Analysis and performance of a picosecond dye laser amplifier chain

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A mathematical analysis is presented for dye laser amplifier chains used to amplify cw mode-locked dye laser pulses up to the gigawatt regime. The model permits a discussion of the important aspects of the problem such as gain saturation, pulse shaping, and the optimization of pumping efficiencies. Experimentally, an amplifier chain has been demonstrated which is simple in design and operation without sacrificing high performance. A frequency doubled *Q*-switched Nd:YAG oscillator alone is used to longitudinally pump three identical Brewster cells with the same flowing dye solution in each. The amplifier boosts the output of a synchronously mode-locked dye laser to obtain $\sim 0.5 \text{ mJ} \lesssim 1 \text{ psec}$ pulses over a $\sim 400 \text{ \AA}$ bandwidth. These pulses are suitable for efficient Raman shifting, frequency mixing, and continuum generation to vastly extend the spectral range of the system. Some experimental results are presented to support the mathematical model.

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INTRODUCTION

One of the more exciting developments in picosecond dye lasers in recent years has been the incorporation of Nd:YAG pumped dye laser amplifier chains to boost the energy of cw mode-locked dye lasers.¹⁻⁵ These systems successfully couple the many advantages of cw mode-locked dye lasers with the higher energies available from solid state lasers to create a very versatile experimental apparatus. However, there have been few publications which present any experimental results or analyze picosecond dye amplifier chains quantitatively.⁶⁻⁸ While the present work contains some overlap with the references cited above, our emphasis and experimental apparatus differ in a number of ways and therefore should be a useful supplement for workers in this area. A description of our experimental apparatus has appeared earlier,⁹ and is also presented here for completeness. A simple mathematical model is then developed which describes simultaneous pump and signal pulse propagation and their energy exchange. This model also describes the saturable absorbers used for isolation of amplifier stages. A discussion of expected pulse shaping effects through the amplifier is then possible, and calculations are compared with experimental results.

For a more thorough and accurate description, the problem is split into the pumping and extraction processes. Approximations appropriate to each time domain are then introduced which permit the inclusion of effects which are essential for a quantitative understanding of amplifier performance. For the extraction or signal amplifying process, a model for gain saturation is developed which includes the effects of angular hole burning, and experimental support for the model is given. For the pumping process a formalism is presented which approximates the effects of amplified spontaneous emission in a simple manner. Experimental evidence is presented which indicates that triplet losses also play a role, and they are also included in the model.

Finally, the problem of efficiency is addressed. It is shown how the tradeoff between energy storage and energy

extraction results in an optimal cell geometry at each amplifier stage.

EXPERIMENTAL APPARATUS

Due to the fast fluorescence decay of organic lasing dyes, the short energetic pulses from a frequency doubled, *Q*-switched Nd:YAG laser provide an almost ideal pump source for dye laser amplifiers in the 6000- $\text{\AA}$ region of the spectrum. While most systems reported employ¹⁻⁴ more costly Nd:YAG oscillator-amplifier combinations with multiple doubling crystals, we find that a single oscillator and doubling crystal is sufficient to insure outputs on the order of 0.5 mJ. The Nd:YAG model we employed, chosen for its excellent temporal triggering stability and good spatial beam profile, provided $\sim 75 \text{ mJ}$ at 532 nm in a $\sim 15 \text{ nsec}$ pulse at a 10-Hz repetition rate. While there was a pronounced mode-beating structure on the pulse, it was very reproducible from pulse to pulse. Also, as will become apparent later, only a relatively short portion of the pulse is effectively used anyway.

Our rhodamine 6G dye oscillator was pumped by a mode-locked argon ion laser and used both a Brewster prism and a wedge interference filter for tuning and limiting the bandwidth. The output pulsewidth was monitored with a real time rotating prism autocorrelation device, while the spectrum was monitored with an optical multichannel analyzer. The rf driver for the argon mode-locker was counted down with appropriate adjustable delays to drive both the flash lamp and *Q* switch of the Nd:YAG laser with subnanosecond timing precision. A fine adjustment on the *Q*-switch delay over a time range slightly greater than the dye laser pulse period (13.2 nsec) was essential to adjust the relative timing of the pump and signal pulse for optimal amplification.

The amplifier system is shown in Fig. 1 and follows the basic multistage concept for high power laser pumped dye laser systems.¹⁰ The $\sim 8\text{-mm-diam}$, $1.06\text{-}\mu\text{m}$ output beam is rotated by a half-wave plate (HWP) to 45° polarization and

FIG. 1. Experimental configuration of amplifier system.

then telescoped by T1 to ~ 5.5 mm diam to insure good doubling efficiency ($\approx 35\%$) in the 5-cm-type II KDP crystal which follows and attain the diameter appropriate for the third amplifier stage. Harmonic beamsplitters HB1–3 remove $\sim 99.8\%$ of the fundamental and beamsplitter BS1 sends 80% of the energy down a delay line to the third stage A3, while the remaining 20% is telescoped by T2 to ~ 2.5 mm diam and split approximately in half by BS2 and sent to the first and second stages A1 and A2, again with appropriate delays. All three stages of the system are longitudinally pumped. As we will explain later, we believe this provides higher gains, better beam homogeneity, and easier alignment than transverse pumping. The 532-nm pump is reflected into and out of the beam path using dichroic mirrors D1–6 which pass most of the signal. The three flowing dye cells are identical 1.8-cm bore clear tubes with nickel-plated brass end caps to hold the fused quartz brewster windows. This permits the use of high quality windows and also allows easy and economical replacement of damaged windows. The system design allows the same ($\sim 10^{-5}$ M Rhodamine 640 in methanol) solution of dye to flow in all three cells with one low capacity gear pump. Good filtration for the flow system was necessary to insure the removal of all air bubbles from the optical path. Saturable absorbers SA1–2 are identical flowing ethylene glycol jets with a high concentration of malachite green having a small signal transmission of $\lesssim 10^{-7}$ at 630 nm. This provides good control of the leading edge to prevent pulse broadening¹¹ but still passes $\sim 75\%$ of the pulse energy. Without the saturable absorbers flowing, the amplifier chain emits a low divergence superradiant pulse of several millijoules with no input. Mirrors M1 and M2 are 5 cm radius of curvature while M3 and M4 are 20 and 50 cm, respectively, to expand the beam to the appropriate diameter for the last stage. Reflecting optics and brewster angles at all interfaces are used to eliminate any reflections which would limit the gain in a given stage.

From a practical point of view, the system is designed with simplicity of operation in mind. To simplify the alignment process, no spatial filters have been employed in our design. The high small signal gains ($\approx 10^5$ in a single stage) allow saturable absorber isolation instead, even after the first stage. The transient character of the gain, combined with the threshold effect of the saturable absorber-amplifier combination make it unnecessary to select out a single dye oscilla-

tor pulse for amplification; a timing adjustment of the Nd:YAG oscillator *Q*-switch is sufficient to effectively isolate a single pulse. Since the cw dye oscillator beam and the first stage pump are collinear and have comparable average powers, the first stage alignment is routinely performed visually in seconds. Following SA1, the amplified dye pulse is easily seen even at its low repetition rate, and alignment of the remaining cells proceeds in a similar fashion. This procedure can be repeated quickly any time the pump or signal lasers are adjusted. Only one circulation system and dye solution is used for all three stages. This is possible with the proper choice of pump beam cross-sectional areas and energies in the various stages, and results in fewer parts and solutions to maintain with negligible sacrifice of amplification efficiency. Several solvents as well as dyes were tried. Dye selection is discussed in the next section, while methanol or ethanol were found to be the most suitable solvents. Water-Ammonyx LO solutions work equally well as far as amplification is concerned, but seemed to make the optics more susceptible to damage at the solution interface and also left deposits after extensive use.

