Role of initial coherence in the generation of harmonics and sidebands from a strongly driven two-level atom

F. I. Gauthey,* C. H. Keitel, P. L. Knight, and A. Maquet*
Optics Section, Blackett Laboratory, Imperial College, London SW7 2BZ, England
(Received 18 November 1994)

We investigate the coherent and incoherent contributions of the scattering spectrum of strongly driven two-level atoms as a function of the initial preparation of the atomic system. The initial "phasing" of the coherent superposition of the excited and ground states is shown to influence strongly the generation of both harmonics and hyper-Raman lines. In particular, we point out conditions under which harmonic generation can be inhibited at the expense of the hyper-Raman lines. Our numerical findings are supported by approximate analytical evaluations in the dressed state picture.

PACS number(s): 42.50.Hz, 42.65.Ky, 42.50.Ar

I. INTRODUCTION

The coherent interaction of a single laser mode with a single atom as represented by a single dipole transition and situated in ordinary vacuum is considered nowadays as a simple and well understood problem. Mollow [1] first predicted that the scattering spectrum of such an atom displays a narrow elastic contribution at the laser frequency and an inelastic triplet structure, in which the linewidths are governed by the atomic relaxation constants and the splitting by the amplitude of the driving field. The underlying physics has been satisfactorily explained by Haroche [2], Haroche and Hartmann [3], and Cohen-Tannoudji and Reynaud [4] and others in the picture of dressed states, i.e., the eigenstates of the Hamiltonian containing the laser field, the atom, and the interaction between both. The area has been described comprehensively in several review articles [5–7].

The situation changed substantially when strong field effects were considered and especially when experimentalists achieved laser field strengths leading to multiphoton absorption and high harmonic generation, a development which has recently been summarized in various review articles [8–10]. A two-level model is an accurate representation of the atomic dynamics provided that all other levels are so weakly coupled that they play a negligible role in the transitions (i.e., that detunings are always very much larger than dipole laser couplings). In very strong fields this is likely not to be valid, and many levels, and continua (leading to ionization), will play an important part in a precise description of the atomic response. Nevertheless, the two-level model remains a useful (because soluble) idealization which has been widely employed to clarify conceptual matters (especially those of the coherences and correlations of central interest here), while recognizing that the two-level model is not a good quantitative model of the atomic response to very intense fields [11–14]. Of course, to describe multiphoton absorption and radiation of high harmonics, it is necessary to abandon the rotating-wave approximation (RWA), although we recognize that, if counterrotating terms are important in electric dipole transitions, then it is likely that other levels beyond the two under consideration are likely to contribute substantially to the atomic dynamics. Sundaram and Milonni [15] pointed out in a more appropriate approach without the RWA that in such strong fields the scattering spectrum of a two-level atom displays a similar succession of harmonic multiples of the laser field frequency as a more realistic model, i.e., an initial exponential reduction in harmonic intensity, then a plateau and a cutoff.

In this context there has also been substantial discussion on the question of whether the scattering spectrum of a strongly driven atom needs in fact to be evaluated from the full two-time correlation function of the dipole [7,16–18]. For resonant light scattering of the Mollow type, both contributions to this complete spectrum, the coherent or mean (one-time) dipole part and the incoherent or quantum fluctuation part, are of significance. Their importance in high harmonic generation has been investigated recently by many authors [13–22]. While these papers investigate in some detail how the various parameters of the laser excitation and the spatial distribution of atoms affect these spectral structures, less attention, it appears, has been paid to the influence of the single atom parameters. In particular, initial coherences of the atom were shown to significantly affect the absorption and dispersion properties of the atom in the low intensity regime; see, e.g., [23].

The object of this paper is to investigate the influence of the initial preparation of the atomic system on the two kinds of contributions of the harmonic spectrum. Particular emphasis is placed on atomic systems which are prepared in a coherent superposition of two atomic levels via a relatively weak laser field prior to the interaction with a superstrong short laser pulse. While the

*Permanent address: Laboratoire de Chimie-Physique-Matière et Rayonnement, Université Pierre et Marie Curie, 11 rue Pierre et Marie Curie, Paris Cedex 05, France.

total spectrum is in fact not very sensitive to the initial condition, we find that the coherent contribution to the harmonic generation can display striking variations as a function of the phase of the initial dipole and even totally disappear at a particular phase. This could facilitate the identification of hyper-Raman lines [21,22] which are usually overwhelmed by intense harmonics. In the analytical calculations in the dressed state picture, we note a total cancellation of interfering dressed transitions in the coherent part apart from a single term that is weighted by a sine in the initial dipole phase.

In Sec. II we derive the emitted light spectrum in the Heisenberg picture and identify its coherent and incoherent contributions. This is followed in Sec. III by an analysis of the expressions for the coherent and incoherent spectra for various initial states, which are the ground state, the excited state, and in particular superpositions of both. We also present the results of a numerical model without the RWA in this section and describe and interpret these results in the following Sec. IV. The analytical model in Sec. V in the dressed state picture and within the RWA is then employed to obtain qualitative insights concerning our numerical findings and the paper ends with our conclusions. Two Appendixes contain mathematical details of couplings and of the RWA.

II. EMITTED LIGHT SPECTRUM

A. General expression

The central concepts of the definition of the spectrum of a driven atomic system have been investigated, among others, by Mollow [1], Cohen-Tannoudji [5], Knight and Milonni [7], Cresser [18], and Feneuille [24]. This problem necessitates a treatment involving the quantum nature of light, and a description of the spectrum from a quantum point of view. The atom is coupled to the multimode quantized light field, which is given in the dipole approximation at the position of the atom by

$$\mathbf{E} = i\sum_{\mathbf{k},\lambda}\left(\frac{2\pi\hbar\omega_k}{V}\right)^{1/2}(a_{\mathbf{k}\lambda} - a^{\dagger}_{\mathbf{k}\lambda})\hat{\epsilon}_{\mathbf{k}\lambda}, \tag{1}$$

where $a_{\mathbf{k}\lambda}$ and $a^{\dagger}_{\mathbf{k}\lambda}$ are the photon annihilation and creation operators, respectively, for a plane wave with wave vector $\mathbf{k}$ and polarization λ. V is the volume of quantization and we have chosen a linear polarization basis, have taken the unit polarization vectors $\hat{\epsilon}_{\mathbf{k}\lambda}$ to be real, and neglected the extension of the atom compared with the wavelength of the laser field.

We will denote the quantized field amplitude for each mode $(\mathbf{k}, \lambda)$ as $\mathbf{E}_{\mathbf{k}\lambda} = (2\pi\hbar\omega_k/V)^{1/2}\hat{\epsilon}_{\mathbf{k}\lambda}$. The Hamiltonian describing the interaction of the radiation field with the single electron of the two-level system is given in the dipole approximation by

$$H = H_0 + H_R - \mathbf{d}\cdot\mathbf{E}. \tag{2}$$

Here H_0 and H_R denote the Hamilton operators for the atom and the field, respectively, and $\mathbf{d}$ is the dipole operator, which we take to be linearly polarized as we will only consider the case of a linearly polarized incident laser field relevant for the situation of harmonic generation.

The dipole operator may be expressed in terms of the atomic transition operators $\sigma_{nm} = |n\rangle\langle m|$:

$$\mathbf{d} = x\hat{\epsilon}_x = \mu(\sigma_{12} + \sigma_{21})\hat{\epsilon}_x, \tag{3}$$

where $\mu = \mu_{12} = \mu_{21}$ is the electric-dipole transition matrix element between the two states and $\hat{\epsilon}_x$ is the unit polarization vector of the dipole, taken to be oriented along the direction of polarization of the laser field. We denote $|1\rangle$ and $|2\rangle$ as the ground and excited states, respectively, so that σ_{12} and σ_{21} are the lowering and raising operators. The Hamiltonian can therefore be written as

$$H = E_1\sigma_{11} + E_2\sigma_{22} + \sum_{\mathbf{k},\lambda}\hbar\omega_k a^{\dagger}_{\mathbf{k}\lambda}a_{\mathbf{k}\lambda} - i\hbar\sum_{\mathbf{k},\lambda}C_{\mathbf{k}\lambda}\mu(\sigma_{12} + \sigma_{21})(a_{\mathbf{k}\lambda} - a^{\dagger}_{\mathbf{k}\lambda}), \tag{4}$$

where $C_{\mathbf{k}\lambda} = \mathbf{E}_{\mathbf{k}\lambda}\cdot\hat{\epsilon}_x/\hbar$. We have omitted the zero point energy of the field, and $(E_2 - E_1)/\hbar = \omega_0$ is the atomic transition frequency between the two levels.

According to the approach advocated by Scully and Lamb [17], and by Glauber [16], the quantum spectrum of emitted light during the time interval $[0, T]$ is defined in the Heisenberg picture as [18]

$$S(\omega, T) = \int_0^T dt'' \int_0^T dt' \langle \mathbf{E}^-(t'')\mathbf{E}^+(t')\rangle \times \exp[-i\omega(t'' - t')], \tag{5}$$

where $\mathbf{E}^+(t) = i\sum_{\mathbf{k},\lambda}\mathbf{E}_{\mathbf{k}\lambda}a_{\mathbf{k}\lambda}(t)$ is the positive frequency part of the field operator (1) and $\mathbf{E}^-(t) = (\mathbf{E}^+)^{\dagger}(t)$. This form of the spectrum is related to the correspondence principle spectrum defined in [13,14] and has the attractive feature of yielding a formal expression equivalent to the classical definition of the spectrum. Considering the case when the laser pulse is applied during the interval $[0, T]$ and when the correlation function of the field tends sufficiently fast to zero before the end of the interval T, the spectrum (5) does not change significantly after this time and may also be written as

$$S(\omega) = \int_{-\infty}^{\infty} dt'' \int_{-\infty}^{\infty} dt' \langle \mathbf{E}^-(t'')\mathbf{E}^+(t')\rangle \times \exp[-i\omega(t'' - t')]. \tag{6}$$

The field correlation function can be obtained by determining the evolution of the light field plus source-atom system as it would occur in the absence of any detector. The evolution equation for the annihilation operator is given in the Heisenberg picture as

$$\dot{a}_{\mathbf{k}\lambda}(t) = -i\omega_k a_{\mathbf{k}\lambda}(t) + C_{\mathbf{k}\lambda}x(t), \tag{7}$$

where $x(t)$ represents the dipole operator in the Heisen-

berg picture: $x(t) = U^\dagger(t)xU(t)$ with $U(t)$ the evolution operator of the system. We would like to remark here that, had we analyzed the case of a rapidly evolving dipole, we would have arrived at a spectrum which is governed by the autocorrelation of the dipole acceleration [10,19,20,25]. In the present situation where we analyze the evolution within a two-level model and explicitly exclude ionization, it is straightforward to relate the acceleration autocorrelation to that of the dipole through a factor ω^4 (Ref. [15]). The solution of Eq. (7) is found to be

$$a_{\mathbf{k}\lambda}(t) = a_{\mathbf{k}\lambda}(0)\exp[-i\omega_k t] + C_{\mathbf{k}\lambda}\int_0^t dt_1 x(t_1)\exp[i\omega_k(t_1-t)]. \quad (8)$$

We see from Eq. (8) that the annihilation operators contain a free evolving term and a source term. When replacing these expressions in the positive and negative frequency parts of the field operator (1) we find

$$\mathbf{E}^+(t) = i\sum_{\mathbf{k},\lambda}\mathbf{E}_{\mathbf{k}\lambda}a_{\mathbf{k}\lambda}(0)\exp(-i\omega_k t) + i\sum_{\mathbf{k},\lambda}\mathbf{E}_{\mathbf{k}\lambda}C_{\mathbf{k}\lambda}\int_0^t dt_1 x(t_1)\exp[i\omega_k(t_1-t)]. \quad (9)$$

Therefore the electric field operator consists of two rather different components and we write it as $\mathbf{E}^\pm(t) = \mathbf{E}_0^\pm(t) + \mathbf{E}_S^\pm(t)$. The first term on the right hand side corresponds to the field present even in the absence of the atom, i.e., the freely evolving field, whereas the second term is the source field or radiation reaction field of the point dipole.

