Excitation of laser working medium by light from an auxiliary subpoisson laser

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It is shown that the rigorously prescribed pumping claimed in Refs. 1 and 2 to be needed to initiate subpoisson lasing is in fact unnecessary. It suffices to suppress only the low-frequency excitation noise; this can be done by exciting the active medium by an auxiliary subpoisson laser.

1. INTRODUCTION

If the atoms on the active energy levels of a laser medium are regularly excited without fluctuations, such a laser is capable of emitting radiation with "good" quantum properties.[1,2] It is of interest to know whether the favorable conditions are preserved if exciting light comes from an auxiliary subpoisson laser of the type, e.g., discussed in Ref. 1. The point is the photon flux from such a laser is not very likely to become misadjusted. If we begin to count the number of photons passing through the cross section of a light beam per unit time, we observe that this number fluctuates almost in accordance with the Poisson law, deviating from it by a very small factor, e.g., 10^{-10}. Clearly, such a photon flux can at best populate the working level of an atom, with atom-number fluctuations barely smaller than in a Poisson process. It is therefore clear beforehand that the initial conditions of Refs. 1 and 2 are not met, meaning that we cannot extend the physical conclusions of these studies to our conditions. There are nevertheless grounds for assuming that we can expect success also in this case. Recall that Refs. 1 and 2 treat, in final analysis, only suppressed low-frequency photon noise. It is possible perhaps that there is no need then to strive for strictly regular excitation of active atoms, but only atoms with suppressed low-frequence fluctuations. This can be accomplished, in principle, by excitation with light from a subpoisson laser. The qualitative aspect of the problem is here perhaps clear, and we plan to elucidate the quantitative aspect below.

To come closer to a real situation, we assume here that the atoms are excited to active levels from the ground state with a definite number of atoms, and from an intermediate state that is quite randomly populated by some natural processes.

2. EXCITING LASER LIGHT

We take the source of the exciting light to be a laser of the same type as in Ref. 1. Let us recall the main details of this theory, since they will be used in the exposition that follows. The medium consists of four-level atoms, and all N atoms are initially in the ground state $|0\rangle_{\rm I}$ (see Fig. 1a). Owing to the powerful and sufficiently short light pulse at resonance with the $|0\rangle_{\rm I} \to |3\rangle_{\rm I}$ transition, and to rapid relaxation $|3\rangle_{\rm I} \to |2\rangle_{\rm I}$, all N atoms are thrown at some instant of time to an upper laser level $|2\rangle_{\rm I}$. The energy of these atoms is converted next into the energy of N photons of lasing on the transition $|1\rangle_{\rm I} \to |2\rangle_{\rm I}$ and the relaxation at a rate $\gamma_2^{(1)}$ to the initial ground state. This is followed by a new excitation pulse, etc. By suitable choice of the temporal parameters we can ensure subpoisson lasing in this case. The energy scheme of the exciting laser is shown in Fig. 1a.

If radiation from such a laser is directed to a photocathode and the photocurrent spectrum is tracked, the theory predicts the following:

$$i_\omega^{(2)} = i_{\rm shot}^{(2)} \left[1 - q \frac{C_{\rm I}^2}{C_{\rm I}^2 + \omega^2} \right]. \qquad (1)$$

The first term here is the shot noise of the photocurrent, and the second is the excess noise which turns out in our conditions to be negative and suppresses the shot noise at near-zero frequencies. $C_{\rm I}$ is the cavity width of the exciting laser, and q is the quantum efficiency of the photocathode.

The equation for the lasing-field density matrix ρ can be written in the form[3]

$$\dot\rho = r_{\rm I}(\hat L_b - \tfrac{1}{2} \hat L_b^2)\rho - C_{\rm I} \hat R_b \rho. \qquad (2)$$

The first term on the right, proportional to the average excitation rate $r_{\rm I}$ of the state $|2\rangle_{\rm I}$ is determined by the active laser medium, and the second term is determined by the resonance properties of the optical cavity. The operator $\hat L_b$ is given by

$$\hat L_b + 1 = 2\underset{\rightarrow}{b^+}\underset{\leftarrow}{b}[\underset{\rightarrow}{b}b^+ + \underset{\leftarrow}{b}b^+ + \tfrac{1}{2}\beta_3(\underset{\rightarrow}{b}b^+ - \underset{\leftarrow}{b^+})]^{-1}, \qquad (3)$$

b and b^+ are the photon annihilation and creation operators in the laser mode:

$$[b, b^+] = 1.$$

An expression for β_3 is given in the Appendix [Eq. (A13)]. The arrows under the operators indicate the side from which these operators act on the expression to their right. The operator

