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Evolution of chirped light pulses and the steady state regime in passively mode-locked femtosecond dye lasers

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Résumé. — Un modèle numérique pour les lasers à colorant à blocage en phase des modes passifs est présenté qui permet d'étudier l'évolution temporelle du module et de la phase des impulsions lumineuses ultrabrèves et de déterminer les paramètres d'impulsion et la fréquence laser du régime stationnaire sous différentes conditions. L'équation de boucle déduite prend en compte le système complet de la matrice densité et des équations de Maxwell pour l'interaction lumière-matière où aucune hypothèse restreignante sur les paramètres d'impulsion n'est donc nécessaire. Une forme possible de photoisomère du colorant absorbant, la dispersion de vitesse de groupe, l'automodulation de phase dépendant de l'intensité et les pertes linéaires sont comprises dans le modèle pour simuler les mécanismes les plus essentiels responsables du régime pulsé stationnaire dans les lasers à colorant femtosecondes actuels. De plus, d'un point de vue plus général le blocage en phase des modes passifs est analysé relativement à la propagation d'impulsions cohérentes dans le milieu résonant.

Abstract. — A numerical model of passive mode-locking of dye lasers is presented that allows one to study the temporal evolution of modulus and phase of ultrashort light pulses as well as to determine the pulse parameters and the laser frequency of the steady state under various conditions. The round trip equation derived takes into account the full system of density matrix and Maxwell equations for the light-matter interaction where accordingly no limiting assumptions on the pulse parameters are necessary. A possibly occurring photoisomer form of the absorber dye, group velocity dispersion, intensity dependent self-phase modulation and linear loss are included in the model to simulate the most essential mechanisms responsible for the steady state pulse regime in present femtosecond dye lasers. In addition, from a more general point of view passive mode-locking is analysed with respect to coherent pulse propagation through the resonant media.

1. Introduction.

1.1 EXPERIMENTAL RESULTS. — Today the most convenient tool to generate subpicosecond optical pulses is the CW mode-locked dye laser, where the mode-locking can be achieved by passive or by hybrid methods [1]. Passive mode-locking with a saturable absorber is the simplest and cheapest laser system, since it requires no mode-locked pump laser. This method is still the source of the shortest directly generated laser pulses [2]. The reduction of the pulse duration in the last few years has been principally due to the optimization of the resonator design of the Rhodamine 6G laser employing DODCI as a saturable absorber. The two significant steps forward in generating femtosecond pulses were the introduction of the colliding pulse mode-locked (CPM) ring laser [3], which for the first time generated pulses of less than 100 fs duration, and the

recognition of the role of the phase modulations in such cavity configurations [4], which resulted in the generation of pulses shorter than 60 fs [5]. The temporal modulation of the phase (chirp) was found to be caused by saturation of the absorber below resonance [6, 7] as well as by self-phase modulation due to Kerr-effect type nonlinearity in the dye solvent [2, 8]. The most advantageous and widely used technique to control the chirp and consequently the pulse duration is the introduction of one or more dispersive prisms in the resonator [2, 5, 9]. Due to the combined action of the angular dispersion leading to anomalous group-velocity dispersion [10] and of the variable intracavity glasspath exhibiting normal dispersion the net group-velocity dispersion can be easily tuned. A quantitative estimation of the net dispersion under experimental conditions is, however, rather complicated because of the influence of focusing elements in the resonator [11].

A common and not completely understood feature when using intracavity prisms is the asymmetrical operational characteristics of the mode-locked regime when the intracavity dispersion is adjusted [2, 7]. The point of minimum pulse width is characterized by decreased stability as can be seen from figures 1a, b [12].

More recently pulses of less than 100 fs duration were obtained in lasers incorporating other high transmission dispersive elements such as dielectric mirrors [13] and Gires-Turnois interferometers [14]. The somewhat different stability and phase modulation behaviour in this case is probably a result of higher order dispersion terms, which are responsible for modification of the nonlinear phase modulations (see for comparative analysis [15, 16] and for discussion [17, 18] and Refs therein).

A common feature of all experiments with the Rhodamine 6G/DODCI laser is that in the best operating regime the laser wavelength is shifted to the long wavelength side of the DODCI absorption spectrum, where the photoisomer of DODCI [19] has a larger absorption cross section. An experimental estimation of the steady state photoisomer accumulation in the femtosecond regime [20] revealed that its contribution to the absorption (at the laser wavelength) is of the same order as that of the ground form. Similarly, the first femtosecond operation in a dispersion optimized configuration using other absorbers (HICI, DASBTI) was also attributed to the corresponding photoisomers [16].

The shortest pulses obtained nowadays using hybrid modelocking techniques employ again the Rhodamine 6G/DODCI dye combination and CPM regime as well as dispersion optimization [21]. As opposed to the pure synchronous pumping these schemes no longer possess wavelength tunability. Although they do not produce pulses as short as those generated by passive mode-locking they still have the advantage of providing somewhat higher output power and of easier synchronization to a subsequent amplifying system.

