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Competition between resonant multiphoton ionization and third-harmonic generation: A mean-field model

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In connection with a recent experiment of vacuum-ultraviolet generation in multiphoton resonant ionization of xenon at increasing pressure, a model is elaborated to account for the simultaneous effect of a laser excitation tuned near a three-photon resonance and a pressure-induced emission from the resonant state to the ground state. Under certain assumptions usual in various super-radiance treatments, a system of Bloch equations is derived and numerically solved. The dependence of the ionization probability and third-harmonic production versus the interaction time, gas pressure, dynamical detuning, and laser intensity is extensively studied. A transition between a high and low ionization rate is observed when the pressure increases simultaneously with the disappearance of Rabi oscillations and a saturation of third-harmonic generation. An order-of-magnitude calculation reveals that such phenomena are accessible to experiment and a qualitative comparison with the above-mentioned results in xenon is found to be satisfactory.

I. INTRODUCTION

Recently, third-harmonic generation with a strong suppression of the ionization yield has been reported in a resonant multiphoton ionization experiment of rare gases at a pressure higher than 10^{-1} Torr.[1] The explanation proposed by the authors is the building of a "giant dipole" under the effect of the coherent laser excitation.[2] On the other hand, models involving a competition between "superfluorescent emission" and incoherent decay have been developed for several years.[3] Such effects should be present too in multiphoton ionization experiments[4]: The atomic system is shown to undergo an abrupt transition between a high and a low degree of ionization, and under certain conditions an hysteresis cycle can appear, pointing out the bistable behavior of the system. The theoretical approach of Payne *et al.*[2] is based upon the usual third-harmonic generation theory. It emphasizes the propagation effects of the generated wave which, as well as the focusing, are certainly of importance when dealing with vacuum-ultraviolet (vuv) radiation in a rather dense medium.[5] On the other hand, the bistability models[3,4] use a set of semiclassical Bloch equations derived from a mean-field approximation. They lead to reliable predictions concerning the spectrum and the statistics of the emitted light.

The alternative approach proposed here is an extension of this model, which emphasizes the dynamics of the atomic variables, while the latter treatment was dealing mainly with stationary solutions. The elimination of the radiated field variables is made possible by the introduction of a damping constant which forces this field to follow adiabatically the atomic motion. The main hypothesis here lies in neglecting stimulated emission and absorption. Thus the present treatment of pressure-induced emission is analogous to the super-radiance theory in an optically thin medium,[6,7] including the ionization loss terms. It can naturally apply to a time-varying excitation or a non-zero-bandwidth laser. The resulting mathematical formalism is a set of nonlinear differential equations that have been numerically integrated. The solution clearly exhibits the dependence of ionization and third-harmonic generation yields versus the various physical parameters, namely, the interaction time, dynamical detuning, pressure, and laser intensity.

Section II deals with the theoretical background and the validity range of the present treatment. The differential system of Bloch equations is derived in Sec. III. Then we develop the computer results, and analytical expressions are obtained in the cases of strong or low emission (Sec. IV). A comparison with available experimental data and a general discussion of expected magnitude orders is finally presented in Sec. V.

II. THEORETICAL APPROACH

In the problem of third-harmonic generation competing with resonant multiphoton ionization, the atoms interact strongly with two waves, usually considered separately.

A. Laser-atom interaction

The incident laser yield, which is tuned near the three-photon resonance, is conveniently described in a classical way because spontaneous emission is negligible in this mode,

$$E_1(t)=\epsilon(t)e^{i(k_1 z-\omega_1 t)}+\text{c.c.} , \tag{1}$$

where $\epsilon(t)$ is a slowly varying envelope; note that we do not allow for spatial variations, but we can describe a multimode field within this formalism. Neglecting nonresonant level populations, the single-atom multiphoton ionization is governed by density matrix equations previously established by several authors.[8] Within the two-level model, the coupling of the ground state and the resonant state is described by an effective Hamiltonian, while incoherent losses towards the continuum are depicted by a non-Hermitian Liouville operator. Such a formulation has been used in laser theory.[9]

Let $|j{:}g\rangle$ and $|j{:}e\rangle$ be the ground state and the resonant state of the jth atom. We define the individual operators:

$$R_{3j}=\tfrac{1}{2}(\,|j{:}e\rangle\langle j{:}e\,|-|j{:}g\rangle\langle j{:}g\,|\,) , \tag{2a}$$

$$R_j^-=|j{:}g\rangle\langle j{:}e\,| , \tag{2b}$$

$$R_j^+=|j{:}e\rangle\langle j{:}g\,| . \tag{2c}$$

In the interaction representation, the time evolution of the density matrix is given by

$$\frac{dW}{dt}=\frac{1}{i\hbar}[H_1,W]+[\Gamma_1,W]_+ \tag{3}$$

with

$$H_1=\hbar\sum_j\left[-\Delta R_{3j}+\frac{r_{eg}\epsilon^3}{\hbar^3}e^{3ik_1z_j}R_j^+ + \frac{r_{ge}\epsilon^{*3}}{\hbar^3}e^{-3ik_1z_j}R_j^-\right] , \tag{4}$$

$$\Gamma_1=-\frac{\Gamma}{2}\sum_j|j{:}e\rangle\langle j{:}e\,| , \tag{5}$$

Δ is the dynamical detuning,

$$\Delta=3\omega_1-\frac{E_{eg}}{\hbar}-\frac{|\epsilon(t)|^2}{\hbar^2}\left[\sum_l|\mu_{le}|^2\frac{2\omega_{le}}{\omega_1^2-\omega_{le}^2}-\sum_m|\mu_{mg}|^2\frac{2\omega_{mg}}{\omega_1^2-\omega_{mg}^2}\right] , \tag{6}$$

