Appl. Phys. B 30, 135–142 (1983)

Applied Physics B Photophysics and Laser Chemistry

Theory of Synchronously Pumped Dye Lasers

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Received 3 November 1982/Accepted 19 November 1982

Abstract. An analysis of cw synchronously pumped dye lasers is presented. Under the assumption that the cavity (tuning element) bandwidth is much wider than the bandwidth of the transform limited pulses generated, the pulse forming dynamics is rigorously treated. It is shown that for a finite mismatch between the lengths of the dye and the pump lasers, a steady-state pulse develops in the dye laser cavity with a conserved pulseshape. The characteristics (energy, shape, peak power, duration) of these pulses of ultimate width are quantitatively determined as a function of cavity mismatch. An analytical solution for the pulse envelope is determined, which yields $I(t) \propto \mathrm{Sech}^2(t/t_p)$ to a good approximation.

PACS: 42.55, 42.60, 42.80

Synchronously pumped dye lasers have recently been widely used for picosecond spectroscopy. Previously published theoretical studies of this method of mode locking have either been numerical or qualitative [1–9]. A quantitative description of the properties of the ultrashort pulses generated in a synchronously pumped mode-locked dye laser necessitates an accurate description of the gain dynamics in the dye medium, taking into account the cavity configuration of a typical dye laser. In this paper, the evolution of the intracavity intensity from spontaneous emission through many round trips is rigorously analyzed under the conditions of a periodic and synchronous excitation of the dye medium and an existing cavity mismatch, for a typical dye laser. It is shown that in an ideal (no cavity length and/or pump pulse perturbations, no bandwidth limitation) synchronously pumped laser, a steady state develops in which the dye pulseshape is indefinitely conserved, the saturable gain mechanism resulting in an effective shift of the pulse and compensating the existing mismatch.

In Sect. 1.1–4 the properties of these ultimate pulses are quantitatively determined almost exactly in a semianalytical formalism. In Sect. 1.5 an approximate analytical solution is found which yields the pulseshape $I(t) = I_m \mathrm{Sech}^2(t/t_p)$ with I_m and t_p given in terms of the fundamental system parameters. In Sect. 1.6, the condition for the formation of a satellite pulse following the main pulse is given.

In Sect. 2, computed results are presented. The approximate (sech^2) solution is found to be an excellent approximation. The critical dependence of the pulsewidth and energy on the mismatch between the dye and pump cavity lengths is elucidated. It is shown that under typical conditions, dye laser pulses of width $\leqq 2$ ps can be generated only if the mismatch is $\leqq 5\,\mu$m. The reduction of pulsewidth in this region is at the expense of a substantial decrease in pulse energy and susceptibility to the formation of satellite pulses. The application of the theory to practical experimental situations and its limitations are also discussed in Sect. 2.

1. Theory

1.1. Interaction of a Dye Laser Pulse and a Pump Pulse in the Dye Medium

Consider a jet stream, three mirror folded cavity dye laser [10] which is pumped by a cw periodic train of mode locked pulses of width 50 ps→100 ps. When the cavity lengths of the pump laser and the dye laser are set close to equal, a dye laser pulse is generated, which is much shorter than the pump pulses near perfect synchronism. A typical situation is described in Fig. 1.

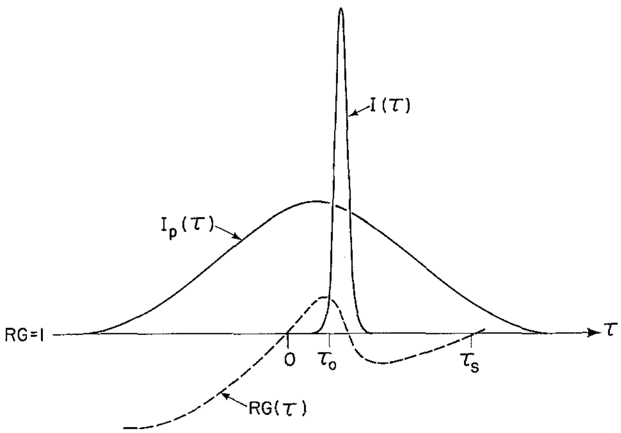

Fig. 1. Typical relative positions of the pump and dye pulses in local time. Dashed curve describes the gain $G(\tau)$ for the dye pulse. $\tau=0$ is the threshold instant and τ_0 is a local time at which the dye pulse begins to become appreciable. If the pump pulse has excessive energy, a second threshold is reached at τ_s after which a satellite pulse is to be formed

In the rate equation approximation and neglecting excited state and triplet absorptions[1], it can be shown [7] that the gain the dye pulse experiences at a transit through the dye jet satisfies

$$\frac{dG}{d\tau}+\sigma G\{I(\tau)(G-1)+I_p(\tau)[(G/G_{\rm eq})^\eta-1]\}=0\,, \tag{1}$$

where $\eta=\sigma_p/\sigma$, $G_{\rm eq}=\exp(\sigma_e n_T d)$, $\sigma\cong\sigma_e$ effective (including the effect of angular hole burning [11]) emission cross-section at the dye laser wavelength, σ_p: effective absorption cross-section at the pump wavelength, n_T: number of dye molecules/cm^3, d: pathlength inside the dye medium. In (1), it is assumed that the pump and the dye pulses are collinear in the dye and the fluorescence within the short duration of the intense pump pulse is negligible. Typically $\sigma_e n_T d\cong 3$ and $G(\tau)\ll G_{\rm eq}$, which correspond to a complete but unsaturated pump absorption. Then, (1) can be written to a very good approximation as

