High harmonic spectra via dominant interaction Hamiltonians

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High Harmonic Generation (HHG) is one of the basic processes of non-linear light-matter interaction involving an electron under the simultaneous influence of a strong laser field and an atomic potential. Initial experimental observations with atoms 1, 2 were soon followed by theoretical work 3, 4, for an early review, see, e.g., 3. More recently HHG has been also investigated in molecules 5, 6 and clusters 7, 8.

The enormous impact of HHG up to recent proposals for imaging of molecular orbitals 9 and the generation of attosecond pulses 10 is not the least due to a very simple description with the so-called three-step model 3, 11. According to this model the electron tunnels out of the combined nuclear plus laser potential, and is then accelerated and pushed back to the nucleus by the laser field where its energy can be converted upon recombination into a high energy photon. This is possible up to a maximal cutoff energy of $E_{\text{max}} = 3.17U_p + I_p$, where $U_p = E^2/(4\omega^2)$ is the ponderomotive energy in terms of the maximal laser field strength $E$ and the photon frequency $\omega$, while $I_p$ the ionization potential. $E_{\text{max}}$ was derived by considering classically the excursion of the electron in the laser field only 3. On the other hand in purely classical HHG calculations the cutoff energy does not play a role since HHG is a quantum mechanical interference effect, essentially between electronic quantum amplitudes in the ground state and in the continuum driven by the laser, as the three step model reveals qualitatively. With a full semiclassical calculation based on trajectories in time, excellent agreement with the quantum result is achieved 12. However, this also implies that a quantitative description of HHG can only be achieved by solving numerically the non-separable dynamics of an electron in the Coulomb field of a nucleus with potential $V_a$ and in the external laser field ($V_{\omega}$).

In view of the universal relevance of HHG as mentioned above it would be very desirable to have an analytical approach which is also quantitative. To this end we introduce the idea of dominant interaction Hamiltonians (DIH) to disentangle the non-separable dynamics by splitting it into spatial regions where either one of the two potentials dominates which is then taken as the only interaction in that region. For regions where the laser dominates (far away from the nucleus) the dynamics is simply that of a free electron in the laser field, the so called Volkov-dynamics, governed by the Hamiltonian $H_{\omega} = p^2/2 + V_{\omega}$. In the opposite case we have the electron only under the influence of the Coulomb potential $V_a$ which is trivially integrable for one electron with $H_a = p^2/2 + V_a$.

To demonstrate how the concept of DIH works, we will use the simplest realization of HHG, a one dimensional electron dynamics in a soft core potential $V_a$ along the linearly polarized laser field, defined by the Hamiltonian (atomic units (a.u.) are used unless stated otherwise)

$$H = \frac{p^2}{2} + V_a + V_{\omega},$$  \hspace{1cm} (1)

where $V_a(x) = -(x^2 + a)^{-1/2}$ with $a = 2$ a.u. such that the ground state energy agrees with hydrogen ($-E_b = 1/2 = I_p$). The laser interaction is defined as $V_{\omega} = xE\cos{\omega}t$, where for convenience we use here a 3.5-cycle laser pulse with $\omega = 0.0378$ a.u. 12. Our observable of interest, the HH spectrum $\sigma(\omega)$ can formulated as the Fourier transform $\sigma(\omega) = \int dt \, d(t) \exp[i\omega t]$ of the dipole acceleration

$$d(t) = -\langle\Psi(t)|\partial V_a/\partial x|\Psi(t)\rangle.$$  \hspace{1cm} (2)

The exact $\Psi(t)$ is the solution of the time-dependent Schrödinger equation with Hamiltonian Eq. (1) and leads to the familiar HH spectrum shown in Fig. 1 (a) which was extracted from the acceleration in Fig. 2 (a). It has been obtained under scattering conditions, i.e., with a Gaussian electron wavepacket $\Psi_i(x,0) = \langle x|g(w_i)\rangle$, where

$$\langle x|g(w_i)\rangle \equiv \left(\frac{\gamma}{\pi}\right)^{1/2} \exp\left\{-\frac{\gamma}{2}(x-q_i)^2 + i p_i(x-q_i)\right\}.$$  \hspace{1cm} (3)
We have introduced the short notation \( \mathbf{w} = (p, q) \) for a point in phase space. The wavepacket \( \Psi_i \) with width parameter \( \gamma = 0.05 \) a.u. is initially located at rest \( (p_i = 0) \) at a distance of \( q_i = \mathcal{E}/\omega^2 \) (70 a.u.) from the proton which corresponds to half the quiver amplitude.

