

OBSERVATION OF MODE LOCKING AND ULTRASHORT OPTICAL PULSES INDUCED BY ANTISOTROPIC MOLECULAR LIQUIDS

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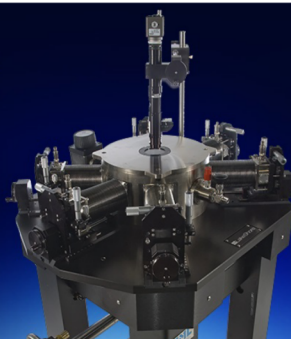
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beam comparison technique. The incident laser light was split into two beams of equal intensity which traversed the fluorescence cell in adjacent tracks. The TPF display beam was reflected upon itself by the 100% mirror in the cell; the second, unreflected beam provided a reference intensity $f = 1/2$. The intensity of various portions of the display beam were compared to the reference beam by photographing the former through a calibrated filter, producing the same exposure as the reference beam.

The measured background intensity $f_b = 1 \pm 0.3$ is consistent with strong mode locking and high picosecond pulse content. This result is consistent with that reported elsewhere¹ in which a contrast ratio $R \approx 10^2$ was reported for a similar laser with a SHG coincidence measurement. The measured contrast ratio in the present work was $R = 2 \pm 0.2$, a result indicating partial mode locking and agreeing with earlier TPF measurements of the second harmonic of a similar laser.⁴ The value of $f_p = 3$ at the expected positions was not observed.

We also measured TPF tracks produced by a single-mode dye Q-switched ruby laser in a solution of 9, 10 diphenylanthracene in benzene. A ratio of (6.1 ± 0.8) was found between the display and reference tracks, indicating peak intensity $f_p = 3.1 \pm 0.4$ as expected from the theory.

In conclusion we point out that the values $R \sim 2$ and $f_b \sim 1$ observed in the multimode laser TPF

displays are consistent with Nd:glass laser emission in the form of 2–4-psec pulses having substantial and random intensity fluctuations on a scale ~ 0.2 psec. Such internal structure is indicated by the observed spectral width of $100\text{--}200\text{ cm}^{-1}$. The apparent absence of the expected intensity peak with $f_p = 3$, $R = 3$, with $\sim 20\text{-}\mu$ width may be due to the limited recording resolution and is currently under investigation.

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OBSERVATION OF MODE LOCKING AND ULTRASHORT OPTICAL PULSES INDUCED BY ANTISOTROPIC MOLECULAR LIQUIDS*

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Mode locking and ultrashort pulses have been produced in a giant-pulse ruby laser with heated nitrobenzene ($T > 110^\circ\text{C}$) or α -chloronaphthalene ($T > 62^\circ\text{C}$) inside the optical resonator. 10^{-11} -sec pulses were observed with the two-photon fluorescence technique.

In a previous article¹ two of the present authors analyzed the effect of anisotropic molecular liquid on a multilongitudinal mode laser. It was shown that for sufficiently high oscillation power and at a temperature where the molecular reorientation time

τ is comparable to the inverse of the laser gain linewidth $\Delta\nu_{\text{gain}}$, the dielectric nonlinearity of the anisotropic molecular liquid should give rise to mode coupling and, consequently, to ultrashort pulses.

In this Letter we describe the experimental observation of mode locking and ultrashort pulses in a giant-pulse ruby laser induced by the presence of

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either α -chloronaphthalene or nitrobenzene inside the optical resonator.

The experimental setup consisted of a giant-pulse ruby laser employing a rotating prism with an average power density of about 10 MW/cm^2 and a 1-m cavity. Internal reflections were minimized through the use of Brewster-angle surfaces and a wedge output mirror. A Brewster-angle, 5-cm cell containing the liquid was placed inside the resonator. The liquid temperature could be controlled over the range -95°C to 140°C .

