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Lasers without inversion: Two-photon stimulated emission in a three-level atom

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Although the two-photon spontaneous emission is electric dipole forbidden, there is a two-photon stimulated emission whose lifetime is much longer in comparison to those of excitations spontaneously emitted by the excited states of the atom. Conditions are established under which the two-photon stimulated emission prevails, indicating that significant amplification is likely to occur near the two-photon frequency.

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There has recently been considerable interest in the study of lasers without population inversion [1–12]. In a recent paper Harris [1] has considered the difference between emission and absorption spectra due to Fano interferences [2] between two lifetime-broadened discrete levels which decay into an identical continuum [1–3]. Many schemes for laser action without population inversion have been proposed [4–12].

The present work differs from previous studies [1,3–13] in that we consider the two-photon-stimulated emission which occurs at the excited state $|1\rangle$ of the three-level atom shown in Fig. 1, where the two-photon transition $|0\rangle \leftrightarrow |1\rangle$ is electric dipole forbidden. The atom is pumped by two laser fields operating in the $|0\rangle \leftrightarrow |2\rangle$ and $|2\rangle \leftrightarrow |1\rangle$ transitions which are electric dipole allowed. In the low-intensity limit of both laser fields an excitation is induced by both laser fields at the two-photon frequency, whose lifetime induced by the laser field b operating in the $|0\rangle \leftrightarrow |2\rangle$ transition is long by comparison to those of excitations spontaneously emitted by the excited states. When certain conditions prevail, a splitting occurs in the excitation spectra at the two-photon frequency, where the intensity of the two resolved peaks take negative values indicating that significant amplification is likely to occur at the corresponding frequencies. Since our processes occur at finite frequencies, they are dynamic in origin. Therefore, we have made use of a suitable mathematical formalism, which has recently been used in similar circumstances [14,15].

The Hamiltonian for the atomic system shown in Fig. 1 in the electric dipole and rotating-wave approximation may be taken as

$$H = \omega_{20} a_2^\dagger a_2 + \omega_{10} a_1^\dagger a_1 + \frac{i}{2} g_b [a_0^\dagger a_2 \exp(-i\omega_b t) - a_2^\dagger a_0 \exp(i\omega_b t)] + \frac{i}{2} g_a [a_2^\dagger a_1 \exp(-i\omega_a t) - a_1^\dagger a_2 \exp(i\omega_a t)] + \sum_{\mathbf{k},\lambda} ck \beta_{\mathbf{k}\lambda}^\dagger \beta_{\mathbf{k}\lambda} + \frac{1}{2} i\omega_p \sum_{\substack{\mathbf{k},\lambda \\ i,j}} \left[f_{ij}(\mathbf{k},\lambda) \frac{\omega_{ji}}{ck} \right]^{1/2} (a_i^\dagger a_j \beta_{\mathbf{k}\lambda}^\dagger - a_j^\dagger a_i \beta_{k\lambda}) , \quad (1)$$

where $a_i^\dagger, a_i$ are the usual Fermi-Dirac operators describing the electron states $i = 1$, 2, and 3, and $n_i = a_i^\dagger a_i$ is the number operator. The functions $f_{ij}(\mathbf{k},\lambda)$ are the oscillator strengths for the atomic transitions $|i\rangle \leftrightarrow |j\rangle$ and ω_p is the atomic plasma frequency; units with $\hbar = 1$ are used throughout. The atom is pumped by two laser fields with frequency modes ω_a and ω_b defined as $\omega_a = \omega_{12} + \Delta_a$ and $\omega_b = \omega_{20} + \Delta_b$, where $\omega_{12} = \omega_1 - \omega_2$ and $\omega_{20} = \omega_2 - \omega_0$ are the transition frequencies while Δ_a and Δ_b are the detunings of the laser fields a and b, respectively. The electronic transitions $|0\rangle \leftrightarrow |2\rangle$ and $|2\rangle \leftrightarrow |1\rangle$ are electric dipole allowed while the transition $|0\rangle \leftrightarrow |1\rangle$ is forbidden. g_a and g_b denote the classical Rabi frequencies for the lasers a and b, respectively. The operators $\beta_{\mathbf{k}\lambda}^\dagger, \beta_{\mathbf{k}\lambda}$ describe the vacuum field, which is quantized with wave vector $\mathbf{k}$, frequency ck, and transverse polarization $\lambda = 1,2$. In writing Eq. (1) we have taken into consideration the relation $n_1 + n_2 + n_3 = 1$.

