Non-relativistic quantum electrodynamics for strong laser-atom interaction

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Abstract

A formulation of quantum electrodynamics is given that applies to atoms in a strong laser field by perturbation theory in a non-relativistic regime. Dipole approximation is assumed. With the current wavelengths, squeezing can be proved to be negligible and then, just the linear term in the Hamiltonian can be retained. The dual Dyson series, here discussed by referring it to the Birkhoff theorem for singularly perturbed linear differential equations, can be applied and a perturbation series obtained transforming the Hamiltonian by a Pauli-Fierz transformation. But, if just few photons are present high-order harmonics cannot be generated. So, it is proven that odd high-order harmonics only appear when the laser field is intense and one can substitute the creation and annihilation operators by the square root of the mean number of photons taken to be huge, the field retaining its coherency property as observed experimentally for harmonics. In this case, the Hamiltonian for perturbation theory comes to the Kramers-Henneberger form. From this Hamiltonian it is shown that just odd harmonics of the laser frequency contribute to the spectrum for a spherically symmetric potential. This contribution is dipolar when the free-electron quiver motion amplitude is larger than the atomic radius. For a Coulomb potential one has that the outer electron is periodically kicked, and so a prove is given that the same should happen to Rydberg atoms in intense microwave fields. The distribution representing the kicking has a Fourier series with just odd terms. Using a modified Rayleigh-Schrödinger perturbation theory, it is shown that under the same condition of validity of the quiver motion amplitude to atomic radius ratio, the atomic wave function is only slightly modified by the laser field due to the way the energy levels rearrange themselves. This gives a prove of stabilization in the limit of laser frequency going to infinity. Then, perturbation theory can be applied when the product of the laser frequency and the square root of the ratio between the ionization energy and the ponderomotive energy, that is the Keldysh parameter, becomes smaller with respect to the shifted distance between the energy levels of the atom.

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I. INTRODUCTION

The availability of powerful sources of laser light has permitted in recent years the study of light-matter interaction in regimes where known approximations fail. Indeed, a number of new phenomena has appeared as high-order harmonics of the laser frequency, ionization with a number of photons well above the requested threshold and possibly, there are also strong theoretical indications of existence of stabilization, meaning by this that the ionization rate goes to zero by increasing the intensity of the laser field \([1]\). There has been a lot of theoretical work following the appearance of those effects and, as a general approach, people aims to develop models that account for the physics of strong-laser atom interaction trying to support them both by numerical and experimental work. So, a very successful model for harmonic generation has been firstly put forward in \([2]\). This is the recollision model where is assumed that the outer electron goes into continuum due to the laser field and here is accelerated by the field, this step being described classically, then it recombines with the core generating the harmonics. A quantum improved version, understanding the emitted radiation as bremsstrahlung radiation due to the electron approaching the core has been given in \([3]\). This improvement has the advantage that an explanation of the appearance solely of odd harmonics of the laser frequency is given. Another quantum account of the recollision model has been given in \([4]\). To obtain these quantum models a number of reasonable assumptions have been made as the disregarding of intermediate states, the absence of resonances and so on, that need to be justified. These quantum versions of the recollision model prove to be a satisfactory explanation in all the experiments carried out so far with harmonics. Then, any theory starting from quantum electrodynamics has to cope with such a successful understanding of the situation at hand.

Since the initial work by Kulander et al. \([5]\) a lot of numerical work is currently performed to understand strong laser-atom interaction. Anyhow, although this work is highly interesting our aim in this paper is quite different. That is, we are trying to rederive the recollision model from purely quantum arguments without any other approximation than the schemes of current experiments in high-intensity laser field require. Our conclusions concern the justification needed to understand the theoretical models discussed in Ref. \([3,4]\). In this light, we take for granted that numerical computations support the recollision model as the experiments do.

After the pioneering work in \([6]\), a lot of studies have also been carried out assuming a simple two-level model. So, one of the open problems in this field of research is if such a simple model really accounts for the physics of harmonic generation. In this light, the general structure of the spectrum has been obtained by Floquet theory and a parallel with the recollision model has been given in \([6,8]\) for the so called cut-off law, that is the rule to obtain the region of the spectrum where the intensity of the harmonics goes rapidly down. It should be said that such a model cannot account for ionization and
so appears somewhat rough at best.

In the light of the above approaches, in this paper we will try to change the point of view beginning directly from quantum electrodynamics, taking as a starting point our work in Ref. [9] that here is deepened and improved. Indeed, modelling is an important way of understanding but, if one is able to solve the equations of the full theory without resorting to numerical methods, again a deep way to understand physics is also given. So, our main aim is to study in depth what is the physics of strong laser-atom interaction by directly solving the Schrödinger equation. The theory we obtain is anyway enough general to be possibly applicable to other physical situations where strong electromagnetic fields play a role as e.g for Rydberg atoms.

It is interesting to note that, in view of using the dual Dyson series presented in [10] and discussed here by using the Birkhoff theorem [12], the Pauli-Fierz transformation and its classical counterpart, the Kramers-Henneberger transformation, [13] turn out to play a dominant role. Indeed, when unitary transformations are applied, using probability amplitudes permits to extract physics without worrying about. This is a standard approach e.g. in quantum field theory for the interaction picture. Things do not change for the dual Dyson series through the Pauli-Fierz transformation.

Some approximations to start with are needed, but we take them directly from the experiments to tailor the Hamiltonian of quantum electrodynamics to the kind of problems we want to discuss. So, we take the electromagnetic field to be second quantized and the atom field in the non-relativistic approximation. The quadratic term of the vector potential in the Hamiltonian is considered just to see, through a single mode approximation, that taking it into account would shift the frequency of the laser in the harmonics. So, the condition to neglect it is also given. Finally, as the wavelengths of interest are much larger than the atomic radius, the dipole or long wavelength approximation is also taken. Then, a Pauli-Fierz transformation is applied to study strong laser-atom interaction. We will show that only if a large number of photons is present and the amplitude of the quiver motion of the free-electron is much larger than the atomic radius, a condition normally met in this kind of experiments and a key approximation that we will assume in this paper, than just odd harmonics are generated, assuming the atomic potential to be spherically symmetric. Indeed, for a large number of photons the Pauli-Fierz transformation reduces to the Kramers-Henneberger transformation [14] as should be.

An interesting result obtained in this way is that, the atom appears to be a kicked quantum system, being this result rigorously derived from the Hamiltonian of quantum electrodynamics. The kicking hypothesis has been assumed for Rydberg atoms in a microwave field firstly in Ref. [15] in order to explain the experimental results about ionization. Indeed, kicking can mean localization as a counterpart of classical chaos [16]. This is one of the main results of this paper pointing toward the merging of such different fields of research.
A kicked Hamiltonian can in principle be solved exactly. Anyhow, we want to study an atom in a strong laser field doing perturbation theory. So, we will obtain a perturbation series having the product $\omega \gamma$ between the Keldysh parameter $\gamma = \sqrt{\frac{I_B}{2U_p}}$, being $I_B$ the ionization energy and $U_p = \frac{e^2E^2}{4m\omega^2}$ the ponderomotive energy for an electric field $E$, and the laser frequency $\omega$ smaller than the distances, properly shifted by the laser field, between the energy levels. This accomplish the task to obtain a perturbation theory dual to the standard time-dependent theory applied to an atom in a weak electromagnetic field. Using this approach, we are able to show that the rate of above threshold ionization determines the duration of the harmonics in the spectrum and then, also the form of the spectrum itself by Fourier transform. Beside, if a rough model for the outer electron in the field of the rest of the atom is taken having a Coulomb form with a $Z_{eff}$ for the atomic number to give the correct ionization energy, it can be seen that the duration of harmonics agrees fairly well with experimental results as given e.g. in [17]. This simply means to scale all the formulas for hydrogen-like atoms by ionization potential, ponderomotive energy and laser frequency.

The applicability of perturbation theory is indeed possible as, through a modified Rayleigh-Schrödinger perturbation theory one can show that the energy levels of the atom are shifted in such a way to change very little the wave function giving a prove of stabilization in the limit of laser frequency increasingly large. This is what we call “rigidity” of the wave function, very similar to the behavior of a superconductor due to the presence of the binding energy of the Cooper pair. This should give a hint, in the framework of quantum chaos, to study the change of statistics of such energy levels, possibly by some numerical work.

