Pulsed single-mode dye laser for coherent control experiments

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We describe a pulsed single-mode dye laser whose output characteristics satisfy the stringent requirements of coherent control experiments. This simple, low cost laser exhibits 1.6× transform-limited frequency performance with a single-shot linewidth of 58 MHz, and a time averaged linewidth of 240 MHz. The spatial mode is nearly Gaussian and has a beam quality parameter $M^2 = 1.2$. We modified the pulse shape using saturable absorption in the dye amplifier. The resultant pulses are temporally smooth and have a duration of approximately 20 ns. © *1997 American Institute of Physics.* [S0034-6748(97)04506-1]

I. INTRODUCTION

The dye laser has played a distinguished role in the history of optical spectroscopy.[1] Its broad tunability made it the clear choice of atomic spectroscopists, providing much higher spectral brightness than was available with conventional lamp sources. While newer solid state sources have demonstrated large tuning ranges (most notably titanium doped sapphire), these gain media are limited at this time to the near infrared region of the spectrum. Optical parametric oscillators show the broadest frequency tunability and may in the future become the tunable source of choice. Until that time, the dye laser will continue to be the workhorse for experiments requiring tunability in the visible. Consequently, much work has gone in to designing and engineering dye laser sources. Although stabilizer cw dye lasers provide extremely high resolution (<1 MHz), many applications require higher peak powers either for nonlinear frequency conversion or to induce transitions between weakly coupled states. The use of pulsed dye lasers is then required.

In the past, pulsed dye lasers were mainly characterized by their linewidth. This was consistent with the job for which they were used: preferentially exciting a given quantum level in favor of all other quantum levels. Given the local level spacing of the target atom (we will use the term ''atom'' here to generically refer to a quantum system with a discrete spectrum), one can place an upper limit on the necessary linewidth for selective excitation. The other main issue is then the total power required to insure efficient transfer of population to the target level. New research in coherent control chemistry has shown that a properly designed laser pulse can drive internal atomic dynamics to a predetermined target state.[2,3] The cost of such flexibility is usually realized in the laser source whose pulses must be near the theoretical ideal. Any deviations from this ideal can cause reduced efficiency in the production of the target atomic state.

Because the coherent control process makes use of quantum interference, the applied laser fields must be highly coherent.[4] That is to say that linewidth considerations alone do not answer the feasibility issue of using dye lasers to coherently control atomic dynamics. Besides just single-longitudinal-mode (SLM) operation, the laser pulse must exhibit near transform-limited frequency performance. Also, since many coherent control schemes involve more than one laser, the spatial profile must be clean and controllable so that overlapping multiple beams in an interaction region is possible. And finally, the temporal smoothness of the laser pulse can play a critical role in some interactions.[5] Any sharp features in the temporal profile can contribute to reduced conversion of the initial to the target state.

In this article we describe a pulsed dye laser source which fulfills these requirements and has been successfully used in a coherent control experiment. One of the main conclusions from the current work is that the quality of the pump laser and the dye laser are strongly correlated, both spatially and temporally. In what follows, we examine each of the critical laser parameters and discuss what pump and oscillator features are important in achieving high quality performance in the resultant dye pulse.

II. GENERAL LAYOUT AND DISCUSSION

Figure 1 shows a schematic layout of our oscillator and amplifier. The properties of the frequency-doubled Nd:YAG pump are critical to dye laser performance and so we describe its design first. The Nd:YAG used is a Quantel model YG581-10 actively Q-switched system providing approximately 60 mJ of 532 nm energy in a single-transverse, multilongitudinal mode output. A typical temporal profile produced by this system is shown in Fig. 2(a). The amplitude modulation is indicative of the multilongitudinal mode oscillation, and the pulse profile varies significantly from shot to shot. As will be discussed in Sec. III on the temporal profile, this gives rise to an unacceptable temporal profile of the dye laser. To remedy this, we modified the cavity in two ways. Since Nd:YAG is predominantly homogeneously broadened, spatial hole burning can be one of the main obstacles in obtaining single-mode oscillation.[6] This was eliminated in the linear cavity geometry by making use of a ''twisted-ribbon mode''[7] in the cavity. This is achieved by surrounding the gain medium with λ/4 waveplates with the output coupler on one side and a linear polarizer on the other. This generates an axially uniform mode energy density inside the gain medium and hence eliminates spatial hole burning. The second modification was to replace the Pockels cell Q switch with a LiF crystal with F_2^- color centers acting as a slow saturable absorber Q switch.[8] This combined with an intracavity etalon produced single-longitudinal-mode output with a