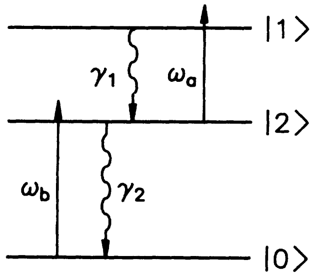

FIG. 1. Energy-level diagram of a three-level atom or ion.

The first two terms on the right-hand side (rhs) of Eq. (1) describe the free atomic fields, while the third and fourth terms denote the interaction of the two atomic transitions with the laser fields a and b, respectively, where the laser fields have been treated classically. The last two terms designate the free vacuum (signal) field and its interaction with the atomic levels, respectively.

The spectral function describing the excitation spectra for an electron in the excited state $|1\rangle$ is determined [16] by the imaginary parts of the Fourier transform of the single-electron Green function $G_{1,1}(\omega) = \langle\langle a_1 ; a_1^\dagger \rangle\rangle$. Using the Hamiltonian (1) we derive the equations of motion [16] for $G_{1,1}(\omega)$:

$$d_1 G_{1,1}(\omega)=\frac{1}{2\pi}-\frac{i}{2}g_a G_{2a,1}(\omega)\ , \tag{2}$$

$$d_{2a}G_{2a,1}(\omega)=\frac{i}{2}g_a G_{1,1}(\omega)-\frac{i}{2}g_b G_{0ab,1}(\omega)\ , \tag{3}$$

$$d_{ab}G_{0ab,1}(\omega)=\frac{i}{2}g_b G_{2a,1}(\omega)\ , \tag{4}$$

where

$$G_{2a,1}(\omega)=\langle\langle a_2\exp(i\omega_a t);a_1^\dagger\rangle\rangle,$$

$$G_{0ab,1}(\omega)=\langle\langle a_0\exp(i\omega_{ab} t);a_1^\dagger\rangle\rangle,$$

$$d_1=\omega-\omega_{10}+i\gamma_1/2,\quad d_{2a}=\omega-\omega_{ab}+\Delta_b+i\gamma_2/2,$$

$$d_{ab}=\omega-\omega_a-\omega_b,\quad \gamma_1=\tfrac{4}{3}(\omega_{12}/c)^3|\mathbf{P}_{21}|^2,$$

$$\gamma_2=\tfrac{4}{3}(\omega_{20}/c)^3|\mathbf{P}_{02}|^2,\omega_{ab}=\omega_a+\omega_b,$$

while $\mathbf{P}_{21}$ and $\mathbf{P}_{02}$ are the transition dipole moments. The functions γ_1 and γ_2 denote the spontaneous emission probabilities for the radiative decays $|1\rangle\rightarrow|2\rangle$ and $|2\rangle\rightarrow|0\rangle$, respectively, while $2/\gamma_1$ and $2/\gamma_2$ define the radiative lifetimes of the corresponding excited states $|1\rangle$ and $|2\rangle$. The solution of Eqs. (2)-(4) yields

$$G_{1,1}(\omega)=(d_{2a}d_{ab}-g_b^2/4)/2\pi D\ , \tag{5}$$

$$G_{2a,1}(\omega)=ig_a d_{ab}/4\pi D\ , \tag{6}$$

$$G_{0ab,1}(\omega)=-g_a g_b/8\pi D\ , \tag{7}$$

where

$$D=d_1 d_{2a}d_{ab}-\frac{g_b^2}{4}d_1-\frac{g_a^2}{4}d_{ab}\ .$$

The Green function $G_{1,1}(\omega)$ describes the excitation spectra of an electron in the excited state $|1\rangle$ while $G_{2a,1}(\omega)$ represents the induced process where a photon of the laser field a is absorbed by the excited state $|2\rangle$. The Green function $G_{0ab,1}(\omega)$ describes the physical process where two photons, namely, one photon from each of the laser fields a and b, are absorbed by the ground state $|0\rangle$ of the atom.