The system is taken to be prepared in some state at $t = 0$, the initial time at which the laser is turned on. An appropriate choice for the initial state of the field is one in which there is no field radiated prior to $t = 0$. Hence, we consider only the laser mode is excited at the initial time.

Let us now determine the correlation function of the field in order to calculate the spectrum as given in (6). By distinguishing between the freely evolving and source parts of each component $\mathbf{E}^+(t)$ or $\mathbf{E}^-(t)$ of the total field, we can write the correlation function as

$$\langle\mathbf{E}^-(t'')\mathbf{E}^+(t')\rangle = \langle\mathbf{E}_0^-(t'')\mathbf{E}_0^+(t')\rangle + \langle\mathbf{E}_S^-(t'')\mathbf{E}_0^+(t')\rangle + \langle\mathbf{E}_0^-(t'')\mathbf{E}_S^+(t')\rangle + \langle\mathbf{E}_S^-(t'')\mathbf{E}_S^+(t')\rangle. \quad (10)$$

This expression shows that the correlation function is made up of three different contributions: first the correlation function of the free field, then the two complex conjugate correlation functions between the free field and the source field, which we can relate to an interference term, and finally the correlation function of the source field.

We now write the field correlation function (10) in terms of the positive and negative frequency parts of the field given in (9):

$$\langle\mathbf{E}^-(t'')\mathbf{E}^+(t')\rangle = \mathbf{E}_{\mathbf{L}\ell}\mathbf{E}_{\mathbf{L}\ell}\langle a_{\mathbf{L}\ell}^\dagger(0)a_{\mathbf{L}\ell}(0)\rangle e^{i\omega_k t''}e^{-i\omega_{k'}t'} + \sum_{\mathbf{k},\lambda}\mathbf{E}_{\mathbf{k}\lambda}\mathbf{E}_{\mathbf{L}\ell}C_{\mathbf{k}\lambda}\int_0^{t''}dt_2\langle x(t_2)e^{-i\omega_k(t_2-t'')}a_{\mathbf{L}\ell}(0)\rangle e^{-i\omega_L t'} + \text{c.c.} + \sum_{\mathbf{k},\lambda,\mathbf{k}',\lambda'}\mathbf{E}_{\mathbf{k}\lambda}\mathbf{E}_{\mathbf{k}'\lambda'}C_{\mathbf{k}\lambda}C_{\mathbf{k}'\lambda'}\left\langle\int_0^{t''}dt_2\int_0^{t'}dt_1 x(t_2)x(t_1)e^{-i\omega_k(t_2-t'')}e^{i\omega_{k'}(t_1-t')}\right\rangle, \quad (11)$$

and we note that the only mode contributing to the freely evolving part of each operator is the one populated at time $t = 0$, that is, the laser mode characterized by a wave vector $\mathbf{L}$ and a polarization ℓ.

Substituting the field correlation function (11) in the expression for the spectrum (6), we find that the total spectrum resulting from the three contributions mentioned above becomes

$$S(\omega) = (\mathbf{E}_{\mathbf{L}\ell})^2[\delta(\omega-\omega_L)]\langle a_{\mathbf{L}\ell}^\dagger(0)a_{\mathbf{L}\ell}(0)\rangle + 2[\delta(\omega-\omega_L)]\,\mathrm{Re}\left[\sum_{\mathbf{k}\lambda}A_{\mathbf{k}\lambda}\int_{-\infty}^{\infty}dt''e^{-i\omega t''}\int_0^{t''}dt_2\langle x(t_2)a_{\mathbf{L}\ell}(0)\rangle e^{-i\omega_k(t_2-t'')}\right] + \sum_{\mathbf{k},\lambda,\mathbf{k}',\lambda'}B_{\mathbf{k}\mathbf{k}'\lambda\lambda'}\int_{-\infty}^{\infty}dt''e^{-i\omega t''}\int_{-\infty}^{\infty}dt'e^{i\omega t'}\int_0^{t''}dt_2\int_0^{t'}dt_1\langle x(t_2)x(t_1)\rangle e^{-i\omega_k(t_2-t'')}e^{i\omega_{k'}(t_1-t')}, \quad (12)$$

where $A_{\mathbf{k}\lambda} = \mathbf{E}_{\mathbf{k}\lambda}\mathbf{E}_{\mathbf{L}\ell}C_{\mathbf{k}\lambda}$ and $B_{\mathbf{k}\mathbf{k}'\lambda\lambda'} = \mathbf{E}_{\mathbf{k}\lambda}\mathbf{E}_{\mathbf{k}'\lambda'}C_{\mathbf{k}\lambda}C_{\mathbf{k}'\lambda'}$. Due to the above choice of the initial state, the first contribution to the spectrum, which is that of the freely evolving field alone, shows a δ function $\delta(\omega-\omega_L)$ and thus only gives a nonvanishing value at the laser frequency. The second contribution, related to the free-field–source interferences, also shows a δ function at the laser frequency for the same reason and hence only accounts for this particular frequency. This has happened because one of the two inputs for the two-time expectation value was simply the coherent driving field and the corresponding integral of the oscillating term $\exp[i(\omega-\omega_L)t]$ generates the δ function $\delta(\omega-\omega_L)$. As we are interested in the multiphoton response of the system, and in the measurement of harmonic generation as well as hyper-Raman processes, we will not include these two terms in our final expression of the spectrum. Hence the spectrum which we consider is given by the spectrum

radiated by the source field:

$$S(\omega) = \sum_{\mathbf{k},\lambda,\mathbf{k}',\lambda'} B_{\mathbf{k}\mathbf{k}'\lambda\lambda'} \int_{-\infty}^{\infty} dt'' e^{-i\omega t''} \int_{-\infty}^{\infty} dt' e^{i\omega t'} \times \int_0^{t''} dt_2 \int_0^{t'} dt_1 \langle x(t_2)x(t_1)\rangle \times e^{-i\omega_k(t_2-t'')} e^{i\omega_{k'}(t_1-t')}. \quad (13)$$

At this point it is reasonable to make the Markov approximation as usual. It is based on recognizing that the vacuum field contains contributions over a very wide range of frequencies, and this range is broad compared with the inverse of the response time of the atomic dipoles. In this way, the sums over the modes can be extended to their continuous limit as $V \longrightarrow \infty$:

$$\frac{1}{V}\sum_{\mathbf{k},\lambda} \longrightarrow \frac{1}{(2\pi)^3}\int d^3k \sum_{\lambda}.$$

The factors $B_{\mathbf{k}\mathbf{k}'\lambda\lambda'}$ appearing in the expression of the spectrum (13) correspond to the densities of modes of the environment which can be assumed to be weakly frequency dependent. The integrals over the wave vectors define in each case a function of t_1 or t_2 which is sharply peaked around t_1 or t_2 equal to t' or t'', respectively, and thus the evolutions of the lowering or raising dipole operators can be taken to be described by their freely evolving form. Hence the integrals over t_1 and t_2 can be replaced by

$$\langle x(t'')x(t')\rangle \int_0^{t''} dt_2 \times \int_0^{t'} dt_1 e^{-i(\omega_k-\omega_0)(t_2-t'')} e^{i(\omega_{k'}-\omega_0)(t_1-t')}.$$

This expression is formally integrated as

$$\langle x(t'')x(t')\rangle \left[\frac{e^{-i(\omega_k-\omega_0)t''}-1}{i(\omega_k-\omega_0)}\right]\left[\frac{e^{i(\omega_{k'}-\omega_0)t'}-1}{i(\omega_{k'}-\omega_0)}\right].$$

After sufficiently long times [6,26] (i.e., t' and t'' much larger than $1/\omega_0$), it becomes

$$\langle x(t'')x(t')\rangle \left[\mathrm{P}\left(\frac{i}{(\omega_k-\omega_0)}\right)+\pi\delta(\omega_k-\omega_0)\right] \times \left[\left(\mathrm{P}\frac{-i}{(\omega_{k'}-\omega_0)}\right)+\pi\delta(\omega_{k'}-\omega_0)\right].$$

P and δ denote the principal part and the δ function as usual. We will not consider the negligible principal part term and the proportionality factor which also includes the sum over the polarizations. This leads us to the scattering spectrum as adopted by many authors [7,14,15,26–32] and in general obtained by use of the correspondence principle [13,14]:

$$S(\omega) = \int_{-\infty}^{\infty} dt'' \int_{-\infty}^{\infty} dt' e^{-i\omega(t''-t')}\langle x(t'')x(t')\rangle. \quad (14)$$

As we are in the Heisenberg picture, the correlation function is calculated with respect to the initial state of the system.

B. Coherent and incoherent spectra

Let us consider a complete and orthonormal set of states $\{|\Psi_m\rangle\}$ containing the initial state $|\Psi(0)\rangle = |\Psi_i\rangle$ of the atomic system. After inserting the closure relation over the states $\sum |\Psi_m\rangle\langle\Psi_m| = 1$ into the dipole correlation function we can write, in the Heisenberg picture,

$$\langle x(t'')x(t')\rangle = \langle\Psi_i|x(t'')|\Psi_i\rangle\langle\Psi_i|x(t')|\Psi_i\rangle + \sum_{m\neq i}\langle\Psi_i|x(t'')|\Psi_m\rangle\langle\Psi_m|x(t')|\Psi_i\rangle. \quad (15)$$

The spectrum (14) can therefore be written as

$$S(\omega) = S_C(\omega) + S_I(\omega), \quad (16)$$

where

$$S_C(\omega) = \left|\int dt e^{-i\omega t}\langle\Psi_i|x(t)|\Psi_i\rangle\right|^2,$$
$$S_I(\omega) = \sum_{m\neq i}\left|\int dt e^{-i\omega t}\langle\Psi_i|x(t)|\Psi_m\rangle\right|^2, \quad (17)$$

which shows a separation into the coherent spectrum $S_C(\omega)$ or spectrum of the mean dipole, and the incoherent spectrum $S_I(\omega)$ due to the quantum dipole fluctuations. Later on in this paper we will refer to the respective time dependent matrix elements $\langle\Psi_i|x(t)|\Psi_i\rangle$ and $\langle\Psi_i|x(t)|\Psi_m\rangle$ $(m \neq i)$ as the "coherent" and "incoherent" amplitudes of the spectrum. Let us remark here that, in all calculations of the correlation function, it is necessary to distinguish between the matrix element of the dipole fluctuations $\langle\Psi_i|x(t)|\Psi_m\rangle$ and its complex conjugate for determining the incoherent amplitude of the spectrum. Indeed, the definition of the Fourier transform in (17) implies that the incoherent amplitude of the spectrum in the case of an initial preparation in a state $|\Psi_i\rangle$ is given by the following matrix element $\langle\Psi_i|x(t)|\Psi_m\rangle$ and not by its complex conjugate. At this point we would like to emphasize that the initial state $|\Psi_i\rangle$ influences the spectrum, as will be the major topic of this paper, though the basis choice $\{|\Psi_m\rangle\}$ does not. However, to separate the spectrum uniquely into a coherent and incoherent contribution we needed to choose an orthonormal set including the initial state $|\Psi_i\rangle$. We next calculate both parts of the spectrum for different cases of initial preparation of the system, that is, in its ground or excited state, or in a coherent superposition of both.