The output of the amplifier system has energy up to 0.7 mJ with no pulse broadening unless the saturable absorbers are intentionally misadjusted by a large amount. Figure 2 shows a typical autocorrelation of the output pulse, having a FWHM of 1.36 psec or a pulsewidth of 0.88 psec if we assumed a sech^2 pulse shape. Fluctuation in the amplified pulse energy was measured at $\sim \pm 6\%$ *P-P* which was actually a little less than the frequency doubled pump laser energy fluctuation at the time of the measurement. The spatial profile of the pulse is very good at all points throughout the amplifier chain.

MATHEMATICAL MODEL

To analyze the gross behavior of a given amplifier cell, we employ as a first approximation the four-level dye model shown in Fig. 3. We assume infinitely fast relaxation from S_1^* to the upper lasing level S_1 , and from the lower lasing level S_0^* to the ground state S_0 . If we tentatively neglect the triplet state T_0 and other excited states, this results in the population being in either S_0 or S_1 . We will refer to the populations of these levels as N_1 and N_2 , respectively, since we effectively have a two level system where $N_1 + N_2 = N$, the total dye concentration. Neglecting group velocity dispersion and assuming the transverse relaxation time T_2 is much shorter than times of interest, we write the incoherent traveling wave rate equations:

$$\begin{aligned} \frac{\partial \varphi_1}{\partial t} + v_g \frac{\partial \varphi_1}{\partial z} &= v_g (\sigma_1^{\text{em}} N_2 - \sigma_1^{\text{abs}} N_1) \varphi_1, \\ \frac{\partial \varphi_2}{\partial t} + v_g \frac{\partial \varphi_2}{\partial z} &= v_g (\sigma_2^{\text{em}} N_2 - \sigma_2^{\text{abs}} N_1) \varphi_2, \end{aligned} \quad (1)$$

FIG. 2. Typical autocorrelation of amplifier output with a FWHM of 1.36 psec or a sech^2 pulse width of 0.88 psec.

FIG. 3. Energy level scheme for the four level dye model with triplet absorption.

$$\frac{\partial N_1}{\partial t} = v_g(\sigma_1^{em} \varphi_1 + \sigma_2^{em} \varphi_2) N_2 - v_g(\sigma_1^{abs} \varphi_1 + \sigma_2^{abs} \varphi_2) N_1 + \frac{N_2}{\tau},$$

$$\frac{\partial N_2}{\partial t} = -\frac{\partial N_1}{\partial t}.$$

Here $\varphi_{1,2}$ are the pump and signal photon densities, v_g is the group velocity in the solvent, and $\sigma_{1,2}^{em}$ and $\sigma_{1,2}^{abs}$ are the emission and absorption cross sections at the pump (1) and signal (2) frequencies, and τ is the relaxation time of the dye.

Although nonlinear, these equations can be combined and integrated in space over the cell length L to yield an ordinary differential equation. The basic steps are given in Appendix A with the result being

$$\frac{dG}{dt} = v_g(\sigma_2^{em} + \sigma_2^{abs})G [\varphi_{10}(t)(1 - e^{-\kappa NL}G) - \varphi_{20}(t)(G - 1)] - \frac{1}{\tau}G [\ln G + \sigma_2^{abs}NL], \quad (2)$$

where G is the appropriately retarded gain

$$G(t) = \frac{\varphi_2(t' = t + \frac{L}{v_g}, z = L)}{\varphi_{20}(t)},$$

and $\varphi_{10}(t)$ and $\varphi_{20}(t)$ are the pump and signal photon densities at the entrance plane. Here

$$\kappa = \frac{(\sigma_2^{em} \sigma_1^{abs} - \sigma_2^{abs} \sigma_1^{em})}{(\sigma_2^{em} + \sigma_2^{abs})}$$

and

$$\gamma = \frac{(\sigma_1^{em} + \sigma_1^{abs})}{(\sigma_2^{em} + \sigma_2^{abs})}.$$

Equation (2) is the most elementary equation governing the energy exchange between the pump and signal pulses, and describes most of the essential features of pulse amplification in longitudinal pumping. Before proceeding any further, however, accurate values of the emission and absorption cross sections of the amplifier dye are needed at

both the pump and signal frequencies.

It is well known that in high power pulsed operation, a given lasing dye will almost always have its peak lasing wavelength blue-shifted from its cw lasing value. This results from the much larger ground state population in the cw case and the overlap of the emission and absorption cross sections; at the peak emission cross section, the absorption cross section still has a significant value except in dyes with an extraordinary Stokes shift. Since our dye oscillator uses Rhodamine 6G (also called R590, depending on the distributor) its lasing range is centered at about 6000 Å. Rhodamine 101 (R640) has its peak emission cross section in just this range with a methanol solvent, and is thus an appropriate choice for the high power pulsed amplifier cells. It is known to have an excellent quantum efficiency and has been used in our system for many months without replacement, which attests to its good chemical stability. Other workable dyes are Kiton Red (Kiton Red 620) or Rhodamine B (R610) which are slightly yellower, and for operation in the green or red extremes of the oscillator lasing range, Rhodamine 6G or DCM, respectively, have been successfully employed. It is also necessary for the dye to have a reasonable absorption cross section at the pump frequency. Figure 4 shows the absorption cross section and the emission lineshape of Rhodamine 101, taken from the measurements of Batchelder.¹² The absorption cross section can be measured in a straightforward manner, but the emission cross section must be deduced from a knowledge of the fluorescence lineshape $F(\lambda)$, the fluorescence decay time τ , and the quantum yield Φ of the dye using the relation¹³

$$\sigma^{em}(\lambda) = \frac{\lambda^4 F(\lambda) \Phi}{8\pi c n^2 \tau}, \quad (3)$$

where n is the index of the solvent and $F(\lambda)$ is normalized such that $\int_0^\infty F(\lambda) d\lambda = 1$. Since accurate knowledge of the fluorescence decay time of Rhodamine 101 is not available, we instead rely on an alternative estimate of the emission cross section¹⁴ which says that since transition matrix element for emission and absorption should not vary much, the peak emission and absorption cross section should scale proportionally to their frequencies

FIG. 4. Absorption cross section and emission lineshape for Rhodamine 101 (R640). The fluorescence lineshape $F(\lambda)$ is evaluated by the methods discussed in the text when the value read from the graph above is multiplied by the quantity $7.80 \times 10^{-4} \text{ cm}/\lambda^2$, where λ is in cm.

$$\frac{\sigma_{\max}^{\text{em}}}{\nu_{\max}^{\text{em}}} = \frac{\sigma_{\max}^{\text{abs}}}{\nu_{\max}^{\text{abs}}}.$$

Since the quantum efficiency of Rhodamine 101 is known to be very high,¹⁵ we assume that a realistic value is 90%, and this in turn allows us to estimate the fluorescence decay time from Eq. (3). The result is 4.3 nsec, in good agreement with rough measurements.¹⁵ These are the values which we will use in the remainder of this paper; cross sections can be inferred from Fig. 4 at all wavelengths. In passing, we note that the estimated quantum efficiency was also chosen to give a reasonable value for the triplet crossing time.

$$\tau_{\text{trip}} \approx \tau \frac{\Phi}{(1 - \Phi)},$$

of 40 nsec which is consistent with observations to be presented in the next section.

Returning to the analysis, for convenience of discussion, we split Eq. (2) into the pumping and extraction processes, which is equivalent to the statement that negligible pumping occurs during the signal pulse duration. Thus for the amplification process itself, we let $\tau \rightarrow \infty$, $\varphi_{10}(t) \rightarrow 0$ and obtain the well-known solution^{16,17}

$$G(t) = \left[1 + e^{-F(t)} \left(\frac{1}{G_0} - 1 \right) \right]^{-1}, \quad (4)$$

where

$$F(t) = \nu_g (\sigma_2^{\text{em}} + \sigma_2^{\text{abs}}) \int_{-\infty}^t \varphi_{20}(t) dt,$$

which yields the well-known result for the net gain

$$G_{\text{actual}} = \frac{1}{S} \ln [1 + G_0(e^S - 1)]. \quad (5)$$

Here,

$$S = F(t = \infty) = \frac{E_{\text{sig}} (\sigma_2^{\text{em}} + \sigma_2^{\text{abs}})}{A h \nu_{\text{sig}}},$$

where E_{sig} is the signal energy and A is the signal beam area. G_0 as used above is now an "initial condition" gain given by integrating for $G_0(t)$ using

$$\begin{aligned} \frac{dG_0}{dt} &= \nu_g (\sigma_2^{\text{em}} + \sigma_2^{\text{abs}}) G_0 \varphi_{10}(t) (1 - e^{-\kappa N L} G_0) \\ &\quad - \frac{1}{\tau} G_0 (\ln G_0 + \sigma_2^{\text{abs}} N L), \end{aligned} \quad (6)$$

and evaluating $G_0(t)$ at the arrival time of the signal pulse.

