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Pressure-induced effects in two-level atoms: New approach and simple physical interpretation

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Pressure-induced effects in nonlinear optics and spectroscopy are studied using a nonstandard-dressed-atom formalism for a set of two-level atoms. We consider a two-level atom interacting with two radiation fields whose frequencies differ by δ and undergoing dephasing collisions with a perturber bath. The atom-field detunings are large compared with all relaxation rates in the problem. The system is described by a Hamiltonian of an atom dressed by a single-mode field with a coupling constant slowly varying in time at frequency δ. We show that the collisionally aided excitation of the initially unpopulated dressed states is modulated in time. All the characteristics of the pressure-induced effects are deduced from the properties of this time-modulated excitation. Apart from already known phenomena (pressure-induced extra resonances in four-wave mixing, pressure-induced phase shifts, etc.) that are reinterpreted using this approach, we present a new effect that can be described as fluorescence beats. Finally, we show that this approach gives a firm basis for use of a model involving collision-induced gratings and we interpret the pressure-induced effects in the context of this model.

Coherent effects induced by collisional damping are among the most puzzling effects observed in nonlinear optics and spectroscopy. These effects have been first considered by Bloembergen and his co-workers[1] in the framework of the theory of nonlinear susceptibilities. The predictions of this approach have been successfully verified in the case of the PIER 4 resonances (pressure-induced extra resonances in four-wave mixing) both in the case of three-level atoms[2] and in the case of two-level atoms.[3] Several other phenomena have been predicted using this approach (or, equivalently, using the optical Bloch equations) and subsequently observed. Among these phenomena are collision-induced gain in two-wave mixing,[4] collision-aided self-focusing and self-defocusing,[5] and collision-induced phase shift.[6] However, in spite of its success, it has proven difficult to give a completely satisfactory physical picture of the pressure-induced resonances using the optical Bloch equations alone.

Other approaches or pictures have been used to clarify the underlying physics. For example, the connection with optical pumping has been stressed and has permitted one to give simple physical interpretations of several experimental results.[7] This approach is especially useful in the case where the Zeeman degeneracy of the levels has to be considered.[7,8] A dressed-atom method has also proven to be very fruitful. More precisely, the dressed-atom approach has permitted one to show how a PIER 4 coherence can be created by collisions in a three-level atom.[9] Also the manifestation of the conservation of energy[10] and the link with other processes such as collision redistribution of radiation[11] or collision-aided excitation of atomic levels[12] is particularly clear in this model.

In the case of a two-level atom having upper level b and lower level a, several difficulties arise in treating the problem of its interaction with two quantized fields. In particular, the notion of multiplets of dressed states[13] is generally lost because a system in the atomic state b in presence of n_1 photons of the first mode of the field and n_2 photons of the second mode (state $|b,n_1,n_2\rangle$) is coupled to $|a,n_1+1,n_2\rangle$ and $|a,n_1,n_2+1\rangle$, which are coupled to $|b,n_1+1,n_2-1\rangle$ and $|b,n_1-1,n_2+1\rangle$, etc. One can try to avoid this difficulty by quantizing only one field and treating the second field as a classical light source. However, in this situation one usually makes the secular approximation to decouple the evolution of the population of the dressed states from their coherences.[13] Unfortunately, this approximation leads to incorrect results when one considers two fields whose frequency difference is of the order of the natural width of the excited state.[14] In particular, the pressure-induced effects tend to vanish in this approximation. There are methods to overcome this difficulty. One can avoid the secular approximation or use complex dressed states.[10,15] However, none of these solutions is really simple.

We propose here a different approach. The theory is applicable under the experimental conditions where the pressure-induced effects are observed. Generally, the experiments are done in a range of frequencies where the frequency difference between the two electromagnetic fields is much smaller than the frequency detuning from resonance. In this limit, one can describe the electromagnetic field as a field oscillating at the mean frequency of the two beams and slowly modulated in time. If this average field is quantized, the coupling constant between the two-level atom and the field is slowly modulated in time. In the framework of the usual dressed-atom model,[13] the collision-aided excitation of the "upper dressed state" is accompanied by a temporal modulation of the population of this state. All the characteristics of the pressure-induced effects can then be easily interpreted using methods that have already been developed to describe the interaction of an atomic system with a modulated excitation beam.[16] The main advantage of this approach is that the very simple mathematics makes the physics extremely clear.

This paper is organized as follows. In the first part (Sec. I), we recall the classical theory of the pressure-induced effects using the optical Bloch equations. We separate the pressure-induced terms from the collision-free terms and we study the general characteristics of each of these contributions. In the second part (Sec. II), we present a semiclassical dressed-atom model and calculate the dressed-state populations and the electric dipole moment with this model. We show that the separation between the pressure-induced and the collision-free terms has a precise physical meaning in this approach. All the pressure-induced terms are associated with a collisionally aided excitation of a dressed-atom level, while the collision-free terms are associated with the modification of the dressed-state wave functions. Apart from the characteristics of the already well-known pressure-induced effects, such as PIER 4 resonances, two-wave mixing gain, etc., which are easily recovered and explained, we calculate a collision-induced resonance in the fluorescence beats emitted by a two-level atom interacting with two fields. We also show that a rigorous basis can be given to the interpretation of the pressure-induced effects in nonlinear optics in terms of collision-induced gratings.[17]

I. PRESSURE-INDUCED EFFECTS IN THE OPTICAL BLOCH EQUATION APPROACH

A. Notations and assumptions

We consider a set of two-level atoms (ground state a, excited state b, energy difference $E_b - E_a = \hbar\omega_0$) interacting with two nonresonant electric fields of same polarization:[18]

$$E_1(t) = \mathcal{E}_1 \cos(\omega_1 t + \varphi_1) , \tag{1a}$$

$$E_2(t) = \mathcal{E}_2 \cos(\omega_2 t + \varphi_2) . \tag{1b}$$

We denote by δ the difference between the frequencies of the two fields

$$\delta = \omega_1 - \omega_2 , \tag{2}$$

and always assume that

$$|\delta| \ll |\omega_1 - \omega_0| . \tag{3}$$

The frequency detuning from resonance Δ is thus almost the same for the two fields,

$$\Delta = \omega_1 - \omega_0 \simeq \omega_2 - \omega_0 . \tag{4}$$

The matrix element of the electric dipole moment between states a and b is called d (which is assumed to be real). We consider here the case where the fields are sufficiently weak, such that

$$|d| \mathcal{E}_i \ll \hbar |\Delta| \tag{5}$$

for $i = 1, 2$.

