GENERATION OF ULTRA-SHORT DURATION COHERENT LIGHT PULSES

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Generation of ultra-short light pulses by the mode locking method is analyzed theoretically. The conditions for locking of many axial modes are found and the decisive role of dispersion of the medium in the resonator in restricting the number of synchronized modes and the minimal duration of the pulse is demonstrated. Ways of further reducing the light pulse duration down to 10^{-13}–10^{-14} sec by compensating for the medium dispersion and by broadening the negative absorption band are proposed.

1. INTRODUCTION

TWO effective methods of obtaining short light flashes of high power are available at present. The first is based on the shortening of the light-flash duration upon propagation in a nonlinearly amplifying medium [1,2] or in a medium having a nonlinear gain and absorption [2,3]. This method yields light flashes of duration 2×10^{-9} sec and power 7 GW [2,3]. In nonlinear amplification, the minimum flash duration is determined by the reciprocal of the spectral line width of the active medium [4,5]. The other method is based on the effect of synchronization of the radiation in several axial modes (mode locking) of a quantum generator.[6,7] As a result of the "locking" of the modes, a number of coherent light fields equally spaced in frequency is produced, and the total field due to interference constitutes a sequence of pulses separated by a distance $2\pi/\Omega$ (Ω - frequency difference of neighboring modes) and having a duration $2\pi/n\Omega$ (n - number of coherent modes). This method yielded light flashes of (directly measured) duration on the order of 10^{-10} sec.[8,9]

In this article we consider the problem of generation, by mode locking, of ultra-short light flashes of duration on the order of the optical oscillation period (10^{-13}–10^{-14} sec). The conditions for locking many axial modes are obtained, and it is shown that the dispersion of the medium inside the resonator plays a major role in the limitation of the number of synchronized modes and of the minimum pulse duration. This is of special interest as a check on the validity of the assumption made by DeMaria and Stetser [10] with respect to mode locking in the entire region of negative absorption of glass with Nd^{3+}, the width of which is 200 Å. It is pointed out that generation of ultra-short flashes of coherent light is made possible by compensation of the dispersion of the medium and broadening of the negative-absorption band.

2. CONDITIONS FOR THE LOCKING OF MANY MODES

The locking of several modes is realized effectively by Q-switching of the resonator at a frequency Ω which is close to the difference between the frequencies of neighboring axial modes. This question was investigated theoretically in [6,11,12]. The generation of ultra-short light pulses is possible when a very large number of modes (10^3 and more) is locked, and the conditions for this process differ greatly from the conditions for the locking of several modes.

If the effective conductivity of the resonator varies like $\sigma = \sigma_0[1 + \rho \cos(\Omega t + \varphi)]$, then the equations describing the mode interaction are

$$\ddot{E}_n + \frac{\omega}{Q}\dot{E}_n + \frac{\omega}{Q}\rho\cos(\Omega t + \varphi)(\dot{E}_{n+1} + \dot{E}_{n-1}) + \omega_n^2 E_n = -4\pi\ddot{P}_n, \quad (1)$$

where E_n is the field intensity in the n-th mode, ω_n is the natural frequency of the n-th mode of the resonator, P_n the polarization of the active medium at the frequency of the n-th mode, and Q the figure of merit of the resonator. The solution of (1) for the field E_n will be sought in the form

$$E_n = A_n(t)\cos[\Omega_n t + \varphi_n(t)],$$

where A_n and φ_n are the amplitude and phase of the field in the n-th mode, and vary slowly during the period. Similarly, we represent the polarization of the medium in the form [13]

$$P_n = C_n \cos(\Omega_n t + \varphi_n) + S_n \sin(\Omega_n t + \varphi_n).$$

We can then obtain for the oscillation phases $\psi_n = \Omega_n t + \varphi_n$ the following equations:

$$\dot{\psi}_n = \Omega_n{}^0 - \frac{\omega\rho}{4Q}\left[\frac{A_{n+1}}{A_n}\sin(\psi_{n+1} - \psi_n - \Omega t - \varphi) - \frac{A_{n-1}}{A_n}\sin(\psi_n - \psi_{n-1} - \Omega t - \varphi)\right], \quad (2)$$

