

## Quantum Optics

### Statistical Properties of Light from a Dye Laser

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One of the great successes of Quantum Optics in the Sixties was the theoretical derivation of the statistical properties of laser light and the subsequent detailed experimental confirmation. The laser was thereby established as a source of light with statistical properties fundamentally different from all thermal light sources.

The well known theoretical model which was successful in explaining all details of the photon statistics of a single mode laser near threshold is the simple "Van der Pol oscillator in rotating wave approximation".

$$\dot{\beta} = ((a_1 + ia_2) - (A_1 + iA_2)|\beta|^2)\beta + \zeta(t). \quad (1)$$

Here  $\beta(t)$  is the complex amplitude of the laser mode,  $a_1$ ,  $a_2$ ,  $A_1$ ,  $A_2$  are real parameters,  $a_2$  and  $A_2$  are different from zero only for nonzero detuning,  $a_1$  is the pump parameter and positive (negative) above (below) threshold,  $A_1 > 0$  provides for stabilization above threshold due to saturation.  $\zeta(t)$  in (1) is a Gaussian white noise source with the properties  $\langle \zeta(t) \rangle = 0$ ,  $\langle \zeta(t) \zeta(0) \rangle = 0$ ,  $\langle \zeta^*(t) \zeta(0) \rangle = Q\delta(t)$ , which turns (1) into a stochastic differential equation. Henceforth, we always use the Stratonovich calculus of such equations.

Equation (1) should apply to any single mode laser, and is in this sense universal, provided the laser is sufficiently close to its threshold, where corrections due to higher order nonlinearities or time derivatives, and deviations of  $\zeta(t)$  from white noise become irrelevant. In particular, the form of (1) is independent of any microscopic details of the laser process, and already follows from general symmetry principles of the theory of continuous instabilities. Recently, Kaminishi et al. [1] reported very interesting experimental results on the photon statistics of a dye laser near threshold. Their results differ completely from what one would expect on the basis of model (1). For instance in [1] it was found that the relative mean square of the intensity fluctuations  $\langle \Delta I^2 \rangle / \langle I \rangle^2$  increases up to values of 1000 for sufficiently small average photon number  $\langle n \rangle$ , whereas model (1) predicts an upper bound  $\langle \Delta I^2 \rangle / \langle I \rangle^2 \leq 1$ . The model which adequately describes a dye laser near threshold must therefore differ from (1) by terms which do not become irrelevant, even very close to the laser threshold.

In the present paper we wish to propose such a model, which turns out to be exactly solvable, and compare its predictions with the available experimental data. The good agreement which we find supports the idea that the dye laser threshold belongs to the new class of continuous instabilities with fluctuating control parameter which is described, again in a universal way, by our model. More experimental work along the lines of [1] which we hope to stimulate by the results presented here, would allow to test the very detailed quantitative predictions of the model.

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### Light Control by Light with an Example in Coherent Pump Dynamics, Propagation, Transverse and Diffraction Effects in Three-Level Superfluorescence

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Superfluorescence [1] (SF), is the dynamical radiation process which evolves from a collection of atoms or molecules prepared initially in the fully inverted state, and which subsequently undergoes collective, spontaneous relaxation [2]. Since Dicke's work [2], much theoretical and experimental effort has been devoted to SF [3].

With the exception of Bowden and Sung [4], all theoretical work has dealt exclusively with the relaxation process from a prepared state of complete inversion in a two-level manifold of atomic energy levels, and thus do not consider the dynamic effects of the pumping process. Yet, all reported experimental work [5-10] has utilized optical pumping on a minimum manifold of three atomic or molecular energy levels by laser pulse injection into the nonlinear

medium, which subsequently superfluoresces. The two-level analysis is only valid for  $\tau_R \gg \tau_p$ , (where  $\tau_R$  is the characteristic SF time and  $\tau_p$  is the pump pulse temporal width), and has not been realized over the full range of reported data.