In Fig. 1 we show a typical two-photon fluorescence display of the output pulse at $.694 \mu$ with no liquid within the resonator. The (longitudinal) uni-

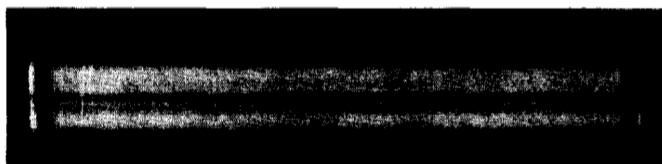


Fig. 1. The two-photon fluorescence display of rhodamine 6G with no anisotropic liquid inside the laser resonator. The longitudinal uniformity indicates the lack of ultrashort pulses. The mirror was placed so that pulses separated by a round-trip cavity time would overlap in the dye.

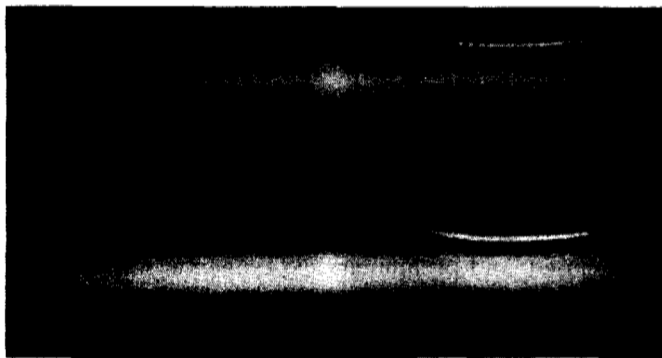


Fig. 2. Typical two-photon fluorescence displays obtained when a 5-cm cell containing α -chloronaphthalene at 62°C is present inside the resonator. The pulse length is approximately $1.2 \times 10^{-11} \text{ sec}$.

formity of the exposure indicates that no ultrashort pulses are present. The introduction of α -chloronaphthalene inside the resonator gives rise to ultrashort pulses as seen in Fig. 2. The temperature of the liquid is 62°C , and the relaxation time τ is estimated to be in the range 2×10^{-11} to $4 \times 10^{-11} \text{ sec}$. The pulse duration is estimated from Fig. 2 to be $\tau_{\text{pulse}} \sim 1.2 \times 10^{-11} \text{ sec}$.

Similar results were obtained using nitrobenzene except that ultrashort pulses occurred reliably *only* when its temperature was raised above 110°C . At

this temperature, $\tau \sim 1.3 \times 10^{-11} \text{ sec}$ (compared to a room temperature value of $4.3 \times 10^{-11} \text{ sec}$). The gain linewidth $(\Delta\nu)_{\text{gain}}$ was measured with a Fabry-Perot etalon at $\sim 1.8 \text{ cm}^{-1}$. The expected pulse width is $\sim (1/(\Delta\nu)_{\text{gain}}) 1.8 \times 10^{-11} \text{ sec}$, which is comparable to the measured value $\tau_{\text{pulse}} \sim 1.2 \times 10^{-11}$, indicating mode locking across the full gain line-width.

A number of interesting observations made in the course of this investigation concerns the process of stimulated Raman emission (SRE) in the anisotropic molecular liquid inside the optical resonator. When the cell containing α -chloronaphthalene was placed outside the resonator and in the absence of ultrashort pulses, we observed self-trapping⁴ accompanied by SRE in the α -chloronaphthalene. When the cell is introduced inside the resonator no SRE is observed. This is most likely due to the increase in threshold for self-trapping which occurs when τ_{pulse} becomes comparable to or shorter than the molecular relaxation time τ .⁵

With nitrobenzene inside the resonator we observed SRE both at $T < 110^\circ\text{C}$, where ultrashort pulses are infrequent, and at $T > 110^\circ\text{C}$ where ultrashort pulses occur regularly.

The following observations suggest that in both of these cases the SRE takes place *without* self-trapping:

- (a) a very nearly exponential dependence of the Raman intensity on that of the laser, with a gain coefficient comparable to the theoretical value; and
- (b) the lack of SRE in α -chloronaphthalene which has nearly the same threshold for self-trapping and relaxation time as nitrobenzene.