where Ω_n^0 is the frequency of the oscillations of the field in the n-th mode in the absence of locking ($\rho = 0$). The frequency Ω_n^0 is determined by the natural frequency of the resonator ω_n, accurate to effects involving the pulling of the generation frequency towards the center of the amplification line, and the repulsion of the neighboring modes. We have left out from (2) terms responsible for the coupling of phases of the oscillation modes as a result of the nonlinear field dependence of the active-medium polarization. The nonlinear terms omitted from (2) are of the form $\omega f(x_{j,j+1})/QT_1 \times (\Omega_{n+1}^0 - \Omega_n^0)$ [12], where T_1 is the lifetime of the particles at the upper level, $x_{j,j+1} = \sin\Phi_{j,j+1}$, and $\Phi_{j,j+1}$ are the phase differences between the neighboring modes, and $f \sim 1$. When the condition

$$1 > \rho \gg 1/T_1(\Omega_{n+1}^0 - \Omega_n{}^0)$$

is satisfied, it is possible to neglect the indicated nonlinear terms in (2). In the cases of practical interest, where mode locking takes place under external modulation or self-locking of modes takes place as a result of an additional element inside the resonator (for example, a bleachable solution), the depth of modulation ρ is much larger than $1/T_1(\Omega_{n0+1} - \Omega_n^0)$, which amounts to 10^{-4}—10^{-6} for luminescent-crystal and glass lasers. In addition, to estimate the number of synchronized modes we can confine ourselves to an approximation in which the amplitudes A_n are assumed equal.

The equations for the phase difference between neighboring modes, $\Phi_{n;\,n+1} = \psi_{n+1} - \psi_n - \Omega t - \varphi$, then take the form

$$\dot{\Phi}_{n;\,n+1} = (\Omega_{n+1}^0 - \Omega_n{}^0 - \Omega) - \frac{\omega\rho}{4Q}(x_{n-1;\,n} - 2x_{n;\,n+1} + x_{n+1;\,n+2}), \quad (3)$$

where $x_{n;n+1} = \sin\Phi_{n;\,n+1}$. The conditions for synchronized generation of radiation in N modes (which we shall designate 1, 2, ..., N) is obviously

$$\dot{\Phi}_{n;\,n+1} = 0, \qquad n = 1, 2, \ldots, N, \quad (4)$$

with $\Phi_{0;\,1} = \Phi_{N;\,N+1} = 0$. From the system of N − 1 equations (3) with conditions (4) we can determine the values of $x_{n;\,n+1}$ and the phase φ_n of the field in each mode. Indeed, introducing the notation

$$a_{n;\,n+1} = \frac{4Q}{\omega\rho}(\Omega_{n+1}^0 - \Omega_n{}^0 - \Omega), \quad (5)$$

we obtain for $x_{n;\,n+1}$ the system of equations

$$x_{n-1;\,n} - 2x_{n;\,n+1} + x_{n+1;\,n+2} = a_{n;\,n+1}, \quad n = 1, 2, \ldots, N, \quad (6)$$

with $x_{01} = x_{N;\,N+1} = 0$. The solution of the system of equations (6) is

$$x_{12} = -N^{-1}[(N-1)a_{12} + (N-2)a_{23} + \ldots + a_{N-1;\,N}],$$

$$x_{n;\,n+1} = (n-1)a_{12} + (n-2)a_{23} + \ldots + a_{n-1;\,n} + nx_{12}. \quad (7)$$

The locking region for all N modes is defined by the condition[1)]

$$|x_{n;\,n+1}| < 1 \quad \text{for all} \quad n = 1, 2, \ldots, N-1. \quad (8)$$

If the generation frequencies ω_k of the laser modes are strictly equidistant in the absence of locking, i.e., $\Omega_{n+1}^0 - \Omega_n^0 = \Omega'$ for all n = 1, 2, ..., N − 1, then, by choosing the modulation frequency $\Omega = \Omega'$, we obtain $a_{n;\,n+1} = 0$ and consequently $x_{n;\,n+1} = 0$. In this ideal case conditions (8) are certainly satisfied, and in this approximation we get locking of all modes for which the self-excitation condition is satisfied. In practice, however, the initial mode generation frequencies Ω_n^0 (i.e., the generation frequencies prior to locking) are not equidistant, and this limits the number of synchronized modes, and consequently the limiting duration of the ultra-short flash.