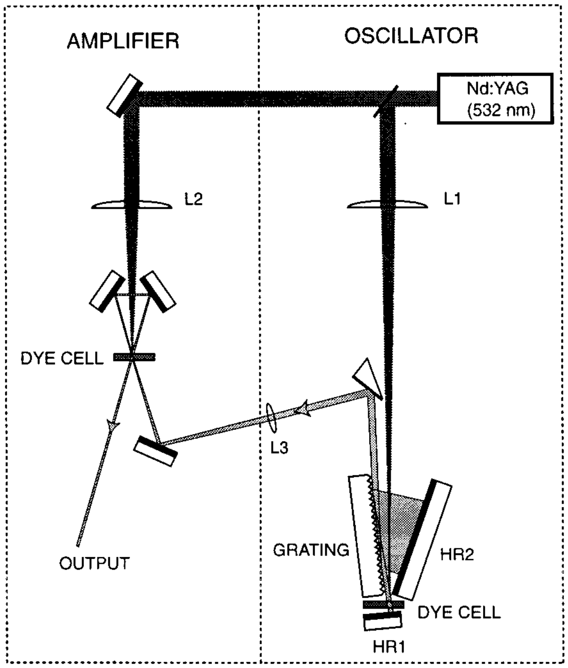

FIG. 1. Schematic layout of the oscillator and amplifier system. L1 is a 250 mm focal length lens and L2 is a 150 mm focal length lens. HR1 and HR2 are high reflectivity mirrors.

smooth temporal profile shown in Fig. 2(b). The resultant 532 nm energy was limited to 12 mJ. The Nd:YAG beam was focused with a $f=250$ mm lens to a spot before the dye cell with small translations of the lens along the optical axis

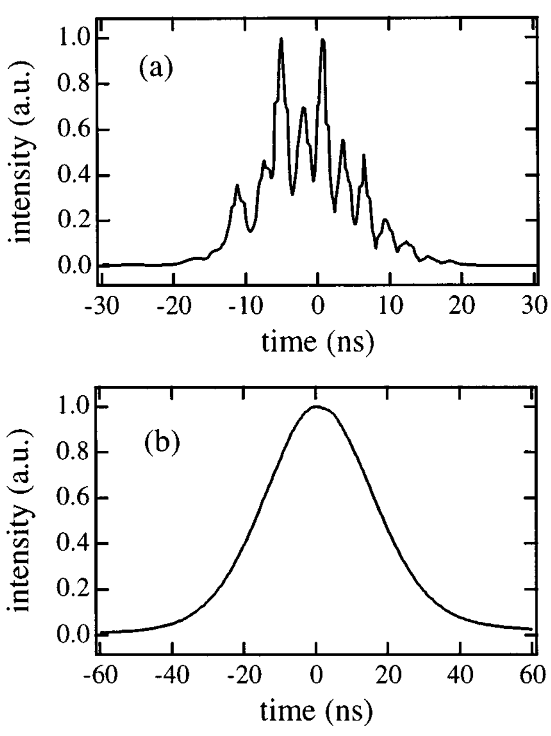

FIG. 2. Temporal profile of the Nd:YAG pump laser. In (a), the laser is actively *Q* switched and is multilongitudinal mode, and in (b) the laser is passively *Q* switched and is single-longitudinal mode.

enabling variation of the size of the gain region. Focusing before the cell is preferable as it ensures that the energy is not focused onto the dye laser mirrors.

The dye oscillator is based on the original design of Shoshan[9] and Littman[10,11] which makes use of a single diffraction grating used in grazing incidence to provide frequency selectivity. This is a convenient design because it involves only a few optical components and is easy to work with and build. The grating is a 1800 lines per mm holographic grating manufactured by Optometrics Corporation. The choice of lines per mm depends on the operating wavelength of the laser. Higher diffraction orders are formed when either the lasing wavelength or the number of lines per mm is decreased. This has two negative consequences. One is that the higher diffracted orders increase the loss of the cavity and hence reduce conversion efficiency. Second, it is possible that in the grazing incidence mode of operation there may be a Littrow diffracted order within the dye bandwidth. This can cause more than one color to lase simultaneously. By choosing the number of lines per mm appropriately, these effects can be avoided.

