Stimulated emission at the difference-frequency generation

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Received: 29 April 1996/Accepted: 17 July 1996

Abstract. We consider the stimulated emission process that occurs at the difference-frequency generation of a bichromatic field interacting with a three-level atom, where one of the laser fields is strong while the other is weak. It is shown that at the difference-frequency generation an induced peak occurs which can exhibit both gain and attenuation. Conditions under which this takes place are established and discussed herein.

PACS: 42.50Hz, 32.80Wt.

Considerable attention has been given recently to the study of lasing without population inversion in the threeand four-level atomic systems [1*—*12]. Atomic coherence effects form the basis of the most commonly discussed schemes for light amplification and lasing without population inversion, which were reviewed recently by Kocharovskaya [13], Scully [14] and Kocharovskaya and Mandel [15]. The observation of light amplification without population inversion has been reported in a number of four-level atomic systems [16*—*19].

It has recently been shown in [20], hereafter referred to as I, that in the low-intensity limit of a bichromatic field interacting with a three-level atom in the lambda configuration as shown in Fig. 1, significant amplification is likely to occur at the difference-frequency generation $\omega = \omega_a - \omega_b$ without population inversion. In Fig. 1, the atom is pumped by two laser fields with frequency modes $\omega_a = \omega_{10} + \Delta_a$ and $\omega_b = \omega_{12} + \Delta_b$, where $\omega_{10} = \omega_a$ $\omega_a = \omega_{10} + \Delta_a$ and $\omega_b = \omega_{12} + \Delta_b$, where $\omega_{10} = \omega_1$
 $\sim \omega_0$ and $\omega_{12} = \omega_1 - \omega_2$ are the transition frequencies, while Δ_a and Δ_b are the detunings of the laser fields *a* and *b*, respectively. The electronic transitions $|0\rangle \leftrightarrow |1\rangle$ and $|1\rangle \leftrightarrow |2\rangle$ are electric-dipole-allowed, while g_a and g_b denote the classical Rabi frequencies for the laser fields *a* and *b*, respectively; units with $h = 1$ are used throughout. It is shown in I that although the spontaneous emission

 $|2\rangle \rightarrow |0\rangle$ is electric-dipole-forbidden and the excited state $|2\rangle$ is a metastable one having a very long lifetime, there is, at low intensities, a two-photon-stimulated emission at $\omega = \omega_a - \omega_b$, whose lifetime is induced by the laser field *a* operating in the $|0\rangle \leftrightarrow |1\rangle$ transition and is much longer than that of excitations spontaneously emitted by the excited state $|1\rangle$ provided that the condition $g_b \gg g_a$ is satisfied. The low-intensity limit for both laser fields occurs when $\gamma_1^2 \gg g_a^2$ and $\gamma_2^2 \gg g_b^2$, which imply that both transitions are not saturated. γ_1 and γ_2 designate the spontaneous emission probabilities for the radiative decay processes $|1\rangle \rightarrow |0\rangle$ and $|1\rangle \rightarrow |2\rangle$, respectively, while $2/\gamma = 2/(\gamma_1 + \gamma_2)$ denotes the spontaneous radiative lifetime of the excited state $|1\rangle$. It is found in I that the intensity of the two-photon-stimulated emission at $\omega = \omega_a - \omega_b$ increases negatively and, consequently, the amplification increases as the value of the ratio g_b/g_a increases for $\Delta_b = 0$ and $\Delta_a \neq 0$. Numerical calculations have shown that for $g_b/g_a \gg 1$, $\Delta_a \neq 0$ and $0 \leq \Delta_b/\Delta_a < 1$, significant amplification is likely to occur at the differencefrequency generation $\omega = \omega_a - \omega_b$. Similar predictions have been made for the process of stimulated emission that occurs in a number of cases when laser fields at low intensities interact with three- and four-level atomic systems [21, 22].