Relaxation of the upper level b results only from spontaneous emission and occurs at rate Γ. Relaxation of the a-b coherence results from spontaneous emission (with rate $\Gamma/2$) and from dephasing collisions between the active atoms and a buffer gas.[19] In the impact limit, the coherence relaxation rate is $(\Gamma/2) + \gamma$ where $\gamma = \gamma' + i\gamma''$ is a (complex) collisional rate parameter (γ varies linearly with the buffer gas pressure p).

B. Calculation of the atomic density matrix to second order in the input fields

In this section, we wish to calculate the upper-state population ρ_{bb} to second order in the field amplitudes.

The calculation is carried out in terms of atomic density matrix elements ρ_{ij} which evolve as

$$\frac{d}{dt}\rho_{ij} = \frac{1}{i\hbar}[H, \rho]_{ij} + \left[\frac{d}{dt}\rho_{ij}\right]_{\text{relax}} , \tag{6}$$

where H is the sum of the free Hamiltonian H_0 and of the electric dipole interaction V between the atom and the input fields E_1 and E_2. The relaxation terms in the optical Bloch equations (6) are

$$\left[\frac{d}{dt}\rho_{bb}\right]_{\text{relax}} = -\Gamma\rho_{bb} , \tag{7a}$$

$$\left[\frac{d}{dt}\rho_{aa}\right]_{\text{relax}} = \Gamma\rho_{bb} , \tag{7b}$$

$$\left[\frac{d}{dt}\rho_{ba}\right]_{\text{relax}} = -\left[\frac{\Gamma}{2} + \gamma\right]\rho_{ba} . \tag{7c}$$

To second order in the input fields, the solution of (6) in the limit $\Gamma, |\gamma| \ll |\Delta|$ is

$$\rho_{bb} = \rho_{bb}^{(\text{fr})} + \rho_{bb}^{(\text{coll})} , \tag{8a}$$

with

$$\rho_{bb}^{(\text{fr})} = \frac{d^2}{4\hbar^2\Delta^2}[\mathcal{E}_1^2 + \mathcal{E}_2^2 + 2\mathcal{E}_1\mathcal{E}_2\cos(\delta t + \varphi)] , \tag{8b}$$

$$\rho_{bb}^{(\text{coll})} = \frac{\gamma'}{\Gamma}\left[\frac{d^2(\mathcal{E}_1^2 + \mathcal{E}_2^2)}{2\hbar^2\Delta^2} + \frac{d^2\mathcal{E}_1\mathcal{E}_2}{\hbar^2\Delta^2}\left[\frac{\Gamma^2}{\Gamma^2 + \delta^2}\cos(\delta t + \varphi) + \frac{\Gamma\delta}{\Gamma^2 + \delta^2}\sin(\delta t + \varphi)\right]\right] , \tag{8c}$$

where $\delta=\omega_1-\omega_2$ and $\varphi=\varphi_1-\varphi_2$. We have separated in the expression for ρ_{bb} the collision-free term given in (8b) from the part proportional to γ' which is collision dependent. This term (8c) gives the pressure-induced contribution to the population of the upper state. We see that a resonance centered at $\delta=0$ can be observed in the pressure-induced term. We discuss this resonance below.

C. The electric dipole moment to third order in the input fields

We now consider the coherence ρ_{ba} and more precisely the mean value of the electric dipole moment

$$\langle d\rangle=d(\rho_{ab}+\rho_{ba}) . \tag{9}$$

To third order in the input fields, we find

$$\langle d\rangle=\langle d\rangle^{(\mathrm{lin})}+\langle d\rangle^{(\mathrm{fr})}+\langle d\rangle^{(\mathrm{coll})} , \tag{10}$$

where $\langle d\rangle^{(\mathrm{lin})}$ is associated with the linear response of the medium given by

$$\begin{aligned}\langle d\rangle^{(\mathrm{lin})}=&-\frac{d^2}{\hbar\Delta}[\mathcal{E}_1\cos(\omega_1t+\varphi_1)+\mathcal{E}_2\cos(\omega_2t+\varphi_2)]\\&+\frac{d^2}{\hbar\Delta}\frac{\frac{\Gamma}{2}+\gamma'}{\Delta}[\mathcal{E}_1\sin(\omega_1t+\varphi_1)\\&+\mathcal{E}_2\sin(\omega_2t+\varphi_2)] .\end{aligned} \tag{11}$$

The first term of (11) corresponds to the linear dispersion of the medium and the second one to the linear absorption. The quantities $\langle d\rangle^{(\mathrm{fr})}$ and $\langle d\rangle^{(\mathrm{coll})}$ are terms in the expansion of $\langle d\rangle$ which are of third order in the field amplitudes, representing the collision-free and pressure-induced terms, respectively. Starting from (8b), we calculate $\langle d\rangle^{(\mathrm{fr})}$ to be

$$\begin{aligned}\langle d\rangle^{(\mathrm{fr})}=&\langle d(\omega_1)\rangle^{(\mathrm{fr})}+\langle d(\omega_2)\rangle^{(\mathrm{fr})}\\&+\langle d(2\omega_1-\omega_2)\rangle^{(\mathrm{fr})}+\langle d(2\omega_2-\omega_1)\rangle^{(\mathrm{fr})} ,\end{aligned} \tag{12}$$

with

$$\langle d(\omega_1)\rangle^{(\mathrm{fr})}=\frac{d^4}{2\hbar^3\Delta^3}(\mathcal{E}_1^2+2\mathcal{E}_2^2)\mathcal{E}_1\cos(\omega_1t+\varphi_1) , \tag{13a}$$

$$\langle d(\omega_2)\rangle^{(\mathrm{fr})}=\frac{d^4}{2\hbar^3\Delta^3}(\mathcal{E}_2^2+2\mathcal{E}_1^2)\mathcal{E}_2\cos(\omega_2t+\varphi_2) , \tag{13b}$$

$$\langle d(2\omega_1-\omega_2)\rangle^{(\mathrm{fr})}=\frac{d^4}{2\hbar^3\Delta^3}\mathcal{E}_1^2\mathcal{E}_2\cos[(2\omega_1-\omega_2)t+2\varphi_1-\varphi_2] , \tag{13c}$$

$$\langle d(2\omega_2-\omega_1)\rangle^{(\mathrm{fr})}=\frac{d^4}{2\hbar^3\Delta^3}\mathcal{E}_2^2\mathcal{E}_1\cos[(2\omega_2-\omega_1)t+2\varphi_2-\varphi_1] . \tag{13d}$$

In the expressions (13), we have kept only the dominant terms.[20] The term (13a) corresponds to the nonlinear modification of the susceptibility of the field E_1. Similarly, (13b) is associated to the nonlinear modification of the susceptibility of the field E_2. The terms (13c) and (13d) are related to the possible generation of new frequencies $(2\omega_1-\omega_2)$ and $(2\omega_2-\omega_1)$ as a result of the nonlinear interaction between the atoms and the incident fields.