The rear high reflector HR1 is a high damage threshold tunable laser mirror made by CVI Laser Corporation, model TLM1. This mirror was epoxied to a piezoelectric transducer (PZT) stack, Thorlabs model AE0505D16, which provides a displacement of ≈ 1 μm at a drive voltage of 10 V. This enabled the use of a standard laboratory power supply to make cavity length changes (on the order of the wavelength) which proved to be very useful in single-mode operation. The tuning mirror, HR2, is manufactured by Optometrics and is 5 cm in length matched to the grating length. Since dispersion in the cavity depends on the length of the illuminated part of the grating (for fixed number of lines per mm), the 5 cm length is preferred over more compact gratings. The tuning mirror is epoxied to a mirror mount (Thorlabs KC1-PZ) which contains a similar PZT stack mentioned above in conjunction with the standard 1/4-80 adjustment screw. The PZT allowed for high resolution tuning through narrow spectral features. The quartz dye cell is manufactured by NSG Precision cells, type 48 and has a path length of 1 mm.

III. LINEWIDTH

In order to excite particular atomic resonances with prescribed detunings, the lasers need to be wavelength tunable. Continuous tuning (without mode hopping) is preferred so that contiguous spectral regions can be investigated. Furthermore, to fully achieve the coherent control objectives, near transform-limited single-shot pulses must be used. Frequency jitter and drift must be kept small, and amplified spontaneous emission (ASE), which often plagues pulsed dye laser systems, must be negligible.

There are many commercial dyes available, enabling laser wavelengths to span the visible and well into the infrared and ultraviolet regions of the spectrum, with essentially no gaps. However, when a broadband dye is used in a very low-*Q* cavity (typical of the grazing incidence design) the entire tuning range is not realized. For a given pumping level, if the laser operates SLM on the wings of the tuning curve, it may be multilongitudinal mode at the gain peak. Consequently, the effective bandwidth of these dye lasers can be reduced, causing gaps in the spectrum covered by existing dyes. This can be remedied however by modifying the chemical environment of the dye.[1] The gain maximum can be adjusted by changing the concentration, the solvent, or the *p*H of the solution. A shift of the gain maximum can also be achieved by changing the length of the gain region,[12] although this is not a practical method. The concentration is typically adjusted to provide good pump absorption while still providing a near cylindrically symmetric gain region within the dye. These two competing requirements are usually balanced to yield acceptable results for both with an absorption level ~80% leaving no room for wavelength tuning in terms of concentration. The solvent, however, is modified easily within the constraints of solubility. For instance, a simple switch from methanol to ethanol resulted in a 4 nm blue shift. Using a 50:50 mixture of H_2O and ethanol solution (with a small addition of the surfactant Ammonyx LO to prevent dye aggregate formation) gave an additional ~5 nm blue shift. And as a final technique that works well with the rhodamine dyes, the *p*H of the solution can be varied. This is achieved by addition of a small quantity of base (saturated solution of KOH in ethanol) to blue shift by up to 10 nm, or acid (HCl in water) to red shift by up to 10 nm. The use of both solvent and *p*H tuning are typically not needed for a single dye, but we regularly use both techniques to shift the dye tuning curve and/or to maximize energy output into a given wavelength. For the particular laser that we describe here, a 3×10^{-4} M solution of rhodamine 610 in 2 ℓ of methanol with 1/2 ℓ of 95:5 H_2O:Ammonyx LO gave a single-mode tuning range of 582–599 nm with a peak near the Na *D*-lines transition of ≈589 nm. The same dye solution was used in the amplifier as in the oscillator.

Given the particular dye environment for achieving the specified wavelength, the main issue becomes attainment of SLM operation. Single-mode output has been previously reported for this cavity.[11,13] The most important parameters are cavity length and angle of grazing incidence. By use of longitudinal pumping, short gain regions can lead to short cavities with lengths ~4 to 5 cm. The angle of incidence is typically 89°–89.5°. The size of the gain region also can affect single-mode performance. Different longitudinal modes travel slightly different paths in the dye cavity.[14] The gain region acts as a slit in the ''spectrometer'' formed by the grating and tuning mirror. The larger the slit, the lower the spectral resolution. We have verified that a cavity geometry that operates single mode for one spot size can act multimode for a larger gain region. This would seem to imply that very small gain regions should be used. However, diffraction can give rise to unacceptably large losses. Thus, the size of the gain region can be an important cavity parameter. We report performance with a pump beam having a Gaussian waist size of 220 μm.