$$\frac{dG}{d\tau}+\sigma G[I(\tau)(G-1)-I_p(\tau)]=0\,. \tag{2}$$

As shown in Appendix A, G is here redefined such that (2) takes into account the two transits through the dye jet in a round trip by means of the appropriate initial condition. With τ_0 defined as the instant in local time after which the dye pulse rapidly develops, the dye pulse can be neglected for $\tau\leqq\tau_0$ and the round trip gain can be written as

$$G(\tau)=G_2(\tau_0)G_1(\tau)\,, \tag{3}$$

[1] Triplet absorption can be approximately taken into account by including it in linear cavity losses

where $G_1(\tau)$ describes the rise of gain from the ground state absorption level $G_a=\exp(-\sigma_a n_T d)$ (σ_a: absorption cross-section at the dye laser wavelength) under the excitation of the pump pulse, i.e., $G_1(\tau)$ is the solution of (2) (with $I=0$) with initial condition G_a, and is given by

$$G_1(\tau)=G_a X_p(\tau)\,, \tag{3a}$$

where $X_p(\tau)=\exp(\sigma E_p(\tau))$ and $E_p(\tau)=\int_{-\infty}^{\tau} I_p(t)dt$. In the above, a sufficiently long cavity length is assumed such that when the pump pulse arrives at the dye jet (dye pulse propagating to the output mirror and back) the excited dye molecules are all relaxed to the ground state (fluorescence lifetime $\leqq$ cavity transit time), which in general is a good approximation.

The initial condition that supplements (2) for $\tau_0\leqq\tau$ is thus $G(\tau_0)=G_2(\tau_0)G_1(\tau_0)$ where $G_1(\tau_0)$ is given by (3a) and $G_2(\tau_0)$ describes the initial value of gain for the second pass and depends on the saturation level of the dye molecules, after the first transit of the dye pulse. (The time between the two transits and the corresponding relaxation is generally small.) It is given by (Appendix B)

$$G_2(\tau_0)=G_a\{XX_p/[1+G_1(\tau_0)(X-1)]\}^\gamma\,, \tag{4}$$

where

$$\gamma=\exp(-T_m/T_f)\,,\qquad X_p=\exp[\sigma E_p(\infty)]\,,$$

$$X=\exp[\sigma E(\infty)]\,,\quad\text{and}\quad E(\tau)=\int_{-\infty}^{\tau} I(t)dt\,.$$

In the above T_m is roughly two-way travel time to the spherical end mirror ($\sim$300 ps) and $T_f=S_1\to S_0$ fluorescence lifetime.

For $\tau>\tau_0$, the dye pulse develops to a peak intensity much higher than the pump pulse within a few ps. Hence, the pump pulse can be neglected in the dye pulse region and the solution of (2) (with $I_p=0$) with initial value $G(\tau_0)$ can be written

$$G(\tau)=G(\tau_0)X(\tau)/[1+G(\tau_0)(X(\tau)-1)]\,, \tag{5}$$

where $X(\tau)=\exp[\sigma E(\tau)]$.

Starting with intensity $I(\tau)$ and experiencing gain $G(\tau)$ and linear cavity losses (defined by reflectivity R), the final dye laser pulse at the end of a full cavity round trip, I_f, is given by (Appendix A)

$$I_f(\tau+\Delta)=RG(\tau)I(\tau)\,. \tag{6}$$

In (6), $\Delta=(L-L_p)/c$ is the mismatch parameter accounting for the difference in arrival times of the pump and dye pulses at the dye medium due to imperfect synchrony, where L and L_p are the dye and pump laser cavity lengths, respectively, and c is the speed of light.

1.2. Stable Pulse Energy

It is shown in Appendix C that in an ideal synchronously pumped dye laser in which the bandwidth of tuning elements ≫ bandwidth of the generated transform limited pulses and $\Delta > 0$, a steady-state develops in which the pulse shape and therefore its energy are conserved. This can be mathematically stated as

$$\int_{-\infty}^{\infty} I_f(\tau)d\tau = \int_{-\infty}^{\infty} I(\tau)d\tau = E(\infty).$$

Substituting (5) and (6) into the above equation and integrating the right hand side, we obtain for the conserved pulse energy

$$X^{1/R} = 1 + G(\tau_0)(X-1). \tag{7}$$

Substituting the expression for $G(\tau_0)$ from (3), (3a), and (4), we can rewrite (7) as

$$X^{1/R} = 1 + G_a G_1(\tau_0) \cdot \{XX_p/[1+G_1(\tau_0)(X-1)]\}^{\gamma}(X-1). \tag{8}$$

In general, $\sigma E(\infty) \ll 1$ and (8) can be approximately solved to the first order as

$$\sigma E(\infty) = 2R[RG_a G_1(\tau_0)X_p^{\gamma} - 1]/\{1 + R^2 G_a G_1(\tau_0)X_p^{\gamma}[1 - 2\gamma(G_1(\tau_0) - 1)]\}. \tag{8a}$$

Given $G_1(\tau_0)$ one can solve the algebraic equation (8) or use (8a) for the pulse energy. Even though the dye pulse can be neglected for $\tau < \tau_0$, the time τ_0 [and hence $G_1(\tau_0)$] depends on the mismatch Δ. In a typical experimental situation, a certain mismatch exists, which leads to a particular τ_0. Hence, the evolution of the dye pulse under the conditions of a given mismatch and pump pulse must be studied in order to determine the corresponding τ_0 [or $G(\tau_0)$]. This will be treated in the next section.