The purpose of the present study is to investigate the high-intensity limit where only one of the two-transitions is saturated, namely, when the conditions $g_b^2 \gg \gamma_2^2$, $g_a^2 \ll \gamma_1^2$ is saturated, namely, when the conditions $g_b \gg \gamma_2$, $g_a \ll \gamma_1$
and $g_b^2 \gg g_a^2$ are satisfied, which imply that laser field *b* operating the transition $|2\rangle \leftrightarrow |1\rangle$, which is saturated, is strong while laser field *a* operating the transition $|0\rangle \leftrightarrow |1\rangle$, which is not saturated, is weak. The induced spectra obtained at this limit would be compared with those derived in I at low intensities, where both laser fields were weak and both transitions were not saturated. Since the present work is an extension of I and for purpose of brevity, all derived expressions in I shall not be repeated herein. Instead, statements of equations from I will be by reference to the numbers of I set forth therein; all notations contained therein are identical to those used in I as well.

The expressions for the Green functions $G_{2,2}(\omega)$, $G_{1b^+}, 2(\omega)$ and $G_{0b^+a}, 2(\omega)$ defined by (5–7) of I, respectively tively, have the same denominator denoted by the function $D_2(\omega)$ and differ only as far as the expressions for the

Issued as NRCC No. 39126

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Fig. 1. Energy-level diagram of the three-level atom or ion

numerators are concerned. In the limit when the conditions $g_b^2 \gg \gamma_2^2$, $g_a^2 \ll \gamma_1^2$ and $g_b^2 \gg g_a^2$ are satisfied, the function $D_2(\omega)$ defined in I may take the form

$$
D_2(\omega) = (X_+ + i\gamma/4) (X_- + i\gamma/4) (X + i\gamma_d/2),
$$
 (1)

where $X_{\pm} = X + A_a - A_b/2 + B/2$, $X = \omega - \omega_a + \omega_b$ $y = y_1 + y_2$, $y_d = y_a/\lambda$, $g^2 = g_b^2 + (\Delta_b + i\gamma/2)^2$, $y_a = g_a^2/\gamma$, $\lambda = 1 + 4\bar{\phi}^2/\gamma^2 A_{ab}^2$, $\phi = A_a A_{ab} - g_b^2/4$ and $A_{ab} = A_a - A_b$. The notation here is identical to that used in I. Substituting (1) into (5*—*7) of I, we expand the numerators in power series of the parameters $\gamma_2^2/g_b^2 \ll 1$, $g_a^2/\gamma_1^2 \ll 1$ and $g_a^2/g_b^2 \ll 1$ at the roots of the corresponding denominators, then we take the imaginary parts of the derived expressions to obtain the spectral functions in the form

$$
I_{2,2}(\omega) = -\left(\frac{2\phi}{\gamma A_{ab}^2}\right) \left(\frac{\gamma_a^2/4 + X\gamma\gamma_d A_{ab}/4\phi}{X^2 + \gamma_d^2/4}\right) + \frac{2}{\gamma} \left(\frac{(1 + A_b/g)\gamma^2/16 - X_+ \gamma^2/8g}{X_+^2 + \gamma^2/16} + \frac{(1 - A_b/g)\gamma^2/16 + X_- \gamma^2/8g}{X_-^2 + \gamma^2/16}\right),
$$
 (2)

$$
I_{1b^+,2}(\omega) = -\left(\frac{2\phi g_b}{\lambda \gamma^2 A_{ab}^2}\right) \left(\frac{\gamma_a^2/4 + X\gamma \gamma_d A_{ab}/4\phi}{X^2 + \gamma_d^2/4}\right)
$$

spectra of an electron in the metastable state $|2\rangle$ while that of (3) represents the induced spectra arising from the physical process where a laser photon *b* is emitted by the excited state $|1\rangle$. The spectral function (4) designates a Raman-type two-photon-induced process where a laser photon *a* is absorbed while a laser photon *b* is emitted simultaneously by the ground state $|0\rangle$ of the atom.

Each right-hand side (r.h.s.) of the spectral functions (2*—*4) consists of two terms, the first of which represents an excitation that is induced by the weak laser field *a*, is an asymmetric Lorentzian line that is peaked at the difference-frequency generation $\omega = \omega_a - \omega_b$, and has a spectral width $\gamma_d/2$. The induced peak vanishes in the absence of the laser field *a*, namely, when $g_a \to 0$ as well as when $\Delta_a = \Delta_b$, namely, when the two-photon resonance condition $\omega_{20} = \omega_a - \omega_b$ is applicable. At frequencies $\omega \neq \omega_a - \omega_b$, the asymmetry of the induced peak depends on the values of the function $\gamma A_{ab}/4\phi$. The second terms on the r.h.s. of the spectral functions (2*—*4) describe the spectra of two sidebands, which are induced by the strong laser field *b*, having asymmetric Lorentzian profiles that are peaked at the frequencies $X_{+} = 0$, namely, at $\omega = \omega_a - \omega_b - A_a + A_b/2 + g/2$, respectively, and have equal spectral widths of the order of $\gamma/4$.