μ being the electric dipole amplitude. The sum over l, i.e., over atomic states coupled by one photon to the resonant state, includes the continuum states, and if the $|j{:}e\rangle$ state can be photoionized, this sum must be considered as a principal value. Γ is the ionization rate of the resonant state, which may be ionized with one photon or (preferably) more. At last, let

$$\Omega=2r_{eg}\epsilon^3/\hbar^3 \tag{7a}$$

be the three-photon Rabi frequency, with

$$r_{eg}=-\sum_{lm}\frac{\mu_{el}\mu_{lm}\mu_{mg}}{(\omega_{lg}-2\omega_1)(\omega_{mg}-\omega_1)} . \tag{7b}$$

Note that incoherent decay phenomena, such as spontaneous emission or collisional transfer towards other levels than those considered here, can be described by terms like Γ. Here it is assumed that the nonradiative transfer between the resonant state and the ground state is negligible compared to the laser-induced transition rate; the effect of a thermal reservoir could have been accounted for through some appropriate operator.[10]

B. Radiated field-atom interaction

The third-harmonic field, assumed to originate in spontaneous coherent emission, is described quantum mechanically. The main treatment we will refer to is the Bonifacio *et al.* super-radiance master equation.[11,12] The radiated field, quantized inside the active volume itself, may be said to be a single-mode field if this volume is characterized by a Fresnel number much greater than unity. Moreover, a damping term Λ_F is introduced to account for the escape of photons from the medium. This loss factor is essential because it forces the radiated field to follow adiabatically the motion of atoms. Let W be the atom-plus-field density matrix; one has, in the interaction picture

$$\frac{dW}{dt}=\frac{1}{i\hbar}[H_3,W]+\Lambda_F W \tag{8}$$

with

$$H_3=\hbar G\left[a\sum_j e^{ik_3z_j}R_j^+ + a^\dagger\sum_j e^{-ik_3z_j}R_j^-\right] \tag{9}$$

and

$$\Lambda_F W = \kappa([aW, a^\dagger] + [a, Wa^\dagger]) . \quad (10)$$

In these formulas, G is the coupling constant between the atom and the third-harmonic radiated field, κ is a damping factor, and a, $a^\dagger$ are the usual annihilation and creation operators of the field mode k_3.

C. Overall evolution equation

From (3) and (8), the atom-plus-laser-field plus radiated field evolution equation becomes

$$\frac{dW}{dt} = \frac{1}{i\hbar}[H_1, W] + [\Gamma_1, W]_+ + \frac{1}{i\hbar}[H_3, W] + \Lambda_F W . \quad (11)$$

Since the attention is focused on cooperative phenomena, we define the classical collective dipole operators[6] and the number of particles,

$$R^\pm = \sum_j e^{3ik_1 z_j} R_j^\pm , \quad (12a)$$

$$R_3 = \sum_j R_{3j} = \tfrac{1}{2}(N_e - N_g) , \quad (12b)$$

$$N_g = \sum_j | j{:}g \rangle \langle j{:}g | , \quad (12c)$$

$$N_e = \sum_j | j{:}e \rangle \langle j{:}e | , \quad (12d)$$

$$N = N_g + N_e . \quad (12e)$$

Equation (11) will yield the time evolution of the mean values $\langle R^+(t) \rangle$, $\langle N_g(t) \rangle$, $\langle N_e(t) \rangle$, and $\langle a(t) \rangle$. Throughout this paper, we assume that the phase-matching condition is fulfilled, i.e.,

$$k_3 = 3k_1 . \quad (13)$$

This requirement is discussed in the Appendix. Moreover, the following basic hypotheses are made:

$$\kappa \gg \sqrt{N} G , \quad (14a)$$

$$\kappa \gg \Gamma, \Omega . \quad (14b)$$

They imply that stimulated emission and absorption are neglected, which is consistent with the phase-matching assumption. Note that condition (14a) is certainly very difficult to fulfill in the uv region, except for very small systems. However, this approach allows us to exhibit a quite general behavior.