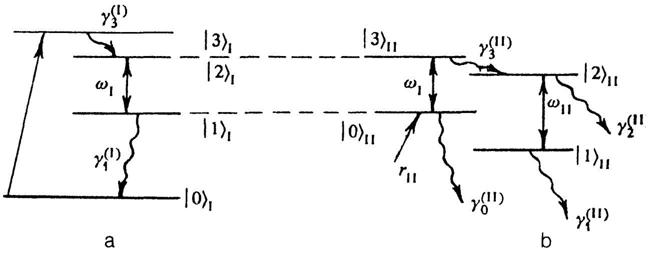

FIG. 1.

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$$\hat{R}_b = \underset{\rightarrow\leftarrow}{b\,b^+} - \frac{1}{2}(\underset{\rightarrow}{b^+}b + b^+\underset{\leftarrow}{b}) \tag{4}$$

determines the damping of the field oscillator. The operator $\hat{L}_b^2$ in (2) appears under the condition that the excitation to the working level is regular. Under Poisson excitation of the atoms, this term is simply absent.

The theory above leads to an explicit expression for the photocurrent spectrum, which takes in the single-mode approximation the following form:[1]

$$i_\omega^{(2)} = i_{\text{shot}}^{(2)}\left[1 + \frac{2q}{n_\text{I}}\operatorname{Re}\int_0^\infty g_\text{I}(t)e^{i\omega t}\right], \tag{5}$$

where

$$n_\text{I} = \langle b^+ b\rangle, \quad g_\text{I}(t) = \langle b^+ b^+(t) b(t) b\rangle, \quad b \equiv b(0). \tag{6}$$

Equation (5) can be recalculated into Eq. (1).

We shall hereafter designate as primary the laser whose emission is used to excite the active medium of the secondary laser.

3. SECONDARY-LASER ACTIVE MEDIUM AND SCHEME FOR ITS EXCITATION

We shall assume that the secondary laser is placed inside the primary one, as shown in Fig. 2. The emission of the primary (exciting) laser is directed to the right, and that of the secondary (excited) to the left. We assume that the mirrors of the secondary laser are transparent to the frequency ω_I. In such a system the primary laser light interacts with two media: with its own medium when the primary lasing is produced, and with the medium of the secondary when the latter is excited. At the same time, the secondary lasing interacts only with its own medium. This asymmetry can be achieved, for example, if the frequencies ω_I and ω_II differ greatly.

The energy structure of the secondary-laser working atoms is shown in Fig. 1b. It is assumed that the state $|0\rangle_\text{II}$ is populated incoherently and perfectly randomly via some excitation mechanism in the medium, and all the remaining states $|1\rangle_\text{II}$, $|2\rangle_\text{II}$, $|3\rangle_\text{II}$ are empty. The frequency of the transition $|3\rangle_\text{II} \leftrightarrow |0\rangle_\text{II}$ agrees with the frequency of the primary lasing and with the frequency of the atomic $|1\rangle_\text{I} \leftrightarrow |2\rangle_\text{I}$ transition. The primary lasing acts therefore on this transition, and the atoms are produced on the upper laser level $|2\rangle_\text{II}$. Secondary lasing sets in when the threshold condition is reached.

We attempt now to explain why this particular scheme of exciting the secondary laser is chosen. Firstly, it makes it easy enough to generalize the single-mode laser mathematical scheme from the preceding section to include our case. We can now solve a two-dimensional laser problem with allowance for the fact that the wave of frequency ω_I interacts with the primary and the excited media, and the wave of frequency ω_II interacts only with the latter. Secondly, getting ahead of ourselves, we state that the conditions are best for us when the primary wave is effectively used by secondary medium. Thus, had we decided to excite the secondary laser using the primary as the external one, we would need an optically thick layer of the secondary medium. On passing through such a layer, however, the quantum properties of the primary light vanish rapidly, so that such a scheme cannot satisfy our requirements.

