Continuously Tunable Picosecond-Pulse Organic-Dye Laser*

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(Received 24 June 1968; in final form 19 August 1968)

Picosecond-pulse outputs, tunable over a broad spectral range, have been demonstrated in an organic-dye laser. Evidence is presented that no more than a small fraction of the available bandwidth cooperates in mode locking to produce individual short pulses.

Mode locking of organic-dye lasers has been reported recently by several groups[1–3] using mode-locked lasers as pumping sources. Most recently, self-mode locking of a flash-lamp-pumped dye laser has been described.[4] These studies reported the oscilloscopic observation, with subnanosecond resolution, of trains of pulses of considerable modulation, with periods of the round-trip cavity times. The appearance of these possibly instrmentally broadened pulses with proper temporal periodicity is a necessary condition for the existence of some degree of mode locking. We report here the observation of 1×10^{-11} sec pulses generated by a laser-pumped dye laser. Furthermore, without degrading the pulsewidth, continuously tunable outputs of 6 cm^{-1} bandwidth have been demonstrated.

A mode-locked neodymium glass laser was constructed for use as a pumping source for the organic-dye laser. All intracavity reflective elements were set at the Brewster angle and mode locking was effected by the use of a bleachable absorber near the 100% reflector in a cell positioned[5] to minimize the intensity of satellite spikes on the main pulsetrain. The pulse was structured in groups of 10 or 11 individual pulses separated by 1×10^{-10} sec, an interval determined by the internal round-trip time in the cavity output reflector. The surfaces of this glass reflector were wedged at 2′ and one surface was coated with a 30% dielectric reflector creating a low-Q resonator. These groups of pulses themselves were separated by 3×10^{-9} sec, $2L/C$ of the cavity with typically 10–15 sets of pulses between the half-power points. Picosecond pulses were detected by the two-photon excitation of fluorescence technique.[6] A solution of Rhodamine 6G in ethyl alcohol was employed as the fluorescent medium. The second harmonic of this laser generated by an ADP crystal revealed the same pattern of pulses with the same temporal duration of $4\pm1\times10^{-12}$ sec. A solution of Esculin in ethyl alcohol was employed in this case as the two-photon fluorescent medium. The spectral bandwidth of the second-harmonic output was 150 cm^{-1}.

These second-harmonic pulses were used to transversely pump a Rhodamine 6G dye cavity. An ethanolic solution of the dye with decadic absorption coefficient equal to 10 at 530 mμ was contained in a cell constructed with internal and external window surfaces disposed at their respective Brewster angles. The internal length of the cell was 1.15 cm. Laser action in unwanted directions was suppressed by degrading all optical surfaces excepting the windows. Wedged dielectric coated mirrors of ~99% reflectivity and of 60%–90% output reflectivity were employed.

When the dye-laser cavity optical length L_l was made equal to (or to a small submultiple of) the pump cavity optical length L_p and the Rhodamine laser dye cell put at the end of the cavity, trains of fully modulated pulses of periods equal to (or to small submultiples of) the pump pulse were observed with instrumental temporal half-width of 7×10^{-10} sec using a planar photodiode and traveling wave oscilloscope. With $L_p=L_l$ and the laser cell in intermediate harmonic positions L/n, $n=3$, 4; two pulses separated by $2L/nc$ occurred for each pumping pulse with one predominantly intense, as may be expected if one considers that pulses traveling in opposite directions in the cavity reach the dye cell together only once per pump pulse. With the cell centered and $L_l=mL_p/n$; $m/n=2$, 1, $\frac{2}{3}$, $\frac{1}{2}$; $2n/m$ pulses are seen on the traveling wave oscilloscope for each pumping pulse, spaced at 3, $\frac{3}{2}$, 1, and $\frac{3}{4}$ nsec, respectively. These results are an extension of findings reported earlier.[2,3] When the pulse interval approaches the pulse resolution time, the overlap of the instrumentally broadened pulses reduces the apparent modulation drastically. When two or more pulses per pump pulse occur, the first two are of approximately equal intensity and any remaining ones show rapidly diminishing intensity, but with large statistical variations in decay rates on successive pictures. The fluorescent lifetime of Rhodamine 6G has been recently redetermined as 5.5×10^{-9} sec for an approximately equal concentration solution.[7] This time is greater than any of the periods described above, a condition which would diminish the effectiveness of this dye were it used as a passively bleachable absorber to engender mode locking with the cavity dimensions and configurations described above.

* This research is part of Project Defender under the joint sponsorship of the Advanced Research Projects Agency, The Office of Naval Research and the Department of Defense.
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However, this is a driven system with periodic gain modulation. The superradiant lifetime over the pumping intensity range employed was found to be greater than 1×10^{-9} and less than 3×10^{-9} sec by removing the cavity reflectors and observing the ability of the emitted radiation to follow stimulation of diminishing periodicity. The effective cavity lifetime including the driven dye-laser cell was inferred from the multipulse decay rates to be between 1 and 2 nsec.