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. At this limit an expansion of Eqs. (5)-(7) into power series of $g_a^2/\gamma_1^2\ll 1$ and $g_b^2/\gamma_2^2\ll 1$ yields

$$G_{1,1}(\omega)=\frac{1}{2\pi}\left[\frac{1}{d_1}+\frac{\gamma_b(\nu-1+2i\eta_b)}{d\gamma_1\lambda_b\lambda_{ab}}\right], \tag{8}$$

$$G_{2a,1}(\omega)=-\frac{g_a\gamma_b(\nu+2im)}{2\pi d\gamma_1\gamma_2\lambda_{ab}\lambda_b^2}\ , \tag{9}$$

$$G_{0ab,1}(\omega)=\frac{g_a g_b(\nu+2im)}{2\pi d\gamma_1\gamma_2\lambda_b\lambda_{ab}}\ , \tag{10}$$

where $d=\omega-\omega_a-\omega_b+i\gamma_b/2\lambda_b$, $\nu=1-4\eta_b\eta_{ab}$, $m=\eta_b+\eta_{ab}$, $\eta_b=\Delta_b/\gamma_2$, $\eta_{ab}=(\Delta_a+\Delta_b)/\gamma_1$, $\gamma_b=g_b^2/\gamma_2$, $\gamma_a=g_a^2/(\gamma_1-\gamma_2)$, $\lambda_b=1+4\eta_b^2$, $\lambda_{ab}=1+4\eta_{ab}^2$, $\lambda_a=1+4\eta_a^2$, and $\eta_a=\Delta_a/(\gamma_1-\gamma_2)$.

Expression (8) represents the excitation spectra of an electron in the excited state $|1\rangle$, where the two terms describe a spontaneous and an induced excitation whose lifetimes are equal to $2/\gamma_1$ and $2\lambda_b/\gamma_b$, respectively. Expressions (9) and (10) describe the absorption of a laser photon a by the excited state $|2\rangle$ and the absorption of two photons, one from each laser field, by the ground state $|0\rangle$, respectively. The last term on the rhs of Eq. (8) as well as Eqs. (9) and (10) represents contributions to the same excitation, which is induced by both laser fields at the two-photon frequency $\omega=\omega_a+\omega_b$ and has a long lifetime $2\lambda_b/\gamma_b$ in comparison to those of $2/\gamma_1$ and $2/\gamma_2$, respectively, that are short in duration, namely, $2\lambda_b/\gamma_b\gg 2/\gamma_1$ and $2\lambda_b/\gamma_b\gg 2/\gamma_2$. The lifetime $2\lambda_b/\gamma_b$ is induced only by the laser field b and, therefore, the induced excitation may be interpreted as arising from the dynamic interference of the laser field b with the induced two-photon spectrum at the frequency $\omega=\omega_a+\omega_b$. In deriving Eqs. (8)-(10), we have discarded terms describing spontaneous processes whose amplitudes of occurrence are of the orders of $g_a^2/\gamma_1^2\ll 1$ and $g_b^2/\gamma_2^2\ll 1$, respectively. We have also made the following approximations: $\gamma_1-\gamma_a/\lambda_a\approx\gamma_1$ and $\gamma_2+\gamma_a/\lambda_a-\gamma_b/\lambda_b\approx\gamma_2$ since at the low-intensity limit $\gamma_1\gg\gamma_a/\lambda_a$ and $\gamma_2\gg(\gamma_a/\lambda_a-\gamma_b/\lambda_b)$ for $\gamma_1>\gamma_2$.

Taking the imaginary parts of Eqs. (8)-(10), we may write the total spectral function $I_1\equiv I_1(\omega)$ for an electron in the excited state $|1\rangle$ as

$$I_1=-2\pi\,\mathrm{Im}[G_{1,1}(\omega)+G_{2a,1}(\omega)+G_{0ab,1}(\omega)]$$
$$=I_1^{\mathrm{sp}}(\omega)+I_{\mathrm{ind}}^{ab}(\omega)\ ,$$