4. BASIC KINETIC EQUATION

A derivation of the kinetic equation for the density matrix of the electromagnetic lasing field is given in the Appendix. The procedure is exactly the same as used by Lamb and Scully.[4] In the general case this is a rather complicated but readily analyzable equation. We however, will be interested here not in the general case but in the simplest case which is at the same time of greatest interest for us. Firstly, we put $\gamma_2^{(\text{II})} = 0$. Clearly, spontaneous radiation from the upper working level to the extraneous introduces an additional uncertainty in the number of lasing photons. We eliminate this uncertainty by putting $\gamma_2^{(\text{II})} = 0$. Secondly, we shall assume that no nonlinear processes are induced by the interaction of the primary radiation with the secondary region. We shall assume that in Eqs. (A15) and (A16)

$$(\beta_0 + \beta_3)n_\text{I} \ll 1$$

[see Eq. (A13)] and confine ourselves to linear term in Eqs. (A15) and (A16). This means that the primary radiation performs only one function, namely, it excites an atom from the state $|0\rangle_\text{II}$ into the state $|3\rangle_\text{II}$. The basic kinetic equation is noticeably simplified under these assumptions and can be written in the form

$$\dot{\rho} = r_\text{I}(\hat{L}_b - \frac{1}{2}\hat{L}_b^2)\rho - (C_\text{I} + r_\text{II}\beta_0)\hat{R}_b\rho + r_\text{II}\beta_0\hat{L}_a \underset{\rightarrow\leftarrow}{b\,b^+}\rho - C_\text{II}\hat{R}_a\rho. \tag{7}$$

The operators $\hat{R}_a$ and $\hat{L}_a$ are obtained from $\hat{R}_b$ and $\hat{L}_b$ [see (3) and (4)] by replacing b and b^+ by the secondary-lasing annihilation and creation operators a and a^+. r_II is the aver-

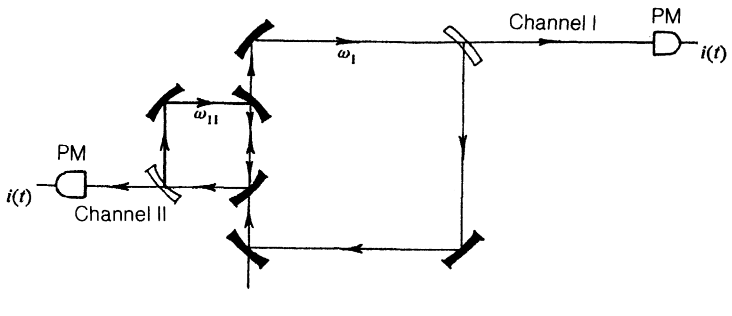

FIG. 2.

age rate of excitation of the level $|0\rangle_{\rm II}$, $C_{\rm II}$ is the damping rate of the secondary lasing from the optical cavity of the secondary laser, and an expression for β_0 is given in the Appendix. Simply taking the trace over the secondary lasing it is easy to obtain from (7) an equation for the primary lasing. It differs from Eq. (2) only in that the field damping rate $C_{\rm I}$ is replaced by $C_{\rm I} + r_{\rm II}\beta_0$. If the primary radiation is detected, we obtain in lieu of the initial Eq. (1)

$$i_\omega^{(2)} = i_{\rm shot}^{(2)}\left[1 - q\frac{C_{\rm I}(C_{\rm I} + r_{\rm II}\beta_0)}{(C_{\rm I} + r_{\rm II}\beta_0)^2 + \omega^2}\right]. \quad (8)$$

Thus, if the additional loss to absorption in the secondary medium is small, i.e., $r_{\rm II}\beta_0 \ll C_{\rm I}$, the situation for the primary radiation remains unchanged: if the shot noise was suppressed to zero, the suppression is preserved. In the other limiting case $r_{\rm II}\beta_0 \gg C_{\rm I}$, the emission of the primary laser will have Poisson statistics without a noticeable suppression of the shot noise.

To analyze the secondary lasing[1)] we go over in Eqs. (7) to a diagonal representation of the density matrix. According to Glauber[5] this can be introduced by the integral equation

$$\rho(t) = \int d^2\alpha \int d^2\beta P(\alpha, \beta, t)|\alpha, \beta\rangle\langle\alpha, \beta|,$$
$$|\alpha, \beta\rangle = |\alpha\rangle|\beta\rangle, \quad a|\alpha\rangle = \alpha|\alpha\rangle, \quad b|\beta\rangle = \beta|\beta\rangle. \quad (9)$$

