COHERENT EFFECTS IN PULSED LASER EMISSION

É. M. BELENOV, A. N. ORAEVSKIĬ, and V. A. SHCHEGLOV

P. N. Lebedev Physics Institute, USSR Academy of Sciences

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Coherent molecular emission effects are considered in the generation of short optical pulses comparable with or shorter than the polarization relaxation time of the laser medium. It is shown that cooperative effects appear in molecular emission under certain conditions and the medium is capable of emission up to the state of complete anti-inversion.

1. Powerful ultrashort optical pulses are currently obtainable from lasers. Resonant interaction of such pulses with matter leads to coherent effects in molecular emission similar to Dicke's cooperative effect in spontaneous emission processes[1]. In particular, the medium may radiate even without inversion, i.e., in the case when the population of the lower level N_1 exceeds that of the upper level N_2.

Coherent emission from an uninverted medium is possible provided certain phase relations are observed between the field and the molecular polarization. Here the field-matter interaction time τ should be shorter than the coherence-loss time T_2 of the substance: $\tau < T_2$. Such time relationships can be realized directly in a Q-switched laser. In fact, if the photon lifetime T_r in the resonator is shorter than the polarization relaxation time T_2 of active molecules, then the emission pulse length τ is sufficiently short. As we show below, the medium in this case can radiate practically up to the complete anti-inversion of the working particles and, consequently, the energy of the laser pulse is close to the total energy stored in the material.

In solid-state lasers (ruby, neodymium, semiconductor) the polarization relaxation time at room temperature is not less than 10^{-12} sec. In this case to observe the coherent effects we must use resonators whose Q is so low ($Q/\omega = T_r < T_2$) that it is impossible to exceed the laser excitation threshold. The cooperative effects in molecular emission cannot be observed in practically feasible cases ($T_r \sim 10^{-8}$–10^{-9} sec). As a consequence the energy emitted by a solid state laser does not exceed one-half of the energy stored in the active material[2]. At the same time gas lasers have a comparatively long polarization relaxation time. For example $T_2 \lesssim 10^{-7}$ sec in a CO_2 laser. Under certain conditions cooperative effects in molecular emission can be observed in a Q-switched gas laser. In such a case the conversion of energy stored in the material into coherent emission can reach 100%.

The purpose of this paper is to consider the pulsed generation of a laser when the coherent molecule emission effects become significant.

2. The system of equations for the laser field $\mathscr{E}(t) = E(t)\cos(\omega t + \varphi(t))$, the polarization $\mathscr{P}(t) = P(t)\cos(\omega t + \psi(t))$, and the number of particles $N = N(t)$ can be reduced to the following form[3]:

$$\dot{Y} = -\mu Y + \mu\eta V \sin\Phi, \tag{1}$$

$$\dot{\Phi} = \left[\mu_2 n \frac{Y}{V} + \mu\eta \frac{V}{Y}\right]\cos\Phi, \tag{2}$$

$$\dot{V} = -\mu_2 V + \mu_2 n Y \sin\Phi, \tag{3}$$

$$\dot{n} = -\mu_1 n + \mu_1 n_0 - \mu_2 Y V \sin\Phi. \tag{4}$$

Here

$$Y = \frac{\rho T_2}{\hbar}E,\quad \mu = \frac{1}{\omega T_p},\quad \eta = \frac{2\pi}{\mu\hbar}\rho^2 N_0,\quad V = \frac{P}{\rho N_0},$$

$$\mu_2 = \frac{1}{\omega T_2},\quad \mu_1 = \frac{1}{\omega T_1},\quad n = \frac{N}{N_0},\quad \Phi = \psi - \varphi,$$

N_0 is the value of population inversion at Q-switch actuation time, η is pump power excess over threshold $\eta = 1$, and T_r, T_1, T_2 are relaxation times for the field, polarization and the number of particles respectively. Differentiation of (1)–(4) is performed with respect to dimensionless time ωt.

