PARAMETERS THAT DETERMINE THE WAVELENGTH OF A PASSIVE MODE-LOCKED DYE LASER

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We introduce a new stability criterion that should be added to previous theoretical models of passive mode locked lasers in order to predict emission wavelength, which is to require that the pulse spectrum reproduces itself after one transit. The CPM laser consisting of rhodamine 6G as gain medium and DODCI as saturable absorber is studied. The relative population balance of the fundamental and photoisomer species of the DODCI are computed as a function of the pulse energy and wavelength. The necessity of a spectral stability criterion follows from the dependence of the gain profile on the pulse parameters. Use of the criterion is shown to agree with previous experimental results, such as the red spectral region of emission and dependence of emission wavelength on absorber concentration. This criterion is expected to be an useful tool in order to select other dye combinations and predict the laser behavior.

1. Introduction

Passive mode-locking of a CW dye laser was first demonstrated in 1972 [1] and the original choice of dyes, Rhodamine 6G as the gain medium and DODCI as the saturable absorber has been commonly used since then.

The primitive cavity designs were tunable, but the bandwidth limitation introduced by the tuning element prevented the obtention of the shortest pulses. Further designs led to pulses in the subpicosecond regime using the colliding pulse scheme [2]. The control of the group velocity dispersion through prisms [3,4] gave rise to even further pulsewidth reduction down below 30 fs [5]. It was also found that CPM systems presented an emission wavelength determined primarily by the absorber concentration [6].

Theoretical investigations were first conducted by New [7]. He showed the need to meet several stability criteria, such as requiring a net gain for the peak of the pulse but with net loss at both leading and trailing edges. Analytical solutions for the equations describing the behavior of passively mode-locked dye lasers were given by Haus [8] who also introduced an additional condition to satisfy the self starting requirements [9]. In more recent works Martinez et al. [10] and Haus et al. [12] included frequency modulation terms that gave rise to new analytical solutions. These solutions consisted of chirped pulses that depended on the group velocity dispersion and other frequency pulling terms. In these works the closed form solutions could be found after assuming the wavelength of emission of the laser. The model showed a good description of the behavior of the Rhodamine 6G-DODCI combination of dyes, but cannot be used to predict the behavior of other dye combinations unless the emission wavelength is determined independently.

A straightforward approach to this problem would be simply to choose the wavelength were net gain is maximum, but saturation of dyes makes gain very

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sensitive to changes in pulse features. This can be seen in fig. 1(a), which shows how the shape of the gain profile changes for different pulse energies, shifting the position of the maximum about 20 nm by increasing the degree of saturation. A subtler way in which the degree of saturation changes arises from changes in pulse wavelength. This alters the saturation energies, by changing the cross sections of the dyes as seen by the pulse. For a pulse wavelength of 580 nm, gain saturation energy is 2.1 mJ/cm^2, DODCI's is 0.21 mJ/cm^2 and photoisomer's is 4.8 mJ/cm^2, while for 620 nm same species saturation energies are 3.4, 2.7 and 0.24 mJ/cm^2 respectively.

