Theory of dressed states in quantum optics

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The dual Dyson series [M. Frasca, Phys. Rev. A **58** 3439 (1998)] is used to develop a general perturbative method for the study of atom-field interaction in quantum optics. In fact, both the Dyson series and its dual, by renormalization-group methods to remove secular terms from the perturbation series, give the opportunity for a full study of the solution of the Schrödinger equation in different ranges of the parameters of the given Hamiltonian. In view of recent experiments with strong laser fields, this approach seems well-suited to give a clarification and an improvement of the applications of the dressed states as currently done through the eigenstates of the atom-field interaction, showing that these are just the leading order of the dual Dyson series when the Hamiltonian is expressed in the interaction picture. In order to best exploit the method, a study is accomplished of the well-known Jaynes-Cummings model in the rotating-wave approximation, whose exact solution is known, comparing the perturbative solutions obtained by the Dyson series and its dual with the same approximations obtained by Taylor expanding the exact solution. Finally, a full perturbative study of high-order harmonic generation is given, obtaining, through analytical expressions, a clear account of the power spectrum using a two-level model, even if the method can be successfully applied to a more general model that can account for ionization too. The analysis shows that to account for the power spectrum it is necessary to go to first order in the perturbative analysis. The spectrum obtained gives a way to measure experimentally the shift of the energy levels of the atom interacting with the laser field by looking at the shifting of hyper-Raman lines. [S1050-2947(99)03807-X]

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I. INTRODUCTION

Recent experiments on atoms using strong laser fields [1] have shown the appearance of a wealth of effects, e.g., high-order harmonics generation, in the interaction between light and atoms. This situation forced researchers to find different approaches to describe the outcomes of those experiments. Numerical studies of the time-dependent Schrödinger equation [2] have shown that the two-level model still proves to be very useful to describe all the features of harmonics generation [3], even if the rotating-wave approximation must be abandoned. Indeed, recent work [4,5] indicates, by comparing results from a two-level model using Floquet states and numerical work on the Schrödinger equation, that the simple two-level model is fairly effective in describing the physical situation at hand. So far, no perturbative solution seems to be known of this two-level model beyond Floquet states for the case of a strong laser field. But, a study by Meystre of an atom in a Fabry-Perot cavity [6] used the same model of Ref. [5] and gave a perturbative analytical solution to such a model in a strong coupling regime. In fact, the analytical solution given by Meystre and its higher-order corrections has been successfully obtained in Ref. [7], showing that the levels of the atom undergoes a shift. Being the same model, now we have at hand a way to observe experimentally such a shift through hyper-Raman lines in harmonic generation, if one is able to properly account for the spectrum.

An understanding of interaction between an atom and a strong electromagnetic field has been possible in recent years through the introduction of the dressed-atom picture [8]. This approach assumes that the field couples the levels of the atom in such a way that the interaction is between this ''dressed'' atom and the field itself. The computation of the corresponding dressed states, as currently found in literature, involves the computation of the eigenstates and the eigenvalues of the term of interaction between the atom and the field in the Hamiltonian, either the computation of the eigenstates of the full Hamiltonian, taking into account in this way the field too. From a physical standpoint the dressed-atom picture is quite general as it assumes that the photons of the field surround the atom as to modify the way the atom itself responds to the field; then it should concern a fully second quantized theory. But, the computation of the eigenstates of the full Hamiltonian or just the atom-field interaction term, which we take to be the dressed states, often reveals itself as an approximation scheme whose understanding is the main aim of this paper. So far, no reason has been known for the nice working of such dressed states in applied mathematics. A recently devised approach [9], the dual Dyson perturbation series, turns out to be both an explanation and an improvement of the computation of dressed states permitting the computation of higher-order corrections to a leading-order solution obtained through such dressed states. As a by-product one has a clear physical understanding of what are the parameters involved in such approximate dressed states and what is going to be neglected. So, by this improvement of the computation of dressed states, we are able to find an analytical perturbative solution to the two-level model to analyze high-order harmonic generation showing that this is a first-order effect, that is, the leading-order solution found by Meystre is not enough to get the right spectrum. Then, the result properly accounts for the relevance of population distribution as discussed in Ref. [5] and an analytical closed expression is given.

The dual Dyson series that accounts for the dressed states as defined above can be derived from the time-dependent

Schrödinger equation by using the duality principle in perturbation theory and the quantum adiabatic approximation [9]. In this way one realizes that the dual Dyson series is the same one as Ref. [10]. The results one gets from what should work just for quantum adiabatic processes can appear somewhat unexpected, as it will be shown for the Jaynes-Cummings model in the rotating wave approximation (RWA) for whom an exact solution is known. But, this just agrees with the results of Ref. [9].

So, the existence of a dual Dyson series can improve the study of atom-field interaction. In fact, one can accomplish a perturbative analysis of models in quantum optics in different regions of the parameter space that for a Jaynes-Cummings model can be easily identified, when spontaneous emission is neglected, with the ratio between the detuning and the Rabi frequency. Then, by generalizing the computation of dressed states through the dual Dyson series on one side and by the standard Dyson series on the other, we can reach the main aim of this paper: A general perturbative method to study atom-field interaction in quantum optics at different values of the parameters of the Hamiltonian.

The completeness of our approach is strongly tied with the recent results obtained in quantum optics through the renormalization-group methods for perturbation theory [7]. These methods permit the resummation of the so-called secularities that appear in perturbation theory. Indeed, we are able to derive an energy level shift of the atom in high-order harmonic generation that has an effect on hyper-Raman lines. As shown in Ref. [5], when the two levels of the atom are equally populated, only hyper-Raman lines should be observed. Then, in view of this situation, such an energy-level shift turns out to be significant.

It should be pointed out that, although the extension of this approach to the method of the master equation [8] should be straigthforward, it is not considered in this paper. So, e.g., the effect of vacuum fluctuations of the field modes is neglected.