We now consider the pressure-induced terms which are calculated starting from (8c). Using the same decomposition as in (12),

$$\begin{aligned}\langle d\rangle^{(\mathrm{coll})}=&\langle d(\omega_1)\rangle^{(\mathrm{coll})}+\langle d(\omega_2)\rangle^{(\mathrm{coll})}\\&+\langle d(2\omega_1-\omega_2)\rangle^{(\mathrm{coll})}+\langle d(2\omega_2-\omega_1)\rangle^{(\mathrm{coll})} .\end{aligned} \tag{14}$$

We find

$$\langle d(\omega_1)\rangle^{(\mathrm{coll})}=\frac{\gamma'}{\Gamma}\frac{d^4}{\hbar^3\Delta^3}[(\mathcal{E}_1^2+\mathcal{E}_2^2)\mathcal{E}_1\cos(\omega_1t+\varphi_1)]+\frac{\gamma'}{\Gamma^2+\delta^2}\frac{d^4\mathcal{E}_2^2}{\hbar^3\Delta^3}[\Gamma\mathcal{E}_1\cos(\omega_1t+\varphi_1)+\delta\mathcal{E}_1\sin(\omega_1t+\varphi_1)] , \tag{15a}$$

$$\langle d(\omega_2)\rangle^{(\mathrm{coll})}=\frac{\gamma'}{\Gamma}\frac{d^4}{\hbar^3\Delta^3}[(\mathcal{E}_1^2+\mathcal{E}_2^2)\mathcal{E}_2\cos(\omega_2t+\varphi_2)]+\frac{\gamma'}{\Gamma^2+\delta^2}\frac{d^4\mathcal{E}_1^2}{\hbar^3\Delta^3}[\Gamma\mathcal{E}_2\cos(\omega_2t+\varphi_2)-\delta\mathcal{E}_2\sin(\omega_2t+\varphi_2)] , \tag{15b}$$

$$\langle d(2\omega_1-\omega_2)\rangle^{(\mathrm{coll})}=\frac{\gamma'}{\Gamma^2+\delta^2}\frac{d^4\mathcal{E}_1^2\mathcal{E}_2}{\hbar^3\Delta^3}\{\Gamma\cos[(2\omega_1-\omega_2)t+2\varphi_1-\varphi_2]+\delta\sin[(2\omega_1-\omega_2)t+2\varphi_1-\varphi_2]\} , \tag{15c}$$

$$\langle d(2\omega_2-\omega_1)\rangle^{(\mathrm{coll})}=\frac{\gamma'}{\Gamma^2+\delta^2}\frac{d^4\mathcal{E}_2^2\mathcal{E}_1}{\hbar^3\Delta^3}\{\Gamma\cos[(2\omega_2-\omega_1)t+2\varphi_2-\varphi_1]-\delta\sin[(2\omega_2-\omega_1)t+2\varphi_2-\varphi_1]\} . \tag{15d}$$

All these terms are proportional to γ' and thus increase linearly with the buffer gas pressure. They are associated with the pressure-induced contributions to the nonlinear susceptibility of beam E_1 [Eq. (15a)], of beam E_2 [Eq. (15b)], and with the generation of new frequencies $2\omega_1-\omega_2$ and $2\omega_2-\omega_1$.

D. Discussion

In all the quantities considered above (population of the upper level, nonlinear contribution to the electric dipole moment, etc.) we have separated the collision-free terms from the pressure-induced ones. It can be seen that some common features are always exhibited. First, the variation with δ exhibits a resonance behavior (around $\delta=0$) only for the pressure-induced terms. The collision-free terms are insensitive to a small variation of δ. Second, the collision-free components of the electric dipole moment are in phase with the incident fields, while a dephasing depending on the ratio δ/Γ always exists for the pressure-induced terms. All these features will be physically interpreted in Sec. II using a new dressed-atom approach. We will also show that the separation between the collision-free and the pressure-induced terms is not only mathematically convenient but can equally well be associated with a different atomic excitation mechanism.

Finally, we note that several effects studied during the recent years are described by formulas (13) and (15). If we set $\varphi_1=-\mathbf{k}_1\cdot\mathbf{r}$ and $\varphi_2=-\mathbf{k}_2\cdot\mathbf{r}$, then E_1 and E_2 are associated with two beams propagating in directions $\mathbf{k}_1$ and $\mathbf{k}_2$, respectively. Formulas (15c) and (15d), together with (13c) and (13d), describe four-wave mixing generation in the directions $2\mathbf{k}_1-\mathbf{k}_2$ and $2\mathbf{k}_2-\mathbf{k}_1$. The PIER 4 resonance[3] centered at $\delta=0$ is apparent in expressions (15c) and (15d). Formulas (13c) and (13d) are associated with the background terms observed when $|\delta|\gg\Gamma$ or when collisions can be neglected ($\gamma'\simeq 0$).

Formula (13a) is associated with the collision-free component of the nonlinear susceptibility $\chi_1=\chi_1'+i\chi_1''$ of beam E_1. Since $\langle d(\omega_1)\rangle^{(\mathrm{fr})}$ is in phase with E_1, this term only describes a modification of the real part of the susceptibility χ_1'. We note that this term induces a phase shift for the field E_1 proportional to $\mathcal{E}_1^2+2\mathcal{E}_2^2$, which is not a symmetric function of $\mathcal{E}_1$ and $\mathcal{E}_2$ (independent of the value of δ). On the other hand, in the pressure-induced term (15a), we can separate a dispersive part which contributes to χ_1' and an absorptive part which contributes to χ_1'' [this is the $\sin(\omega_1 t+\varphi_1)$ term]. Let us first discuss the pressure-induced contribution to χ_1'. We note that, contrary to the collision-free term (13a), the pressure-induced phase shift for field E_1 is proportional to $\mathcal{E}_1^2+\mathcal{E}_2^2$ when $|\delta|\gg\Gamma$, and is then a symmetric function of $\mathcal{E}_1$ and $\mathcal{E}_2$. On the other hand, the asymmetry still exists when $|\delta|\lesssim\Gamma$. It has been shown that these results explain the nonreciprocity in the output of a four-wave mixing ring oscillator pumped by two asymmetric pump beams.[6] Formula (15a) also shows that the pressure-induced contribution to χ'' can be either positive or negative depending on the sign of δ. A pressure-induced amplification has thus been predicted and observed.[4] The corresponding process has been called pressure-induced two-wave mixing or pressure-induced Rayleigh gain, since χ'' attains its extrema when $|\delta|\sim\Gamma$.