Let us finally point out that a number of methods have been devised starting from Volkov states, that are the solution of the Schrödinger equation for a free particle in a plane wave. The prototype of these approaches is given in Ref. [18]. Anyhow, our point of view assumes different asymptotic states that are naturally derived from dual Dyson series as an entangled state between the radiation and the atomic state: This is just the leading order approximation. Then, e.g. no hypothesis a priori on the atomic potential being short ranged is needed. On the other side, no general proof is known of Volkov states being the proper asymptotic states for perturbation theory in a strong electromagnetic field. Beside, it appears very difficult to understand why just odd harmonics are experimentally observed as this approach keeps both odd and even harmonics [19]. Anyhow, an improved version has been recently given in Ref. [20] by the Volkov states in a second quantized field giving just odd harmonics. This approach satisfy the principle of duality in perturbation theory but what is changed is the initial state used to obtain the perturbation series and is generally presented as a non-perturbative method through the results for the computation of the amplitudes given in Ref. [18].

The paper is so structured. In sec. I we give a brief presentation of duality for the Schrödinger equation and discuss the Birkhoff theorem using the
dual Dyson series. In sec.II we give the Hamiltonian formulation of quantum electrodynamics to start with and show why the quadratic term is negligible. Then we perform a Pauli-Fierz transformation to obtain the dual Hamiltonian and show how it reduces to the Kramers-Henneberger form. A discussion of the two-level approximation with respect to the full theory is also given. In sec.IV we apply the approximation of large amplitude of the quiver motion in the laser field with the respect to the atomic radius to show how kicking arises and why the atomic wave function turns out to be rigid in the sense given above. In sec.V perturbation theory is done in the tunnelling regime deriving the rate of ionization and consequently the spectrum of the harmonics. Finally, in sec.VI the conclusions are given.

II. DUALITY AND BIRKHOFF THEOREM

The duality principle in perturbation theory, when applied to the Schrödinger equation written as (here and in the following $\hbar = c = 1$)

$$ (H_0 + H_1(t))|\psi\rangle = i\frac{\partial|\psi\rangle}{\partial t} $$

states that, by choosing $H_0$ as unperturbed Hamiltonian, the perturbation series has a development parameter exactly the inverse of the series obtained by taking $H_1$ as unperturbed Hamiltonian. Formally, this latter case means that we are multiplying $H_1$ by an ordering parameter $\lambda$ going to infinity. Duality principle is enough, as shown in Ref. [10,11], to prove that the dual to the Dyson series is just the adiabatic approximation and its higher order corrections. Indeed, from the theory of singular perturbation we can rederive a similar result for the equations of probability amplitudes by the Birkhoff theorem [12], where “singular” means that the small parameter multiplies the higher derivative in the equation. In this way, a dual Dyson series can be obtained also for the probability amplitudes. In fact, the dual perturbation series is obtained by solving at the leading order the equation

$$ \lambda H_1(t)|\psi^{(0)}\rangle = i\frac{\partial|\psi^{(0)}\rangle}{\partial t} $$

taking $\lambda \to \infty$. But this can be stated as a singular perturbation problem. Indeed, using the eigenstates of the unperturbed part $H_0|n >= E_n|n >$ and putting $|\psi(t) >= \sum_n e^{-iE_n \epsilon} a_n(t)|n >$, one has for eq. (1) the system of linear differential equations for the amplitudes

$$ i\epsilon \frac{da_n(t)}{dt} = \sum_m e^{-i(E_n - E_m)\epsilon} \langle m|H_1(t)|n \rangle a_n(t) $$

being now $\epsilon = \frac{1}{\lambda}$. If the Hamiltonian $H_0$ has a finite set of $N$ eigenstates and eigenvalues and considering the amplitudes $a_n(t)$ as elements of a vector $\alpha(t)$, we can apply a result obtained by Birkhoff on 1908 in singular perturbation
theory \([12]\), that states that the equation (3) has a fundamental set of solutions \(b_j(t)\) with \(1 \leq j \leq N\) if the eigenvalues \(\lambda_j(t)\) are distinct, given by

\[
b_{kj}(t) = \exp \left[ -i \int_{t_0}^{t} \lambda_j(t') dt' \right] \exp[i\gamma_j(t)] u_{kj}(t) + O(\epsilon) \tag{4}\]

\(k\)-th element of the vector \(b_j(t)\), \(\gamma_j(t) = \int_{t_0}^{t} u_j^\dagger(t') i \frac{d}{dt} u_j(t')\) the geometrical part of the phase, \(u_j(t)\) the eigenstates of the matrix having elements \(A_{mn} = e^{-i(E_n - E_m)t} \langle m | H_1(t) | n \rangle\). The condition of no crossing of eigenvalues \(\lambda_j(t)\) is also required. The final leading order approximation is then written as

\[
a(t) = \sum_{j=1}^{N} \alpha_j b_j(t) \tag{5}\]

with the coefficient \(\alpha_j\) given by the initial conditions \(a(0)\). Again, we realize that formally this is the adiabatic approximation applied to the operator \(e^{iH_0t} H_1(t) e^{-iH_0t}\). Then, we arrived at similar conclusions as in Ref. \([11]\) for dressed states where, instead, duality principle was used, in the limit \(\lambda \to \infty\). But, the use of duality principle permits to get rid of the limitation on the spectrum of \(H_0\) being bounded to \(N\) eigenstates and eigenvalues and then, it appears as a more general tool to treat also this kind of problems. So, we relax the condition on the spectrum of \(H_0\) required by the Birkhoff theorem, as duality permits to give an alternative way to derive it.

Indeed, one can build the dual Dyson series by also doing an unitary transformation that removes the perturbation \(H_1(t)\) in eq.(1). This defines a dual interaction picture. In fact, one can see that the unitary transformation operator must be a solution of

\[
\lambda H_1(t) U_F(t) = i \frac{\partial U_F(t)}{\partial t} \tag{6}\]

and so \(U_F(t)\) turns out to be the evolution operator of the adiabatic approximation in the limit \(\lambda \to \infty\), proving the equivalence. But, this equation can also be solved exactly in some cases, without resorting to the adiabatic approximation giving the series

\[
|\psi(t)\rangle = U_F(t) \mathcal{T} \exp \left[ -i \int_{t_0}^{t} dt' U_F^\dagger(t') H_0 U_F(t') \right] |\psi(t_0)\rangle \tag{7}\]

with \(\mathcal{T}\) the time-ordering operator.

III. NON-RELATIVISTIC FORMULATION OF QUANTUM ELECTRODYNAMICS

A. Full Hamiltonian and the Quadratic Term

The starting assumptions about the formulation of quantum electrodynamics that we need are the following. Firstly, we takes a non-relativistic
approximation and neglect spin effects. Secondly, we cut-off the wavelengths limiting the analysis to the long ones that is, we take the dipole approximation: currently, this is in agreement with the experimental results for harmonic generation. Finally, we do second quantization on the electromagnetic field as we want to understand the properties of the scattered light in experiments with strong fields. So, we write the Hamiltonian in the Coulomb gauge \cite{13} (here and in the following $\hbar = c = 1$)

\[ H = \sum_{\lambda, k} \omega_k a_\lambda^\dagger(k) a_\lambda(k) + \frac{p^2}{2m} + V(x) + \frac{e}{m} A p + \frac{e^2}{2m} A^2 \]  

(8)

with $\lambda = 1, 2$ meaning a sum over polarizations, $V(x)$ the atomic potential, $a_\lambda^\dagger(k), a_\lambda(k)$ the creation and annihilation operators for the mode $k$ with polarization $\lambda$. So, we have

\[ A = \sum_{\lambda, k} g_k (\epsilon_\lambda a_\lambda^\dagger(k) + \epsilon_\lambda a_\lambda(k)) \]  

(9)

being $\epsilon_\lambda$ the complex polarization vector, $g_k = \sqrt{\frac{2\pi}{V \omega_k}}$ and a normalization in a box of volume $V$ is assumed everywhere.