The paper is so structured. In Sec. II we give a general description of the methods and show why the eigenstates of the perturbation are important for strong fields. In Sec. III a study of the Jaynes-Cummings model in RWA is accomplished in order to have a pedagogical description of the methods and a comparation with an exact solution. In Sec. IV the question of high-order harmonic generation is discussed through the methods so far introduced.

II. A GENERAL METHOD FOR PERTURBATIVE ANALYSIS

A. General theory

In Ref. [9] we have introduced the duality principle in perturbation theory. By duality we mean that, for a given differential equation, it is possible to compute both a perturbation series in λ and $1/\lambda$, λ being the characteristic parameter of the equation. This is accomplished by a proper choice of the leading-order equation. So, e.g., for the Duffing equation

$$\ddot{x}+x+\lambda x^3=0, \tag{1}$$

one can compute a series in λ and $1/\lambda$ by taking, at leading order, in the former case $\ddot{x}+x=0$ and in the latter case $\ddot{x}+\lambda x^3=0$. It is easy to see that the duality principle is true independently by our ability to do the computations of the equations one gets from the perturbation series.

In turn, the existence of a duality principle in perturbation theory means that a perturbative analysis is possible in different regions of the parameter space of the given equation. This situation could turn out to be very useful in quantum mechanics if one is able to obtain a dual Dyson series. This is indeed the case.

So, let us consider the time-dependent Schrödinger equation,

$$H(t)|\psi\rangle=i\frac{\partial|\psi\rangle}{\partial t}, \tag{2}$$

where $H(t)$ is the Hamiltonian and $\hbar=1$ here and in the following. The Dyson series is a perturbative solution of this equation given by

$$|\psi(t)\rangle=\left(I-i\int_{t_0}^{t}dt_1\,H(t_1)-\int_{t_0}^{t}dt_1\int_{t_0}^{t_1}dt_2\,H(t_1)H(t_2)+\cdots\right)|\psi(t_0)\rangle \tag{3}$$

or, by introducing the time-ordering operator $\mathcal{T}$,

$$|\psi(t)\rangle=\mathcal{T}\exp\left(-i\int_{t_0}^{t}dt'\,H(t')\right)|\psi(t_0)\rangle. \tag{4}$$

The dual series can be obtained, through the duality principle, by assuming that the Hamiltonian $H(t)$ has a discrete spectrum, that is, $H(t)|n,t\rangle=E_n(t)|n,t\rangle$ with $|n,t\rangle$ the eigenstate corresponding to the eigenvalue $E_n(t)$. Then, the dual Dyson series is the one given in Ref. [10], that is,

$$|\psi(t)\rangle=U_A(t)\mathcal{T}\exp\left(-i\int_{t_0}^{t}d\hat{t}\,H'(\hat{t})\right)|\psi(t_0)\rangle \tag{5}$$

being

$$U_A(t)=\sum_n e^{i\gamma_n(t)-i\int_{t_0}^{t}dt'E_n(t')}|n,t\rangle\langle n,t_0|, \tag{6}$$

the adiabatic unitary evolution operator, for the Berry phase $\dot{\gamma}_n(t)=\langle n,t|i\partial/\partial t|n,t\rangle$ and

$$H'(t)=-\sum_{n,m,n\neq m}e^{-i[\gamma_m(t)-\gamma_n(t)]}e^{i\int_{t_0}^{t}dt'[E_m(t')-E_n(t')]}\times\left\langle m,t\left|i\hbar\frac{\partial}{\partial t}\right||n,t\right\rangle|m,t_0\rangle\langle n,t_0|. \tag{7}$$

This result proves that the well-known adiabatic approximation and its higher-order corrections can be very effective in building asymptotic approximations to the solution of the Schrödinger equation, as is, on the other side, the Dyson series.

Let us now consider a perturbed quantum system with Hamiltonian

$$H=H_0+V(t), \tag{8}$$

where H_0 is the Hamiltonian of the unperturbed system and $V(t)$ is the perturbation. In the interaction picture one has

$$H_I(t)=e^{iH_0t}V(t)e^{-iH_0t}. \tag{9}$$

It is now possible to study the given system in different regions of the parameter space through the Dyson series and its dual. In the former case we have standard textbook time-dependent perturbation theory. In the latter case we have to compute

$$H_I(t)|n,t\rangle_I=E_n^{(I)}(t)|n,t\rangle_I. \tag{10}$$

But $H_I(t)$ is just the interaction $V(t)$ transformed by a unitary transformation. Then, the eigenvalues $E_n^{(I)}(t)$ are those of the perturbation $V(t)$ and the eigenstates $|n,t\rangle_I$ are just a unitary transformation away from the corresponding eigenstates. These are the dressed states as generally computed in the current literature: It is just the leading-order approximation of a dual Dyson series. But now we have a more general theory and higher-order corrections can be computed. Besides, we realize why the dressed states are so effective in a strong-field regime being obtained from the dual Dyson series that has a development parameter exactly inverse of the one of the Dyson series.

It should be pointed out that both Dyson series and its dual can have the same kind of problems. One of the most important is surely the question of secularities: In any case, resummation of secular terms can be achieved through the renormalization group methods as pointed out, for quantum optics, in Ref. [7].