C. Multiatom spectra

For a sample of N atoms the spectrum $S_N(\omega)$ arises from the sum over the contributing dipoles $x_n(t)$ and reads

$$S_N(\omega) = \sum_{n,p=1}^{N} \int_{-\infty}^{\infty} dt'' \int_{-\infty}^{\infty} dt' e^{-i\omega(t''-t')} \langle x_n(t'') x_p(t') \rangle = C_N(\omega) + I_N(\omega). \quad (18)$$

Here we define the coherent part of the spectrum (with respect to *interatom* coherence) C_N and the incoherent part I_N as follows [13]:

$$I_N(\omega) = \sum_{n=1}^{N} \int_{-\infty}^{\infty} dt'' \int_{-\infty}^{\infty} dt' e^{-i\omega(t''-t')} \langle x_n(t'') x_n(t') \rangle \propto N,$$

$$C_N(\omega) = \sum_{n,p=1, n\neq p}^{N} \int_{-\infty}^{\infty} dt'' \times \int_{-\infty}^{\infty} dt' e^{-i\omega(t''-t')} \langle x_n(t'') x_p(t') \rangle \propto N^2. \quad (19)$$

We may now split the above coherent and incoherent parts with respect to the *interatom* coherence into their contributions of the coherent and incoherent parts with respect to *intraatom* coherence [19,20], i.e., into the mean dipole and quantum fluctuation contributions. In the case of the incoherent spectrum we have both N-dependent mean dipole and fluctuation terms:

$$I_N(\omega) = \int_{-\infty}^{\infty} dt'' \int_{-\infty}^{\infty} dt' e^{-i\omega(t''-t')} \times \sum_{n=1}^{N} \left[\langle \Psi_i | x_n(t'') | \Psi_i \rangle \langle \Psi_i | x_n(t') | \Psi_i \rangle + \sum_{m\neq i} \langle \Psi_i | x_n(t'') | \Psi_m \rangle \langle \Psi_m | x_n(t') | \Psi_i \rangle \right], \quad (20)$$

while for the coherent spectrum we obtain, under the assumption that the atoms are not correlated to each other,

$$C_N(\omega) = \int_{-\infty}^{\infty} dt'' \int_{-\infty}^{\infty} dt' e^{-i\omega(t''-t')} \times \sum_{n,p}^{N} \langle \Psi_i | x_n(t'') | \Psi_i \rangle \langle \Psi_i | x_p(t') | \Psi_i \rangle. \quad (21)$$

Thus all the N^2 dependence is contained in the mean dipole term of the atoms which usually overwhelms the N-dependent quantum fluctuation term. In the following we will therefore only discuss single atom features, keeping in mind that the mean dipole contributions detected in the forward direction are N^2 dependent and that the quantum fluctuations are N dependent.

III. EFFECT OF INITIAL PREPARATION

A. Preparation in one of the two levels of the system

In the case of initial preparation in one of the two levels of the system, we can simply apply Eq. (15) and obtain the corresponding dipole correlation functions. In the case of preparation in the ground state $|1\rangle$, we find

$$\langle x(t'')x(t') \rangle = \langle 1|x(t'')|1\rangle\langle 1|x(t')|1\rangle + \langle 1|x(t'')|2\rangle\langle 2|x(t')|1\rangle \quad (22)$$

and, in the case of preparation in the upper state $|2\rangle$, we have

$$\langle x(t'')x(t') \rangle = \langle 2|x(t'')|2\rangle\langle 2|x(t')|2\rangle + \langle 2|x(t'')|1\rangle\langle 1|x(t')|2\rangle. \quad (23)$$

We see that, due to the choice of the closure relation in the $\{|1\rangle, |2\rangle\}$ basis, which is orthonormal and includes in both cases the initial state $|\Psi_i\rangle$, each correlation function separates into a coherent (diagonal) and incoherent (off-diagonal) contribution.

The spectra are calculated according to Sec. II B in the $\{|1\rangle, |2\rangle\}$ basis set, and result in a coherent part given by the dipole mean value, and an incoherent part corresponding to the dipole fluctuations. The numerical treatment and the discussion of the results are presented in Secs. III C and IV, respectively.

B. Preparation in a coherent superposition of states

Let us consider an initial state $|\Psi(0)\rangle$ prepared in such a way that there is maximal initial coherence between the two levels $|1\rangle$ and $|2\rangle$, by the action (say) of a preparation pulse, applied prior to $t = 0$ which is the turn-on time of the main pulse, so that

$$|\Psi(0)\rangle = \frac{1}{\sqrt{2}}(|1\rangle + e^{i\varphi}|2\rangle), \quad (24)$$

where φ is an adjustable phase. In dependence on the initial preparation, we shall see that the choice of the basis is fundamental for understanding the physical meaning of the spectrum, that is, to distinguish between the coherent and incoherent amplitudes as previously defined in Sec. II B.

To that end let us follow Eq. (15) and now consider the orthonormal set of states $\{|\Psi(0)\rangle, |\Psi^{\perp}(0)\rangle\}$, where the state $|\Psi^{\perp}(0)\rangle$ is the orthonormal supplement to the initial state $|\Psi(0)\rangle$ given in Eq. (24):

$$|\Psi^{\perp}(0)\rangle = \frac{1}{\sqrt{2}}(|1\rangle - e^{i\varphi}|2\rangle). \quad (25)$$

$\{|\Psi(0)\rangle, |\Psi^{\perp}(0)\rangle\}$ is complete and orthornormal. According to Sec. II B, the corresponding correlation function can thus be expressed as

$$\langle x(t'')x(t') \rangle = \langle \Psi(0)|x(t'')|\Psi(0)\rangle\langle \Psi(0)|x(t')|\Psi(0)\rangle + \langle \Psi(0)|x(t'')|\Psi^{\perp}(0)\rangle\langle \Psi^{\perp}(0)|x(t')|\Psi(0)\rangle \quad (26)$$

and the scattering spectrum is then given in this basis by

$$S_{12}(\omega) = \left| \int dt e^{-i\omega t} \langle \Psi(0)|x(t)|\Psi(0)\rangle \right|^2 + \left| \int dt e^{-i\omega t} \langle \Psi(0)|x(t)|\Psi^{\perp}(0)\rangle \right|^2, \quad (27)$$

where we recognize the first and second terms, respectively, as the coherent and incoherent parts of the spectrum, which are both expressed in the Heisenberg picture. Using any other basis for the closure relation we would not have obtained this particular separation of the spectrum.

The expressions for the average of the dipole moment and its fluctuations are developed in the former basis $\{|1\rangle, |2\rangle\}$. Using the fact that the trace of the time dependent operator is equal to zero, $\mathrm{Tr}[x(t)] = 0$, we find that the coherent amplitude of the spectrum is given by

$$\langle \Psi(0)|x(t)|\Psi(0)\rangle = \frac{1}{2}\left[e^{i\varphi}\langle 1|x(t)|2\rangle + e^{-i\varphi}\langle 2|x(t)|1\rangle\right] \quad (28)$$

whereas the incoherent amplitude is expressed as

$$\langle \Psi(0)|x(t)|\Psi^{\perp}(0)\rangle = \frac{1}{2}\left[\langle 1|x(t)|1\rangle - \langle 2|x(t)|2\rangle\right] + \frac{1}{2}[-e^{i\varphi}\langle 1|x(t)|2\rangle + e^{-i\varphi}\langle 2|x(t)|1\rangle]. \quad (29)$$

We see that only the incoherent amplitude contains diagonal elements with respect to the $\{|1\rangle, |2\rangle\}$ basis, whereas both coherent and incoherent amplitudes contain off-diagonal elements, the latter being associated with the initial phase φ.

C. Model and numerical calculations

We assume the laser driving field to be in a coherent state so that we can adopt a semiclassical model in which the quantum atomic system interacts with the classical electric field $\mathbf{E} = \mathbf{E}_0 \sin(\omega_L t)$ with ω_L the laser frequency and $\mathbf{E}_0$ its amplitude, oriented along the polarization axis $\hat{\epsilon}_x$. This model is furthermore justified as we only consider the spectrum radiated by the source, which remains unchanged if we replace the quantized driving field by an approximate classical field. The interaction Hamiltonian in the dipole approximation is

$$H_I = -\mathbf{d}\cdot\mathbf{E}(t) = \hbar\Omega_0 \sin(\omega_L t), \quad (30)$$

where $\mathbf{d}$ is the quantum dipole operator defined in Sec. II A. We have introduced in the above expression the Rabi frequency $\Omega_0 = -\mathbf{E}_0 \cdot \mathbf{d}/\hbar = -\mathbf{E}_0 \cdot \mu\hat{\epsilon}_x/\hbar$, and will also denote $\Delta = \omega_0 - \omega_L$ as the detuning between the atomic and the laser frequencies.

The total Hamiltonian is $H = H_0 + H_R + H_I$, and the time evolution of the system is governed by the Schrödinger equation

$$i\hbar\frac{\partial|\Psi\rangle}{\partial t} = H|\Psi\rangle. \quad (31)$$

As we have seen in the previous sections, both coherent and incoherent spectral amplitudes can be developed in the two-level $\{|1\rangle, |2\rangle\}$ basis, whatever the case of initial preparation. Hence we only need to determine the time dependent matrix elements of the dipole in this basis. In the Schrödinger picture, the wave function at time t is expressed as

$$|\Psi(t)\rangle = U(t)|\Psi(0)\rangle = c_1(t)|1\rangle + c_2(t)|2\rangle. \quad (32)$$

If the electron starts in the lower level, we denote the time dependent wave function as $|\Psi(t)\rangle = U(t)|1\rangle = a(t)|1\rangle + b(t)|2\rangle$. If the initial state of the system is the upper level, we write $|\Psi(t)\rangle = U(t)|2\rangle$ and this is equal to $a'(t)|1\rangle + b'(t)|2\rangle$. Hence the evolution operator is expressed in the $\{|1\rangle, |2\rangle\}$ basis as

$$U(t) = \begin{pmatrix} a(t) & a'(t) \\ b(t) & b'(t) \end{pmatrix}. \quad (33)$$

The equations of motion for the time dependent coefficients $c_1(t)$ and $c_2(t)$ are derived from the Schrödinger equation:

$$i\hbar\dot{c}_1(t) = E_1 c_1(t) + \hbar\Omega_0 \sin(\omega_L t)c_2(t),$$
$$i\hbar\dot{c}_2(t) = E_2 c_2(t) + \hbar\Omega_0 \sin(\omega_L t)c_1(t). \quad (34)$$

For convenience we will take $E_1 = -\hbar\omega_0/2$ and $E_2 = +\hbar\omega_0/2$.

