensional wavepacket located at the quiver distance, for the same wavapacket and field parameters as in [4]. These results show that a clear HHG spectrum with cut off located at about $I_p + 2U_p$ is obtained.
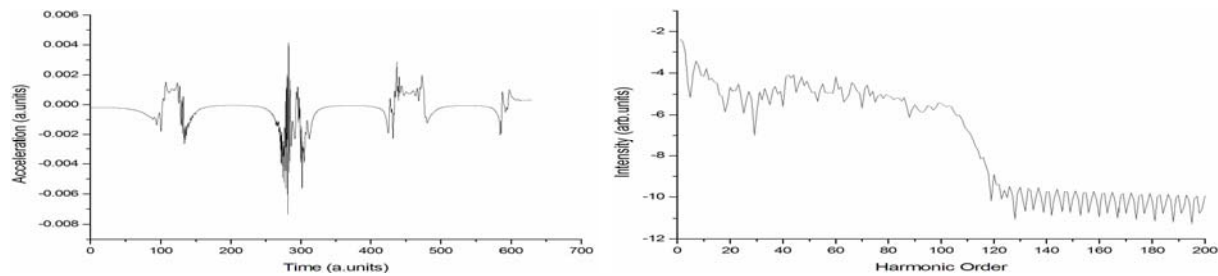

Fig. 3 Dipole moment in acceleration form (left panel) and corresponding high-order harmonic spectra (right panel), with 50 trajectories for the quantum propagation. The strength and frequency of the laser field are 0.1 a.u and 0.0378 a.u., respectively. The wave packet starts at 70 a.u. away from the core with spatial width of 10 a.u., and zero momentum with width 0.1 a.u. in momentum space.

4. Conclusions

The work presented in this paper constitutes the first ever computation of HHG spectra using the Coupled-Coherent States (CCS) method. The outcome exhibits the expected features, such as the plateau and the cutoff, which are related to the quantum interference of electron trajectories as predicted by the three-step model. It also agrees with the results in the literature obtained for reduced-dimensionality models employing other semiclassical propagators in phase space, which characterize the plateau as a quantum-interference effect [4].

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