Equation (11) leads to the following general system involving mean values:

$$\frac{d}{dt}\langle R^+ \rangle = -\left[i\Delta + \frac{\Gamma}{2} \right] \langle R^+ \rangle - i\Omega^* \langle R_3 \rangle - 2iG \langle a^\dagger R_3 \rangle , \quad (15a)$$

$$\frac{d}{dt}\langle N_g \rangle = i\frac{\Omega}{2}\langle R^+ \rangle - i\frac{\Omega^*}{2}\langle R^- \rangle + iG\langle aR^+ \rangle - iG\langle a^\dagger R^- \rangle , \quad (15b)$$

$$\frac{d}{dt}\langle N_e \rangle = -\Gamma \langle N_e \rangle - \frac{d}{dt}\langle N_g \rangle , \quad (15c)$$

$$\frac{d}{dt}\langle a \rangle = -iG\langle R^- \rangle - \kappa \langle a \rangle . \quad (15d)$$

III. SYSTEM OF BLOCH EQUATIONS

The above system is not of Bloch type because it involves mean values of products such as $a^\dagger R_3$. It can be put into a more convenient form by using a few approximations already discussed by Bonifacio *et al.*[11,12]

A. "Adiabatic approximation"

From (15d), $\langle a(t) \rangle$ can be expressed as a function of $\langle R^- \rangle$

$$\langle a(t) \rangle = -iG \int_0^t ds\, e^{-\kappa s} \langle R^-(t-s) \rangle . \quad (16)$$

As it will be shown later, $\langle R^-(t) \rangle$ evolves slowly during times of the order of $1/\kappa$ if the conditions $\kappa \gg \Omega, \Gamma, \sqrt{N} G$ are fulfilled. The radiated field thus follows adiabatically the atomic dipole

$$\langle a(t) \rangle = -i\frac{G}{\kappa}\langle R^-(t) \rangle . \quad (17)$$

B. Decorrelation approximation

Under certain conditions, the a and R operators involved in (15) can be decorrelated. First, note that Bonifacio *et al.*[11] have shown, provided the condition $\kappa \gg \sqrt{N} G$ is met, that

$$\langle a^\dagger R^- \rangle = i\frac{G}{\kappa}\langle R^+ R^- \rangle . \quad (18)$$

Second, the validity of the decorrelation

$$\langle R^+ R^-(t) \rangle = \langle R^+(t) \rangle \langle R^-(t) \rangle \quad (19)$$

has been studied numerically. In super-radiance theory, it has been shown that (19) is quite satisfactory for a system initially in a "Dicke state," i.e., a system excited by a coherent $\pi/2$ pulse. The decorrelation is only approximate when a total population inversion is created at time $t=0$, owing to the large quantum fluctuations necessary to initiate the super-radiant pulse.

The validity range of the decorrelation can be studied in the present case. When a full population inversion is reached, the system is driven by the laser field, and the coherent spontaneous emission has a negligible effect on its evolution. Thus the fluctuations otherwise necessary to initiate the super-radiant emission are not of importance here.

From a more quantitative point of view, the departure from the classical situation (where a whole decorrelation is assumed) is shown to be important only when[12]

$$\theta \lesssim \frac{1}{\sqrt{N}} , \tag{20a}$$

θ being the "tipping angle" of the Bloch vector, defined by

$$\langle R_3 \rangle = \frac{N}{2} \cos\theta . \tag{20b}$$

Let us now compare the strengths of the laser driving field and of the radiated field in (15a),

$$\rho = \left| \frac{\Omega^* \langle R_3 \rangle}{2G \langle a^\dagger R_3 \rangle} \right| \simeq \left| \frac{\kappa \Omega}{2G^2 \langle R^+ \rangle} \right| = \frac{\kappa\Omega}{2NG^2} \frac{1}{\sin\theta} . \tag{21}$$

Introducing the boundary value (20a) in (21) we obtain the ratio

$$\rho \simeq \sqrt{N} \frac{\kappa\Omega}{2NG^2} . \tag{22}$$

Provided that the dimensionless quantity $(\kappa\Omega)/(2NG^2)$, hereafter called ω, is much larger than $1/\sqrt{N}$, the laser interaction dominates the evolution when spontaneous emission alone would create important quantum fluctuations. This condition ($\rho \gg 1$) is certainly easy to fulfill practically.

Within these approximations, the following nonlinear differential system can be written:

$$\frac{d}{dt}\langle R^+ \rangle = -\left[i\Delta + \frac{\Gamma}{2} \right] \langle R^+ \rangle - i\frac{\Omega^*}{2}(\langle N_e \rangle - \langle N_g \rangle) + \frac{G^2}{\kappa} \langle R^+ \rangle (\langle N_e \rangle - \langle N_g \rangle) , \tag{23a}$$

$$\frac{d}{dt}\langle N_g \rangle = i\frac{\Omega}{2}\langle R^+ \rangle - i\frac{\Omega^*}{2}\langle R^- \rangle + \frac{2G^2}{\kappa}\langle R^+ \rangle \langle R^- \rangle , \tag{23b}$$

$$\frac{d}{dt}\langle N_e \rangle = -\frac{d}{dt}\langle N_g \rangle - \Gamma \langle N_e \rangle . \tag{23c}$$

We define the "super-radiance rate" as

$$\frac{1}{T_{\mathrm{SR}}} = \frac{2N_0 G^2}{\kappa} , \tag{24}$$

where N_0 is the initial number of atoms. It can also be expressed in terms of the single-atom radiative decay from e to g (ν_{rad}), of the generated wavelength (λ_3) and of the radius of the active volume (a) as[13]

$$\frac{1}{T_{\mathrm{SR}}} = \frac{3}{8\pi^2} N_0 \left[\frac{\lambda_3}{a} \right]^2 \nu_{\mathrm{rad}} = N_0 \mu \nu_{\mathrm{rad}} , \tag{25a}$$

where

$$\mu = \frac{3}{8\pi^2} \left[\frac{\lambda_3}{a} \right]^2 \tag{25b}$$

is a geometrical factor.