At frequencies $\omega = \omega_a - \omega_b$ and at $X_{\pm} = 0$, the spectral function (3) disappears, namely, I_{1b} ⁺, 2($\omega_a - \omega_b$) = 0 and $I_{1b^2,2}(\omega_a - \omega_b - A_a + A_b/2 \pm g/2) = 0$; this result can be also verified from the original expression defined by (6) of I. Therefore, the spectral function (3) will be ignored since it vanishes in the frequency regime of our interest. Then we may write the total spectral functions (2) and (4) as

$$
I_2(\omega) = I_{2,2}(\omega) + I_{0b^+a,2}(\omega) = I_{\text{ind}}(\omega) + I_+(\omega) + I_-(\omega),
$$
\n(5)\n
$$
I_{\text{ind}}(\omega) = -\left(\frac{2\phi}{\gamma A_{ab}^2}\right) \left(1 - \frac{g_b}{g_a}\right) \left(\frac{\gamma_a^2/4\lambda^2 + X\gamma_d A_{ab}/4\phi}{X^2 + \gamma_d^2/4}\right),
$$
\n(6)

$$
I_{\pm}(\omega) = \frac{2}{\gamma} \left(\frac{(1 \pm A_b/g - g_a g_b P_{\pm}/\lambda \gamma^2 A_{ab}^2) \gamma^2 / 16 \mp \gamma^2 X_{\pm} (1 \pm g_a g_b R_{\pm} g/\gamma^2 A_{ab}^2) / 8g}{X_{\pm}^2 + \gamma^2 / 16} \right). \tag{7}
$$

$$
+\left(\frac{g_b}{2g}\right)\left(\frac{X_+}{X_+^2+\gamma^2/16}-\frac{X_-}{X_-^2+\gamma^2/16}\right),\qquad(3)
$$

$$
I_{0b^+a,2}(\omega) = \left(\frac{2\phi g_b}{\gamma g_a \Delta_{ab}^2}\right) \left(\frac{\gamma_a^2/4 + X\gamma \gamma_d \Delta_{ab}/4\phi}{X^2 + \gamma_d^2/4}\right) - \left(\frac{2g_a g_b}{\lambda \gamma^3 \Delta_{ab}^2}\right) \left(\frac{P_+\gamma^2/16 + \gamma^2 X_+ R_+ / 8}{X_+^2 + \gamma^2/16}\right) + \frac{P_-\gamma^2/16 + \gamma^2 X_- R_- / 8}{X_-^2 + \gamma^2/16}\right),
$$
 (4)

where $I_{2,2}(\omega) = -2\pi \operatorname{Im} \{ G_{2,2}(\omega) \}, I_{1b^+,2}(\omega) = -2\pi \operatorname{Im} \{ G_{2,2}(\omega) \}$ $\{G_{1b^+,2}(\omega)\}, \quad I_{0b^+a,2}(\omega) = -2\pi \operatorname{Im} \{\widetilde{G}_{0b^+a,2}(\omega)\}, \quad P_{\pm} = \phi A_{\pm}$ $\frac{1}{2}\gamma^2 A_{ab}/4g$, $R_{\pm} = A_{\pm} A_{ab} \mp \phi/g$ and $A_{\pm} = 1 \pm (A_a + A_a)$ Δ _{*ab*})/*g*. The spectral function (2) describes the excitation Thus, the spectral function (5) describes a triplet, namely, an induced peak at the difference-frequency generation $\omega = \omega_a - \omega_b$ and two sidebands at the frequencies $\omega = \omega_a - \omega_b - A_a + A_b/2 + g/2$ and $\omega = \omega_a - \omega_b - A_a + A_b/2 - g/2$, respectively. Since $g_b^2 \gg g_a^2$, the multiplicative factor $[1 - g_b/g_a]$ in (6) may be replaced by $[-g_b/g_a]$ indicating that near the frequency $\omega = \omega_a - \omega_b$ the two-photon process described by the first term on the r.h.s. of (4) prevails over the corresponding one of (2).

