Chaos and nonlinearities in high harmonic generation

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1. Introduction

The presence of a strong laser field of frequency $\omega_L$ induces large modifications to the wave function of atoms and makes new phenomena of great basic as well as applicative importance possible. Among these, we mention high harmonic generation (HHG), which occurs when matter emits electromagnetic radiation whose spectrum is formed by a broad plateau of odd harmonics of $\omega_L$, followed by a rapid quenching of the radiation [1, 2].

Assuming that only one electron is active, the full Hamiltonian of the laser–atom interaction is

$$\hat{H}(t) = \hat{H}_0 + ef(t)E_0 \cdot \hat{r} \sin(\omega_L t)$$

(1)

written in the dipole approximation and in the length gauge. Here, $\hat{H}_0$ is the laser-free atomic Hamiltonian, $E_0$ the peak electric field and $0 \leq f(t) \leq 1$ a function describing the envelope of the laser pulse. The spectrum of the radiation scattered by the atom is given by

$$S(\omega) = \frac{4e^2}{3\hbar^3} |\langle r(t)|\rangle|^2 (2)$$

with $S(\omega)d\omega$ the energy irradiated in the frequency range $\omega$ to $\omega + d\omega$ during the whole laser shot, $r(\omega)$ the Fourier decomposition of the quantum averaged electron position: $r(t) \equiv \langle t|\hat{r}|t\rangle$ and $|t\rangle$ the time-dependent atomic state.

The broad plateau points toward the possibility of designing new high-frequency lasing devices and can be exploited for the creation of electromagnetic pulses in the attosecond ($10^{-18}$ s) regime. Moreover, HHG is a powerful tool for obtaining information on the structure and behaviour of the atoms: for example, it has been shown that the wave function of a molecule can be directly observed or that the position of the emitted electron can be traced back by the analysis of...
HHG \[3–8\]. In light of these remarkable successes in spectroscopy, we must ask if it is possible to use a laser field and HHG for testing the postulates of quantum theory.

In this context, the hypothesis of testing the postulate of linearity formulated by Weinberg is particularly interesting \[9\]. The core of the idea is that an intrinsic nonlinearity of quantum mechanics causes a shift of the energy levels and detunes the transition which is resonant in traditional theory. Precise measurements of the nominally resonant interaction between a weak laser field and the atoms are liable to reveal the presence of such terms that must be small, however, since up to now their effects (if any) have been masked or not discerned.

The cogent way to deal with laser–matter interaction is within second quantization \[10\], but if the number of photons in the laser mode is large, the use of a classical description of the field is a safe approximation \[1\]. Moreover, in the treatment of laser–matter interaction it is not rare to use phenomenological or effective Hamiltonians mimicking the complex mechanisms of the field–matter interaction by introducing nonlinear terms that make the analytical treatment simpler. Here, it is worth mentioning the so-called neoclassical theory, which attempts to introduce the radiation reaction back to the charges in the quantum world \[11\] by resorting to a non-quantized (hence classical) description of the field. The task is accomplished by observing that the total electric field acting upon the charges includes a term generated by the same accelerated charges. The Hamiltonian thus depends upon the experimental results \[13\]. Actually, within the realm of quantum electrodynamics, the theory and experiments present an agreement rarely reached in other branches of science; thus, it is seldom discussed whether there is an intrinsically present nonlinear term in the Schrödinger equation.

In the past, other forms of nonlinearities, perhaps with minor ontological significance but surely of larger scope and utility, were introduced into the Hamiltonian for studying composite adiabatic passage and the Landau–Zener effect in the Bose–Einstein condensate—or in density functional theory of both the time-independent and dependent variety \[14–18\]. The possibility that the sudden collapse of the wave function into an eigenstate after a measurement might be the manifestation of a nonlinearity of some sort is also interesting \[19\].

This paper suggests the search for a nonlinear term, wherever its origin comes from, by exploiting the fast modification induced by a laser field on the state of a quantum system. The root of the idea lies in the fact that cumulative effects introduced by the oscillations of the wave function are liable to develop into detectable modifications of measurable quantities. Of course the presence of a nonlinearity is not a matter of logic but of experimental test and measurement accuracy.