This cooperative emission rate allows the definition of four reduced parameters,

$$\frac{t}{T_{\mathrm{SR}}} = \tau , \tag{26a}$$

$$\Delta T_{\mathrm{SR}} = \delta , \tag{26b}$$

$$\Gamma T_{\mathrm{SR}} = \nu , \tag{26c}$$

$$\Omega T_{\mathrm{SR}} = \omega . \tag{26d}$$

We suppose from now on that ω is real without true loss of generality. Using four new real functions

$$\langle R^+(t) \rangle = N_0 [r_1(t) + i r_2(t)] , \tag{27a}$$

$$\langle N_g(t) \rangle = N_0 n_g(t) , \tag{27b}$$

$$\langle N_e(t) \rangle = N_0 n_e(t) , \tag{27c}$$

the system (23) can be written

$$\frac{d}{d\tau} r_1 = -\frac{\nu}{2} r_1 + \delta r_2 + \tfrac{1}{2} r_1 (n_e - n_g) , \tag{28a}$$

$$\frac{d}{d\tau}r_2 = -\delta r_1 - \frac{\nu}{2}r_2 + \tfrac{1}{2}(-\omega + r_2)(n_e - n_g) \,, \tag{28b}$$

$$\frac{d}{d\tau}n_g = -\omega r_2 + r_1^2 + r_2^2 \,, \tag{28c}$$

$$\frac{d}{d\tau}n_e = -\frac{d}{d\tau}n_g - \nu n_e \,. \tag{28d}$$

A new equation can be readily derived from this system,

$$\frac{d}{d\tau}(n_e n_g - r_1^2 - r_2^2) = -\nu(n_e n_g - r_1^2 - r_2^2) \,. \tag{29}$$

Therefore, if initially

$$n_e n_g = r_1^2 + r_2^2 \,, \tag{30}$$

which condition is of course fulfilled for a system in its ground state at $t=0$, then the equality (30) holds at any time. This condition means that no depolarization occurs during the interaction. It would be different if elastic collisions had been included in the effective Hamiltonian, the square of the dipole being then smaller than the product of populations.

To conclude this section, note that usually one also writes the third component of the equivalent vector

$$r_3(t) = \tfrac{1}{2}(n_e - n_g) \,. \tag{31}$$

Here, as pointed out by Georges *et al.*,[8] the classical vector model of Feynman, Vernon, and Hellwarth[14] does not apply because the ionization reduces the length of this vector. However, this physical representation remains useful. For example, the square of the transverse component is

$$r_\perp^2 = r_1^2 + r_2^2 \,. \tag{32}$$

Up to the factor N_0, this is the number of third-harmonic photons emitted per unit time. The total length of the rotating vector is

$$(r_1^2 + r_2^2 + r_3^2)^{1/2} = \tfrac{1}{2}(n_e + n_g) \,, \tag{33}$$

which is the number of neutral atoms still present in the medium divided by $2N_0$.

IV. NUMERICAL SOLUTION

The system (28) has been solved by numerical integration using a standard fourth-order Runge-Kutta method. The limiting case where radiative terms are disregarded leads to the well-known resonant multiphoton ionization solutions.[15] The condition (30) has been checked at each step of integration.

Our results are summarized in Figs. 1–5. In each case, a square pulse is assumed, and the laser intensity is such that $\Omega/\Gamma = 10$. The set of reduced parameters (26) is particularly convenient because it allows for a calculation without specifying the atomic values.

The time dependence of the atomic level populations g,e and ionization probability c is plotted in Fig. 1(a). Figure 1(b) represents the corresponding emitted intensity i and number of photons p. The parameter ω is 10. Note that curve e, population of the excited state, is the derivative of curve c, number of ions, and curve i, instantaneous intensity, is the derivative of p, time-integrated number of photons. In the present case, it is clear that the behavior is mainly determined by the laser driving field. The number of emitted photons remains relatively small.

Figure 2 represents the opposite case, where $\omega = 0.31$, which corresponds to the same laser intensity as in Fig. 1 but to a pressure 32 times higher. The oscillating behavior of the populations disappears during the first six Rabi periods. In this case, the radiative damping is so strong that it may be said to be "overcritical."[16] But ionization

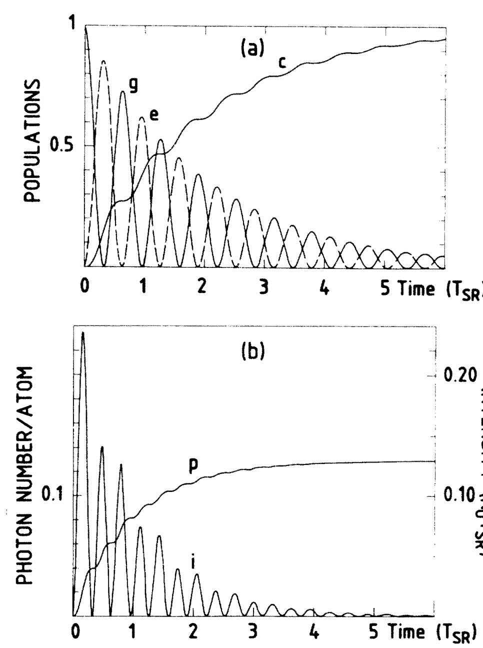

FIG. 1. (a) Time evolution of the atom and (b) the radiated field during the interaction. The reduced parameters defined in (26) are $\omega = 10$, $\nu = 1$, and $\delta = 0$. Abscissas: time $\tau = t/T_{SR}$. Ordinates: (a) g, ground-state population; e, excited-state population; c, ion number per atom. (b) p, photon number per atom, the corresponding scale being on the left; i, radiated intensity with the scale on the right, in units $N_0^2 \mu \nu_{rad}$.