Alternatively, we can use Eq. (2) to describe propagation through saturable absorbers. In this case, φ_1 would be zero and the initial gain G_0 would be a number less than one representing the leading edge attenuation factor of the saturable absorber. Since the saturable absorber lifetime τ is very short, it cannot be ignored and we must solve Eq. (2) numerically. We take the value of τ for malachite green in ethylene glycol to be 5.9 psec.¹⁸ If G_0 is known for each amplifier cell and saturable absorber in the system at the arrival time of the signal pulse, the energy amplification and signal distortion in time can be determined by successively solving Eq. (2) at each section and using the output, appropriately attenuated by linear losses of the intermediate optics, as the input to the next stage.

The results of such a calculation are shown in Figs. 5, 6, 7, and 8, where the input signal pulse is taken to have a 2-psec FWHM sech^2 intensity profile. The data used for G_0 are typical measured values for the small signal gains or attenuations of each amplifier section. In Fig. 5, the curves show the input pulse shape, and the shape after the first, second, and third stages. Since there are only two saturable absorbers, the first stage is taken as having no saturable absorber, while the second and third stage each include a saturable absorber on their input ends. The curves are normalized to have the same amplitude, but the pulse energies are 1.0×10^{-9} , 1.0×10^{-5} , 4.2×10^{-5} , and 7.1×10^{-4} J, respectively. These are the same orders of magnitude as the observed energies, but exact agreement with the experimental gain distribution among the three cells cannot be expected for reasons to be discussed in the next section. In addition, the actual distribution is extremely wavelength sensitive, although the net combined gain is not. Figure 6 shows the computed autocorrelation curves of the input, and the second and third stage outputs. The first stage output autocorrelation is not shown since its shape varies only slightly from the input autocorrelation. These figures correspond to normal adjustment of the amplifier apparatus and the numerical values used in the calculation reflect this. Several features are of interest; the most interesting perhaps is the pulse compression at the amplifier output. This results from the attenuation of the leading edge by the saturable absorbers and the depleted amplification of the trailing edge; this is the same process acting in passively mode-locked dye lasers and it is well known that pulse shortening can occur.¹⁹ The extreme saturation of the second stage in this calculation has led to a tail before entering the third stage. Either a reduction in the first stage gain or a larger optical density of the saturable absorber would eliminate this effect without appreciably changing the energy. However, note that the tail has disappeared after the third stage and the compression has occurred despite the tail. It is also worth noting that the pulse effectively creeps forward in time as it propagates through the system, with the peak traveling at a superluminal velocity.²⁰

FIG. 5. Calculated pulse shape upon propagation through amplifier chain. Curves 1-4 represent the input and after each of the three successive stages. Energies are given in text.

Figures 7 and 8 show the same result with the first saturable absorber intentionally misadjusted by moving the saturable absorber jet ≈ 0.5 cm away from the focal point of the first set of focussing optics. This reduces the intensity so that it does not exceed the saturation intensity of the saturable absorber dye by as large an amount and effectively softens the leading edge of the pulse going into the second stage. This hinders the compression process and after the third stage the pulse displays an undesirable lobe on the tail with no compression at all.

Experimentally, we found some of the expectations of the simple analysis above to be exaggerated. In particular, we found little pulse compression, although routine operation resulted in an output pulse which was almost always the same or only slightly shorter than the input pulse. However, when the first saturable absorber jet was intentionally misadjusted as described above, very noticeable pulse *broadening* resulted. This is shown in Fig. 9. The upper trace is the input to the amplifier, with the dye oscillator slightly misadjusted to get pulses which were a little broader than their minimum value, just to see if any compression would be visible. The second trace is the amplifier output with normal adjustment and is almost a perfect replica of the input pulse, an unexpected result in light of the extreme nonlinearity in the amplification process. The third trace shows the amplifier output with the first saturable absorber jet moved out of focus as explained above, and the broadening is easily seen. The lack of compression seems to indicate that there is a significant pulse broadening process which competes with the compression mechanism described above. The most obvious candidate is dispersion, although our estimates, even in conjunction with a reasonable amount of self-phase modulation,²¹ indicate that this is not sufficiently strong in this time regime to prevent a *small* amount of pulse compression from taking place. Another possibility is the finite vibrational relaxation rate of the ground state. Under strong saturated amplification, this can "bottle-neck" a significant fraction of the available stored amplifier energy, eventually releasing it in the tail of the amplified pulse with a timescale characteristic of the vibrational relaxation.²²

FIG. 6. Calculated autocorrelations at different points through amplifier chain. Curves 1-3 are the input and after the second and third stages, respectively.

FIG. 7. Calculated pulse shapes of misadjusted amplifier chain (see text); curves have same meaning as in Fig. 5.

GAIN SATURATION

Equation (5) is the usual result quoted for short pulse saturation behavior, but it assumes an ensemble of dye molecules all with the same cross sections $\sigma_{em,abs}$. If we assume the dye molecule has a well defined axis for its dipole transition moment²³ and a linearly polarized incident beam, we would actually have for a given molecule

$$\sigma_{em,abs} \rightarrow \sigma_{em,abs}(x) = 3\bar{\sigma}_{em,abs} x^2,$$

where $x = \cos \vartheta$ and ϑ is the angle between the dipole axis of the molecule and the signal polarization. Here $\bar{\sigma}_{em,abs}$ are the usual quoted angularly averaged values of the cross sections. We must then also take into account the angular distribution of excited dye molecules, $N_{1,2} \rightarrow N_{1,2}(x)$, while the overall dye concentration N maintains an isotropic angular distribution. If we consider only the short time scale behavior, we ignore pumping and look only at the signal propagation, replacing Eq. (1) with

$$\frac{\partial \varphi}{\partial t} + v_g \frac{\partial \varphi}{\partial z} = \frac{3v_g}{2} \int_{-1}^1 [\bar{\sigma}_{em} N_2(x) - \bar{\sigma}_{abs} N_1(x)] x^2 dx \varphi, \quad (7a)$$

$$\frac{\partial N_1(x)}{\partial t} = 3v_g [\bar{\sigma}_{em} N_2(x) - \bar{\sigma}_{abs} N_1(x)] x^2 \varphi, \quad (7b)$$

FIG. 8. Calculated autocorrelations of misadjusted amplifier chain (see text); curves have same meaning as in Fig. 6.

$$\frac{\partial N_2(x)}{\partial t} = -\frac{\partial N_1(x)}{\partial t} \quad (7c)$$

Making the usual change of variables to local "time," $\xi \equiv v_g t - z$, we have

$$\frac{\partial \varphi}{\partial z} = \frac{3}{2} \int_{-1}^1 \Delta x^2 dx \varphi, \quad (8a)$$

$$\frac{\partial \Delta}{\partial \xi} = -3[\bar{\sigma}_{em} + \bar{\sigma}_{abs}] x^2 \Delta \varphi, \quad (8b)$$

where $\Delta = [\bar{\sigma}_{em} N_2(x) - \bar{\sigma}_{abs} N_1(x)]$.