II. NONSTANDARD DRESSED-ATOM APPROACH

A. Description of the model

We again consider a two-level atom interacting with two fields $E_1(t)$ and $E_2(t)$ given by the expressions (1a) and (1b). As shown above, the frequency difference between the two beams δ is an important quantity for the experimental observations. It is thus interesting to describe the total field $E(t)=E_1(t)+E_2(t)$ as a field oscillating at a mean frequency $[\simeq(\omega_1+\omega_2)/2]$, with a modulation at frequency $\simeq\delta=(\omega_1-\omega_2)$. We thus transform

$$E(t)=\mathcal{E}_1\cos(\omega_1 t+\varphi_1)+\mathcal{E}_2\cos(\omega_2 t+\varphi_2) \tag{16}$$

into

$$E(t)=\mathcal{E}\cos(\omega t+\Phi)\ , \tag{17}$$

with

$$\mathcal{E}=[\mathcal{E}_1^2+\mathcal{E}_2^2+2\mathcal{E}_1\mathcal{E}_2\cos(\delta t+\varphi)]^{1/2}\ , \tag{18a}$$

$$\varphi=\varphi_1-\varphi_2\ , \tag{18b}$$

$$\omega=(\omega_1+\omega_2)/2\ , \tag{18c}$$

$$\Phi=\tfrac{1}{2}(\varphi_1+\varphi_2)+\alpha\ , \tag{18d}$$

$$\alpha=\tan^{-1}\left[\frac{\mathcal{E}_1-\mathcal{E}_2}{\mathcal{E}_1+\mathcal{E}_2}\tan\left[\frac{\delta t}{2}+\frac{\varphi_1-\varphi_2}{2}\right]\right]\ . \tag{18e}$$

If the amplitude $\mathcal{E}$ and the phase Φ of the field (17) were fixed quantities, it would be straightforward to quantize the field and to consider the eigenstates of the atom dressed by this field.[13] Actually, the Hamiltonian of the dressed atom is

$$H'=H_0'+V'\ , \tag{19}$$

with

$$H_0'=\hbar\omega_0|b\rangle\langle b|+\hbar\omega a^\dagger a\ , \tag{20a}$$

$$V'=g(S^+ae^{-i\Phi}+S^-a^\dagger e^{i\Phi})\ , \tag{20b}$$

where S^+ and S^- are the operators $|b\rangle\langle a|$ and $|a\rangle\langle b|$ and g is a coupling constant between the atom and the field that is equal to $\hbar\Omega_1/2\sqrt{\langle N\rangle}$, where $\langle N\rangle$ is the mean number of photons in the mode ω and Ω_1 is the resonant Rabi frequency, equal to

$$\Omega_1=-\frac{d\mathcal{E}}{\hbar}\ . \tag{21}$$

For constant $\mathcal{E}$, the eigenstates of H' are

$$|2(N)\rangle=(\cos\theta)|a,N+1\rangle-e^{-i\Phi}(\sin\theta)|b,N\rangle\ , \tag{22a}$$

$$|1(N)\rangle=e^{i\Phi}(\sin\theta)|a,N+1\rangle+(\cos\theta)|b,N\rangle\ , \tag{22b}$$

where $|a,N+1\rangle$ and $|b,N\rangle$ are eigenstates of H_0' with energies $(N+1)\hbar\omega$ and $\hbar\omega_0+N\hbar\omega$, and θ is defined by the relation

$$\mathrm{cotan}(2\theta)=-\frac{\Delta}{\Omega_1},\quad 0\le 2\theta<\pi\ . \tag{23}$$

In the perturbative limit ($|d|\mathcal{E}\ll\hbar|\Delta|$) considered here, the eigenstates $|2(N)\rangle$ and $|1(N)\rangle$ can be written as[21]

$$|2(N)\rangle=|a,N+1\rangle+\frac{\Omega_1 e^{-i\Phi}}{2\Delta}|b,N\rangle\ , \tag{24a}$$

$$|1(N)\rangle=|b,N\rangle-\frac{\Omega_1 e^{i\Phi}}{2\Delta}|a,N+1\rangle\ . \tag{24b}$$

In fact, in our system, $\mathcal{E}$ and Φ are not constants but vary very slowly with time [according to Eqs. (18a) and (18d)].

However, as long as the assumption of our study ($|\delta| \sim \Gamma, |\Delta| \gg \Gamma, d\mathcal{E}_1/\hbar, d\mathcal{E}_2/\hbar$) is valid, we can still quantize the field of frequency ω by the procedure given above. In this case, the amplitude and the phase of the atom-field coupling vary very slowly and $|1(N)\rangle$ and $|2(N)\rangle$ are adiabatic eigenstates of the system.[22]

Since there is actually no field having frequency $(\omega_1+\omega_2)/2$, but two fields at frequencies ω_1 and ω_2, one can consider this approach as a way to avoid the mathematical difficulties found in the problem of a two-level atom dressed by two electromagnetic fields. Another possibility would have been to quantize only one field and to treat the second field classically. However, this method does not respect the symmetry between the two fields.[23] The approach presented here has the first obvious advantage to maintain the symmetry between the two fields.

Another interest of the method is that $|1(N)\rangle$ and $|2(N)\rangle$ behave as quasistationary states of the system. Provided that $|\delta/\Delta| \ll 1$ and secular approximation, the only coupling between $|1(N)\rangle$ and $|2(N)\rangle$ is collisional in nature. In the following we shall assume that these conditions are fulfiled and consequently we shall use the formulas established for the collisional relaxation of a two-level atom dressed by a single-mode field.[24]

B. Population of the upper level: Fluorescence beats emitted by a two-level atom

In the absence of collisions, the only states that are populated are the $|2(N)\rangle$ states. Collisions induce a transfer from $|2(N)\rangle$ to $|1(N)\rangle$ with a rate equal to

$$w=\gamma'\frac{\Omega_1^2}{2\Delta^2} . \tag{25}$$

Since w is proportional to both γ' and Ω_1^2, the transfer to the $|1(N)\rangle$ state increases with the buffer gas pressure and with the light intensity.