To change to an equation in the diagonal representation, we must make the following changes:

$$\underset{\rightarrow}{a}, \underset{\rightarrow}{b} \to \alpha, \beta,$$
$$\underset{\leftarrow}{a^+}, \underset{\leftarrow}{b^+} \to \alpha^*, \beta^*,$$
$$\underset{\leftarrow}{a}, \underset{\leftarrow}{b} \to \left(\alpha - \frac{\partial}{\partial\alpha^*}\right), \quad \left(\beta - \frac{\partial}{\partial\beta^*}\right),$$
$$\underset{\rightarrow}{a^+}, \underset{\rightarrow}{b^+} \to \left(\alpha^* - \frac{\partial}{\partial\alpha}\right), \quad \left(\beta^* - \frac{\partial}{\partial\beta}\right). \quad (10)$$

It is easily seen that this results in a partial differential equation which is, generally speaking, of infinite order in the complex amplitudes α and β. Since we are dealing with quantum (subpoisson) light, there is no need to confine ourselves to the lowest order, as in the case of classical light for which the diffusion approximation is always very good. In our specific problem the simplification is effected by a transition to the approximation of small relative fluctuations of the photon number:

$$|\alpha|^2 = n_{\rm II} + \varepsilon, \quad \varepsilon \ll n_{\rm II},$$
$$|\beta|^2 = n_{\rm I} + \mu, \quad \mu \ll n_{\rm I}. \quad (11)$$

For the amplitude density matrix

$$R(\varepsilon, \mu, t) = \int d\varphi \int d\psi P(\alpha, \beta, t), \quad (12)$$

where φ and ψ are the phases of the complex amplitudes α and β we obtain the following equation:

$$\frac{\partial R}{\partial t} = C_{\rm II}\frac{\partial}{\partial\varepsilon}\left[\left(\varepsilon - \frac{n_{\rm II}}{n_{\rm I}}\mu\right)R\right] + (C_{\rm I} + r_{\rm II}\beta_0)\frac{\partial}{\partial\mu}(\mu R)$$
$$- \frac{1}{2}(C_{\rm I} + r_{\rm II}\beta_0)n_{\rm I}\frac{\partial^2 R}{\partial\mu^2}. \quad (13)$$

To write down this equation it was necessary to use the quasiclassical-approach equations

$$\dot n_{\rm I} = r_{\rm I} - \left(C_{\rm I} + \frac{r_{\rm II}\beta_0}{1 + (\beta_0 + \beta_3)n_{\rm I}}\right)n_{\rm I},$$
$$\dot n_{\rm II} = \left(\frac{r_{\rm II}\beta_0\beta_2}{1 + (\beta_1 + \beta_2)n_{\rm II}}n_{\rm I} - C_{\rm II}\right)n_{\rm II},$$

which under our conditions $(\beta_0 + \beta_3)n_{\rm I} \ll 1$ and $\gamma_2 = 0(\beta_2 \to \infty)$ take a simpler form (see the Appendix for the expressions for β_1 and β_2)

$$\dot n_{\rm I} = r_{\rm I} - (C_{\rm I} + r_{\rm II}\beta_0)n_{\rm I},$$
$$\dot n_{\rm II} = r_{\rm II}\beta_0 n_{\rm I} - C_{\rm II}n_{\rm II}.$$

The conditions for stationary lasing are obtained from the requirements $\dot n_{\rm I} = \dot n_{\rm II} = 0$

$$\frac{n_{\rm I}}{n_{\rm II}} = \frac{C_{\rm II}}{r_{\rm II}\beta_0}, \quad n_{\rm I} = \frac{r_{\rm I}}{C_{\rm I} + r_{\rm II}\beta_0}.$$

We emphasize once more that Eq. (13) was obtained without using the diffusion approximation. In this sense, this equation is exact.

5. SECONDARY-LASING PHOTOCURRENT SPECTRUM

The general equation for the photocurrent spectrum in the case of secondary lasing will, of course, be the same as for primary lasing: it is only necessary to replace in (5) $n_{\rm I}$ and $g_{\rm I}$ by $n_{\rm II}$ and $g_{\rm II}$, where

$$n_{\rm II} = \langle a^+a\rangle, \quad g_{\rm II}(t) = \langle a^+a^+(t)a(t)a\rangle, \quad a \equiv a(0). \quad (14)$$

We rewrite $g_{\rm II}$ in the diagonal representation

$$g_{\rm II}(t) = \langle|\alpha(t)|^2|\alpha(0)|^2\rangle = n_{\rm II}^2 + \langle\varepsilon\varepsilon(t)\rangle, \quad \varepsilon \equiv \varepsilon(0). \quad (15)$$