Let us remark at this point that strong fields may partly ionize the atom and generate substantial level widths of the order of $\hbar\omega_0$ for laser field amplitudes relevant for harmonic generation of the order of 10^{-2} a.u. [33]. Nevertheless, we omit these widths in our approximate consideration and assume the pulses to be short compared to the atomic relaxation times, i.e., we do not take into account relaxation effects and hence our model associates infinite lifetimes for the ground and upper level.

We will analyze separately the amplitudes corresponding to the coherent and incoherent parts of the spectrum as defined in Sec. II B, for each particular initial preparation of the system. The corresponding time dependent dipole matrix elements are calculated in the Heisenberg picture, with the dipole operator expressed as $x(t) = U^{\dagger}(t)xU(t)$. The notation μ will denote the time independent dipole transition matrix element $\langle 1|x|2\rangle = \langle 2|x|1\rangle$. The coherent and incoherent amplitudes for all cases of different initial preparation can be determined by only knowing the different matrix elements of the time dependent dipole operator in the $\{|1\rangle, |2\rangle\}$ bare state basis.

In the case of ground state preparation, the coherent and incoherent amplitudes are given, respectively, by

$$\langle 1|x(t)|1\rangle = \mu[a^*(t)b(t) + b^*(t)a(t)], \quad (35)$$
$$\langle 1|x(t)|2\rangle = \mu[a^*(t)b'(t) + b^*(t)a'(t)]. \quad (36)$$

In the case of upper state preparation, the amplitudes corresponding to the coherent and incoherent parts of the spectrum are given, respectively, by (37) and (38),

and can be derived from those obtained in the case of ground level preparation. The trace of the time dependent operator being equal to zero, we find that the coherent amplitude is given by

$$\langle 2|x(t)|2\rangle = \mu[a'^*(t)b'(t) + a'(t)b'^*(t)] = -\langle 1|x(t)|1\rangle \tag{37}$$

and due to the Hermiticity of $x(t)$ we find for the incoherent amplitude

$$\langle 2|x(t)|1\rangle = \mu[a'^*(t)b(t) + b'^*(t)a(t)] = [\langle 1|x(t)|2\rangle]^*. \tag{38}$$

In the case of an initial preparation in a coherent superposition of states, the expressions for the coherent and incoherent amplitudes of the spectrum appearing in the correlation function of the dipole (26) can be developed in the $\{|1\rangle, |2\rangle\}$ basis according to (28) and (29). We find that the coherent part is expressed as follows:

$$\langle \Psi(0)|x(t)|\Psi(0)\rangle = \frac{\mu}{2}\left\{e^{i\varphi}\left[a^*(t)b'(t) + b^*(t)a'(t)\right] + \text{c.c.}\right\} \tag{39}$$

and for the incoherent part we find

$$\begin{aligned}\langle \Psi(0)|x(t)|\Psi^{\perp}(0)\rangle &= \mu\left[a^*(t)b(t) + \text{c.c.}\right] \\ &+ \frac{\mu}{2}\left\{e^{-i\varphi}\left[a'^*(t)b(t) + b'^*(t)a(t)\right] - \text{c.c.}\right\}.\end{aligned} \tag{40}$$

It is interesting to remark here that, in the case of initial coherence with an initial phase φ, the coherent amplitude consists of terms which are all φ dependent, whereas the incoherent amplitude contains φ independent as well as φ dependent terms. This helps us already at this stage to gain insights as to the origin of the strong dependence of the coherent amplitude on the initial phase of preparation.

The system of evolution equations (34) is solved numerically without making the RWA approximation with an adaptative time step fourth order Runge-Kutta method [34]. The Fourier transformations are performed using the fast-Fourier-transform algorithm [34] with a time step $dt = T_p/256$, where T_p is the optical period. The laser pulses considered have a duration of 64 cycles and correspond to monochromatic square pulses.

IV. DISCUSSION OF NUMERICAL RESULTS

The main object of this section is to present the structures of coherent and incoherent spectral contributions for a driven two-level system as a function of the initial preparation. We study the cases both of initial preparation in one of the two states and of preparation in a coherent superposition. The atomic frequency in all cases is $\omega_0 = 0.3$ a.u. while the Rabi frequency in comparison is chosen as large as 0.1 a.u. A significant problem of this numerical analysis has been the existence of background noise which arises due to the presence of δ functions in the time evolution of the dipole operator without relaxation. We have addressed this rather technical problem in Appendix A and in further detail in a previous contribution by two of us [35].

A. Preparation in the ground or excited state

We would like to begin our discussion with the conceptual case of initial preparation of the atom in the ground state and then the excited state. We have applied a square laser pulse of 64 cycles, a Rabi frequency Ω_0 of 0.1 a.u., and a driving frequency ω_L equal to 0.145 a.u. which yields a detuning of $\Delta = \omega_0 - \omega_L = 0.155$ a.u. It has been noted that a sudden turn-on (as in a squared pulse) may lead to the generation of unphysical even harmonics for hydrogenic systems [10] which we do not observe for our driven two-level system without relaxation. We assume that the relation between turn-on time and atomic lifetime is of significance for even harmonic generation. In the absence of atomic relaxation, this ratio is independent of the turn-on time and this may be why we find that even harmonic generation does not depend on the turn-on time (see also [15]).

The full spectrum obtained for a ground level preparation is given in Fig. 1(a), and shows harmonics at odd multiples of the laser frequency as well as hyper-Raman peaks positioned as side peaks of each harmonic peak. Hence the spectrum appears as a succession of triplets centered around the odd harmonics of the laser frequency. The triplet structure can be understood due to the transitions among the corresponding dressed states as will be explained in more detail in Sec. V. Let us nevertheless point out now that the position of the sidebands is governed by the Rabi frequency and the detuning, so that the side peaks are positioned at the frequencies $n\omega_L \pm \Omega$, where $\Omega = \sqrt{\Omega_0^2 + \Delta^2}$ denotes the generalized Rabi frequency and n is an odd integer number. For very intense laser fields Ω may happen to be larger than ω_L with the consequence of the overlapping of the triplets. The corresponding coherent and incoherent parts of the spectrum of Fig. 1(a) are given in Figs. 1(b) and 1(c). The comparison to the full spectrum reveals that both coherent and incoherent spectra contribute to the harmonic and hyper-Raman lines. However, for the particular laser excitation considered, the major contribution to both kinds of peak comes from the coherent spectrum.

A related analysis has been made for the case of upper level preparation. In the full spectrum [Fig. 2(a)], the peak corresponding to the Rabi shifted atomic frequency becomes predominant. We find that hyper-Raman lines have much larger amplitudes compared to the case of ground level preparation [Fig. 1(a)], whereas the harmonic peak amplitudes tend to remain unchanged. When we analyze this effect with respect to the coherent and incoherent parts [see Figs. 2(b) and 2(c)] we see that the coherent spectra obtained when taking $|1\rangle$ or $|2\rangle$ as initial states are identical [Figs. 1(b) and 2(b)]. This can simply be related to the vanishing trace of the dipole operator

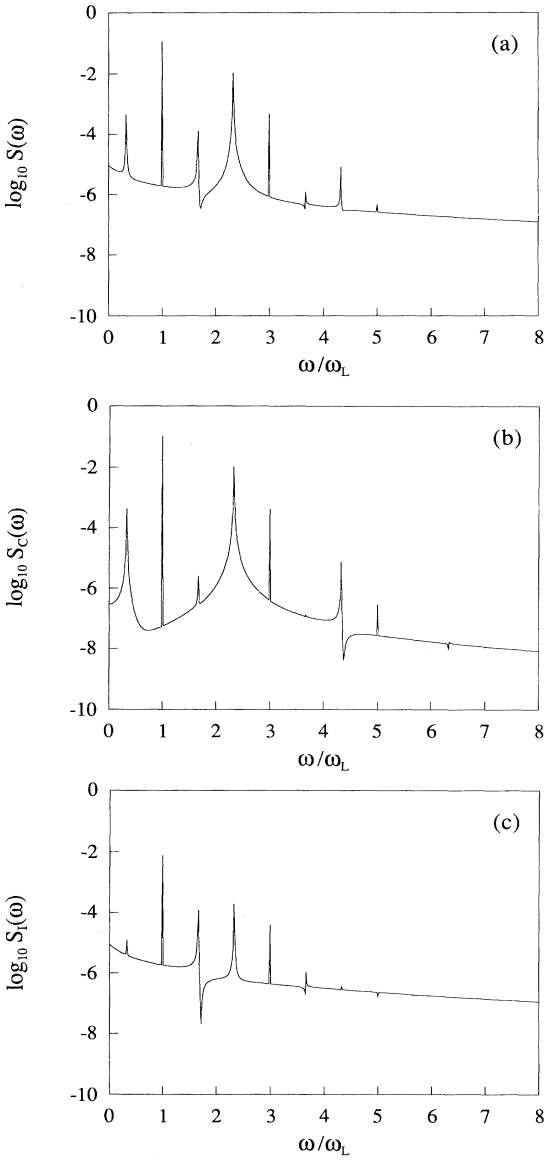

FIG. 1. The logarithm of the full spectrum $S(\omega)$ (a), its coherent part $S_C(\omega)$ (b), and incoherent part $S_I(\omega)$ (c) are shown under conditions of excitation from its ground level with a Rabi frequency of 0.1 a.u. and a laser frequency of 0.145 a.u., which gives a detuning of $\Delta = 0.155$ a.u. The Fourier transforms are computed for pulses that are 64 cycles of the laser frequency in duration, and the frequency unit is the laser frequency. Since $\Omega/\omega_L = 1.27$ the triplets overlap each other. For this reason the hyper-Raman line at $\omega_L - \Omega$ is not visible and we note a peak at $-\omega_L + \Omega$. The hyper-Raman line at $5\omega_L + \Omega$ is only visible in the coherent part of the spectrum.

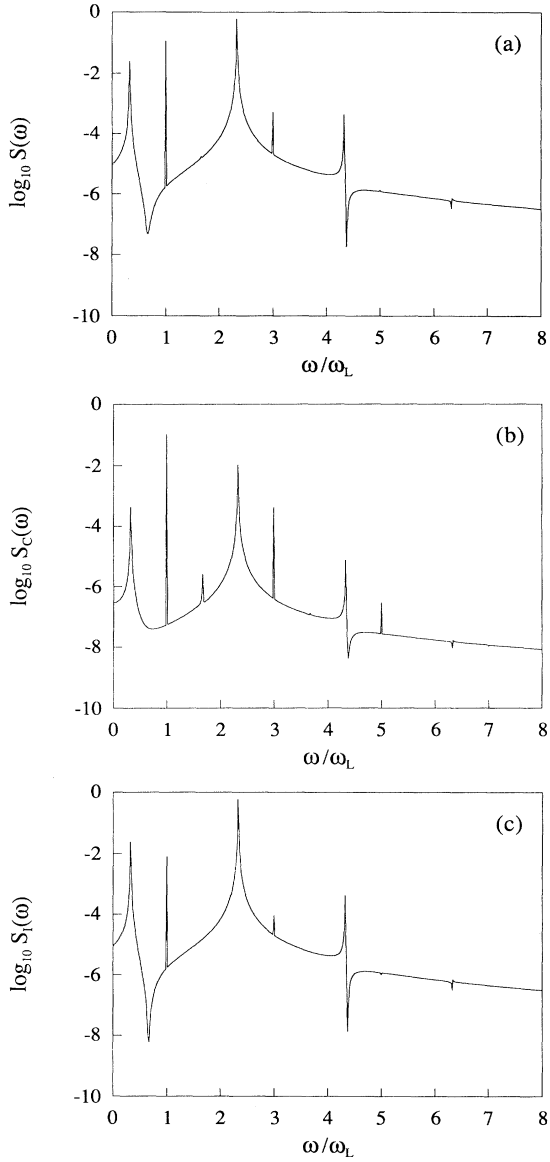

FIG. 2. The logarithm of the full spectrum $S(\omega)$ (a), its coherent part $S_C(\omega)$ (b), and incoherent part $S_I(\omega)$ (c) are shown under the same conditions of excitation of the system as in Fig. 1, but from its upper level.

