Theoretical explanation of the first experimentally observed laser without inversion in a two-level scheme

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The theory of lasing in an ensemble of driven two-level atoms is shown to agree satisfactorily with the experimental observation of lasing without inversion by Grandclément, Grynberg, and Pinard [Phys. Rev. Lett. **59**, 40 (1987); **59**, 44 (1987)]. It is demonstrated that the lasing is dominated by the multiphoton amplification processes induced in the medium.

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I. INTRODUCTION

Recently, there has been a considerable interest in the so-called "laser without inversion" (LWI), i.e., in schemes that allow for lasing even in the absence of the population inversion between levels involved in the lasing transition [1–3]. It has been shown that some of these schemes may be understood as a typical lasing with inversion, provided an appropriate change of the basis is made [4, 5]. Other proposed models, e.g., [3], seem to defy such an interpretation.

Most of the LWI schemes involve coherent pumping of some atomic transition by a "pump" laser. In such a case a proper understanding of the LWI action can be obtained in the dressed-state (DS) basis, i.e., the basis which diagonalizes the atom–pump-laser interaction [6]. This approach explains the lasing in the so-called double-Λ configuration [2] showing that it is due to the population inversion between DS's [4, 5].

To understand the advantage of the DS basis it is sufficient to consider a two-level atom of transition frequency ω_0 driven by a pump laser of frequency ω_L. The dressed-atom energy eigenstates are shown in Fig. 1(a). The DS doublets, separated by ω_L, are split by the generalized Rabi frequency Ω (I assume $\hbar = 1$); $\Omega^2 = \Omega_0^2 + \Delta_1^2$, where Ω_0 is the resonant Rabi frequency and $\Delta_1 = \omega_0 - \omega_L$ is the pump-laser detuning. For $\Delta_1 \neq 0$, stationary populations of DS are different in value; in particular, for $\Delta_1 > 0$ [as in Fig. 1(a)] there is a population inversion between the lower state of a given doublet (say $|-, n+1\rangle$) and the upper state of the neighboring doublet ($|+, n\rangle$). Thus one may expect gain to occur at the frequency $\omega = \omega_L - \Omega$. Indeed the linear-gain curve [[7]; see Fig. 1(b)] shows gain at $\omega = \omega_L - \Omega$; it predicts also strong absorption at $\omega = \omega_L + \Omega$, easily understood in the DS basis too [Fig. 1(a)]. The above gain indeed may lead to lasing as demonstrated experimentally [8] and independently calculated [9, 10].

Similarly, one may notice a population inversion between $|-, n+1\rangle$ and $|+, n-1\rangle$ DS's. These DS's may be connected by a two-photon transition, thus one may expect gain at $\omega = \omega_L - \Omega/2$. Indeed such a two-photon optical DS laser has been predicted theoretically [10] and very recently observed experimentally [11]. Note, however, that no gain at $\omega = \omega_L - \Omega/2$ appears in Fig. 1(b)—that is, because the curve in Fig. 1(b) corresponds to a linear (weak-field) gain, while the two-photon process requires a nonzero threshold to appear. The theory of nonlinear gain [12], valid for an arbitrary strong probe pulse, correctly predicts gain at $\omega = \omega_L - \Omega/2$ (and for $\omega = \omega_L - \Omega/n$ for n-photon processes, $n \geq 2$). Therefore, it is clear that it is essentially the nonlinear gain, and not its low-field limit, which determines the possibility of lasing. We shall return to this point later on.

Careful inspection of Fig. 1(b) reveals, on the other hand, an additional dispersive-shaped structure centered around $\omega = \omega_L$ with a positive gain at the blue-frequency side (for $\Delta_1 < 0$ on the red side). It is clear from Fig. 1(a) that at $\omega \approx \omega_L$ the transitions involved are that between equally populated DS's, e.g., $|+, n+1\rangle$ and $|+, n\rangle$. One may show, working in the DS basis, that this gain is due to the coherence between the DS's of the same doublet. This coherence appears only when the so-called nonsecular terms in the spontaneous decay of DS's are taken into account. The gain has been only recently nicely interpreted as coming from the interference of several diagrams in the perturbative limit [13].

The question immediately arises whether this gain may be utilized for producing laser action? If so, then one would have yet another (as compared to [1–5] or to the DS lasers [8, 10]) LWI as the populations of levels involved in the transition are exactly equal. The answer indeed is yes, such a LWI was observed a few years ago by Grandclément, Grynberg, and Pinard [14] in collisionally broadened Na vapor. It has been originally interpreted perturbatively in terms of the two-wave-mixing process. The aim of this paper is to present a dynamical theory of this LWI and to show that the understanding of this laser action requires a nonperturbative approach.