Here we note two circumstances where we can make a very simple approximation. If Δ were initially isotropic in x and the signal were to deplete the inversion only slightly, Δ would remain roughly isotropic in x . Alternatively, if the absorption or stimulated emission occurs on a time scale slow compared to orientational relaxation times (as might occur in pumping processes) then Δ would also remain isotropic. In either case we approximate Δ with a quantity $\tilde{\Delta}$ which is independent of x to obtain

$$\frac{\partial \varphi}{\partial z} = \tilde{\Delta} \varphi, \quad (9a)$$

$$\frac{\partial \tilde{\Delta}}{\partial \xi} = -\frac{9}{5} [\bar{\sigma}_{em} + \bar{\sigma}_{abs}] \tilde{\Delta} \varphi. \quad (9b)$$

These are identical to the ordinary rate equations, except we would change the results given by Eqs. (2) and (3) by simply letting

$$[\sigma_{em} + \sigma_{abs}] \rightarrow \frac{9}{5} [\bar{\sigma}_{em} + \bar{\sigma}_{abs}].$$

$$\frac{S}{\ln G_0} \int_1^{G_{actual}} \frac{d\xi}{\left[1 - (1-\alpha) \frac{\beta}{\alpha} - (1-\beta) \left(\frac{\pi}{12S\xi} \right)^{1/2} \text{erf}(\sqrt{3S\xi}) + \frac{\beta}{\alpha} \left(\frac{\pi}{12S\xi\alpha} \right)^{1/2} \text{erf}(\sqrt{3S\xi\alpha}) \right]} = 1. \quad (11)$$

Here $\beta = -(\ln R_0 / \ln G_0)$ where R_0 is the initial or leading edge loss factor due to triplet absorption, and $\alpha = \bar{\sigma}_{trip abs} / (\bar{\sigma}_{em} + \bar{\sigma}_{abs})$. Figures 12 and 13 show experimental results for the second and third amplifier stages re-

FIG. 9. Experimental test for compression. Upper trace is input autocorrelation with slight intentional broadening. Middle trace is amplifier chain output under normal adjustment, lower trace is with amplifier chain misadjusted as explained in text.

We refer to this approximation to the effects of angular hole burning as the "modified" theory. This approximation says that the gain is depleted (or excited, if we had been discussing a pumping process) as if the molecules had cross sections 9/5 as large as the ensemble average, and the effect occurs because the molecules which are oriented to make the largest contribution to the gain are the first ones to be depleted (or excited).

Returning to the exact treatment, it is shown in Appendix B that the set Eq. (8) integrates out to yield the result for the net energy gain

$$\frac{S}{\ln G_0} \int_1^{G_{actual}} \frac{d\xi}{\left[1 - \left(\frac{\pi}{12S\xi} \right)^{1/2} \text{erf}(\sqrt{3S\xi}) \right]} = 1, \quad (10)$$

where $S = [E_{sig}(\bar{\sigma}_{em} + \bar{\sigma}_{abs}) / h\nu_{sig} A]$ just as before. Equation (10) is analogous to the simpler Eq. (5) but includes angular hole burning.

Figure 10 compares the results of Eq. (5), Eq. (5) with the 9/5 modification, and Eq. (10) for a hypothetical amplifier cell with an initial gain $G_0 = 10^5$ and a typical cross-section value of $(\bar{\sigma}_{em} + \bar{\sigma}_{abs}) = 4 \times 10^{-16} \text{ cm}^2$. The deviation from the simple theory can be as much as $\sim 2.5 \text{ dB}$, and even the simple 9/5 modified result is a sizable improvement, especially in the mildly saturated regime as expected. Figure 11 shows the analogous result for a saturable absorber with an initial attenuation of 10^{-6} .

For comparison with experimental results, we also included triplet losses and for simplicity assume the initial inversion and triplet populations are approximately uniform in z . As shown in Appendix B, Eq. (10) then is modified to

spectively. The dashed curve for the 595 nm results is Eq. (5), while the solid curve is Eq. (11). In both cases, the theoretical small signal gains were fit to the experimentally measured small signal gains. Both the measured and theoretical curves also include the linear losses of the dichroics before and after each amplifier stage, so the actual gains are higher. The triplet losses for Eq. (11) were set to $R_0 = 0.6$, while the triplet absorption cross section was assumed to be $\sim 7.5 \times 10^{-17} \text{ cm}^2$ which is consistent with other results to be given in the next section. The singlet cross section sums $(\bar{\sigma}_{em} + \bar{\sigma}_{abs})$ were taken from Fig. 4, and had the values 4.45 and $2.44 \times 10^{-16} \text{ cm}^2$ at 595 and 610 nm, respectively. As can be seen, the agreement is quite good with the more complete theory. We have ignored the effects of excited state singlet absorption on the saturation, but we believe these to be small.

As will be discussed in the last section, gain saturation is important for efficient amplification, but it is also desirable for amplitude stability. Since the small signal gain (neglecting saturable absorbers) for our system is $\sim 140 \text{ dB}$, the gain is severely saturated. A small decrease in the input signal, for example, will not affect the output much since in the first two

FIG. 10. Saturation behavior for a hypothetical amplifier stage with $G_0 = 10^5$ and $(\bar{\sigma}_{em} + \bar{\sigma}_{abs}) = 4 \times 10^{-16} \text{ cm}^2$. The dashed curve represents Eq. (5), the dotted curve is Eq. (5) with the 9/5 modification, and the solid curve is the exact result, Eq. (10).

stages the signal still achieves the saturation intensity necessary to extract the majority of the stored inversion energy in the last stage. The saturable absorbers tend to reduce this effect, but we have still observed a significant decrease in amplitude variation upon traversal through the system; 50% reduction in input typically causes only a 10% or less reduction in the output. This behavior also makes the actual gain much more flat across the spectrum than an exponential dependence of small signal gain on cross section would suggest.

PUMPING AND AMPLIFIED SPONTANEOUS EMISSION

To analyze the pumping process, we note that Eq. (6) leaves out angular hole burning, triplet state population buildup, and also the depleting effect of amplified spontaneous emission (ASE). For the pumping process, it is adequate to use the 9/5 "modified" result as mentioned in the last

FIG. 11. Saturation behavior for a hypothetical saturable absorber with $G_0 = 10^{-6}$ and $(\bar{\sigma}_{em} + \bar{\sigma}_{abs}) = 4 \times 10^{-16} \text{ cm}^2$. The dashed curve represents Eq. (5), the dotted curve is Eq. (5) with the 9/5 modification, and the solid curve is the exact result, Eq. (10).

FIG. 12. Second stage saturation behavior at 595 nm. Dashed curve represents Eq. (5); solid curve is Eq. (11). Linear losses are included in measured values, so actual gain is higher.

section. The inclusion of the triplet state population buildup can also be included in a straightforward manner, and this is outlined in Appendix C. The principal effect of the triplet state population is to increase the losses for the signal pulse propagation. That the triplet state attains a significant population is evidenced by the experimental curves shown in Fig. 14. The solid curve is the pump pulse, while the dotted curve is on the on-axis ASE flux. The latter is easily shown to be roughly proportional $G(\ln G)^{-1/2}$. Instead of an increase in the gain on the third lobe of the pumping compared to the gain in the first lobe which would be expected from an integrating effect on the inversion, we actually see a *decrease* in the gain. This results from the increase in the losses at later times resulting from absorption by the triplet state population which has accumulated by intersystem crossing ever since the upper singlet state first became populated. This

FIG. 13. Third stage saturation behavior at 595 and 610 nm. Dashed curve represents Eq. (5); solid curve is Eq. (11). Linear losses are included in measured values, so actual gain is higher.