To see how much relevant is the quadratic term, we refer to the experimental result that just harmonics of the laser frequency are observed. Then, specializing the above Hamiltonian to a single mode \cite{13}, we derive the Heisenberg motion equation for the creation and annihilation operators. So, we take

\[ H = \omega a^\dagger a + \frac{p^2}{2m} + V(x) + \frac{e}{m} A p + \frac{e^2}{2m} A^2. \]  

(10)

Assuming a linear polarization, that is

\[ A = \sqrt{\frac{2\pi}{V \omega}} \epsilon (a^\dagger + a) \]  

(11)

we can use the same argument given in Ref. \cite{21} that the laser frequency should be shifted by squeezing. Indeed, one has the Heisenberg equations for $a$ and $a^\dagger$

\[ \frac{da}{dt} = -i \omega a - ie^2g^2 \frac{m}{e} (a + a^\dagger) - i eg \frac{m}{e} p \]

\[ \frac{da^\dagger}{dt} = i \omega a + ie^2g^2 \frac{m}{e} (a + a^\dagger) + i eg \frac{m}{e} p \]  

(12)

and without going into details of computation, by duality, as a first approximation, one neglects the atomic Hamiltonian and then, managing $p$ as a c-number, the time dependence of the creation and annihilation operators is harmonic with frequency $\Omega = \sqrt{\omega^2 + \frac{4\pi e^2}{mV}}$ and not just $\omega$ as should be required by the experimental results. So, squeezing terms can be neglected otherwise a shifted frequency of the harmonics would be observed \cite{21}. This effect should be more pronounced as greater is the order of the harmonic. But, considering this shift with current frequencies and gas densities really negligible,
the quadratic term of the field in the Hamiltonian (8) can be systematically neglected giving the final Hamiltonian

\[ H = \sum_{\lambda, k} \omega_k a_\lambda^\dagger(k) a_\lambda(k) + \frac{P^2}{2m} + V(x) + \frac{e}{m} \sum_{\lambda, k} g_k (\epsilon_\lambda^* a_\lambda^\dagger(k) + \epsilon_\lambda a_\lambda(k)) p. \] (13)

This will be the starting point for our further analysis.

B. Pauli-Fierz Transformation and Kramers-Henneberger Hamiltonian

We now try to approach the Hamiltonian (13) using duality principle, that is, we try to compute the dual Dyson series. So, one can remove the perturbation by a Pauli-Fierz transformation given by

\[ U_{PF} = \exp \left[ \sum_{k, \lambda} \left( \beta^*_\lambda(k) a_\lambda(k) - \beta_\lambda(k) a_\lambda^\dagger(k) \right) \right] \] (14)

where one has

\[ \beta_\lambda(k) = \frac{e}{m \omega_k} g_k \epsilon^*_\lambda \cdot p \] (15)

and obtains the Hamiltonian [13]

\[ H_{PF} = \sum_{k, \lambda} \omega_k a_\lambda^\dagger(k) a_\lambda(k) + \frac{P^2}{2m} + V \left[ x - i \sum_{k, \lambda} \left( \beta^*_\lambda(k) a_\lambda(k) - \beta_\lambda(k) a_\lambda^\dagger(k) \right) \right] \] (16)

being \( m^* \) the renormalized mass due to the field. Here we try another way, to agree with duality in perturbation theory. That is, firstly we consider the Hamiltonian [13] in the interaction picture giving

\[ H_I = \frac{P^2}{2m} + V(x) + \frac{e}{m} \sum_{\lambda, k} g_k \left[ \epsilon_\lambda^* a_\lambda^\dagger(k) e^{i\omega_k t} + \epsilon_\lambda a_\lambda(k) e^{-i\omega_k t} \right] \cdot p. \] (17)

Then, we solve the leading order equation

\[ \frac{e}{m} \sum_{\lambda, k} g_k \left[ \epsilon_\lambda^* a_\lambda^\dagger(k) e^{i\omega_k t} + \epsilon_\lambda a_\lambda(k) e^{-i\omega_k t} \right] \cdot p U_F(t) = i \frac{\partial U_F(t)}{\partial t}. \] (18)

The solution is standard and can be written as

\[ U_F(t) = \exp \left[ - \sum_{k, \lambda} \gamma_{k, \lambda}(t) (\epsilon^*_\lambda \cdot p) a_\lambda^\dagger(k) \right] \exp \left[ \sum_{k, \lambda} \gamma^*_\lambda(t) (\epsilon_\lambda \cdot p) a_\lambda(k) \right] \exp \left[ \sum_{k, \lambda} a_{k, \lambda}(t) (\epsilon^*_\lambda \cdot p)(\epsilon_\lambda \cdot p) \right] \] (19)

with

\[ \gamma_{k, \lambda} = \frac{e}{m} \sqrt{\frac{2\pi}{\omega_k V}} \frac{e^{i\omega_k t} - 1}{\omega_k} \] (20)
and

$$\alpha_{k,\lambda} = -\frac{1}{2}\gamma_{k,\lambda}(t)^2.$$  \hfill (21)

So, we have found in this way a time-dependent version of the above Pauli-Fierz transformation. But now, by eq. (7) we have a dual Dyson series for quantum electrodynamics. From this, one has e.g. that the leading order wave-function in the interaction picture, if we assume the atom in the ground state, is given by

$$|\psi^{(0)}(t)\rangle_I = U_F(t)|1s\rangle|\alpha\rangle$$ \hfill (22)

being $|1s\rangle$ the atomic state and $|\alpha\rangle$ the initial state of the laser field described by a coherent state. As it should be expected this is an entangled state between the atomic state and the field state \cite{21}. It is easy to realize, using the momentum representation for the atomic state, that the scattered light is indeed coherent and preserves the property of the initial state for the field.

On this basis, we expect the light of the harmonics to be coherent as well \cite{21}. In fact, in order to evaluate higher orders in the dual Dyson series we rewrite the Pauli-Fierz transformation as

$$U_F(t) = \exp\left[\sum_{k,\lambda} \left(\gamma^*_{k,\lambda}(t)\epsilon_{\lambda}\cdot p) a_{\lambda}(k) - \gamma_{k,\lambda}(t)\epsilon^*_{\lambda}(p) a^\dagger_{\lambda}(k)\right)\right],$$  \hfill (23)

obtaining the transformed Hamiltonian for higher order computations

$$H_F = U_F^\dagger(t)(\frac{p^2}{2m} + V(x)) U_F(t) = \frac{p^2}{2m} + V [x + X(t)]$$ \hfill (24)

being

$$X(t) = -i \sum_{k,\lambda} \left(\gamma^*_{k,\lambda}(t)\epsilon_{\lambda} a_{\lambda}(k) - \gamma_{k,\lambda}(t)\epsilon^*_{\lambda} a^\dagger_{\lambda}(k)\right).$$ \hfill (25)

Now, we specialize the above construction assuming that initially the atom is exposed to an intense laser field. It is well-known that the creation and annihilation operators can be expressed through the number operator $n_{k,\lambda} = a^\dagger_{k,\lambda} a_{k,\lambda}$ and a phase operator by the Susskind and Glogower \cite{23} or the Pegg and Barnett construction \cite{24}. But, the distribution probability of the phase can be made to coincide in such a way to give for a coherent state a definite phase having quantum fluctuations that go to zero for a very large mean number of photon, that is the case of the intensity of the laser field of interest. Then, one can keep just one mode and operate the substitution \cite{23}

$$a_{k,\lambda} \rightarrow \sqrt{n_{k,\lambda}} e^{i\phi_{k,\lambda}}.$$ \hfill (26)

The Hamiltonian $H_F$ is then reduced to the Kramers-Henneberger form \cite{14} for a classical field that is,

$$H_{HK} = \frac{p^2}{2m} + V [x - \lambda L \epsilon(\sin(\omega t - \phi) + \sin(\phi))]$$ \hfill (27)
where a linear polarization has been assumed, all subscripts have been dropped and $\lambda_L$, the free-electron maximum excursion in a monochromatic field, can be rewritten as $\lambda_L = \frac{eE}{m\omega^2}$, being $E$ the intensity of the laser field. The above Hamiltonian can be obtained by an unitary transformation, the Kramers-Henneberger transformation, for an atom in a monochromatic classical field. Then, we simply neglect quantum fluctuation beyond the leading order due to the characteristic of the electromagnetic field. The relevance of this limit for a coherent state is that it makes consistent the arguments that follow about the properties of the harmonics. We just note that the advantage of such a derivation is that it gives us the coherence property of the scattered laser light in atomic scattering experiments, and permits to understand the way from a quantum electrodynamics formulation to a classical Hamiltonian that fully accounts for the situation at hand.

As a general approach, in agreement with the duality principle in perturbation theory [10], we see that to study strong laser-atom interaction we apply unitary transformations to remove the big part of the Hamiltonian that in this case is due to the field. We now apply the same idea also to the two-level model going to a dual interaction picture.