We consider that the polarization relaxation rate μ_2 is small: $\mu_2 \ll \mu$. In this case the motions described by the system (1)–(4) can be broken down into fast and slow[4]. The fast coordinates Y and Φ quasistatically follow the slow coordinates assuming the values $Y = \eta V$ and $\Phi = \pm\pi/2$, and the medium according to (1) is emissive when the phase shift between the field and polarization $\Phi = \pi/2$ and absorptive when $\Phi \approx -\pi/2$. Eqs. (1)–(2) linearized in the neighborhood of the point $Y = \eta V$, $\Phi = +\pi/2$ show that the solution $\Phi = \pi/2$ is stable when the inequality

$$\mu/\eta > -n\mu_2. \tag{5}$$

is satisfied.[1)] Inequality (5) can be violated only for negative n, i.e., depending on phase relations between the field and polarization the medium can radiate without being inverted. Assuming that $n < 0$ we write (5) in the form $|n| < T_2/T_r\eta$. For solid state lasers and the excitation threshold excess by a factor of ten ($\eta = 10$) $|n| < 10^{-12}/10^{-8}\eta \approx 10^{-5}$. Thus outside of the dependence on phase relations the medium of a solid state laser at room temperature radiates only up to the time when working level populations become equal. In a gas laser $|n| < 10^{-7}/10^{-8}\eta = 10/\eta$ and consequently the laser medium can radiate until complete anti-inversion is reached even if threshold is exceeded by a factor of ten ($\eta = 10$).

We compute the parameters of an optical pulse. For this purpose we rewrite (3) and (4) in the form

$$\dot{R} = -2\mu_2 R + 2\mu_2\eta n R, \tag{6}$$

1)We note that condition (5) determining the stability of the solution $\sin\Phi = +1$ is valid for any relations between μ and μ_2. The solution $\sin\Phi = +1$ is stable if the factor in front of $\cos\Phi$ in (2) is positive. This is what is required by inequality (5).

$$\dot{n} = -\mu_2\eta R. \quad (7)$$

In the derivation of (6)—(7) we introduced $R = V^2$ and neglected spontaneous level decay ($\mu_1 = 0$) during the pulse. A differential equation for n is readily obtained from (6)—(7):

$$\ddot{n} = -2\mu_2\dot{n} + 2\mu_2\eta n\dot{n},$$

Given initial conditions n = 0 and n = 1, this reduces to the first-order equation

$$\dot{n} = -2\mu_2 n + \mu_2\eta n^2 + 2\mu_2(1 - \eta/2). \quad (8)$$

We consider that at t = 0 the laser field is maximum: $Y = Y_m$. Since the polarization of the medium is proportional to the instantaneous value of the field ($\eta V = Y$) the population inversion at field maximum is

$$n(0) = 1/\eta. \quad (9)$$

The time variation of population is described by the solution of (8):

$$n = \frac{1}{\eta} + \frac{\eta - 1}{\eta}\,\mathrm{th}\,\mu_2(\eta - 1)t. \quad (10)$$

when $n \to 1$ $t \to -\infty$, and $n \to -1 + 2/\eta$ when $t \to +\infty$.

If inequality (5) holds, $N_0(1 - 1/\eta)$ particles radiate coherently out of a total of N_0 active particles. When the excess over threshold is high enough all particles radiate coherently in contrast to one-half radiating particles in typical solid state lasers.

We find the shape of the emission pulse. Differentiating (10) with respect to time and using (7) that can be represented in the form

$$Y^2 = -\frac{\eta}{\mu_2}\dot{n},$$

we find the time variation of Y

$$Y(t) = (\eta - 1)/\mathrm{ch}\,\mu_2(\eta - 1)t. \quad (11)$$

The maximum value of the field is $Y_m = (\eta - 1)$ and the pulse length is

$$\tau \approx 1/\mu_2(\eta - 1). \quad (12)$$

Switching to dimensional variables, (12) can be rewritten in the form

$$\tau \approx T_2/(\eta - 1). \quad (13)$$

Thus given a sufficient threshold excess, the pulse length is η times shorter than the polarization relaxation time T_2. However if η is so high as to violate (5) the generation process no longer has the single-pulse character discussed above and the laser emission assumes a spiking character. We now briefly discuss this case.