B. An example

To give a clear insight of the working of the above analysis for a differential equation, let us consider the standard textbook example,

$$\psi''(x)+\alpha^2(x)\psi(x)=0, \tag{11}$$

which can be written in the form (the i factor is introduced just for convenience),

$$i\frac{d}{dx}\begin{pmatrix}\psi(x)\\ \phi(x)\end{pmatrix}=\begin{pmatrix}0 & i\\ -i\alpha^2(x) & 0\end{pmatrix}\begin{pmatrix}\psi(x)\\ \phi(x)\end{pmatrix}=L(x)\begin{pmatrix}\psi(x)\\ \phi(x)\end{pmatrix}. \tag{12}$$

We can apply Dyson series and its dual. Dyson series is not normally applied to the above equation. Indeed, it gives the expansion

$$\begin{pmatrix}\psi(x)\\ \phi(x)\end{pmatrix}=\left[I-i\int_{x_0}^{x}dx'\begin{pmatrix}0 & i\\ -i\alpha^2(x') & 0\end{pmatrix}-\int_{x_0}^{x}dx'\int_{x_0}^{x'}dx''\begin{pmatrix}\alpha^2(x'') & 0\\ 0 & \alpha^2(x')\end{pmatrix}+\cdots\right]\times\begin{pmatrix}\psi(x_0)\\ \phi(x_0)\end{pmatrix}. \tag{13}$$

In order to compute the dual Dyson series, we need to compute the eigenvectors and eigenvalues of the matrix $L(x)$. So, for the eigenvalue $\alpha(x)$, one has the eigenvector

$$|1,x\rangle=\frac{1}{\sqrt{-2i\alpha(x)}}\begin{pmatrix}1\\ -i\alpha(x)\end{pmatrix} \tag{14}$$

and for the eigenvalue $-\alpha(x)$,

$$|2,x\rangle=\frac{1}{\sqrt{2i\alpha(x)}}\begin{pmatrix}1\\ i\alpha(x)\end{pmatrix}. \tag{15}$$

Then, one has for the Berry phases $\langle 2,x|id/dx|2,x\rangle=\langle 1,x|id/dx|1,x\rangle=0$ and the unitary evolution operator (6),

$$U_A(x,x_0)=\frac{1}{\sqrt{\alpha(x)\alpha(x_0)}}\begin{pmatrix}\alpha(x_0)\cos\left(\int_{x_0}^{x}dx'\alpha(x')\right) & \sin\left(\int_{x_0}^{x}dx'\alpha(x')\right)\\ -\alpha(x)\alpha(x_0)\sin\left(\int_{x_0}^{x}dx'\alpha(x')\right) & \alpha(x)\cos\left(\int_{x_0}^{x}dx'\alpha(x')\right)\end{pmatrix}. \tag{16}$$

It is straightforward to see that

$$\begin{pmatrix}\psi(x)\\ \phi(x)\end{pmatrix}\approx U_A(x,x_0)\begin{pmatrix}\psi(x_0)\\ \phi(x_0)\end{pmatrix} \tag{17}$$

gives the well-known Wentzel-Kramers-Brillouin-Jeffreys (WKBJ) result,

$$\psi(x)\approx\frac{C_1}{\sqrt{\alpha(x)}}\cos\left(\int_{x_0}^{x}dx'\alpha(x')\right)+\frac{C_2}{\sqrt{\alpha(x)}}\sin\left(\int_{x_0}^{x}dx'\alpha(x')\right). \tag{18}$$

In this derivation we have omitted the problem connected to turning points. We just note that, if there are points where $\alpha(x)=0$, Berry phases are no more zero as these are degeneracy points.

This example shows the full power of the adiabatic approximation in finding asymptotic approximations to a given differential equation, without any requirement of slow variation of the parameters of the equation. In the following we will show how to find higher-order corrections too.

C. Duality and Berry's asymptotics

Duality principle has been introduced in Ref. [9] to resolve problems both with infinitely small and large perturbations. As such, there is a region of the parameter space that is not possible to analyze by perturbation methods. But, it is not difficult to realize that, as a by-product, an alternative solution to the Schrödinger equation for its unitary evolution through Eq. (5) is obtained. This has no trivial consequences as, differently from the Dyson series, a superadiabatic scheme could be applied instead as devised by Berry [11] that could give nonperturbative informations on the dual series.

A superadiabatic scheme proves to be very useful when the full Hamiltonian is considered with no *a priori* large or small parts, as shown in Ref. [12] to describe stimulated Raman adiabatic passage by a three-level model. Indeed, the idea is to iterate the scheme to compute the adiabatic series giving $U_A(t)$ and $H'(t)$, by computing $U'_A(t)$ for $H'(t)$, and the Hamiltonian $H''(t)$ through the eigenstates of $H'(t)$. In principle, the procedure can be repeated to the step one wants, giving the unitary evolution $U(t) \sim U_A(t)U'_A(t)U''_A(t)\dots U^{(n)}(t)$ and it is tempting to stop to a given step to obtain an approximation to the unitary evolution but, actually, the procedure is shown to diverge. Anyhow, an optimal step n_c exists for which an eigenstate basis set can be built by the approximated $U(t)$ to approximate the solution of the Schrödinger equation. Divergence is due to the fact that off-diagonal terms computed by the Hamiltonians are systematically neglected.

Indeed, to address the question of dressed states we consider a Hamiltonian like

$$H=\frac{\omega_0}{2}\sigma_3+V(t)\sigma_1, \tag{19}$$

where $V(t)$ is a generic perturbation, σ_1 and σ_3 are Pauli matrices and ω_0 is the level separation of the model. The regimes of interest are fully perturbative as $V(t)$ is assumed to be very large. So, the initial Hamiltonian to apply the superadiabatic scheme is given, in interaction picture, by

$$H_I=e^{i\omega_0 t\sigma_3}V(t)\sigma_1. \tag{20}$$

In this case, the superadiabatic scheme just stops to the second step. Indeed, at the first step one has $U_A(t) = e^{i(\omega_0/2)\sigma_3 t}e^{-i\sigma_1\int_0^t dt'\,V(t')}$ and, at the second step, $U'_A(t) = U_A^\dagger(t)$. So, the product of unitary evolution operators is stopped and nothing new is obtained. Anyhow, the Berry's scheme can prove to be very useful in a nonperturbative regime, that is, when $V(t)$ and ω_0 are of the same order of magnitude and exponentially small factors can be retained. Then, we can conclude that a superadiabatic scheme turns out to be useful in an intermediate regime, being in this way a bridge between the small and large perturbation theory linked in turn by the duality principle. This matter deserves further investigation.