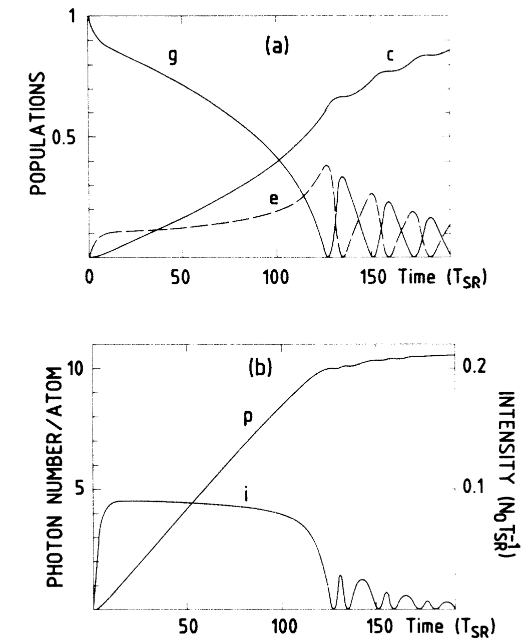

FIG. 2. Time evolution of the atom and radiated field. Same situation as in Fig. 1, except for the pressure which is 32 times higher. Therefore $\omega=3.125\times10^{-1}$, $\nu=3.125\times10^{-2}$, and $\delta=0$. The abscissa scale is such that equal lengths in Fig. 1 and this figure correspond to equal interaction times t and reduced times τ in a ratio of 32.

meanwhile reduces the number of neutral atoms and the cooperative emission, proportional to $n_e n_g$, decreases rapidly when roughly 60% of the atoms are ionized. Then the damped Rabi oscillations reappear. Note that they are asymmetrical: Upward transition is only owing to three-photon absorption, while downward transition is owing to stimulated emission plus spontaneous decay. The generated photons are mainly produced in the first, nonoscillating stage. If the interaction time is shorter than the critical time defined by the transition to the oscillating regime, the ionization probability can strongly decrease compared to its low pressure value.

Figures 3 and 4 are dispersion curves, the zero abscissa corresponding to the dynamical resonance. The solid curves represent the number of emitted photons per atom and the broken lines represent the corresponding ion number.

In Fig. 3, the interaction time is varied, while laser intensity and pressure are kept constant such that $\omega=10^3$, $\nu=10^2$. The generated harmonic signal exhibits the classical Rabi splitting. In this case, the maximum number of photons is not obtained at exact resonance.

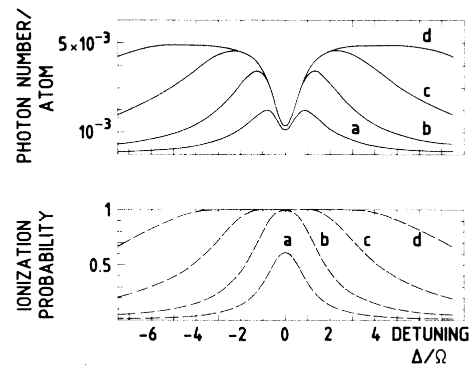

FIG. 3. Dispersion curves of the photon number (solid lines) and the ion number (dashed lines) per atom. Reduced parameters: $\omega=10^3$ and $\nu=10^2$. In the abscissa, Δ is expressed in units of Ω. The various curves correspond to the following interaction times: a, $t=0.02\ T_{SR}=20/\Omega$; b, $t=0.1\ T_{SR}=100/\Omega$; c, $t=0.5\ T_{SR}=500/\Omega$; and d, $t=2.5\ T_{SR}=2500/\Omega$.

In Fig. 4, the time interaction is equal to $100/\Omega$, and the various curves correspond to increasing densities. The ionization yield, saturated at low pressure, decreases while the photon number increases and the "Rabi profile" broadens, no splitting being present if $\omega \lesssim 0.3$.