The state $|1(N)\rangle$ decays toward the state $|2(N-1)\rangle$ with a rate Γ (owing to spontaneous emission).[25] The population Π_1 of the level $|1(N)\rangle$ is found by solving the rate equation

$$\frac{d}{dt}\Pi_1=-\Gamma\Pi_1+w . \tag{26}$$

The Rabi frequency Ω_1 given by Eqs. (21) and (18a) is now a function of time, resulting in a time-dependent rate w given by

$$w=\gamma'\frac{d^2}{2\hbar^2\Delta^2}[\mathcal{E}_1^2+\mathcal{E}_2^2+2\mathcal{E}_1\mathcal{E}_2\cos(\delta t+\varphi)] . \tag{27}$$

The nontransient solution of (26) is thus

$$\Pi_1=\frac{\gamma'}{\Gamma}\left[\frac{d^2(\mathcal{E}_1^2+\mathcal{E}_2^2)}{2\hbar^2\Delta^2}+\frac{d^2\mathcal{E}_1\mathcal{E}_2}{\hbar^2\Delta^2}\left[\frac{\Gamma^2}{\Gamma^2+\delta^2}\cos(\delta t+\varphi)+\frac{\Gamma\delta}{\Gamma^2+\delta^2}\sin(\delta t+\varphi)\right]\right] . \tag{28}$$

One recognizes in (28) the result (8b) found for $\rho_{bb}^{(\text{coll})}$. The pressure-induced term in the solution of the optical Bloch equations just corresponds to the population of the upper level of the dressed atom. We now can explain the features of this term. First, we note that the population Π_1 is driven by an excitation modulated in time at the frequency δ. We therefore understand the origin of the modulation at frequency δ and also of the resonance occurring at $\delta=0$. Secondly, since the upper state has a lifetime equal to Γ^{-1}, it is natural to find a resonance whose width is Γ and which does not exhibit any pressure broadening.[19] Finally, the fact that $\rho_{bb}^{(\text{coll})}$ (or Π_1) is not in phase with the applied field just corresponds to the finite response time of the population of the $|1(N)\rangle$ level.

A system in the $|1(N)\rangle$ state decays toward the $|2(N-1)\rangle$ state by emitting a photon at the atomic frequency ω_0 (Fig. 1). The intensity of the light scattered at this frequency ω_0 is thus proportional to Π_1. We see on formula (28) that the scattered intensity should exhibit an oscillation at a frequency equal to δ. A new type of fluorescence beats is thus predicted in the case of two-level atoms interacting with two single-mode fields. These fluorescence beats are pressure induced since they depend on Π_1, which is proportional to the pressure broadening γ' of the atomic transition $a\rightarrow b$. Using Eq. (28), we find that the modulation depth of the fluorescence beats is given by

$$(\Pi_1)_{\max}-(\Pi_1)_{\min}=2\frac{d^2\mathcal{E}_1\mathcal{E}_2}{\hbar^2\Delta^2}\frac{\gamma'}{(\Gamma^2+\delta^2)^{1/2}} . \tag{29}$$

The modulation depth profile as a function of δ exhibits a resonant behavior centered at $\delta=0$, having width (full width at half maximum) equal to $2\sqrt{3}\Gamma$.

Also we note that these fluorescence beats are obtained in a situation where the excitation is not resonant, the energy defect between the atomic energy and the photon energy being supplied by the collisions.[10] There is of course a relationship between these pressure-induced fluorescence beats and the pressure-induced optical pumping considered in previous papers.[7]

C. Mean value of the electric dipole moment

For a two-level atom dressed by a single-mode field, the mean value of the electric dipole moment is given by[13]

$$\langle d\rangle=(\Pi_1-\Pi_2)d\sin(2\theta)\cos(\omega t+\phi) , \tag{30}$$

where Π_1 and Π_2 are the populations of the levels $|1(N)\rangle$ and $|2(N)\rangle$ (normalized by $\Pi_1+\Pi_2=1$) and θ is defined by (23).

The mean value of the dipole moment is proportional to the difference of population between the levels $|1(N)\rangle$ and $|2(N)\rangle$. The factor $d\sin(2\theta)$ comes from the matrix element of the electric dipole moment between the states

$|i(N)\rangle$ and $|i(N-1)\rangle$ (where $i=1,2$) for which the Bohr frequency is equal to ω.

Using $\Pi_1+\Pi_2=1$, we transform (30) into

$$\langle d\rangle=(2\Pi_1-1)d\sin(2\theta)\cos(\omega t+\phi)\ . \tag{31}$$

In the perturbative limit ($|\Omega_1|/|\Delta|\ll 1$), the terms of third order in the input field can be obtained in two ways. We can first multiply Π_1, which is a second-order term by $\sin(2\theta)\sim 2\theta$, which is a first-order term. Since Π_1 is proportional to γ', we thus obtain a pressure-induced term. We can also consider the term -1 in $(2\Pi_1-1)$ and develop $\sin(2\theta)$ to third order in Ω_1. This contribution to the dipole moment is obviously independent of the pressure broadening and thus leads to the collision-free terms for the electric dipole moment.

Let us first consider these collision-free terms. As mentioned, we have to develop $\sin(2\theta)$ as a function of Ω_1/Δ. We start from

$$\sin(2\theta)=\frac{\tan(2\theta)}{[1+\tan^2(2\theta)]^{1/2}}\simeq\tan(2\theta)\left[1-\frac{\tan^2(2\theta)}{2}\right]\ , \tag{32}$$

and using (23), we find

$$\sin(2\theta)=-\frac{\Omega_1}{\Delta}\left[1-\frac{\Omega_1^2}{2\Delta^2}\right]=-\frac{\Omega_1}{\Delta}+\frac{\Omega_1^3}{2\Delta^3}\ . \tag{33}$$

We do not consider the term linear in Ω_1 (which leads to the linear response), and using (21), (31), and (33) we calculate the collision-free third-order component of the electric dipole moment

$$\langle d\rangle^{(\mathrm{fr})}=\frac{d^4}{2\hbar^3\Delta^3}\mathcal{E}^3\cos(\omega t+\phi)\ . \tag{34}$$