III. PERTURBATIVE ANALYSIS OF THE JAYNES-CUMMINGS MODEL

The Jaynes-Cummings model is widely used in quantum optics. Its Hamiltonian, in the RWA, is given by [8]

$$H_{JC}=\omega a^+a+\frac{\omega_0}{2}(|2\rangle\langle 2|-|1\rangle\langle 1|) +g(|2\rangle\langle 1|a^+ +|1\rangle\langle 2|a), \tag{21}$$

representing a two-level atom coupled with a single-mode radiation of frequency ω through the constant g. The reason to consider it here is that the exact solution is known and can be compared with the results of our perturbative analysis.

In the interaction picture one has the Hamiltonian,

$$H_{JC}^{(I)}=g(e^{i\Delta t}|2\rangle\langle 1|a^+ +e^{-i\Delta t}|1\rangle\langle 2|a), \tag{22}$$

where $\Delta=\omega_0-\omega$ is the detuning that here we assume different from 0 for the sake of generality. As it can be seen from the form of $H_{JC}^{(I)}$, the critical parameter in the model is the ratio g/Δ. This means that an eventual perturbation series and its dual will have this parameter and its inverse as a development parameter. Now, we proceed to compute those series from the exact solution.

The exact solution of the Schrödinger equation in interaction picture

$$H_{JC}^{(I)}|\psi\rangle_I=i\frac{\partial|\psi\rangle_I}{\partial t} \tag{23}$$

can be found by looking for a solution in the form

$$|\psi\rangle_I=\sum_n c_{1,n+1}(t)|1,n+1\rangle+c_{2,n}(t)|2,n\rangle, \tag{24}$$

where n is the photon number. So, the probability amplitudes are given by [8]

$$c_{1,n+1}(t)=\left\{c_{1,n+1}(0)\left[\cos\left(\frac{\Omega_n t}{2}\right)+\frac{i\Delta}{\Omega_n}\sin\left(\frac{\Omega_n t}{2}\right)\right] -\frac{2ig\sqrt{n+1}}{\Omega_n}c_{2,n}(0)\sin\left(\frac{\Omega_n t}{2}\right)\right\}e^{-i\Delta t/2},$$

$$c_{2,n}(t)=\left\{c_{2,n}(0)\left[\cos\left(\frac{\Omega_n t}{2}\right)-\frac{i\Delta}{\Omega_n}\sin\left(\frac{\Omega_n t}{2}\right)\right] -\frac{2ig\sqrt{n+1}}{\Omega_n}c_{1,n+1}(0)\sin\left(\frac{\Omega_n t}{2}\right)\right\}e^{i\Delta t/2}, \tag{25}$$

where $\Omega_n=\sqrt{\Delta^2+\mathcal{R}_n^2}$ and $\mathcal{R}_n=2g\sqrt{n+1}$ is the Rabi frequency. As expected, Δ and g are the only parameters, their ratio enters the only meaningful development parameter. The Dyson series is obtained by expanding the above solution in Taylor series of $\lambda=\mathcal{R}_n/\Delta$ giving till second order,

$$c_{1,n+1}(t)=\left\{c_{1,n+1}(0)\left[1+i\frac{\lambda^2}{4}[\Delta t+i(1-e^{-i\Delta t})]\right] -\frac{\lambda}{2}c_{2,n}(0)(1-e^{-i\Delta t})+O(\lambda^3)\right\},$$

$$c_{2,n}(t)=\left\{c_{2,n}(0)\left[1-i\frac{\lambda^2}{4}[\Delta t+i(e^{i\Delta t}-1)]\right]-\frac{\lambda}{2}c_{1,n+1}(0) \times(e^{i\Delta t}-1)+O(\lambda^3)\right\}. \tag{26}$$

It is easy to see that at second order in the development parameter a secularity appears, which is a term that grows without bound in the limit $t\to\infty$. In perturbation theory, unless we are not able to get rid of the secularity, the series is not very useful. This can be accomplished through the renormalization-group methods described in Ref. [7]. But here, the problem can be easily traced back to the Taylor expansion of the function $\sin(\sqrt{1+\epsilon^2}t)$ in ϵ, having $\sqrt{1+\epsilon^2}=1+\epsilon^2/2+O(\epsilon^4)$. So, we can eliminate it by simply substituting Δ with $\Delta+\mathcal{R}_n^2/2\Delta$ everywhere in the approximate solution into the exponentials of Eq. (26).

It is not difficult to get back the result (26) through the Dyson series (3). So, as expected, this series gives an analysis of the Jaynes-Cummings model when the detuning Δ is larger enough than the Rabi frequency $\mathcal{R}_n$.

Now, let us repeat the above discussion in the opposite limit with the Rabi frequency larger than the detuning. Again, by Taylor expanding the exact solution, one has

$$c_{1,n+1}(t)=\left(c_{1,n+1}(0)\left[\cos\left(\frac{\mathcal{R}_n}{2}t\right)+\frac{i}{\lambda}\sin\left(\frac{\mathcal{R}_n}{2}t\right)-\frac{1}{2\lambda^2}\frac{\mathcal{R}_n}{2}t\sin\left(\frac{\mathcal{R}_n}{2}t\right)\right]-ic_{2,n}(0)\left\{\sin\left(\frac{\mathcal{R}_n}{2}t\right)-\frac{1}{2\lambda^2}\left[\sin\left(\frac{\mathcal{R}_n}{2}t\right)-\frac{\mathcal{R}_n}{2}t\cos\left(\frac{\mathcal{R}_n}{2}t\right)\right]\right\}+O\left(\frac{1}{\lambda^3}\right)\right)e^{-i\Delta t/2}, \tag{27}$$