Evidently, to express the spectrum in explicit form, we must find the mean value $\langle\varepsilon\varepsilon(t)\rangle$, which can be represented in the form[5]

$$\langle\varepsilon\varepsilon(t)\rangle = \int\int d\varepsilon_1 d\varepsilon_2 \int d\mu_1 d\mu_2 \varepsilon_1\varepsilon_2 R(\varepsilon_1, \mu_1, t=0)$$
$$\times G(\varepsilon_1, \mu_1, t=0|\varepsilon_2\mu_2 t), \quad (16)$$

where $R(\varepsilon,\mu,t)$ is the physical solution of the problem (13), and $G(12)$ is the solution of the same problem under the condition

$$G(\varepsilon_1\mu_1 t_1|\varepsilon_2\mu_2 t_1) = \delta(\varepsilon_1 - \varepsilon_2)\delta(\mu_1 - \mu_2). \quad (17)$$

Using (13) and (16) we readily obtain an equation for the sought mean value in the form:

$$\frac{d}{dt}\langle\varepsilon\varepsilon(t)\rangle = -C_{\rm II}\langle\varepsilon\varepsilon(t)\rangle + \frac{n_{\rm II}}{n_{\rm I}}C_{\rm II}\langle\varepsilon\mu(t)\rangle. \quad (18)$$

To complete the problem, we need an equation for $\langle\varepsilon\mu(t)\rangle$, which can be obtained quite similarly:

$$\frac{d}{dt}\langle\varepsilon\mu(t)\rangle = -(C_{\rm I} + r_{\rm II}\beta_0)\langle\varepsilon\mu(t)\rangle. \quad (19)$$

The set of differential equations (18) and (19) is easy to solve. The solution is

$$\langle\varepsilon\varepsilon(t)\rangle = \langle\varepsilon^2\rangle\exp(-C_{II}t)$$

$$+ \langle\varepsilon\mu\rangle \frac{n_{II}}{n_I} \frac{C_{II}}{C_{II} - C_I - r_{II}\beta_0} \{\exp[-(C_I + r_{II}\beta_0)t]\exp(-C_{II}t)\}. \quad (20)$$

It is not the final form, since the mean values $\langle\varepsilon^2\rangle$ and $\langle\varepsilon\mu\rangle$ have not been determined. They can be likewise obtained with the aid of Eq. (13), which leads to the system

$$\frac{d}{dt}\langle\varepsilon^2\rangle = -2C_{II}\langle\varepsilon^2\rangle + 2C_{II}\frac{n_{II}}{n_I}\langle\varepsilon\mu\rangle = 0,$$

$$\frac{d}{dt}\langle\varepsilon\mu\rangle = -(C_{II} + C_I + r_{II}\beta_0)\langle\varepsilon\mu\rangle + C_{II}\frac{n_{II}}{n_I}\langle\mu^2\rangle = 0, \quad (21)$$

$$\frac{d}{dt}\langle\mu^2\rangle = -2(C_I + r_{II}\beta_0)\langle\mu^2\rangle - (C_I + r_{II}\beta_0)n_I = 0.$$

We have equated to zero all the derivatives, since we are interested only in stationary solutions. Solving the resultant algebraic set of equations, we obtain ultimately

$$\langle\varepsilon\varepsilon(t)\rangle = \xi_{II}n_{II}\frac{1}{C_{II} - C_I - r_{II}\beta_0}$$

$$\times \{C_{II}\exp[-(C_I + r_{II}\beta_0)t] - (C_I + r_{II}\beta_0)\exp(-C_{II}t)\}. \quad (22)$$

Here

$$\xi_{II} = -\frac{1}{2}\frac{r_{II}\beta_0}{C_{II} + C_I + r_{II}\beta_0} \quad (23)$$

is the Mandel parameter that defines the integral photon fluctuations

$$\Delta n_{II}^2 = n_{II}(1 + \xi_{II}).$$

The effective coupling of the primary and secondary lasers depends on the parameter $r_{II}\beta_0$, which is physically the coefficient of absorption of the primary radiation by the secondary medium in the absence of lasing. The greater the absorptivity, the higher the populations of the working levels of the secondary lasers. If the coupling is very ineffective and $r_{II}\beta_0 \ll C_I, C_{II}$, it is readily seen that the secondary lasing will have only a very small quantum effect, $\zeta_{II} \ll 1$, remaining practically of the Poisson type. In the other limiting case of effective excitation of the working medium we have $r_{II}\beta_0 \gg C_I, C_{II}$, the shot noise of the second lasing effectively vanish in the frequency region $\omega < C_{II}$ are effectively suppressed ($\zeta_{II} = -1/2$), just as in the primary lasing prior to inclusion of the secondary medium. The equation for the photocurrent spectrum will coincide with Eq. (1) with C_I replaced by C_{II}, since

