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Off-resonant activation of optical emission

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A B S T R A C T

Recent reports have identified a three-wave optically parametric mechanism for the active enhancement of fluorescence using off-resonant radiation. In this Letter it is shown by numerical simulation that the output of a laser system optically pumped just below threshold, can be strongly enhanced by this mechanism, using an ancillary beam of moderate intensity. The electrodynamics and kinetics of the nonlinear optical mechanism are analyzed, model calculations performed, and the output is illustrated graphically. The response demonstrates a novel method for achieving all-optical transistor action.

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1. Introduction

In the sphere of parametric nonlinear optics, a facility to achieve switching and coherent frequency conversion is well known and its mechanisms are widely employed. The high efficiency of such processes are largely due to the coherence of the fundamental interactions, sustained by wave-vector matching and compounded by the development of stimulated emission in the output. Recently it has emerged that the basic process of optical emission can itself engage with off-resonant laser light, the strength of this mechanism owing to the stimulated forward scattering character of the coupling. In this Letter it is shown that the output of a laser system optically pumped just below threshold can be strongly enhanced by this mechanism, given the throughput of an ancillary beam with moderate intensity. Part of the interest in such a scheme lies in its possible relevance to all-optical transistor action which, like its electronic antecedent, can effect the switching or amplification of a source under the control of a signal input. Several other novel schemes for the realization of this type of action have recently been proposed, including one based on saturated absorption [1], electromagnetic induced transparency (EIT) [2–5], the optical Kerr effect [6–8], nonlinear transmission through coupling with surface plasmons [9–11], and beam filament rotation by application of a signal beam [12,13].

It can be anticipated that the desired characteristics of any practicable all-optical transistor will depend crucially on the engagement of a strongly nonlinear optical response. The scheme proposed in the present work is based on a third-order nonlinearity – its effect enhanced by stimulated emission – operating within a system designed to exploit the highly nonlinear response observed at the threshold for laser emission. In

contrast to the work of Hwang et al. [1], stimulated emission here primarily occurs in the course of forward scattering by a throughput beam whose optical frequency is purposely off-set from resonance. Detailed analysis shows that this beam, acting as the input signal, modifies the kinetics of emission and can thereby lead to an enhanced output [14,15]. The results of calculations for three-level laser systems highlight a significant potential for all-optical transistor action.

2. Method and results

Consider a typical three-level laser system optically pumped within a microcavity. The kinetics of emission are primarily determined by a pump rate R_p driving population from the ground state E_0 into a metastable upper level E_2, with stimulated emission as E_2 decays to E_1, and ultrafast relaxation from E_1 (Fig. 1). Following Siegman [16], the standard rate equations for the temporal behaviour of the cavity photon number, n, and the E_2 population, N_2, are as follows:

$$\frac{dn}{dt} = K(n+1)N_2 - \gamma_c n, \tag{1}$$

$$\frac{dN_2}{dt} = R_p - nKN_2 - \gamma_2 N_2. \tag{2}$$

Here, the population of E_1 is assumed to be negligible; K denotes the coupling coefficient for the laser transition, and γ_c, γ_2 signify the cavity and population decay rates, respectively. Under steady-state conditions, Eqs. (1) and (2) give the result;

$$n = \frac{R_p - g\gamma_c p + \left[\left(R_p - g\gamma_c p\right)^2 + 4\gamma_c R_p\right]^{\frac{1}{2}}}{2\gamma_c}. \tag{3}$$

