

## PICOSECOND PULSE EMISSION FROM A MODE-LOCKED $\text{Nd}^{+++}$ $\text{POCl}_3$ LIQUID LASER

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A mode-locked  $\text{Nd}^{+++}$  ( $\text{POCl}_3$ ) liquid laser has been constructed which emits picosecond pulses in a train lasting as long as microseconds. The spectral bandwidth exceeds  $100 \text{ cm}^{-1}$  about the output wavelength of  $1.052 \mu$ . Stimulated Raman scattering has been observed to be generated inside the laser.

Samelson and Lempicki [1] first demonstrated mode-locking of a  $\text{Nd}^{++}$  ( $\text{SeOCl}_2$ ) liquid laser. They reported pulse durations of 10 to 40 psec and pulse train durations of  $\approx 100$  nsec. We have obtained picosecond pulse emission in a train lasting microseconds from an  $\text{Nd}^{+++}$  ( $\text{POCl}_3$ ) liquid laser. The significant improvement is attributed to a carefully designed optical cavity which eliminates spurious reflections. Results indicate that the liquid laser has advantages over the much used  $\text{Nd}^{+++}$  (glass) laser [2] for some picosecond pulse applications.

The  $\text{Nd}^{+++}$  ( $\text{POCl}_3$ ) liquid laser design is shown in fig. 1. The composition of the  $\text{Nd}^{+++}$  ( $\text{POCl}_3$ ) solutions is (in moles per liter)  $\text{Nd} - 0.3$ ;  $\text{ZrCl}_4 - 0.45$ ;  $\text{POCl}_3 - 9$  and a reaction product (dimer)  $\text{P}_2\text{O}_3\text{Cl}_4 - 0.9$ . The liquid is contained in a fused quartz cell with end faces cut at the quartz-air Brewster angle. The length of the active medium is 25 cm and the diameter is 1.0 cm. The mirror  $M_1$  has a 10 m radius of curvature and is 100% reflecting at  $1.05 \mu$ . Several flat output mirrors  $M_2$  are used; their reflectivity varies between

4% and 80%, and, most important, the rear uncoated face of each mirror is tilted at an angle of from  $1^\circ$  to  $5^\circ$  with respect to the coated face to avoid feedback. The  $\text{Nd}^{+++}$  ( $\text{POCl}_3$ ) liquid solution has a refractive index of  $n = 1.478$  at  $1.05 \mu$ , and thus provides a good index match with the fused quartz ( $n = 1.45$ ). An  $\text{Nd}^{+++}$  ( $\text{SeOCl}_2$ ) solution ( $n = 1.67$ ) might lead to spurious reflections because of the greater mismatch with quartz. The laser is Q-switched with Kodak 9860 dye with transmission between 60 and 70%. The Kodak cell is antireflection coated at  $1.06 \mu$  and is tilted at  $45^\circ$  with respect to the laser axis to avoid feedback. Spectra of the laser output are obtained on sensitized Kodak 1Z plates mounted in a 2 m Bauch and Lomb spectrograph with a resolution of  $0.01 \text{ \AA}$ . The pulse train is detected by a TRG 105B photodiode which is connected to a Tektronix 519 scope (resolution 0.5 nsec). Pulse trains from the liquid laser are compared relative to  $\text{Nd}$ :glass laser pulse trains using this photodiode-scope combination. The  $\text{Nd}$ :glass laser used in this comparison consists of a Brewster-Brewster cut

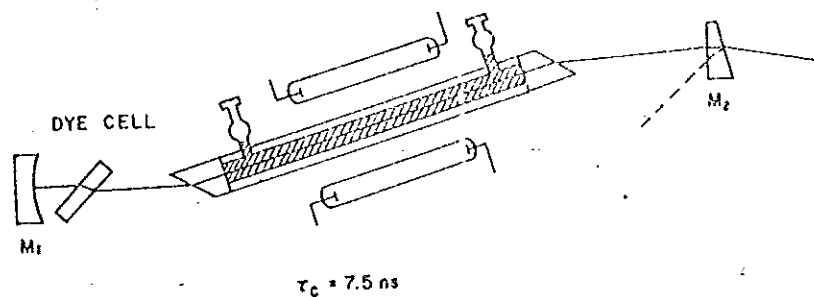


Fig. 1. Schematic arrangement of mode-locked liquid laser.