$$c_{2,n}(t)=\left(c_{2,n}(0)\left[\cos\left(\frac{\mathcal{R}_n}{2}t\right)-\frac{i}{\lambda}\sin\left(\frac{\mathcal{R}_n}{2}t\right)-\frac{1}{2\lambda^2}\frac{\mathcal{R}_n}{2}t\sin\left(\frac{\mathcal{R}_n}{2}t\right)\right]-ic_{1,n+1}(0)\left\{\sin\left(\frac{\mathcal{R}_n}{2}t\right)-\frac{1}{2\lambda^2}\left[\sin\left(\frac{\mathcal{R}_n}{2}t\right)-\frac{\mathcal{R}_n}{2}t\cos\left(\frac{\mathcal{R}_n}{2}t\right)\right]\right\}+O\left(\frac{1}{\lambda^3}\right)\right)e^{i\Delta t/2},$$

with the same problem of a secularity at second order. Indeed, this series can be obtained by the dual Dyson series (5) showing what could seem an unexpected result from the adiabatic approximation, but in agreement with the results of Ref. [9].

To compute the dual Dyson series we need the eigenstates and eigenvalues of $H_{JC}^{(I)}$. It is easily found that for the eigenvalue $g\sqrt{n+1}$ we have the eigenstate

$$|a,n,t\rangle=\frac{1}{\sqrt{2}}(e^{-i\Delta t}|1,n+1\rangle+|2,n\rangle), \tag{28}$$

and for the eigenvalue $-g\sqrt{n+1}$ we have the eigenstate

$$|b,n,t\rangle=\frac{1}{\sqrt{2}}(|1,n+1\rangle-e^{i\Delta t}|2,n\rangle), \tag{29}$$

which are easily recognized as the dressed states of Ref. [8] for the Jaynes-Cummings model with a nonzero detuning. Berry phases are then easily computed to give

$$\dot{\gamma}_a=\left\langle a,n,t\left|i\frac{\partial}{\partial t}\right|a,n,t\right\rangle=\frac{\Delta}{2}, \tag{30}$$

$$\dot{\gamma}_b=\left\langle b,n,t\left|i\frac{\partial}{\partial t}\right|b,n,t\right\rangle=-\frac{\Delta}{2}. \tag{31}$$

Then, after some algebra using the dressed states computed above, the unitary evolution operator (6) is given by

$$\begin{aligned}U_0(t)&=e^{i(\Delta/2)t-ig\sqrt{n+1}t}|a,n,t\rangle\langle a,n,0|+e^{-i(\Delta/2)t+ig\sqrt{n+1}t}|b,n,t\rangle\langle b,n,0|\\&=\cos\left(\frac{\mathcal{R}_n}{2}t\right)(e^{-i(\Delta/2)t}|1,n+1\rangle\langle 1,n+1|+e^{i(\Delta/2)t}|2,n\rangle\langle 2,n|)-i\sin\left(\frac{\mathcal{R}_n}{2}t\right)(e^{-i(\Delta/2)t}|1,n+1\rangle\langle 2,n|+e^{i(\Delta/2)t}|2,n\rangle\langle 1,n+1|)\end{aligned} \tag{32}$$

that, for $|\psi(0)\rangle=c_{1,n+1}(0)|1,n+1\rangle+c_{2,n}(0)|2,n\rangle$, gives

$$|\psi(t)\rangle_I\approx\left[\cos\left(\frac{\mathcal{R}_n}{2}t\right)c_{1,n+1}(0)-i\sin\left(\frac{\mathcal{R}_n}{2}t\right)c_{2,n}(0)\right]\times e^{-i(\Delta/2)t}|1,n+1\rangle+\left[\cos\left(\frac{\mathcal{R}_n}{2}t\right)c_{2,n}(0)-i\sin\left(\frac{\mathcal{R}_n}{2}t\right)c_{1,n+1}(0)\right]\times e^{i(\Delta/2)t}|2,n\rangle \tag{33}$$

that is the exact form of Eqs. (27) when higher-order terms beyond the leading one are neglected, i.e., when $\lambda\to\infty$, as expected from the results of Ref. [9].

In order to go to higher orders, we have to compute $H'(t)$ from Eq. (7). Again, using the above expressions for the dressed states, one gets

$$H'(t)=-\frac{\Delta}{2}[\cos(\mathcal{R}_n t)(|1,n+1\rangle\langle 1,n+1|-|2,n\rangle\langle 2,n|)-i\sin(\mathcal{R}_n t)(|1,n+1\rangle\langle 2,n|-|2,n\rangle\langle 1,n+1|)], \tag{34}$$

so that, the first-order correction to the leading-order evolution operator $U_0(t)$ of Eq. (32) is given by

$$\begin{aligned}U_1(t)&=-iU_0(t)\int_0^t dt_1\,H'(t_1)\\&=i\frac{1}{\lambda}\sin\left(\frac{\mathcal{R}_n}{2}t\right)(e^{-i(\Delta/2)t}|1,n+1\rangle\langle 1,n+1|-e^{i(\Delta/2)t}|2,n\rangle\langle 2,n|)\end{aligned} \tag{35}$$

that gives the first-order correction,

$$|\delta_1\psi(t)\rangle_I = i\frac{1}{\lambda}\sin\left(\frac{\mathcal{R}_n}{2}t\right)(e^{-i(\Delta/2)t}c_{1,n+1}(0)|1,n+1\rangle - e^{i(\Delta/2)t}c_{2,n}(0)|2,n\rangle), \quad (36)$$

again in agreement with the Taylor expansion as given in Eqs. (27), to order $1/\lambda$. So, in the same way we have at the second order,

$$U_2(t) = -U_0(t)\int_0^t dt_1\, H'(t_1)\int_0^{t_1} dt_2\, H'(t_2)$$
$$= i\frac{1}{2\lambda^2}\left\{\left[\sin\left(\frac{\mathcal{R}_n}{2}t\right) - \frac{\mathcal{R}_n}{2}t\cos\left(\frac{\mathcal{R}_n}{2}t\right)\right]\times(e^{i(\Delta/2)t}|2,n\rangle\langle 1,n+1| + e^{-i(\Delta/2)t}|1,n+1\rangle\langle 2,n|) - i\frac{\mathcal{R}_n}{2}t\sin\left(\frac{\mathcal{R}_n}{2}t\right)(e^{-i(\Delta/2)t}|1,n+1\rangle\langle 1,n+1| + e^{i(\Delta/2)t}|2,n\rangle\langle 2,n|)\right\}. \quad (37)$$