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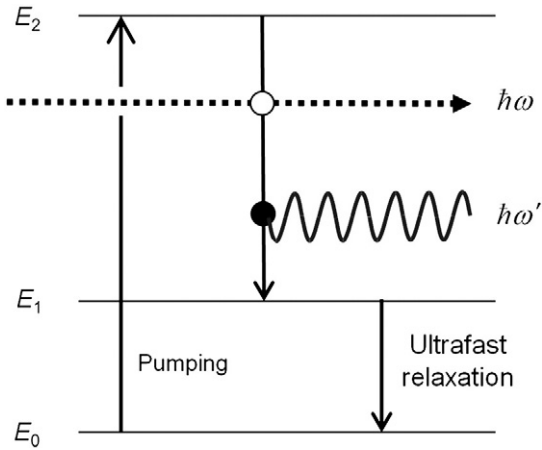
Fig. 1. Energy level diagram of the three-level laser system: black vertical arrows denote electronic transitions, the wavy line represents emission ($\hbar\omega'$), and the off-resonant beam ($\hbar\omega$) is the dashed arrow. The black and open dots symbolize one and two matter–photon interactions, respectively.

where $g = \gamma_2/\gamma_{rad}$ and $K = \gamma_{rad}/p$ (in which γ_{rad} is the radiative decay rate and p is the number of resonant cavity modes). The relaxation from E_2 into E_1 is not entirely radiative, i.e. $\gamma_2 \neq \gamma_{rad}$, since non-radiative relaxation processes (lattice phonons, wall collisions etc.) contribute to the decay. For present calculational purposes, given that the level E_2 decay rate γ_2 subsumes – but is dominated by – the rate of radiative decay, we assume $g = 5/4$ in the absence of the off-resonant input signal considered below. From Eq. (3), employing typical values $p = 10^{10}$ and $\gamma_c = 10^8 s^{-1}$, the familiar vertical climb in cavity photons at laser threshold emerges, as graphically illustrated by Fig. 2 (solid line).

All-optical control of such a pumped active medium may be achieved by nonlinear optical engagement of the laser emission with stimulated
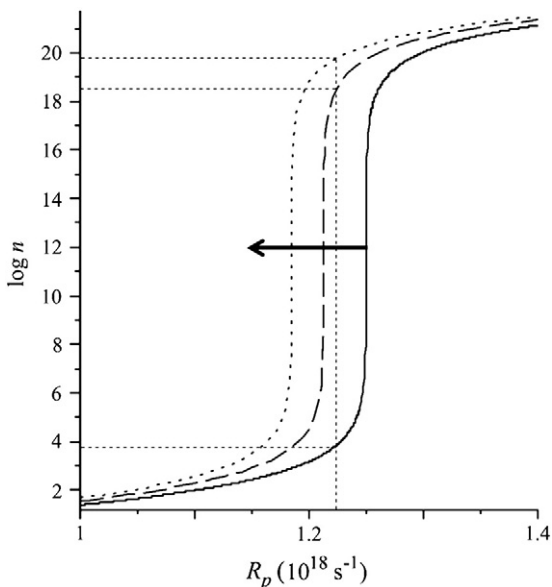

Fig. 2. Plot of log n, where n is the number of cavity photons, against the pumping rate, R_p, for absent (solid line) and present signal beam; example irradiances of the latter are 2×10^{11} W cm^{-2} (dashed) and 4×10^{11} W cm^{-2} (dotted). Horizontal arrow illustrates a movement of the lasing threshold to the left for increasing laser intensities. The vertical dotted line represents a constant R_p at which, on introduction of the signal beam, transistor action produces above threshold operation (denoted by the upper pair of horizontal dotted lines).

elastic forward scattering of off-resonant (signal) laser pulses, effecting a modification to the dipole transition moment for the $E_2 \rightarrow E_1$ laser transition [14,15]. The mechanism fundamentally entails three matter–photon interactions (Fig. 1); photons annihilated and created into the signal radiation mode (which thus emerges unchanged) are coupled with the photon emission. The intensity of emission, $I'(\Omega')$, (or power per unit solid angle) follows from Fermi's Rule [17] multiplied by the emitted photon energy, $\hbar\omega'$ [18]. Hence, the net intensity is determined from $I'(\Omega')d\Omega' = 2\pi\rho\omega'|M^{(1)} + M^{(3)}|^2$, where ρ is the density of radiation states; $M^{(1)}$ and $M^{(3)}$ are the quantum amplitudes for first- and third-order interaction processes, respectively — the former being the usual result for optical emission, the latter its leading nonlinear correction. The sought optical effects depend on the relative signs of the first- and third-order amplitudes, which are of primarily real character; a common sign will lead to emission enhancement, opposite signs its suppression. To proceed, the following detailed result has been determined in recent work [14];