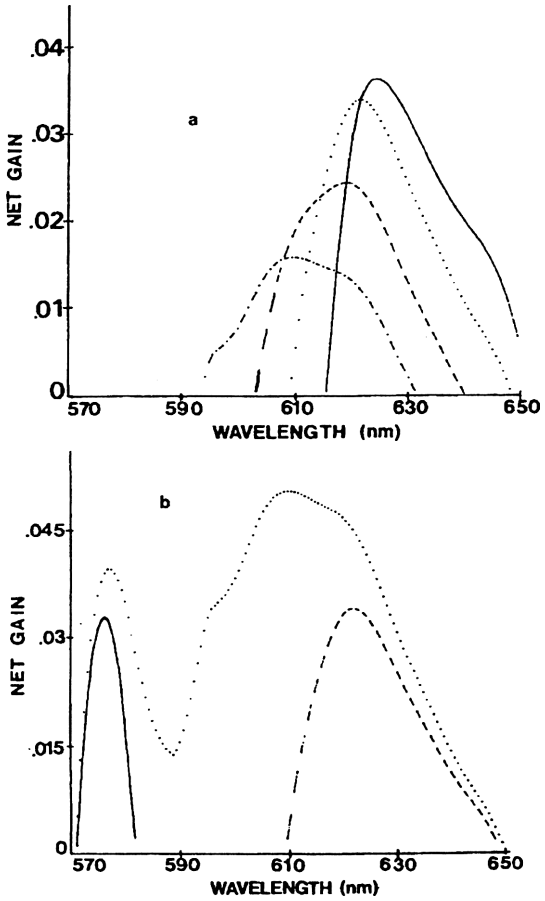

Fig. 1. (a) Net gain at the peak of the pulse for fixed wavelength (620 nm) and different energies (solid 2 nJ, dots 4 nJ, dashed 6 nJ and dash-dot 8 nJ). (b) Net gain at the peak of the pulse for fixed energy (4 nJ) and different wavelengths (solid 580 nm, dots 615 nm and dashed 620 nm).

This effect gives the dramatic result shown in fig. 1(b), where net gain is displayed for three pulse wavelengths. It is clear then that we must ask for self consistency in our model.

In this work we will try to introduce a new stability criterion that should be added to previous models in order to obtain a self consistent solution that also predicts the wavelength of the pulse to be emitted. This criterion is simply to request that the pulse spectrum repeats itself after each transit. In order to take into account this simple condition, one must consider the dependence of the dyes population inversions on the pulse energy and wavelength. This dependence should be added in a self consistent manner to the model that predicts the wavelength. In this work we will simplify this condition to requesting that the emission wavelength is such that the gain spectrum has a maximum at such wavelength after saturation by half of the pulse (pulse peak). Small signal gain is unimportant, as it only represents loss at the leading edge of the pulse. The starting wavelength may be different from the operating one, but as the pulse builds up from noise, the wavelength shifts to maximum gain at the peak of the pulse. Actually this is only a first estimate on the wavelength because, as shown by Martinez et al. [11], some frequency pulling might be necessary to achieve a stable solution with a chirped pulse. As such terms will only introduce a minor shift in the wavelength in typical operating conditions [12] it will not change the fundamental features presented here. Assuming typical energies for the pulse, we will use this criterion to predict the dependence of the emission wavelength on different parameters such as dye concentration, pump power and relative focusing on the gain and absorber dyes. Parameter ranges where stable solutions are found can also be predicted in this manner. The examples discussed will assume that Rhodamine 6G and DODCI are used as gain and absorber respectively (curves from [13] and [14]), and values of parameters of a typical CPM ring laser [5], such as a beam waist radius of 10 μm in the gain jet. Unless stated otherwise, a pulse energy of 6 nJ is assumed, and a spot area three times smaller in absorber jet.

2. Steady state regime

In order to compute net gain at the peak of the pulse, one must take in account the cross sections of the involved processes and the saturation dependent population of the states of the three dyes in the system (R6G, DODCI and its photoisomer). Both absorption and amplification are proportional to the population difference of the respective medium, being in the proportionality factor (the cross sections), where the wavelength dependence of the gain profile appears. We will derive expressions for the population inversion per unit area, including in such way the path traversed through the medium. During the passage of the pulse this population difference evolves after the following expression. Detailed derivation can be found in ref. [8],

$$n = n_0 \exp[-2U(t)/U_s] . \quad (1)$$

Here $U(t)$ is the cumulative energy of the pulse up to time t and U_s is the saturation energy. The factor 2 accounts for the pulse being so short (less than 100 fs) that no relaxation occurs during the passage of the pulse [15], but should be dropped if modeling a picosecond system. Between the passage of two successive pulses, population difference grows asymptotically to a certain value with a proper time constant. Populations at the arrival of the pulse are determined from requirement of a steady state regime.