A more explicit representation of the pressure dependence of these signals is given in Fig. 5. The

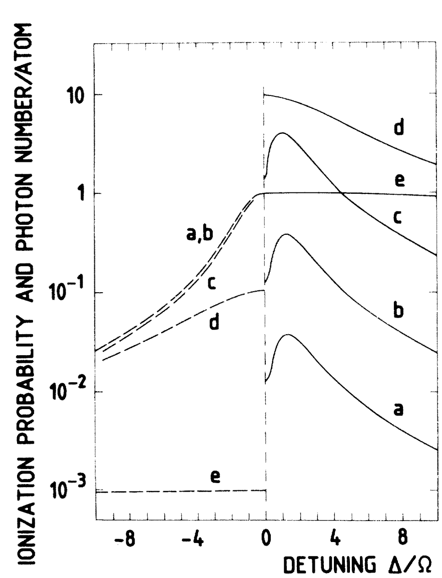

FIG. 4. Dispersion curves of the ion number per atom (dashed lines) and photon number per atom (solid lines). The interaction time is equal to $100/\Omega$, and the laser intensity is such that $\Omega/\Gamma=10$. a, $\nu=10$; b, $\nu=1$; c, $\nu=10^{-1}$; d, $\nu=10^{-2}$; and e, $\nu=10^{-3}$. These curves are symmetrical with respect to the axis $\Delta=0$.

number of photons and ions produced per atom at dynamical resonance is plotted versus the parameter ν^{-1}, directly proportional to the pressure. The various curves correspond to various interaction times.

Essentially, two different regimes can be distinguished. If $\Omega T_{SR} \gg 1$, i.e., at low pressure, the effect of collective emission is small. In this case, the emitted intensity being $r_1^2+r_2^2$ in reduced units, the number of emitted photons can be obtained from

$$\frac{N_{ph}}{N_0} = \int_0^\tau [r_1^2(\sigma)+r_2^2(\sigma)]d\sigma = \int_0^\tau n_e(\sigma)n_g(\sigma)d\sigma \,. \tag{34}$$

Using the results of resonant multiphoton ionization,[15]

$$n_e(t)=\frac{\Omega^2}{2P}e^{-(\Gamma/2)t}\left[\cosh\left[\sqrt{P}\sin\frac{\varphi}{2}t\right]-\cos\left[\sqrt{P}\cos\frac{\varphi}{2}t\right]\right], \tag{35a}$$

$$n_g(t)=\frac{e^{-(\Gamma/2)t}}{P}\left[\frac{1}{2}\left[P+\Delta^2+\frac{\Gamma^2}{4}\right]\cosh\left[\sqrt{P}\sin\frac{\varphi}{2}t\right]+\sqrt{P}\left[\Delta\cos\frac{\varphi}{2}+\frac{\Gamma}{2}\sin\frac{\varphi}{2}\right]\sinh\left[\sqrt{P}\sin\frac{\varphi}{2}t\right]\right.$$
$$\left.+\frac{1}{2}\left[P-\Delta^2-\frac{\Gamma^2}{4}\right]\cos\left[\sqrt{P}\cos\frac{\varphi}{2}t\right]+\sqrt{P}\left[-\Delta\sin\frac{\varphi}{2}+\frac{\Gamma}{2}\cos\frac{\varphi}{2}\right]\sin\left[\sqrt{P}\cos\frac{\varphi}{2}t\right]\right], \tag{35b}$$

where

$$P\cos\varphi=\Omega^2+\Delta^2-\frac{\Gamma^2}{4} \tag{35c}$$

and

$$P\sin\varphi=\Gamma\Delta \,. \tag{35d}$$

The integration involved in (34) can be performed analytically. For an interaction time corresponding to saturation, one obtains

$$\frac{N_{ph}^{(s)}}{N_0}=\frac{1}{2\Gamma T_{SR}}\frac{1}{\left[\Omega^2+\Delta^2-\frac{\Gamma^2}{4}\right]^2+\Gamma^2\Delta^2}\left[\Delta^4+\left[\frac{\Gamma^2}{4}+\frac{7}{4}\Omega^2\right]\Delta^2+\frac{\Omega^4}{4}-\frac{9}{16}\Omega^2\Gamma^2\right.$$
$$\left.+\frac{\Gamma^4}{16}+\frac{\Omega^4\Gamma^2(\Delta^2+\Omega^2+\frac{3}{4}\Gamma^2)}{(\Delta^2+\Omega^2+\frac{3}{4}\Gamma^2)^2+\Delta^2\Gamma^2}\right], \tag{36a}$$

$$\simeq\frac{1}{2\Gamma T_{SR}}\left[1-\frac{\Omega^2}{4}\frac{\Delta^2+3\Omega^2+\frac{\Gamma^2}{4}}{\left[\Delta^2+\Omega^2-\frac{\Gamma^2}{4}\right]^2+\Delta^2\Gamma^2}\right]. \tag{36b}$$

At resonance, the approximate formula (36b) holds if $\Gamma \gg \Omega \gtrsim 1/T_{SR}$ or if $\Omega \gg \Gamma \gtrsim 1/T_{SR}$. Far from resonance, it is valid if $|\Delta| \gg \max(\Gamma,\Omega) \gg 1/T_{SR}$ whatever the ratio Γ/Ω. When $\Omega \gg \Gamma$, the third-harmonic saturation signal is

$$\frac{N_{ph}^{(s)}}{N_0}=\frac{1}{8\Gamma T_{SR}} \tag{37}$$

at dynamical resonance, and

$$\frac{N_{ph}^{(s)}}{N_0}=\frac{1}{2\Gamma T_{SR}} \tag{38}$$

if the saturation is reached far from resonance, this last value being an absolute maximum for N_{ph}/N_0, whatever the parameters.