2. Theory

Since the sought effect is small, an optimal laser–atom coupling is required, and it occurs at the resonance between the laser photon energy and the energy difference between the two atomic eigenstates. In this condition, all the atomic dynamics may be safely described by taking into account only these two states, and \(|\psi\rangle\) assumes the form

\[
|\psi\rangle = c_0(t)|0\rangle + c_1(t)|1\rangle. \tag{3}
\]

Here, \(|0\rangle\) and \(|1\rangle\) are the two coupled eigenstates of the bare Hamiltonian with energy respectively \(-\hbar \omega_0/2\) and \(\hbar \omega_0/2\); they are assumed with well-defined parity so that \(r_{kk} = 0\) with \(r_{kk} \equiv \langle m|\hat{r}|n\rangle\). Substitution of \(|\psi\rangle\) in the Schrödinger equation gives the differential equations

\[
\frac{ih \hat{\psi}}{\psi} = \mathbb{H}(\psi) = \mathbb{E}(\psi)(t) \tag{4}
\]

with

\[
c = \left( \begin{array}{c} c_0 \\ c_1 \end{array} \right) = \left( \begin{array}{c} \langle 0|\psi\rangle \\ \langle 1|\psi\rangle \end{array} \right). \tag{5}
\]

\[
\mathbb{H} \rightarrow \mathbb{H}_{\text{lin}} = \frac{\hbar}{2} \left( \begin{array}{cc} -\omega_0 & 0 \\ 0 & \omega_0 \end{array} \right) + \mathbb{U}_0 f(t) \left( \begin{array}{cc} 0 & \sin(\omega_1 t) \\ \sin(\omega_1 t) & 0 \end{array} \right). \tag{6}
\]

\[
\mathbb{U}_0 \equiv -e E_0 \cdot \nu_0 \text{ and } \nu_0 = \nu_0 \text{. Equation (4) must be solved with the assigned initial condition } \psi(0). \text{ The mean dipole moment is given by}
\]

\[
r(t) \equiv \langle \hat{r}\psi(t) \rangle = [c_0^2 c_1 + c_0 c_1^2] \nu_0. \tag{7}
\]

The wave function contains four parameters which can be reduced to three by exploiting the normalization condition \(\langle \psi|\psi \rangle = 1\). However, only the relative phase \(\phi(t) \equiv \phi_0(t) - \phi_0(t)\) appears in the expression for the dipole moment. Thus, for the purposes of this paper, knowledge of the relative argument of the states \(\phi(t)\) contains all the information needed. Therefore, for knowledge of the instantaneous electric dipole moment the determination of the two parameters \(P(t) = |c_1(t)|^2\) and \(\phi(t)\) is sufficient. Moreover, in \[14\] it is proved that in correspondence with the two-state system it is possible to find a classical Hamiltonian with a pair of parameters \(\phi, P\) as canonical variables and coordinates of the phase space.

The linearity of the Hamiltonian \(\mathbb{H}_{\text{lin}}\) conforms to the superposition principle and implies that a small variation of the initial condition \(\psi(0) \rightarrow \psi(0) + \delta \psi(0)\) produces a small variation of the final state.

To explore the effects of a nonlinearity on the Schrödinger equation, we introduce the following form for the Hamiltonian:

\[
\mathbb{H} = \frac{\hbar}{2} \left( \begin{array}{cc} -\omega_0 & 2\varpi |c_1|^2 \\ 2\varpi |c_1|^2 & \omega_0 \end{array} \right) + U_0 f(t) \left( \begin{array}{cc} 0 & \sin(\omega_1 t) \\ \sin(\omega_1 t) & 0 \end{array} \right). \tag{8}
\]

with \(\varpi\) the small nonlinearity parameter whose dimension is the inverse of time. Of course, now the superposition principle is not valid any more and new interpretation might be required \[14, 20\]. However, if \(\varpi \ll \omega_0\), the whole traditional
interpretation machinery may be safely retained, at least with very high confidence.

Often, the physicist is interested in the final output of an experiment or simulation; therefore, we concentrate on the determination of the ultimate value of the quantities as a function of the initial condition. For simplicity in what follows, we set \( \phi(0) = 0 \) and let \( P_1(t) \) change; therefore, at the final time \( t_f \) we plot both \( P(t_f) \) and \( \phi(t_f) \) versus \( P_1(0) \). To retain valuable information on the absolute phases we also plot \( \phi(t_f) \) and \( \phi_1(t_f) \) versus \( P_1(0) \).