C. Two-level Approximation

The two-level model generally adopted in the study of harmonic generation can be cast in the form (e.g. [8])

$$H = \frac{\Delta}{2} \sigma_3 + \Omega \sigma_1 \cos(\omega t)$$

being $\Delta$ the distance between the two-level, $\Omega$ the strength of the field, $\omega$ the frequency of the laser field and $\sigma_1$, $\sigma_3$ the Pauli matrices. The first point to note is that, in order to be in agreement with the recollision model, one of the two levels should be in the continuum or, at best, $\frac{\Delta}{2}$ is the ionization energy $I_B$ [7]. In this way, one can rederive the well-known cut-off law for the frequency of the maximum harmonic $I_B + 3U_p$, being $U_p = \frac{e^2E^2}{4m\omega^2}$ the ponderomotive energy that in this model should be proportional to $\Omega$ as pointed out in Ref. [7]. But, in this model the continuum is missing and then, ionization cannot be described. This means to lose any connection between ionization and harmonic generation that, as we will see, plays a role in the duration of the harmonics itself. Finally, let us apply a Pauli-Fierz transformation (or Kramers-Henneberger transformation) by removing the oscillating term as

$$U_F(t) = \exp \left[ -i{\sigma_1}{\frac{\Omega}{\omega}} \sin(\omega t) \right]$$

(29)

to obtain the transformed Hamiltonian

$$H_F = \frac{\Delta}{2} J_0 \left( \frac{2\Omega}{\omega} \right) + \Delta \sigma_3 \sum_{n=1}^{\infty} J_{2n} \left( \frac{2\Omega}{\omega} \right) \cos(2n\omega t) + \Delta \sigma_2 \sum_{n=0}^{\infty} J_{2n+1} \left( \frac{2\Omega}{\omega} \right) \sin((2n + 1)\omega t).$$

(30)
From this Hamiltonian is quite easy to realize that both odd and even harmonics play the same role and the only way to make ones or the other appear is to choose proper initial conditions. To show that things are really in this way, we turn back to Hamiltonian (28) and apply the Birkhoff theorem to rederive the spectrum already obtained in Ref. [11], given in a general form by the Floquet theory in Ref. [8].

So, we have to solve the equation

\[
\frac{\Delta}{2} \sigma_3 + \Omega \sigma_1 \cos(\omega t) \int |\psi(t)\rangle = i \frac{d|\psi(t)\rangle}{dt}
\]

(31)

that in the interaction picture becomes the system of equations

\[
\begin{pmatrix}
0 & \Omega e^{-i\Delta t} \cos(\omega t) \\
\Omega e^{-i\Delta t} \cos(\omega t) & 0
\end{pmatrix}
\begin{pmatrix}
|a_1(t)\rangle \\
|a_2(t)\rangle
\end{pmatrix} = \begin{pmatrix}
|a_1(t)\rangle \\
|a_2(t)\rangle
\end{pmatrix}
\]

(32)

to which the Birkhoff theorem can be applied. It should be pointed out that this approximation is consistent with \(\Delta \ll \Omega\) and we take \(\omega \ll \Omega, \Delta\), in agreement with the identifications of Ref. [7] and in the way the model should compare with experiments. Otherwise, the Birkhoff theorem should be applied differently. Under these conditions, we show that even harmonics can be present depending on the initial conditions. These harmonics are known as hyper-Raman lines in the current literature [8,26].

Firstly, we note that eq.(32) is the same as eq.(31) when use has been made of the eigenstates of \(\sigma_3\). Then, we can simply use the results of Ref. [11] about the dressed states of Hamiltonian (28) and obtain the Birkhoff basis as

\[
b_1(t) = e^{-i\frac{\Delta}{2} t} e^{i\frac{\Omega}{2} \sin(\omega t)} \frac{1}{\sqrt{2}} \begin{pmatrix} 1 \\ -e^{-i\Delta t} \end{pmatrix}
\]

(33)

and

\[
b_2(t) = e^{i\frac{\Delta}{2} t} e^{-i\frac{\Omega}{2} \sin(\omega t)} \frac{1}{\sqrt{2}} \begin{pmatrix} e^{i\Delta t} \\ 1 \end{pmatrix}
\]

(34)

Then, we conclude that the Birkhoff theorem gives the same results obtained by other approaches in Ref. [11,28]. The spectrum is then given by [11]

\[
\langle \psi(t) | \sigma_1 | \psi(t) \rangle \approx a_2(0)a_1^*(0)e^{-i\Delta_R t} + a_2^*(0)a_1(0)e^{i\Delta_R t}
\]

\[
+ (|a_1(0)|^2 - |a_2(0)|^2)\Delta \sum_{n=0}^{\infty} J_{2n+1} \left( \frac{2\Omega}{\omega} \right) \frac{\cos((2n+1)\omega t) - 1}{(n + \frac{1}{2})\omega}
\]

\[
+ i(a_2^*(0)a_1(0)e^{i\Delta_R t} - a_2(0)a_1^*(0)e^{-i\Delta_R t})\Delta \sum_{n=1}^{\infty} J_{2n} \left( \frac{2\Omega}{\omega} \right) \frac{\sin(2n\omega t)}{n\omega}
\]

(35)

with \(\Delta_R = \Delta J_0 \left( \frac{2\Omega}{\omega} \right)\) the renormalized level separation. This form of the spectrum is in agreement with the one derived by the Floquet theory as given in [8]. In the range of validity of this approximation as stated above, this proves the assertion that the even harmonics should be considered on the
same ground as the odd ones for the two-level model. As we are going to show, this makes the applicability of this approximation to harmonic generation somewhat inappropriate as the appearance of just odd harmonics in the experiments has a deep physical meaning that here is overlooked. In fact, the two-level model relies just on the initial conditions to select the properties of the spectrum.

**IV. PROPERTIES OF ATOMS IN INTENSE LASER FIELDS**

**A. Odd Harmonics and Kicking for an Electron in a Coulomb Field**

We now turn our attention to the classical limit of the Pauli-Fierz Hamiltonian that is, the Kramers-Henneberger Hamiltonian that here we rewrite assuming the phase \( \phi = 0 \) obtaining

\[
H_{HK} = \frac{p^2}{2m} + V [x - \lambda_L \epsilon \sin(\omega t)].
\]  

(36)

The potential has the Fourier series

\[
V [x - \lambda_L \epsilon \sin(\omega t)] = \sum_{k=0}^{\infty} v_k(x) e^{ik\omega t} + (-1)^n e^{-ik\omega t}
\]  

(37)

where

\[
v_k(x) = \int_{-1}^{1} \frac{dx'}{\pi} V(x - \epsilon \lambda_L x') \frac{T_k(x')}{\sqrt{1 - x'^2}}
\]  

(38)

and being \( T_k(x) \) the \( k \)-th Chebyshev polynomial of first kind. In this section we study the terms having \( k > 1 \), deserving the analysis of the static contribution with \( k = 0 \), i.e. the dressed potential, for the next section.

We now take the unperturbed potential \( V(x) \) to be spherical symmetric. It easily realized that one has different parity for even components being \( v_{2n}(-x) = v_{2n}(x) \), and odd components being \( v_{2n+1}(-x) = -v_{2n+1}(x) \). But, the unperturbed part of the Hamiltonian is invariant by parity transformation while, the odd time-dependent part breaks this symmetry. So, by perturbation theory one has at first order

\[
\langle \psi(t)|x|\psi(t) \rangle = \langle \psi^{(0)}(t)|x|\psi^{(1)}(t) \rangle + c.c.
\]

being \( |\psi^{(0)}\rangle \) the initial atomic state such that \( \langle \psi^{(0)}(t)|x|\psi^{(0)}(t) \rangle = 0 \), and the only way not to obtain a null spectrum through perturbation theory is from the odd components. This result turns out to be in agreement with the general one obtained through Floquet states in Ref. \[27\].