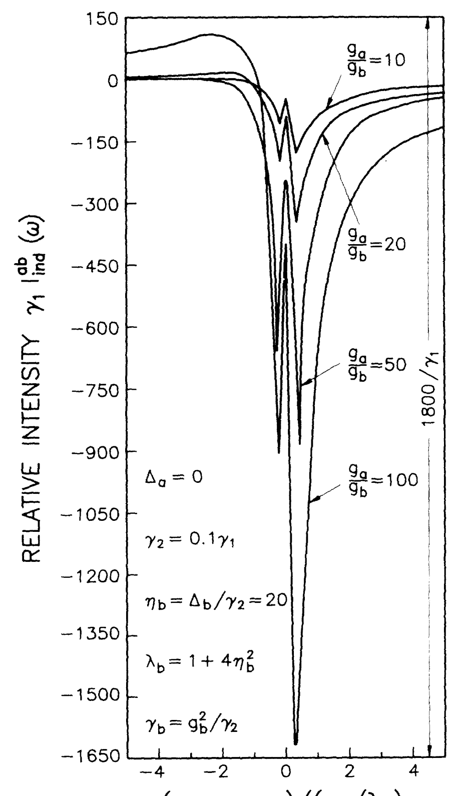

FIG. 2. Two-photon stimulated emission spectra in the low-intensity limit of both laser fields. The relative induced intensity $\gamma_1 I_{\mathrm{ind}}^{ab}(\omega)$ in units of $1/\gamma_1$ computed from Eq. (12) is plotted vs the relative frequency $(\omega-\omega_a-\omega_b)/(\gamma_b/\lambda_b)$ for $\Delta_a=0$, $\eta_b=20$, and various values of $g_a/g_b=10$, 20, 50, and 100, respectively.

where

$$I_1^{\text{sp}}(\omega)=\left[\frac{2}{\gamma_1}\right]\frac{\gamma_1^2/4}{(\omega-\omega_{10})^2+\gamma_1^2/4}\,, \tag{11}$$

$$I_{\text{ind}}^{ab}(\omega)=\left[\frac{2}{\gamma_1\lambda_{ab}}\right]\times\frac{f_1(\gamma_b/2\lambda_b)^2-(\omega-\omega_a-\omega_b)f_1'\gamma_b/\lambda_b}{(\omega-\omega_a-\omega_b)^2+(\gamma_b/2\lambda_b)^2}\,, \tag{12}$$

with $f_1=-1+\nu(1+\sigma)$, $\sigma=g_a/g_b-g_a/\gamma_2\lambda_b$, and $f_1'=\eta_b+m\sigma$. Expression (11) describes the spontaneous process for the excited state $|1\rangle$ which is a symmetric Lorentzian line that is peaked at $\omega=\omega_{10}=\omega_a+\omega_b-\Delta_a-\Delta_b$ and has a spectral width of the order of $\gamma_1/2$. The spectral function (12) represents the induced excitation, which is an asymmetric Lorentzian line that is peaked at $\omega=\omega_a+\omega_b$ and has a spectral width equal to $\gamma_b/2\lambda_b$. At frequencies $\omega^{\ddagger}\omega_a+\omega_b$, the asymmetry of the spectral line depends on values of the function f_1'. The maximum intensity (height) of the induced peak at $\omega=\omega_a+\omega_b$ is equal to $2f_1/\gamma_1\lambda_{ab}$, which may take positive or negative values indicating that the physical process of induced absorption (attenuation) and stimulated emission (amplification) is likely to occur at $\omega=\omega_a+\omega_b$.

When the laser field b is at resonance with the $|0\rangle\leftrightarrow|2\rangle$ transition, namely, when $\Delta_b=0$, then $\nu=1$, $\lambda_b=1$, $f_1=\sigma$, and $f_1'=\sigma\eta_a$. In this case f_1 takes always positive values ($f_1>0$), which implies that the intensity (height) of the induced peak at $\omega=\omega_a+\omega_b$ is positive and, therefore, the spectral function at $\omega=\omega_a+\omega_b$ describes an induced absorption process. At frequencies $\omega^{\ddagger}\omega_a+\omega_b$ and for $\Delta_b=0$, the intensity (height) of the induced peak will take positive and negative values at frequencies $\omega<\omega_a+\omega_b$ and at $\omega>\omega_a+\omega_b$, respectively, the extent of which will depend on the values of the function $f_1'=\sigma\eta_a$ that are causing the asymmetry of the spectral lines.

