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**Title:** High efficiency picosecond pulse generation in the 675-930 nm region from a dye laser synchronously pumped by an Argon-ion laser

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Picosecond pulses tunable from 675 to 930 nm have been obtained from a dye-laser synchronously pumped at 514.5 nm by a mode-locked Argon-ion laser. Peak energy conversion efficiencies between 10% and 29% are observed with pulse durations between 1.7 ps and 16 ps as measured by autocorrelation.

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INTRODUCTION

In this letter we describe the generation of wavelength-tunable picosecond pulses in the far-red to near-infrared by synchronously pumping a dye laser with the 514.5 nm line of a mode-locked Argon-ion laser. Near IR picosecond pulse generation by synch-pumping has been reported earlier,1 using the less intense 647.1 nm line of a Krypton laser. An alternative technique for obtaining infrared laser emission with an Argon-ion laser is to cascade two dye-lasers, so that the output of the first dye laser (typically in the orange-red) is used to pump the second dye-laser.2,3 This technique however suffers from low overall efficiency and stability. Generation of picosecond pulses at even longer wavelengths is possible by pumping at 1.06 μm with the fundamental of a Nd laser, as shown recently by Seilmeier et al.4

Several new commercially available dyes5 can now provide deep red to near-infrared dye laser operation with green pump light. Figure 1 shows the absorption spectrum, measured with a Hewlett Packard model 8450 parallel-detector spectrometer, of four such dyes: Pyridine-1 (also designated LDS-698), Styryl-8 (LDS-751), Styryl-9 (LDS-820) and LDS-821 (a dye very similar to Styryl-9 but with a longer lifetime under photochemical excitation). Earlier reports6,7 indicate that these new compounds lase extremely efficiently in the deep red to the near-
infrared when continuously pumped in the green, with efficiency as high as 25% reported\(^6\) for Pyridine-1.

We report here the characterization of the efficient synch-pumped operation of these dyes using standard commercial equipment.

**EXPERIMENT AND RESULTS**

The pump laser in this experiment is an Argon-ion laser, Spectra-Physics (SP) model 171, mode-locked at 514.5 nm to produce 150 ps (FWHM) pulses at \(^8\)0 MHz (output power 0.8 watt) or \(^\sim\)240 MHz (output power 1.8 watts). The dye laser (SP model 375) uses a tuning wedge or a one (or two)-plate birefringent filter. Internal reflectors and output coupler mirrors are matched to the spectral band of interest for maximum power or shortest pulse as shown in Table 1.

Alignment of the dye-laser does not present any major problems. Dye fluorescence up to 850 nm can still be seen with the naked eye in a dark room. Above 850 nm alignment of the cavity requires the use of an IR phosphor coated card.\(^8\)

**Table 1.** Dye-laser operating parameters.

<table>
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<tr>
<th>Dye</th>
<th>Concentration (mmole/liter)</th>
<th>Optics</th>
<th>Range (nm)</th>
<th>Output Power (mW)</th>
<th>Pulse Width (ps)</th>
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<td>1.8</td>
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<td>125**</td>
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<td>190</td>
<td>12.0</td>
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<td>700-812</td>
<td>190</td>
<td>12.0</td>
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<td></td>
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<td>G0058-905</td>
<td>700-812</td>
<td>190</td>
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<td>Styryl-9</td>
<td>1.85</td>
<td>G3845-011 G0058-905</td>
<td>775-840</td>
<td>180*</td>
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<td>790-931</td>
<td>630</td>
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* Operation at 80 MHz with a one-plate birefringent filter.
** Operation at 80 MHz with a two-plate birefringent filter.
***Recorded at peak power.

All others at 240 MHz with a tuning wedge.

The pulse widths are measured with an autocorrelator (SP model 409) with angle phase-matched KDP. They are always shorter when operating at 80 MHz. The wavelengths are measured using a calibrated 1/4 m grating spectrometer with 0.5 nm resolution, the power is recorded with a Scientech model 30-0001 laser power meter.

The dyes dissolve easily in dimethyl sulfoxide (DMSO) and propylene carbonate (PC), but have a limited solubility in ethylene glycol (EG), the standard solvent for jet-stream dye laser operation. Due to health hazards\(^9\) associated with DMSO/dye solutions, we avoided their use. A mixture of 15% PC (Aldrich, 99%) and 85% EG (Mallinkrodt, analytical reagent) was found to support the requisite concentration of dye without precipitation. This solvent mixture,
when maintained at room temperature, possesses sufficient viscosity for a stable jet stream.\textsuperscript{10}

\begin{figure}
\centering
\includegraphics[width=0.5\textwidth]{fig2}
\caption{Output power versus wavelength for mode-locked operation (240 MHz) with 1.8 W pump power and wedge tuning.}
\end{figure}

In Fig. 2 the average mode-locked output power is plotted as a function of the lasing wavelength. For 1.8 W of pump power at 240 MHz we observe an average output power peaking at 520 mW for both Styryl-9 and LDS-821. The maximum efficiency for Pyridine-1 is only 19% when sync-pumped, which is less than the 25% reported\textsuperscript{6} when pumped CW all-lines. This is probably due to the fact that Pyridine-1 absorbs more in the blue than in the green, as shown in Fig. 1.

Typically, the average power decreases by a factor of 2.5 when operating at 80 MHz with 0.8 W pump power instead of at 240 MHz and 1.8 W. In general, power goes up by \textasciitilde 20\% and temporal pulse width increases when using the birefringent filter (one or two plates) instead of the tuning wedge. The concentration has been optimized for maximum output power, but concentration changes of \pm 10\% do not materially affect the efficiency.

Pyridine-1 and LDS-821 show no noticeable photochemical degradation after 550 watts-hours. The Styryl-9 power output dropped by \textasciitilde 15\% after 100 watts-hours. Degradation measurements were not made for Styryl-8.

CONCLUSION

In summary, we have demonstrated the generation of picosecond pulses in the 675-930 nm spectral range, using an Argon-ion laser as a pump. This range has been previously accessible only to the less intense Krypton-ion laser. Peak efficiency as high as 29\% is measured for both Styryl-9 and LDS-821 at 809 nm, making them some of the most efficient argon-ion pumped laser dyes.

We thank the National Science Foundation, Chemistry and the Office of Naval Research, Chemistry for the financial support which has made this work possible, and the Swiss National Foundation for fellowship support to P. Bado. The authors thank R. Steppel of Exciton Chemical Co. for helpful discussions.

References

5. Exciton Chemical Co., Dayton, OH.
8. IR phosphor coated cards from Banner Engineering Corp., Minneapolis, MN, were found superior to other similar products.
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