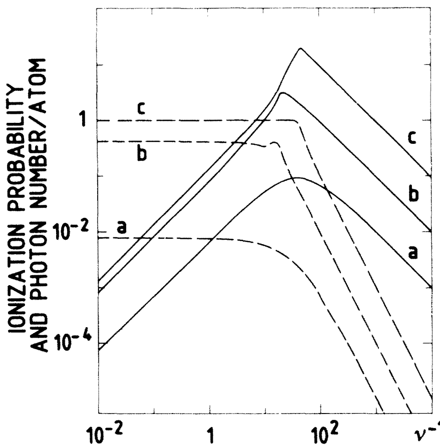

FIG. 5. Constant intensity ν^{-1} dependence (i.e., pressure dependence) of the ion number (dashed lines) and photon number (solid lines) per atom. The chosen value for Ω/Γ is 10. Using the order of magnitude given in Sec. V and the intensity value $I=10^{11}$ $\mathrm{W\,cm^{-2}}$, we get $\nu\sim 1$ when $p\sim 4\times 10^{-4}$ Torr. The various curves correspond to various interaction times. a, $t=1/\Omega$; b, $t=10/\Omega$; and c, $t=100/\Omega$.

As could be guessed, the smaller the ionization rate is, the larger the radiated energy is. Thus an experimental situation where the excited level is ionized with two or more photons is preferable. This condition was met in the Miller *et al.* experiment.[1] Furthermore, the following discussion assumes generally that Ω is much larger than Γ. Last, concerning the low-pressure results, we note that $1/2\Gamma T_{\mathrm{SR}}$ is a linear function of pressure, and thus the total number of emitted photons varies quadratically with the atomic density, as expected in cooperative emission phenomena.

A striking feature in Fig. 5 is the abrupt decrease of the ionization yield for atomic densities such that $\Omega T_{\mathrm{SR}}\lesssim 1$. In the limiting case $1/T_{\mathrm{SR}}\gg\Omega$ and Γ, the ionization probability decreases as N_0^{-2} and thus the total ion number is inversely proportional to the pressure

$$N_i \simeq N_0\Omega^2\Gamma T_{\mathrm{SR}}^2 t=\frac{1}{N_0}\frac{\Omega^2\Gamma}{\mu^2\nu_{\mathrm{rad}}^2}t \tag{39}$$

with the above definition of μ (25b).

On the other hand, the number of photons per atom behaves like ν or N_0^{-1}, and the total number of photons is approximately independent of the pressure,

$$N_{\mathrm{ph}} \simeq N_0\Omega^2 T_{\mathrm{SR}} t=\frac{\Omega^2}{\mu\nu_{\mathrm{rad}}}t \ . \tag{40}$$

These expressions are valid if $\Omega\gg\Gamma$ and only as long as the oscillatory regime is not reached. This condition can be roughly expressed by writing that the photon yield given in (37) is greater than the corresponding value (40),

$$\omega^2\tau\lesssim\frac{1}{8\nu}$$

or (41)

$$\Gamma\Omega^2 T_{\mathrm{SR}}^2 t\lesssim\tfrac{1}{8} \ .$$

It should be emphasized that relation (41) is valid only if the isolated atom ionization process is saturated with the chosen values of the intensity and interaction time. Below saturation, the number of photons emitted at resonance is given by

$$\frac{N_{\mathrm{ph}}}{N_0}\simeq\frac{1}{8\Gamma T_{\mathrm{SR}}}(1-e^{-\Gamma t}) \ , \tag{42}$$

which is valid if t is greater than the Rabi period.

Equating this expression to the one given by (40) leads to an approximate relation characterizing the appearance of Rabi oscillations,

$$\Omega^2\Gamma T_{\mathrm{SR}}^2 t=\tfrac{1}{8}(1-e^{-\Gamma t}) \ . \tag{43}$$

It is interesting to note that if

$$\Omega T_{\mathrm{SR}}\gtrsim\frac{1}{\sqrt{8}} \ , \tag{44}$$

Eq. (43) has no solution and the regime is purely oscillating. Conversely, if by a pressure increase or intensity decrease the opposite condition is met, then (43) defines an optimum laser intensity if pressure and interaction time are kept fixed or equivalently an optimum interaction time for a given intensity and pressure.

The effect of pressure and laser intensity is not completely equivalent. If the laser power is kept fixed, then for sufficient pressure the nonoscillatory regime is reached, and with increasing pressure one observes no spectacular effect (as long as no avalanche phenomena occur) because the number of emitted photons saturates. On the other hand, at a constant atomic density, the photon number is a more complex function of the laser intensity I. At low intensity, it is proportional to I^3 [see (40)]; it then reaches a maximum and finally decreases like I^{-2} if the resonant level is two-photon ionized.

V. DISCUSSION AND CONCLUSION

It is instructive to compare our results to experimental observations of Miller *et al.*[1] No accurate value is available for the Rabi frequency or ionization rate in xenon, so this comparison is essentially qualitative. The reported photon number is roughly linear with pressure below 0.2 Torr.

