Temporal smoothing of multimode dye-laser pulses

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Pulses from a broadband dye laser operating in many modes have been temporally smoothed by passing them through a strongly saturated dye amplifier in the presence of a strong, nonsaturable absorber. The standard nonlinear coupled partial differential equations based on the rate-equation approximation have been used to model the amplifier. Both analytic and numerical solutions for the equations show that under attainable conditions the intensity of the amplified pulse follows the temporal shape of the pump pulse.

The intensity of light from a multimode dye laser is generally strongly modulated in time owing to the beating between all pairs of modes within the laser bandwidth. The usual exception to this is the case of frequency-modulated (FM) laser operation, which is achieved by placing a periodic phase modulator inside the laser cavity.[1] In the ideal case this type of laser emits multimode light with constant intensity. Nonideal FM mode locking has also been observed to occur spontaneously in a dye laser, leading to a lessening, but not to the entire elimination, of mode beating.[2]

In this Letter we report a method that acts externally to the laser cavity and that produces a nearly ideal FM-locked dye-laser field, starting from a pulsed dye-laser field with arbitrary mode phases and amplitudes. The intensity of light pulses generated in this manner is temporally smooth, while the spectrum of the pulses remains broad. The method is based on passing the pulses through a strongly saturated dye amplifier to which a strong, nonsaturable absorber has been added and which is pumped by smooth laser pulses. This study is related to an earlier one by Curry *et al.*, who observed a stabilization of dye-laser pulse energy under conditions similar to those studied here.[3] However, changes to the pulse shape were not reported.

We have theoretically modeled the amplifier system as a four-level gain medium plus a nonsaturable absorber with absorption coefficient α. We start with the standard nonlinear coupled partial differential equations

$$\frac{\partial I(t,z)}{\partial z} = [\sigma N_2(t,z) - \alpha] I(t,z), \tag{1}$$

$$\frac{\partial N_2(t,z)}{\partial t} = \sigma_p I_p(t,z) N - [\sigma_p I_p(t,z) + \sigma I(t,z) + A_{21}] N_2(t,z), \tag{2}$$

where $I(t, z)$ is the total photon flux of the light being amplified, $N_2(t, z)$ is the population inversion density of the gain medium, $\sigma_p I_p(t, z)$ is the pump rate, σ is the emission cross section, A_{21} is the spontaneous decay rate of the upper level, N is the concentration of the active molecules, and t is the retarded time variable. Equation (2) can be integrated to give

$$N_2(t,z) = \int_0^t \exp\left\{-\int_{t'}^t [\sigma_p I_p(t'',z) + \sigma I(t'',z) + A_{21}] \mathrm{d}t''\right\} \sigma_p I_p(t',z) N \mathrm{d}t', \tag{3}$$

where $N_2(0, z)$ has been assumed to be zero. If the upper-level relaxation rate is much larger than the rate of change of both $I_p(t, z)$ and $I(t, z)$, i.e., if $(\sigma_p I_p + \sigma I + A_{21}) \gg (1/I_p)\partial I_p/\partial t$ and $(1/I)\partial I/\partial t$, then the exponential factor in the integrand of Eq. (3) is strongly peaked around $t' = t$. This allows one to replace $I_p(t', z)$ by $I_p(t, z)$ and, by a similar argument, to evaluate the remaining integral, which gives

$$N_2(t,z) \simeq \frac{N\sigma_p I_p(t,z)}{\sigma_p I_p(t,z) + \sigma I(t,z) + A_{21}}. \tag{4}$$

Expression (4) is substituted into Eq. (1). A procedure similar to that used to obtain expression (4) can then be applied, yielding

$$I(t,z) \simeq \frac{\sigma}{\alpha} \frac{I(t,z) N \sigma_p I_p(t,z)}{\sigma_p I_p(t,z) + \sigma I(t,z) + A_{21}} \tag{5}$$

under the conditions $\alpha z \gg 1$ as well as $\alpha \gg (1/I_p)\partial I_p/\partial z$, $(1/I)\partial I/\partial z$. The initial condition $I(0, t)$ has dropped out since $\alpha z \gg 1$. This is a quadratic equation in $I(t, z)$, the nontrivial solution of which is

$$I(t,z) \simeq (N/\alpha - 1/\sigma)\sigma_p I_p(t,z) - A_{21}/\sigma. \tag{6}$$