Then, one has

$$|\delta_2\psi(t)\rangle_I = i\frac{1}{2\lambda^2}\left\{\left[\sin\left(\frac{\mathcal{R}_n}{2}t\right) - \frac{\mathcal{R}_n}{2}t\cos\left(\frac{\mathcal{R}_n}{2}t\right)\right]\times(e^{i(\Delta/2)t}c_{1,n+1}(0)|2,n\rangle + e^{-i(\Delta/2)t}c_{2,n}(0)|1,n+1\rangle) - i\frac{\mathcal{R}_n}{2}t\sin\left(\frac{\mathcal{R}_n}{2}t\right)(e^{-i(\Delta/2)t}c_{1,n+1}(0)\times|1,n+1\rangle + e^{i(\Delta/2)t}c_{2,n}(0)|2,n\rangle)\right\}. \quad (38)$$

The agreement with the Taylor expansion as given in Eqs. (27), to order $1/\lambda^2$, is complete.

As expected from the results of Ref. [9], the adiabatic approximation and its higher-order corrections turn out to be a nice method for asympotic analysis of the Schrödinger equation, being the dual of the well-known Dyson series and explaining in this way the nice working of the method of dressed states currently used in quantum optics. No slowly varying of the parameters of the Hamiltonian is involved as one could expect for the adiabatic approximation.

IV. PERTURBATIVE ANALYSIS OF MODELS FOR HIGH-ORDER HARMONIC GENERATION

A. Models

Several models are currently used to account for high-order harmonic generation. The first model considered [2] has been a one-dimensional model described by the Schrödinger equation,

$$\left[-\frac{1}{2}\frac{\partial^2}{\partial x^2} + V(x) - x\epsilon_0(t)\sin\omega_L t\right]\Psi(x,t) = i\frac{\partial\Psi(x,t)}{\partial t}, \quad (39)$$

where $\epsilon_0(t)$ is a function taking in account the time to rise the laser field to its maximum value, ω_L is the frequency of the laser field, and $V(x)$ is a simple representative binding potential for the atom. A choice currently found in literature is $V(x) = -1/\sqrt{1+x^2}$. Beside numerical methods that are very computer demanding, other methods as Floquet theory have also been applied [13] for the full three-dimensional case. A fruitful understanding of harmonic generation through semiclassical ideas has also been yielded in Ref. [14]. By these semiclassical results, a nonperturbative quantum model has been obtained [15]. Besides, an approach by second quantization has also been given where a hint was put forward that harmonic generation is a first-order effect [16]. Analytical expressions are barely given as all these models have been solved numerically or nonperturbatively so as to require at some step numerical computation. Another model is a simpler two-level system described by the Hamiltonian [3–5],

$$H = \frac{\omega_0}{2}(|2\rangle\langle 2| - |1\rangle\langle 1|) - x\epsilon_0(t)\begin{Bmatrix}\sin\omega_L t\\ \cos\omega_L t\end{Bmatrix} \quad (40)$$

and

$$x = -d_{12}(|1\rangle\langle 2| + |2\rangle\langle 1|), \quad (41)$$

where d_{12} is the matrix element of the atomic dipole. This model is well-known in quantum mechanics. A first hint to a strong coupling perturbative solution was given by Meystre [6] who used it to describe an atom inside a Fabry-Perot cavity. The series till first order and the way to compute higher orders for strong coupling were finally obtained in Ref. [7] where it was shown that a shift of the levels of the atom occurs.

Indeed, this two-level model seems very effective in describing high-order harmonic generation too. The two physical situations of a Fabry-Perot cavity strongly coupled with an atom and an atom in a strong laser field seems to be described by the same Hamiltonian. But this should not come as a surprise. What really matters here is the existence of the shift of the energy levels of the atom in these situations that, for the case of high-order harmonic generation, can change the spectrum of hyper-Raman lines and so, can be measured experimentally.

Besides, as we are going to show, the leading-order solution found by Meystre is not enough to get the power spectrum computed through the Fourier transform of the equation

$$x(t) = \langle\Psi(t)|x|\Psi(t)\rangle. \quad (42)$$

In fact, by the dual Dyson series one can see that high-order harmonic generation is actually a first-order effect. In this way we are able to reproduce the results obtained in Ref. [5] by the Floquet method, but having an analytical expression to be compared with experiments. As a by-product we have that the hyper-Raman lines can be shifted. Through this approach the computation can be pushed to any order, coping always with definite analytical expressions.

The model (39) can also be treated by this approach. Indeed, an application to multiphoton ionization has been found by Salamin [17]. The leading-order solution should be written as

$$\psi(x,t)\approx e^{ix\int_0^t dt'\epsilon_0(t')\sin\omega_L t'}\phi_n(x), \tag{43}$$

where

$$\left[-\frac{1}{2}\frac{\partial^2}{\partial x^2}+V(x)\right]\phi_n(x)=E_n\phi_n(x). \tag{44}$$

It is easy to see that probability transitions given by $w_{mn}(t)=\int_{-\infty}^{+\infty}dx\,\phi_m(x)\psi(x,t)$ are not trivial and can be computed also for the continuous part of the spectrum. But, as we are going to show using the two-level model and as can be seen by the look of the leading-order solution (43), we need to compute the first-order correction to it to account for high-order harmonic generation. We do not pursue the study of this model further here, as the two-level model can give a satisfactory account of all this matter in a simpler way. We just note that in this way, more complex models than that of Eq. (39), through perturbation methods, could be taken into account.