When the laser field a is at resonance with the $|1\rangle\leftrightarrow|2\rangle$ transition, namely, when $\Delta_a=0$, then $\eta_{ab}=\Delta_b/\gamma_1$ and the height of the induced peak $2f_1/\gamma_1\lambda_{ab}$ assumes maximum negative values provided that the conditions $\Delta_b^2\gg\gamma_1\gamma_2/4$, $g_a>g_b$ and $\gamma_1>\gamma_2$ are satisfied. The conditions $\Delta_a=0$ and $\gamma_1>\gamma_2$ are required because they reduce substantially the numerical value of λ_{ab}. The induced two-photon spectra are illustrated in Figs. 2 and 3, where the relative intensity $\gamma_1 I_{\text{ind}}^{ab}(\omega)$ in units of $1/\gamma_1$ computed from Eq. (12) is plotted versus the relative frequency $(\omega-\omega_a-\omega_b)/(\gamma_b/\lambda_b)$ for $\Delta_a=0$, $\gamma_2=0.1\gamma_1$, and different values of g_a/g_b and $\eta_b=\Delta_b/\gamma_2$, respectively. The induced two-photon spectra in Figs. 2 and 3 are characterized by a splitting which occurs at $\omega=\omega_a+\omega_b$, where the two resolved peaks are located symmetrically from the frequency $\omega=\omega_a+\omega_b$ with unequal negative intensities (heights). The induced peak at $\omega>\omega_a+\omega_b$ has greater negative intensity (height) than that at $\omega<\omega_a+\omega_b$. The

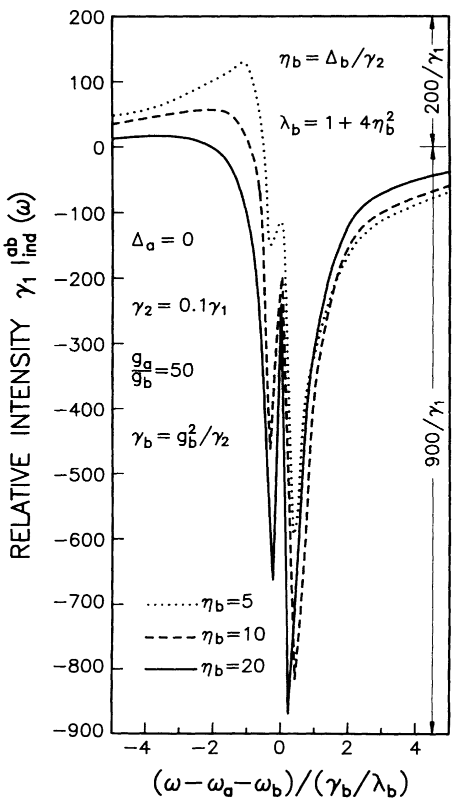

FIG. 3. As in Fig. 2 but for $g_a/g_b=50$ and various values of $\eta_b=5$, 10, and 20, respectively.

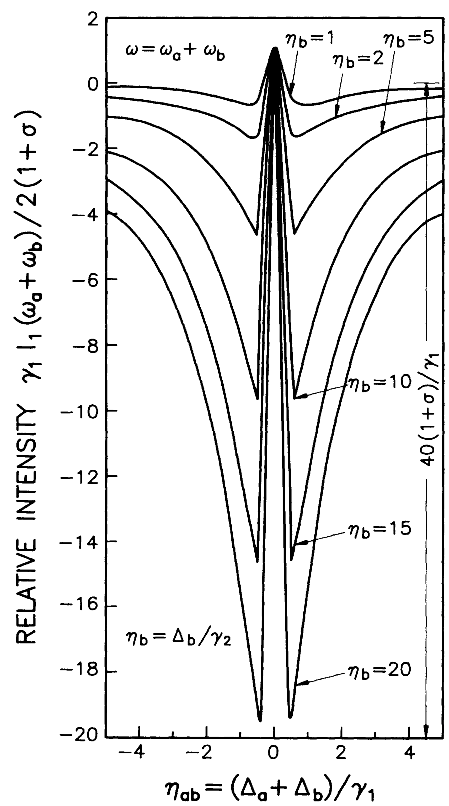

FIG. 4. Two-photon stimulated emission spectra in the low-intensity limit of both laser fields at the two-photon frequency $\omega=\omega_a+\omega_b$. The total relative intensity $\gamma_1 I_1(\omega_a+\omega_b)/2(1+\sigma)$ in units of $2(1+\sigma)/\gamma_1$ for an electron in the excited state $|1\rangle$ is computed from Eq. (13) and is plotted vs the total relative detuning $\eta_{ab}=(\Delta_a+\Delta_b)/\gamma_1$ for various values of $\eta_b=1$, 2, 5, 10, 15, and 20, respectively.