Using (16), (17), and (18a) we transform $\mathcal{E}^3\cos(\omega t+\phi)$ into

$$\begin{aligned}[\mathcal{E}_1^2+\mathcal{E}_2^2+2\mathcal{E}_1\mathcal{E}_2\cos(\delta t+\varphi)][\mathcal{E}_1\cos(\omega_1 t+\varphi_1)+\mathcal{E}_2\cos(\omega_2 t+\varphi_2)]\\ =(\mathcal{E}_1^2+\mathcal{E}_2^2)[\mathcal{E}_1\cos(\omega_1 t+\varphi_1)+\mathcal{E}_2\cos(\omega_2 t+\varphi_2)]\\ +\mathcal{E}_1^2\mathcal{E}_2\{\cos(\omega_2 t+\varphi_2)+\cos[(2\omega_1-\omega_2)t+2\varphi_1-\varphi_2]\}+\mathcal{E}_1\mathcal{E}_2^2\{\cos(\omega_1 t+\varphi_1)+\cos[(2\omega_2-\omega_1)t+2\varphi_2-\varphi_1]\}\ .\end{aligned} \tag{35}$$

Regrouping these terms in formula (34), we find an expression for $\langle d\rangle^{(\mathrm{fr})}$ which is exactly equal to the one obtained in formulas (12) and (13).

It follows from this theory that the collision-free terms in the third-order component of the electric dipole moment are associated with the modification of the wave function of the $|2(N)\rangle$ state. Since the wave function follows instantaneously the variation of the applied fields, we understand why the terms appearing in $\langle d\rangle^{(\mathrm{fr})}$ are in phase with the applied fields. Also, since this process is basically nonresonant, we do not expect to find any resonance when δ is scanned. Finally, since the level $|1(N)\rangle$ is not involved in this process, it is reasonable that the decay rate of this $|1(N)\rangle$ level does not appear in the final result.

We now consider the pressure-induced terms in $\langle d\rangle$. These terms are obtained by replacing in (31) Π_1 by its value given in (28) and $\sin(2\theta)$ by its first-order approximation $(-\Omega_1/\Delta)$. This leads to

$$\langle d\rangle^{(\mathrm{coll})}=\frac{2\gamma'}{\Gamma}\left[\frac{d^2(\mathcal{E}_1^2+\mathcal{E}_2^2)}{2\hbar^2\Delta^2}+\frac{d^2\mathcal{E}_1\mathcal{E}_2}{\hbar^2\Delta^2}\left[\frac{\Gamma^2}{\Gamma^2+\delta^2}\cos(\delta t+\varphi)+\frac{\Gamma\delta}{\Gamma^2+\delta^2}\sin(\delta t+\varphi)\right]\right]\frac{d^2\mathcal{E}}{\hbar\Delta}\cos(\omega t+\phi)\ . \tag{36}$$

Using (16) and (17), we write

$$\mathcal{E}\cos(\omega t+\phi)=\mathcal{E}_1\cos(\omega_1 t+\varphi_1)+\mathcal{E}_2\cos(\omega_2 t+\varphi_2)\ . \tag{37}$$

Finally, we transform (36) using trigonometric relations. For example,

$$\cos(\delta t+\varphi)\cos(\omega_1 t+\varphi_1)=\tfrac{1}{2}\{\cos(\omega_2 t+\varphi_2)+\cos[(2\omega_1-\omega_2)t+2\varphi_1-\varphi_2]\}\ . \tag{38}$$

By regrouping the terms in (36), we find an expression for $\langle d\rangle^{(\mathrm{coll})}$ which exactly coincides with the one obtained in formulas (14) and (15).

The dressed-atom approach presented here permits a clear physical separation between the collision-free terms and the pressure-induced ones. The former are associated with the modification of the wave functions and follow instantaneously the applied fields, while the latter are associated with a collisionally aided excitation of an initially unpopulated level of the dressed atom. Because of the finite lifetime Γ^{-1} of this level, the pressure-induced component of the electric dipole moment exhibits a dephasing with respect to the applied fields. Also, we find in $\langle d\rangle^{(\mathrm{coll})}$ all the characteristics already discussed for the pressure-induced population Π_1 and which are related to the fact that the excitation w is not static but is modulated at the frequency δ.

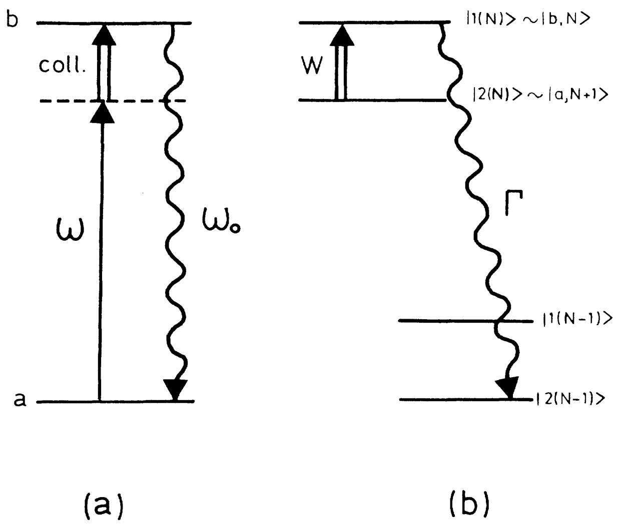

FIG. 1. Effect of a dephasing collision in the bare-atom picture (a) and in the dressed-atom picture (b). Schematically, the collision provides the energy necessary to reach the upper state in (a) and induces transition inside a multiplet of the dressed-state energy diagram in (b). Once the atom is the upper state, it decays by emitting a spontaneous photon of frequency ω_0 with a rate Γ.

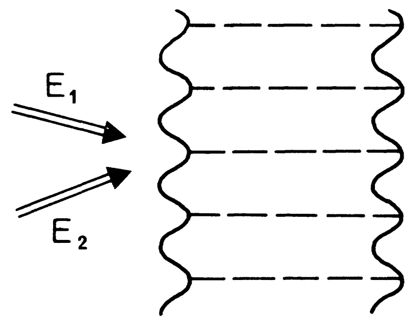

FIG. 3. Spatial modulation of intensity (light grating) created by the incident beams E_1 and E_2. When these beams have different frequencies the grating moves inside the medium. The probability of collisionally aided excitation is maximum at a light grating antinode. Because of the atomic lifetime Γ^{-1}, the dressed-atom grating is retarded with respect to the light grating.

D. Physical interpretation of the PIER 4 resonances and of the Rayleigh gain

As shown above, all the pressure-induced effects are associated with the collision-aided excitation of the $|1(N)\rangle$ level. We are able to explain these pressure-induced effects through this collisional excitation.