B. Perturbative analysis for high-order harmonic generation

To fix the ideas, we consider the two-level model of Ref. [5], that is, Eq. (40) with a cosine perturbation. Dyson series using probability amplitudes and its dual solution to first order of this model through operatorial methods were given in Ref. [7]. So, we avoid the analysis by the Dyson series of this model discussed in depth in [7] and references therein. Instead, we use the dual Dyson series to show that it is equivalent to the operatorial method used in [7] and presented initially in Ref. [18].

The rising of the laser field accounted for by the function $\epsilon_0(t)$ is taken as instantaneous to make the computations simpler; that is, we take $\epsilon_0(t)=\Omega=\text{const}$.

In the interaction picture, the Hamiltonian (40) is given by

$$H_I=\Omega d_{12}\cos\omega_L t(e^{-i\omega_0 t}|1\rangle\langle 2|+e^{i\omega_0 t}|2\rangle\langle 1|). \tag{45}$$

Then, computing the dual Dyson series, for the eigenvalue $\Omega d_{12}\cos\omega_L t$ we get the eigenvector

$$|b,t\rangle=\frac{1}{\sqrt{2}}(e^{i\omega_0 t}|2\rangle+|1\rangle), \tag{46}$$

and for the eigenvalue $-\Omega d_{12}\cos\omega_L t$ we get the eigenvector

$$|a,t\rangle=\frac{1}{\sqrt{2}}(|2\rangle-e^{-i\omega_0 t}|1\rangle). \tag{47}$$

These are the dressed states for this model. The corresponding Berry phases are given by

$$\dot{\gamma}_b(t)=\frac{\omega_0}{2},$$
$$\dot{\gamma}_a(t)=-\frac{\omega_0}{2}. \tag{48}$$

It is interesting to note here that Berry phases originate from the energies of the levels of the unperturbed atom.

All this gives the unitary evolution

$$U_0(t)=e^{-i(\omega_0/2)t}e^{i(\Omega d_{12}/\omega_L)\sin\omega_L t}|a,t\rangle\langle a,0| + e^{i(\omega_0/2)t}e^{-i(\Omega d_{12}/\omega_L)\sin\omega_L t}|b,t\rangle\langle b,0| \tag{49}$$

that yields in terms of the bare states $|1\rangle$ and $|2\rangle$,

$$U_0(t)=\cos\left(\frac{\Omega d_{12}}{\omega_L}\sin\omega_L t\right)(e^{-i(\omega_0/2)t}|1\rangle\langle 1|+e^{i(\omega_0/2)t}|2\rangle\langle 2|) - i\sin\left(\frac{\Omega d_{12}}{\omega_L}\sin\omega_L t\right)(e^{-i(\omega_0/2)t}|1\rangle\langle 2| + e^{i(\omega_0/2)t}|2\rangle\langle 1|). \tag{50}$$

We can reformulate the above operator as a matrix by taking for the bare states,

$$|1\rangle=\begin{pmatrix}0\\1\end{pmatrix},\quad |2\rangle=\begin{pmatrix}1\\0\end{pmatrix}, \tag{51}$$

so to have

$$U_0(t)=\begin{pmatrix} e^{i(\omega_0/2)t}\cos\left(\frac{\Omega d_{12}}{\omega_L}\sin\omega_L t\right) & -ie^{i(\omega_0/2)t}\sin\left(\frac{\Omega d_{12}}{\omega_L}\sin\omega_L t\right) \\ -ie^{-i(\omega_0/2)t}\sin\left(\frac{\Omega d_{12}}{\omega_L}\sin\omega_L t\right) & e^{-i(\omega_0/2)t}\cos\left(\frac{\Omega d_{12}}{\omega_L}\sin\omega_L t\right)\end{pmatrix}. \tag{52}$$

It is not difficult to see that the above operator can be rewritten through the Pauli matrices $\sigma_1,\sigma_2,\sigma_3$ as

$$U_0(t)=e^{i(\omega_0/2)\sigma_3 t}e^{-i\sigma_1(\Omega d_{12}/\omega_L)\sin\omega_L t}. \tag{53}$$

Then, by eliminating the prefactor due to interaction picture, we are left with the leading-order result of Ref. [7] for the wave function

$$|\Psi(t)\rangle\approx e^{-i\sigma_1(\Omega d_{12}/\omega_L)\sin\omega_L t}|\Psi(0)\rangle. \tag{54}$$

In the same way, we can compute higher-order corrections to the above by computing $H'(t)$ for the dual Dyson series. In the bare states, using again the dressed ones, one has

$$H'(t)=\frac{\omega_0}{2}\left[\cos\left(2\frac{\Omega d_{12}}{\omega_L}\sin\omega_L t\right)(|2\rangle\langle 2|-|1\rangle\langle 1|) - i\sin\left(2\frac{\Omega d_{12}}{\omega_L}\sin\omega_L t\right)(|2\rangle\langle 1|-|1\rangle\langle 2|)\right]. \tag{55}$$

That is,

$$H'(t)=\frac{\omega_0}{2}e^{i\sigma_1(\Omega d_{12}/\omega_L)\sin\omega_L t}\sigma_3 e^{-i\sigma_1(\Omega d_{12}/\omega_L)\sin\omega_L t}, \tag{56}$$

in agreement with the computation of the first-order correction computed through operatorial methods in Ref. [7]. The two series are identical as should be expected.