(37). Changing the initial state from $|1\rangle$ to $|2\rangle$ only has an effect on the incoherent part of the spectrum, where the hyper-Raman peak amplitudes are larger [see Figs. 1(c) and 2(c)].

B. Preparation in a coherent superposition of states

We now turn our interest to the case when the initial state is a superposition of both levels with maximal coherence as given in Sec. III B, and investigate the spectral components as a function of the phase φ. As in the previous case we experience numerical problems due to δ functions arising from the evolution of the dipole without the laser field as discussed in Appendix A (see also [35]).

We here consider the response of the two-level system to the presence of a laser field as a function of the initial phase of preparation φ. We keep the same excitation parameters as in Sec. IV A and first consider the example of the particular phase of initial preparation $\varphi = 0$. The full spectrum in the case of initial coherence is shown on Fig. 3(a). When we compare it to the full spectra obtained in the cases of single level preparations [Figs. 1(a) and 2(a)], we see that it shows peaks positioned at

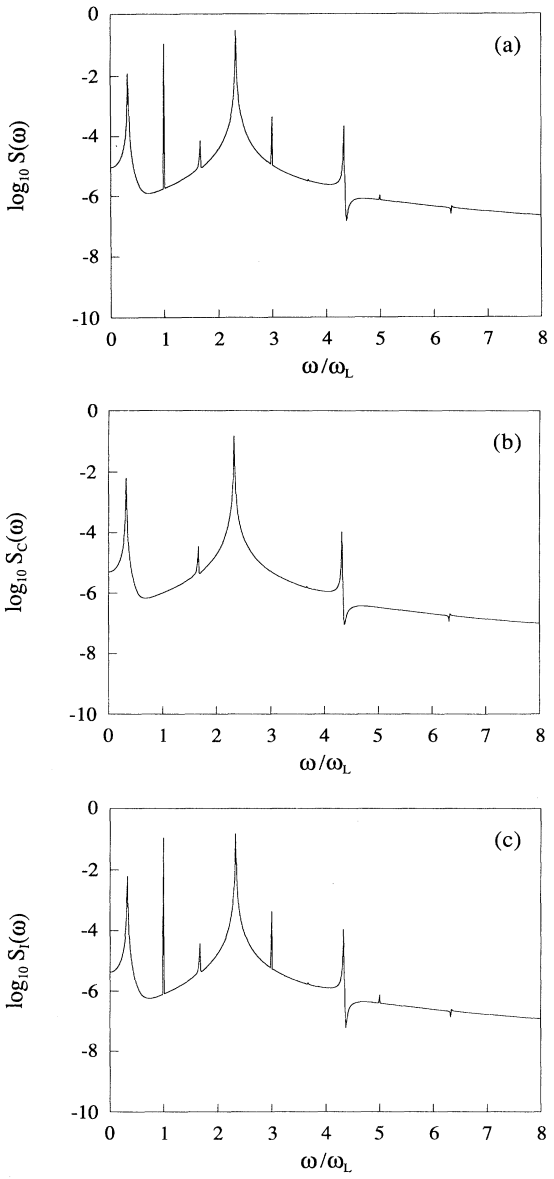

FIG. 3. The logarithm of the full spectrum $S(\omega)$ (a), its coherent part $S_C(\omega)$ (b), and incoherent part $S_I(\omega)$ (c) are shown for the system initially prepared in a coherent superposition of states with a phase $\varphi = 0$, and submitted to a laser pulse excitation of 64 cycles, with a Rabi frequency of 0.1 a.u. and a laser frequency of 0.145 a.u., which gives a detuning $\Delta = 0.155$ a.u.

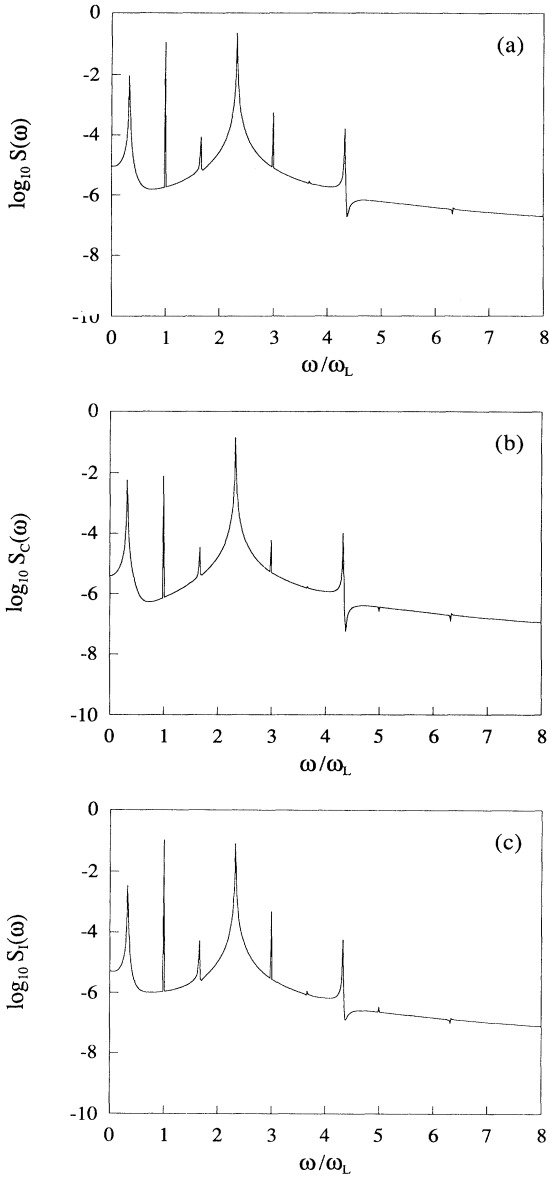

FIG. 4. Same as in Fig. 3, but for the system prepared in a coherent superposition of states with an initial phase $\varphi = \pi/2$.

the same frequencies, and in particular presents the same characteristic shape of a succession of triplets. However, while the harmonic lines seem identical in amplitude, the hyper-Raman peak strengths are different from those obtained in both single level preparation cases, though are closer in strength to those obtained in the case when the system is initially prepared in the upper level $|2\rangle$.

The coherent spectrum [see Fig. 3(b)] shows significant differences when compared to both single state preparation coherent spectra [Figs. 1(b) and 2(b)]. The main difference is that the harmonics disappear from the coherent spectrum. Moreover, the hyper-Raman peaks have much larger amplitudes than in the coherent spectra with no initial coherence. We therefore believe that proper initial preparation may be the key to the observation of hyper-Raman lines. The incoherent spectrum [Fig. 3(c)] shows much more intense harmonics than in the case of single level preparations [see Figs. 1(c) and 2(c)], and hyper-Raman peaks quite similar to those obtained with the upper level preparation. We thus find in this case that the chosen initial phasing has led to a transfer of output energy from the coherent to the incoherent contribution of the spectrum.

We next turn our attention to the effect of a varying phase of initial preparation. Here it appears that the full spectrum is not affected visibly when changing the

phase [see Fig. 4(a) for the case $\varphi = \pi/2$]. We note that the coherent spectra depend upon the initial phase of preparation. The most important feature of this behavior is that both particular initial phases $\varphi = 0$ [see Fig. 3(b)] or $\varphi = \pi$ give rise to the disappearance of all harmonic peaks from the coherent spectra. Any other values of the initial phase [see Fig. 4(b) for the case $\varphi = \pi/2$] give coherent spectra which contain harmonics. Let us remark here that the comparison of coherent spectra obtained for various phases different from the particular cases $\varphi = 0$ or π shows that the differences occur in the amplitudes of the harmonics, which are higher, for example, for $\pi/2$ than for $\pi/4$.

The effect of a varying phase of initial preparation has also been investigated on the incoherent part of the spectrum. Both particular cases $\varphi = 0$ [Fig. 3(c)] or $\varphi = \pi$ give identical incoherent spectra. But taking other values of φ [see Fig. 4(c) for $\varphi = \pi/2$] shows smaller hyper-Raman peaks amplitudes in the incoherent spectra when compared to the cases where $\varphi = 0$ or π. Let us remark here that, apart from the cases where $\varphi = 0$ or π (which correspond to the extreme cases where harmonic generation only comes from the incoherent part of the spectrum), the incoherent spectra obtained for different phases φ give the major contribution to the harmonic lines (as they have much higher amplitudes than those visible in the coherent spectrum).

In Sec. V we will give some analytical insights into the origin of these effects, by determining the dependence of the coherent and incoherent amplitudes of the spectrum upon the initial phase of preparation and in particular we will investigate the disappearance of harmonic lines from the coherent spectra for the specific phases $\varphi = 0$ or π.

The same analysis has been performed for various configurations of the laser pulse excitation. Changing the frequency of the laser or its amplitude showed the same general behavior of the system with respect to its initial phase of preparation φ. It also reproduced the characteristic features expected for each triplet and in particular the shifts of the side peaks with respect to the central peak occurring when varying the generalized Rabi frequency Ω (see Secs. IV A and V and Refs. [36,37]).

We have also investigated the response of the system when submitted to a more realistic short $\sin^2$ pulse corresponding to a laser field $\mathbf{E}(t) = \mathbf{E}_0(t)\sin(\omega_L t)$ with a time dependent amplitude $\mathbf{E}_0(t) = \sin^2(2\pi t/T)$, where T is the duration of the pulse. Our analysis reveals essentially the same behavior of the system when varying its initial phase of preparation φ. Hence, as the response of the system to a varying phase of preparation is not altered by the shape of the laser pulse, and moreover as the best pulse form for the production of stable hyper-Raman lines is a flat top one [22], we restrict the study presented here to the case of a square pulse.