1.3. Parameters Describing the Steady-State Pulse Properties

In Appendix C, it is shown that in an ideal synchronously pumped dye laser cavity, the spontaneously emitted fluorescence builds up into an ultrashort pulse which, after a sufficient number of round trips in the cavity, reaches a steady state with its shape conserved and satisfying

$$I(\tau) = RG(\tau)I(\tau - \Delta) + I_{sp}[G(\tau) - 1], \tag{9}$$

where I_{sp} is the intensity of spontaneous emission within the solid angle subtended by the beam.

As depicted in Fig. 1, the local time (τ) domain is divided into three regions. $\tau = 0$ is defined as the threshold time, where

$$RG(0) = 1. \tag{10}$$

For $\tau \leqq 0$, spontaneous emission is dominant and intensity reaches a steady-state given by

$$I(\tau) = I_{sp}\{G(\tau)(1-R)[1 + RG(\tau - \Delta) \cdot [1 + RG(\tau - 2\Delta)[1 + \ldots[\ldots]]]] - 1\}, \tag{11}$$

$$I(0) = I_{sp}\{G(0)(1-R)[1 + RG(-\Delta) \cdot [1 + RG(-2\Delta)[1 + \ldots[\ldots]]]] - 1\}. \tag{11a}$$

The above series converges to a finite value $I(0) \gg I_{sp}$. Hence, for $\tau \geqq 0$, we can neglect spontaneous emission and the conserved pulseshape satisfies

$$I(\tau) = RG(\tau)I(\tau - \Delta) \tag{12}$$

with $G(\tau)$ satisfying (2).

Given Δ, (12) and (2) can be simultaneously solved using the initial conditions $G(0) = 1/R$ and $I(0)$ as determined from (10). However, since $G = G_1 G_2$ and G_2 is a function of the total pulse energy, pulse energy X must be found to determine the threshold time $\tau = 0$. Hence, in order to determine the relation between τ_0 and Δ, $I(\tau)$ must next be determined over $0 \leqq \tau \leqq \tau_0$. Iterating (12) for n steps of Δ

$$I(n\Delta) = R^n G(n\Delta) \ldots G(\Delta)I(0) = I(0)[RG_2(\tau_0)]^n \exp\left[\sigma \sum_{k=1}^{n} E_p(k\Delta)\right],$$

where (3) and (3a) are used and ground state absorption is neglected ($G_a = 1$). Letting $n\Delta = \tau$,

$$I(\tau) \simeq I(0)[RG_2(\tau_0)]^{\tau/\Delta} \exp\left[\frac{\sigma}{\Delta}\int_0^{\tau} E_p(t)dt\right], \tag{13}$$

assuming that the pump pulse does not change significantly over intervals of the order of Δ. Hence

$$I(\tau_0) = I(0)\left\{[RG_2(\tau_0)]^{\tau_0} \exp\left[\sigma \int_0^{\tau_0} E_p(t)dt\right]\right\}^{1/\Delta}. \tag{14}$$

As shown in Appendix C, $I(\tau)$ increases monotonically with τ until it reaches a high peak value. The local time τ_0 is so far arbitrarily defined to be an instant at which the dye pulse starts to become significant. A study of (2) reveals that $I(\tau)$ tends to deplete (saturate) the gain while the pump pulse $I_p(\tau)$ is increasing it. Initially $I(\tau)$ is negligible, but near the pulse region it rises rapidly and attains intensities $\gg I_p$ within a few ps. Within the pulse region $I_p(\tau)$ is negligible. The boundary between these two regions can be conveniently defined to be at $\tau = \tau_0$ at which

$$\left.\frac{dG}{d\tau}\right|_{\tau=\tau_0} = 0,$$

i.e., when the dye pulse has reached a value the saturation effect of which compensates the excitation by the pump pulse. Then, from (2)

$$I(\tau_0) = I_p(\tau_0)/[G(\tau_0) - 1]. \tag{15}$$

Given the pump pulse and the fundamental dye laser constants, the important parameters that define the properties (energy, shape) of the generated steady-state ultrashort pulse are thus determined from a simultaneous solution of (15), (14), (11a), (10), and (8), and use of (3), (3a), and (4). An appropriate procedure is as follows: Given $G_1(\tau_0)$ (or time τ_0) (8) can be solved for the total pulse energy (X), which through (4) yields $G_2(\tau_0)$. The threshold instant $\tau=0$ is then determined from (10) using (3) and (3a). Using the value of $G_2(\tau_0)$ and (15), one can simultaneously solve (14) and (11a) for Δ. Thus, simultaneous solutions of $G(\tau_0)$ and Δ are found, together with $I(\tau_0)$ and the total pulse energy.

