Solid-state multiple-prism grating dye-laser oscillators

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Compact solid-state multiple-prism grating dye-laser oscillators are shown to yield in excess of 9% conversion efficiency at $\Delta\nu \approx 1.12$ GHz and a tuning range of 47 nm.

Key words: Solid-state dye lasers, tunable lasers, dispersive oscillators, multiple-prism grating oscillators, narrow linewidth.

Recently[1] we reported preliminary results on the use of solid-state dye-laser media in new and compact dispersive oscillators.[2] These dispersive oscillators (shown in Fig. 1) are of the multiple-prism Littrow (MPL) and the hybrid multiple-prism grazing-incidence (HMPGI) grating classes,[3] respectively, each with a Glan–Thompson polarizer output coupler.[2] The polarizer output coupler has its inner surface (facing the gain medium) antireflection coated, and its outer surface is coated for 20% reflection. Because the emission from these dispersive oscillators is highly polarized parallel to the plane of incidence as a result of the influence exerted by the multiple-prism grating assembly,[3] the function of the output coupler polarizer is to reduce the level of unpolarized amplified spontaneous emission.[4] Hence oscillators of this class have been shown to yield very low levels of amplified spontaneous emission.[4]

In the preliminary experiments[1] two types of solid-state dye-laser media were used. The first matrix was tetraethoxysilane doped with Rhodamine 590 and contained in an optical cell that provided a 10-mm active length. The other medium was a dye-doped organically modified silicate (ORMOSIL).[5,6] The geometry of this medium was a circular wedge 20 mm in diameter, with a thickness of 6–8 mm. In both cases the dye concentration was 2 mM. The excitation geometry was transverse, and a coaxial flash-lamp-pumped dye laser (Candela SLL-250) capable of delivering in excess of 50 mJ in a 160-ns pulse was used. Using tetraethoxysilane and ORMOSIL gain media in dispersive oscillators,[4] we obtained[1] energies ≤ 1 mJ and linewidths $\Delta\nu \approx 3$ GHz.

In this Note a significant improvement in the performance of solid-state dye-laser oscillators is reported. In these experiments the dispersive oscillator configurations illustrated in Fig. 1 are used in conjunction with modified poly(methyl methacrylate) (MPMMA) gain media[7] in the short-pulse regime. The results reported here indicate that solid-state dispersive dye-laser oscillators offer performance comparable to the highly successful liquid-phase dye-laser oscillators. Indeed the solid-state dye-laser oscillators described here should find wide applicability in fields such as spectroscopy.

In these experiments excitation was accomplished with a N_2 laser-pumped liquid C152 dye laser. This laser is pumped transversely and is composed of a simple mirror–mirror cavity, including a Glan–Thompson intracavity polarizer to induce p-polarized emission. When a dye concentration of 10 mM is used, this laser yields approximately 2 mJ in a 3–4-ns-pulse full-width at half-maximum (FWHM) at $\lambda \approx 520$ nm.

Output energies were measured with a calibrated pyroelectric energy detector (Gentec ED200) and a 7834 Tektronix oscilloscope. Temporal pulses were detected with a Hamamatsu biplanar phototube (R1193U) and a Tektronix SCD1000 transient digitizer. The rise time of the transient digitizer is ≤ 350 ps. Interferometric measurements were performed with a Burleigh RC110 Fabry–Perot interferometer with a finesse ≥ 30 and a free spectral range (FSR) of 7.49 GHz. Wavelength ranges were determined with a Spex 1681 spectrometer, and the beam divergence was measured photographically.

The MPMMA active medium has a 20-mm diameter with a cross section of trapezoidal geometry with

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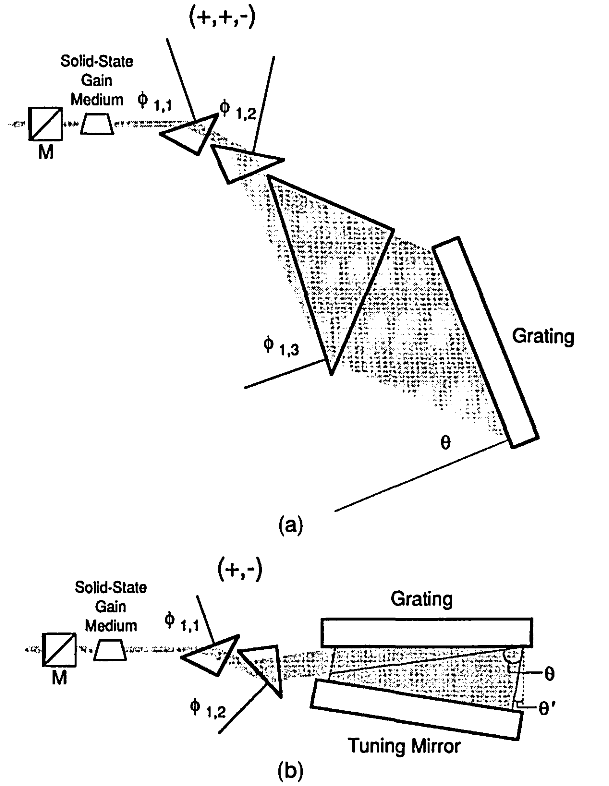