V. DRESSED STATE ANALYTICAL APPROACH WITHIN THE RWA

In Sec. III C we have given the formulations of the coherent and incoherent amplitudes of the spectrum, for different initial preparations, as a function of the time dependent coefficients of the wave function. These coefficients are determined according to the evolution equations (34), which can only be solved exactly numerically. Nevertheless, it is possible to obtain analytical solutions for these equations within the RWA (see Appendix B), and the coherent and incoherent parts of a fluorescence triplet of the spectrum can be directly expressed analytically in the bare state basis. Such an analytical approach can help in order to gain some intuition into the nature of the spectrum, and in particular to understand the disappearance of harmonics in the coherent spectrum for specific phases of initial preparation. Nevertheless, the bare state picture is not sufficient for us to understand the succession of triplets in the spectrum [38,39]. Hence we have chosen to perform analytical calculations in the dressed states basis, which presents the most natural set of states as the eigenbasis of the complete Hamiltonian, and indeed confirms the interpretation of the spectrum as a succession of triplets centered around the odd harmonics of the laser frequency. Considering that the laser is in a coherent state we will consider a semiclassical approach of the dressed picture.

A. The dressed states basis

The semiclassical dressed states (Ref. [40] and Fig. 5), are obtained by diagonalizing the RWA interaction Hamiltonian, expressed in the rotating frame after transformation of the equations of motion [Eq. (B1)] in the case of a sinusoidal laser field $\mathbf{E}(t) = \mathbf{E}_0 \sin(\omega t)$. The wave function in the rotating frame is thus given by $|\Psi_R\rangle = R|\Psi\rangle$, where R is the transformation operator of rotation, and the corresponding Hamiltonian is, with respect to the $\{|1\rangle, |2\rangle\}$ basis,

$$\tilde{H}_{\mathrm{RWA}} = \frac{\hbar}{2}\begin{pmatrix} -\Delta & \Omega_0 e^{-i\pi/2} \\ \Omega_0 e^{i\pi/2} & \Delta \end{pmatrix}, \tag{41}$$

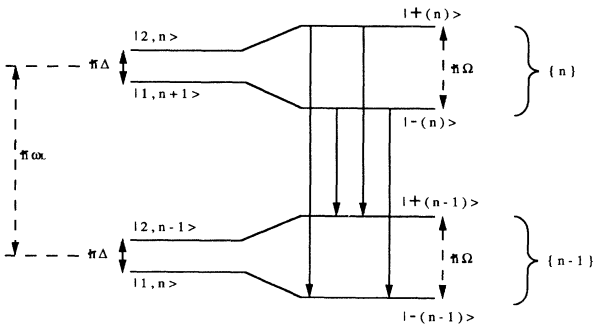

FIG. 5. Unperturbed levels (left part) and dressed states (right part) energy diagram of a two-level system with bare states denoted $|1\rangle$, $|2\rangle$. We show here two particular "multiplicities" of the laser, $\{n\}$ and $\{n-1\}$, where n represents the number of laser photons. $\Delta = \omega_0 - \omega_L$ is the laser detuning on the $|1\rangle \longrightarrow |2\rangle$ transition and is here taken positive. Ω is the generalized Rabi frequency. The single headed arrows represent the allowed transitions between the dressed states of two adjacent multiplicities.

where $\Delta = \omega_0 - \omega_L$ is the detuning and Ω_0 is the resonant Rabi frequency. We would like to stress here the phase factor $e^{i\pi/2}$ which arises from the choice of a sinusoidal structure of the field rather than a cosinusoidal one.

The eigenvectors of this Hamiltonian are known as the semiclassical dressed states, and are found to be

$$|+\rangle = \cos(\theta/2)\exp(-i\pi/4)|1\rangle + \sin(\theta/2)\exp(i\pi/4)|2\rangle,$$
$$|-\rangle = -\sin(\theta/2)\exp(-i\pi/4)|1\rangle + \cos(\theta/2)\exp(i\pi/4)|2\rangle \tag{42}$$

with $\tan(\theta) = -\Omega_0/\Delta$, $\cos(\theta) = -\Delta/\Omega$, and $\sin(\theta) = \Omega_0/\Omega$, where $\Omega = \sqrt{\Omega_0^2 + \Delta^2}$ is the generalized Rabi frequency. The corresponding energies for each state are $E_+ = \hbar\Omega/2$ and $E_- = -\hbar\Omega/2$.

B. Time dependent dipole operator in the dressed states basis

Let us express the time dependent dipole operator $x(t)$ in terms of its positive and negative frequency parts:

$$x(t) = x^+(t) + x^-(t), \tag{43}$$

where $x^+(t) = U^\dagger(t)\sigma_{12}U(t)$, $x^-(t) = U^\dagger(t)\sigma_{21}U(t)$, and σ_{12} and σ_{21} denote the lowering and raising atomic operators, respectively. The expression for the positive frequency part $x^+(t)$ of the dipole in the dressed state basis is

$$\begin{aligned} x^+(t) = {} & \frac{i\mu}{2}\frac{\Omega_0}{\Omega}\exp(-i\omega_L t)\{|+\rangle\langle+| - |-\rangle\langle-|\} \\ & + \frac{i\mu}{2}\left(1 - \frac{\Delta}{\Omega}\right)\exp[-i(\omega_L - \Omega)t]|+\rangle\langle-| \\ & - \frac{i\mu}{2}\left(1 + \frac{\Delta}{\Omega}\right)\exp[-i(\omega_L + \Omega)t]|-\rangle\langle+|, \end{aligned} \tag{44}$$

while the negative frequency part $x^-(t)$ is the corresponding dual operator given by $x^-(t) = [x^+(t)]^\dagger$. We can see from Eq. (44) that the dressed state basis is much more favorable for the purpose of an intuitive understanding than the bare state basis, because every one of the three terms in the previous equations can be associated with one of the peaks of a triplet. As an example the term oscillating as $\cos[(\omega_L + \Omega)t]$ gives rise to a peak in the spectrum at the position $(\omega_L + \Omega)$ which arises due to the time evolution of the coherence between the $|+\rangle$ dressed state of the $\{n\}$ multiplicity (where n represents the number of laser photons) and the $|-\rangle$ dressed state of the $\{n-1\}$ multiplicity (Ref. [40] and Fig. 5). Harmonic generation and fluorescence are interpreted as a radiative cascade between different multiplicities of the dressed state picture. We also see that, as opposed to the sidebands, the central contribution to the spectrum arises from two coherences and we can expect interferences.

C. Coherent and incoherent contributions to the spectrum

In this section we determine the analytical expressions for the coherent and incoherent amplitudes of the spectrum for each case of initial preparation, with regard to the transitions between dressed states and to their corresponding frequencies.

1. Preparation in the ground or excited state

The ground state $|1\rangle$ and the upper state $|2\rangle$ are expressed in the dressed state basis as, respectively,

$$|1\rangle = e^{i\pi/4}\cos(\theta/2)|+\rangle - e^{i\pi/4}\sin(\theta/2)|-\rangle,$$
$$|2\rangle = e^{-i\pi/4}\sin(\theta/2)|+\rangle + e^{-i\pi/4}\cos(\theta/2)|-\rangle. \tag{45}$$

Let us first consider the case of ground state preparation. The coherent amplitude of the spectrum or mean value of the dipole operator is given by

$$\begin{aligned} \langle 1|x(t)|1\rangle = {} & \frac{\cos(\theta)}{2}\{\langle+|x(t)|+\rangle - \langle-|x(t)|-\rangle\} \\ & - \frac{\sin(\theta)}{2}\{\langle+|x(t)|-\rangle + \langle-|x(t)|+\rangle\}. \end{aligned} \tag{46}$$

After expressing the matrix elements of the dipole operator in the dressed state basis as given from Eq. (44) we find that the coherent amplitude in the case of ground state preparation is given by

$$\begin{aligned} \langle 1|x(t)|1\rangle = \mu\frac{\Omega_0}{\Omega}\Bigg\{ & -\frac{\Delta}{\Omega}\sin(\omega_L t) \\ & - \frac{1}{2}\left(1 - \frac{\Delta}{\Omega}\right)\sin[(\omega_L - \Omega)t] \\ & + \frac{1}{2}\left(1 + \frac{\Delta}{\Omega}\right)\sin[(\omega_L + \Omega)t]\Bigg\}. \end{aligned} \tag{47}$$

The Fourier transform of the mean value of the dipole $\langle 1|x(t)|1\rangle$ will give the coherent part of the spectrum. It will obviously have peaks at ω_L, $\omega_L + \Omega$, and $\omega_L - \Omega$. (Here the detuning determines the relative strengths of the peaks.) The transitions at the frequency ω_L correspond to the harmonic peaks and arise from the transitions between the two dressed levels $|+\rangle$ (or the two dressed levels $|-\rangle$) of two adjacent multiplicities $\{n\} \longrightarrow \{n-1\}$; the transitions at $\omega_L + \Omega$ and $\omega_L - \Omega$ correspond to the hyper-Raman peaks observed as sidebands of each triplet. The peak located at $\omega_L + \Omega$ is generated

by the transition between the upper dressed state $|+\rangle$ of a multiplicity $\{n\}$ to the lower dressed state $|-\rangle$ of the multiplicity $\{n-1\}$ while the peak located at $\omega_L - \Omega$ corresponds to the transition between the lower dressed state $|-\rangle$ of a multiplicity $\{n\}$ to the upper dressed state of the multiplicity $\{n-1\}$ (see Fig. 5).

The incoherent amplitude is given by the fluctuations of the dipole between the two bare states. It is expressed in terms of dressed states as

$$\begin{aligned}\langle 1|x(t)|2\rangle = &-i\frac{\sin(\theta)}{2}\{\langle +|x(t)|+\rangle - \langle -|x(t)|-\rangle\} \\ &-i\frac{1+\cos(\theta)}{2}\langle +|x(t)|-\rangle \\ &+i\frac{1-\cos(\theta)}{2}\langle -|x(t)|+\rangle. \end{aligned} \tag{48}$$

When replacing the matrix elements by their expressions, we find

$$\begin{aligned}\langle 1|x(t)|2\rangle = &-i\mu\left(\frac{\Omega_0}{\Omega}\right)^2 \sin[\omega_L t] + \frac{\mu}{2}\left(1-\frac{\Delta}{\Omega}\right)\left\{\cos[(\omega_L-\Omega)t] + i\frac{\Delta}{\Omega}\sin[(\omega_L-\Omega)t]\right\} \\ &+\frac{\mu}{2}\left(1+\frac{\Delta}{\Omega}\right)\left\{\cos[(\omega_L+\Omega)t] - i\frac{\Delta}{\Omega}\sin[(\omega_L+\Omega)t]\right\}. \end{aligned} \tag{49}$$

Again we find the three contributions corresponding to the harmonic line and hyper-Raman peaks of the triplet around the fundamental. We understand that there is a succession of dressed states with field quantum number n and all the high order triplets arise in an approach without the RWA due to transitions among dressed state multiplicities that differ from each other by an odd photon number of the field.

The coherent and incoherent amplitudes of the spectrum in the case of upper level preparation are easily derived from those obtained in the case of ground level preparation as was pointed out in Sec. III C.