FIG. 14. Experimental pump pulse profile and the resulting on-axis ASE profile. Note that the third ASE lobe is much smaller than the first, indicating some triplet state absorption buildup.

attests to the necessity of including triplet losses for quantitative agreement. The result from Appendix C is the replacement of the single differential equation Eq. (6) with the pair of equations

$$\begin{aligned} \frac{dG_0}{dt} = & \frac{9}{5} (\sigma_2^{em} + \sigma_2^{abs}) G_0 \varphi_{10}(t) \\ & \times \left[1 - e^{-\kappa NL G_0^\gamma R^{-[\gamma + (\kappa/\sigma_{trip abs})]}} \right] \\ & - \left[\frac{1}{\tau} + \frac{1}{\tau_{trip}} \frac{(\sigma_{trip abs} + \sigma_2^{em})}{(\sigma_2^{em} + \sigma_2^{abs})} \right] G_0 \\ & \times \left(\ln G_0 + \sigma_2^{abs} NL + \frac{(\sigma_2^{abs} - \sigma_{trip abs})}{\sigma_{trip abs}} \ln R \right), \quad (12a) \end{aligned}$$

$$\begin{aligned} \frac{dR}{dt} = & - \frac{R}{\tau_{trip} (\sigma_2^{em} + \sigma_2^{abs})} [\sigma_{trip abs} \ln G_0 \\ & + (\sigma_2^{abs} - \sigma_{trip abs}) (\ln R + \sigma_{trip abs} \sigma_2^{abs} NL)], \quad (12b) \end{aligned}$$

where R is the reduction factor of the gain due to absorption from the triplet state, and τ_{trip} is the triplet crossing time, and $\sigma_{trip abs}$ is the absorption cross section from the triplet state T_0 .

A detailed theory of ASE has been well developed and studied numerically in earlier work.²⁴ To facilitate a simpler but less accurate calculation, we treat the dynamic effects of ASE by adopting a simple approximation based on Fig. 15. If the gain constant is given by γ and is constant throughout an arbitrary convex volume V , it is a straightforward exercise to calculate the total number of photons per unit time leaving the volume V . The gain of a photon emitted at $\mathbf{x}'$ and propagating to $\mathbf{x}$ is $e^{\gamma|\mathbf{x} - \mathbf{x}'|}$, but only a fraction $(\mathbf{x} - \mathbf{x}') \cdot d\mathbf{A} / 4\pi|\mathbf{x} - \mathbf{x}'|^3$ are emitted in this direction. Then the effective average lifetime including induced emissions from ASE is obtained by averaging over all possible points of emission within the volume V . The result is thus

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau} \frac{1}{4\pi V} \int_V d^3x' \int_A \frac{e^{\gamma|\mathbf{x} - \mathbf{x}'|} (\mathbf{x} - \mathbf{x}') \cdot d\mathbf{A}}{|\mathbf{x} - \mathbf{x}'|^3}. \quad (13)$$

This is easily put in a form which separates out the contribution from ASE by employing Green's identity

$$\int_V \psi \nabla^2 \varphi d^3x + \int_V \nabla \psi \cdot \nabla \varphi d^3x = \int_A \psi \nabla \varphi \cdot d\mathbf{A}$$

and letting $\psi = e^{\gamma|\mathbf{x} - \mathbf{x}'|}$ and $\varphi = -(1/|\mathbf{x} - \mathbf{x}'|)$. Then, noting that $\nabla^2 \varphi = 4\pi\delta(\mathbf{x} - \mathbf{x}')$, we obtain after some manipulation

$$\frac{1}{\tau_{eff}} = \frac{1}{\tau} + \frac{\gamma}{\tau} \int_0^\infty e^{\gamma l} S(l) dl, \quad (14)$$

where $S(l)$ is the "length distribution function" of the shape V given by

$$S(l) = \frac{1}{4\pi l^2 V} \int_V d^3x \int_V d^3x' \delta(l - |\mathbf{x} - \mathbf{x}'|). \quad (15)$$

The second term in Eq. (14) accounts for the induced emission from ASE, and the geometrical dependence is now all contained in $S(l)$. As an example, for a sphere of diameter D ,

$$S(l) = 1 - \frac{3}{2} \left[\frac{l}{D} \right] + \frac{1}{2} \left[\frac{l}{D} \right]^3,$$

and for an infinitely long cylinder of diameter D , a tedious calculation yields

$$S_\infty(l) = \begin{cases} 1 + \frac{4}{3\pi} \left[\frac{l}{D} \right] F\left(\frac{l}{D}\right) & \text{if } l < D, \\ 1 + \frac{4}{3\pi} F\left(\frac{D}{l}\right) & \text{if } l > D, \end{cases} \quad (16)$$

where

$$F(x) = \frac{[\mathbf{K}(x) - \mathbf{E}(x)]}{x^2} - [\mathbf{K}(x) + \mathbf{E}(x)],$$

where $\mathbf{K}$ and $\mathbf{E}$ are the usual abbreviations for the elliptic integrals. For the finite cylinder of length L which is of interest to us here, as long as $L/D \gg 1$, it is an excellent approximation to let

$$S(l) = S_\infty(l) \cdot \left(1 - \frac{l}{L}\right).$$

The integral in Eq. (15) can then be performed yielding a preliminary asymptotic result for the effective lifetime

FIG. 15. Arbitrary convex volume used in ASE depletion model.

$$\frac{1}{\tau_{\text{eff}}} \approx \frac{1}{\tau} \left[1 + \frac{2}{3} \gamma L \left(\frac{D}{L} \right) + \frac{1}{8} \frac{e^{\gamma L}}{\gamma L} \left(\frac{D}{L} \right)^2 \left(1 + \frac{4}{\gamma L} + \frac{18}{(\gamma L)^2} + \dots \right) \right], \quad (17)$$

which holds for $D/L \ll 1$ and $\gamma L \gg 1$. In deriving Eq. (13), we have ignored the frequency dependence of γ . If we ignore ground state population, the emission lineshape will be given by $\gamma(\nu)$ when appropriately normalized, and in Eq. (17) we should replace every appearance of $1/\tau(\dots)$ with

$$\frac{1}{\tau} \frac{\int_{-\infty}^{\infty} d\nu \gamma(\nu)(\dots)}{\int_{-\infty}^{\infty} d\nu \gamma(\nu)}.$$

The only term this has a significant effect on is the one containing $e^{\gamma L}$. If we assume that the emission line shape is Gaussian, asymptotically for $\gamma L \gg 1$, this term is reduced by an additional factor of $(\gamma_{\text{max}} L)^{-1/2}$. We then let $\gamma_{\text{max}} L \equiv \ln G$ to obtain

$$\frac{1}{\tau_{\text{eff}}} \approx \frac{1}{\tau} \left[1 + \frac{2}{3} \left(\frac{D}{L} \right) \ln G + \frac{1}{8} \frac{D^2}{L^2} G (\ln G)^{-3/2} \times \left(1 + \frac{4}{\ln G} + \frac{18}{(\ln G)^2} + \dots \right) \right]. \quad (18)$$

The G which should be inserted above is the actual small signal gain (including the gain saturating effect of the ASE) which would be experienced by a signal propagating the length of the amplifier cell at the peak gain wavelength. We note that this expression is similar to the results of others who were interested in upper limits on amplifier gain.²⁵ The most severe approximation involved in the derivation was that the gain was assumed to be uniformly saturated in space by the ASE. For a given ASE depleted G , we expect Eq. (18) to slightly underestimate the effective rate $1/\tau_{\text{eff}}$ only because in the real case the inversion will be localized along the length of the cell in a manner dependent upon the pumping process. This means that, compared to the uniform case, at some points in the cell a larger solid angle of emission will experience a large gain, while at other points a smaller solid

angle sees the large gain. For a given inversion nonuniformity, the tradeoff in solid angle is linear but the gain tradeoff is exponential, so for the same integrated gain inversion nonuniformities should slightly increase the total ASE over the uniform case. As an example, however, in one-dimensional calculations which employ an average solid angle of emission, the result for τ_{eff} is independent of the spatial gain distribution. We stress that the model above does include the ASE depletion by replacing a complicated calculation with an x dependent depleted $\gamma(x)$ with a constant depleted γ such that $\gamma \equiv \frac{1}{L} \int_0^L \gamma(x) dz$.