Fig. 1. (a) Solid-state multiple-prism Littrow (MPL) dye-laser oscillator, (b) solid-state hybrid multiple-prism grazing-incidence (HMPGI) dye-laser oscillator. Excitation was accomplished semilongitudinally, with the excitation beam incident at an angle of a few degrees relative to the optical axis of the cavity. Both oscillator configurations incorporate a Glan–Thompson polarizer output coupler.[2]

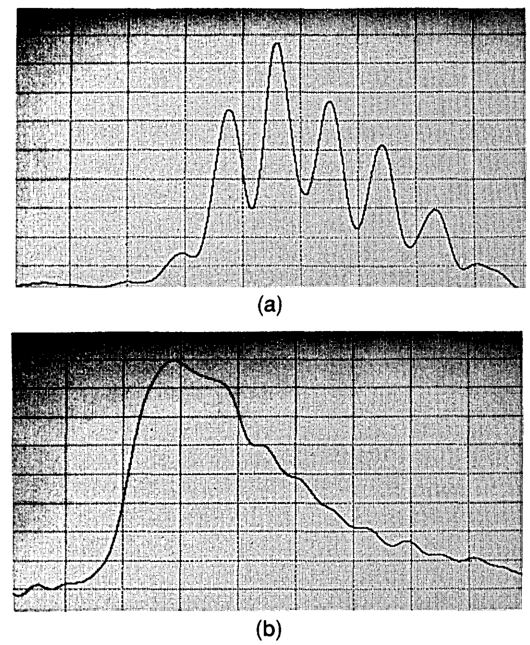

Fig. 2. (a) Temporal pulse from the MPL oscillator showing double-longitudinal-mode oscillation at $\Delta\nu \approx 1.12$ GHz, (b) temporal pulse from the HMPGI oscillator showing near single-longitundinal-mode oscillation. The full-width laser linewidth determined interferometrically for this case was $\Delta\nu \leq 500$ MHz. In both cases the time scale is 1 ns/div.

sides 10- and 7-mm thick. The two circular surfaces were optically polished to a surface quality better than $\lambda/4$. The grating used in these oscillators was a holographic grating with 3000 lines/mm and was 5 cm in length. In the case of the MPL oscillator the multiple-prism expander was composed of three quartz prisms deployed in a (+, +, −) compensating configuration.[8] The first two prisms each have a hypotenuse of 15 mm, and the third prism has a hypotenuse of 50 mm. The apex angle for all prisms is 42.7°. In the HMPGI oscillator, only the two small prisms are used and are deployed in a (+, −) compensating configuration. Multiple-prism beam expander design and characteristics are discussed in detail elsewhere.[8] The Glan–Thompson polarizer output coupler has an extinction ratio of 5×10^{-5}. In the case of the HMPGI cavity the tuning mirror is a broadband reflector.

The physical cavity lengths for the MPL and the HMPGI oscillators are ~10 and ~9 cm, respectively. Excitation was accomplished semilongitudinally, through the polarizer output coupler, at an angle of a few degrees relative to the optical axis of the oscillator. To limit optical damage of the dye in the solid matrix, the incident energy on the gain medium was limited to ≤1 mJ, which resulted in an incident energy density <1 J/cm^2. The beam waist at the gain region was ≈200 μm.

Measured results for the MPL and the HMPGI solid-state dye-laser oscillators are given in Table 1. The measured efficiencies were 9.3% and 4% for the MPL and the HMPGI oscillators, respectively. Laser linewidth values 1.12 GHz $< \Delta\nu <$ 1.20 GHz correspond to double-longitudinal-mode oscillation, as illustrated in Fig. 2(a). For reduced gain conditions the HMPGI oscillator yielded single-longitudinal-mode oscillation with temporal pulses, as shown in Fig. 2(b). Single-longitudinal-mode oscillation is inferred here because the temporal profile shows little or no modulation.[9] The linewidth of the single-longitudinal-mode oscillation was measured to be ≤500 MHz with a Fabry–Perot interferometer with a

Table 1. Performance of Solid-State Dispersive Dye Laser Oscillators

Cavity	Excitation	Gain Medium Matrix	$\Delta\nu$ (GHz)	$\Delta\theta$ (mrad)	Tuning Range (nm)	% Efficiency	C (mM)
MPL	Semilongitudinal	MPMMA	1.12	≈2.9	563–610	9.3	0.5
HMPGI	Semilongitudinal	MPMMA	1.20	≈2.6	565–603	4	0.5

FSR of 7.49 GHz. Measurements of sub-gigahertz linewidths (in the 300–500-MHz range) from liquid dye lasers emitting pulses in the 2–7-ns range have been reported by several authors[10–12] using Fabry–Perot interferometers with 2 GHz $\leq$ FSR $\leq$ 7.5 GHz. The cavity lengths of the MPL and the HMPGI oscillators used in the present experiments are more compact than those reported by Hansch[10] and Saikan[11] and are similar to the cavity length reported by Littman.[12] Measurement of laser linewidths from lasers emitting in the nanosecond regime is discussed in Ref. 13.