The gain medium is the simplest, in steady state regime, pumping between the passage of two successive pulses exactly compensates the depletion of excited state population produced by the preceding pulse. This depletion depends on the energy and color of the pulse. A yellow pulse will decrease excited state population more efficiently than a red one of equal energy,

$$n_i = \frac{t I_p [1-\exp(-T/2t)]}{[1-\exp(-T/2t)][1+\exp(2U/U_1)]/2} . \quad (2)$$

Here n_i is the excited state population at the arrival of the pulse, U is the pulse energy, t is relaxation time for Rhodamine 6G and U_1 is its saturation energy. T is the period of the mode-locked pulse train, and I_p is the pump power.

Population of the DODCI can be transferred to the photoisomer by the pulse and population of photoisomer also experiences back isomerization [16]. Fig. 2(a) depicts the path by which such mixing of populations occurs. It is a simplification of the real processes that take place and involve a short lived twisted state. Absorption of energy during the passage of the pulse causes transition of both species to their excited levels. As the pulse is very short, all other processes may be neglected during the pulse, but they are important between the passage of two successive pulses. Desactivation from excited states with time constants as defined in the figure include both radiative and non-radiative processes. A very complete and recent description of DODCI can be found

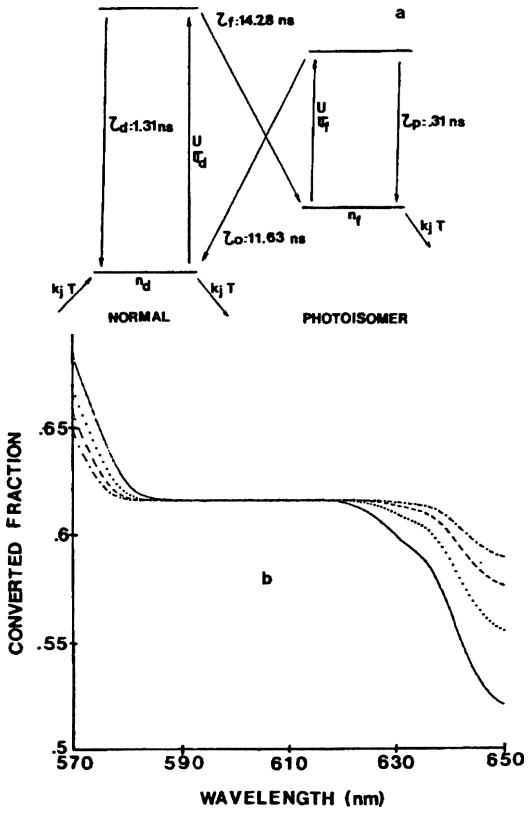

Fig. 2. (a) Simplified diagram of levels and transitions of the saturable absorber DODCI and its photoisomer. Time constants used in computation are defined. (b) Fraction of DODCI concentration that is converted to photoisomer as a function of pulse wavelength for different pulse energies (solid 6 nJ, dots 8 nJ, dashed 10 nJ and dash-dot 12 nJ).

in ref. [17]. Ground states are also affected by the circulation of the dye solution, which tends to increase normal species population.

When the laser is working, a fraction of the absorber concentration will be converted to the photoisomer form. Restricting our attention to ground state of normal form, and requiring its population not to change after one transit, an expression can be found for the converted fraction as a function of the energy and wavelength of the incoming pulse,

$$\frac{n_f}{n_d+n_f} = \frac{[1-\exp(-2U/U_d)]\tau_d}{2(\tau_d+\tau_f)} \times\left(k_j T+\frac{[1-\exp(2U/U_f)]\tau_p}{2(\tau_0+\tau_p)} + \frac{[1-\exp(-2U/U_d)]\tau_d}{2(\tau_d+\tau_f)}\right)^{-1}. \quad (3)$$

n_d and n_f are the population of normal and photoisomer species respectively, U_d and U_f are their saturation energies (wavelength dependent) and k_j accounts for the apparent relaxation due to dye renewal in the jet. Typical values give relaxation times around 700 ns. A plot of numerical computation of this expression is displayed in fig. 2(b) as a function of wavelength for different pulse energies. It can be seen that in the zone where both the photoisomer and the normal species saturate, converted fraction takes the value 0.616, regardless the wavelength and the energy of the pulse. When only one of the species is saturated, the converted fraction becomes more sensitive to the incoming pulse. For a yellow pulse, DODCI converts very efficiently to the photoisomer, but back isomerization is not so efficient because of the smaller cross section of the photoisomer in that wavelength. Converted fraction approaches both saturated value only for pulses with larger energies. An analogous but inverted situation rises when the pulse is red.