Next, we construct the semiclassical HH spectrum. While the full quantum spectrum only serves as a reference for accuracy of our approximation, the semiclassical propagation is part of the DIH approach to be developed since the latter requires local information in phase space as we will see. Such information is contained in the classical trajectories underlying the semiclassical propagator developed by Herman and Kluk [13], (see also [14] and [15]),

\[
K(x, x', t) = \int \frac{d^2 w_0}{2\pi} \langle x|g(w_0)\rangle \sqrt{R} e^{iS} \langle g(w_0)|x'\rangle,
\]

(4)

with Gaussians \( \langle x|g(w)\rangle \) which have for convenience the same width as the initial state Eq. (3). The interpretation of Eq. (4) is straightforward: The quantum transition amplitude from point \( x' \) at time \( t = 0 \) to \( x \) at time \( t \) is constructed through classical trajectories which start at \( \mathbf{w}_0 \) at time \( t = 0 \) and reach under the dynamics of the Hamiltonian the phase space point \( \mathbf{w}_t \) at time \( t \). The preexponential weight factor of such a trajectory in phase space is given by

\[
R = \frac{1}{2} \det \left( m_{p_0p_0} + m_{q_0q_0} - i\gamma m_{q_0p_0} + \frac{i}{\gamma} m_{p_0q_0} \right),
\]

(5)

which is composed out of the four blocks \( m_{ab} \equiv \partial a/\partial b \) of the monodromy matrix [13]. Note that for the present one-dimensional case, the \( m_{ab} \) are scalars and no determinant has to be taken. The semiclassical amplitude is then given by \( \sqrt{R} e^{iS} \), where \( S(t) \) is the action along the trajectory. The integration is performed over all phase space points \( \mathbf{w}_0 \) which serve as initial conditions of classical trajectories \( \mathbf{w}_t \equiv (p_t(p_0, q_0, t), q_t = q(p_0, q_0, t)) \). Convergence is achieved with a finite number of trajectories through the Gaussian envelope, as illustrated in Fig. 2. The HH spectrum obtained with the semiclassical wavefunction \( \psi(x, t) = \int dx' K(x, x', t) \psi_i(x') \) is in excellent agreement with the quantum spectrum, see Fig. 1. While providing a lot of insight into the dynamics which creates HH the semiclassical approach is at least as numerically involved as solving the Schrödinger equation directly, since the trajectories \( \mathbf{w}_t \) cannot be obtained analytically, and moreover, the full classical dynamics of
this problem is chaotic \cite{12}, although the HH spectrum is very regular.

This underscores the motivation for the DIH concept, where for each dominant interaction, the trajectories are ideally known analytically, or at least can be obtained with little numerical effort. The key point of the DIH approach is to define an appropriate phase space boundary between the electron dynamics governed by the atomic potential $V_a$ and the laser potential $V_E$. For the present one dimensional case, the boundary reduces to isolated points $w_c$ in phase space. The physical process we have to describe is the trapping of the freely oscillating electron in the laser field, due to the atomic potential. This will be most likely if the electron is slow close to the nucleus, i.e., we set $p_c = 0$. The trapping region is defined in a natural way as the range where the soft-core potential is always stronger than the laser potential. For $V_a$ and $V_E$ from Eq. (1), this region results to be the interval $[−x_c, x_c]$, with $x_c = 3.0083$ (see Fig. 4).