Under typical conditions the A_{21} term is negligible, and thus the output intensity $I(t, z)$ is predicted to follow the pump intensity $I_p(t, z)$. This means that pumping the amplifier with smooth pulses will produce smooth output pulses regardless of the input-pulse shape. Note also that the output intensity is independent of the input intensity. Substituting expression (6) into expression (4) leaves us with a simple expression for $N_2(t, z)$:

$$N_2(t,z) \simeq \alpha/\sigma, \tag{7}$$

which is valid for $\alpha < N\sigma$.

We have also solved Eqs. (1) and (2) numerically to check whether the analytic solutions in expressions (6) and (7), which are asymptotic limits, are accurate under experimentally realizable conditions. The follow-

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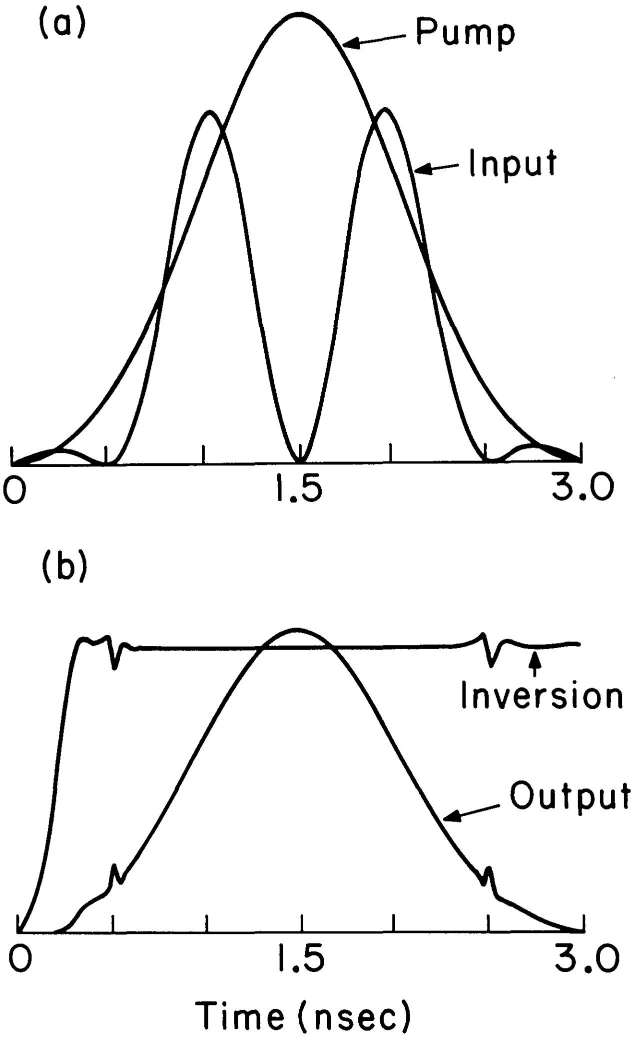

Fig. 1. (a) Temporal profiles of the pump pulse and input pulse used in the numerical calculation. (b) Computed output intensity and gain medium population inversion at the output face.

