BROADLY TUNABLE, NARROW LINEWIDTH DYE LASER EMISSION IN THE NEAR INFRARED*

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An efficient organic dye laser with a linewidth of less than 2 Å has been developed as a source of tunable high power radiation in the wavelength range 7500–9200 Å. Utilizing a 2 MW ruby laser pump, infrared output powers in excess of 200 kW have been attained for the range 7650–8900 Å with peak power levels above 400 kW. The tuning range for a single dye varies from 550 to 1030 Å.

The generation of broadly tunable, narrow linewidth, high power radiation in the near infrared is important for a number of scientific applications. The infrared radiation may be utilized as a spectroscopic source or in laser—matter interactions that require a high power, wavelength tunable light source. Frequency doubling of the near infrared radiation leads to a tunable source of high spectral brightness in the near ultraviolet and visible blue [1]. Also, difference frequency experiments can be performed which involve mixing tunable near infrared radiation with a fixed frequency laser source to generate tunable mid-infrared radiation [2–4].

We wish to report the operating characteristics of an efficient, narrow bandwidth infrared dye laser source. This source employs polymethine dyes [5] and is capable of a wide tuning range for a single dye, previously reported only for dye lasers operating in the visible spectrum. Broadband infrared dye lasers reported to date produce peak powers of about 1 MW in a 100–300 Å bandwidth for a 20 MW laser pump. Upon the introduction of wavelength dispersive elements to the dye laser cavity, narrowband infrared output is obtained with reported tuning ranges of 100–500 Å for a single dye [1–6].

A linearly polarized ruby laser, passively *Q*-switched with a solution of cryptocyanine in acetonitrile, is used as the dye laser pump in a longitudinal pumping configuration. The laser cavity employed is shown in fig. 1. Mirror M1 transmits 85% of the ruby radiation into the dye laser cavity where it impinges upon a 1 cm path length dye cell which is placed at Brewster's angle. However, M1 also reflects more than 99% of the infrared dye radiation from 7500–10 000 Å. The output of the laser is taken through mirror M2 which has a reflectivity of approximately 60% throughout the wavelength range of interest. A 60° glass prism set for an angle of incidence of approximately 80° preferentially transmits light of a horizontal polarization and also acts as a beam expander for the diffraction grating which forms one end

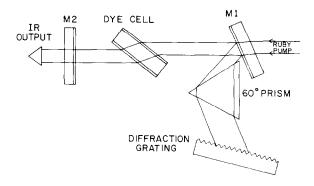


Fig. 1. Schmatic diagram of infrared dye laser.

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Table 1						
Tuning range for useful polymethine dyes in the spectral region from 7500 to 9000 A						

Dye	Abbreviation	Concentration	Tuning range	Tuning range, power ≥ 200 kW
3,3'-dimethyl-2,2'-oxatricarbocyanine iodide	OXA	$8 \times 10^{-5} M$	7500-8100 A	7650-8050 A
$1,3,3,1^{\prime},3^{\prime},3^{\prime}\text{-hexamethyl-2},2^{\prime}\text{-indotricar bocyanine iodide}$	HEXA	$6 \times 10^{-5} \mathrm{M}$	78008830 A	7890 -8570 A
3,3'-diethyl-2,2'-thiatricarbocyanine iodide	DTTC	$6 \times 10^{-5} \mathrm{M}$	7900-8705 A	8040-8450 A
3.3'-diethyl- $2.2'$ - $(4.5.4',5'$ -dibenzo) thiatricarbocyanine iodide	DIBENZO	$6 \times 10^{-5} \mathrm{M}$	8340-9000 A	8550-8900 Â
1,1'-diethyl-2,2'-quinotricarbocyanine iodide	QUINO	$8 \times 10^{-5} \mathrm{M}$	8650-9200 A	

of the cavity. The 1800 lines/mm grating is used in a first order Littrow arrangement and wavelength tuning is achieved by rotation of the grating. The grating is approximately twice as efficient when the dye radiation is polarized perpendicular to the grating grooves than when the polarization is parallel to the grooves. The combination of the Brewster angled dye cell, the 80° incident angle prism orientation, and vertical orientation of the diffraction grating grooves results in a dye laser cavity which predominantly transmits horizontally polarized infrared radiation. The dye laser operates just as well when the pump beam enters the laser cavity at a small angle to the main cavity axis [6]. This scheme ensures spatial separation of the pump beam and IR dye laser output which is often desirable.