At the difference-frequency generation $\omega = \omega_a - \omega_b$, the spectral function (6) becomes equal to

$$
I_{\rm ind}(\omega_a - \omega_b) = -\frac{2\phi}{\gamma A_{ab}^2} \left(1 - \frac{g_b}{g_a}\right)
$$

$$
= -\frac{2}{\gamma} \left(1 - \frac{g_b}{g_a} \right) \left(\frac{A_a}{A_{ab}} - \frac{g_b^2}{4A_{ab}^2} \right), \tag{8}
$$

which designates the relative intensity (height) of the induced peak in question. Since $g_b^2 \gg g_a^2$, the relative intensity (height) of the induced peak $I_{ind}(\omega_a - \omega_b)$ takes positive values when $g_b^2 < 4\Delta_a\Delta_{ab}$ and negative ones when $g_b^2 > 4 \Delta_a \Delta_{ab}$, indicating that the physical process of in- duced absorption (attenuation) and stimulated emission (amplification) is likely to occur at $\omega = \omega_a - \omega_b$. The relative intensity $I_{\text{ind}}(\omega_a - \omega_b)$ becomes zero when $g_b^2 = 4A_a A_{ab}$, implying the vanishing of the induced peak. When the weak laser field *a* operating in the $|0\rangle \leftrightarrow |1\rangle$ transition is at resonance, namely, when $\Delta_a = 0$ but $\Delta_b \neq 0$, (8) is reduced to

$$
I_{\text{ind}}(\omega_a - \omega_b) = \frac{1}{2\gamma} \left(\frac{g_b}{A_b}\right)^2 \left(1 - \frac{g_b}{g_a}\right)
$$

$$
\approx -\frac{1}{2\gamma} \left(\frac{g_b}{g_a}\right) \left(\frac{g_b}{A_b}\right)^2 < 0,
$$
(9)

which takes always negative values. In the opposite case when $\Delta_b = 0$ but $\Delta_a \neq 0$, then from (8) we have

$$
I_{\text{ind}}(\omega_a - \omega_b) = \frac{-2}{\gamma} \left(1 - \frac{g_b}{g_a} \right) \left(1 - \frac{g_b^2}{4A_a^2} \right)
$$

$$
\approx \frac{2}{\gamma} \left(\frac{g_b}{g_a} \right) \left(1 - \frac{g_b^2}{4A_a^2} \right),\tag{10}
$$

which takes positive values for $g_b^2 < 4\Delta_a^2$, negative ones for $g_b^2 > 4A_a^2$ and vanishes for $g_b^2 = 4A_a^2$. This behaviour is demonstrated in Figs. 2*—*4, where the induced relative intensity $(g_a/g_b)\gamma I_{\text{ind}}(\omega)$ computed from (6) in units of $(g_b/g_a)/\gamma$ is plotted vs the relative frequency $(\omega - \omega_a + \omega_b)/\gamma$.

 Figures 2 and 3 illustrate the induced two-photon spectra for $g_b^2 \gg g_a^2$, $g_b = 5\gamma$, $\Delta_a = 0$ and for various values of the relative detuning $\eta_b = \Delta_b/\gamma = 1 - 0.125$ and for $\Delta_b = 0$ and $\eta_a = \Delta_a/\gamma = 1 - 0.125$, respectively. Inspection of Fig. 2(3) indicates that for $\Delta_a = 0$ ($\Delta_b = 0$) small values of the relative detuning η_b (η_a) favour the relative intensities (heights) of the induced peaks to take negative values. This implies that as the value of the relative detuning decreases from 1, the relative intensity increases negatively, indicating that stimulated emission (amplification) increases at the two-photon frequency $\omega = \omega_a - \omega_b.$

 Figure 4 illustrates the induced two-photon spectra for $g_b^2 \gg g_a^2$, $g_b = 5\gamma$, $\eta_a = \Delta_a/\gamma = 0.5$ and for various values of the ratio Δ_b/Δ_a . It is shown that the relative intensities (heights) of the induced two-photon increase negatively, provided that Δ_b/Δ_a takes values between zero and one, namely, for $0 \leq \frac{d}{2b}/\Delta_a < 1$. Since smaller values of $\Delta_{ab} = \Delta_a - \Delta_b < 1$ favour higher negative relative inten- sities (heights) for the induced peaks, an inspection of (6) reveals that $\omega \neq \omega_a - \omega_b$, the asymmetry of the spectral lines is negligible and, therefore, the spectral lines in Figs. 2*—*4 take Lorenzian shapes, the relative intensities (heights) of which are determined by (9), (10) and (8), respectively.