1.4. Numerical Solution of Pulseshape

In order to determine the pulseshape for $\tau \geqq \tau_0$ one can iteratively solve

$$G(\tau+\Delta)=G(\tau)\{1-\sigma\Delta[I(\tau)(G(\tau)-1)-I_p(\tau)]\}$$
$$I(\tau+\Delta)=RG(\tau)I(\tau)$$

in steps of Δ, starting at τ_0 with the initial conditions $G(\tau_0)$ and $I(\tau_0)$. Such an approach has been recently described also by Catherall et al. [1]. Note that the value of Δ which corresponds to the initial gain $G(\tau_0)$ is as determined in Sect. 1.3 and it must be only a small fraction of the pulsewidth in order that the pulse is stably maintained. (The saturable gain mechanism can yield a forward shift of the pulse only a small fraction of pulsewidth.)

1.5. Approximate Analytical Solution

The numerical solution of the previous sections yields the pulseshape and energy of an ideal conventional synchronously pumped dye laser as a function of cavity mismatch and pump pulse and dye laser cavity characteristics (linear losses, cavity configuration, dye constants etc.) almost exactly. In this section, we introduce an approximation which leads to the analytical determination of the steady-state pulse characteristics.

Let $t=\tau-\tau_0$ and $I(\tau_0)=I_0$, $G(\tau_0)=G_0$. Neglecting the pump pulse I_p in the vicinity of the dye pulse, the steady-state pulse satisfies

$$I(t+\Delta)=RG(t)I(t),$$

where $G(t)$ is given by (5). Iterating the above equation in steps of Δ, and letting $n\Delta=t$ and $G(t)=\exp[g(t)]$, we obtain

$$I(t)=I_0\left\{R^t\exp\left[\int_0^t g(t')dt'\right]\right\}^{1/\Delta}, \tag{16}$$

assuming that g varies slowly over intervals of Δ. Typically, cavity mismatches are of the order $\lesssim 10\,\mu\text{m}$ and pulsewidths are ~ 1 ps. Hence, $\Delta \ll$ pulsewidth and the change in I and g over Δ is small.

For small total pulse energies, gain $G(t)$ is only slightly depleted; then, we can approximate (2) by

$$\frac{dG}{dt}+\sigma GI(t)(G_0-1)=0. \tag{17}$$

The solution of (17) is

$$g(t)=g_0-\sigma\int_0^t(G_0-1)I(t')dt'. \tag{18}$$

Substituting into (16)

$$I(t)=I_0\exp\left\{2\int_0^t\left[\alpha-\beta\int_0^{t'}\sigma I(t'')dt''\right]dt'\right\},$$

where

$$\alpha=\ln(RG_0)/2\Delta, \quad \beta=(G_0-1)/2\Delta. \tag{19}$$

Let $E(t)=\int_0^t I(t')dt'$. Then,

$$\frac{dE}{dt}=I_0\exp\left[2\int_0^t(\alpha-\beta\sigma E)dt'\right].$$

Differentiating

$$\frac{d^2E}{dt}=2\frac{dE}{dt}(\alpha-\beta\sigma E)=\frac{d}{dt}(2\alpha E-\beta\sigma E^2)$$

which we can rewrite as

$$\frac{dE}{dt}-I_0=2\alpha E-\beta\sigma E^2. \tag{20}$$

The above equation (Ricatti) for pulse energy is analytically solvable. Its solution yields

$$E(t)=I_0[1-\exp(-2qt)]/[q_1+q_2\exp(-2qt)], \tag{21}$$

where

$$q=(\alpha^2+\beta\sigma I_0)^{1/2}\simeq\alpha$$
$$q_1=q-\alpha \simeq\beta_0\sigma I_0/2\alpha$$
$$q_2=q+\alpha \simeq 2\alpha.$$

After differentiation of (21) the corresponding pulseshape is given by

$$I(t)=I_m\,\mathrm{Sech}^2[q(t-t_m)], \tag{22}$$

where

$$I_m=q^2/\sigma\beta=(\ln RG_0)^2/2\sigma\Delta(G_0-1),$$
$$t_m=\ln(q_2/q_1)/2q.$$

The full width at half-maximum pulsewidth is

$$t_p=1.7627/q\simeq 3.5\Delta/\ln(RG_0). \tag{23}$$

Hence, the pulseshape is "sech^2" with the peak of the pulse occurring at τ_0+t_m. Notice that the important pulse characteristics (I_m, t_p) are only very slightly dependent on I_0 (therefore on I_{sp}). This is expected since as $I(t)$ increases around $t=0$, $G(t)$ does not change until the pulse attains appreciable energy in the leading edge. Hence, a considerably different value of I_0 would equally well correspond to approximately the same G_0, while for a given G_0 one must still get a unique pulseshape and energy. The value of I_0 affects t_m, resulting in an unimportant shift of the pulse peak relative to τ_0.

With these analytical results and the relation between Δ and G_0 determined in Sect. 1.3, all the properties of stable pulses generated by conventional synchronously pumped dye lasers are quantitatively determined.