The introduction of a nonlinearity within the theory brings chaos into the scenario. Classical physics gave birth and definition to chaos, devised tools—few of intuitive, many of less immediate comprehension—for its identification and classification. Of course, this is not the place to give even a small account of the theory: the interested reader is referred to the available literature \([21, 22]\). Here, for self-consistency’s sake, we outline the Poincaré map used in this paper. The temporal evolution of a 1D periodically driven classical dynamical system is uniquely determined by the trajectory of the representative point in the 2D phase space of coordinate \( (q(t), p(t)) \); the Poincaré map is a representation of \( (q(nT), p(nT)) \), with \( T \) the period. If the motion of the system is periodic, then the stroboscopic points appear fixed on a few points or lie along a regular trajectory. If this is not the case, the points may spread over the accessible space, or may display other patterns which qualitatively characterize the motion as chaotic (with all its facets). Of course Poincaré’s maps find its role in classical physics where position and momentum can simultaneously be determined.

Inspired by the Poincaré map, to ascertain if the solution of the new Hamiltonian is strongly sensitive to the initial condition, we mark the pair \( (\phi(t), P(t)) \) in the parameter space stroboscopically calculated at the times \( t = nT_1 \), with the \( n \) integer and \( T_1 = 2\pi/\omega_1 \), the laser period. If the points appear distributed along a regular curve, from the knowledge of the initial state we can extrapolate the final state; borrowing the terminology from the classical world, we may say that a chaotic motion does not evolve from the particular \( e(0) \). If they fill the parameter space, from the knowledge of the initial state we may not predict the final state and chaos rules the evolution \([21]\). We must emphasize that the type of chaos dealt with here is of a quantum nature and stems from the introduction of nonlinear terms into the Schrödinger equation.

3. Results

3.1. HHG

The time-dependent Schrödinger equation with a Hamiltonian \((8)\) has been solved numerically; the free parameters entering the calculations have been chosen to disclose the physics of the model without imposing tedious and prohibitively long calculations. In fact, by dimensional considerations we expect that the modifications induced by the nonlinearity need a time \( \tau \approx 1/\omega_0 \) to become sizeable.

In order for analytical, albeit simplified, support for this statement to be obtained, it is convenient for a moment to restrict the considerations to the case where no laser is present: \( U_0 = 0 \).

By writing the Hamiltonian as a linear combination of the Pauli matrices, the equation for the time evolution of the state is

\[
\dot{\psi} = \left[ \mathbf{\omega} P_1 \sigma_z - \frac{\omega_0}{2} \sigma_z \right] \psi
\]

(9)

that can be solved by the iteration. Let the matrix \( \mathbf{M} \) be the solution of the differential equation

\[
\dot{\mathbf{M}} = \mathbf{\omega} P_1 \sigma_z \mathbf{M}
\]

(10)

which is

\[
\mathbf{M} = e^{-i\mathbf{F}(t)\sigma_z}, \quad F(t) = \int_0^t P(t) d\theta \leq t
\]

(11)

and perform the unitary transform

\[
\psi(t) = \mathbf{M} \mathbf{s}(t).
\]

(12)

Substitution in equation \((9)\) gives an equation for \( s \)

\[
\dot{s} = -\frac{\omega_0}{2} \left[ \cos(2\pi F(t)) \sigma_z + \sin(2\pi F(t)) \sigma_y \right] s.
\]

(13)

Since \( F(t) \leq t \), for \( \tau \ll 1 \) the previous equation can be approximately solved:

\[
s(t) = e^{i\mathbf{F}(t)\sigma_z} \mathbf{c}_1(0), \quad G(t) = \int_0^t \cos(2\pi F(t)) d\theta
\]

(14)

so that the state of the atom is analytically found, at least formally. If \( \tau \approx 1 \) the modifications due to \( \mathbf{F}(t) \) into \( \mathbf{M} \) become large, and the iteration must be continued.