$$\langle\varepsilon\varepsilon(t)\rangle = -\frac{1}{2}n_I\exp(-C_{II}t).$$

6. CONCLUSION

It can be concluded from the foregoing that excitation of a lasing medium by an auxiliary subpoisson laser leads to subpoisson lasing, and if the shot noise is completely suppressed in the exciting laser it remains completely suppressed at zero frequencies. The reason is that the atoms on the working levels deviate very little from having a Poisson distribution. The decisive role if played here by the fact that since there is no low-frequency noise in the exciting light, there is none in the atom excitation itself. This apparently ensures absence of low-frequency noise in the lasing itself.

Note the difference between the case considered here and the one discussed, for example, by Kolobov *et al.* (private communication).[2)] They stipulated the feasibility of both perfectly random and regular pumping of the working atoms on the laser levels. The theory involves a certain parameter $0 \leqslant p \leqslant 1$ that defines the degree of deviation of the pump from regular. At the same time, however, the source of the noise in the populations of the atomic level is regarded as having a "white" spectrum. In our case, however, the population noise is governed by the light of the auxiliary laser, and by virtue of our requirements is not spectrally white, having suppressed low-frequency components. Hence the different results: in our case, although $p < 1$ (using terminology of Kolobov *et al.*), the suppression of the shot noise is nonetheless complete, whereas in their private communication it is only partial and depends on the extent to which p differs from unity.

APPENDIX

Basic kinetic equation

The two-mode lasing equation described in the main text will be derived in the spirit of a paper by Lamb and Scully.[4] The lasing density matrix $\rho(t)$ is altered because one lasing mode interacts with two media—with one during the primary lasing and with the second by its action on the secondary laser. The second mode (secondary lasing wave) interacts with the secondary medium, and both waves attenuate from their optical cavities:

$$\dot\rho = (\dot\rho)_I + (\dot\rho)_{II} + (\dot\rho)_{\rm damp}. \quad (A1)$$

The damping terms take the well known form

$$(\dot\rho)_{\rm damp} = -C_I\hat R_b\rho - C_{II}\hat R_a\rho, \quad (A2)$$

where

$$\hat R_a = \underset{\rightarrow}{a}\,\underset{\leftarrow}{a^+} - \frac{1}{2}(\underset{\rightarrow}{a^+a} + \underset{\leftarrow}{a^+a}),$$
$$\hat R_b = \underset{\rightarrow}{b}\,\underset{\leftarrow}{b^+} + \frac{1}{2}(\underset{\rightarrow}{b^+b} + \underset{\leftarrow}{b^+b}), \quad (A3)$$

b^+ and b are the creation and annihilation operators of the primary-lasing photons, a^+ and a are the same for the secondary lasing:

$$[a, a^+] = [b, b^+] = 1.$$

The term $(\dot\rho)_I$ is connected with the alteration of the lasing field by the interaction with primary medium and can be written, in accordance with Ref. 3, in the form

$$(\dot\rho)_I = r_I(\hat L_b - \frac{1}{2}\hat L_b^2)\rho, \quad (A4)$$

where the operator $\hat L_b$ is given by

$$\hat L_b + 1 = 2\underset{\rightarrow}{b^+}\underset{\leftarrow}{b}[\underset{\rightarrow}{bb^+} + \underset{\leftarrow}{bb^+} + \frac{1}{2}\beta_3(\underset{\rightarrow}{bb^+} - \underset{\leftarrow}{bb^+})^2]^{-1}, \quad (A5)$$

while the term $\hat L_b^2$ appears when the working atoms are purposefully excited to an upper laser level, and is zero if the excitation is random. The quantity β_3 is a certain nonlinear parameter which will be explicitly written out below together with others. The arrows below the operators show the direction from which these operators should act on the expression to the right of them.