In the work, we present through numerical simulations, including both transverse and longitudinal reshaping, the effects of coherent pump dynamics on SF emission from an optically-pumped three-level system. Contrary to [4], we do not confine our solution to the mean field regime and the linearized regime (short time) but we have adopted the propagational model advanced by Feld and co-authors [5] where both transients and propagation effects are rigorously studied. Quantum fluctuations [11] are not discussed in the treatment; instead, a classical (not random) uniform tipping angle concept is used throughout for initiating the polarization to simulate the fluorescence initiation. This approach is well-established for both transient two- and steady-state three level [5, 12] analysis. Since transverse effects are considered, the obtained results also extend the pumpless analysis that previously modelled the SF Cs experiment [14].

The model comprises a collection of identical three-level atoms, each having the energy level scheme such that the 1→3 transition is induced by a coherent pulse injected at frequency  $\omega_0$  tuned to the transition. The properties of this pumping pulse are specified initially. The transition 3→2 evolves by spontaneous emission at a much lower frequency  $\omega$ . The energy level spacing is such that the fields equations can be treated as separate. The energy levels 2→1 are not coupled radiatively due to parity considerations, and spontaneous relaxation from 3→2 is simulated by the choice of a small, uniform initial transverse polarization characterized by the parameter  $\phi_0 \sim 10^{-3}$  [12].

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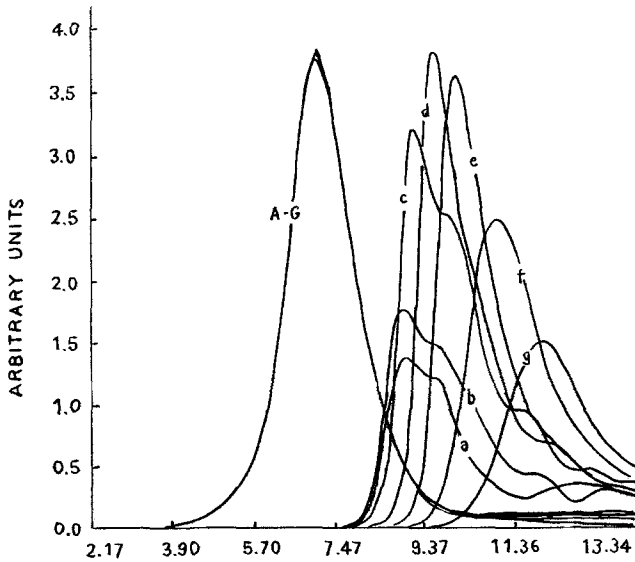


Fig. 1. Radially integrated normalized intensity profiles for the SF and injected pulses at  $z = 5.3$  cm penetration depth for seven different initial pump radial width at half-maximum  $r_p$ . The SF pulses are indicated by lower case letters, whereas the corresponding injected pump pulses are labeled by upper-case letters. The injection pulses are initially radially and temporally Gaussian with initial on-axis areas  $\theta_p = 2\pi$  with temporal widths  $\tau_w = 4$  ns. The level spacings are such that  $(\epsilon_3 - \epsilon_1)/(\epsilon_3 - \epsilon_2) = 126.6$ . The relaxation and dephasing times are taken as identical for all transitions, and are given as  $T_1 = 80$  ns and  $T_2 = 70$  ns, respectively. The SF effective gain  $g_s = 758.3 \text{ cm}^{-1}$  and the pump transition effective gain  $g_p = 14.6 \text{ cm}^{-1}$ . The initial radial widths at half-maximum for the injected pulses are: A  $r_p = 0.57$  cm; B  $r_p = 0.43$  cm; C  $r_p = 0.24$  cm; D  $r_p = 0.18$  cm; E  $r_p = 0.15$  cm; F  $r_p = 0.11$  cm; G  $r_p = 0.09$  cm. The corresponding geometrical Fresnel numbers are: a  $F = 8.46$ ; b  $F = 4.79$ ; c  $F = 1.47$ ; d  $F = 0.85$ ; e  $F = 0.57$ ; f  $F = 0.35$ ; g  $F = 0.21$

The atomic variables are chosen consistent by with the particular choice of  $\phi_0$  and the population in the ground state nearly unity [4, 13].