To avoid ramifications in the discussion, we have chosen a CW laser field: \( f(t) = 1 \) lasting \( t_1 = 150T_1 \) and with an atomic energy difference \( h\omega_0 = 0.21 \) au = \( 5.7 \) eV, \( \omega_L = \omega_0/3 \) (three photon resonance). The value of the field intensity is important because it rules the flip of the electron between the states. Our choice is \( U_0 = 2.5 \cdot 10^{-1} \) au; assuming \( r_0 = a_0 \) (\( a_0 \) is the Bohr radius), then the electric field intensity of the laser is \( E_0 = 2.5 \cdot 10^{-1}/e\alpha_0^2 \) for definiteness the polarization is taken along the \( z \) axis. More difficult to choose is the nonlinearity parameter: a too small value of \( \mathbf{v} \) would require too long numerical calculations. Therefore, we set \( \mathbf{v} = 3 \cdot 10^{-2} \) au, which is physically very large, but that retains the duration of the pulse length within reasonable borders.

We start our discussion with the two extreme cases \((U_0 = 2.5 \cdot 10^{-1} \) au, \( \mathbf{v} = 0 \) and \( U_0 = 0, \mathbf{v} = 3 \cdot 10^{-2} \) au\). The first case \((\mathbf{v} = 0)\) corresponds to the laser-driven (linear) two-level atom; in figure 1 we plot, in the first row, \( P(t_1) \) and \( \phi(t_1) \) versus \( P_1(0) \), and in the third row, \( P(t_1) \) versus \( \phi(t_1) \). All the curves are regular and indicate that the outputs of the calculations are predictable in the sense discussed at the end of section 2. The central row gives the value of the absolute phases \( \phi(t_1) \) (\( k = 0.1 \)). The excitation probability given by \( P(t_1) \) is, in principle, easily measurable by using a probe laser field that couples the state \( |1\rangle \) to the continuum. The non-chaotic behaviour is confirmed by the Poincaré stroboscopic map in the upper part of figure 2, where the representative points are distributed along regular curves. The same figure shows the Fourier power spectrum of the dipole moment obtained with the initial condition \( P(0) = 1 \), the odd harmonics and the hyper-Raman lines \([23]\) are visible.
Having gained information on what is to be expected from the standard case, we pass to the second novel nonlinear case, but with $U_0 = 0$. In this case, the time evolution of the vector state $c$ is determined by the nonlinear term alone. In figure 3, we show the equivalent plots of figure 1. As a function of $P_1(t)$, the final population $P_1(t_f)$ presents rapid oscillations (which can be as large as 0.2) absent in the linear case; therefore, because of the nonlinear term, the excited state is not stationary and a spontaneous decay appears. This effect, if real, coexists with the traditional spontaneous decay induced by the vacuum electromagnetic field, and accurate measurements of the decay time of the excited atoms might give an upper limit to the value of $\omega$. The lifetime of the $2p_{1/2}$ state of the hydrogen atom is $T = 1.6 \times 10^{-9}$ s [24]; we argue, therefore, that $1/\omega \geq 10^{-9}$ s. The time $1/\omega$ sets a limit to the information storage in a quantum system; for longer durations the information about
the state deteriorates. The absolute phases $\phi(t_f)$ do not show exotic behaviour, and have little role in the dipole moment. The randomness of $P_1(t_f)$ versus $\phi(t_f)$ indicates that the final state of the atom is unpredictable from the knowledge of the initial one. No unpredictability, here we mean that any particular value of $P_1(0)$ and $\phi(0)$ can be determined, but only by an actual solution of the full time-dependent Schroedinger equation and not by extrapolation of the curves from known points. By measuring the final excitation probability it is possible to check the presence of the nonlinear term. In figure 4, the Poincaré map with the initial condition $P_1(0) = 1$ does not show any particular chaotic behaviour. The power spectrum is very interesting; the energy difference between the levels is blue shifted from the nominal value $\hbar \omega_0 = 3 \hbar \omega_1$ to the new value $\hbar \omega_{01} = 3.17 \hbar \omega_1$.