3. The criterion

From the preceding discussion, it is clear that net gain in the peak of the pulse will depend very strongly on the pulse that is travelling in the cavity. This can be seen in fig. 1(b), where the net gain is displayed as a function of wavelength for pulses of fixed energy and three different wavelengths. If the pulse is yellow, a maximum appears around 580 nm, while with a red pulse the maximum is around 620 nm. For pulse wavelength between these, both maxima are present. The exact positions of these maxima are very sensitive to the energy and wavelength of the incoming pulse. The dependence on pulse energy is shown in fig. 1(a). Keeping the wavelength fixed at 620 nm, a larger pulse energy shifts the position of the maximum gain to shorter wavelength.

If a steady state regime is to be achieved, with a pulse of well defined energy and wavelength, the position of the maximum of net gain at the peak of the pulse must coincide with the wavelength of the pulse, otherwise a frequency pulling term would appear and the pulse would not repeat itself after a transit. This condition represents a stability criterion on emission wavelength, and severely restricts the possible emission wavelengths of the system. An immediate representation that will help to make clear the influence of the different parameters is shown in fig. 3. The position of the maximum for a fixed pulse energy is plotted as a function of the pulse wavelength for different sets of parameters. It can be easily seen that there is only one wavelength that fulfills our stability criterion; the one where each curve intersects the diagonal near 620 nm. Crossing the diagonal from upside ensures stability against perturbation of the system, as can be understood considering that a perturbation of either sign shifts maximum position as to produce a restoring frequency pulling.

It should be noted, however, that this only means a possible emission wavelength of the system. In order to predict emission wavelength the criterion must be used in conjunction with a model that predicts the rest of the features of the steady state solution, such as pulse energy (for example [11], completed in a self consistent way). We study the effect of different parameters of the laser system upon the emission wavelength, as given by the criterion for fixed pulse energy i.e. we are disregarding changes in the pulse caused by the parameter we are varying. If taken in account, that would change the exact dependence of emission wavelength upon laser parameters, but not the fundamental features presented here.