3. LIMITING DURATION OF ULTRA-SHORT FLASH

The initial generation frequencies Ω_n^0 are determined by the natural frequency ω_n of the resonator, and also by the small shifts due to the pulling of the generation frequency towards the center of the negative-absorption line and due to the nonlinear mode interaction. The mode locking is affected only by the unequal spacing between the generation frequencies Ω_n^0. The main contri-

[1)] We neglect here the influence of the "unlocked" modes. The time behavior of the phase differences $\Phi_{j,j+1}$ of those modes for which the self-excitation conditions are satisfied and for which (8) does not hold is described, as before, by Eq. (3), but in this case the quantities $x_{j,j+1}$ will be periodic functions of the time. In the stationary modes, which we are considering in the present paper, the influence of these oscillating terms can be neglected if we average over a sufficiently long time. Moreover, the oscillating terms, as follows from (3), are connected only with the phases of two or three closest modes, and their contribution to the determination of the number of locked modes is negligibly small when $N \approx 10^3 - 10^4$.

bution to this unequal spacing is made by the unequal spacing of the natural frequencies ω_n of the resonator, due to the dispersion of the medium[2] inside the resonator. Allowance for the pulling effect reduces to a simultaneous allowance for the dispersion of the active particles. The non-equidistant spacing of the frequencies Ω_n^0 due to the mode interaction in the absence of external modulation is negligibly small compared with the non-equidistant spacing due to the dispersion of the medium. An estimate of this effect will be presented below. Thus, we can assume that the non-equidistant spacing of the frequencies Ω_n^0 is determined by the non-equidistant spacing of the natural frequencies ω_n of the resonator filled with the active medium: $\Omega_{n+1}^0 - \Omega_n^0 = \omega_{n+1} - \omega_n$.

The natural frequencies of the resonator ω_k are determined by the resonance condition[3]

$$\omega_k n(\omega_k) = \pi ck/L, \tag{9}$$

where L is the length of the resonator, c is the velocity of light in vacuum, and $n(\omega_k)$ is the refractive index of the medium inside the resonator at a frequency ω_k. Usually there are several substances (active medium, shutter, etc.) inside the resonator, with refractive indices $n_i(\omega_k)$ and lengths l_i. In this case the expression for $n(\omega_k)$ is

$$n(\omega_k) = \frac{1}{L}\sum_i l_i n_i(\omega_k). \tag{10}$$

In the considered region of negative-absorption frequencies, the change of the refractive index can be represented by

$$n(\omega_k) = n_0 + \frac{\partial n}{\partial \omega}(\omega_k - \omega_0) + \frac{1}{2}\frac{\partial^2 n}{\partial \omega^2}(\omega_k - \omega_0)^2, \tag{11}$$

where $n_0 = n(\omega_0)$ and the derivatives are taken at the point $\omega = \omega_0$. The non-equidistant spacing of neighboring pairs of natural frequencies is given by[4]

$$\frac{\omega_{k+2} - \omega_{k+1}}{\omega_{k+1} - \omega_k} = 1 - 2\Omega\mathcal{L}(n), \qquad \mathcal{L}\Omega \ll 1, \tag{12}$$

where

$$\mathcal{L}(n) = \left(\frac{\partial n}{\partial \omega} + \frac{\omega_0}{2}\frac{\partial^2 n}{\partial \omega^2}\right) \Big/ \left(n_0 + \omega_0 \frac{\partial n}{\partial \omega}\right).$$

The frequency difference $\omega_{k+1} - \omega_k$ of all the pairs of modes can be expressed with the aid of (12) in terms of the frequency difference of two selected modes (p + 1 and p):

$$\frac{\omega_{k+1} - \omega_k}{\omega_{p+1} - \omega_p} = 1 - 2\Omega\mathcal{L}(n)(k - p). \tag{13}$$

Then the general expression for the coefficients $a_{k;\,k+1}$ in the locking conditions (7) and (8) takes the form

$$a_{k;\,k+1} = \frac{4Q}{\omega\rho}[(\Omega_{p+1}^0 - \Omega_p{}^0 - \Omega) - 2\Omega^2\mathcal{L}(n)(k - p)], \tag{14}$$

where $\Omega_{p+1}^0 - \Omega_p^0 \approx \Omega$. The explicit expression (14) for the coefficients $a_{k;\,k+1}$ makes it possible to calculate $x_{n;\,n+1}$ accurately. Substituting (14) in (7) and summing, we obtain the locking conditions (8) in explicit form

$$\frac{2Q}{\omega\rho} n(N - n) \left| (\Omega_{p+1}^0 - \Omega_p{}^0 - \Omega) + 2\Omega^2\mathcal{L}(n)\left(p - \frac{N+n}{3}\right)\right| < 1 \tag{15}$$

for all n = 1, 2, ..., N − 1.