Five polymethine dyes which cover the wavelength region 7500 to 9000 Å were investigated as listed in table 1. The output wavelength for a single dye is tunable over a 550 to 1030 Å range. Dimethyl sulphoxide (DMSO) was found to be the most efficient solvent. However stability is an important factor since a dye solution in DMSO will lose its lasing efficiency over a period of several days when exposed to conventional fluorescent lighting. On the other hand dye solutions in DMSO stored in amber bottles lase with high efficiency after storage for a number of months. Spectrophotometric-grade DMSO purchased from one manufacturer would not produce a lasing dye solution unless first purged with nitrogen. DMSO from a second manufacturer produced highly efficient lasing solutions for

both spectrophotometric and conventional laboratory grades. Tuning ranges obtained with the latter DMSO were twice as wide as those obtained with the nitrogen-purged solvent, and energy conversion efficiency was three times higher.

The power tuning curves for the infrared dyes are shown in fig. 2. Spectra were recorded with a 1 meter spectrograph and a thermopile of 0.2 mJ resolution was used to measure pulse energies. The temporal behavior of the ruby and dye lasers were recorded using silicon photodiodes and a Tektronix 7904 oscilloscope. Peak dye laser output power levels in excess of 200 kW

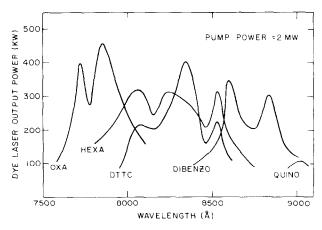


Fig. 2. Plot of dye laser peak output power as a function of wavelength.

and pulse durations of 8 to 30 nsec are obtained throughout the range 7650–8900 Å with a linewidth of less than 2 Å using a horizontally polarized 2 MW ruby pump. In order to preclude the production of multiple ruby laser pulses for a single flashlamp excitation, the maximum power used to pump the dye laser was limited to 2 MW in a single 25–30 nsec pulse. Substantially narrower linewidths are possible through the addition of Fabry–Pérot etalons to the dye cavity. The largest tuning range (1030 Å) is obtained by using a $6 \times 10^{-5} \mathrm{M}$ solution of HEXA in DMSO.

Peak energy conversion efficiency was approximately 15% when the ruby pumping radiation was horizontally polarized. Without wavelength selection, in a broadband dye laser cavity, the peak energy conversion efficiency increases to 35% for a 60 mJ single pulse ruby input and the emission linewidth increases to as much as 300 Å.

Dye laser peak power and energy conversion efficiency were also investigated for pumping by a ruby laser which had been internally apertured to a 2 mm diameter beam. This laser operated in a single longitudinal mode with lowest order transverse mode structure. Peak ruby pumping power for this configuration was 1.4 MW in a single horizontally polarized 20 nsec pulse. This pump yielded a maximum dye laser energy conversion efficiency of 18% for a 28 mJ pumping pulse, and dye laser output powers of up to 300 kW in a 17 nsec pulse were achieved.

For a broadband cavity the dye laser pulse is found to follow the ruby pumping pulse. For a narrowband cavity the dye laser pulsewidth largely approximates the pumping pulsewidth in the high-gain regions of the tuning range. For low-gain regions such as those encountered near the limits of a dye's tuning range the pulsewidth drops to 25 to 50% of the ruby pulsewidth.

Although energy conversion efficiency declines in the wings of the tuning range, this decrease in pulsewidth partially compensates for the reduced energy and produces a high power (\geq 100 kW) pulse. Power thus remains relatively high until the dye ceases to lase. (This pulsewidth narrowing is also found throughout the tuning range of low-gain dyes such as QUINO.)

In summary, we have demonstrated highly efficient, narrow linewidth laser action throughout a broad wavelength range in the near infrared. Output power as a function of wavelength is given for a number of dyes, and the temporal nature of the dye laser pulses is studied. Tuning ranges for infrared laser dyes are achieved which are double the maximum range previously reported.

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References

- [1] S.M. Hamadini and G. Magyar, Opt. Commun. 4 (1971) 310.
- [2] C.F. Dewey Jr. and L.O. Hocker, Appl. Phys. Letters 18 (1971) 58.
- [3] D.C. Hanna, R.C. Smith and C.R. Stanley, Opt. Commun. 4 (1971) 300.
- [4] D.W. Meltzer and L.S. Goldberg, Opt. Commun. 5 (1972) 209
- [5] Y. Miyazoe and M. Maeda, Appl. Phys. Letters 12 (1968) 206.
- [6] D.J. Bradley, G.M. Gale, M. Moore and P.D. Smith, Phys. Letters 26A (1968) 378.