1.2 THEORETICAL DESCRIPTION. — The first analysis of passive mode-locking in the case when the saturable absorber has a long relaxation time compared with the pulse duration (slow absorber) was performed by New [22]. He showed that through the combined action of the saturable absorber and the saturable gain rapid pulse evolution can occur; where the pulse experiences a net loss on both its leading and trailing edge and a net gain in its centre. A steady state regime is, however, possible only in the presence of dispersion, since otherwise the leading edge will be sharpened indefinitely by the absorber response. Accordingly the steady state can be understood as a counterbalancing between pulse shortening and spreading mechanisms as well as

Fig. 1a, b. — Optimization of the intracavity dispersion. The autocorrelation trace in (b) is recorded after adding about 0.1 cm glass path as compared to (a) by translating the intracavity SQ1 prism [12]. Further increase of the glass path causes abrupt narrowing of the pulse spectrum and loss of the stable mode-locked regime.

between amplification and losses. The spreading is, in general, due to the bandwidth limiting properties of the cavity, which can be represented by an effective filter element. Using such a filter Haus [23] obtained the first closed form analytical solutions

(hyperbolic secant) for the pulse shape in the approximation of small pulse energy and of slight pulse shaping introduced by any of the cavity elements. These restrictions were left out in [24] and more general solutions were obtained on the basis of the rate equation approximation (REA). The model of Haus was further extended to include the CPM effects in the saturable absorber [25], which essentially lead to its enhanced saturation.

The phase modulation observed experimentally could be adequately accounted for in the theory only by considering the off-resonant light-matter interaction [26, 27]. In [26] down chirp was calculated due to the off-resonant interaction with the absorber molecules. In [27] minimum pulse duration was predicted around zero group velocity dispersion and asymmetrical behaviour of the pulse parameters was observed with respect to the dispersion sign. Addition of nonlinear index of refraction was found to increase the pulse width and did not influence the chirp. Accordingly the authors concluded (contrary to [2]) that the Kerr effect type nonlinearity is detrimental to achievement of the shortest pulses. Introducing a number of simplifying assumptions it was possible in [28] to estimate the phase shaping due to balancing only between dispersion and nonlinearity. As a result additional pulse shortening was predicted for favourable contribution of these two shaping mechanisms.

Here we present a new approach to the calculation of steady state complex pulse solutions in the passively mode-locked dye laser. We follow [29] in taking into consideration the finite phase memory of the resonant media but leave the assumption of small pulse energy as well as the assumption of a given (sech) pulse shape and phase function using instead a numerical iterative procedure. As opposed to all other models this approach allows one to predict the laser frequency in the mode-locked regime, which should not necessarily coincide with the maximum of the small signal net gain. We investigate here the dependence of the complex pulse parameters in the case of two near-resonant media (amplifier and absorber), the effect of addition of a second absorber (which may be interpreted as a photoisomer form) and the influence of linear dispersion and nonlinear refractive index elements. Finally, the validity of the obtained solutions as well as the possibility of coherent pulse propagation are discussed.

2. Basic equations for the light-matter interaction and the round trip model.

We describe the saturable media in the resonator by the density matrix formalism. The dye molecules are supposed to be represented by two-level systems with homogeneously broadened lines and with negli-

gible energy relaxation during the passage of the pulse. Furthermore, the propagation of the radiation is treated as one dimensional and only a single space coordinate (z) is needed. In addition, the effects of host medium dispersion are ignored. The system of density matrix and field equations in this case reads :

$$\left(\frac{\partial}{\partial t} - i\omega_j + 1/\tau_j \right) \rho_{12}^j = i\mu_{12}^j \Delta^j E \quad (1)$$

$$\frac{\partial \Delta^j}{\partial t} = \frac{-2i}{\hbar} (\mu_{12}^j \rho_{21}^j - \mu_{21}^j \rho_{12}^j) E \quad (2)$$

$$\left(\frac{-\partial^2}{\partial z^2} + \frac{\epsilon_j}{c^2} \frac{\partial^2}{\partial t^2} \right) E = -\gamma_j \mu_0 \left(\mu_{21}^j \frac{\partial^2 \rho_{12}^j}{\partial t^2} + \mu_{12}^j \frac{\partial^2 \rho_{21}^j}{\partial t^2} \right) \quad (3)$$

(γ_j : particle number density of the molecules of type j ; ω_j : centre frequency of the transition; τ_j : phase decay time; $\Delta^j = \rho_{22}^j - \rho_{11}^j$: inversion probability; μ_{12}^j : transition moment). On the time scale considered the concept of decomposing the electric field and the off-diagonal elements of the density matrix into slowly varying amplitudes and rapidly oscillating terms should still be justified and they are introduced by

$$E(z, t) = \frac{1}{2} \bar{E}(z, t) \exp[i(\omega_L t - k_L z)] + \text{c.c.} \quad (4)$$

$$\rho_{12}^j(z, t) = \bar{\rho}_{12}^j(t) \exp[i(\omega_L t - k_L z)], \quad (5)$$

where $\bar{E}(z, t) = |\bar{E}(z, t)| \exp[i\varphi(z, t)]$, ω_L is the carrier frequency of the pulse and k_L is the wave number at ω_L .