To apply the above scheme to the Coulomb potential one has to consider the integrals

\[
v_k(x) = \frac{Ze^2}{\lambda_L} \int_{-1}^{1} \frac{dx'}{\pi} \frac{1}{\sqrt{\left(\frac{x}{\lambda_L} - x'\right)^2 + \left(\frac{y}{\lambda_L}\right)^2 + \left(\frac{z}{\lambda_L}\right)^2}} \frac{T_k(x')}{\sqrt{1 - x'^2}}
\]  

(39)

but we have to study them around the origin of coordinates where the Coulomb potential turns out to be singular. Indeed, at small distances the
theory given above has its shortcomings due to the dipole approximation and the neglecting of the relativity. Then, some regularization is needed for the above integrals. So, let us study the integrals (39) at the point $y = z = a_0$, introducing in this way the cut-off $\eta = \frac{a_0}{\lambda_L} \ll 1$, and we are reduced to the analysis of the leading term

$$v_k(x) = -\frac{Ze^2}{\lambda_L} \int_{-1}^{1} \frac{dx'}{\pi} \frac{1}{\sqrt{\left(\frac{x}{\lambda_L} - x'\right)^2 + 2\eta^2\sqrt{1 - x'^2}}} T_k(x')$$

very near the origin. These integrals turn out to be zero for odd Chebyshev polynomials when one takes $x = 0$ and, for even Chebyshev polynomials one gets an extremum such that the integral is bounded. The extremum depends on $\eta$ but increases slowly. Then, we can remove the even terms, after we take $v_{2n}$ at their values at the origin, by an unitary transformation. The same cannot be done for odd terms as they go to zero at the origin. These integrals can be studied by a Taylor series at $x = 0$ giving for the dipolar term

$$v_{\text{dip}}^k (x) = -\epsilon \frac{Ze^2}{\lambda_L} \left(-1\right)^n (2n + 1) \frac{x}{a_B}$$

and the integral gives the cut-off dependent approximation

$$\int_{-1}^{1} \frac{dx'}{\pi} \frac{1}{\left(x'^2 + 2\eta^2\right)^{\frac{3}{2}}} \frac{T_k(x')x'}{\sqrt{1 - x'^2}} \approx -\frac{2}{\pi} \log(\eta)(2n + 1)(-1)^n.$$

that has a logarithmic divergence for the cut-off $\eta$ going to zero. If we introduce the ratio $\frac{a_B}{\lambda_L} \ll 1$ that defines the region where we want to study the physics of the model, we can reabsorb the divergence $-\log(\eta)$ as a renormalization into it and introduce the physical ratio $\epsilon = \frac{2a_B}{\pi\lambda_L}$. This means that we have two bare constants $\epsilon_0$ and $\eta$ and this goes to zero through the relation $\eta = e^{-\frac{\epsilon}{\epsilon_0}}$ with $\epsilon$ the physical ratio. Then, the limit $\epsilon_0 \to 0$ implies $\eta \to 0$. Through this redefinition, the dipolar terms are

$$v_{\text{dip}}^k (x) = -\epsilon \frac{Ze^2}{\lambda_L} \left(-1\right)^n (2n + 1) \frac{x}{a_B}$$

In this way, we can now prove that the electron in a Coulomb field and an intense laser field, such to have the parameter $\epsilon \ll 1$, undergoes kicking turning the problem into one of quantum chaos [16].

The question of a kicked hydrogen atom has been put forward to explain experiments on ionization of Rydberg atoms in an intense microwave field through the mechanism of dynamical localization [15,16]. Pioneering work has also been done on strong fields and atoms, using kicked models e.g. in Ref. [29]. But, a rigorous proof of this behavior derived directly from quantum electrodynamics has not been given yet. Here, we accomplish this task by completing the analysis of the time-dependent part of the Kramers-Henneberger Fourier components. To complete this derivation, we point out that a periodic distribution as $\delta_T(t) = \sum_{n=-\infty}^{+\infty} \delta(t - nT)$ that has period $T$, can be defined...
Through the coefficients of its Fourier series. So, e.g. \( \delta_T(t) = \frac{1}{T} \sum_{n=-\infty}^{+\infty} c_n e^{i2\pi nt/T} \) has all constant coefficients given by \( \frac{1}{T} \). To have convergence of the Fourier series in the sense of distributions, the coefficients \( f_n \) must satisfy the criterion of being slowly varying \([30]\), i.e. \( |f_n| \leq M|n|^k \), being \( M \) and \( k \) two constants.

Then, by using eqs. (13) and one gets the dipolar potential as

\[
\tilde{V}_{KH} = -\frac{Ze^2}{\lambda_L a_B} \frac{x}{\omega} \sum_{k=-\infty}^{+\infty} (2k+1) e^{i(2k+1)\omega t} = 2\frac{Ze^2}{\lambda_L a_B} \frac{x}{\omega} \sum_{k=0}^{+\infty} (2k+1) \sin((2k+1)\omega t). \tag{44}
\]

The Fourier series appearing here has convergence just in the sense of distributions. We now give an explicit form for the dipolar term. Indeed, one has

\[
\tilde{\delta}_T(t) = \sum_{k=-\infty}^{+\infty} (-1)^k \delta(t - kT) = \frac{2}{T} \sum_{k=-\infty}^{+\infty} e^{i(2k+1)\omega t} \tag{45}
\]

so that, the dipolar term is defined through the derivative of a periodic distribution of period \( \frac{T}{2} \). One can write at last

\[
\tilde{V}_{KH} = -\frac{\pi}{\omega} \frac{Ze^2}{\lambda_L a_B} \frac{x}{\omega} \frac{d}{dt} \tilde{\delta}_T(t) + \cdots \tag{46}
\]

and we draw the conclusion that the motion of an atom in an intense laser field undergoes kicking with period \( \frac{T}{2} \) so that, localization, a typical effect of quantum chaos, can happen.

Now, we show that the Rydberg atoms used in ionization experiments with microwave are indeed in a regime where the ratio between the atomic dimensions and the amplitude of the quiver motion of the free-electron \( \lambda_L \) in the laser field is largely smaller than unity, and the above theory applies. As a starting point we take Ref. [31] for a typical experiment. The less favourable case is for an intensity of the microwave field of 2.5 V/cm, at 12.4 GHz and the Rydberg atom has \( n_0 = 98 \). This gives for the ratio \( \frac{n_0^2 a_B}{\lambda_L} = \frac{n_0^2 \omega}{\sqrt{8I_B U_p}} \) the value 0.0027, largely lesser than unity. Instead, the most favourable case is given by the intensity of the electric field of 21 V/cm, at 18 Ghz and \( n_0 = 64 \) giving a ratio of 0.00029 improving the situation of about a magnitude order. Then, we can conclude that Rydberg atoms are kicked by the microwave field and localization can happen, in agreement with all the current researches. This result comes directly from quantum electrodynamics and so, it is fully justified.

For the case of the harmonic generation the situation is still better because the Keldysh parameter \( \gamma \) given by \( \frac{\gamma}{\sqrt{2 \pi I_p}} \) can be lesser than one permitting a straightforward application of the perturbation theory. A typical example of an experiment in this regime is given in Ref. [17]. This possibility can be fully exploited if a set of unperturbed states can be found to do perturbation theory. This is indeed the case as we will see in the next section.
B. Rigidity of the Atomic Wave Function

In order to do perturbation theory, one generally needs a full set of orthonormal functions to start with, representing the unperturbed system. But, here one has to diagonalize the Hamiltonian

$$H_0 = \frac{p^2}{2m} + \int_{-1}^{1} \frac{dx'}{\pi} V(x - \epsilon \lambda L x') \frac{1}{\sqrt{1 - x'^2}}$$

that is generally an impossible task analytically.

What we want to do is to analyze this problem in the limit where the amplitude of the quiver motion of the free-electron in the laser field is much larger than the atomic radius, further specializing the above problem to the Coulomb potential.

To evaluate the degree of deformation due to the laser field in this approximation we apply a modified Rayleigh-Schrödinger approximation as obtainable from the time-dependent perturbation series. This can be accomplished easily if one makes the multipolar expansion of the dressed Coulomb potential in eq.(47) as

$$V_{KH} = -\frac{Ze^2}{r} \left[ 1 + \sum_{n=1}^{+\infty} A_n \left( \frac{\lambda L}{r} \right)^{2n} P_{2n} \left( \frac{x}{r} \right) \right]$$

being $A_n = \int_{-1}^{1} dx x^{2n} / (\pi \sqrt{1 - x^2})$ and $P_n$ the $n$-th Legendre polynomial. So, we approach the static part of the Kramers-Henneberg Hamiltonian differently from the time-dependent part. The reason to do that is that we expect very large shifts of the energy levels and that just very few terms of the multipolar series (48) really contributes to the matrix elements in the Rayleigh-Schrödinger series.