$$I'\left(\Omega'\right) = \left(\frac{\omega'^4}{8\pi^2\varepsilon_0 c^3}\right)\left[e_i' e_j' \mu_i^{12}\overline{\mu}_j^{12} + (I/c\varepsilon_0)e_i' e_j e_k e_l'\chi_{ijk}^{12}\left(\omega';-\omega,\omega\right)\overline{\mu}_l^{12}\right.$$
$$\left. + \left(I^2/4c^2\varepsilon_0^2\right)e_i' e_j e_k e_l' e_m e_n \chi_{ijk}^{12}\left(\omega';-\omega,\omega\right)\overline{\chi}_{lmn}^{12}\left(\omega';-\omega,\omega\right)\right] , \quad (4)$$

where successive terms in square brackets originate from $|M^{(1)}|^2$, $M^{(1)}\overline{M}^{(3)}$ (plus its conjugate) and $|M^{(3)}|^2$, respectively. The decay transition dipole moment is designated by the shorthand notation $\boldsymbol{\mu}^{12} = \langle 1|\boldsymbol{\mu}|2\rangle$ — with $|1\rangle$ and $|2\rangle$ denoting the states of levels E_1 and E_2, respectively. Eq. (4) deploys the implied summation convention for repeated (Cartesian) indices, and I is the intensity (irradiance) of the input signal, with $\mathbf{e}'$ and $\mathbf{e}$ representing the linear polarization unit vectors of emission and signal photons, respectively.

The key parameter in Eq. (4) is the nonlinear transition (inelastic) tensor, $\chi_{ijk}^{12}(\omega';-\omega,\omega)$, mediating radiative decay of the molecular excited state. Unlike a parametric susceptibility, this tensor registers an inelastic (energy loss) process — in consequence of which, the first frequency parameter ω', registering emitter decay, differs from the sum of those which follow in the argument of χ. The explicit form of this tensor is determined from well-attested and reported methods [18–21], and the result is given by:

$$\chi_{ijk}^{12}\left(\omega';-\omega,\omega\right) = \sum_r \sum_{s\neq 2}\left(\frac{\mu_i^{1s}\mu_j^{sr}\mu_k^{r2}}{E_{s2}(E_{r2}-\hbar\omega)} + \frac{\mu_i^{1s}\mu_k^{sr}\mu_j^{r2}}{E_{s2}(E_{r2}+\hbar\omega)}\right)$$
$$+ \sum_r \sum_s \left(\frac{\mu_j^{1s}\mu_i^{sr}\mu_k^{r2}}{(E_{s2}-\hbar\omega+\hbar\omega')(E_{r2}-\hbar\omega)} + \frac{\mu_k^{1s}\mu_i^{sr}\mu_j^{r2}}{(E_{s2}+\hbar\omega+\hbar\omega')(E_{r2}+\hbar\omega)}\right)$$
$$+ \sum_{r\neq 1} \sum_s \left(\frac{\mu_j^{1s}\mu_k^{sr}\mu_i^{r2}}{(E_{s2}-\hbar\omega+\hbar\omega')(E_{r2}+\hbar\omega')} + \frac{\mu_k^{1s}\mu_j^{sr}\mu_i^{r2}}{(E_{s2}+\hbar\omega+\hbar\omega')(E_{r2}+\hbar\omega')}\right), \quad (5)$$

where ω is the signal beam frequency, and the transition moments are defined in the same manner as $\boldsymbol{\mu}^{12}$; r and s are intermediate states, equating to either 0, 1 or 2 in the three-level system (except where precluded in certain summations, as indicated above), and $E_{xy} = E_x - E_y$ is an energy difference between two such states. In passing, we observe that the nonlinear mechanism may alternatively be interpreted in terms of a 'dressing' of the emitter states by the throughput beam, manifest in a modification to the $E_2 \rightarrow E_1$ transition moment; although a different derivation method ensues, the same expression as Eq. (5) will emerge.