ing values of the parameters were used in the calculations: $N = 7.5 \times 10^{16}$ cm^{-3}, $\sigma = 2.2 \times 10^{-16}$ cm^2, $\sigma_p = 3.9 \times 10^{-16}$ cm^2, $A_{21} = 2 \times 10^8$ sec^{-1}, $\alpha = 7$ cm^{-1}, $z = 1.5$ cm, and $I_p = 1.2 \times 10^8$ W/cm^2 (peak value). The initial population inversion was assumed to be zero. The assumed temporal profiles of the pump $I_p(t, z)$ and input $I(t, 0)$ pulses are shown in Fig. 1(a). The peak value of $I(t, 0)$ is 5×10^8 W/cm^2. The amplifier was assumed to be transversely pumped. Figure 1(b) shows the results of the calculation. Except for some rather minor transients the results are in excellent agreement with the predictions of expressions (6) and (7), i.e., the output pulse follows the pump pulse and the inversion remains constant. It is also worth pointing out that the numerical values of $I(t, z)$ and $N_2(t, z)$ are in good agreement with expressions (6) and (7). Since the values of the parameters used in the calculations are readily achieved in the experiment, the proposed method can be used for obtaining broadband dye-laser pulses with smooth temporal profile.

We have experimentally verified the predictions of the theoretical model. The frequency-doubled output from a Nd:YAG laser operating in a single longitudinal mode[4] provided temporally smooth 6-nsec pump pulses for the dye-laser–amplifier system. The input pulses for the smoothing amplifier were generated in a grazing-incidence Rh6G dye-laser oscillator, which lased in a few longitudinal modes spaced by 1.3 GHz. The pulses from the laser oscillator (~0.1 mJ) were spectrally and spatially filtered to eliminate amplified spontaneous emission (ASE) and then amplified in a smoothing amplifier, transversely pumped by a 28-mJ frequency-doubled Nd:YAG-laser pulse. As an amplifying-absorbing medium we have used a solution of Rh6G (2×10^{-4} *M*) and Malachite Green[5] in methanol. The output from the smoothing amplifier (0.3 mJ) was spectrally filtered to eliminate ASE and then optically delayed for detection purposes. A portion of the in-

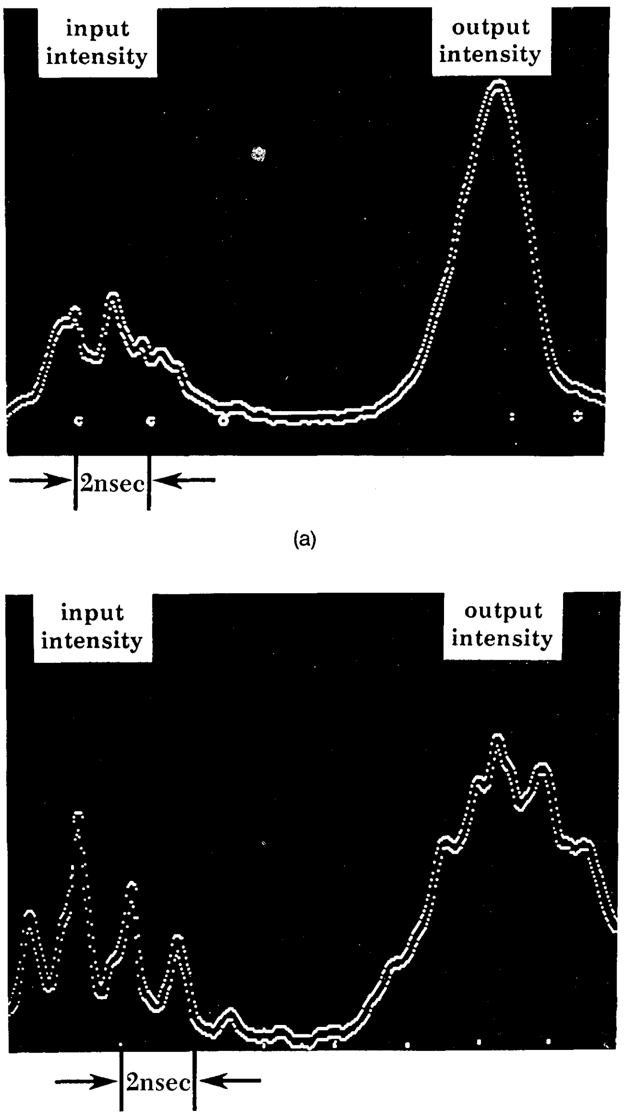

Fig. 2. Input and output pulses recorded with a transient digitizer: (a) concentration of nonsaturable absorber optimized for best smoothing, (b) the same system but without a nonsaturable absorber.