Following Haus [23] we assume only small changes $\delta \bar{E}$ of the pulse during a single transit through any of the cavity elements, which is experimentally verified in the femtosecond dye lasers [20, 27]. Thus the effect of the resonator components on the pulse shaping can be considered by the following relation :

$$\bar{E}'(\eta) = \bar{E}(\eta) + \delta_j \bar{E}(\eta), \quad (6)$$

where the prime denotes the modified amplitude after the interaction and η is the time in a frame moving with the group velocity v of the pulse ($\eta = t - z/v$). An approximate expression for $\delta_j \bar{E}(\eta)$ based on equations (1)-(5) was derived in [30] :

$$\delta_j \bar{E}(\eta) = \frac{g_j}{2\tau_j} \int_{-\infty}^{\eta} d\eta' \bar{E}(\eta') \times \exp \left[\frac{\eta' - \eta}{\tilde{\gamma}_j \tau_j} - \epsilon_j(\eta') \right]. \quad (7)$$

Here g_j is the small signal gain (absorption) coefficient at the leading edge of the pulse; $\tilde{\gamma}_j =$

$[1 + i\tau_j(\omega_L - \omega_j)]^{-1}$ is the complex lineshape factor and

$$\varepsilon_j(\eta) = \beta_j \tilde{J}_j^r \int_{-\infty}^{\eta} d\eta' |\bar{E}(\eta')|^2 \quad (\tilde{J}_j^r = \text{Re} \{ \tilde{J}_j \}) \quad (8)$$

is the pulse energy normalized to the saturation energy of the corresponding transition at ω_L , where $\beta_j = |\mu_{12}|^2 \tau_j / \hbar^2$. Note that the phase memory of the resonant media is taken into account in (7) and no passive filter is necessary for the resonator model, because the dispersion of the transition itself acts as a bandwidth limiting factor. A principal departure of equation (7) from the concept of REA and passive filter [23-27] is the temporal change of the filter properties due to the saturation taken into account in (7). Equation (7) can now be specified for the three media to be considered. We normalize all energy terms to the saturation energy of the gain medium at the laser wavelength. The amplification coefficient of the gain medium (a) is modified to account for the fact that the energy relaxation time of this dye (T_a) can be comparable to the cavity round trip time U [23]:

$$\delta_a \bar{E}(\eta) = \frac{g_a(1 - e^{-U/T_a})}{2\tau_a(1 - e^{-\varepsilon - U/T_a})} \times \int_{-\infty}^{\eta} d\eta' \bar{E}(\eta') \exp \left[\frac{\eta' - \eta}{\tilde{J}_a \tau_a} - \varepsilon_a(\eta') \right] \quad (9)$$

where $\varepsilon = \varepsilon_a(\infty)$. The two types of absorbing molecules (e.g. ground (b) and photoisomer (c) forms of a dye) generally exhibit different saturation energies and

$$\delta_{b,c} \bar{E}(\eta) = \frac{g_{b,c}}{2\tau_{b,c}} \int_{-\infty}^{\eta} d\eta' \bar{E}(\eta') \times \exp \left[\frac{\eta' - \eta}{\tilde{J}_{b,c} \tau_{b,c}} - m_{b,c} \varepsilon_a(\eta') \right] \quad (10)$$

($m_{b,c} = m_{b,c}^0 \tilde{J}_{b,c}^r / \tilde{J}_a^r$ are stability (saturation) parameters [22], where $m_{b,c}^0 = q\sigma_{b,c}/\sigma_a$ are the ratios of the corresponding interaction cross sections $\sigma_j = \mu_0 |\mu_{12}|^2 \tau_j \omega_L^2 / \hbar k_L$ multiplied by a focusing factor q ; $g_b = -\kappa(1-p)$ and $g_c = -\kappa p m_c^0 / m_b^0$, where κ is an effective absorption coefficient, which coincides with the total absorption if the two isomers have equal absorption profiles, and p is the photoisomer percentage).