1.6. Satellite Pulses

Another important characteristic of synchronously pumped dye lasers is the condition for satellite formation. If the pump pulse has sufficient energy a second threshold may be reached to develop a satellite pulse following the main pulse, as depicted in Fig. 1. The condition for the formation of a satellite pulse and its position in local time with respect to the main pulse is determined as follows:

At the end of the dye pulse the gain is depleted to a value G_{sat}, given by the final value of (5)

$$G_{sat}=G(\tau_0)X/[1+G(\tau_0)(X-1)]. \tag{24}$$

$G(\tau)$ then increases due to the remaining energy in the pump pulse, in accordance with (2) ($I=0$). Hence, for $\tau \geqq \tau_0$

$$G(\tau)=G_{sat}\exp\{\sigma[E_p(\tau)-E_p(\tau_0)]\}. \tag{25}$$

The condition for the formation of a satellite is $RG(\infty)>1$, which, using (25), can be written

$$RG_{sat}\exp\{\sigma[E_p(\infty)-E_p(\tau_0)]\}>1. \tag{26}$$

If the above inequality is valid, the satellite appears slightly after $\tau=\tau_s$, where

$$RG_{sat}\exp\{\sigma[E_p(\tau_s)-E_p(\tau_0)]\}=1. \tag{27}$$

Given Δ, the solution in Sects. 1.1–1.3 yield the values of τ_0, $G(\tau_0)$ and the total pulse energy X. With the use of these values in (24) and (27), the position of a possible satellite pulse is determined. It is seen that R must be decreased (higher linear losses) to eliminate the satellite pulse since both τ_0 and τ_s are then shifted towards the end of the pumping pulse.

2. Numerical Results and Discussion

In Fig. 2, computed exact pulsewidth, peak power and pulse energy are plotted as a function of cavity mismatch for a typical experimental situation. The peak 2nd harmonic intensity, to be obtained in an autocorrelation pulsewidth measurement set-up, and the approximate (sech^2) solutions are also given.

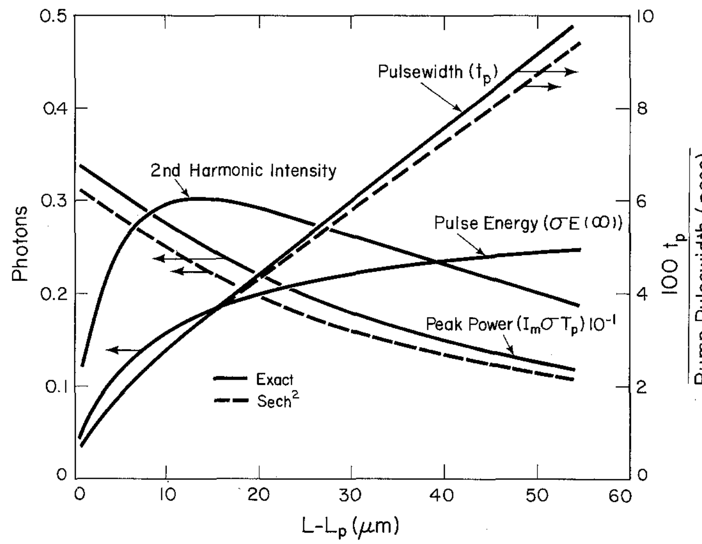

Fig. 2. Characteristics of synchronously pumped dye laser pulses as a function of cavity mismatch. [L: dye laser cavity length, L_p: pump laser cavity length, I_m: intracavity peak power, t_p: pulsewidth, and $\sigma E(\infty)$: intracavity energy/pulse.] The following constants are used: $R=0.5$, $G_a=1$, $I_p(t)=\sigma E_p(2\sigma T_p)^{-1}\operatorname{sech}^2(t/T_p)$, $\sigma E_p=0.5$ photons, $I_{sp}=5\times10^{17}$ ph/cm^2 s, and $\sigma T_p=1.13\times10^{-26}$ cm^2 s. 2nd harmonic intensity is in arbitrary units

Since, typically, Δ is a very small fraction of a ps whereas pump pulses are $\cong 100$ ps, (11a) is simplified for computations as follows: Dividing the region $\tau\leqq 0$ into N intervals, each $K\Delta$ long, such that $\tau=-NK\Delta$ is before the pump pulse, assume $G=$const in each interval. From iteration of (9) in the j^{th} interval over K steps of Δ, it follows that

$$I_j=(RG_j)^K I_{j-1}+I_{sp}[1-(RG_j)^K](G_j-1)/(1-RG_j). \tag{11b}$$

N^{th} iteration of j yields $I(0)$ [$I(0)=I_N$], starting with

$$I_0=I(-NK\Delta)=I(-\infty)$$
$$=I_{sp}[G_2(\tau_0)-1]/[1-RG_2(\tau_0)].$$

In Fig. 3, pulseshapes obtained for a mismatch of $\cong 3\,\mu$m are plotted. It is seen from Figs. 2 and 3 that the sech^2 solution is a very good approximation. Notice that any bandwidth limitation due to a tuning element or dispersion would lead to broader pulses than these ultimate pulses for the ideal synchronously pumped dye laser considered in this paper.

These theoretical results are in excellent agreement with experimental observations [12–15]. The sharp decrease in the 2nd harmonic intensity over a few µm as $\Delta\to 0$ is quantitatively demonstrated. It is seen that pulses with $\leqq 1$ ps duration are possible only if the mismatch $\leqq 2\,\mu$m, for a typical pulsewidth of 100 ps,