Continuous wavelength tuning is sometimes required in coherent control experiments. If the axis of rotation of the tuning mirror is not chosen properly, the act of rotating the mirror will change the cavity length in an uncontrolled fashion which will lead to mode hopping. It has been shown however[15,16] that if the axis of rotation is chosen properly, the rotation can cause a change in cavity length that exactly tracks the frequency change so that the longitudinal modes move in tandem with the grating passband thereby preventing mode hops. In our system, with no care made to achieve the correct pivot point, we had a single-mode tuning range of <0.05 cm^{-1}. However, with a modest attempt to get the rotation axis to coincide with the special location gave a much extended tuning range of 1.7 cm^{-1}. This was sufficient for our application. However, as shown by Zhang *et al.*[16] tuning ranges of 190 cm^{-1} can be achieved if care is taken to more stringently satisfy the pivot position.

To investigate the single-shot linewidth, we utilized a modified plane-parallel Fabry–Perot interferometer (Burleigh RC 110) with a free spectral range of 250 MHz and finesse ~10 and observed circular fringes with a laser beam analyzer (Spiricon model LBA-100). We digitized single shots and measured the linewidth. Furthermore, the temporal profile was measured as well. This enabled us to calculate the theoretical transform-limited linewidth without making any assumption about the pulse shape. This helps to eliminate any systematic error caused by lack of knowledge of temporal profile. For our pulse with a full width at half-maximum (FWHM) ≈20 ns, we calculated a transform-limited linewidth of 36 MHz and measured 58 MHz. This gives 1.6× transform-limited performance for a single shot. This compares favorably with performance from pulse amplified cw sources where 1.4× transform-limited performance has been achieved.[17] However, the simplicity of this design eliminates the need for a complicated stabilized cw source. Of course, an advantage of a stabilized cw source is the long term frequency stability. To analyze this parameter for our system, the same single-shot spectrum was averaged over 100 shots to produce a time averaged linewidth of 240 MHz. The most like source of this frequency jitter is nonlaminar flow in the dye cell.[18]

The main source of frequency drift in our system was temperature drift in the dye. We measured the required temperature stability by using a heat exchanger in the dye flow system coupled to a constant temperature bath recirculator. This system provided a temperature stability of 0.01 °C with a temperature resolution of 0.1 °C. Using a Na/Ne optogalvanic cell, we monitored a Ne optogalvanic resonance signal as the temperature of the bath was varied. The results are shown in Fig. 3. The Ne resonance at 588 nm has a (Doppler broadened) width of 3 GHz.[19] The best fit Doppler profile gives a temperature FWHM=0.3 °C. This implies a temperature induced frequency drift of 10 GHz/°C. Using the temperature dependent refractive index of ethanol and the length of the gain region, we can calculate a predicted frequency drift of 9 GHz/°C, very close to the measured value. This shows that the temperature of the dye must be kept stable to less than a tenth of a degree Celsius if sub-GHz resolution is required. Without the use of the constant temperature bath, we found two modifications to the dye flow system that helped to minimize the temperature drift. The first consideration was to use a large dye reservoir. We found that 2 ℓ was a reasonable compromise between stability and convenience. The second modification was to arrange the dye pump so that it pulled fluid through the dye cell rather than

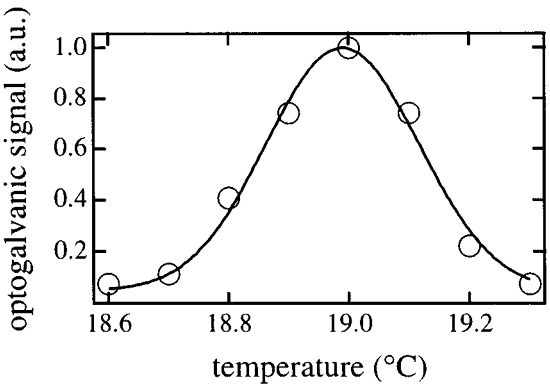

FIG. 3. Optogalvanic signal as a function of temperature of the dye fluid (open circles). The best fit Gaussian (Doppler) profile is shown as the solid line.

forcing it through. This had a considerable reduction in temperature drift because the motor is the largest source of heat in the system.

