SUBPICOSECOND GENERATION VIA HYBRID MODE-LOCKING
OF STYRYL 9 IN THE NEAR INFRA-RED

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By applying a hybrid mode locking technique, pulses as short as 0.55 ps, tunable in the region of 840 nm have been obtained from a cw styryl 9 dye laser. The dye IR-140 which was used as the saturable absorber was initially shown to give complete modulation with styryl 9 in a simple flashlamp-pumped laser system.

1. Introduction

The generation of subpicosecond frequency-tunable pulses in the near infra-red is of importance to the study of the kinetics of the excited states of many semiconductor materials [1]. Generally, sources in this spectral region are obtained by frequency shifting the amplified output of a passively mode-locked cw rhodamine 6G dye laser by a nonlinear technique [2]. However, the simplest approach is to generate the required frequency directly using either passive, synchronous or hybrid mode-locked dye laser systems. The recently introduced styryl dyes [3,4] are especially efficient in the near infra-red and have the added advantage that their peak absorption bands are such that they can be excited with Ar+ radiation at 514 nm.

In this letter we report on the generation of frequency tunable subpicosecond pulses from a cw styryl 9 dye laser using a hybrid mode locking technique. The saturable absorber used had been shown to give complete modulation of styryl 9 when placed in a simple flashlamp pumped laser system.

2. Experimental

As ethanolic solutions of styryl 9 exhibit a rather broad absorption band peaking around 582 nm ($\sigma_{max} \approx 1.7 \times 10^{-16}$ cm$^2$), it was operated initially in a flashlamp pumped configuration, and the effect of a saturable absorber on the laser output examined. The flashlamp pumped cavity arrangement has been described previously [5]. An uncooled $2 \times 10^{-4}$ M ethanolic solution of styryl 9 (LDS 820 - Exciton) was continually circulated in the 127 mm long, 3 mm i.d. quartz dye cell and was excited using a standard double elliptical pumping geometry. Typically the electrical energy deposited into the system was $\sim 150$ J. An overall cavity length of $\sim 30$ cm was used, formed by a 100% $R$ and $\sim 70$% $R$ mirrors in the range 820–880 nm. Tuning was carried out with a 5.5 $\mu$m air gap Fabry-Perot etalon. A static solution of the saturable absorber was placed in a 500 $\mu$m thick, optically contacted dye cell placed on the 100% reflector.

Maximum modulation of the styryl 9 output was found using ethanolic solutions of the saturable absorber 5,5'-dichloro-11-diphenylamino-3,3'-diethyl-10,12-ethylene thiatricarbocyanine perchlorate (IR-140 Kodak). Fig. 1(a) shows an example of $\sim 60$% modulation obtained in a 500 ns output train at 850 nm for the dye laser cavity arrangement described above. Typically the modulation achieved was of this order and although total modulation could be obtained (fig. 1(b)) it was critically dependent on operation close to threshold, and for our laser system this was difficult to achieve with any degree of reproducibili-
Fig. 1. (a) Typical output wave form at 850 nm of styryl 9 flashlamp pumped dye laser, mode locked using IR-140 as saturable absorber. (b) Complete modulation achieved with the above system, both 100 ns/small div. (c) As in (b) only 5 ns/small div. single broad bunches of pulses per round trip.

Fig. 2. Fluorescence decay of styryl 9 in ethanolic solution.

This behaviour can be explained by considering the necessary parameters to achieve stable passive mode-locking in dye laser systems [6]. Synchroscan streak camera measurements of the fluorescence lifetime of styryl 9 in ethanol indicated a fluorescence decay time of 640 ps (see fig. 2). However, the cavity round-trip time was 2.5 ns and under such circumstances it is possible for the amplifier gain to recover completely during a cavity period. If these conditions prevail the evolution of a single mode locked pulse is not in a stable regime [6] and the laser output will tend to consist of broad structured pulses. In fig. 1(c) it can be seen that although modulation was practically complete the generated pulses appeared relatively broad and no further time-resolved measurements of the mode locked output were carried out because the physical dimensions of the flashlamp pumped laser cavity did not permit any further reduction of the cavity period to assist in the generation of single picosecond pulses.

The action of IR-140 was then examined in a hybrid mode locking arrangement in a synchronously pumped c.w. styryl 9 dye laser system, where the cavity parameters were more easily controlled. A standard linear cavity was used for the synchronously pumped c.w. dye laser [7]. The acousto-optically mode locked Ar+ laser pump source provided an average operating pump power of ~900 mW in pulses of 80 ps at ~82 MHz repetition rate. The styryl 9 was made up as a 2 × 10^{-3} M solution in the mixed solvents 1 : 4 propylene carbonate : ethylene glycol, and flowed in a 100 μm jet placed between 5 cm radius of curvature, nominally 100% reflecting mirrors (800–900 nm), in a conventional three mirror cavity arrangement. The output mirror had a reflectivity of 80% at 850 nm increasing to 90% at 900 nm, and wavelength selectivity was achieved with an intracavity dielectric tuning wedge. The maximum average power with styryl 9 was 90 mW at 830 nm and the tuning range extended from 800 nm (20 mW) to 890 nm (10 mW). Typically the pulse durations, in the re-
Fig. 3. Autocorrelation traces of (a) 0.9 ps pulse obtained from a synchronously mode locked styryl 9 dye laser, (b) 0.55 ps pulse from hybrid mode locked operation of system in (a).

Fig. 4. The dependence of the pulse duration on the absorber dye concentration for hybrid mode locked operation at 840 nm.

830–840 nm, in hybrid operation. For a similar enhancement to be achieved at other wavelengths readjustment of the saturable absorber concentration is necessary. Although the inclusion of the absorbing dye substantially reduces the useful tuning range of the laser to 815–850 nm, the stability was appreciably enhanced by the presence of the absorber. No detectable deterioration of the absorbing dye was observed after many hours (>100) operation of the hybrid system.

In conclusion, we have shown that the laser dye IR-140 is a suitable saturable absorber to use in a hybrid mode locking technique with a synchronously pumped c.w. styryl 9 dye laser system. Stable, subpicosecond pulses tunable in the near infra-red region around 840 nm are generated which should be especially useful in band gap studies of some semiconductor species.

References