II. THEORY AND COMPARISON WITH EXPERIMENT

All necessary details concerning the experiment discussed may be found in [14]. It is worth stressing that the arrangement used reduced the Doppler effect to the

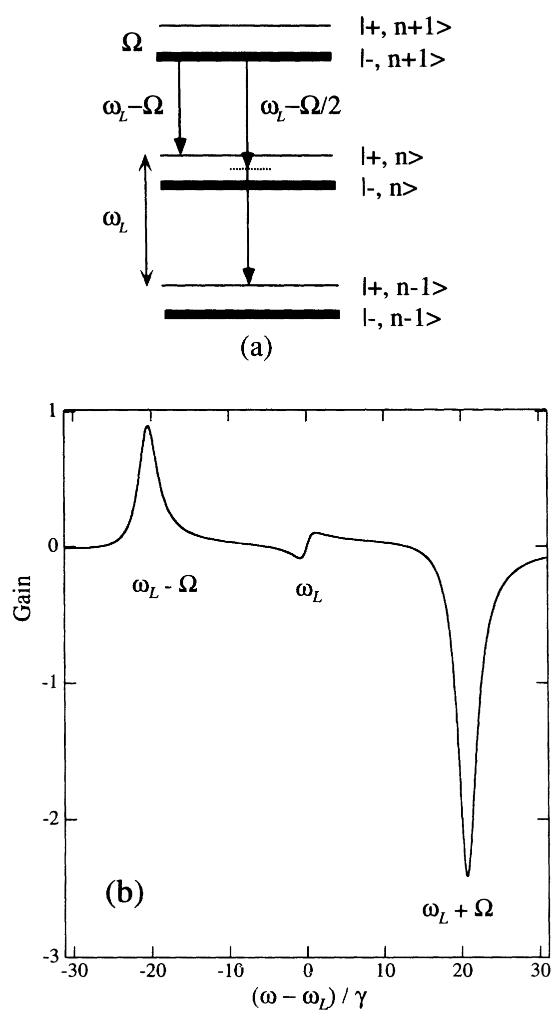

FIG. 1. (a) Dressed-atom doublets represented by lines whose thicknesses are proportional to the relative populations. Gain lines are indicated by arrows. (b) Typical linear-gain curve for $\Omega_0/\gamma = 20$, $\Delta_1/\gamma = 5$; ω is the weak-probe frequency.

value comparable to the inverse of the natural lifetime of the excited state. Therefore, a theory neglecting the Doppler effect should work quite well. We have recently formulated the general theory of lasing in the system of two-level atoms driven by the pump laser of arbitrary intensity and detuning [15] (to be referred to as I). The theory, originally developed as the extension of the previous work [10] on one- and two-photon DS lasers described above, allowed us to discuss the competition between different multiphoton processes created in the medium. Thus this theory is ideally suited to discuss lasing in the presently considered scheme, after extending it (see below) to include collisional broadening.

I briefly sketch here the main points of the theory, referring the reader to [15] for more details. In short the density matrix of the atom-field system is assumed to fulfill the following master equation in the frame rotating with the pump-laser frequency ω_L:

$$\dot{\rho} = -i\,[\mathcal{H},\rho] + \mathcal{L}_F\rho + \mathcal{L}_A\rho, \qquad (1)$$

where

$$\mathcal{H} = \frac{1}{2}\sum_{\mu}^{N}\{\Delta_1\sigma_{3\mu} + \Omega_0(\sigma_\mu + \sigma_\mu^\dagger) + g_\mu\sigma_\mu^\dagger a + g_\mu^* a^\dagger\sigma_\mu\} + \Delta_2 a^\dagger a\,, \qquad (2)$$

$$\mathcal{L}_F\rho = \Gamma(2a\rho a^\dagger - a^\dagger a\rho - \rho a^\dagger a)\,, \qquad (3)$$

and

$$\mathcal{L}_A\rho = \gamma\sum_\mu(2\sigma_\mu\rho\sigma_\mu^\dagger - \sigma_\mu^\dagger\sigma_\mu\rho - \rho\sigma_\mu^\dagger\sigma_\mu) + \frac{\gamma_c}{2}\sum_\mu(\sigma_{3\mu}\rho\sigma_{3\mu} - \rho)\,. \qquad (4)$$