Continuing, we may suppose $\omega' < \omega$, and that these frequencies determine an offset, $\Delta E = E_{20} - \hbar\omega - \hbar\omega'$, a small fraction of the

energy for a typical electronic transition. Under these conditions the fourth term of Eq. (5) is dominant, delivering the following:

$$e'_i e_j e_k \chi^{12}_{ijk}\left(\omega'; -\omega, \omega\right) \approx \frac{\mu^3}{\Delta E(\Delta E + \hbar\omega')}. \tag{6}$$

Here and henceforth, it is assumed that the relevant transition dipole moment components, now simply represented as μ, have broadly similar magnitudes and direction − in calculations on specific systems, this approximation can of course be surrendered for greater accuracy. It should be observed that both factors in the denominator of Eq. (6) have negative values, so that the resulting tensor is always positive and, thus, denotes enhanced emission; under other conditions the susceptibility components may assume a negative value, representative of reduced emission. On insertion of Eq. (6) into Eq. (4), typical values of $I'(\Omega')$ may now be calculated for various signal beam intensities.

As indicated above, it is evident that the initial and final terms on the right-hand side of Eq. (4) correspond to spontaneous emission and the nonlinear coupling process, respectively. However, it is the second term (linear in I), signifying a quantum interference of these two processes, that represents the leading correction. With this in mind, the degree of enhancement (or in other cases any suppression) of the emission can be measured by taking the ratio of the second term against the first; the corresponding parameter η may be approximated as:

$$\eta = \frac{I\mu^2}{c\varepsilon_0 \Delta E(\Delta E + \hbar\omega')}. \tag{7}$$

Returning to Eq. (3), it is clear that the variable g will be affected by introduction of the input signal beam, since the radiative decay rate, γ_{rad}, and population decay rate, γ_2, both thereby suffer change (but to differing degrees); the non-radiative decay rate, γ_{nr}, can be assumed to be constant. By simple manipulation, an expression for g is given by;

$$g(I) = 1 + \frac{1 - Y(I)}{Y(I) + \eta Y(I)}, \tag{8}$$

where $Y(I) = \gamma_{\text{rad}}(I)/\gamma_2(I)$ and $\gamma_{nr}/\gamma_2(I) = 1 - Y(I)$. With the previous condition $g = 5/4$ for $I = 0$, and adopting indicative values $\mu = 16 \times 10^{-30}$ C m, $\Delta E = 10^{-20}$ J and $\hbar\omega' = 10^{-19}$ J, inserting Eq. (8) into Eq. (3) generates the results illustrated by two further curves on Fig. 2. For a constant pumping rate at a level indicated by the dotted vertical line, the system operates below threshold when no signal laser is present; on introduction of an off-resonant beam with an irradiance approaching 2×10^{11} W cm^{-2}, the output climbs by fourteen orders of magnitude, rising to sixteen orders if the signal input is doubled − typical of a laser operating at threshold. Transistor action with respect to the signal beam is clearly exhibited.

3. Discussion

The present theoretical analysis, based on off-resonant activation of laser emission, has identified a potentially new basis for achieving all-optical transistor action. The relatively simple optical configuration offers advantages of ultrafast response with high repetition rate, high efficiency, and a straightforward experimental setup. Moreover, the general scheme is based on a fundamental principle that is not limited to operation with any one specific material. In the broad context of research on all-optical methods of switching and amplification, such a scheme now invites experimental exploration with a view to potential applications. The realization of a material system that can prove suitable for implementing this mechanism will require the identification of active media for which the key tensor parameters can be optimized; all the necessary theory is now in place.

Acknowledgement

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