The picosecond pulse structure of the dye laser was observed using the alcoholic solution of Esculin. Multiple exposures were often required and so for convenience the two-photon fluorescence cells were sometimes positioned in the cavity, a procedure empirically justified by the finding of no significant change in the observed pulse shapes. More dispersive, scattering, and generally nonlinearly behaving solvents than ethanol might be expected to cause complications. Several of the cavity configurations mentioned above were tried all yielding similar picosecond pulses patterns. The additional expediency of increasing the f No. of the camera lens with inexpensive positive portrait lenses allowed the use of less sensitive but higher gamma emulsions (Polaroid type 52, $\gamma\sim3.5$ or type 146-L, $\gamma\sim2.5$) to photograph the two-photon-excited fluorescence. This is an aid in densitometrically determining the contrast ratio (see below), as the fractional error in contrast ratio, for an error in optical density, is inversely proportional to gamma.

The structure of the picosecond pulses closely followed that observed for the pump, except that the pulsewidth was $1.1\pm0.1\times10^{-11}$ sec. The contrast ratio was determined to be 2 ± 0.2, and the contrast ratio of the pump pulses was found to be 1.8 ± 0.2. Several approaches have been taken to attempt to realize more of the full mode-locking potential of the ~150 cm^{-1} dye laser bandwidth which might yield pulses as short as $\sim2\times10^{-12}$ sec in duration. However, no significant changes in pulsewidths were noted when the spatial width of the pumping pulse was narrowed from 12 to 1 mm by focusing with a cylindrical lens in order to bound the 10^{-11} sec pulse length, when fine variations were made in the position of the pulse in the dye cell and the position of the cell itself, or when operating the dye laser from 10 to as much as 20 times above threshold. Higher output mirror reflectivities produced only trivially shorter pulses within the estimated deviation.

One condition that might contribute to the pulsewidth is a frequency sweeping of the laser output, a circumstance reported for certain cyanine dye lasers[1,8] It should also be noted that because of the pulse structure of each laser burst in these experiments, each of the picosecond pulses displayed in the two-photon fluorescence technique is actually an average over $>10^2$ discreet pulses. Each pulse may be narrower than the average but broadened by aperiodicity within each 10^{-9} sec group of pulses or from one such 10^{-9} sec pulse to the next. Furthermore, as has recently been discussed,[9,10] two-photon fluorescence pictures of picosecond pulses with contrast ratios of less than three, the maximum value which can be obtained as for example in the case of fully mode-locked operation, are fraught with uncertainties about the actual nature of the pulses. Contrast ratios of two which are observed here might indicate a free running oscillation[10] or random (initial condition) mode-locked laser.[9] It should further be noted that all experimental problems as, for example, scattering and imperfect overlap of the beams, would tend to reduce the observed contrast ratio. Because of this uncertainty we can quote only the bounds of the average peak power between a maximum and a minimum had there been no picosecond pulse content: for the pump $8\times10^6<P(\text{watts})<10^8$; for the dye laser $2\times10^6<P(\text{watts})<2\times10^7$ using an output mirror reflectivity of 60%. A smaller number of picosecond pulses might be expected to yield higher powers. The full beam angle at half-power density was measured as 2.0 mrad by a self-calibrating technique[11] which obviates the problem of film reciprocity.

Efficiently confining the spectral output of the dye laser to 6 cm^{-1}, by replacing one of the cavity mirrors by a diffraction grating as described in an earlier publication,[12] we have demonstrated a continuously tunable source of $1.1\pm0.1\times10^{-11}$ sec pulses over a half-power tuning range of 600 cm^{-1}. Narrowing the spectral output much beyond this would, of course, broaden the pulsewidths. The pulse pattern again resembled the pumping pulse structure and had a measured contrast ratio of 2 ± 0.2. The Rhodamine cell was centered in the cavity with $L_l=L_p$. Using a grating of 2160 lines/mm of approximately constant first-order reflectivity of 58% in the range of interest in the Littrow position and a 100% reflector, the energy output in the zeroth order at the peak of the tuning curve was greater than 50% of that obtained with the 60% dielectric reflector replacing the grating. The full beam angle at half-power density was found to be 4.8 mrad.

It should be noted that pulse duration with narrow spectral output is identical to that obtained with no grating when the full 150 cm^{-1} bandwidth was generated. This indicates that no more than 6 cm^{-1} of the available bandwidth cooperated in mode locking to produce any one short pulse.

The technique described in this paper, providing tunable short pulses, is expected to work well with many families of dyes, both in liquid and plastic[12] hosts covering the entire range from the near ultraviolet to the near infrared. The flash-lamp-pumped self-mode-locked dye laser[4] would also be amenable to this technique. Single tunable picosecond pulses or pulses with other intervals should also be obtainable.

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