So, the solution for the system (40) till first order can be written as [7]

$$|\Psi(t)\rangle=e^{-i\sigma_1(\Omega d_{12}/\omega_L)\sin\omega_L t}\Bigg[I-i\frac{\omega_0}{2}J_0\left(\frac{2\Omega d_{12}}{\omega_L}\right)t\sigma_3 - i\omega_0\sum_{n=1}^{\infty}J_{2n}\left(\frac{2\Omega d_{12}}{\omega_L}\right)\frac{\sin(2n\omega_L t)}{2n\omega_L}\sigma_3 + i\omega_0\sum_{n=0}^{\infty}J_{2n+1}\left(\frac{2\Omega d_{12}}{\omega_L}\right)\frac{\cos[(2n+1)\omega_L t]-1}{(2n+1)\omega_L}\sigma_2 + \cdots\Bigg]|\Psi(0)\rangle, \tag{57}$$

where use has been made of the operatorial identity,

$$e^{\pm i\sigma_k z\sin\phi}=J_0(z)+2\sum_{n=1}^{\infty}J_{2n}(z)\cos(2n\phi) \pm 2i\sigma_k\sum_{n=0}^{\infty}J_{2n+1}(z)\sin[(2n+1)\phi], \tag{58}$$

with σ_k, one of the Pauli matrices and $J_n(z)$, Bessel functions of integer order. The secular term in Eq. (57) can be resummed away by renormalization-group methods, as shown in Ref. [7], giving the renormalized levels of the atom in the laser field. Then, the solution one has to use to compute the power spectrum is

$$|\Psi(t)\rangle=e^{-i\sigma_1(\Omega d_{12}/\omega_L)\sin\omega_L t}\Bigg[I-i\omega_0\sum_{n=1}^{\infty}J_{2n}\left(\frac{2\Omega d_{12}}{\omega_L}\right)\times\frac{\sin(2n\omega_L t)}{2n\omega_L}\sigma_3+i\omega_0\sum_{n=0}^{\infty}J_{2n+1}\left(\frac{2\Omega d_{12}}{\omega_L}\right)\times\frac{\cos[(2n+1)\omega_L t]-1}{(2n+1)\omega_L}\sigma_2+\cdots\Bigg]\times e^{-i(\omega_0/2)J_0(2\Omega d_{12}/\omega_L)t\sigma_3}|\Psi(0)\rangle. \tag{59}$$

It is easy to see that if we just limit our analysis to Eq. (54), we are not able to obtain the spectrum of the harmonics. In fact, one would have from Eq. (42) $x(t)=\langle\Psi(t)|x|\Psi(t)\rangle=-d_{12}\langle\Psi(0)|\sigma_1|\Psi(0)\rangle=\text{const}$. Instead, using Eq. (59) one has at first order,

$$x(t)=-d_{12}\Bigg[c_2c_1^*e^{-i\omega_{0R}t}+c_2^*c_1e^{i\omega_{0R}t}+(|c_1|^2-|c_2|^2)\omega_0\times\sum_{n=0}^{\infty}J_{2n+1}\left(\frac{2\Omega d_{12}}{\omega_L}\right)\frac{\cos[(2n+1)\omega_L t]-1}{\left(n+\frac{1}{2}\right)\omega_L}+i(c_2^*c_1e^{i\omega_{0R}t}-c_2c_1^*e^{-i\omega_{0R}t})\omega_0\times\sum_{n=1}^{\infty}J_{2n}\left(\frac{2\Omega d_{12}}{\omega_L}\right)\frac{\sin(2n\omega_L t)}{n\omega_L}\Bigg], \tag{60}$$

where $\omega_{0R}=\omega_0 J_0(2\Omega d_{12}/\omega_L)$ is the renormalized separation of the two levels of the atom and introducing the population distribution c_1 and c_2 for the bare levels of the atom through the initial state $|\Psi(0)\rangle$. This is exactly the form one must have for the high-order harmonic generation using this model, as shown in Ref. [5] by the Floquet method. In fact, we have odd harmonics of intensity $(|c_1|^2-|c_2|^2)^2$, while the latter term is due to the hyper-Raman lines at $\omega_{0R}\pm 2n\omega_L$ of intensity $|c_2|^2|c_1|^2$. But now, an explicit analytic expression for the power spectrum is given, so that a clear understanding of all the parameters involved into atom and strong laser field interaction is obtained. Particularly, we have an exact expression for the shift of the levels of the atom that now we know how to measure: If we take initially $|c_1|=|c_2|$ we will be just left with hyper-Raman lines into the spectrum. These lines can be shifted by varying the ratio $2\Omega d_{12}/\omega_L$, given by the parameters to be controlled into the experiment. By observing how these lines move we can obtain a measure of the shifts of the atom levels. By the expression of $x(t)$, it is clear that the only lines that can be moved are indeed the hyper-Raman lines.

The advantages above the Floquet method used in Ref. [5] are evident as we have an explicit analytical formula for the spectrum with all the functional dependencies on the parameters entering into the model explicitly expressed.

V. CONCLUSIONS

The way dressed states are currently treated in quantum optics becomes to compute the eigenstates and eigenvalues pertaining to either the full Hamiltonian of a given system or the interaction term. In this paper we have shown how the dual Dyson series can give both an understanding of this approach and a tool to improve it. Indeed, the existence of a dual Dyson series permits, as we have shown, a perturbative study of atom-field interaction in different regions of the parameter space of a given model. The case of the Jaynes-Cummings model, fully exploited in this paper, is an example in this sense. Besides, the dual Dyson series is nothing else than the quantum adiabatic approximation and its higher-order corrections that in this way prove to be a very powerful tool to obtain asymptotic approximations to the Schrödinger equation.

The theory is applied to the analysis of models for high-order harmonic generation giving an explicit expression for the power spectrum. In this way new experiments can be thought where, properly changing the parameters involved, measures of the energy levels of the atom in interaction in a strong laser field could be accomplished. Besides, a control on the amplitudes of the harmonics and the position of the cutoff on the spectrum could be obtained.

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