The contribution of a nonlinear index medium (e.g. intensity dependent refractive index of the dye solvents) and of the linear dispersion elements (e.g. intracavity prisms) can be considered by expansion of the corresponding polarization terms in the non-resonant wave equation [31]. Neglecting higher order terms (nonlinear Schrödinger equation) we have

for the pulse shaping in the nonlinear refractive index element

$$\delta_c \bar{E}(\eta) = -in_c \tau_a \beta_a \tilde{J}_a^r |\bar{E}(\eta)|^2 \bar{E}(\eta) \quad (11)$$

and in the group velocity dispersion element

$$\delta_d \bar{E}(\eta) = \frac{ir}{2} \frac{d^2 \bar{E}(\eta)}{d\eta^2}. \quad (12)$$

The normalized parameters are defined by

$$n_c = \frac{q_c L_c k_L n_2}{\tau_a \beta_a \tilde{J}_a^r n_0} \quad (13)$$

and

$$r = \frac{4\pi^2 c L_d d^2 n_0}{\omega_L^3 \tau_a^2 d\lambda^2} \quad (14)$$

($n = n_0 + n_2 |\bar{E}(\eta)|^2$ is the nonlinear refractive index, $L_{c,d}$ are the lengths of the nonlinear and of the dispersive media, and the factor q_c takes into account possible focusing or colliding pulse effects in the nonlinear media). The effect of a small linear intensity loss γ (the outcoupling mirror) can be introduced in a similar manner:

$$\delta_f \bar{E}(\eta) = -\frac{\gamma}{2} \bar{E}(\eta). \quad (15)$$

Equations (10)-(12) and (15) are now combined in a unidirectional ring model shown in figure 2, where the relative position of each element is of no importance, because the corresponding transfer operators are expanded only to first order terms. For certain values of the laser parameters steady state solutions of the self-consistency equation

$$\bar{E}'(\eta + h) = \bar{E}(\eta) + \sum_{i=a,b,c,d,e,f} \delta_i \bar{E}(\eta) \quad (16)$$

(h : possible shift in the local time) exist, which

Fig. 2. — Schematic of the resonator model, a) amplifier, b, c) absorbers, d) dispersive element, e) nonlinearity, f) linear loss.

means that due to the counterbalancing action of the different elements the pulse reproduces itself in modulus and in phase after one round trip. Analytical solutions of (16) could be obtained only in the limit of small pulse energies by imposing additional restrictions on the pulse duration [29]. In the next section we present the results of an alternative approach, which does not employ further approximations and where the round trip equation (16) is treated numerically.

3. Results of the numerical simulation.

3.1 EVOLUTION OF THE STEADY STATE. — To obtain steady state solutions equation (16) is numerically iterated assuming an initial pulse shape or starting from noise. A similar approach was described in [32] in the case of real pulse solutions and REA. The treatment is greatly complicated, however, in the case of off-resonant interaction and complex pulse solutions because of the evolution of the laser centre (or mean) frequency. Up to now none of the models dealing with phase modulated pulses has predicted unambiguously the laser frequency. In the off-resonant REA approach the derived system of algebraic equations contained separate parameters corresponding to the laser centre frequency and a deviation from it [26]. In a similar theory [27] the value of the laser frequency was fixed *a priori* in the equations, although this parameter is of primary importance for the generation of chirp in the near-resonant interaction discussed. When considering phase memory effects the problem is additionally complicated and approximate estimations were made in [29] only in two limiting cases : at the peak of the small signal gain and at maximum energy (minimum net loss). From a general point of view the decomposition of the instantaneous frequency into a carrier frequency and instantaneous chirp is not unique. The results that we obtained using for simplification a fixed carrier frequency were considerably influenced by the value of this frequency due to the accumulation of « constant » chirp during the evolution process. That is why we imposed an additional requirement of vanishing mean frequency shift for the steady state solution besides the usual one of nearly constant pulse parameters after a certain number of round trips :

$$\tau_a(\bar{\omega} - \omega_L) = \frac{\tau_a \int_{-\infty}^{\infty} \frac{d\varphi}{d\eta} |\bar{E}(\eta')|^2 d\eta'}{\int_{-\infty}^{\infty} |\bar{E}(\eta')|^2 d\eta'} = 0. \quad (17)$$

This condition can be satisfied by correction of ω_L during the evolution process and corresponding correction of the Lorentzian lineshape functions.

The steady state is characterized by the steady state energy ε_{st} , the normalized pulse duration (FWHM) T/τ_a , the normalized relative mean frequency $\tau_a(\bar{\omega} - \omega_L)$ and the mean chirp

$$\tau_a^2 \bar{\omega} = \tau_a^2 \frac{\int_{-\infty}^{\infty} \frac{d^2\varphi}{d\eta^2} |\bar{E}(\eta')|^2 d\eta'}{\int_{-\infty}^{\infty} |\bar{E}(\eta')|^2 d\eta'} \quad (18)$$

which can be regarded as a measure for a possible pulse compression.

Fig. 3a. — Evolution of the pulse parameters as a function of the round trip number : energy ε (solid line), normalized pulse duration T/τ_a (dashed line), normalized mean frequency shift $\tau_a(\bar{\omega} - \omega_L)$ (dotted line). In the normalized mean frequency shift ω_L is the mean frequency of the steady state.