Slow drifts can be eliminated by active cavity stabilization using the technique of Raymond *et al.*[20] This scheme makes use of the different intracavity paths taken by different longitudinal modes which creates angular deviations in the output (this can also give rise to transverse mode structure which will be discussed later). By using a two-element detector, an error signal can be derived from this angular separation. We have implemented this technique and achieved the short time averaged linewidth for much extended periods.

Finally, using a one meter Jarrell–Ash Czerny–Turner spectrometer with a Princeton Applied Research CCD camera and optical multichannel analyzer we measured the ASE intensity 2 nm from the lasing line. Although the measurement was limited by short-term baseline drift in the detection system, we obtained an upper limit of 10^{-4} for the ratio of the ASE intensity to the peak intensity of the amplified laser.

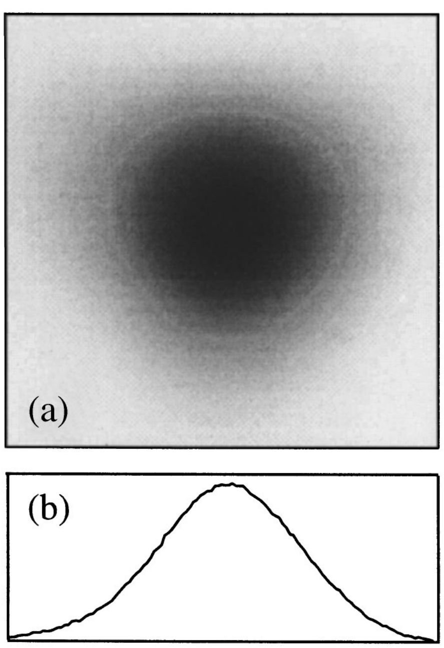

FIG. 4. (a) Spatial profile of the dye laser. (b) Cross section of the profile taken horizontally through the center of the beam showing its near Gaussian spatial quality.

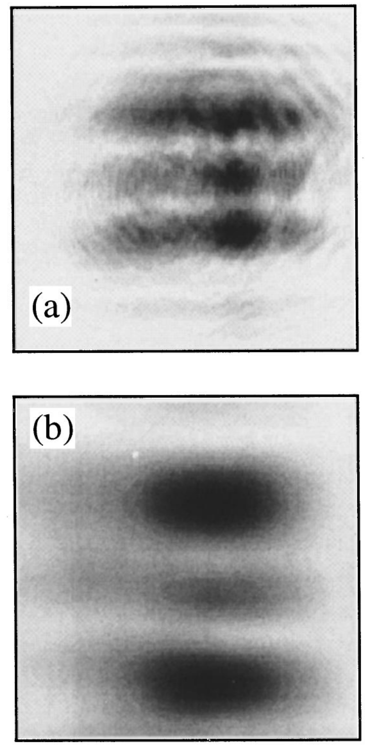

FIG. 5. Spatial profiles of the pump and dye lasers showing the impact of gain guiding in the oscillator. In (a) is the Nd:YAG mode incident on the dye cell. In (b) is the resultant dye beam.

IV. SPATIAL PROFILE

The bare resonator cavity as described contains no beam shaping optical elements. The grating and tuning mirror can be replaced by a single flat mirror for mode calculations. The model cavity then contains two flat mirrors which is a marginally stable design with large diffractive losses. When the dye is pumped however, gain guiding serves to shape the

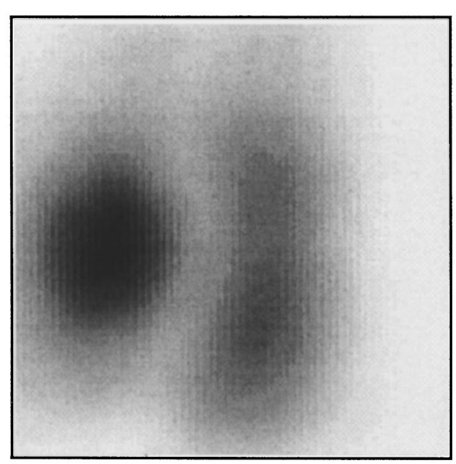

FIG. 6. Spatial profile of the dye beam in a quasisingle-longitudinal-mode laser. The two lobes are present when the laser is running in two longitudinal modes.