As noted above, the excitation of level $|1(N)\rangle$ is modulated at a frequency $\delta=\omega_1-\omega_2$ equal to the beat frequency between the two applied fields. The population Π_1 of the level $|1(N)\rangle$ has thus a static component and a component oscillating at frequency δ. The interaction of the atom with a third field of frequency ω_3 leads to a dipole having components at ω_3 (because of the static component of the population) and sidebands at $\omega_3+\delta$ and $\omega_3-\delta$. In the case where $\omega_3=\omega_1$, the frequencies of the sidebands are $2\omega_1-\omega_2$ and ω_2. The first component is associated with a four-wave mixing generation at $2\omega_1-\omega_2$, while the second is associated with the modification of the propagation of beam E_2 by beam E_1. When $|\delta|$ becomes very large compared to Γ, the modulation of the excitation is too fast by comparison with the time response of the excited state and the modulated component becomes vanishingly small. It follows that the pressure-induced component of the four-wave mixing generation goes to 0. (However, the four-wave mixing generation does not disappear because of the collision-free background terms which are independent of δ.)

Let us now look more closely at $\langle d(\omega_1)\rangle^{(\mathrm{coll})}$ and $\langle d(\omega_2)\rangle^{(\mathrm{coll})}$ given by formulas (15a) and (15b). Because of the finite response time Γ^{-1} of the excited level, $\langle d(\omega_1)\rangle^{(\mathrm{coll})}$ is generally not in phase with $\mathcal{E}_1\cos(\omega_1 t+\varphi_1)$ and $\langle d(\omega_2)\rangle^{(\mathrm{coll})}$ is not in phase with $\mathcal{E}_2\cos(\omega_2 t+\varphi_2)$. More precisely, if $\langle d(\omega_1)\rangle^{(\mathrm{coll})}$ is in advance with respect to $E_1(t)$, then $\langle d(\omega_2)\rangle^{(\mathrm{coll})}$ is retarded with respect to $E_2(t)$ (see Fig. 2). This shows that E_1 can be amplified while E_2 experiences an extra-absorption (or

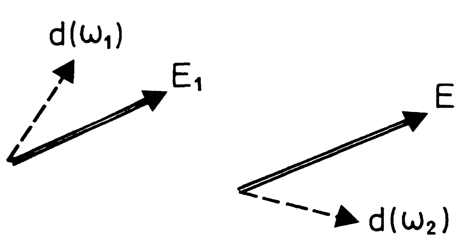

FIG. 2. Relative positions of the fields and of the dipole components in a phase diagram. Because of the pressure-induced terms, $d(\omega_1)$ and $d(\omega_2)$ are not in phase with the fields oscillating at frequencies ω_1 and ω_2. One of the dipole components is retarded (which leads to an extra-absorption), while the other is advanced (which leads to an amplification).

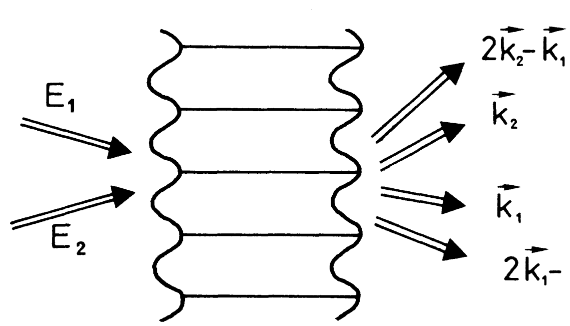

FIG. 4. Diffraction of the beams E_1 and E_2 on the atomic grating created by these two beams.

vice versa) and that energy is transfered from one beam to the other. This is the origin of the pressure-induced two-wave mixing.[4]

E. Dressed-atom gratings

Additional insight can be found by examining the spatial variation of the dipole moment.[17] Let us consider two beams E_1 and E_2 propagating in the directions $\mathbf{k}_1$ and $\mathbf{k}_2$ implying values $\varphi_1 = -\mathbf{k}_1 \cdot \mathbf{r}$ and $\varphi_2 = -\mathbf{k}_2 \cdot \mathbf{r}$. The two beams E_1 and E_2 create a light grating (i.e., a light beam having spatial modulation) (Fig. 3). This grating has a spatial period equal to $2\pi/|\mathbf{k}_1 - \mathbf{k}_2|$ and moves with a velocity proportional to $\omega_1 - \omega_2$. Because of the spatial modulation of the light intensity, the collisionally aided process is also spatially dependent and a grating of atoms excited in the $|1(N)\rangle$ level is formed. The existence of such a grating is manifest in formula (28), giving the population Π_1 of level $|1(N)\rangle$ when we replace φ_1 and φ_2 in that equation by $-\mathbf{k}_1 \cdot \mathbf{r}$ and $-\mathbf{k}_2 \cdot \mathbf{r}$, respectively. Let us now consider the behavior of this population grating for three different values of $\omega_1 - \omega_2$. When $\omega_1 - \omega_2 = 0$, the light spatial modulation is stationary. The collisionally aided excitation is maximum at an antinode of the light spatial modulation and minimum at a node. The population grating, like the light grating, remains stationary and the modulation depth of the population grating is maximum. When $|\omega_1 - \omega_2| \sim \Gamma$, the light grating and the population grating move together but they no longer coincide [because of the finite lifetime Γ^{-1} of the $|1(N)\rangle$ state, a system in the $|1(N)\rangle$ state does not return immediately to a $|2(N-1)\rangle$ state]. The nodes and antinodes of the population grating are thus retarded with respect to those of the light grating. Finally, when $|\omega_1 - \omega_2| \gg \Gamma$, the light grating moves so rapidly inside the medium that the atoms are almost uniformaly excited. In this case, the modulation depth of the population grating becomes vanishingly small.