The ASE depletion can now be included dynamically to a fairly good approximation by using Eq. (6) for the gain at the peak wavelength but letting $\tau \rightarrow \tau_{\text{eff}}$ given by Eq. (18). With this and the previously mentioned 9/5 modification, Eq. (6) gives the gross features of the longitudinal pumping process. To reduce the timing sensitivity, it is desirable to achieve a steady state, which inherently requires energy dissipation after a certain point in time. There are two important features which accomplish this and effectively put a "ceiling" on the gain. One is the strong dependence of τ_{eff} on gain and the second is the fact that once the pump has longitudinally bleached all the way through the dye cell, no further gain can result. Both of these effects make a "steady state" obtain rapidly and make pulse timing less critical. Figure 16 shows the peak gain of the first stage of our system from a numerical integration of the modified equation Eq. (6). The gain almost follows the pump linearly and this is confirmed in Fig. 14 by the on-axis ASE flux.

EFFICIENCY OPTIMIZATION

To roughly analyze the efficiency of a cell, we consider steady state solutions of Eq. (6) since we are only interested in trends. The factor $(1 - e^{-\kappa NL G_0^{\gamma}})$ in Eq. (6) represents the fraction of pump power absorbed and we assume that as we vary all quantities below, we always adjust the dye concentration N so that a good fraction of the pump power is absorbed, i.e., $(1 - e^{-\kappa NL G_0^{\gamma}}) = \text{const} = f$. For simplicity we also ignore the second term in the expression Eq. (18) for τ_{eff} since $D/L \ll 1$. We then obtain

$$G_0 = C_1 \exp \left[\frac{C_2}{A \left[1 + \frac{A G_0}{2\pi L^2} (\ln G_0)^{-3/2} \left(1 + \frac{4}{\ln G_0} + \frac{18}{(\ln G_0)^2} + \dots \right) \right]} \right] \quad (19)$$

and we have from Eq. (3)

$$G_{\text{actual}} = \frac{A}{C_3} \ln [1 + G_0 (e^{C_1/A} - 1)], \quad (20)$$

where C_1 is a constant depending only on f and cross sections, and C_2 and C_3 are constants proportional to pump power and signal energies, respectively.

$$C_1 = (1 - f) \left[\frac{\sigma_2^{\text{abs}}}{\sigma_1^{\text{abs}}} \right],$$

$$C_2 = \frac{P_{\text{pump}} \tau (\sigma_2^{\text{em}} \sigma_1^{\text{abs}} - \sigma_2^{\text{abs}} \sigma_1^{\text{em}}) f}{h\nu_{\text{pump}} \sigma_1^{\text{abs}}},$$

$$C_3 = \frac{E_{\text{sig}} (\sigma_2^{\text{em}} + \sigma_2^{\text{abs}})}{h\nu_{\text{sig}}}.$$

A is the cross-sectional area of the active region of the cell, and we assume here a perfect match between signal and pump beam diameters. P_{pump} is the incident pump power on the amplifier entrance face.

Figure 17 shows the small signal gain in dB and the actual gain on a linear scale obtained from Eqs. (19) and (20) as a function of active diameter for various values of pump energy, all with fixed signal energy of 1.5×10^{-6} J and a 10-cm cell. These are typical values for the second stage of our

FIG. 16. Transient gain calculation at 595 nm for first cell including ASE depletion and triplet losses. Pump is measured doubled Nd:YAG power going into first stage.

system. While the small signal gain always increases as $D \rightarrow 0$, for the actual gain there is always an optimal diameter for each pump and signal level. This results from a competition between energy storage and energy extraction and can be explained qualitatively as follows. The most efficient extraction of a fixed amount of energy occurs when the diameter is as small as possible; the signal reaches the saturation intensity very quickly and extracts almost all of the stored energy. However, to distribute the same total energy $E_{sto} \approx N_2 A L h \nu_{sig}$ in such a thin cell requires that the small signal gain, which is roughly proportional to $e^{N_2 \sigma^{em} L}$ increase as $G_0 \sim \exp(E_{sto} \sigma^{em} / h \nu_{sig} A)$ as A becomes smaller. For any finite length Eq. (6) will not let G_0 increase this rapidly because of ASE. As soon as $G_0 (\ln G_0)^{-3/2} \sim 2\pi L^2 / A$, further reduction in A does not increase G_0 as quickly as $\exp(E_{sto} \sigma^{em} / h \nu_{sig} A)$ anymore and the stored energy available for saturated pulse extraction begins to decline. The best design therefore represents a balance between the best diameter for energy storage and the small diameters needed for efficient energy extraction, and the most efficient cell will be

FIG. 17. Calculated small signal and actual gain at 595 nm as a function of active diameter for input energy of 1.5×10^{-6} J and cell length of 10 cm. Curves 1–5 represent results for pump pulse energies of 2, 4, 6, 8, and 10 mJ, respectively, distributed over a 15-nsec pulse.

one where ASE is just beginning to play a large role. We can also infer from this argument that the existence of an optimal diameter actually results from practical considerations on cell length, but the length dependence is weak ($\sim$ logarithmic) so a substantial improvement would require absurdly long cells. In passing, we note an advantage of longitudinal pumping over transverse pumping. Any mismatch between signal beam and active region cross-sectional areas both wastes energy and reduces gain from ASE considerations, since the aspect ratio D/L increases. The spatial characteristics of longitudinal pumping permit a good match without the adverse effects of reflecting and aberrating wall nearby.

To both *store* and *extract* the largest energies possible, we set the cell diameter at the optimal value as we vary all quantities. In principle, we solve Eq. (19) for $G_0(A)$, insert this in Eq. (20) and maximize G_{actual} with respect to A for each pump energy and then see how this varies with pump energy for various values of input signal energy. This is shown in Fig. 18, all for $L = 10$ cm and signal energies ranging from 10^{-9} to 10^{-4} J. It is immediately obvious why a multiple cell arrangement with ASE isolation is needed if reasonable efficiency is to be expected. The majority of the energy can then be allocated to the last stage where the storage and extraction problems have a more harmonious solution. We can also conclude that if more pump energy were made available, a fourth stage would be desirable providing linear losses are not too high, but almost comparable results should be obtained if the third stage were increased in area slightly and the energy were deposited there. In any event, only a few mJ would be available even for 100 mJ of pump energy in a 15-nsec pulse.

CONCLUSION

In closing, we have shown that a three-stage picosecond dye laser amplifier chain can be constructed using a Nd:YAG oscillator alone with longitudinal pumping of all cells and that energies on the order of 0.5 mJ are easily attained with excellent beam quality. We have shown that the extreme saturation behavior can be well understood and, when

FIG. 18. Optimized extracted energy at 595 nm vs pump pulse energy using approximate steady state analysis. Curves 1–6 are for signal energies of 10^{-9} , 10^{-8} , 10^{-7} , 10^{-6} , 10^{-5} , and 10^{-4} J, respectively, and the pump energy is distributed over a 15-nsec pulse.

properly employed, that it causes efficient energy extraction, good amplitude stability, and negligible pulse broadening. Finally, we have used a simple model for ASE depletion and saturated amplification to show how energy storage and the energy extraction problems can be balanced out to obtain the optimal cell geometry for a given stage.