3. We consider that $\eta \gg \mu/\mu_2$ and, furthermore, we neglect spontaneous decay of polarization which is valid if pulse length τ is shorter than time T_2. In this case the system (1)—(4) assumes the form

$$\dot{Y} = -\mu Y + \mu\eta V \sin\Phi, \quad (14)$$

$$\dot{\Phi} = \left[\mu_2 n\frac{Y}{V} + \mu\eta\frac{V}{Y}\right]\cos\Phi, \quad (15)$$

$$\dot{V} = \mu_2 nY\sin\Phi, \quad (16)$$

$$\dot{n} = -\mu_2 YV\sin\Phi. \quad (17)$$

The last two equations are integrated directly

$$V = \sin\left(\mu_2\int Y\sin\Phi\,dt\right), \quad n = \cos\left(\mu_2\int Y\sin\Phi\,dt\right). \quad (18)$$

We first consider the limiting case of infinitely large resonator Q. The solution $\sin\Phi = +1$ is stable in the initial emission stage up to the point where the expression $\lambda = [\mu_2 nY/V + \mu\eta V/Y]$ becomes equal to zero. Here the field Y(t) satisfies the equation

$$\dot{Y} = \mu\eta\sin\left(\mu_2\int_{-\infty}^{t} Y\,dt\right), \quad (19)$$

whose solution has the form

$$Y(t) = \frac{2}{\mu_2\tau}\Big/\mathrm{ch}\frac{t}{\tau}, \quad \tau = 1/\sqrt{\mu\mu_2\eta}, \quad (20)$$

corresponding to a pulse shape propagating in an induced self-bleaching medium[5]. We can obtain an explicit time-dependence of λ for a field of type (10):

$$\lambda(t) = \frac{1}{\tau}\left[\cos\left(\frac{1}{2}\mu_2\int_{-\infty}^{t} Y\,dt\right)\right]^{-1}\sin^2\left(\frac{1}{2}\mu_2\int_{-\infty}^{t} Y\,dt\right). \quad (21)$$

For t = 0

$$\theta = \mu_2\int_{-\infty}^{t} Y\,dt = \pi,$$

and consequently λ changes sign, going from $+\infty$ to $-\infty$, at the point t = 0. In order for solution (20) to be stable over the whole range of θ, $\sin\Phi$ must change instantaneously from +1 to −1 at $\theta = \pi$ (more precisely, at $\theta = \pi + 2\pi m$, where m is an integer). Since $|\lambda| \to \infty$ when $\theta \to \pi$, any fluctuations of the system, however small, are sufficient to switch the phase of Φ in this manner. Thus when $0 \le \theta \le \pi$, $\sin\Phi = +1$ and the medium amplifies; when $\pi \le \theta \le 2\pi$, $\sin\Phi = -1$, and the medium absorbs. The pulse shape is determined uniquely by Eq. (20) at all stages of emission. Consequently when damping is neglected the energy stored in the material is pumped into the field, and conversely, when damping is taken into account the energy is gradually removed from the resonator so that emission has the character of damped pulsations.

In the case when the spike length $\tau = 1/\sqrt{\mu\mu_2\eta}$ is small relative to the photon lifetime $\frac{1}{2}\mu$ in the resonator, the energy ΔU per spike emitted by the generator can be readily found. Considering that

$$\Delta U \sim 2\mu\int_{-\infty}^{+\infty} Y^2(t)\,dt \quad (22)$$

and that the function Y(t) approaches (20) we obtain $\Delta U = 2\mu\tau\hbar\omega N$. The population inversion $N = N_2 - N_1$ decreases here at the end of the spike by $\Delta N = 4\mu\tau N$. In the approximation of $\mu\tau \ll 1$ we can also easily obtain the recurrent formulas relating the parameters of the m-th and (m + 1)-st spikes in an emission pulse. Taking into account the fact that the coefficients η_m and η_{m+1} of pump excess over threshold are proportional to the population inversion, we obtain

$$\eta_{m+1} = \eta_m(1 - 4\mu\tau_m), \quad \tau_m = 1/\sqrt{\mu\mu_2\tau_m}. \quad (23)$$

The population inversion at the beginning of the (m + 1)-st spike and the energy emitted in the (m + 1)-st spike are

$$N_{m+1} = N_m(1 - 4\mu\tau_m), \quad \Delta U_{m+1} = 2\mu\tau_m\hbar\omega N_m.$$

Since η decreases in the generation process, the spike length increases with m according to

$$\tau_{m+1} = \tau_m(1 + 2\mu\tau_m). \quad (24)$$

When m is sufficiently large so that the coefficient η_m becomes comparable with μ/μ_2 (here the length of the m-th spike in a pulse is of the order of photon lifetime in the resonator) (23) and (24) are no longer valid. In this case the character of laser generation is intermediate between the π- and the 2π-pulse emission and the solution of the system (14)—(17) seems to be possible only by numerical methods. We note that some results of the numerical solution of a system of material and field equations of a laser for $T_r \sim T_2$ are given in[6].

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