We now derive the term $(\dot{\rho})_{\rm II}$, which is determined by the interaction between the two-mode lasing and the secondary medium, the energy structure of which is shown in Fig. 1b. The increment of the field density matrix within a time T through interaction with an atom excited at the instant t_0 to a level $|0\rangle_{\rm II}$ takes the form

$$\rho(t_0+T)-\rho(t_0)=\sigma_0+\sigma_1+\sigma_2-F_{00}(t_0). \tag{A6}$$

The definitions of σ are

$$\sigma_i=\int_{t_0}^{t_0+T}\gamma_i^{(\rm II)}F_{ii}(t')dt',\qquad i=0,1,2,3,$$
$$\sigma_{ik}=\int_{t_0}^{t_0+T}\gamma_{ik}^{(\rm II)}F_{ik}(t')dt',\qquad ik=03,\,30,\,12,\,21. \tag{A7}$$

Here F_{ik} is the density matrix of the lasing field and a single atoms excited initially from a level $|0\rangle_{\rm II}$, and the relaxation constants $\gamma^{(\rm II)}$ are defined in Fig. 1b. For the matrix elements F_{ik} (these are field-variable operators) we can write the system of equations

$$\begin{aligned}
\dot F_{00}&=-\gamma_0F_{00}+ig^*F_{03}b-igb^+F_{30},\\
\dot F_{11}&=-\gamma_1F_{11}-i\bar g a^+F_{21}+i\bar g^+F_{12}a,\\
\dot F_{22}&=-\gamma_2F_{22}+\gamma_3F_{33}-i\bar g aF_{12}+i\bar g F_{21}a^+,\\
\dot F_{33}&=-\gamma_3F_{33}-ig^*bF_{03}+igF_{30}b^+,\\
\dot F_{03}&=-\gamma_{03}F_{03}-igb^+F_{33}+igF_{00}b^+,\\
\dot F_{30}&=-\gamma_{03}F_{30}-ig^*bF_{00}+ig^*F_{33}b,\\
\dot F_{12}&=-\gamma_{12}F_{12}-i\bar g a^+F_{22}+i\bar g F_{11}a^+,\\
\dot F_{21}&=-\gamma_{12}F_{21}-i\bar g^*aF_{11}+i\bar g^*F_{22}a.
\end{aligned} \tag{A8}$$

We integrate these equations from the right and from the left from t_0 to t_0+T for $T\gg\gamma^{-1}$:

$$\begin{aligned}
-F_{aa}(t_0)&=-\sigma_0+\frac{ig^*}{\gamma_{03}}\sigma_{03}b-\frac{ig}{\gamma_{03}}b^+\sigma_{30},\\
0&=-\sigma_1-\frac{i\bar g}{\gamma_{12}}a^+\sigma_{21}+\frac{i\bar g^*}{\gamma_{12}}\sigma_{12}a,\\
0&=-\sigma_2+\sigma_3-\frac{i\bar g^*}{\gamma_{12}}a\sigma_{12}+\frac{i\bar g}{\gamma_{12}}\sigma_{21}a^+,\\
0&=-\sigma_3-\frac{ig^*}{\gamma_{03}}b\sigma_{03}+\frac{ig}{\gamma_{03}}\sigma_{30}b^+,\\
0&=-\sigma_{03}-\frac{ig}{\gamma_3}b^+\sigma_3+\frac{ig}{\gamma_0}\sigma_0b^+,\\
0&=-\sigma_{30}-\frac{ig^*}{\gamma_0}b\sigma_0+\frac{ig^*}{\gamma_3}\sigma_3b,\\
0&=-\sigma_{12}-\frac{i\bar g}{\gamma_2}a^+\sigma_2+\frac{i\bar g}{\gamma_1}\sigma_1a^+,\\
0&=-\sigma_{21}-\frac{i\bar g^*}{\gamma_1}a\sigma_1+\frac{i\bar g^*}{\gamma_2}\sigma_2a.
\end{aligned} \tag{A9}$$

We have omitted the relaxation-constant superscript II that relates them to the secondary medium. The constants of the dipole interaction of the atom with the field is

$$\begin{aligned}
g&=-i\left(\frac{\omega_{\rm I}}{2L^3}\right)^{1/2}d_{03}^{(\rm II)}\exp(i\mathbf{k}_{\rm I}\mathbf{r}),\\
\bar g&=-i\left(\frac{\omega_{\rm II}}{2L^3}\right)^{1/2}d_{12}^{(\rm II)}\exp(i\mathbf{k}_{\rm II}\mathbf{r}).
\end{aligned} \tag{A10}$$