In Eqs. (2)–(4) a ($a^\dagger$) denotes the anihilation (creation) operator of the cavity mode, $\Delta_2 = \omega_c - \omega_L$ is the mismatch between the cavity resonant frequency and the pump-laser frequency, Γ is a cavity half width at half maximum, while g_μ is the strength of μth-atom–cavity-mode coupling. Write $g_\mu = g\exp(-i\varphi_\mu)$. I shall assume the same strength g of atom-cavity coupling for all atoms while phases φ_μ (related to phases of atomic dipoles) will be taken random. Such an assumption is valid for a single-mode traveling-wave cavity or a "mean field" approach for quasidegenerate multimode cavities (see [10] for discussion). Atomic operators (in the standard atomic basis) σ_μ, $\sigma_\mu^\dagger$, and $\sigma_{3\mu}$ denote, respectively, the atomic lowering, the atomic rising, and the atomic inversion operators. The spontaneous atomic decay rate is 2γ while γ_c denotes the rate of collisional dephasing.

In the first step Eqs. (2)–(4) are transformed to the DS basis with the semiclassical DS defined as $|+\rangle_\mu = \cos\alpha|1\rangle_\mu + \sin\alpha|0\rangle_\mu$, $|-\rangle_\mu = -\sin\alpha|1\rangle_\mu + \cos\alpha|0\rangle_\mu$ in terms of the upper $|1\rangle_\mu$ and the lower $|0\rangle_\mu$ μth-atom state. The angle α is defined through the relations $\Omega_0 = \Omega\sin 2\alpha$ and $\Delta_1 = \Omega\cos 2\alpha$. Next, to treat properly the collective behavior of the sample of atoms, we introduce the macroscopic variables: $S^{(k)} = \sum_\mu\langle\sigma_\mu\exp(-ik\phi_\mu)\rangle$ and $S_3^{(k)} = \sum_\mu\langle\sigma_{3\mu}\exp(-ik\phi_\mu)\rangle$, where the $\langle\,\rangle$ symbol stands for both the quantum average and the average over a random distribution of φ_μ. The variables $S^{(k)}$ and $S_3^{(k)}$ have a direct physical meaning. In particular, $S^{(0)}$ denotes the coherence between DS's of the same doublet while $S_3^{(0)}$ is simply a DS inversion [16]. The k-photon [as seen from the power of the $\exp(-i\phi_\mu)$ factor resulting from the atom-cavity interaction via g_μ—see (2)] polarizations $S^{(k)}$ for $k > 0$ ($k < 0$) build up in transitions from the upper, $|+\rangle$ (lower, $|-\rangle$) to the lower (upper) DS's, respectively. The k-photon processes that link upper to upper and lower to lower DS's are described by $S_3^{(k)}$. In particular, single-photon DS lasing is due to the build up of $S^{(-1)}$ ($S^{(1)}$) for $\Delta_1 > 0$ ($\Delta_1 < 0$) while for the two-photon lasing $S^{(-2)}$ ($S^{(2)}$) is dominant.

Making the usual semiclassical approximation, i.e., decorelating atomic and field observables, utilizing the above definitions and Eqs. (2)–(4) the equations of motion of the system are obtained,

$$\dot{S}^{(k)} = -(\gamma_1 + i\Omega)S^{(k)} + iF_+S_3^{(k-1)}A - iF_-A^*S_3^{(k+1)} -2iF_0(A^*S^{(k+1)} + AS^{(k-1)}) +\gamma_0\delta_{k0} + \gamma_3 S_3^{(k)} - \gamma_4 S^{(-k)*} , \quad (5)$$

$$\dot{S}_3^{(k)} = -\gamma_2(S_3^{(k)} - \bar{S}_3\delta_{k0}) +2iF_+(A^*S^{(k+1)} - AS^{(-k+1)*}) +2iF_-(A^*S^{(-k-1)*} - AS^{(k-1)}) +2\gamma_3(S^{(k)} + S^{(-k)}) \quad (6)$$

$$\dot{A} = -(\Gamma + i\Delta_2)A - iF_+S^{(1)} + iF_-S^{(-1)*} - iF_0S_3^{(1)}, \quad (7)$$

where $A = \langle a \rangle$, the dressed-atoms–cavity coupling strengths are given by $F_\pm = (C/4)(1 \pm \cos 2\alpha)$, $F_0 = (C/4)\sin 2\alpha$ with $C = gN^{1/2}$ (N is the number of active atoms). γ_i, $i = 1, \ldots, 5$ are the damping constants of two-level atoms in the DS basis given by

$$\gamma_0 = \gamma \sin 2\alpha , \quad \gamma_1 = \frac{1}{2}\left[\gamma\left(2 + \sin^2 2\alpha\right) + \gamma_c\left(2 - \sin^2 2\alpha\right)\right] , \quad \gamma_2 = \gamma\left(1 + \cos^2 2\alpha\right) + \gamma_c \sin^2 2\alpha , \quad \gamma_3 = \frac{\gamma - \gamma_c}{2}\sin 2\alpha\cos 2\alpha , \quad \gamma_4 = \frac{\gamma - \gamma_c}{2}\sin^2 2\alpha , \quad (8)$$

while $\bar{S}_3 = -2\gamma\cos(2\alpha)/\gamma_2$ is a stationary DS inversion in the secular approximation.