splitting of the spectral line is due to the large values of the function f_1^c Figure 2 depicts the induced two-photon spectra for $\eta_b=20$ and for various values of $g_a/g_b=10$, 20, 50, and 100, respectively. It is shown in Fig. 2 that the negative intensities (heights) of the induced peaks increase and, consequently, the amplification increases as the value of g_a/g_b increases for a given value of $\eta_b^2 \gg (\gamma_1/\gamma_2)$. Figure 3 illustrates the induced two-photon spectra for a given value of $g_a/g_b=50$ and for different detunings of $\eta_b=5$, 10, and 20, respectively. It is shown that the negative intensities (heights) of the induced peaks increase as the value of the detuning increases for a given value of g_a/g_b. Comparison between the negative intensities (heights) of the induced two-photon peaks in Figs. 2 and 3 with the intensity (height) $2/\gamma_1$ of the peak described by Eq. (11) at $\omega=\omega_{10}=\omega_a+\omega_b-\Delta_a-\Delta_b$, implies that significant amplification is likely to occur at the corresponding frequencies.

At frequencies $\omega=\omega_a+\omega_b$, the total spectral function for an electron in the excited state $|1\rangle$, $I_1(\omega)=I_1^{\mathrm{sp}}(\omega)+I_{\mathrm{ind}}^{ab}(\omega)$, becomes a function of the detuning $\eta_{ab}=(\Delta_a+\Delta_b)/\gamma_1$, namely,

$$I_1(\omega_a+\omega_b)=\frac{2(1+f_1)}{\gamma_1\lambda_{ab}}=\frac{2v(1+\sigma)}{\gamma_1}\frac{1/4}{\eta_{ab}^2+1/4}. \qquad (13)$$

The spectral function (13) describes the variation of the total spectral intensity at $\omega=\omega_a+\omega_b$ as a function of the detuning η_{ab}. It is illustrated in Fig. 4, where the relative intensity $\gamma_1 I_1(\omega_a+\omega_b)/2(1+\sigma)$ in units of $2(1+\sigma)/\gamma_1$ computed from Eq. (13) is plotted versus the detuning η_{ab}. For practical purposes $\sigma=g_a/g_b-g_a/\gamma_2\lambda_b$ may be replaced by $\sigma=g_a/g_b$ since $g_a/\gamma_2\lambda_b \ll 1$. Then the intensities (heights) of the induced peaks in Fig. 4 are proportional to $g_a/g_b>1$, and they increase negatively as the value of the detuning η_b increases. In Fig. 4 the splitting of the spectral lines occurs at $\eta_{ab}=0$, where the intensity is positive and equal to $I_1(\omega_a+\omega_b)=2(1+\sigma)/\gamma_1$, while for values of $\eta_{ab}>0$ and $\eta_{ab}<0$, the resolved peaks have equal negative intensities (heights) for a given value of η_b.

In conclusion, the spectral function (12) and Figs. 2 and 3 indicate that the two-photon-stimulated emission prevails provided that the conditions $\Delta_a=0$, $\eta_b^2 \gg \gamma_1/\gamma_2$, and $g_a>g_b$ are satisfied; therefore, significant amplification is expected to occur near the two-photon frequency $\omega=\omega_a+\omega_b$ The spectral function (13) and Fig. 4 imply that at the two-photon frequency $\omega=\omega_a+\omega_b$, the total intensities (heights) of the two peaks vary negatively for values of $\eta_{ab}>0$ and $\eta_{ab}<0$, respectively, for a given value of η_b. It is hoped that experiments will be encouraged in this direction to provide insight into the fundamental problem concerning the two-photon-stimulated emission process in a three-level atom, where laser amplification can be achieved without the need of population inversion between optical excitations.

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