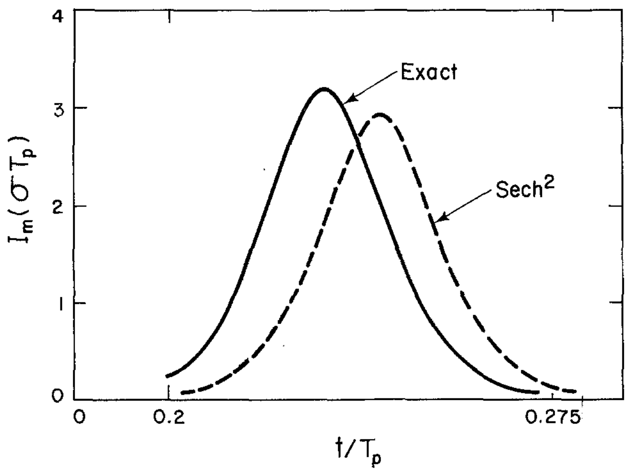

Fig. 3. Pulses for the exact and the sech^2 solutions and their position relative to the pump pulse, for mismatch $L-L_p=3.13\,\mu$m. (Peak of the pump pulse occurs at $t=0$.) Parameters are as in Fig. 2

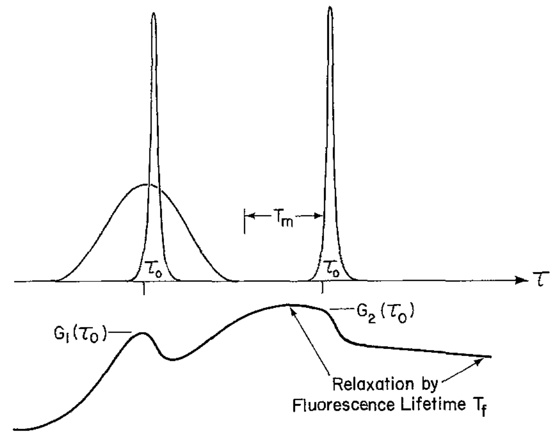

Fig. 4. Gain dynamics for the two transits of the dye pulse through the dye jet in a cavity round trip. T_m is the time elapsed between the end of the pump pulse and the arrival time for the second transit, and is typically ~ 250 ps. $G_1(\tau_0)$ and $G_2(\tau_0)$ are the initial values of gain for the respective transits

and that there is a substantial decrease in energy/pulse as $\Delta \to 0$. Pulse energy decreases since, then, the pulse forms at earlier times τ_0 for which $G(\tau_0)$ is smaller, see (7). Since more energy remains in the trailing edge of the pump pulse as τ_0 shifts to earlier times, susceptibility to satellite pulse formation also increases as $\Delta \to 0$. Total pump energy ($E_p(\infty)$) and/or R must be decreased in order to supress the satellite in this shorter pulse region ($\Delta \to 0$).

A consistent observation in synchronously pumped dye lasers is the exponentially shaped intensity autocorrelation functions. Under practical experimental conditions, cavity length, dye jet thickness and pump pulse fluctuations result in perturbations in cavity mismatch. Since the pulse properties (pulsewidth and energy) are sensitively dependent on Δ as seen above, in particular for $\Delta \to 0$, random fluctuations in Δ must result in significant variations in pulse characteristics. With the assumption that these fluctuations are on a time scale longer than the pulse evolution time, the present analysis can be applied to such a non-ideal laser by setting Δ a random variable in the solution. This can be shown [16] to result in exponentially autocorrelation traces and must be taken into account in experimental pulsewidth determinations.

It should be noted that such perturbations in Δ and cavity bandwidth limitation (not considered here) may result in a quasistable solution for $\Delta < 0$ near $\Delta = 0$, but in accordance with the present results, experimental observations indicate stable mode-locking in the $\Delta \geqq 0$ region.

Acknowledgements. The author is grateful to N. Amer and R. Gerlach for helpful discussions. This work was supported by the Office of Energy Research, Characterization and Measurement Division of the U.S. Department of Energy Contract No. DE-AC03-76SF00098.

Appendix A

A round trip inside the astigmatically compensated three mirror folded dye laser cavity [10] is composed of two transits through the dye jet. As described in Fig. 4, in the first transit the dye pulse coincides with the pump pulse which typically has a duration $\leqq 100$ ps. The dye pulse then travels to the spherical end mirror and makes a reverse transit through the dye jet, typically ~ 330 ps later. In general, by this second pass the pump pulse has left the dye medium.

It can be shown that [7] after the first transit through the dye jet, the dye pulse which starts as $I(\tau)$ (before first transit) becomes

$$I_1(\tau) = G_1(\tau) I(\tau), \tag{A.1}$$

where, from Sect. 1.1

$$\frac{dG_1}{d\tau} + \sigma G_1 [I(\tau)(G_1 - 1) - I_p(\tau)] = 0. \tag{A.2}$$

$I_1(\tau)$ is the pulse incident on the jet for the second transit, at the end of which, the pulse becomes

$$I_2(\tau) = G_2(\tau) I_1(\tau) = G_1(\tau) G_2(\tau) I(\tau), \tag{A.3}$$

where

$$\frac{dG_2}{d\tau} + \sigma G_2 I_1(\tau)(G_2 - 1) = 0. \tag{A.4}$$

Letting $G(\tau) = G_1(\tau) G_2(\tau)$,

$$\frac{1}{G}\frac{dG}{d\tau} = \frac{1}{G_1}\frac{dG_1}{d\tau} + \frac{1}{G_2}\frac{dG_2}{d\tau},$$

from which, using (A.2 and 4),

$$\frac{dG}{d\tau} + \sigma G [I(\tau)(G - 1) - I_p(\tau)] = 0. \tag{A.5}$$

After the second transit, the pulse $I_2(\tau)$ travels to the output mirror and undergoing total linear losses of $T = 1 - R$, it is reflected back at the dye jet as $I_f(\tau) = R I_2(\tau)$ for the first transit of the next roundtrip. Hence, starting with $I(\tau)$, the final pulse at the end of a full round trip is

$$I_f(\tau) = R G(\tau) I(\tau), \tag{A.6}$$

where $G(\tau)$ satisfies (A.5) subject to the initial condition $G(\tau_0) = G_1(\tau_0)G_2(\tau_0)$.