The approximate method to solve the above infinite hierarchy of equations has been presented in I. For the cavity tuned around ω_L (i.e., Δ_2 of the order of γ_i) effects due solely to one-photon processes may be obtained by neglecting all $S^{(k)}$, $S_3^{(k)}$ for $|k| > 1$. In the systematic improvement one may take into account variables with $|k| \le K$, neglecting those with $|k| > K$, $K = 2, 3, \ldots$.

In the limit of large Ω one may adiabatically eliminate rapidly oscillating polarizations $S^{(k)}$, thus reducing the number of equations to solve (at the price of complicating their structure) [17]. Here, the results of a straightforward numerical integration of Eqs. (5)–(7) are presented; the adiabatic equations lead, for the experimental parameters, to about 10% error for the stationary field strength [17]. The known values of parameters (see Fig. 2 caption) correspond to the experiment [14], the values for Γ and C are chosen to assure lasing. The stationary solutions for $S^{(k)}$ and $S_3^{(k)}$ in the absence of the cavity field A, together with a small value for A, have been chosen as initial conditions for integration [18]. The number of integrated equations has been varied (by increasing K) until the convergence has been obtained.

In the experimental regime ($\Omega_0 \ll \Delta_1$) one might expect that the lasing is dominated by one-photon processes. Amazingly, the convergence required inclusion of the variables up to $K = 3$. This means that not only two-photon but also three-photon amplification processes play a *significant* role.

Figure 2(a) presents the turn on of the laser intensity as a function of time. Note that the curves corresponding to different approximations coincide for low LWI intensity; this regime is dominated by the linear gain at first and then by the one-photon nonlinear processes. As the laser intensity increases, the "one-photon theory" becomes insufficient, the two-photon gain "takes over." At still higher values of laser intensity the two-photon theory becomes unstable with erratic oscillations of intensity while the fully converged three-photon theory (i.e., for $K = 3$) predicts a smooth intensity increase until a stable stationary solution is reached. Note the huge difference between stationary solution values predicted by a one-photon theory and the converged solution.

Inspection of stationary values of different polarizations obtained has shown that the difference between $K = 1$, $K = 2$, and $K = 3$ results is due to relatively large values of polarizations $S^{(k)}$, $k = \pm 2, \pm 3$ (i.e., polarizations describing transitions between unequally populated dressed states). By comparison, polarizations $S_3^{(k)}$, $k = 2, 3$ are practically negligible (despite be-