2. *Preparation in a coherent superposition*

In the case of initial coherence, the initial state $|\Psi(0)\rangle$ [Eq. (24)] is expressed in the dressed state basis as

$$\begin{aligned}|\Psi(0)\rangle = &\frac{1}{\sqrt{2}}e^{i\pi/4}\left\{\cos(\theta/2) + e^{i(\varphi-\pi/2)}\sin(\theta/2)\right\}|+\rangle \\ &-\frac{1}{\sqrt{2}}e^{i\pi/4}\left\{\sin(\theta/2) - e^{i(\varphi-\pi/2)}\cos(\theta/2)\right\}|-\rangle \end{aligned} \tag{50}$$

and its orthogonal supplement $|\Psi^{\perp}(0)\rangle$ is given by

$$\begin{aligned}|\Psi^{\perp}(0)\rangle = &\frac{1}{\sqrt{2}}e^{i\pi/4}\left\{\cos(\theta/2) - e^{i(\varphi-\pi/2)}\sin(\theta/2)\right\}|+\rangle \\ &-\frac{1}{\sqrt{2}}e^{i\pi/4}\Big\{\sin(\theta/2) \\ &+e^{i(\varphi-\pi/2)}\cos(\theta/2)\Big\}|-\rangle. \end{aligned} \tag{51}$$

The coherent amplitude for the spectrum, which corresponds to the dipole expectation value, is formally given in the Heisenberg picture as

$$\begin{aligned}\langle\Psi(0)|x(t)|\Psi(0)\rangle = &\frac{\sin(\theta)\sin(\varphi)}{2}\{\langle +|x(t)|+\rangle - \langle -|x(t)|-\rangle\} + \frac{1}{2}\{\cos(\theta)\sin(\varphi) - i\cos(\varphi)\}\langle +|x(t)|-\rangle \\ &+\frac{1}{2}\{\cos(\theta)\sin(\varphi) + i\cos(\varphi)\}\langle -|x(t)|+\rangle. \end{aligned} \tag{52}$$

After expressing the matrix elements according to the expression for the dipole operator in the dressed state basis (44), we find that the coherent amplitude is equal to

$$\begin{aligned}\langle\Psi(0)|x(t)|\Psi(0)\rangle = &\mu\left(\frac{\Omega_0}{\Omega}\right)^2 \sin(\omega_L t)\sin(\varphi) + \frac{\mu}{2}\left(1-\frac{\Delta}{\Omega}\right)\left\{\cos[(\omega_L-\Omega)t]\cos(\varphi) - \frac{\Delta}{\Omega}\sin[(\omega_L-\Omega)t]\sin(\varphi)\right\} \\ &+\frac{\mu}{2}\left(1+\frac{\Delta}{\Omega}\right)\left\{\cos[(\omega_L+\Omega)t]\cos(\varphi) + \frac{\Delta}{\Omega}\sin[(\omega_L+\Omega)t]\sin(\varphi)\right\}. \end{aligned} \tag{53}$$

The coherent part of the spectrum, given after Fourier transforming this expression, will have peaks at ω_L and $\omega_L \pm \Omega$. It is of interest to notice here that the contributions to the harmonic lines at the frequency of the laser (coming from the transitions between the same type of dressed states, that is, upper levels or lower levels, of adjacent multiplicities) are totally dependent upon the initial phase of preparation (see also [38,39]). Hence we see that, for the particular phases $\varphi = 0, \pi$ the harmonic contributions cancel out, and this is in agreement with our numerical results (see Sec. IV).

The incoherent amplitude, given by the fluctuations of the dipole between the initial state and the orthogonal state, is expressed in the dressed state basis as

$$\langle\Psi(0)|x(t)|\Psi^{\perp}(0)\rangle = \frac{1}{2}\{\cos(\theta)+i\sin(\theta)\cos(\varphi)\}\{\langle+|x(t)|+\rangle-\langle-|x(t)|-\rangle\}$$
$$-\frac{1}{2}\{\sin(\theta)+\sin(\varphi)-i\cos(\theta)\cos(\varphi)\}\langle+|x(t)|-\rangle$$
$$-\frac{1}{2}\{\sin(\theta)-\sin(\varphi)-i\cos(\theta)\cos(\varphi)\}\langle-|x(t)|+\rangle \quad (54)$$

and it is finally expressed according to (44) as

$$\langle\Psi(0)|x(t)|\Psi^{\perp}(0)\rangle = \mu\left(\frac{\Omega_0}{\Omega}\right)\sin(\omega_L t)\left\{-\frac{\Delta}{\Omega}+i\left(\frac{\Omega_0}{\Omega}\right)\cos(\varphi)\right\}$$
$$+\frac{\mu}{2}\left(1-\frac{\Delta}{\Omega}\right)\sin[(\omega_L-\Omega)t]\left\{-\left(\frac{\Omega_0}{\Omega}\right)-i\frac{\Delta}{\Omega}\cos(\varphi)\right\}$$
$$+\frac{\mu}{2}\left(1-\frac{\Delta}{\Omega}\right)\cos[(\omega_L-\Omega)t]\{-i\sin(\varphi)\}+\frac{\mu}{2}\left(1+\frac{\Delta}{\Omega}\right)\sin[(\omega_L+\Omega)t]\left\{\left(\frac{\Omega_0}{\Omega}\right)+i\frac{\Delta}{\Omega}\cos(\varphi)\right\}$$
$$+\frac{\mu}{2}\left(1+\frac{\Delta}{\Omega}\right)\cos[(\omega_L+\Omega)t]\{-i\sin(\varphi)\}. \quad (55)$$

We see here that the first term of the incoherent amplitude, which gives the incoherent contribution to the harmonic lines, does not depend totally on the initial phase of preparation, as opposed to the coherent contribution to harmonic generation which was shown to depend entirely on the phase in Eq. (53). Out of resonance, the harmonic lines do not vanish from the incoherent spectrum when varying the phase of initial preparation.

VI. SUMMARY AND CONCLUSIONS

In this paper we have investigated the influence of initial preparation on the harmonic spectrum of a strongly driven two-level system. The general structure of a succession of triplets was shown to remain mostly robust though quantitative changes are visible. This occurs in spite of the intuitive understanding that initial conditions within the atom are expected to be "forgotten" rather quickly given the large number of Rabi oscillations in a very strong laser pulse.

The most interesting effects arise in the case of initial "phasing" of the atom, i.e., the preparation into a coherent superposition. Although the total spectrum shows no dramatic influence on the particular phase choice, its coherent and incoherent contributions turn out to display significant dependences.

The coherent part was expected to be dominant in forward scattering because of its N^2 dependence on the atom density N. Just here we found harmonic generation can be transformed from vanishing to existent as a function of the initial phase. This seems significant because this could clearly improve the detection of hyper-Raman lines which are usually dominated by the strong harmonic peaks.

The approximate analytical calculation in the dressed state basis allowed us to understand the succession of triplet structures of one harmonic and two Rabi shifted sidebands due to transitions among dressed states. We have associated the various terms for the time dependent dipole operator in the dressed state basis with the corresponding spectral peaks. Thus it was also clear to see analytically that the harmonics can disappear for a proper phasing of the initial dipole in the coherent contribution of the spectrum. As opposed to the sidebands, the harmonics arise due to transitions between the same dressed states, i.e., in a fully quantum mechanical treatment between those that only differ from each other by the photon number of the field. The corresponding contributions due to the $|+\rangle \longrightarrow |+\rangle$ and $|-\rangle \longrightarrow |-\rangle$ dressed state transitions depend on the initial phasing, and we find that they can cancel each other out in the coherent contribution to the spectrum for a proper choice of the phase and thus lead to the disappearance of harmonics.

ACKNOWLEDGMENTS

The financial support of the European Community and of the U.K. Engineering and Physical Sciences Research Council is gratefully acknowledged. We also would like to thank B. J. Dalton, J. H. Eberly, T. Millack, K. Rzążewski, R. Taïeb, and V. Véniard for valuable discussions.

APPENDIX A

In this appendix we discuss the numerical problems experienced due to δ functions arising in the time evolution of the dipole operator without relaxation. Those can be understood by considering the spectrum without the presence of the field. One can obtain a reduction of background noise in the driven two-level system by subtracting the spectral components due to δ functions arising in the spectrum without laser field.

In the absence of a laser field, the evolution equations for the time dependent amplitudes $c_1(t)$ and $c_2(t)$ reduce to

$$i\hbar\frac{dc_1(t)}{dt} = E_1 c_1(t),$$
$$i\hbar\frac{dc_2(t)}{dt} = E_2 c_2(t), \quad (A1)$$

and they have for solutions:

$$c_1(t) = c_1(0)\exp(-iE_1t/\hbar),$$
$$c_2(t) = c_2(0)\exp(-iE_2t/\hbar). \quad \text{(A2)}$$

Using the notations introduced in Sec. III C, we find that their expression in the case of ground state preparation is given by $a(t) = \exp(-iE_1t/\hbar)$ and $b(t) = 0$ whereas in the case of the excited state preparation they are equal to $a'(t) = 0$ and $b'(t) = \exp(-iE_2t/\hbar)$. The evolution operator is thus given by

$$U(t) = \begin{pmatrix} e^{-iE_1t/h} & 0 \\ 0 & e^{-iE_2t/h} \end{pmatrix}. \quad \text{(A3)}$$

Hence the matrix elements corresponding to the mean dipole are equal to zero:

$$\langle 1|x(t)|1\rangle = \langle 2|x(t)|2\rangle = 0, \quad \text{(A4)}$$

whereas the dipole fluctuation matrix elements are equal to

$$\langle 1|x(t)|2\rangle = [\langle 2|x(t)|1\rangle]^* = \mu\ \exp[-i(E_2 - E_1)t/\hbar]. \quad \text{(A5)}$$

Hence the full spectrum, in the absence of a laser field, reduces to the incoherent spectrum related to the fluctuations of the dipole. In the case of a ground state preparation, the spectrum is then given by

$$S(\omega) = \left|\int dt e^{-i\omega t}\langle 1|x(t)|2\rangle\right|^2 = \left|\int dt e^{-i(\omega+\omega_0)t}\mu\right|^2. \quad \text{(A6)}$$

It is proportional to a δ function $\delta(\omega + \omega_0)$ and will thus give a peak in the unphysical negative frequency range at $\omega = -\omega_0$.

In the case of the initial preparation in the excited level, the spectrum in the absence of the laser field is proportional to the δ function $\delta(\omega - \omega_0)$ and will thus give a peak in the positive frequency range at $\omega = +\omega_0$. Thus, in the absence of a laser field, the spectrum shows an atomic peak at $\omega = -\omega_0$ in the case of initial ground state $|1\rangle$ preparation [see Fig. 6(a)], and at $\omega = +\omega_0$ when the system is initially in the upper level $|2\rangle$ [Fig. 6(b)]. These peaks correspond to the δ functions $\delta(\omega + \omega_0)$ and $\delta(\omega - \omega_0)$ associated with the incoherent spectra in the absence of a laser field, which is the only part contributing to the full spectrum as it is related to the fluctuations of the dipole which have a nonzero value even in the absence of excitation, whereas the mean dipole moment giving the coherent part of the spectrum is equal to zero. Nevertheless, the peaks obtained in our present calculation are broader and show very high wings compared to the δ function shapes expected. This is due to a numerical artifact in the Fourier transform coming from the fact that the time step used for the numerical algorithms employed is adapted to the laser frequency (taken as $\omega_L = 0.145$ a.u.) and thus not to the atomic frequency for the particular values chosen in this example. The problem of the background and of its reduction has already been discussed in previous work on the ground level preparation [35].