We used the electric-dipole and rotating-wave approximations and couple the atomic dipole moments to classical field amplitudes. Frequency detuning for both pump and SF emission are also included in the model. The matter equations of motion which describe the atomic system are coupled to the Maxwell's equations through the off-diagonal matrix elements associated with each transition field and are described elsewhere [14]. Not only do our results relate strongly to previous calculations [4] and experimental results in SF [10, 19], but we demonstrate a new manifestation of light control by light. We show how characteristics of the SF emitting at one-frequency can be controlled by specifying certain initial characteristics of the pump pulse injected at another frequency. In particular, our calculations show that the injected coherent pump initial characteristics, such as on-axis area, temporal and radial width (and associated gain-length Fresnel number [17]), and shape alter the SF pulse characteristics [14]. The effects of changing the effective gain of either the SF or the pump transition and the density of active atoms are also studied [14]. For sufficiently large effective gain and/or large input pump area, the two light pulses overlap and the two-photon processes (RCR-resonant coherent Raman) make strong contributions to the mutual pulse development [14]. Dependencies of this type have been recently observed in methyl-fluoride [10] and in barium [19b]. Furthermore, under other conditions, we obtained a SF pulse of temporal width much less than that of the pump even though the two pulses temporally

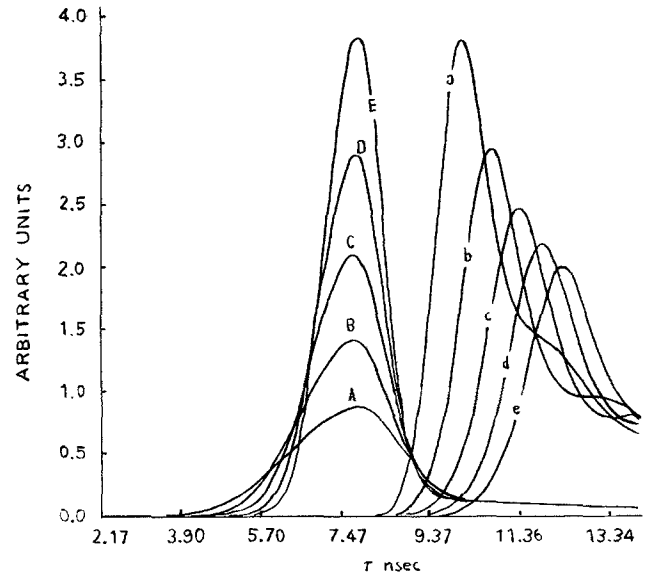


Fig. 2. Radially integrated normalized intensity profiles for the SF and injected pulses at  $z = 5.3$  cm penetration depth for five different initial temporal widths of the injected pulse. The initial on-axis area of the injected pulse  $\theta_p = \pi$  and the pump transition and SF effective gains are  $g_p = 17.5 \text{ cm}$  and  $g_s = 641.7 \text{ cm}^{-1}$ , respectively. All other parameters are the same as those for Fig. 1. The injected pulse initial temporal widths at half-maximum are: A  $\tau_w = 4$  ns; B  $\tau_w = 3.3$  ns; C  $\tau_w = 2.9$  ns; D  $\tau_w = 2.5$  ns; E  $\tau_w = 2.2$  ns

overlap. This calculation agrees qualitatively with the results of [10].

Calculational methods, discussed elsewhere [16, 17], were used to compute the effects on SF pulse evolution for various conditions for the injection signal such that  $\tau_R \ll \tau_p$ . Five examples follow.

In Figs. 1 and 2 are shown results of the numerical calculations for the radially integrated intensity profiles vs. retarded time, for the co-propagating SF and injected pulses at a penetration depth of  $z = 5.3$  cm in the nonlinear medium. These profiles correspond to what is observed with a wide aperture energy detector.