The modulation frequency Ω should lie in the interval $(\Omega_2 - \Omega_1;\ \Omega_N - \Omega_{N-1})$, and for any pair of modes (which we shall denote by p + 1 and p) the modulation frequency will coincide with $\Omega_{p+1}^0 - \Omega_p^0$ accurate to $2\Omega^2\mathcal{L}(n)$. Consequently, the first term $(\Omega_{p+1}^0 - \Omega_p^0 - \Omega)$ in (15) is negligibly small compared with the second. Then conditions (15) reduce to the following:

$$\left| n(N - n)\left(p - \frac{N+n}{3}\right)\right| < \frac{\omega\rho}{2Q}\frac{1}{2\Omega^2\mathcal{L}(n)}, \qquad n = 1, 2, \ldots, N - 1. \tag{16}$$

The left side of the expression has a maximum when

$$n = p \pm \sqrt{p^2 - N(p - N/3)}.$$

Substituting n(p) in (16) we obtain a condition that limits the number of synchronized modes N(p). The optimal case is when the modulation frequency is in best resonance with the central pair of modes (p = N/2), for when p = N/2 the left side of (16) with n = n(p) reaches a minimum. As a result we get the condition that limits the number of synchronized modes:

$$N < \left(9\sqrt{3}\frac{\omega\rho}{Q\Omega^2\mathcal{L}(n)}\right)^{1/3} = N_{max}. \tag{17}$$

The limiting duration of the ultra-short pulse is $\tau_{min} = 2\pi/\Omega N_{max}$ and, consequently, it is determined by the expression[5]

[2] We have in mind lasers using luminescent crystals and glasses.

[3] The radiative shift of the neutral frequency [14] can be neglected.

[4] The quadratic term $\partial^2 n/\partial\omega^2$ makes a contribution that is linear in ω to the non-equidistance of the modes. This fact was kindly pointed out to us by V. I. Bespalov.

[5] If $\Omega N_{max} \gtrsim \Delta\omega$ ($\Delta\omega$ – width of the negative-absorption band), then the dispersion of the medium does not limit the extremal pulse duration.

$$\tau_{min} = 2\pi\left(9\sqrt{3}\frac{\omega\rho\Omega}{Q\mathcal{L}(n)}\right)^{-1/3}. \quad (18)$$

By way of an example, let us consider a neodymium-glass laser with the following parameters: $\omega/Q \approx 10^8$ sec^{-1}; $\Omega = 10^9$ sec^{-1}; $\rho \approx 0.1$; $\omega_0 \approx 2\times 10^{15}$ sec^{-1}; $n_0 \approx 1.5$. The only dispersive medium is the glass, with $l/L = 0.1$ and $|\partial n/\partial\omega| \approx 10^{-17}$ sec. The dispersion of the glass is due to absorption bands in the ultraviolet. In this case, $\omega\partial^2 n/\partial\omega^2 \approx \partial n/\partial\omega$ at frequencies in the visible and in the infrared. The negative dispersion of the active particles is $\partial n^-/\partial\omega \approx \alpha/k\Delta\omega$, where α is the gain of the medium in cm^{-1}, k the wave vector, and $\Delta\omega$ the width of the negative-absorption band. In the case of neodymium glass ($\alpha \approx 0.1$ cm^{-1}, $\Delta\omega \approx 4\times 10^{13}$ sec^{-1}, $k \approx 10^4$ cm^{-1}), the dispersion of the ions is $\partial n^-/\partial\omega \approx 1.5\times 10^{-19}$, i.e., it is small compared with the dispersion of the glass. In this case the maximum number of synchronized modes $N_{max} \approx 530$, and the minimal duration of the pulse is $\tau_{min} \approx 1.2\times 10^{-11}$ sec. This exceeds by more than one order of magnitude the limiting duration 2×10^{-13} sec corresponding to locking of all the axial modes within the limits of the negative-absorption band of the neodymium glass.