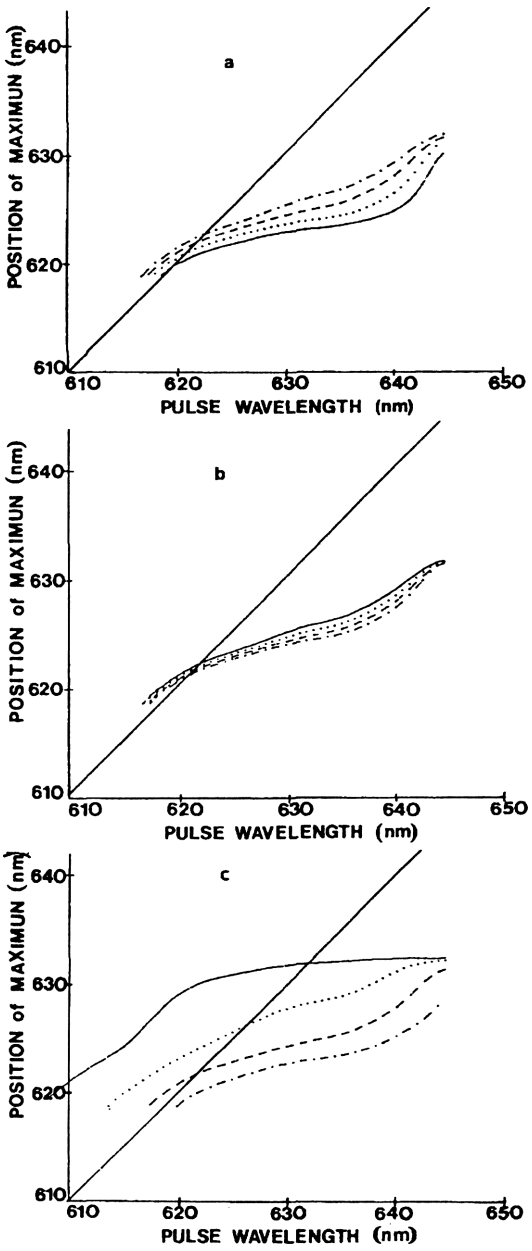

Fig. 3. Representation of stability criterion. Position of maximum gain as a function of the pulse wavelength. Stable wavelength is the one where the curve intersects the diagonal. (a) Effect of absorber concentration. Curve of maxima for different absorber concentration (solid 3.0, dots 3.6, dashed 4.2, dash-dot 4.8×10^{15} l/cm^2). (b) Effect of pump power. Curve of maxima for different pump powers (solid 2.86, dots 3.04, dashed 3.22, dash-dot 3.40×10^{23} phot/cm^2/s). (c) Effect of relative focusing. Curve of maxima for different relative focusing in the gain and absorber medium (focusing ratio: solid 1, dots 2, dashed 3, dash-dot 4).

4. Dependence on laser parameters

Any change in laser parameters alters the position of the maximum gain for given pulse energy and wavelength, and thus changes the shape of the curve of maxima. The modified curve will intersect the diagonal in some different wavelength, or may not intersect it at all. This means the system would not work for such particular choice of parameters.

It is well known, though it was rather surprising at first, that the emission wavelength of the passive mode locked dye laser is determined primarily by the concentration of the absorber dye [6]. Fig. 3(a) shows how the curve of the maxima is affected by the concentration of DODCI. Increasing the absorber concentration shifts the only stable operating wavelength further to the red. This behavior can be interpreted as follows: for a red pulse the photoisomer is deeply saturated because of its larger cross section, and its absorption at the peak of the pulse may be neglected. Absorption of the normal species is still important, as it saturates near the peak of the pulse. Increasing absorber concentration increases the loss for the short wavelength side and thus shifts the maximum to longer wavelength. For a sufficiently small absorber concentration, the curve of maxima keeps below the diagonal and no stable solution is found near 620 nm, but another one is found around 580 nm.

In fig. 3(b) we study the effect of pump power. Increasing pump power shifts down the curve of maxima. This behavior offers a new interpretation for bistability reported on CPM systems [18]. For a given absorber concentration they increase pump power until the curve of maxima is pushed below the diagonal and laser wavelength jumps to yellow, where the threshold for continuous operation has been surpassed. In that situation the fraction of DODCI concentration that is converted to the photoisomer is larger than the one in the mode locked regime. Increased absorption in the red thus prevents the laser from jumping to red again, even if pump power is lowered below the point where the first transition occurred.

Another parameter that affects emission wavelength is the relative focusing on the gain and absorber. As it can be seen in fig. 3(c) (focusing ratio is defined as gain beam area over absorber beam

area), stronger focusing in the absorber shifts the emission wavelength to orange. As discussed in connection with fig. 3(a), we are only interested in absorption by the normal species of DODCI. Strong focusing favours saturation with an overall effect of decreasing absorber concentration at the peak of the pulse.

5. Conclusion

In conclusion we have shown the need of an additional stability criterion on emission wavelength of passive mode-locked dye lasers, consisting in requesting the pulse spectrum to repeat itself after each transit. Self consistency is essential because the spectral profile of the net gain depends dramatically upon pulse features as energy and central wavelength. Influence of different laser parameters according to this new criterion is shown to agree with reported behavior. In order to give detailed dependence of emission wavelength upon laser parameters, the criterion should be used in conjunction with a model that predicts pulse features from this parameters. Nevertheless, this criterion alone can be useful to predict possible emission wavelength of new and yet not experimented gain/saturable absorber dye combinations, and with minor modifications it would be suitable for the case of femtosecond hybrid mode locked lasers.

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