Now we are ready to study the problem of the nonlinear atom driven by a laser field. From figure 5 we observe deep oscillations of the excitation probability and of $\phi$ resulting in the unpredictability of the final state. The emitted spectrum in figure 6 presents even and odd harmonics (together with the ubiquitous hyper Raman lines); it is important to note that the introduction...
of the nonlinear term introduces qualitative changes in the observed spectrum which might simply be spotted. Again, the setting of an experimental upper limit on the presence of even harmonics in HHG might give an upper limit to the value of \( \varpi \).

In all the simulations in this paper, the stroboscopic points in the Poincaré map are distributed along regular curves so that all individual simulations appear to be non-chaotic. This is an effect of the low value assigned to \( \varpi \); we have performed many simulations with a larger value of this parameter and seen that the stroboscopic points may be randomly distributed over the space, indicating that the equations of the system are, indeed, chaotic. Nevertheless, the sensitiveness of the final state from the initial state is always present in the nonlinear model; the final chaotic-like distribution of \( P(t_f) \) and \( \phi(t_f) \) is a cumulative effect of the whole evolution of the system.

### 3.2. Parametric dependence

The value of the two load parameters in the Hamiltonian (8): the laser–atom detuning \( \omega_0 - 3\omega_L \) and the nonlinearity
E Fiordilino

Parameter \( \varpi \) might strongly affect the results. For our problem, it is of paramount importance to investigate the robustness of the output against small variations of \( \varpi \), because the experimental determination of its value may be more or less difficult. In figure 7 we display the result of a robustness check obtained by slightly changing \( \varpi \). The value of the final excitation probability and of the phases versus \( \varpi \) are plotted in the neighbourhood of the central value \( \varpi = 3 \cdot 10^{-2} \text{ au} \). We notice that our choice is good but not optimal: it lies where \( P_f(t_f) \) does not change very much with \( \varpi \) but where the phases do have a jump. A better choice would be \( \varpi = 2.8 \cdot 10^{-2} \text{ au} \) where \( P_f(t_f) \) is at a minimum and \( \phi(t_f) \) is almost constant. Nevertheless, the results seem quite acceptable.

The role of the laser frequency and consequently of the detuning within the linear theory is well known, since it has been investigated in the huge amount of literature dedicated to the two-level atom. The Hamiltonian is nonlinear at all orders in \( L \omega \) and therefore should produce a very sensitive dependence of the solution upon small variations of \( \varpi \). This can be ascertained by the direct inspection of an iterative solution of

Figure 7. The behaviour of the excitation probability \( P_f(t_f) \) and of the phases as a function of \( \varpi \) in au; the value \( \varpi = 3 \cdot 10^{-2} \text{ au} \) is the one used in all previous simulations.

Figure 8. The behaviour of the excitation probability \( P_f(t_f) \) and of the phases as a function of \( \omega_L/\omega_{L0} \) with \( \omega_{L0} \) the central frequency used in all previous simulations. The plots show a very sensitive dependence of the results upon the detuning.
the equation outlined for the example in [25]. Thus, here we confine ourselves to a few notes. In figure 8 we display the usual four final quantities as a function of the laser frequency. Large variations in the results are evident, and sensitiveness of this sort deserves attention as it suggests the non-stability of the equation.

4. Conclusions

In conclusion, a laser field, resonant with a pair of atomic transitions, enhances the dynamics of the electrons and might make the presence of very small, and as yet, unrecognized nonlinear terms in the Hamiltonian visible. We have discussed a particular choice of nonlinearity and showed that it induces spontaneous decay, line shifts and modifications of the HHG spectrum. These effects coexist, are entwined to similar phenomena and can escape determination; careful experiments are, therefore, due for the detection of nonlinear terms.

As a last remark, classical mechanics is, by construction, a purely deterministic theory and initial conditions set the final output of an experiment. Instead, quantum mechanics is the realm of probabilistic prediction. The recent theory of deterministic chaos has somehow shuffled the cards by introducing a type of unpredictability in classical theory that is generated, in nonlinear systems, by unavoidable quantitative imprecisions in the experimental determination of any initial state. Instead, standard quantum theory is refractory to chaos and the wave function is deterministically obtained from the initial one. Of course, in quantum theory, the knowledge of the initial state can be incomplete and the density matrix description should be resorted to; but the case discussed in this paper seems related to a different situation, which requires discussion on a different basis.

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References