Fig. 2. Difference-frequency-generation-induced spectra in the presence of the strong laser field *b* operating in the $|2\rangle \leftrightarrow |1\rangle$ transition. The relative intensity $(g_a/g_b) \gamma I_{\text{ind}}(\omega)$ in units of $(g_b/g_a)/\gamma$ computed from (6) is plotted vs the relative frequency $(\omega - \omega_a + \omega_b)/\gamma_a$ for $\Delta_a = 0$, $g_b = 5\gamma$ and for various values of the relative detunings $\eta_b = \Delta_b/\gamma = 1, 0.5, 0.25, 0.167$ and 0.125, respectively

In conclusion, it is shown that at high intensities for the laser field *b* and at low intensities of the laser field *a*, namely, for $g_b^2 \gg \gamma^2$, $g_a^2 \ll \gamma^2$ and, hence $g_b^2 \gg g_a^2$, the conditions $g_b^2 < 4\Delta_a \Delta_{ab}$ and $g_b^2 > 4\Delta_a \Delta_{ab}$ favour the processes of induced absorption and stimulated emission to occur, respectively, at the frequency $\omega = \omega_a - \omega_b$, while the twophoton-induced peak vanishes when $g_b^2 = 4A_aA_{ab}$. Since $g_b^2 \gg g_a^2$, the relative intensities of the two-photon-induced peaks determined by (8*—*10) are proportional to the ratio g_b/g_a and arise from the two-photon Raman-type process, where a laser photon *b* is emitted while a laser photon *a* is absorbed simultaneously by the ground state $|0\rangle$ of the atom and is described by the first term on the r.h.s. of the spectral function (4). The conditions $g_b > g_a$ and $0 \leq \frac{\Delta_b}{\Delta_a} < 1$ and the fact that the relative intensities of the induced peaks vary proportionally to the ratio $g_b > g_a$ are phenomena identical to those of I at low intensities of both laser fields. The basic difference is that at high intensities of the laser field *b* the appearance of the term $g_b^2/4A_{ab}^2$ shown in (8) and the condition $g_b^2 > 4A_aA_{ab}$ make the occurrence of stimulated emission and, therefore, the amplification at $\omega = \omega_a - \omega_b$ to be more pronounced than at

Fig. 3. As in Fig. 2 but for $\Delta_b = 0$, $g_b = 5\gamma$ and for various values of the relative detunings $\eta_a = A_a / \gamma = 1$, 0.5, 0.25, 0.167 and 0.125, respectively

the low-intensity limit. The disappearance of the induced peak when $g_b^2 = 4A_a A_{ab}$ is a unique property that occurs at high intensities of the laser field *b* as well. The occurrence of the two sidebands at the frequencies $\omega = \omega_a - \omega_b - \Delta_a + \Delta_b/2$ and $\omega = \omega_a - \omega_b - \Delta_b$ cies $\omega = \omega_a - \omega_b - \Delta_a + \Delta_b/2$ and $\omega = \omega_a - \omega_b - \Delta_a$
+ $\Delta_b/2 - g/2$, respectively, is due entirely to the presence of the strong laser field. The difference-frequency-generation approach is of technical importance and provides a means of generating intense tunable radiation in the infrared, like the infrared-visible difference-frequency generation, which is a very attractive experimental method in nonlinear optics [23]. It is hoped that the present work, which is supplemental to I, will stimulate experimental interest in this direction.

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Fig. 4. As in Fig. 2 but for $g_b = 5\gamma$, $\eta_a = \frac{\Delta_a}{\gamma} = 0.5$ and for various values of the ratio $\Delta_b/\Delta_a = 0.70, 0.80, 0.85, 0.90, 0.925$ and 0.95, respectively

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