In figure 3a we show the evolution of the pulse parameters as a function of the round trip number. The evolution is strongly influenced by the net gain, but the steady state is independent of the initial conditions. Identical solutions are established when starting with a definite pulse shape and energy and from noise, which can be simulated by the spontaneous emission of the gain medium (Lorentzian intensity profile in the frequency domain and no phase correlations). The differences due to the initial conditions are smoothed during the first 100...200 round trips. In this first stage a strong yellow shift of the mean laser frequency to the maximum of the small signal net gain takes place depending on the value of the initial pulse energy. Shorter input pulses gain more rapidly energy in this stage. Steady state pulse energy and pulse duration are approximately approached in the first several hundreds of round trips, whereas fluctuations of the phase can occur in some cases for more than a thousand round trips.

A typical complex steady state solution is shown in figure 3b. Once such a steady state is approached the pulse parameters fluctuate by less than 0.1 % per

Fig. 3b. — Phase modulated steady state solution. The normalized pulse intensity (solid line) is defined by : $F(\eta) = \tau_a \beta_a \mathcal{Z}_a^r |\bar{E}(\eta)|^2$. The normalized relative instantaneous frequency (dashed line) is defined by $\tau_a(\omega - \omega_L) = \tau_a \dot{\phi}$.

round trip. As can be expected, due to the different shaping mechanisms at the leading and at the trailing edge, the pulse is rather asymmetrical.

All laser parameters used in the following analysis are summarized in table I.

3.2 RESONANT LIGHT-MATTER INTERACTION AND SINGLE ABSORBER. — In this simplest case (discussed in more detail in [30]) only three elements are taken into account in the resonator model : saturable gain (a) and absorption (b), and linear loss (f). Figure 4 shows the stationary pulse parameters as a function of the small signal gain. The dependence of the normalized energy is similar to the analytical results obtained without consideration of phase memory effects [24]. The steady state pulse

Fig. 4. — Resonant interaction. The normalized steady state pulse energy ε_{st} (solid line) and pulse duration T/τ_a (dashed line) are plotted *versus* the small signal gain g_a for three values of the absorption coefficient κ . For comparison the pulse energy calculated in [24] using the same parameters is given (dotted line).

duration, however, differs considerably from that calculated using REA and is in general shorter. At high net amplification (far above the threshold) it approaches the phase relaxation times of the transitions. The latter can be explained by the saturation of the dispersion of the transitions [30], which in our model is the only bandwidth limiting mechanism in the resonator.

3.3 OFF-RESONANT INTERACTION AND SINGLE ABSORBER. — This case can be investigated using the same three elements in the resonator, but now the two saturable media have different centre fre-

Table I.

Fig.	g_a	κ	m_b^0	m_c^0	τ_b/τ_a	τ_c/τ_a	$\tau_a(\omega_b - \omega_a)$	$\tau_a(\omega_c - \omega_a)$	γ	U/T_a	p	r	n_c
3	0.31	0.6	10	—	3	—	-1.0	—	0.05	1.0	0	0	0
4	v	v	7	—	1	—	0.0	—	0.0527	0.5	0	0	0
5	v	v	10	—	3	—	-1.0	—	0.05	1.0	0	0	0
6	v	0.6	v	—	3	—	-1.0	—	0.05	1.0	0	0	0
7	0.32	0.6	10	7	3	2	-0.2	-1.0	0.05	1.0	v	0	0
8	0.3	0.6	10	7	3	2	0.0	-1.0	0.05	1.0	0.3	v	0
9	0.5	v	v	—	1	—	0.0	—	0.0527	1.0	0	0	0

v : variable.

quencies. The corresponding transfer functions are complex and that is why there appears a phase modulation in the steady state. We focus here our attention on three parameters, which can be varied in experiment and which are of particular importance for the proper regime of the laser : the amplification coefficient (varied by the power of the pump laser), the absorption coefficient (varied by the concentration of the absorber dye or the thickness of the absorber jet) and the saturation parameter m_b^0 (varied by the adjustment of the waists in the resonator). In figure 5 the dependence of the pulse parameters on the small signal gain g_a is presented for two values of the small signal absorption κ . The increase of the pulse energy and the reduction of the pulse duration above the threshold have already been found in previous theoretical models. In general, however, the pulses obtained experimentally exhibit maximum stability and shortest duration near the threshold. A possible explanation for this is that the higher gain regimes can hardly be realized experimentally e.g. because of perturbations of the evolution process by the influence of the spontaneous emission. These effects are not taken into account in our model ; we note that the net amplification at the leading edge of the pulse in the steady state increases with g_a and can take positive values,

which may be detrimental for the pulse stability or which indicate the possibility of multiple pulse regimes.