The initial radial width,  $r_p$ , of the Gaussian injected pulse was varied and the effect upon the SF pulse evolution is shown in Fig. 1. There is clearly indicated an optimum value for  $r_p$  for which the SF peak intensity is a maximum and the SF temporal width  $\tau_s$  is a minimum. It is seen that optimization occurs for a unity value of the conventional SF transition Fresnel number  $F_s$ ,  $F_s \approx 1$ . Thus the gain length  $\mathcal{F}_s = g_s z_{\text{max}}$ . Thus,  $\mathcal{F}_s = g_s/\kappa_s$  is the effective gain,  $g_s$ , to diffraction loss,  $\kappa_s$ , ratio. Thus, under these conditions, the SF pulse intensity reaches its maximum value, as it co-propagates with the injected pulse, at the penetration length,  $z_{\text{max}} = \kappa_s^{-1}$ . This is reasonable, since for  $F_s \gtrsim 1$  the propagation is expected to be plane-wave like, whereas for  $F_s < 1$ , diffraction is expected to play a major role. Other features of the SF pulses are in agreement with the results of [14] for a two-level system without effects of pump dynamics, namely the symmetrisation and smoothing of the SF pulses and increase in the SF peak delay time,  $\tau_D$ , with decreases in Fresnel number  $F_s$ .

Figure 2 shows the SF pulses at equal penetration for various values for the initial temporal width of the injected Gaussian a  $\Pi$ -pulses. All other parameters for the pulse propagation are equal. These results indicate the existence of an approximate linear relationship between the relative time delay,  $\tau_D$ , (between the peak SF and corresponding pump intensity), and the initial temporal width  $\tau_w$  of the pump pulse. This is in qualitative agreement with the analytical prediction made in [4B, Eq. (5.1)], based upon mean field theory.

Thus, reducing  $\tau_{sp}$ , increases the SF delay time and temporal width and decreases the SF peak intensity as well as the pump depletion to the evolving SF.

The initiation process for SF pulse generation was treated in this calculation completely classically and non-statistically. Therefore, these results must be taken as expectation values, or ensemble averages. The statistical treatment with quantum initiation [11] will be treated in a future publication [18].

Furthermore, this work constitutes a simplified model where the importance of pump dynamics effects are correctly simulated for the first time. It can be readily generalized to encompass two-photon pump and concomitant two-cascade or two-parallel SF emission. This effort is in progress to achieve a quantitative comparison with Eu and Sr observations [19a].

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## Theory of Coherent Two-Photon Resonance in Averaged Hamiltonian Approximation

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A theory of coherent two-photon resonance in a multilevel system is developed using the method of averaged Hamiltonian approximation. Assuming that the Hamiltonian of atomic or molecular system interacting with the photon field is

$$\mathcal{H} = \mathcal{H}_s + \mathcal{H}_{int} + \mathcal{H}_{ph} \quad (1)$$

with

$$\mathcal{H}_s = \sum_i \Omega_i N_{ii}, \mathcal{H}_{int} = \sum_{ij} \mu_{ij}^{(k)} A_k N_{ij}, \text{ and } \mathcal{H}_{ph} = \sum_k \mathcal{H}_{ph}^{(k)}.$$

Here  $N_{ij}$  are the standard-basis operators [1] of the system,  $A_k$  operators describes the photon field,  $\Omega_i$  are the energy levels ( $\hbar=1$ ) and  $\mu_{ij}^{(k)}$  are the matrix elements for allowed transitions between levels  $i \rightleftharpoons j$  induced by photons  $k$ .

The dynamics of the system is determined by the time-evolution operator which can be presented in the form

$$U(t) = \exp[-i(\mathcal{H}_s + \mathcal{H}_{ph})t] \cdot T \exp\left[-i \int_0^t \mathcal{H}_{int}(t') dt'\right]. \quad (2)$$

The interacting Hamiltonian is transformed into rotating frame according to

$$\tilde{\mathcal{H}}_{int}(t) = \exp(i(\mathcal{H}_s + \mathcal{H}_{ph})t) \cdot \mathcal{H}_{int} \exp[-i(\mathcal{H}_s + \mathcal{H}_{ph})t]. \quad (3)$$