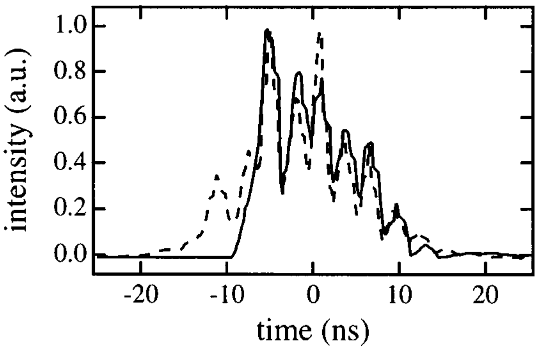

FIG. 7. Temporal profile of a single-longitudinal-mode dye laser (solid line) when pumped by a multilongitudinal mode Nd:YAG laser. Shown as a dashed line is the pump temporal profile [from Fig. 2(a)] that produced the dye laser pulse. The temporal structure in the dye laser pulse follows that of the pump.

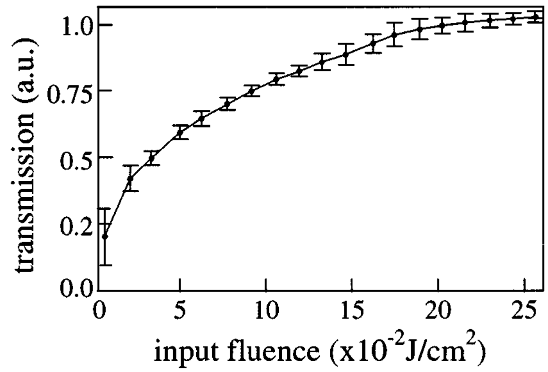

FIG. 9. Normalized transmission of the dye beam as a function of the input dye fluence. The transmission is normalized to the saturated value because the measurement did not account for the reflection losses in the cell. A smooth line is drawn through the data points with error bars indicating the noise standard deviation.

output beam and make the cavity less lossy.[21] Thus the dominant beam shaping within this cavity is done by the gain region which depends on the transverse quality of the Nd:YAG pump beam. To demonstrate the gain guiding of this resonator, we first aligned the dye resonator to achieve the best spatial mode, which is nearly Gaussian and is shown in Fig. 4. We then modified the pump beam (nominally Gaussian) from the Nd:YAG by placing a thin wire horizontally in the beam after the focusing lens. This served to create a beam with horizontal nulls in the focused beam profile at the cell. This is shown in Fig. 5(a). Then, without making any changes to the dye cavity, the output beam from the dye was measured and is shown in Fig. 5(b). The correlation in shape is clearly visible. It is important to note that the dye cavity is not misaligned so that some higher order mode (e.g., TEM_{20}) is lasing as a result of the bare cavity. Rather, the shape of the gain region is determining the beam shape of the output. We also verified this gain guiding by using only slightly distorted pump beams and obtained similar (though less dramatic) results.

The spatial quality of the dye beam is therefore critically linked to the spatial quality of the pump beam. Since the dye beam appears nearly Gaussian, we sought to quantify the near diffraction limited performance by measuring the M^2 beam quality parameter.[22] This entailed measuring the spot size at the focus of a high quality ($f=100$ mm doublet) lens and in the far field of that lens. We measured $M^2=1.2$ which is comparable to the lowest M^2 obtained for a cw argon ion laser in a recent study.[23] We therefore see that the high quality (TEM_{00}) mode of the Nd:YAG pump gives rise to a near diffraction limited mode of the dye beam in this type of cavity. This is one of the main benefits of the use of the longitudinally pumped gain region as opposed to the original transversely pumped design.[9,13]

A final issue affecting transverse mode quality is whether or not the laser is running SLM. We mentioned above that different longitudinal modes travel slightly different paths in the oscillator cavity. This can give rise to slight angular deviations in the direction of the output beam. It also can radically modify transverse mode profiles. We show in Fig. 6 a spatial mode profile obtained when the dye laser was running in two longitudinal modes. This cavity was quasi-SLM in the sense that changing the cavity length (by $\lambda/2$) with the PZT could cause the laser to cycle through SLM operation to double mode operation and back to SLM. When the laser was in double mode operation, the different longitudinal modes actually were separated transversely in space,