The behaviour of the mean laser frequency *versus* the small signal amplification and absorption is in agreement with the characteristics experimentally obtained. It was not possible, however, to obtain for any set of parameters a frequency which is red shifted with respect to the centre of the absorption line. In terms of the net gain this can be explained by the symmetrical lineshapes used (which are a consequence of the model of homogeneously broadened two-level systems). Accordingly there exists always a position for the laser frequency between the centres of the two lines, where the absorption coefficient (as well as the saturation energy of the absorber) is the same as if the frequency was on the opposite side of the absorption curve, but where the gain is higher.

It is interesting to investigate the factors which determine the laser frequency in the mode-locked regime. If we neglect the feedback effect of the phase modulation there are two tendencies : pulling towards the maximum of the amplification line (evident when g_a is increased), and pulling towards the maximum of the absorption line in order to ensure easier saturation of the absorption (evident when κ is increased). There exists a point on the curve $\kappa = 0.5$ in figure 5, where the frequency

Fig. 5. — Off-resonant interaction. Normalized pulse energy ε_{st} (solid line) and pulse duration T/τ_a (dashed line). Normalized relative mean frequency $\tau_a(\omega_L - \omega_a)$ (solid line) and normalized chirp $\tau_a^2 \bar{\omega}$ (dashed line). Note that here (as well as in the next figures) the frequency dependent saturation parameters are smaller than the values defined in the centres of the corresponding lines.

Fig. 6. — Off-resonant interaction. In comparison with figure 5 κ is fixed here and the stability parameter m_b^0 is varied : $-m_b^0 = 15$, ε_{st} (solid line), T/τ_a (dashed line), $\tau_a(\omega_L - \omega_a)$ (solid line), $\tau_a^2 \bar{\omega}$ (dashed line) ; $-m_b^0 = 7.5$, ε_{st} (dotted line), T/τ_a (dash-dotted line), $\tau_a(\omega_L - \omega_a)$ (dotted line), $\tau_a^2 \bar{\omega}$ (dash-dotted line).

dependent saturation parameter m_b becomes less than unity. Above this point the frequency shifts to the yellow side of the gain line. At this point the interaction with the amplifier is resonant and it can be assumed that the chirp is produced only by the absorber. The chirp obtained here is actually a result of counterbalancing between the chirp generation in the absorber and the filter action of the amplifier. Under certain saturation of the two saturable media it is possible to produce negative chirp also in the case when the absorber is excited above and the amplifier below resonance. It should be outlined that in our solutions both media are strongly saturated as opposed to all other models, where the saturation is relatively weak [26, 29]. At energies greater than the saturation energy the dependence of the chirp on the saturation can differ considerably [33] from that in the case of weak saturation. Note, however, that the chirp in the steady state is derived here using a self-consistent method and can not be estimated from calculations of chirping through single passage [33].

The chirp production in the absorber can be investigated by varying the absorber bandwidth. Using e.g. the parameters for figure 6 (the case $m_b^0 = 15$) the increase of the absorber bandwidth ($\tau_b/\tau_a = 2$, instead of 3) results in a 20 % reduction of the pulse duration. The pulse energy is also decreased, which can be explained by the preserved mean frequency leading to decreased small signal net gain. The unchanged laser frequency means only stronger saturation of the absorber but unchanged conditions for the amplifier. Due to this modified absorber saturation the induced chirp is reduced by about 1/3 preserving its negative value. Such a behaviour indicates the primary importance of the absorber for the generation of chirp in this system.

A very important parameter for the established regime is the stability parameter m_b^0 , figure 6. In comparison with figure 5 the solutions for $m_b^0 = 15$ in figure 6 are characterized by increased pulse energy and decreased pulse duration. The higher value of m_b^0 (stronger saturation of the absorber) shifts the frequency towards the maximum of the absorber line. If the parameter m_b^0 is decreased (experimentally this means e.g. to reduce the focusing in the absorber) the frequency dependent saturation parameter m_b takes values smaller than unity and the laser frequency is shifted to the short-wavelength side of the amplifying line (the case $m_b^0 = 7.5$ in Fig. 6). The existence of such solutions having frequencies which provide easier saturation of the amplifier and not of the absorber cannot be explained by the stability criterion established by New [22]. We interpret this shift of the stability region from the region between the two lines to the short-wavelength arm of the amplifying line to be

caused by the temporal modulation of the pulse phase and the action of the phase memory.