The way we compute the Rayleigh-Schrödinger corrections is taken from the time-dependent perturbation theory. Indeed, let us suppose that one can neglect the time dependent part of the Kramers-Henneberger Hamiltonian, so that one is left with the time-independent problem

$$H_A = \frac{p^2}{2m} - \frac{Ze^2}{r} + \delta V_{KH}(x)$$

where we have put

$$\delta V_{KH} = -\frac{Ze^2}{r} \sum_{n=1}^{+\infty} A_n \left( \frac{\lambda L}{r} \right)^{2n} P_{2n} \left( \frac{x}{r} \right)$$

and we want to solve the problem

$$H_A |\psi(t)\rangle = i \frac{\partial |\psi(t)\rangle}{\partial t}.$$ 

This is the way stabilization is studied in the Kramers-Henneberger frame when the limit $\omega \to \infty$ is taken [3,42]. Here we are able to prove that stabilization indeed exists in the limit of large ratio of free-electron quiver motion and Bohr radius due to the rigidity of the atomic wave function.
We take as unperturbed states the ones of the Coulomb problem by setting
\[ |\psi(t)\rangle = \sum_n a_n(t)e^{-iE_nt}|n\rangle \]

being
\[ \left( \frac{p^2}{2m} - \frac{Ze^2}{r} \right)|n\rangle = E_n|n\rangle. \] (53)

One gets for the amplitudes
\[ ia_m(t) = \langle m|\delta V_{KH}(x)|m\rangle a_m(t) + \sum_{n \neq m} e^{-i(E_n-E_m)t}\langle m|\delta V_{KH}(x)|n\rangle a_n(t) \]

and introducing \( b_m(t) = e^{-i\delta E_m t}a_m(t) \), \( \delta E_m = \langle m|\delta V_{KH}(x)|m\rangle \) and \( \tilde{E}_m = E_m + \delta E_m \), we arrive finally at the equations
\[ ib_m(t) = \sum_{n \neq m} e^{-i(\tilde{E}_n-\tilde{E}_m)t}\langle m|\delta V_{KH}(x)|n\rangle b_n(t). \] (55)

Then, if the shifts \( \delta E_m \) are really large in the limit of large ratio between the quiver amplitude of the motion of the free electron and the Bohr radius, the Rayleigh-Schrödinger corrections to the initial wave-function are really small and this state is “rigid” with respect to the perturbation introduced by the laser field. Indeed, one has
\[ b_m(t) = b_m(0) + \sum_{n \neq m} e^{-i(\tilde{E}_n-\tilde{E}_m)t}\frac{\langle m|\delta V_{KH}(x)|n\rangle}{\tilde{E}_n - \tilde{E}_m}b_n(0) + \cdots \]

where an adiabatic switching of the perturbation has been introduced. We see that the first order correction gives a modification of the Rayleigh-Schrödinger perturbation theory as in place of the unperturbed energy levels \( E_m \) there are the modified energy levels \( \tilde{E}_m \). This appears also as an improvement with respect to the Brillouin-Wigner perturbation series. Indeed, one has for the wave function
\[ |\psi(t)\rangle = \sum_n e^{-i\tilde{E}_n t}a_n(0)|n\rangle + \sum_n e^{-i\tilde{E}_n t} \sum_{k \neq n} e^{-i(\tilde{E}_k-\tilde{E}_n)t}\frac{\langle n|\delta V_{KH}(x)|k\rangle a_k(0)|k\rangle}{\tilde{E}_k - \tilde{E}_n} + \cdots. \] (57)

In this way is possible to show that the first-order correction is really small.

For the level shift one gets for the first few levels (we have put \( a_B \) for the Bohr radius, \( l \) for the orbital angular moment and \( l_z \) for the third component of the orbital angular moment):
\[ n=1 \]
\[ \delta E_{001} = \langle l = 0, l_z = 0, n = 1|\delta V_{KH}(x)|l = 0, l_z = 0, n = 1 \rangle = 0 \] (58)
\[ n=2 \]
\[ \delta E_{102} = \langle l = 1, l_z = 0, n = 2 | \delta V_{KH}(x) | l = 1, l_z = 0, n = 2 \rangle = \frac{1}{240} \frac{Ze^2}{a_B} \left( \frac{\lambda_L}{a_B} \right)^2 \]
\[ \delta E_{112} = \delta E_{1 \rightarrow 12} = \langle l = 1, l_z = 1, n = 2 | \delta V_{KH}(x) | l = 1, l_z = 1, n = 2 \rangle = -\frac{1}{480} \frac{Ze^2}{a_B} \left( \frac{\lambda_L}{a_B} \right)^2 \]
\[ \delta E_{002} = \langle l = 0, l_z = 0, n = 2 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 2 \rangle = 0 \]
\[ n = 3 \]
\[ \delta E_{223} = \langle l = 2, l_z = 2, n = 3 | \delta V_{KH}(x) | l = 2, l_z = 2, n = 3 \rangle = -\frac{Ze^2}{a_B} \left( \frac{1}{5760} \left( \frac{\lambda_L}{a_B} \right)^2 + \frac{1}{81680} \left( \frac{\lambda_L}{a_B} \right)^4 \right) \]
\[ \delta E_{213} = \delta E_{2 \rightarrow 13} = \langle l = 2, l_z = 1, n = 3 | \delta V_{KH}(x) | l = 2, l_z = 1, n = 3 \rangle = \frac{Ze^2}{a_B} \left( \frac{1}{11340} \left( \frac{\lambda_L}{a_B} \right)^2 + \frac{1}{204120} \left( \frac{\lambda_L}{a_B} \right)^4 \right) \]
\[ \delta E_{203} = \langle l = 2, l_z = 0, n = 3 | \delta V_{KH}(x) | l = 2, l_z = 0, n = 3 \rangle = -\frac{Ze^2}{a_B} \left( \frac{1}{5670} \left( \frac{\lambda_L}{a_B} \right)^2 + \frac{1}{136080} \left( \frac{\lambda_L}{a_B} \right)^4 \right) \]
\[ \delta E_{103} = \langle l = 1, l_z = 0, n = 3 | \delta V_{KH}(x) | l = 1, l_z = 0, n = 3 \rangle = \frac{1}{810} \frac{Ze^2}{a_B} \left( \frac{\lambda_L}{a_B} \right)^2 \]
\[ \delta E_{113} = \delta E_{1 \rightarrow 13} = \langle l = 1, l_z = 1, n = 3 | \delta V_{KH}(x) | l = 1, l_z = 1, n = 3 \rangle = -\frac{1}{1620} \frac{Ze^2}{a_B} \left( \frac{\lambda_L}{a_B} \right)^2 \]
\[ \delta E_{003} = \langle l = 0, l_z = 0, n = 3 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 3 \rangle = 0 \]
\[ n = 4 \]
\[ \delta E_{334} = \langle l = 3, l_z = 3, n = 4 | \delta V_{KH}(x) | l = 3, l_z = 3, n = 4 \rangle = -\frac{Ze^2}{a_B} \left( \frac{1}{32256} \left( \frac{\lambda_L}{a_B} \right)^2 + \frac{3}{50462720} \left( \frac{\lambda_L}{a_B} \right)^4 + \frac{25}{113359454208} \left( \frac{\lambda_L}{a_B} \right)^6 \right) \]
\[ \delta E_{324} = \delta E_{3 \rightarrow 24} = \langle l = 3, l_z = 2, n = 4 | \delta V_{KH}(x) | l = 3, l_z = 2, n = 4 \rangle = -\frac{Ze^2}{a_B} \left( \frac{1}{7208960} \left( \frac{\lambda_L}{a_B} \right)^4 + \frac{25}{18893242368} \left( \frac{\lambda_L}{a_B} \right)^6 \right) \]
\[ \delta E_{314} = \delta E_{3 \rightarrow 14} = \langle l = 3, l_z = 1, n = 4 | \delta V_{KH}(x) | l = 3, l_z = 1, n = 4 \rangle = -\frac{Ze^2}{a_B} \left( \frac{1}{53760} \left( \frac{\lambda_L}{a_B} \right)^2 + \frac{1}{50462720} \left( \frac{\lambda_L}{a_B} \right)^4 + \frac{125}{37786484736} \left( \frac{\lambda_L}{a_B} \right)^6 \right) \]
\[ \delta E_{304} = \langle l = 3, l_z = 0, n = 4 | \delta V_{KH}(x) | l = 3, l_z = 0, n = 4 \rangle = -\frac{Ze^2}{a_B} \left( \frac{1}{40320} \left( \frac{\lambda_L}{a_B} \right)^2 + \frac{3}{25231360} \left( \frac{\lambda_L}{a_B} \right)^4 - \frac{125}{28339863552} \left( \frac{\lambda_L}{a_B} \right)^6 \right) \]
\[ \delta E_{224} = \delta E_{2 \rightarrow 24} = \langle l = 2, l_z = 2, n = 4 | \delta V_{KH}(x) | l = 2, l_z = 2, n = 4 \rangle = -\frac{Ze^2}{a_B} \left( \frac{1}{13440} \left( \frac{\lambda_L}{a_B} \right)^2 + \frac{3}{4587520} \left( \frac{\lambda_L}{a_B} \right)^4 \right) \]
\[ \delta E_{214} = \delta E_{2 \rightarrow 14} = \langle l = 2, l_z = 1, n = 4 | \delta V_{KH}(x) | l = 2, l_z = 1, n = 4 \rangle = \frac{Ze^2}{a_B} \left( \frac{1}{26880} \left( \frac{\lambda_L}{a_B} \right)^2 + \frac{3}{1146880} \left( \frac{\lambda_L}{a_B} \right)^4 \right) \]
\[ \delta E_{204} = \langle l = 2, l_z = 0, n = 4 | \delta V_{KH}(x) | l = 2, l_z = 0, n = 4 \rangle = -\frac{Ze^2}{a_B} \left( \frac{1}{13440} \left( \frac{\lambda_L}{a_B} \right)^2 + \frac{9}{2293760} \left( \frac{\lambda_L}{a_B} \right)^4 \right) \]
\[
\delta E_{114} = \delta E_{1-14} = \langle l = 1, l_z = 1, n = 4 | \delta V_{KH}(x) | l = 1, l_z = 1, n = 4 \rangle = -\frac{1}{3840} \frac{Ze^2}{a_B} \left( \frac{\lambda_L}{a_B} \right)^2
\]
\[
\delta E_{104} = \langle l = 1, l_z = 0, n = 4 | \delta V_{KH}(x) | l = 1, l_z = 0, n = 2 \rangle = \frac{1}{1920} \frac{Ze^2}{a_B} \left( \frac{\lambda_L}{a_B} \right)^2
\]
\[
\delta E_{004} = \langle l = 0, l_z = 0, n = 4 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 4 \rangle = 0
\]