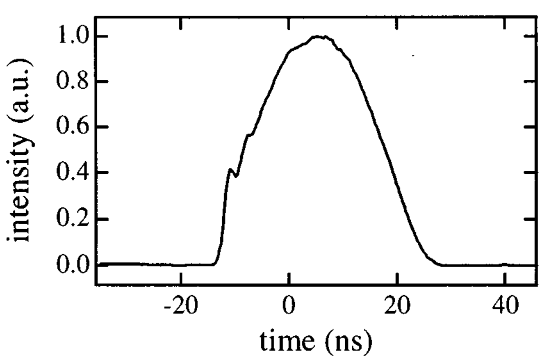

FIG. 8. Temporal profile of a single-longitudinal-mode dye laser when pumped by a single-mode (smooth) pump laser. The rapid rising edge is indicative of the output from the low-Q dye cavity.

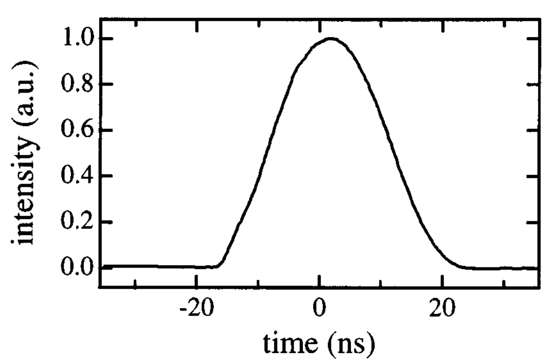

FIG. 10. Temporal profile of the single-mode dye laser beam after saturable absorption in the dye amplifier.

as seen in Fig. 6. By tuning the cavity length so that SLM operation was achieved, the spatial mode returned to its near Gaussian profile.

V. TEMPORAL PROFILE

When a multilongitudinal mode laser pulse is used to pump a dye laser, the temporal profile of the dye laser pulse will be similar to the pump pulse, even if the dye laser is SLM. This behavior is shown in Fig. 7 (temporal pulse shape data were taken with a Hi Voltage Components, Inc. Model PDH-114-P vacuum photodiode and a Tektronix 7912AD digitizing oscilloscope with a 500 MHz bandwidth). If the dye laser is pumped only slightly above threshold then the pulse may be smooth and significantly shorter than the pump pulse.[24] The amplitude fluctuations typically will be larger in this configuration, and the energy output will be reduced. Therefore it is often desirable to run the laser significantly above threshold with the dye laser pulse essentially following the pump pulse profile (for typical nanosecond Nd:YAG, N_2, and excimer pump pulses) as in Fig. 7.

If smooth dye laser pulses are required, then the pump pulse must be smooth as in Fig. 2(b). This is not enough to determine smooth output however. The dye laser must not exhibit mode-beating itself, and so SLM must be attained. While the pulse nominally follows the pump pulse, the low Q of the dye cavity can give rise to relaxation oscillation ''spikes'' (which is the source of the short pulse generation mechanism discussed in Ref. 24). These sharp features were observed experimentally and modeled theoretically by Sorokin *et al.*[25] A typical pulse with fast turn-on feature in Fig. 8. Here the dye laser is running SLM.

In order to eliminate this fast transient (which can be as short as 100 ps),[26] we took advantage of saturable absorption present in the dye amplifier itself.[27] Figure 9 shows the absorption of the dye beam incident upon the unpumped amplifier as a function of the input fluence. A typical saturation curve is obtained. This gave rise to pulse reshaping depending on the amount of input fluence. If the fluence was very small or very large with respect to the saturation fluence, then essentially no reshaping was observed. However, when the input fluence was near 2×10^{-2} J/cm^2, the pulse was modified and had the temporal profile shown in Fig. 10. Here we see that the fast transient has been eliminated and a smooth pulse results. When the amplifier is pumped, the reshaping remains as long as the amount of gain in the amplifier is limited to <3. This way, the amplifier can act as a pulse reshaper combined with a preamplifer. If the pulse reshaping takes place in a region of the dye cell that is unpumped, then subsequent amplification can be large without detrimental impact on the pulse shape.

ACKNOWLEDGMENTS

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