The system (A9) can be solved quite readily. It must be remembered throughout that the operators a, b, and σ do not commute with one another. Solving the system, we obtain

$$\begin{aligned}
\sigma_0&=[1+\tfrac12\beta_2(\underset{\rightarrow}{b}b^++\underset{\leftarrow}{b}b^+)]S_{\rm I}^{-1}\rho(t_0),\\
\sigma_1&=\beta_2\beta_0\underset{\rightarrow}{a^+}\underset{\leftarrow}{a}S_{\rm II}^{-1}\underset{\rightarrow}{b}\underset{\leftarrow}{b^+}S_{\rm I}^{-1}\rho(t_0),\\
\sigma_2&=\beta_0[1+\tfrac12\beta_1(\underset{\rightarrow}{a}a^++\underset{\leftarrow}{a}a^+)]S_{\rm II}^{-1}\underset{\rightarrow}{b}\underset{\leftarrow}{b^+}S_{\rm I}^{-1}\rho(t_0),\\
\sigma_3&=\beta_0\underset{\rightarrow}{b}\underset{\leftarrow}{b^+}S_{\rm I}^{-1}\rho(t_0),
\end{aligned} \tag{A11}$$

where the operators $S_{\rm I}$ and $S_{\rm II}$ are given by

$$S_{\rm I}=1+\frac12(\beta_0+\beta_3)(\underset{\rightarrow}{b}b^++\underset{\leftarrow}{b}b^+)+\frac14\beta_0\beta_3(\underset{\rightarrow}{b}b^+-\underset{\leftarrow}{b}b^+)^2, \tag{A12}$$

$$S_{\rm II}=1+\frac12(\beta_1+\beta_2)(\underset{\rightarrow}{a}a^++\underset{\leftarrow}{a}a^+)+\frac14\beta_1\beta_2(\underset{\rightarrow}{a}a^+-\underset{\leftarrow}{a}a^+)^2.$$

We present now explicit expressions for all the nonlinearity parameters:

$$\begin{aligned}
\beta_0&=\frac{2|g|^2}{\gamma_0^{(\rm II)}\gamma_{03}^{(\rm II)}},\qquad \beta_3=\frac{2|g|^2}{\gamma_3^{(\rm II)}\gamma_{03}^{(\rm II)}},\\
\beta_1&=\frac{2|\bar g|^2}{\gamma_1^{(\rm II)}\gamma_{12}^{(\rm II)}},\qquad \beta_2=\frac{2|\bar g|^2}{\gamma_2^{(\rm II)}\gamma_{12}^{(\rm II)}}.
\end{aligned} \tag{A13}$$

The quantity $(\beta_0+\beta_3)^{-1}$ has the meaning of the number of photons saturating the transition $|0]_{\rm II}\leftrightarrow|3\rangle_{\rm II}$, and accordingly $(\beta_1+\beta_2)^{-1}$ is the number of photons saturating the transition $|1\rangle_{\rm II}\leftrightarrow|2\rangle_{\rm II}$.

Now, using (A6), we can write the increment of the field matrix per atom:

$$\delta\rho=\rho(t_0+T)-\rho(t_0)=\beta_0\hat L\rho(t_0), \tag{A14}$$

$$\begin{aligned}
\hat L=\{&[\underset{\rightarrow}{a^+}\underset{\leftarrow}{a}-\tfrac12(\underset{\rightarrow}{a}a^++\underset{\leftarrow}{a}a^+)-\tfrac14\beta_1(\underset{\rightarrow}{a}a^+-\underset{\leftarrow}{a}a^+)^2]S_{\rm II}^{-1}\underset{\rightarrow}{b}\underset{\leftarrow}{b^+}\\
&+\underset{\rightarrow}{b}\underset{\leftarrow}{b^+}-\tfrac12(\underset{\rightarrow}{b^+}b+\underset{\leftarrow}{b^+}b)-\tfrac14\beta_3(\underset{\rightarrow}{b^+}b-\underset{\leftarrow}{b^+}b)^2\}S_{\rm I}^{-1}.
\end{aligned} \tag{A15}$$

Summing over all the atoms assuming that they are excited to the level $|0\rangle_{\rm II}$ randomly, we obtain the desired equation in the form

$$(\dot\rho)_{\rm II}=r_{\rm II}\beta_0\hat L\rho. \tag{A16}$$

[1)]If primary lasing is regarded as a classical field then the operators b and b^+ in (7) can be replaced by corresponding c-number amplitudes. The result is a closed equation for the secondary lasing similar to Eq. (2). The quantity $r_{II}\beta_0 \underset{\sim}{b}\underset{\sim}{b}^+ \to r_{II}\beta_0 n_{II}$ plays then the role of the average rate of excitation of the upper laser level.

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