(61)

It easily realized that just very few terms of the multipolar series of \( \delta V_{KH} \) really contribute to level shifts making the argument working. Beside, as it should be expected the s-states have no shift.

Indeed, for the matrix elements, assuming as initial state the ground state of the atom one obtains

\[
\langle l = 1, l_z = 1, n = 2 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]
\[
\langle l = 1, l_z = 0, n = 2 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]
\[
\langle l = 0, l_z = 0, n = 2 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]

(62)

\[
\langle l = 2, l_z = 2, n = 3 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = -\frac{Ze^2}{2} \frac{1}{720} \left( \frac{\lambda_L}{a_B} \right)^2
\]
\[
\langle l = 2, l_z = 1, n = 3 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]
\[
\langle l = 2, l_z = 0, n = 3 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = \frac{Ze^2 \sqrt{150}}{10800} \left( \frac{\lambda_L}{a_B} \right)^2
\]
\[
\langle l = 1, l_z = 1, n = 3 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]
\[
\langle l = 1, l_z = 0, n = 3 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]
\[
\langle l = 0, l_z = 0, n = 3 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]

(63)

\[
\langle l = 3, l_z = 3, n = 4 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]
\[
\langle l = 3, l_z = 2, n = 4 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]
\[
\langle l = 3, l_z = 1, n = 4 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]
\[
\langle l = 3, l_z = 0, n = 4 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]
\[
\langle l = 2, l_z = 2, n = 4 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = \frac{Ze^2 13 \sqrt{150}}{150000} \left( \frac{\lambda_L}{a_B} \right)^2
\]
\[
\langle l = 2, l_z = 1, n = 4 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]
\[
\langle l = 2, l_z = 0, n = 4 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = \frac{Ze^2 13}{15000} \left( \frac{\lambda_L}{a_B} \right)^2
\]
\[
\langle l = 1, l_z = 1, n = 4 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]
\[
\langle l = 1, l_z = 0, n = 4 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]
\[
\langle l = 0, l_z = 0, n = 4 | \delta V_{KH}(x) | l = 0, l_z = 0, n = 1 \rangle = 0
\]

(64)

Again we see that just very few terms of \( \delta V_{KH} \) really contribute to the matrix elements. Beside, this contribution is smaller if not zero with respect to the
level shifts. Then, these results strongly support the statement that the first-order correction into the series \((57)\) is really small in the limit \(\frac{\lambda}{a_B} \gg 1\) as it should be. This also supports stabilization when the limit of laser frequency going to infinity is taken, with the ponderomotive energy being constant as in this limit, the time-dependent part of the Kramers-Henneberger Hamiltonian can be neglected [1,32].

Finally, we conclude that a fairly good approximation for the unperturbed Hamiltonian for our aims is

\[
H_A \approx \sum_n \tilde{E}_n |n\rangle \langle n|
\] (65)

where just diagonal terms are kept and the off-diagonal terms

\[
H_A' = \sum_{n \neq m} |m\rangle \langle n| \delta V_{KH} |n\rangle
\] (66)

are neglected. We take the Hamiltonian (65) for time-dependent computations in perturbation theory.

V. PERTURBATION THEORY FOR ATOMS IN INTENSE LASER FIELDS

The experiments carried out to produce harmonics have a small laser frequency with respect to ionization and ponderomotive energy, then also the time dependent components of the Kramers-Henneberger Hamiltonian need to be considered.

Our aim is to make a perturbation analysis of the Hamiltonian

\[
H = \sum_n \tilde{E}_n |n\rangle \langle n| + 2 \varepsilon \omega \gamma \frac{x}{a_B} \sum_{k=0}^{+\infty} (2k + 1) \sin((2k + 1)\omega t)
\] (67)

being

\[
\gamma = \frac{Ze^2}{\lambda L \omega} = \sqrt{\frac{I_B}{2U_p}}
\] (68)

the Keldysh parameter. So, the perturbation theory is applicable in this case just when \(\gamma \ll 1\) that defines the so-called tunnelling regime for the electron in the laser field. But, the situation here is more favourable as we have just to require that the distance between energy levels is much larger than \(\omega \gamma\) and we are able to account for a larger number of experiments than the tunnelling regime would permit.

We write the equations for the probability amplitudes as

\[
i \dot{a}_n(t) = -i \varepsilon \omega \gamma \sum_n e^{-i(\tilde{E}_n - \tilde{E}_m)t} a_m(t) \left( m \frac{x}{a_B} \right) \sum_{k=0}^{+\infty} (2k + 1) \left( e^{i(2k+1)\omega t} - e^{-i(2k+1)\omega t} \right) .\] (69)

Then, a generic term, out of resonance, will be written as
\[ a_m(t) = a_m(0) + \]
\[ i \epsilon \omega \gamma \sum_n a_n(0) \left< m \left| \frac{x}{a_B} \right| n \right> \sum_{k=0}^{+\infty} \left[ (2k+1) \left( \frac{e^{i(2k+1)\omega \cdot \Delta E_m t} - 1}{(2k+1)\omega - \Delta E_n + \Delta E_m} \right) + \cdots \right]. \tag{70} \]

This result shows that when there are two resonant states \( m \) and \( n \) in the laser field, then we have Rabi flopping with Rabi frequency given by
\[ \frac{\Omega_R}{2} = \epsilon \gamma (2k+1) \omega \left< m \left| \frac{x}{a_B} \right| n \right> \] determined by the \((2k+1)\)-th harmonic of the laser frequency. This implies a significant modification of the spectrum of the harmonics with respect to the observed patterns. Otherwise, out of resonance amplitudes are really small due to the large shifts in the limit of large \( \frac{\lambda}{a_B} \) ratio. So, the interesting case is the resonance with the set of continuous states of the atom representing the situation of the recollision model. In this case, instead of flopping, we can have decay, that means ionization, and a probability for the electron to turn back to the core emitting radiation.