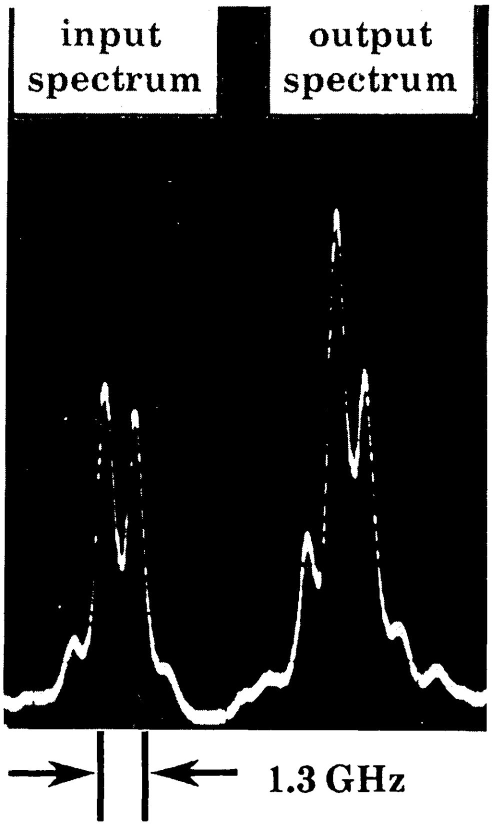

Fig. 3. Example of the single-shot optical spectrum of the input and output pulses.

put pulse was combined with the attenuated, delayed output pulse, and both were detected with a fast vacuum photodiode, whose output was recorded with a 1-GHz-bandwidth transient digitizer.

An example of the measured pulse shapes, illustrating good pulse smoothing, is shown in Fig. 2(a). The bandwidth of a laser pulse was typically larger than 1 GHz. Since the rise time of the detection system was 0.3 nsec, it is not possible to say whether intensity fluctuations faster than this were removed from the pulse in the amplification process, although the theory indicates that they should have been. The best smoothing was obtained when the concentration of Malachite Green was 2.67×10^{-5} M and the net energy gain was comparable with unity. It should be emphasized that in the absence of the nonsaturable absorber good smoothing could not be obtained, as illustrated in Fig. 2(b). Some insight into this situation can be gained by using the steady-state analysis of Curry *et al.*,[3] since the molecules reach steady state faster than the changes in input intensity occur. That analysis shows that the addition of a nonsaturable absorber increases the range of input intensities over which the output intensity is nearly constant.

Additional measurements were performed to detect the changes in the laser-pulse spectrum resulting from the temporal smoothing. Optical spectra of the input and output pulses were recorded with a Fizeau interferometer and a Reticon array.[6] Since the spectrum of the dye laser changed from pulse to pulse, it was necessary to perform single-shot measurements, i.e., both input and output spectra were recorded for the same pulse. Figure 3 shows one example of the results. The spectrum of the smoothed pulse is seen to be different from that of the input pulse. This is not surprising since the lack of mode beating indicates special relations between the amplitudes and phases of the modes in the output pulse.

In conclusion, we have shown that multimode dye-laser pulses can be smoothed temporally by being passed through a saturated amplifier with a nonsaturable absorber added. The amplifier must be pumped by a temporally smooth laser pulse. We have developed a theoretical model for the smoothing process based on the rate-equation approximation. Both numerical calculations and analytic solutions show that under proper conditions the output pulse follows the temporal shape of the pump pulse. Experimental evidence of temporal smoothing has been observed, as were changes of the optical spectrum of the pulse resulting from the amplification process. It should be noted that the efficiency of the smoothing amplifier was much less than unity. Studies are currently being carried out to determine the degree of trade-off between efficiency and smoothing.

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