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APPENDIX A

Making the change of variables $\xi \equiv v_g t - z$ and $\eta \equiv z$, and using $N_1 = N - N_2$ reduces the set Eq. (1) to

$$\frac{\partial \ln \varphi_1}{\partial \eta} = (\sigma_1^{\text{em}} + \sigma_1^{\text{abs}})N_2 - \sigma_1^{\text{abs}}N, \quad (\text{A1})$$

$$\frac{\partial \ln \varphi_2}{\partial \eta} = (\sigma_2^{\text{em}} + \sigma_2^{\text{abs}})N_2 - \sigma_2^{\text{abs}}N, \quad (\text{A2})$$

$$\begin{aligned} \frac{\partial N_2}{\partial \xi} = & - [(\sigma_1^{\text{em}} + \sigma_1^{\text{abs}})\varphi_1 + (\sigma_2^{\text{em}} + \sigma_2^{\text{abs}})\varphi_2]N_2 \\ & + (\sigma_1^{\text{abs}}\varphi_1 + \sigma_2^{\text{abs}}\varphi_2)N - \frac{N_2}{v_g \tau}. \end{aligned} \quad (\text{A3})$$

Combining (A1) and (A2) yields the double equality

$$\begin{aligned} N_2 = & \frac{1}{(\sigma_1^{\text{em}} + \sigma_1^{\text{abs}})} \left(\frac{\partial \ln \varphi_1}{\partial \eta} + \sigma_1^{\text{abs}}N \right) \\ = & \frac{1}{(\sigma_2^{\text{em}} + \sigma_2^{\text{abs}})} \left[\frac{\partial \ln \varphi_2}{\partial \eta} + \sigma_2^{\text{abs}}N \right]. \end{aligned} \quad (\text{A4})$$

Using this result in Eq. (A3) yields

$$\begin{aligned} \frac{\partial^2 \ln \varphi_2}{\partial \eta \partial \xi} = & - \frac{\partial}{\partial \eta} \left[(\sigma_2^{\text{em}} + \sigma_2^{\text{abs}})(\varphi_1 + \varphi_2) - \frac{1}{v_g \tau} \ln \varphi_2 \right] \\ & - \frac{1}{v_g \tau} \sigma_2^{\text{abs}}N. \end{aligned} \quad (\text{A5})$$

Equation (A5) is integrated once in η to yield

$$\begin{aligned} \frac{\partial \ln \varphi_2}{\partial \xi} + (\sigma_2^{\text{em}} + \sigma_2^{\text{abs}})(\varphi_1 + \varphi_2) \\ + \frac{1}{v_g \tau} (\ln \varphi_2 + \sigma_2^{\text{abs}}N\eta) = \Psi(\xi), \end{aligned} \quad (\text{A6})$$

where $\Psi(\xi)$ is an arbitrary function of ξ only. It can be evaluated at $\eta = 0$ where $\varphi_1(\xi, \eta = 0) \equiv \varphi_{10}(\xi)$ and $\varphi_2(\xi, \eta = 0) \equiv \varphi_{20}(\xi)$ are the known input photon densities into the entrance face of the amplifier cell. This yields

$$\begin{aligned} \Psi(\xi) = & \frac{\partial \ln \varphi_{20}(\xi)}{\partial \xi} + (\sigma_2^{\text{em}} + \sigma_2^{\text{abs}})[\varphi_{10}(\xi) + \varphi_{20}(\xi)] \\ & + \frac{1}{v_g \tau} \ln \varphi_{20}(\xi). \end{aligned} \quad (\text{A7})$$

Returning to Eq. (A4), we can integrate this in η also to yield

$$\begin{aligned} \frac{\ln \varphi_1}{(\sigma_1^{\text{em}} + \sigma_1^{\text{abs}})} - \frac{\ln \varphi_2}{(\sigma_2^{\text{em}} + \sigma_2^{\text{abs}})} + \frac{(\sigma_2^{\text{em}}\sigma_1^{\text{abs}} - \sigma_2^{\text{abs}}\sigma_1^{\text{em}})N\eta}{(\sigma_1^{\text{em}} + \sigma_1^{\text{abs}})(\sigma_2^{\text{em}} + \sigma_2^{\text{abs}})} \\ = \Pi(\xi), \end{aligned} \quad (\text{A8})$$

where $\Pi(\xi)$ is an arbitrary function of ξ only. Again this can be evaluated at $\eta = 0$ and then Eq. (A8) holds at all values of ξ and η to yield

$$\varphi_1 = \varphi_{10}(\xi) e^{-\kappa N \eta} \left[\frac{\varphi_2}{\varphi_{20}(\xi)} \right]^\gamma, \quad (\text{A9})$$

where

$$\kappa = \frac{(\sigma_2^{\text{em}}\sigma_1^{\text{abs}} - \sigma_2^{\text{abs}}\sigma_1^{\text{em}})}{(\sigma_2^{\text{em}} + \sigma_2^{\text{abs}})}$$

and

$$\gamma = \frac{(\sigma_1^{\text{em}} + \sigma_1^{\text{abs}})}{(\sigma_2^{\text{em}} + \sigma_2^{\text{abs}})}.$$

If we insert Eq. (A9) into Eq. (A6) using Eq. (A7) for $\Psi(\xi)$ and then evaluate the resulting equation at $\eta = z = L$, we obtain after some simplification the differential equation Eq. (2) in the text.

APPENDIX B

Equation (8b) in the text is integrated in ξ from $-\infty$ to ξ to yield

$$\Delta = \Delta_0(x, z) \exp \left[-3x^2(\bar{\sigma}_{\text{em}} + \bar{\sigma}_{\text{abs}}) \int_{-\infty}^{\xi} \varphi d\xi \right], \quad (\text{B1})$$

where $\Delta_0(x, z)$ is the initial ($\xi \rightarrow -\infty$) value of $\Delta(x, z)$. Placing this into Eq. (8a) and integrating once more from $-\infty$ to ξ yields

$$\frac{\partial F(z, \xi)}{\partial z} = \frac{1}{2} \int_{-\infty}^{\xi} \Delta_0(x, z) [1 - e^{-3F(z, \xi)x^2}] dx, \quad (\text{B2})$$

where $F(z, \xi)$ is the appropriately retarded integrated photon flux past the point z in the cell

$$F(z, \xi) = (\bar{\sigma}_{\text{em}} + \bar{\sigma}_{\text{abs}}) \int_{-\infty}^{\xi} \varphi(z, \xi) d\xi. \quad (\text{B3})$$

We now assume that the initial distribution $\Delta_0(x, z)$ is angularly isotropic, and then Eq. (B2) is readily integrated in x and z to yield

$$\begin{aligned} \int_{F(z=0, \xi)}^{F(z=L, \xi)} \frac{dF}{\left[1 - \left(\frac{\pi}{12F} \right)^{1/2} \text{erf}(\sqrt{3F}) \right]} \\ = \int_0^L \Delta_0(z) dz = \ln G_0, \end{aligned} \quad (\text{B4})$$

and for the net energy gain, we let $\xi \rightarrow \infty$ and perform a simple change of variables in the integral to get the desired result, Eq. (10) in the text:

$$\frac{S}{\ln G_0} \int_1^{G_{\text{actual}}} \frac{d\xi}{\left[1 - \left(\frac{\pi}{12S\xi} \right)^{1/2} \text{erf}(\sqrt{3S\xi}) \right]} = 1, \quad (\text{B5})$$

where $S = [E_{\text{sig}}(\bar{\sigma}_{\text{em}} + \bar{\sigma}_{\text{abs}})/h\nu_{\text{sig}}A]$. This equation also holds for a saturable absorber which can be modeled by the level scheme in Fig. 3 ignoring the triplet state. In this case,

G_0 and G_{actual} will be the leading edge or small signal attenuation and the actual attenuation, respectively.

To approximate the effects of triplet state losses, we assume that in Fig. 3, the upper lasing level S_1^* can decay over to the triplet state T_0 with a decay lifetime τ_{trip} and that once there it can absorb from T_0 with a cross section $\bar{\sigma}_{\text{trip abs}} x^2$, where $x = \cos \vartheta$ as before. We assume also that after an absorption event from T_0 there is no subsequent decay or emission from the upper triplet state manifold on the short signal time scale. Equation (8) in the text then becomes modified to

$$\frac{\partial \varphi}{\partial z} = \frac{3}{2} \int_{-1}^1 \Delta x^2 dx \varphi - \frac{3}{2} \int_{-1}^1 \Delta_T x^2 dx \varphi, \quad (\text{B6})$$

$$\frac{\partial \Delta}{\partial \xi} = -3[\bar{\sigma}_{\text{em}} + \bar{\sigma}_{\text{abs}}] x^2 \Delta \varphi, \quad (\text{B7})$$

$$\frac{\partial \Delta_T}{\partial \xi} = -3\bar{\sigma}_{\text{trip abs}} x^2 \Delta_T \varphi, \quad (\text{B8})$$

where $\Delta_T = N_{\text{trip}} \bar{\sigma}_{\text{trip abs}}$ and Δ is the same as given in the text.