Then, we can apply the Wigner-Weisskopf argument \[33\] to eq. (70). The first approximation is to reduce the system to the two levels really resonating and to assume all the other amplitudes to be small in the sense given above. This means to approximate eq. (70) by
\[ \dot{a}_0(t) \approx -\epsilon \omega \gamma \sum_p e^{-i(E_p - E_0) t} \left< 0 \left| \frac{x}{a_B} \right| p \right> a_p(t) \sum_{k=0}^{+\infty} (2k+1) e^{i(2k+1)\omega t} \tag{72} \]
where we have labelled the eigenstates of continuum for the atom by the momentum \( p \) and \( E_0 = -I_B \). We seek a solution of this equations by setting
\[ a_0(t) = e^{-\frac{\Gamma'}{2} t}. \tag{73} \]

By substituting eq. (73) into eq. (72) one arrives at the condition
\[ \frac{\Gamma'}{2} = i \epsilon^2 \gamma^2 \omega^2 \sum_p \left< 0 \left| \frac{x}{a_B} \right| p \right|^2 \sum_{k=0}^{+\infty} (2k+1)^2 \left\{ 1 - e^{-i(E_p - E_0 - (2k+1)\omega) t + i \frac{\Gamma'}{2} t} \right\} \tag{74} \]
For small \( \Gamma' \) this indeed reduces to a time-independent expression \[34\]
\[ \frac{\Gamma'}{2} = i \epsilon^2 \gamma^2 \omega^2 \sum_p \left< 0 \left| \frac{x}{a_B} \right| p \right|^2 \sum_{k=0}^{+\infty} (2k+1)^2 \left[ \mathcal{P} \frac{1}{(2k+1)\omega - E_p + E_0 - i \delta(E_p - E_0 - (2k+1)\omega)} \right] \tag{75} \]
with \( \mathcal{P} \) meaning the principal value. So, finally, one gets the a.c. Stark shift of the ground state of the atom given by
\[
\frac{\delta \omega}{2} = e^2 \gamma^2 \omega^2 \sum_p \left| \langle 0 \left| \frac{x}{a_B} \right| p \rangle \right|^2 \sum_{k=0}^{+\infty} (2k + 1)^2 \frac{1}{p} \left( \frac{k + 1}{\omega - E_p + E_0} \right) \]

and the decay rate for the ionization of the atom

\[
\Gamma = 2\pi e^2 \gamma^2 \omega^2 \sum_p \left| \langle 0 \left| \frac{x}{a_B} \right| p \rangle \right|^2 \sum_{k=0}^{+\infty} (2k + 1)^2 \delta(E_p - E_0 - (2k + 1)\omega). \tag{77}
\]

Then, for the continuum one has

\[
a_p(t) \approx -i\epsilon\gamma \omega \sum_{k=0}^{+\infty} \left\langle p \left| \frac{x}{a_B} \right| 0 \right\rangle (2k + 1) e^{i[Ep - E_0 - (2k + 1)\omega + \frac{\Gamma^2}{2}t]} - 1 \left( E_p - E_0 - (2k + 1)\omega + \frac{\Gamma^2}{2} \right) \tag{78}
\]

so that, taking

\[
|\psi(t)\rangle = a_0(t) e^{iH_0 t}|0\rangle + \sum_p e^{-iE_p t} a_p(t)|p\rangle \tag{79}
\]

one has for the harmonic spectrum

\[
\langle \psi(t)|x|\psi(t)\rangle = \sum_p \left( e^{i(E_p + I_B)t} a_0(t) a_p^*(t) \left( p \left| \frac{x}{a_B} \right| 0 \right) + e^{-i(E_p + I_B)t} a_p(t) a_0^*(t) \langle 0 \left| x \right| p \rangle \right) \tag{80}
\]

where use has been made of the fact that \( \langle 0|\langle 0 \rangle = \langle p|x|p \rangle = 0 \). Assuming the laser light coming from the far past, eq. (78) can rewritten as

\[
a_p(t) \approx -i\epsilon\gamma \omega \sum_{k=0}^{+\infty} \left\langle p \left| \frac{x}{a_B} \right| 0 \right\rangle (2k + 1) e^{i[Ep - E_0 - (2k + 1)\omega + \frac{\Gamma^2}{2}t]} \left( \frac{\Gamma}{\pi} \right) \left( \frac{E_p - E_0 - (2k + 1)\omega - \frac{\delta\omega}{2}}{\left[ E_p - E_0 - (2k + 1)\omega - \frac{\delta\omega}{2} \right]^2 + \frac{\Gamma^2}{4}} \right) \tag{81}
\]

and we are left with the resonant part of the spectrum given by

\[
\langle \psi(t)|x|\psi(t)\rangle_R \approx -2\pi\epsilon\gamma \omega \sum_p \left| \langle 0 \left| \frac{x}{a_B} \right| p \rangle \right|^2 \sum_{n=0}^{+\infty} (2n + 1) \frac{\frac{\Gamma}{\pi} \left( \frac{E_p - E_0 - (2k + 1)\omega - \frac{\delta\omega}{2}}{\left[ E_p - E_0 - (2k + 1)\omega - \frac{\delta\omega}{2} \right]^2 + \frac{\Gamma^2}{4}} \right)}{\left[ E_p - E_0 - (2k + 1)\omega - \frac{\delta\omega}{2} \right]^2 + \frac{\Gamma^2}{4}} \times e^{-\Gamma|t|} \cos\left[ (2n + 1)\omega t + \frac{\delta\omega}{2} \right]. \tag{82}
\]

This form of the spectrum gives two main results: Firstly, one has all the harmonics shifted by the same quantity \( \frac{\delta\omega}{2} \) and secondly each harmonic has a Lorentzian form that, for very small \( \Gamma \) can be reduced to a Dirac \( \delta \) distribution.

In this case the integration in \( \mathbf{p} \) can be done taking a plane wave as a final state and neglecting the shift of the spectrum, one gets an improved version of the result of Ref. [1], that is

\[
\langle \psi(t)|x|\psi(t)\rangle_R \approx \frac{2}{\pi} \frac{2\pi^2}{3} \frac{Z e^2}{U_p^2} \frac{\omega}{\gamma^2} \sum_{n=n_0}^{+\infty} \frac{x_n^\frac{3}{2}}{(x_n + 2\omega x_n + \frac{\delta\omega}{2})^3} \cos((2n + 1)\omega t)e^{-\Gamma|t|}. \tag{83}
\]

being \( x_n = \frac{(2n + 1)\omega - I_B}{3U_p} \) and \( n_0 \) is a lower cut-off given by the integer value such that \( (2n_0 + 1)\omega - I_B \geq 0 \) firstly foreseen in Ref. [1]. Being scaled in this way through \( U_p, I_B \) and \( \omega \), a simple model of a single electron in an atom
with atomic number $Z_{eff}$ can be adopted for complex atoms in a strong laser field and the above result used also in this case. As shown in Ref. [9], the order of magnitude is correct. The same is true for $\Gamma$ given by

$$\Gamma = \frac{256 \omega^2 \gamma^2}{3\pi^2 U_p} \sum_{n=n_0}^{+\infty} \left[ \frac{I_B}{(2n+1)\omega} \right]^{\frac{3}{2}} \left[ 1 - \frac{I_B}{(2n+1)\omega} \right]^{\frac{1}{2}}$$

(84)

that for the experiment described in Ref. [17] yields for helium .012 eV and for neon .01 eV improving the results given in Ref. [9]. This in turn means that the mean lifetime of the harmonics ranges from 52 to 68 fs and, by e-folding, after a time of few $10^2$ fs the harmonics disappear in agreement with the experimental results. The result is independent from the order of the harmonics. We expect that the value of $\Gamma$ should be in better agreement with the experimental results when $U_p$ increases or being the same, when the Keldysh parameter $\gamma$ decreases marking the so called tunnelling regime.

VI. CONCLUSIONS

We have given a full theory of harmonic generation starting from non-relativistic quantum electrodynamics in the dipole approximation. A relation with quantum chaos is obtained through a kicked model that originates from the model at small distances where a regularization procedure is needed. Perturbation theory can be done in the tunnelling regime, where the Keldysh parameter is small, as the atomic wavefunction turns out to be rigid due to the way the energy levels are shifted by the laser field. In this way a deep connection between ionization rate and harmonic spectrum is given. Finally, a fully understanding of the recollision model is obtained and this was our main aim.
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