We are now in a position to generate the HH spectrum with the DIH approach. Although this can be done completely analytically \cite{16}, we prefer to use here the same propagation scheme of classical trajectories as used for the semiclassical HH spectrum presented above. This allows for strict comparison of the full semiclassical (and quantum) spectrum and the one to be calculated with DIH. The DIH result agrees remarkably well with the exact quantum spectrum as can be seen in Fig. 1 and Fig. 2.

In order to understand why the DIH approach works so well, it is instructive to analyze the initial conditions of the trajectories which switch and get trapped. A little thought reveals that they form bands in the initial phase space (see Fig. 3). To see that we recall that the trajectory for an electron in a laser field with initial conditions $q(0) = q_0$ and $p(0) = p_0$ reads

$$p(t) = p_0 - \frac{\mathcal{E}}{\omega} \sin(\omega t), \quad (6)$$

$$q(t) = q_0 + p_0 t + \frac{\mathcal{E}}{\omega^2} [\cos(\omega t) - 1]. \quad (7)$$

From the condition $p(t_c) = 0$ follows that $t_c \approx n\pi/\omega$ with $n = 1, 2, \ldots$. Then, the conditions for a switch from $V_E$ to $V_a$ are

$$q_c = q_0 + \frac{n\pi}{\omega} p_0 + \frac{\mathcal{E}}{\omega^2} [\cos(n\pi) - 1]$$

$$= q_0 + \frac{n\pi}{\omega} p_0 - \frac{1 - (-1)^n}{2} \frac{2\mathcal{E}}{\omega^2}. \quad (8)$$

This implies that the initial phase space points are given by lines $p_0^{(n)}(q_0)$ with a width $\Delta p_0^{(n)} = \omega/(n\pi)$ as illustrated in Figs. 3 and 5. The explicit expression follows from rearranging Eq. (8),

$$p_0^{(n)}(q_0) = -\frac{\omega}{n\pi} \left(q_0 - \frac{1 - (-1)^n}{2} \frac{2\mathcal{E}}{\omega^2}\right). \quad (9)$$
Interestingly, for the full interaction, the initial conditions for those trajectories which get trapped lie on the same phase space stripes (Fig. 5). We may conclude that the DIH switching condition describes the dynamics relevant for HHG quite well. The small differences in the initial conditions can be attributed to the (small) attraction by the nucleus which the electrons from the full classical trajectories feel on the way inward.

This observation also explains the long standing puzzle why the Coulomb long range nature of the potential plays only a minor role: In fact, HH spectra in qualitative agreement with experiments have been also calculated with zero-range potentials $V_a \propto \delta(x)$ [17]. In the DIH approach, the switching condition would change to $x_c = 0$ but the structure of the initial manifold leading to switching remains the same.

To summarize, we have introduced the concept of dominant interaction Hamiltonians (DIH) to simplify the theoretical description of high harmonic generation by splitting the problem into two integrable ones: the electron under the influence of the laser field and the electron under the influence of the atomic potential. We construct the HH spectrum semiclassically by using classical trajectories: They feel the force of the laser or of the atomic potential and the force is switched at the phase space boundary defining the dominance of each of the two interactions. The dynamics is integrable under either of the two interactions reducing greatly its complexity without loss of accuracy of the spectrum. The simplification manifests itself in the fact that up to one million trajectories are necessary in the present example of HHG to converge the spectrum fully semiclassically while a factor of 100 less is sufficient to converge the DIH spectrum.

Moreover, the DIH provides a natural dynamical extension of the simple man’s approach: In the latter the wave functions of the electron in the laser field and the electron in the ground state of the atomic potential only are simply coherently added to produce qualitatively the HHG spectrum [18]. Here, with the help of DIH we have provided a framework how to dynamically populate one of these states (the ground state) while starting initially with the other one.

While the HH spectrum presented here can be obtained fully analytically [10], the main thrust of the DIH lies in the perspective to describe HHG in more complex systems with the simplification of separating the interactions. Presently, one has to resort to semiclassical techniques since the switching condition is local in phase space. We will explore the possibility of a corresponding quantum condition in future work.

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