The presently available experimental data are those of Stetser and DeMaria [8,10] on the self-locking of neodymium-glass laser modes as a result of nonlinearity in a saturable filter, where it is deduced on the basis of indirect assumptions that all the modes were locked in the negative-absorption band and pulses with 2×10^{-13} sec duration were generated. Our foregoing results are not directly applicable to a laser with a saturable filter for the following reasons: 1) the depth of modulation ρ is a function of the number of synchronized modes; 2) the Q-switching law is not sinusoidal, i.e., the modulation spectrum can include several components at frequencies that are multiples of Ω; 3) the proper dispersion of the saturable filter is not constant, but is modulated in accordance with the same law as the transmission of the filter. Nonetheless, we can obtain on the basis of the foregoing results an upper estimate for the pulse duration in this case, too, under the assumption that the depth of modulation has reached saturation ($\rho \approx 1$), that the modulation at the frequencies $\Omega, 2\Omega, \ldots, m\Omega$ is equivalent to an increase of the parameter $\rho\Omega$ by a factor m, and that the dispersion of the filter itself can be neglected. Then the value of the limiting duration in our example is $\tau_{min} \approx m^{-1/3} \times 0.5\times 10^{-11}$ sec. Even when $m \approx 10$ the limiting duration $\tau_{min} \approx 2\times 10^{-12}$ sec is smaller by one order of magnitude than the duration 2×10^{-13} sec, estimated from the width of the negative-absorption band. However, the obtained estimate is approximate, and to obtain a definite answer it is necessary to measure the dispersion of the medium inside the resonator, to measure the depth and spectrum of the frequencies at which the absorption of the saturable filter are modulated in the mode-locking regime.

It was assumed in the foregoing calculation that the amplitudes of the fields in all the modes are approximately the same. Actually, the field amplitudes in the modes can be smaller at the edges of the line than at the center of the line. Therefore the obtained value of N_{max} is the upper limit of the number of synchronized modes. The actual value of N_{max} may be somewhat lower if ΩN_{max} is close to the width of the negative-absorption band. However, if ΩN_{max} is much smaller than the width of the amplification band (as in the example given above), then the obtained expressions for N_{max} and τ_{min} are more accurate.

Let us return to an estimate of the influence of the mode interaction on the non-equidistant character of the frequencies Ω_n^0 in the absence of external modulation. The maximum non-equidistance due to such an interaction is $\Delta\Omega_{int} \approx \omega(\eta - 1)/T_1\Omega QN$ [12] (T_1 is the longitudinal relaxation time, η is the coefficient of the rise above threshold). The maximum non-equidistance of the frequencies Ω_n^0 due to dispersion, according to (13), is

$$\Delta\Omega_{disp} \approx \frac{2\Omega^2(\partial n/\partial\omega + {}^1/_2\omega_0\,\partial^2 n/\partial\omega^2)}{n_0 + \omega_0\,\partial n/\partial\omega}N.$$

At standard values of T_1, Ω, $\partial n/\partial\omega$ etc., and for arbitrary values of N, we have $\Delta\Omega_{disp} \gg \Delta\Omega_{int}$. Consequently, the dispersion makes the main contribution to the limitation of the number of synchronized modes.

4. POSSIBILITIES OF FURTHER REDUCING THE DURATION

To obtain by the mode-locking method light flashes of duration determined only by the reciprocal width of the negative-absorption band it is necessary to reduce the dispersion of the medium inside the resonator. The simplest method is to use supplementary substances whose dispersion has a sign opposite that of the dispersion of the active medium, of the saturable solution, etc. In principle, such a method can make the total dispersion to vanish in first order:

$$\frac{\partial n}{\partial \omega}+\frac{\omega_0}{2}\frac{\partial^2 n}{\partial \omega^2}=\sum_i \frac{l_i}{L}\left(\frac{\partial n_i}{\partial \omega}+\frac{\omega_0}{2}\frac{\partial^2 n_i}{\partial \omega^2}\right)=0. \quad (19)$$

In this case only the dispersion of higher orders can limit the duration. It is similarly possible to calculate the maximum value of the synchronized modes in this case, too, but in practice the pulse duration will be limited here already by the width of the negative-absorption band.

To generate light pulses with duration shorter than 10^{-13} sec it is necessary to use very broad negative-absorption bands. It is possible to broaden the negative-absorption band by using inside the resonator several working media with non-overlapping amplification bands. For example, the negative-absorption band of Nd^{3+} ions varies in different crystals within a range 500—600 Å.[15] In principle it is possible to use entirely different substances (for example, ruby and neodymium glass) and generate at different wavelengths ultrashort light pulses that are coherent with one another. Interest also attaches to mode locking within the limits of different spectral lines of gases (for example, the numerous rotational-vibrational transitions of CO_2 molecules [16], etc.), since this leads to an appreciable increase of the peak power and to coherence of all the emission lines.

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