3.4 THE EFFECT OF A SECOND SATURABLE ABSORBER. — The excitation of the saturable absorber DODCI results in the production of its photoisomer, which is accumulated until a steady state is reached in the flowing jet, where the back reaction is due to the photoisomer excitation with subsequent relaxation to the ground form [20]. The percentage of the photoisomer is a function of the laser wavelength and the pulse energy. The dependence estimated in [20] can be used to study the evolution of the equilibrium between the two isomer forms. For the purpose of simplification we assume here constant photoisomer percentage during the evolution of the steady state. The resulting dependence of the steady state parameters on the photoisomer percentage, cannot of course, be interpreted as a situation reached under constant values of all other laser parameters (although this situation can be experimentally simulated simply by exciting the absorber additionally with a tunable CW source or by changing the flow velocity). Such an approach should, however, give a qualitative idea of the influence of the photoisomer production when optimizing experimentally the pulse parameters by adjusting e.g. the pump power. It can be seen from figure 7, that the increase of the photoisomer percentage p pulls the laser frequency towards the maximum of the photoisomer absorption. Not very high concentrations of the photoisomer (of the same order as those measured in [20]) are sufficient to shift the laser wavelength to the long-wavelength side of the ground form absorption — a situation, which is relevant to the experimentally observed wavelengths. Further increase of the photoisomer percentage drives the system under the threshold, which can be interpreted as a result of the competition between the two mechanisms determining the laser wavelength (see 3.3).

A number of different regimes with respect to the phase modulation exist when varying the photoisomer percentage and consequently the laser frequency. In the region $p = 0.2 \dots 0.7$ the instantaneous frequency exhibits an initial decrease followed by an increase, whereas the minimum value moves forwards when p is increased. The net integral chirp is determined by the relative saturation of the two absorbers at the laser wavelength. The frequency dependent absorption coefficients on the other hand determine the individual contribution to the shaping of the leading edge. The photoisomer dominates as a shaping mechanism only if $p > 0.5$. The saturation of the two absorbers is of the same order in the discussed range of p . At $p = 0.1$ the mean laser frequency coincides approximately with the maximum of the ground form absorption, which is

Fig. 7. — Effect of the photoisomer production. Normalized pulse energy ϵ_{st} (solid line) and pulse duration T/τ_a (dashed line). Normalized relative mean frequency $\tau_a(\omega_L - \omega_a)$ (solid line) and normalized chirp $\tau_a^2 \bar{\omega}$ (dashed line). At $p = 0$ the laser frequency is already shifted to the yellow side of the amplifying line. The lineshape parameters are estimated from the corresponding absorption profiles [19].

Fig. 8. — Effect of introducing linear dispersion. Normalized pulse energy ϵ_{st} (solid line) and pulse duration T/τ_a (dashed line). Normalized relative mean frequency $\tau_a(\omega_L - \omega_a)$ (solid line) and normalized chirp $\tau_a^2 \bar{\omega}$ (dashed line).

strongly saturated. The integral chirp takes its minimum (negative) value, which could be expected from the analysis in the previous section. In this case the dominating chirping mechanism is, however, the photoisomer, whose contribution to the pulse shaping is negligible. The negative mean chirp, which is a result now of the monotonically decreasing instantaneous frequency across the pulse duration, is accompanied by an abrupt decrease of the pulse duration. Thus, minimum pulse duration on the red shifted arm of the ground form absorption occurs when one of the chirping mechanisms (in this case the photoisomer) is dominant. Nevertheless, the shortest pulses are chirped and not chirpfree.

The general features of the dependence of the steady state on the amplification coefficient, the absorption coefficient and the saturation parameters, which we discussed in 3.3 remain valid also with the addition of the photoisomer. Similarly, in the absence of frequency selective elements it was not possible to predict for any set of parameters a frequency lying on the red arm of the photoisomer absorption profile. As opposed to the case of a single absorber the integral chirp here takes relatively small values in a very large range for all laser parameters. This can be explained by the competition between the chirping action of the two absorbers when the laser wavelength is between the corresponding absorption lines and by the high degree of overlapping of the two relatively broad absorption profiles. An increase of the gain, high enough to pull the frequency to the yellow side of the amplifier line (see Fig. 5), results here (for all other parameters from figure 7 unchanged) in a drastic increase of the integral chirp, because in this case the two chirping mechanisms work in the same direction.

The dynamics of the DODCI-photoisomer equilibrium can be described in the following manner: increasing pulse energy and yellow shifted laser wavelength produce more photoisomer, which causes a frequency shift towards the photoisomer absorption maximum. This red shift of the laser wavelength increases the contribution of the photo-back reaction [20] and reduces the photoisomer production.

3.5 CONTROL OF THE INTRACAVITY CHIRP. — The introduction of linear dispersion affects the phase of the pulse and consequently the interaction with the near-resonant media. As a result a new balance between the different chirp generating processes is established. Figure 8 shows the calculated steady state parameters as a function of the dispersion, which takes both positive and negative values. The minimum pulse duration is obtained around zero group-velocity dispersion. A slight asymmetry similar to that obtained in [27] is also present. The shortest pulses are found to be chirped and not chirp

compensated [28]. This behaviour is probably due to the relatively weak chirp production in the saturable media in this case. No stationary solutions exist in the presence of linear dispersion combined with stronger chirping in the saturable media (e.g. as in the case in Fig. 5 and Fig. 6). The reason is a continuous shift of the laser frequency away from the maximum of the net gain and reaching finally the region below the threshold, where strong fluctuations of all pulse parameters take place. Thus, no equilibrium can be approached because of the non-linear character of the chirp induced in the saturable media and the possibility to compensate only the linear contribution with such a passive dispersive element.