In Fig. 3 we see on the oscillograph both the laser output and the Raman light generated from a nitrobenzene cell at 90°C inside the cavity. Note the fast rise of Raman output with small increase of laser power. Furthermore, the Raman output is accompanied by substantial smoothing in the laser intensity fluctuations, suggesting the appearance of parametric saturation.⁶

The use of a nonlinear dielectric constant inside the laser cavity may be a very practical way of generating ultrashort pulses. Present techniques generally use a saturable absorber, which tends to enhance the filamentary structure of the laser output, often causing damage to laser components. The liquids used here reduced the filamentary nature of the beam even when compared to that without any liquid. This can be seen by comparing the transverse structures in Figs. 1 and 2. We found no damage to components when ultrashort pulses were generated by the anisotropic molecular liquids,

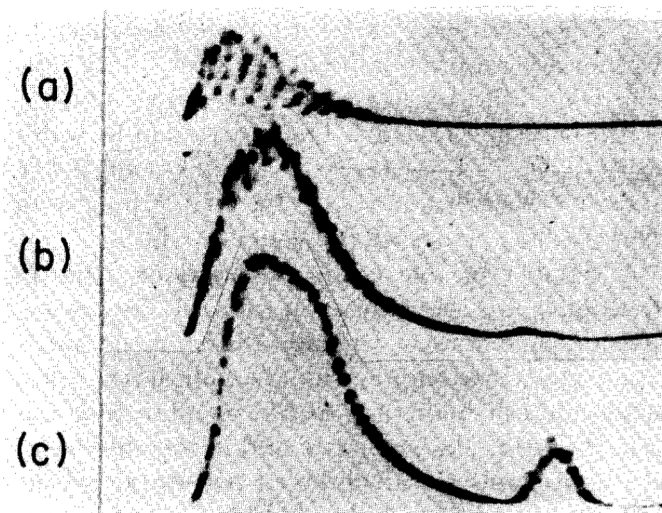


Fig. 3. Laser oscillation and stimulated Raman emission from nitrobenzene at 90°C inside the laser cavity. (a) Single pulse showing the large fluctuations in the laser output below the onset of SRE with a width of 40 nsec. (b) At an increased pumping level the very beginning of SRE is seen in the small pulse after the main laser pulse. The SRE pulse is delayed electronically by 10^{-7} sec so that a simultaneous single-trace display is possible. (c) At even higher pumping level; the laser intensity fluctuations are largely smoothed out.

while it was a frequent problem with saturable absorbers.

The bandwidth of the nonlinear dielectric coupling is determined by the molecular reorientation time, which can be varied by temperature tuning or by changing liquids. The possibility of using this effect for producing ultrashort pulses of variable length is being investigated.

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OPTICAL SAMPLING OF SUBNANOSECOND LIGHT PULSES

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An optical analog of the electronic sampling oscilloscope is proposed, and preliminary experimental results are presented. The sampling pulses, ~ 4 psec in duration, are produced by a mode-locked Nd:glass laser. They are mixed in a KDP crystal with pulses of ~ 800 -psec duration, from a mode locked He-Ne laser. The optical sum frequency is detected and displayed on a conventional oscilloscope. The pulse separations in the two lasers differ in such a way that, as time goes by, the sampling pulse scans the He-Ne pulse. There results, on the conventional oscilloscope, a sampled display of the He-Ne pulse.

Several techniques¹⁻⁵ have recently been developed for the display of picosecond light pulses. The technique we describe here is the optical analog of the electronic sampling oscilloscope.⁶ This technique, although based on the same principle as previous ones,¹⁻⁵ offers the advantages of being free from autocorrelation effects,⁷ of giving immediate oscilloscope displays, and of having high sensitivity.

In the electronic sampling oscilloscope, the repetitive pulse waveform to be displayed is sent to a diode bridge. The latter, normally closed, is opened by a so-called sampling pulse for a very brief moment,

during which a charge proportional to the instantaneous waveform amplitude leaks through, is amplified, and is displayed as a dot on a cathode-ray tube. The ordinate of the dot represents the amplitude of the waveform at a time represented by the abscissa. Successive waveforms are sampled at progressively increasing times and there results a display whose time resolution is determined by the width of the sampling pulse.⁶

In the optical domain the same basic idea can be applied by replacing the diode bridge by a nonlinear optical element. The sampling pulse and the pulse