Magnus [2] has derived the formula for expanding the operator with time-ordering as

$$T \exp\left[-i \int_0^t \tilde{\mathcal{H}}_{int}(t') dt'\right] = \exp[-i(F_0 + F_1 + F_2 + \dots)]. \quad (4)$$

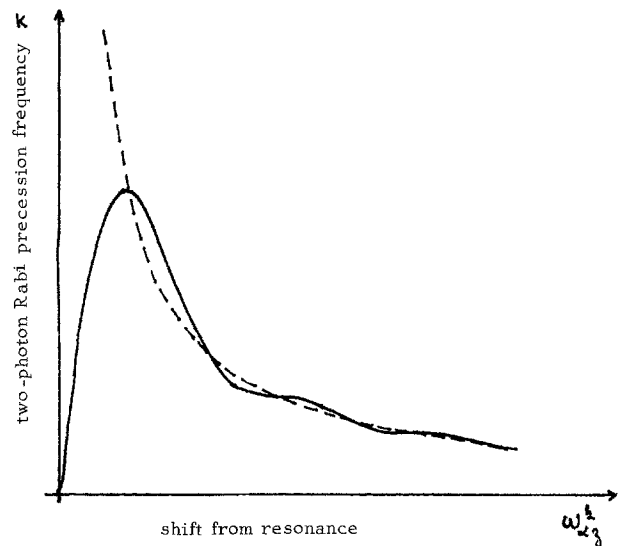


Fig. 1. The figure shows the Rabi two-photon precession frequency calculated from expression (6) (—) and the result from [3] (---)

The detailed calculations of the particular terms of the exponent (4) reveals that  $F_0$  consists of the operators inducing only allowed one-photon transitions,  $F_1$  only two-photon transitions,  $F_2$  only three-photon transition etc. If the effective-averaged Hamiltonian can be defined as  $\mathcal{H}_{av} = \frac{\partial F}{\partial t}$  then two-photon averaged Hamiltonian is

$$\mathcal{H}_{av}^{(2)} = -\frac{1}{4} \sum_{\alpha\gamma} \mu_{\alpha\gamma}^{(k)} \mu_{\alpha\gamma}^{(k')} A_k A_{k'} N_{\alpha\gamma} \left[ \frac{\omega_{\alpha\gamma}^k - \omega_{\alpha\gamma}^{k'}}{\omega_{\alpha\gamma}^k \omega_{\alpha\gamma}^{k'}} \exp[i(\omega_{\alpha\gamma}^k + \omega_{\alpha\gamma}^{k'})t] + \frac{1}{\omega_{\alpha\gamma}^{k'}} \exp(i\omega_{\alpha\gamma}^k t) + \frac{1}{\omega_{\alpha\gamma}^k} \exp(i\omega_{\alpha\gamma}^{k'} t) \right] \quad (5)$$

with  $\omega_{ij}^k = \pm\omega_k - \Omega_i + \Omega_j = \pm\omega_k - \Omega_{ij}$ , where  $\omega_k$  is the frequency of the photon field.

At two-photon resonances when  $\omega_{az}^k + \omega_{zy}^{k'} = 0$  the Hamiltonian (5) gets the form

$$\mathcal{H}_{av}^{(2)} = -\frac{1}{2} \sum_{\substack{\alpha z \gamma \\ kk'}} \mu_{\alpha}^{(k)} \mu_{\gamma}^{(k')} A_k A_{z'} N_{\alpha \gamma} \frac{1}{\omega_{az}^k} [1 - \cos \omega_{az}^k t]. \quad (6)$$

It describes the following two-photon effects

$$\omega_k - \omega_{k'} = \Omega_{az} + \Omega_{z\alpha} = 0$$

Rabi two-photon precession when

$$\omega_k + \omega_{k'} = \Omega_{az} + \Omega_{zy}$$

Raman coherent transitions when

$$\omega_k - \omega_{k'} = \Omega_{az} + \Omega_{zy}.$$

It also exhibits two very interesting features not found in the previous calculations [4, 3]:

1) When approaching the one-photon resonance  $\omega_{az}^k \rightarrow 0$  the two-photon Hamiltonian (6) is disappearing (Fig. 1).