Having in mind that n_c is positive in ethylene glycol, which is a typical solvent used, we investigated the effect of the nonlinear refractive index element in combination with negative group velocity dispersion (the same parameters as in Fig. 8). The nonlinearity alone can produce additional pulse shortening by about 20 %. The pulse shortening is accompanied by an increasing positive integral chirp. Further increase of n_c results in instabilities of the pulse phase and the pulse parameters. The minimum pulse duration when the two mechanisms work is independent of the value of the nonlinearity n_c , although it is approached for different values of the linear dispersion r . The shortest pulses are again phase modulated. We note, however, that for the proper interpretation of the interplay between dispersion and nonlinearity further investigation is needed not only in the cases when the saturable media produce stronger phase modulation but also with consideration of higher order dispersion terms.

3.6 π -SOLUTIONS. — The solutions, which we discussed in figure 4 are presented in a very broad stability region (as compared to the experimental one) and are stable also far above the threshold although the net amplification at the leading edge of the pulse is no longer negative [23, 24, 30]. The latter seems not to be a necessary condition in this numerical approach at least in the cases when no perturbations (e.g. pump laser fluctuations, spontaneous emission) are considered. At high net amplification, however, the pulse area

$$\theta = \frac{|\mu_{12}^a|}{\hbar} \int_{-\infty}^{\infty} d\eta' |\bar{E}(\eta')| \quad (19)$$

of the stationary pulse approaches values in the order of unity and coherent interaction can take place [34]. This possibility seems to be supported by the most recent femtosecond spectroscopic investigations of dyes [35], which revealed that the phase decay times could be much longer than expected. To account for this and also to check the validity of our

approximate solution (7), which only partially takes into account the phase memory, we modified our model in the simplest resonant case using directly the differential equations (1)-(3) instead the approximate solution (7). The integration is performed by only one integration step and assuming small amplification (absorption) coefficients as described in detail in [33]. This modification resulted in somewhat shorter pulse duration and increased pulse energy whereas the qualitative behaviour of figure 4 has

Fig. 9a. — Coherent light matter interaction. Steady state sech pulse obtained using $\kappa = 0.105$ and $m_b^0 = 4$.

Fig. 9b. — Variation of the pulse energy ε_{st} (solid line), area θ (dash-dotted line) and duration T/τ_a (dashed line) versus m_b^0 for two values of the absorption κ , indicated on the corresponding curves.

been preserved. We conclude therefore that equation (7) can adequately describe the light-matter interaction in the energy range used throughout the previous analysis and for pulse durations longer than the phase decay times. The pulse area in the case of coherent light-matter interaction stabilizes at values near π and is constant over a large range of the small signal amplification and the pulse energy. The steady state pulse shown in figure 9a can be very well approximated with a sech-amplitude and is extremely reproducible once the stationary regime is established. The integral parameters, for example the pulse energy, fluctuate only by 0.0001 % during 20 round trips, which lies in the region of computational errors. The symmetrical pulse shape indicates negligible contribution of the absorber in the pulse shaping. Indeed slight influence of the absorption coefficient κ is evident only at the threshold, where the area is slightly under π and the pulse duration — longer than the phase decay times. We note that the method used here for calculation of the steady state is equivalent to the case of coherent propagation of a pulse in a two-component amplifier [36], because the periodical interactions with the saturable media and the linear loss modify the pulse envelope only slightly and the stationary state is approached due to the condition (16). The pulse shown in figure 9a is a

2π pulse for the absorber (self-induced transparency). The deviation of the parameter m_b^0 from 4 (Fig. 9b) results in a deviation of the pulse area in the amplifier from π in such a direction so that the area of the pulse in the absorber tends to 2π . Unlike [36] we do not obtain a minimum pulse duration at $m_b^0 = 4$ but monotonical shortening of the pulse when increasing this parameter. Above the threshold the value of the absorption coefficient κ has an effect upon the pulse area and the pulse duration only if m_b^0 deviates from 4.

At present it can hardly be concluded whether π -pulse shaping mechanisms play an essential role in passively mode-locked dye lasers because some aspects of the interaction of femtosecond light pulses with organic dye molecules are still unknown (e.g. the role of homogeneous and inhomogeneous broadening, the values of τ_i). Work is in progress to use the model presented for the simulation of passive mode-locking also in the case of inhomogeneously broadened resonant media.

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