Let us consider the interaction of E_1 and E_2 with this population grating. The two beams E_1 and E_2 can be diffracted by this grating (Fig. 4). For example, diffraction of beam E_1 in lowest order leads to three diffracted beams, one propagating in direction $\mathbf{k}_1$ (order 0), one propagating in direction $\mathbf{k}_1 + \mathbf{k}_1 - \mathbf{k}_2 = 2\mathbf{k}_1 - \mathbf{k}_2$ (order 1), and one propagating in direction $\mathbf{k}_1 - (\mathbf{k}_1 - \mathbf{k}_2) = \mathbf{k}_2$ (order -1). Because of the Doppler shift of the moving grating, the beam diffracted in order 1 has a frequency $2\omega_1 - \omega_2$ and the beam in order -1 a frequency ω_2. Let us first consider the beam generated in the $2\mathbf{k}_1 - \mathbf{k}_2$ direction. Its amplitude is maximum when the modulation depth of the population grating is maximum. This occurs when $\omega_1 - \omega_2 = 0$. In contrast, when $|\omega_1 - \omega_2| \gg \Gamma$, the modulation depth goes to 0 and there is no longer any pressure-induced contribution to the four-wave mixing generation. This analysis shows that in the presence of collisions, the four-wave mixing generation at $2\mathbf{k}_1 - \mathbf{k}_2$ exhibits a resonance around $\omega_1 - \omega_2 = 0$, a resonance whose width is of the order of Γ. These are the well-known characteristics of the PIER 4 resonances for a set of two-level atoms.[3]

Let us now consider the beam propagating in the $\mathbf{k}_2$ direction which exits the interaction region. This beam comes from the coherent superposition of the diffraction of E_2 at order 0 and of the diffraction of E_1 at order -1. The sum of these two fields is generally not in phase with E_2. This is the origin of the pressure-induced phase shift which has been shown to be of interest in four-wave mixing gyros.[6] Also the sum of the diffracted fields can be larger than the incident field E_2 and, in this case, a pressure-induced amplification is obtained.[4]

Finally, we want to comment on the collision-free terms. As shown above, these terms do not depend on the populations of the dressed states (Π_1 is equal to 0 when $\gamma' = 0$), but on the variation of the dipole moment with Ω_1 [see formulas (31) and (33)]. Apart from the collisionally assisted grating, there is also a grating of dipole moments that is associated with the dressed-atom wave functions and that follows instantaneously the variation of the applied field. It is the diffraction of E_1 and E_2 on this grating of dipole moments which is the origin of emission at $2\omega_1 - \omega_2$ and $2\omega_2 - \omega_1$ when $|\delta| \gg \Gamma$. Also, since the grating of dipole moments is always in phase with the applied fields, the diffraction of E_1 and E_2 gives terms which only contribute to the dispersion of the fields [see formulas (13a) and (13b)].

III. CONCLUSION

We have presented a nonstandard dressed-atom approach to interpret pressure-induced effects for a set of two-level atoms interacting with radiation fields and a buffer gas. To third order in the input fields, this approach gives a physical basis to the separation between the pressure-induced terms and the collision-free terms. The former are associated with a collisionally aided excitation of a dressed state, while the latter correspond to a modification of the dressed-atom wave functions. All the properties of the nonlinear effects (resonant behavior versus δ, etc.) can be easily interpreted from this model.

This model is also useful if one wants to predict what occurs to fifth order in the input fields. It is well known that, in this case, new extra resonances in multiwave mixing can be found which are not collisionally assisted.[26–28] Actually, these resonances are also associated with a relaxation-assisted population of the dressed states, but now the radiative relaxation replaces the collisions. In other words, the atom is now excited through a three-photon hyper-Raman process involving the emission of a spontaneous photon.[27] In the absence of collisions, the population Π_1 of the dressed state $|1(N)\rangle$ results from the spontaneous emission from $|2(N+1)\rangle$. Using the value of the matrix element of the electric dipole moment between the states $|2(N+1)\rangle$ and $|1(N)\rangle$,[13] one finds that (26) has to be replaced by

$$\frac{d}{dt}\Pi_1 = -\Gamma\Pi_1 + \Gamma\frac{d^4\mathcal{E}^4}{16\hbar^4\Delta^4} . \tag{39}$$

Using (18a), one finds the following stationary solution for Π_1:

$$\Pi_1=\frac{d^4}{16\hbar^4\Delta^4}\left[(\mathcal{E}_1^4+\mathcal{E}_2^4+4\mathcal{E}_1^2\mathcal{E}_2^2)+4(\mathcal{E}_1^3\mathcal{E}_2+\mathcal{E}_1\mathcal{E}_1^3)\left[\frac{\Gamma^2}{\Gamma^2+\delta^2}\cos(\delta t+\varphi)+\frac{\Gamma\delta}{\Gamma^2+\delta^2}\sin(\delta t+\varphi)\right]\right.$$
$$\left.+2\mathcal{E}_1^2\mathcal{E}_2^2\left[\frac{\Gamma^2}{\Gamma^2+4\delta^2}\cos2(\delta t+\varphi)+\frac{2\Gamma\delta}{\Gamma^2+4\delta^2}\sin2(\delta t+\varphi)\right]\right] . \tag{40}$$

One sees that in the absence of collisions, fluorescence beats from the "upper state" are still predicted, but now, two Fourier components, one at frequency δ and the other at frequency 2δ, should be observed. Also, two resonances, one of width Γ and the other of width 2Γ, should appear when δ is scanned.

The terms oscillating at frequency 2δ lead to the generation of a dipole moment oscillating at frequency $3\omega_1-2\omega_2$. Consequently the resonant behavior occurring for $\delta=0$ also exists to fifth order in the field amplitude, even in the absence of collisions for the generation of a field at a frequency $3\omega_1-2\omega_2$. Of course, in the presence of collisional damping, there is a corresponding pressure-induced component to fifth order in the field which can be calculated by taking into account the next-order term in the development of w as a function of Ω_1. The important point is that all of these resonances are associated with the fact that the population of the $|1(N)\rangle$ state is obtained through a relaxation-aided process and that the relaxation aided process is modulated in time when the fields have different frequencies.

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[19]The possibility to quench the upper-state population by collisions is neglected in this paper.

[20]More precisely, nonlinear terms smaller by a quantity of the order of Γ/Δ have been neglected. For example, one could add to (13a) a third-order absorption term proportional to $\sin(\omega_1 t+\varphi_1)$. However, this term is much smaller than the dispersion term (because of the large value of Δ) and its contribution can generally be neglected in the nonlinear processes considered here.

[21]The expansion (24) corresponds to a situation where $\omega_0>\omega$. In the opposite situation where $\omega_0<\omega$, one has to exchange the part played by $|1(N)\rangle$ and $|2(N)\rangle$.

[22]A similar idea has been used by J. Dalibard and C. Cohen-Tannoudji, J. Opt. Soc. Am. B **2**, 1707 (1985), to interpret the radiative cooling of atoms in a standing wave.

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[25]The decay rate from $|1(N)\rangle$ to $|1(N-1)\rangle$ is of the order of $\Gamma\Omega_1^2/\Delta^2 \ll \Gamma$ (see Ref. 13).

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