The present theory predicts a p^2 law under these conditions. Several phenomena not included in it (collisions, for instance) can certainly decrease the emission signal. The strong dependence of the shift and broadening toward the pressure observed in the experiment is not reproduced in our model because propagation effects are not taken into account. The shift considered here is simply induced by the laser excitation. Conversely, the broadening obtained in this model

$$\frac{1}{T_{SR}} = N_0 \mu \nu_{\rm rad}$$

is linear with pressure and has the correct order of magnitude. Furthermore, the saturation of the photon number at high density is well reproduced in our model, as well as the simultaneous abrupt decrease of the ionization rate. A cubic dependence of the third-harmonic production versus the laser intensity was reported, which agrees with the expression (40), valid in the strong emission range. It is thus clear that the "regime transition" described here has been observed by Miller *et al.* Of course, the directivity of the coherent emission is in agreement with the present model where the radiated intensity is concentrated within the diffraction angle.

Let us now give typical orders of magnitude. Assuming a single atom radiative width equal to 10^8 s^{-1} for the resonant level, a third-harmonic wavelength of 1450 Å and a tight focusing, limiting the effective region of interaction to a cylinder 200 μm long, one finds that the super-radiant rate defined by (24), is, in s^{-1},

$$\frac{1}{T_{SR}} \sim (2\times 10^{12})p \ ,$$

where p is in Torr units.

A two-photon ionization cross section is typically 10^{-49} $cm^4\,s$ for nonresonant phenomena.[17] Then

$$\nu \sim 4\times 10^{-26}[I({\rm W\,cm^{-2}})]^2/p \ ,$$

where p is in Torr units.

The determination of the Rabi frequency is much more difficult because expressions like (7b) involve severe destructive interferences. Nevertheless, taking for Ω,[18]

$$\Omega = 2\times 10^{-6}[I({\rm W\,cm^{-2}})]^{3/2}({\rm s^{-1}}) \ ,$$

gives

$$\omega \sim 10^{-18}[I({\rm W\,cm^{-2}})]^{3/2}/p \ ,$$

where p is in Torr units.

This gives a more convenient scale for the figures of the preceding section. For a gas pressure of 10^{-2} Torr, the "critical" situation $\Omega T_{SR} = 1/\sqrt{8}$ occurs at $I = 2\times 10^{10}$ W cm^{-2}. This shows that, in spite of the inaccuracy of these numerical values, the phenomena studied here correspond to realistic situations with the high-power lasers now available.

The present work proposes a formulation of pressure-induced third-harmonic generation which, in contrast to the previous treatments, does not involve the resolution of either the Maxwell equations or of a N-atom density matrix equation. Though propagation effects are not taken into account, this single-atom approach is really adequate to bring a physical representation of the processes.

It predicts a transition between two very different behaviors of the atomic evolution, the critical value of pressure can be related simply to the atom and field parameters. Collisions in the impact approximation can be easily included in the present set of equations. Moreover, this formalism can be easily extended to the case of a multimode laser field. Several theoretical developments are thus possible, and new experiments are desirable to test the conclusions of this model.

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APPENDIX

The validity range of the phase-matching condition (13) is studied here. This condition is

$$(n_3 - n_1)\frac{L}{\lambda_3} \ll 1 \ . \tag{A1}$$

L is the length of the medium, n_3 (or n_1) is the refractive index of the medium at frequency $3\omega_1$ (or ω_1). Since the laser frequency is not one-photon resonant, we assume $n_1 \simeq 1$. Moreover, L is much greater than λ_3; thus (A1) implies that n_3 must not be very different from 1.

The Sellmeier formula gives a rough estimate of the maximum refractive index n_3,

$$n_3^2 - 1 \simeq 2(n_3 - 1) \le \frac{4\pi n r_0 c^2}{3\omega_1 \Lambda} \ , \tag{A2}$$

where n is the vapor density, r_0 is the classical radius of the electron, and Λ is the damping factor of the dipole. In cases of practical interest, the three-photon Rabi frequency Ω is greater than the ionization width, and we will adopt for the electromagnetic broadening the value

$$\Lambda = \Omega \ . \tag{A3}$$

Thus the maximum wave-vector mismatch is given by

$$(n_3 - n_1)\frac{L}{\lambda_3} \leq \frac{1}{4\pi^2\alpha}\frac{e^2 r_0 \lambda_3}{|\mu_{eg}|^2}\frac{1}{\Omega T_{SR}} \ , \tag{A4}$$

where α is the fine-structure constant and all other quantities are defined in the text. An order-of-magnitude estimate (see Sec. V) shows that the numerical factor in front of $1/\Omega T_{SR}$ is close to unity. Thus the phase-matching condition in its most severe form should require

$$\Omega T_{SR} \gg 1 \ . \tag{A5}$$

Assuming $\Omega/\Gamma = 10$, the validity range of the present theory should be $\nu^{-1} \ll 10$. We can see in Fig. 5, for instance, that this restriction removes part of the interest of this study. Nevertheless, we must keep in mind that n_3 is close to its maximal value only when ω_3 differs from E_{eg} by a quantity less than Λ. The disagreement between the experiment and this theory is expected to be smaller for a slightly nonresonant excitation. Finally, it is interesting to note that the phase-matching condition is less severe when $|\mu_{eg}|$ is small, e.g., intercombination lines. Of course, these remarks are mainly qualitative, and a more involved study is necessary at exact resonance.

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