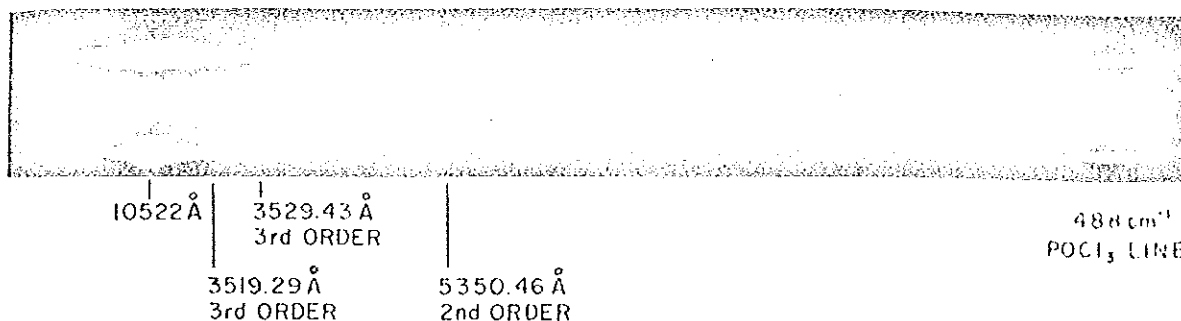


Fig. 2. Spectra of mode-locked liquid laser. Two shots show  $\approx 100 \text{ cm}^{-1}$  central band and Raman shifted light.

$7\frac{1}{2}$  in.  $\times$   $\frac{1}{2}$  in. Owen Illinois glass rod (ED-2), a Korad (K-1) laser head, a 10 m 100% reflectivity rear dielectric mirror, a 50% dielectric wedge output mirror ( $\approx 10^\circ$ ), and a Kodak Q-switch dye (Kodak dye 9860) cell. Pulses are also displayed by the two-photon fluorescence technique using Rhodamine 6G in dichloroethane.

The spectral output of the laser is shown in fig. 2. The spectrum of the laser emission is  $\approx 100 \text{ cm}^{-1}$  wide and is devoid of any structure except for a sharp line  $\approx 2 \text{ cm}^{-1}$  wide near the center of the spectrum at  $1.052\mu$ . A similar sharp line appears in  $\text{Nd}^{+++}$  glass laser spectra [3]. Liquid spectra are, like glass spectra, asymmetric about this line with more light emitted toward the red than the blue. Also present in fig. 2 is stimulated Raman scattering which is generated in the active medium from the  $488 \text{ cm}^{-1}$  line of the  $\text{POCl}_3$  molecule. The stimulated Raman conversion in the mode-locked laser was measured to be as high as 12%. The Raman line center shifts as much as  $10 \text{ \AA}$  from shot to shot, presumably because individual pulses in the laser generate different spectra and those pulses which produce stimulated Raman scattering make up a smaller part of the laser spectrum.