Just as before, Eqs. (B7) and (B8) are integrated in ξ and then inserted into Eq. (B6) which can then also be integrated in ξ to yield

$$\begin{aligned} \frac{\partial F(z, \xi)}{\partial z} &= \frac{1}{2} \int_{-1}^1 \Delta_0(x, z) [1 - e^{-3F(z, \xi) x^2}] dx \\ &\quad - \frac{1}{2} \int_{-1}^1 \Delta_{T0}(x, z) [1 - e^{-3\alpha F(z, \xi) x^2}] dx, \end{aligned} \quad (\text{B9})$$

where $\alpha = \bar{\sigma}_{\text{trip abs}} / (\bar{\sigma}_{\text{em}} + \bar{\sigma}_{\text{abs}})$ and $F(z, \xi)$ has the same meaning as before. Again we use the fact that Δ_0 and Δ_{T0} are isotropic in x and for simplicity we assume that they are also approximately uniform in z . In this case we have $\Delta_{T0}(x, z) = -(\ln R_0/L)$, where R_0 is the initial small signal attenuation factor from the triplet absorption. The initial inversion then becomes $\Delta_0(x, z) = (\ln G_0/L) - (\ln R_0/L)$, where G_0 is the net small signal initial gain. Equation (11) in the text then follows upon integrating Eq. (B9) over the cell length and letting $\xi \rightarrow \infty$.

APPENDIX C

Since we are just considering the pumping process and we assume that at the pump frequency the triplet state absorption is negligible, if we denote the triplet state population by N_T , Eq. (1) in the text becomes modified to

$$\begin{aligned} \frac{\partial \varphi_1}{\partial t} + v_g \frac{\partial \varphi_1}{\partial z} &= v_g (\sigma_1^{\text{em}} N_2 - \sigma_1^{\text{abs}} N_1) \varphi_1, \\ \frac{\partial N_1}{\partial t} &= \frac{9}{5} v_g (\sigma_1^{\text{em}} N_2 - \sigma_1^{\text{abs}} N_1) \varphi_1 + \frac{N_2}{\tau}, \\ \frac{\partial N_2}{\partial t} &= -\frac{\partial N_1}{\partial t} - \frac{N_2}{\tau_{\text{trip}}}, \\ \frac{\partial N_T}{\partial t} &= \frac{N_2}{\tau_{\text{trip}}}, \end{aligned} \quad (\text{C1})$$

where now $N_1 + N_2 + N_T = N$, where N is the total dye concentration. We have already included the 9/5 modification for angular hole burning on a time scale slow compared

to orientational relaxation times.

Just as in Appendix A we make the same change of variables and let $\Delta \equiv [\sigma_1^{\text{em}} N_2 + \sigma_1^{\text{abs}} N_1]$. Then we obtain

$$\frac{\partial \ln \varphi_1}{\partial \eta} = \Delta, \quad (\text{C2})$$

$$\begin{aligned} \frac{\partial \Delta}{\partial \xi} &= -\frac{9}{5} (\sigma_1^{\text{em}} + \sigma_1^{\text{abs}}) \Delta \varphi_1 - \left[\frac{1}{\tau} + \frac{\sigma_1^{\text{em}}}{(\sigma_1^{\text{em}} + \sigma_1^{\text{abs}}) \tau_{\text{trip}}} \right] \\ &\quad \times [\Delta + \sigma_1^{\text{abs}} (N - N_T)], \end{aligned} \quad (\text{C3})$$

$$\frac{\partial N_T}{\partial \xi} = \frac{1}{(\sigma_1^{\text{em}} + \sigma_1^{\text{abs}}) \tau_{\text{trip}}} [\Delta + \sigma_1^{\text{abs}} (N - N_T)]. \quad (\text{C4})$$

We will just outline the procedure from here. Again, we insert Eq. (C2) into Eqs. (C3) and (C4) and integrate from 0 to L in η . Equation (C3) becomes

$$\begin{aligned} \frac{\partial}{\partial \xi} \ln \left[\frac{\varphi_1(\eta = L)}{\varphi_{10}} \right] &= -\frac{9}{5} (\sigma_1^{\text{em}} + \sigma_1^{\text{abs}}) \varphi_{10} \left[\frac{\varphi_1}{\varphi_{10}} - 1 \right] \\ &\quad - \left[\frac{1}{\tau} + \frac{\sigma_1^{\text{em}}}{(\sigma_1^{\text{em}} + \sigma_1^{\text{abs}}) \tau_{\text{trip}}} \right] \\ &\quad \times \left[\ln \left(\frac{\varphi_1}{\varphi_{10}} \right) + \sigma_1^{\text{abs}} NL - \sigma_1^{\text{abs}} \int_0^L N_T d\eta \right], \end{aligned} \quad (\text{C5})$$

while Eq. (C4) becomes

$$\begin{aligned} \frac{\partial}{\partial \xi} \int_0^L N_T d\eta &= \frac{1}{(\sigma_1^{\text{em}} + \sigma_1^{\text{abs}}) \tau_{\text{trip}}} \\ &\quad \times \left[\ln \left(\frac{\varphi_1}{\varphi_{10}} \right) + \sigma_1^{\text{abs}} NL - \sigma_1^{\text{abs}} \int_0^L N_T d\eta \right]. \end{aligned} \quad (\text{C6})$$

Noting that $\varphi_1 = \varphi_{10} e^{\int_0^L \Delta d\eta}$ and that $N_1 + N_2 + N_T = N$ allows us to solve for both $\int_0^L N_1 d\eta$ and $\int_0^L N_2 d\eta$ in terms of the other dependent variables as follows:

$$\begin{aligned} \int_0^L N_1 d\eta &= -\frac{1}{(\sigma_1^{\text{em}} + \sigma_1^{\text{abs}})} \\ &\quad \times \left\{ \ln \left[\frac{\varphi_1(\eta = L)}{\varphi_{10}} \right] + \sigma_1^{\text{em}} NL - \sigma_1^{\text{em}} \int_0^L N_T d\eta \right\}, \quad (\text{C7}) \\ \int_0^L N_2 d\eta &= -\frac{1}{(\sigma_1^{\text{em}} + \sigma_1^{\text{abs}})} \\ &\quad \times \left\{ \ln \left[\frac{\varphi_1(\eta = L)}{\varphi_{10}} \right] + \sigma_1^{\text{abs}} NL - \sigma_1^{\text{abs}} \int_0^L N_T d\eta \right\}. \quad (\text{C8}) \end{aligned}$$

The net signal gain for the *input signal*, including triplet losses, is given by

$$G_0 = e^{\int_0^L (\sigma_2^{\text{em}} N_2 - \sigma_2^{\text{abs}} N_1 - \sigma_{\text{trip abs}} N_T) d\eta}, \quad (\text{C9})$$

where, as before, G_0 is evaluated at the arrival time of the signal pulse and

$$G_0 \equiv \frac{\varphi_2(\xi, \eta = L)}{\varphi_{20}(\xi)}.$$

It follows from Eqs. (C7), (C8), and (C9) that we can obtain an expression for $\varphi_1(\eta = L)/\varphi_{10}$ in terms of $G_0(\xi)$ and $\int_0^L N_T d\eta$ only, which are now considered the dependent variables. This is

$$\frac{\varphi_1(\eta = L)}{\varphi_{10}} = G \gamma e^{-\kappa N L R^{[\gamma + (\kappa/\sigma_{\text{trip abs}})]}}, \quad (\text{C10})$$

where we have let

$$R(\xi) \equiv e^{-\sigma_{\text{trip abs}} \int_0^L N_T d\eta}, \quad (\text{C11})$$

which is the small signal attenuation factor of the signal pulse from triplet losses. Equations (C5) and (C6) can then be rewritten using the results (C10) and (C11), and after some algebra, the results become the pair of equations Eq. (12) in the text.

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