Appendix B

At the end of the first pass of the dye pulse through the dye medium, the trailing edge of the pump pulse keeps increasing the gain. Assuming that the gain value at the end of the first pass of the dye pulse is $G_{1,\mathrm{sat}}$ the dye gain is pumped to a value [solution of (A.2) with $I=0$ and initial value of $G_{1,\mathrm{sat}}$]

$$G_1(\infty)=G_{1,\mathrm{sat}}\exp\left[\sigma\int_{\tau_0}^{\infty} I_p(t)dt\right]. \tag{B.1}$$

Neglecting the pump pulse in the dye pulse vicinity, $G_{1,\mathrm{sat}}$ is the final value of the solution of (A.2) ($I_p=0$) subject to the initial value $G_1(\tau_0)$, and is given by

$$G_{1,\mathrm{sat}}=G_1(\tau_0)X/[1+G_1(\tau_0)(X-1)], \tag{B.2}$$

where $X=\exp\left[\sigma\int_{-\infty}^{\infty} I(t)dt\right]$.

After both pulses exit the dye medium, it can be shown from the rate equations that the logarithmic gain relaxes exponentially by the fluorescence lifetime (T_f) to the steady-state value given by the ground state absorption. Then, defining $G=\exp(g)$, when the dye pulse reappears at the dye jet for the second pass (after an elapsed time of T_m), the logarithmic gain has reached a value

$$g_2(\tau_0)=g_a+(g_1(\infty)-g_a)\exp(-T_m/T_f). \tag{B.3}$$

Defining $\gamma=\exp(-T_m/T_f)$, the initial value of gain for the second transit is therefore given by

$$G_2(\tau_0)=G_a[G_1(\infty)/G_a]^\gamma, \tag{B.4}$$

where $G_a=\exp(g_a)$ is the ground state absorption at the dye laser wavelength, as defined in the main text.

Substituting (B.1 and 3) into (B.4).

$$G_2(\tau_0)=G_a\left\{\left[G_1(\tau_0)X\exp\int_{\tau_0}^{\infty}\sigma I_p(t)dt\right]\Big/G_a[1+G_1(\tau_0)(X-1)]\right\}^\gamma.$$

From (3)

$$G_1(\tau_0)=G_a\exp\left[\sigma\int_{-\infty}^{\tau_0} I_p(t)dt\right],$$

which, substituted into the previous equation, leads to

$$G_2(\tau_0)=G_a\{XX_p/[1+G_1(\tau_0)(X-1)]\}^\gamma, \tag{B.5}$$

where $X_p=\exp\left[\sigma\int_{-\infty}^{\infty} I_p(t)dt\right]$.

Appendix C

It was shown in Appendix A that, neglecting spontaneous emission, the dye laser pulse satisfies (6) over a round trip in the cavity. It can be shown, by iterating (6) over many round trips, that without spontaneous emission a stable pulse cannot be maintained in the cavity for any Δ, since its leading edge will always experience loss for $\tau<0$ (Fig. 1)[2]. Hence, spontaneous emission must be taken into account in the rate equations in order to determine the evolution of the pulse. It is shown below that, for $\Delta>0$, a stable steady-state develops in an ideal synchronously pumped dye laser, in which the saturable gain mechanism effectively shifts the pulse forward, compensating the retardation due to the cavity mismatch.

Consider the region $\tau<\tau_0$ where the effect of the dye pulse on the excited state population is negligible. Neglecting ground state absorption, the interaction in the dye medium is described by

$$\left(\frac{\partial}{\partial x}+\frac{1}{c}\frac{\partial}{\partial t}\right)I=\sigma n_1(I+I_{sp})$$

$$\left(\frac{\partial}{\partial x}+\frac{1}{c}\frac{\partial}{\partial t}\right)I_p=-\sigma_p n_0 I_p$$

$$\frac{dn_0}{dt}=-n_0\sigma_p I_p$$

$$n_0+n_1=n_T,$$

where I_{sp} is the intensity of spontaneous emission into the solid angle subtended by the beam. It follows from the above equations that at the exit face of the dye medium [7], the dye laser intensity $I(\tau)$ satisfies

$$I(\tau)+I_{sp}=[I_0(\tau)+I_{sp}]G_0(\tau), \tag{C.1}$$

where $I_0(\tau)$ is the intensity at the entrance plane of the dye medium and

$$G_0(\tau)\simeq\exp\left[\sigma\int_{-\infty}^{\tau} I_p(t)dt\right]. \tag{C.2}$$

Considering two transits [iterate (C.1) twice using the final pulse of the first pass as the input pulse of the second pass, during which there is no pumping], as in Appendix B, the final pulse at the end of a full cavity round trip becomes

$$I_1(\tau)=RG(\tau)I_0(\tau-\Delta)+I_{sp}[G(\tau)-1], \tag{C.3}$$

where $G(\tau)=G_2(\tau_0)G_0(\tau)$ and $G_2(\tau_0)$ denotes the constant gain in the second transit through the dye medium.