Of special significance are the "clean" pulse trains. The scope trace shows single pulses spaced at the roundtrip time of the cavity (7.5 nsec). The pulse widths are limited in resolution only by the combined resolution time of the photodiode and scope. Typical pulse trains are shown in fig. 3. The pulse train often lasts *microseconds*. Our results contrast with the  $\text{Nd}^{+++}$  glass laser data. When we operate an  $\text{Nd}^{+++}$  glass laser with the same mirror reflectivities, cavity length, and dye concentrations, we obtain pulse trains of  $\approx 200 \text{ nsec}$  and a multiple pulse output. To obtain a clean pulse train with an  $\text{Nd}^{+++}$  glass laser, extra effort is involved to eliminate multi-

ple pulsing [4]. When the liquid laser pulse train lasts microseconds, over one thousand intense pulses have been displayed. Then the peak height of the liquid laser mode-locked pulse train displayed on the photodiode-scope combination is approximately 3 to 10% the peak height of our  $\text{Nd}$ :glass mode locked laser, and the liquid laser emits about 10 to 20 times as many pulses as the glass laser. Results suggest that the liquid laser emits more pulses but of lower power than the glass laser. However, with an output mirror of 50% reflectivity and a dye transmission of 43%, the  $\text{Nd}$ :liquid laser emitted pulses  $\approx 20\%$  of the peak height of a glass laser in a train of  $\approx 300 \text{ nsec}$ . These results are tentative because  $\text{Nd}$  concentrations, pumping arrangements, and active lengths differ in the two lasers. Peak powers of the liquid laser pulses of  $\approx 1 \text{ GW}$  have been obtained thus far.

"Clean" two-photon fluorescence patterns verify that just a single pulse is rattling back and forth in the cavity. The two-photon fluorescence display shows a single bright spot at the mirror. The pulse duration is computed to be  $\approx 3 \text{ psec}$ . The resolution time is limited by photographic techniques. Since the two-photon fluorescence pattern appears to be nearly identical to that of the  $\text{Nd}$  glass laser and since the spectral width is similar, shorter substructure must be present [5].

Experiments were carried out to explain the improvement over the results obtained by Samelson and Lempicki [1]. When parallel glass flats are introduced inside our laser cavity and aligned perpendicular to its axis, the spectrum narrows. As extra flats are added the spectrum becomes as narrow as  $5 \text{ cm}^{-1}$ , and the pulse train displayed on the scope becomes as short as  $\approx 200 \text{ nsec}$ . Furthermore, the width of the pulses is no longer limited by the photodiode scope resolution time and spectra become modulated at the

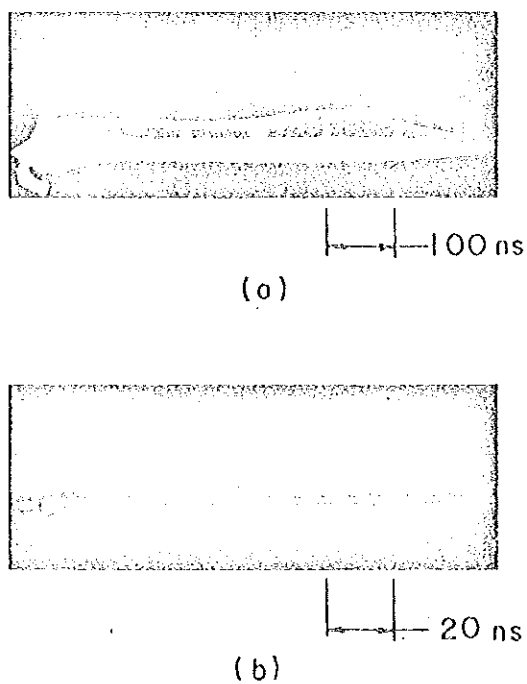


Fig. 3. Typical oscilloscope traces of laser pulse train. a) 100 nsec/cm; b) 20 nsec/cm. Fig. 3b has been air-brushed.

flat spacing. As shown in the experiment of Giordmaine et al. [6]<sup>\*</sup>, extra parallel surfaces

<sup>\*</sup> See also Stetser and DeMaria [7], who obtained a narrower spectrum on insertion of a parallel Fabry-Pérot reflector.

lead to modulated spectra and to emission of more complicated pulse trains as evidenced by two-photon fluorescence patterns. The plane parallel mirrors used by Samelson and Lempicki presumably introduced the same complicating effects.

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