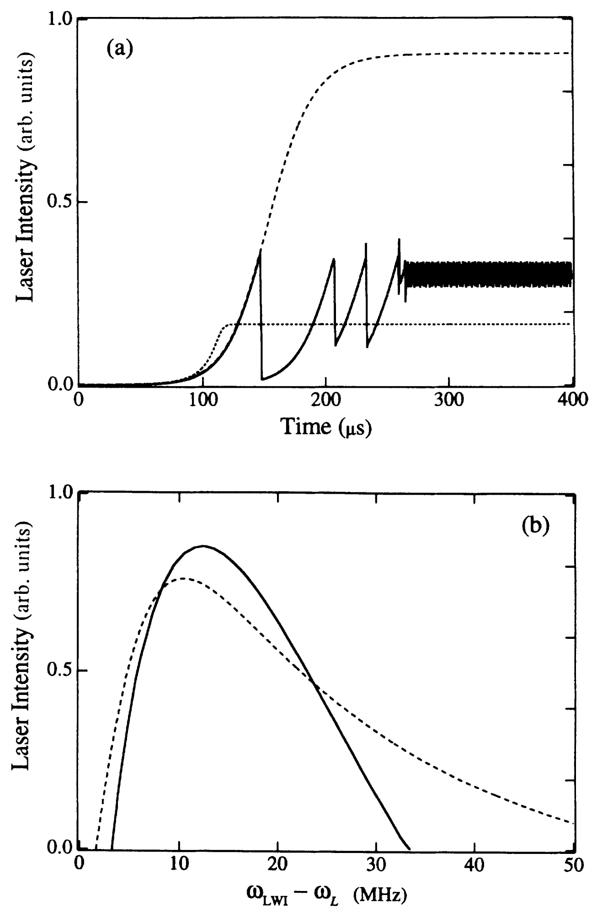

FIG. 2. (a) The turn on of the laser for $\Omega_0 = 400$ MHz, $\Delta_1 = 2$ GHz, $2\gamma = 10$ MHz, $\gamma_c = 20$ MHz, $C \approx 141$ MHz, $\Gamma = 0.05$ MHz, and $\Delta_2 = 30$ MHz. Dotted, solid, and dashed lines correspond to one-photon $K = 1$, $K = 2$, and $K = 3$ (converged) calculations, respectively. (b) The dependence of laser intensity (solid line) on Δ_2; other parameters as in (a); the dashed line shows the linear-gain curve for the same parameters.

ing resonant with the cavity in contrary to strongly off-resonant $S^{(k)}$). This has been further confirmed by forcing $S_3^{(3)} = 0$ at all times. The resulting stationary intensity obtained has been the same as that for exact solution within the accuracy of numerical integrator. Setting $S_3^{(2)} = 0$ at all times affects the final solution by less than 1%. Thus while the initial linear gain is dominated by resonant processes, the nonresonant transitions between unequally populated dressed states become important for the buildup of the laser field in the nonlinear regime. Then one can no longer speak of a single transition in the system studied and the notion of the inversion or the lack of it becomes vague. It is worthwhile to note that this behavior shows some resemblance to the dynamical behavior of the Imamoglu, Field, and Harris [3] laser. In that model an initial lack of inversion is accompanied, for typical values of parameters, with the buildup of inversion at later times (when the lasing mode is sufficiently occupied) [17].

The stationary laser intensity versus its frequency, ω_{LWI} (or rather the frequency difference $\omega_{\mathrm{LWI}} - \omega_L$) is depicted in Fig. 2(b) together with the linear-gain curve for comparison. Due to the frequency pulling effect ω_{LWI} differs from the empty cavity resonance frequency, ω_c. The ω_{LWI} values have been obtained by Fourier-transforming the field amplitude A in the stationary regime, for different input values of Δ_2. The maximum of lasing intensity is slightly shifted with respect to the linear-gain curve. In the experiment [14] a shift of 9 MHz was observed. Much smaller shift obtained in our Doppler-free nonlinear theory provides an indirect confirmation of the interpretation [14] which explains the experimental shift as due to the residual Doppler effect. This effect is also a probable source (the other being an unfortunate choice of Γ and C) of a discrepancy between theoretical and experimental lasing intensity curves at the high Δ_2 side (lasing was observed [14] up to $\Delta_2 = 50$ MHz). Otherwise, the curve predicted [Fig. 2(b)] reproduces remarkably well the experimental data bearing in mind several simplifications of the presented approach (e.g., single-longitudinal-mode approximation).

III. CONCLUSIONS

To conclude, a fully dynamical theoretical description of the first, experimentally observed lasing without inversion has been presented. It shows that the nature of lasing in the arrangement of Ref. [14] is quite complicated; in particular, two- and even three-photon amplification processes play surprisingly a big role in the buildup and maintaining of the lasing. The results indicate again the well-known, albeit sometimes forgotten truth, namely that the laser is an intrinsically nonlinear device and the presence of linear gain is neither the necessary (recall the two-photon DS laser) nor the sufficient (as losses may be bigger) condition for the laser to work.

Finally, it is worth stressing that the laser discussed here appears as a LWI in either pure atomic basis or the dressed-state basis, i.e., in both bases which are suggested by the form of the Hamiltonian (2). This, of course, does not constitute a proof that there does not exist a basis which allows for interpretation of the lasing as occurring in the inverted medium. Rather, it shows the difference in the origin of gain between the system studied and the DS lasers [8, 10] or the double-lambda model [2, 4, 5]. In the case studied here the initial linear gain, as mentioned in the Introduction, may be traced back to the coherence induced by the nonsecular terms in the spontaneous emission in the DS picture. The present model shares this property with both the Imamoglu-Field-Harris laser [3] and with resonantly driven Mollow sideband laser [19].

The important difference between the laser discussed here (or also the DS lasers [8]) and multilevel "laser-without-inversion" schemes [1–5] is that the latter allow for the generation of laser action on a frequency which may be, in principle, very different from that of the pumping light. Thus these schemes are of great interest for practical applications; they may allow one to generate laser radiation in the frequency ranges unaccesible presently by current lasers. Experimental verification of these schemes is still, therefore, one of the big challenges of quantum optics, particularly for large pump-transition–lasing-transition frequency mismatch.

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