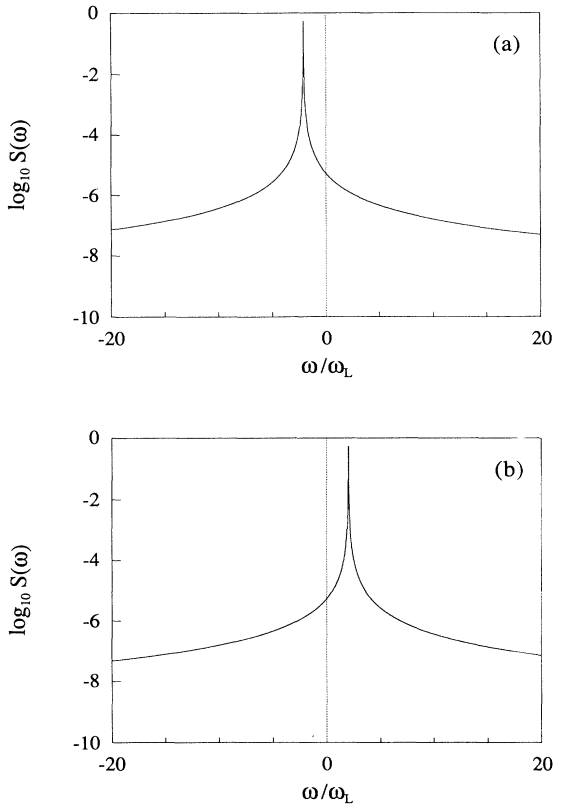

FIG. 6. The logarithm of the full spectrum $S(\omega)$ in the absence of a laser field for the system initially in its ground state (a) and excited state (b). The frequency unit has been chosen as $\omega_L = 0.145$ a.u. and thus allows a comparison with the results with laser excitation.

The corresponding problem arises also for the case of an initial preparation of the atom in a coherent superposition of the laser field. We have considered separately the coherent and incoherent parts of the corresponding spectrum in the positive frequency range. It appears that, even in the absence of laser excitation, both coherent and incoherent spectra contribute visibly to the atomic peak in the full spectrum if the system is initially prepared in a coherent superposition of levels, whereas in the case of single level preparation the incoherent spectrum was the only one contributing to this component. This particular effect is due to initial coherence, as in this case both coherent and incoherent parts of the correlation function contain dipole fluctuation terms between the ground and upper level [see Eqs. (28) and (29)], which give a spectrum even in the absence of an excitation, while for a preparation in a single state, and thus no initial coherence, the fluctuations of the dipole only appear in the incoherent term of the correlation function [see Eq. (22)]. The same analysis has been made, still in the absence of a laser field, for various phases φ and showed no particular

dependence of the full spectra or of both its contributions upon the initial phase.

APPENDIX B

The system of equations of motion (34) for the time dependent coefficients $c_1(t)$ and $c_2(t)$ governing the evolution of the wave function is expressed in the interaction representation as

$$\begin{aligned} i\hbar\frac{d\tilde{c}_1(t)}{dt} &= \tilde{c}_2(t)\frac{\hbar\Omega_0}{2i}\{\exp[i(\omega_L-\omega_0)t] \\ &\quad -\exp[-i(\omega_L+\omega_0)t]\}, \\ i\hbar\frac{d\tilde{c}_2(t)}{dt} &= \tilde{c}_1(t)\frac{\hbar\Omega_0}{2i}\{\exp[i(\omega_L+\omega_0)t] \\ &\quad -\exp[-i(\omega_L-\omega_0)t]\}, \end{aligned} \tag{B1}$$

where $\tilde{c}_1(t) = c_1(t)\exp(-i\omega_0 t/2)$ and $\tilde{c}_2(t) = c_2(t)\exp(i\omega_0 t/2)$.

If we assume that the interaction matrix elements are much smaller than the energies of the unperturbed Hamiltonian, the amplitudes can be replaced by an average over the time τ such that $(1/\omega_L) \ll \tau \ll (1/|V|)$, where $1/\omega_L$ is the optical period and $1/|V|$ the evolution time. Hence, when close to resonance, and for rather small intensities of the laser field, it is possible to make the RWA by neglecting the fast oscillating terms $\exp[i(\omega_L+\omega_0)t]$ and Eqs. (B1) reduce to the standard textbook Rabi equations

$$\begin{aligned} i\hbar\frac{d\tilde{c}_1(t)}{dt} &= \tilde{c}_2(t)\frac{\hbar\Omega_0}{2i}\exp[i(\omega_L-\omega_0)t], \\ i\hbar\frac{d\tilde{c}_2(t)}{dt} &= -\tilde{c}_1(t)\frac{\hbar\Omega_0}{2i}\exp[-i(\omega_L-\omega_0)t]. \end{aligned} \tag{B2}$$

Using the same notation as in Sec. V, we find that, in the Schrödinger picture, the solution of this system for the initial state being the lower level $|1\rangle$ is given by

$$\begin{aligned} a(t) &= \frac{-\Delta+\Omega}{2\Omega}\exp[-i(\Delta+\Omega)t/2]\exp(i\omega_0 t/2) \\ &\quad +\frac{\Delta+\Omega}{2\Omega}\exp[-i(\Delta-\Omega)t/2]\exp(i\omega_0 t/2), \end{aligned} \tag{B3}$$

$$\begin{aligned} b(t) &= i\frac{\Omega_0}{2\Omega}\exp[-i(-\Delta+\Omega)t/2]\exp(-i\omega_0 t/2) \\ &\quad -i\frac{\Omega_0}{2\Omega}\exp[i(\Delta+\Omega)t/2]\exp(-i\omega_0 t/2), \end{aligned} \tag{B4}$$

and for the case when the initial state is the upper level $|2\rangle$

$$\begin{aligned} a'(t) &= -i\frac{\Omega_0}{2\Omega}\exp[-i(\Delta+\Omega)t/2]\exp(i\omega_0 t/2) \\ &\quad +i\frac{\Omega_0}{2\Omega}\exp[-i(\Delta-\Omega)t/2]\exp(i\omega_0 t/2) \end{aligned} \tag{B5}$$

$$\begin{aligned} b'(t) &= \frac{\Delta+\Omega}{2\Omega}\exp[-i(-\Delta+\Omega)t/2]\exp(-i\omega_0 t/2) \\ &\quad +\frac{-\Delta+\Omega}{2\Omega}\exp[i(\Delta+\Omega)t/2]\exp(-i\omega_0 t/2). \end{aligned} \tag{B6}$$

[1] B. R. Mollow, Phys. Rev. **188**, 1969 (1969).
[2] S. Haroche, Ann. Phys. (Paris) **6**, 189 (1971); **6**, 327 (1971).
[3] S. Haroche and F. Hartmann, Phys. Rev. A **6**, 1280 (1972).
[4] C. Cohen-Tannoudji and S. Reynaud, J. Phys. B **10**, 345 (1977); **10**, 365 (1977); **10**, 2311 (1977).
[5] C. Cohen-Tannoudji, in *Atoms in Strong Resonant Fields*, Proceedings of the Les Houches Summer School of Theoretical Physics "Frontiers of Laser Spectroscopy," Les Houches, 1975, edited by R. Balian, S. Haroche, and S. Liberman (North-Holland, Amsterdam, 1977).
[6] L. Allen and J. H. Eberly, *Optical Resonance and Two-Level Atoms* (Wiley-Interscience, New York, 1975); see in particular Chaps. 2, 3, and 7.
[7] P. L. Knight and P. W. Milonni, Phys. Rep. **66**, 21 (1980).
[8] A. L'Huillier, L. A. Lompré, G. Mainfray, and C. Manus, in *Atoms in Intense Laser Fields*, edited by M. Gavrila (Academic Press, Boston, 1992).
[9] The whole field of superintense field-atom interaction is reviewed in *Atoms in Intense Laser Fields* [8].
[10] K. Burnett, V. C. Reed, and P. L. Knight, J. Phys. B **26**, 561 (1993).
[11] K. Rzążewski and J. H. Eberly, J. Mod. Opt. **39**, 795 (1992).
[12] L. Plaja and L. Roso, J. Mod. Opt. **40**, 793 (1993).
[13] J. H. Eberly and M. V. Fedorov, Phys. Rev. A **45**, 4706 (1992).
[14] D. G. Lappas, M. V. Fedorov, and J. H. Eberly, Phys. Rev. A **47**, 1327 (1993).
[15] B. Sundaram and P. W. Milonni, Phys. Rev. A **41**, 6571 (1990).
[16] R. J. Glauber, Phys. Rev. **130**, 2529 (1963).
[17] M. O. Scully and W. E. Lamb, Jr., Phys. Rev. **166**, 246 (1968).
[18] J. D. Cresser, Phys. Rep. **94**, 47 (1983).
[19] K. J. LaGattuta, J. Mod. Opt. **39**, 1181 (1992).
[20] K. J. LaGattuta, Phys. Rev. A **48**, 666 (1993).
[21] B. W. Shore, *The Theory of Coherent Atomic Excitation* (Wiley-Interscience, New York, 1990), Vol. 1, Chap. 10.4.
[22] T. Millack and A. Maquet, J. Mod. Opt. **40**, 2161 (1993), and references therein.
[23] M. O. Scully, Phys. Rev. Lett. **67**, 1855 (1991).
[24] S. Feneuille, Rep. Prog. Phys. **40**, 1257 (1977).
[25] K. Burnett, V. C. Reed, J. Cooper, and P. L. Knight, Phys. Rev. A **45**, 3347 (1992).
[26] M. Sargent III, M. O. Scully, and W. E. Lamb, Jr., *Laser Physics* (Addison-Wesley, Reading, MA, 1974); see in particular pp. 236–239.
[27] K. Wódkiewicz and J. H. Eberly, Ann. Phys. (N.Y.) **101**, 574 (1976).

[28] E. Courtens and A. Szöke, Phys. Rev. A **15**, 1588 (1977).
[29] J. H. Eberly and K. Wódkiewicz, J. Opt. Soc. Am. **67**, 1252 (1977).
[30] M. A. Newbold and G. J. Salamo, Phys. Rev. A **22**, 2098 (1980).
[31] M. Florjańczyk, K. Rzążewski, and J. Zakrzewski, Phys. Rev. A **31**, 1558 (1985).
[32] M. Wilkens and K. Rzążewski, Phys. Rev. A **40**, 3164 (1989).
[33] E. Fiordilino and V. Miceli, J. Mod. Opt. **41**, 1415 (1994).
[34] W. H. Press, B. P. Flannery, S. A. Teukolsky, and W. T. Vetterling, *Numerical Recipes* (Cambridge University Press, Cambridge, England, 1986).
[35] F. I. Gauthey, A. Maquet, R. Taïeb, and V. Véniard (unpublished).
[36] M. S. Kumar, M. Ll. Pons, and J. H. Eberly, Phys. Rev. A **44**, 1995 (1991).
[37] J. H. Eberly and V. D. Popov, Phys. Rev. A **37**, 2012 (1988).
[38] Y. S. Bai, A. G. Yodh, and T. W. Mossberg, Phys. Rev. Lett. **55**, 1277 (1985).
[39] J. E. Golub, Y. S. Bai, and T. W. Mossberg, Phys. Rev. A **37**, 119 (1988).
[40] C. Cohen-Tannoudji, J. Dupont-Roc, and G. Grynberg, *Processus d'interaction entre photons et atomes* (InterEditions/Editions du CNRS, Paris, 1988).