2) There is the time delay between the beginning of the photon pulse and the time when two-photon effects starts. It is proportional to

$$\tau \approx 1/\omega_{az}^k.$$

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## Novel Quantum-Electronics Phenomena and Devices

### Voltage Memory, Provided by Photophysical Hole-Burning of Selectively Laser-Excited Dyes

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Selective laser-excitation of dyes in Langmuir-Blodgett films and in noncrystalline solids, e.g. organic glasses and amorphous polymers, demonstrated new optical methods [1] of detecting acoustic phonons and led to laser-induced unusual spectroscopic observations [2] combined with persistent hole-burning. The results are explained by a photophysical model [1] of matrix-shift variations, caused by phonon-induced transitions in the double-well potentials [3] of the amorphous solid. In addition this nonphotochemical model explained the large thermal line-broadening of selectively laser-excited dyes in amorphous solids. In the present contribution we demonstrate that persistent hole-burning provides not only a sensitive method of probing the effects of electric fields on the dye-matrix system, but also a versatile voltage memory, which on the other hand is confirming the photophysical model.

The fluorescence intensity  $I_0$  of the first vibronic zero-phonon line of perylene molecules, matrix-isolated in a thin film of the amorphous polymer polyvinylbutyral has been reduced to  $I_0^*$  in the course of selective laser-excitation at temperatures between 1.2 K and 30 K. Simultaneously voltages  $U_n$  have been applied consecutively to electrodes at the surfaces of the film. After each hole burning process with high power density  $p_L$  of the exciting He-Cd laser,  $I_0$  has been measured with low  $p_L$  as a function of the voltage  $U$ . It turned out that the curve of fluorescence intensity  $I_0^*$  as a function of the voltage  $U$  (or electric field) (Fig. 1) provides a versatile voltage memory, storing information not only on the value and the sign but also on the chronological order of the voltages  $U_n$  applied during hole burning. The dependence of the effects on the sign of the electric field, despite of the symmetry of the perylene

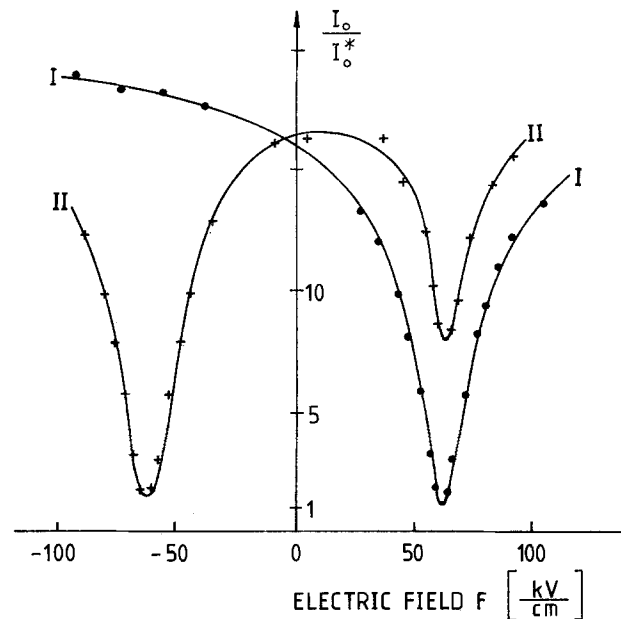


Fig. 1. Fluorescence intensity  $I_0/I_0^*$  of perylene in polyvinylbutyral vs. the electric field  $F$ , after hole burning in the presence of electric fields at 1.3 K. I After hole burning at  $E=60$  kV/cm, II after repeating hole burning at  $E=-60$  kV/cm

molecule suggested conclusions attributing the physical origin of the effects to the amorphous dye-matrix system. The dependence of the voltage memory on the chronological order of the applied voltages  $U_n$  provides a confirmation of the photophysical model of hole burning and refilling, because it demonstrates the matrix-shift variation of the electronic levels of the dye molecules within the inhomogeneous broadening which is due to the statistical distribution of the dye sites.

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