Suppose that an arbitrary pulse $I_0(\tau)$ exists in the cavity. In the next n round trips [iterating (C.5) n times]

$$I_1(\tau)=RG(\tau)I_0(\tau-\Delta)+I_{sp}[G(\tau)-1]$$

$$I_2(\tau)=R^2G(\tau)G(\tau-\Delta)I_0(\tau-2\Delta)+I_{sp}\cdot[RG(\tau)G(\tau-\Delta)+G(\tau)(1-R)-1]$$

$$\vdots$$

$$I_n(\tau)=R^nG(\tau)G(\tau-\Delta)\dots G(\tau-(n-1)\Delta)I_0(\tau-n\Delta)+I_n^{sp}(\tau),$$

where

$$\begin{aligned}I_{n+1}^{sp}(\tau)=I_{sp}[&R^nG(\tau)G(\tau-\Delta)\dots G(\tau-n\Delta)+R^{n-1}\\&\cdot(1-R)G(\tau)G(\tau-\Delta)\dots G(\tau-(n-1)\Delta)\\&+\dots+R^pG(\tau-p\Delta)\dots G(\tau-\Delta)G(\tau)(1-R)+\dots\\&+RG(\tau-\Delta)G(\tau)(1-R)+G(\tau)(1-R)-1].\end{aligned} \tag{C.4}$$

After a sufficient number of round trips (large n), the first term will vanish and

$$I_n(\tau)\xrightarrow[n\to\infty]{} I_n^{sp}(\tau). \tag{C.5}$$

The above indicates that any injected pulse vanishes, and the dye laser intensity evolves from spontaneous emission. Equation (C.4) can be rewritten

$$I_{n+1}^{sp}(\tau)=I_{sp}[R^nG(\tau)G(\tau-n\Delta)+(1-R)R^{n-1}\cdot G(\tau)\dots G(\tau-(n+1)\Delta)]+S$$

$$I_n^{sp}(\tau)=I_{sp}[R^nG(\tau)G(\tau-\Delta)\dots G(\tau(n-1)\Delta)]+S.$$

[2] We consider only $\Delta>0$ since for $\Delta\leqq0$, there is no stable pulse solution in the present model

Hence,

$$I_{n+1}^{sp}(\tau)=I_{sp}R^nG(\tau)G(\tau-\Delta)\ldots G(\tau-(n+1)\Delta)\,[G(\tau-n\Delta)-1]+I_n^{sp}(\tau).$$

For $\tau \ll n\Delta$, the first term vanishes. Hence, for $\tau \leqq q\Delta$, where $q \ll n$,

$$I_{n+1}^{sp}(\tau)=I_n^{sp}(\tau)\equiv I^{sp}(\tau). \tag{C.6}$$

Equations (C.5 and 6) imply that, for $\tau \leqq \tau_0$ and after a sufficient number of round trips, the evolved laser intensity reaches a steady state with its shape conserved during subsequent cavity round trips.
Substituting (C.5 and 6) into (C.3), we obtain, for $\tau \leqq \tau_0$, a conserved pulse shape satisfying

$$I(\tau)=RG(\tau)I(\tau-\Delta)+I_{sp}[G(\tau)-1]. \tag{C.7}$$

In the above, which is valid for $\tau \leqq \tau_0$, $G(\tau)$ was considered independent of the dye laser intensity. For $\tau \geqq \tau_0$, $I(\tau)$ is significant and its effect on $G(\tau)$ must be taken into account. From (C.4) it can be seen that for $\tau \leqq 0$ [where $RG(0)=1$], $I(\tau)=I^{sp}(\tau)$ monotonically increases with τ. Typically, the series (C.4) converges to a value $I(0) \gg I_{sp}$. Hence, considering the development of the spontaneously evolved dye laser intensity over a cavity round trip for $\tau \geqq 0$, we can neglect the spontaneous emission occurring in that round trip. Then, for $\tau \geqq 0$, the intensity at the end of the n^{th} round trip is given by

$$I_n(\tau)=RG(\tau)I_{n-1}(\tau-\Delta),$$

where, from (C.4 and 6), $I_n(\tau)$ has the property that $I_n(\tau)=I_{n-1}(\tau)$ for $\tau \leqq 0$. Redefine $n=1$. In the next N round trips,

$$I_1(0)=I_0(0)$$
$$I_1(\Delta)=RG_1(\Delta)I_0(0)$$
$$I_2(0)=I_1(0)$$
$$I_2(\Delta)=RG_2(\Delta)I_0(0)=I_1(\Delta)$$
$$I_2(2\Delta)=RG_2(2\Delta)I_1(\Delta)=R^2G_2(2\Delta)G_1(\Delta)I_0(0)$$
$$I_3(0)=I_2(0)$$
$$I_3(\Delta)=RG_3(\Delta)I_0(0)=I_2(\Delta)$$
$$I_3(2\Delta)=RG_3(2\Delta)I_2(\Delta)=I_2(2\Delta)$$
$$I_3(3\Delta)=R^3G_3(3\Delta)G_2(2\Delta)G_1(\Delta)I_0(0).$$
$$\vdots$$

Hence, $I_N(\tau)=I_{N-1}(\tau)$ for $\tau<(N-1)\Delta$ and after a sufficient number of round trips (large N), $I(\tau)$ approaches a steady-state, with a conserved pulseshape for all τ.

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