The tuning ranges for the MPL and the HMPGI oscillators were measured at 47- and 38 nm, respectively. Beam divergences were measured to be 2.9 mrad or less. For these oscillators the level of amplified spontaneous emission, as defined in Ref. 8, was measured to be $\approx 10^{-7}$.

Energy conversion efficiency for the MPMMA material was 35% for an incident energy density of ~ 0.7 J/cm^2, at a dye concentration of 0.5 mM, for broadband emission. This is very close to the efficiency observed when one is using a liquid solution in a cell providing the same gain length, at identical concentration. However, increasing the incident energy density to ≥ 1 J/cm increased the energy conversion efficiency to better than 40% in the case of the liquid solution. We achieved this increase by maintaining the pump-laser energy at a constant level while increasing the energy density by reducing the cross-sectional area of the incident beam. Because dye degradation was observed in the MPMMA materials at ~ 1 J/cm^2, the incident energy densities were limited to ~ 0.5 to 0.7 J/cm^2. At this incident energy density, no optical damage was noticed throughout the duration of these experiments. Test results provided by the supplier of the MPMMA material[7] indicate that damage thresholds are in excess of several Joules per centimeters squared.

Under the pumping regime described above, the MPL oscillator yielded better than 9% conversion efficiencies, and the HMPGI oscillator yielded approximately 4% conversion efficiencies. These figures should be compared with 14% and 7% conversion efficiencies achieved in MPL and HMPGI oscillators, respectively, using liquid solutions of Coumarin 500 (at $C \sim 10$ mM) under transverse N_2 laser excitation.[3,14] For high-pulse-repetition-frequency copper-vapor laser excitation, similar MPL and HMPGI oscillators[15] have yielded 5% and 4% conversion efficiencies, respectively, for single-longitudinal-mode oscillation at $\Delta\nu \approx 600$ MHz. In the latter experiments the excitation was transverse, and the dye used was Rhodamine 590, in the liquid phase, at $C \sim 2$ mM.

The results given here indicate that the performance of solid-state dispersive dye-laser oscillators with a MPMMA active medium offer conversion efficiency performance comparable to the well-established oscillators with liquid gain media.

A characteristic of emission from the oscillators with MPMMA is the good beam quality, which approaches TEM_{00} characteristics. In the present experiments the measured beam waist was 200 µm. For a beam waist of 200 µm the Fresnel number is 0.52 for the cavity length used here, at 575 nm. Using the interference equation given in Ref. 8 we can show that these conditions yield a single transverse mode. Note that for the present beam-waist characteristics, the diffraction-limited divergence is 0.92 mrad. The measured beam divergences for the solid-state MPL and the HMPGI oscillators are given in Table 1. By comparison the measured $\Delta\theta$ for the MPL oscillator using a conventional liquid gain medium was ≈ 1.4 mrad.

Using the method described by Duarte,[3,8] we obtain the double-pass dispersive linewidth:

$$\Delta\lambda = \Delta\theta\left[M\left(\frac{\partial\theta}{\partial\lambda}\right)_G + \left(\frac{\partial\Phi}{\partial\lambda}\right)_P\right]^{-1}, \qquad (1)$$

where $\Delta\theta$ is the beam divergence, $(\partial\theta/\partial\lambda)_G$ is the grating dispersion, and $(\partial\Phi/\partial\lambda)_P$ is the prismatic dispersion. In this case $(\partial\theta/\partial\lambda)_G \gg (\partial\Phi/\partial\lambda)_P$, and $M \approx 63$ and $M \approx 23$ for the MPL and the HMPGI configurations, respectively. Using this approach, and the $\Delta\theta$ values given in Table 1, we estimate the calculated double-pass dispersive linewidths to be 7.01 and 3.14 GHz for the MPL and HMPGI configurations, respectively. Measured linewidths for both oscillator configurations were in the 1.12–1.2-GHz range, which corresponds to double-longitudinal-mode oscillation [see Fig. 2(a)]. It is well known that this double-pass analysis yields only an upper limit for the linewidth value[3,16] and that a refined value can be estimated with a multipass approach.[17] In the present case, however, the larger-than-usual beam divergence is a contributing factor to the difference. In this regard the MPL oscillator with a liquid gain medium (Rhodamine 6G in ethanol at $C = 0.5$ mM) yields $\Delta\theta \leq 1.4$ mrad. For this $\Delta\theta$ the double-pass dispersive linewidth estimated with Eq. (1) is 3.4 GHz.

The larger beam divergence observed with the dye-doped MPMMA is probably the result of lensing induced at the active region. However, what is important here is that the transverse mode structure is TEM_{00}, and that enables us to achieve effective control of extracavity beam propagation with standard optics.

Notice that for a cavity length of 10 cm the corresponding inter-longitudinal-mode spacing ($\Delta\nu = c/2L$) is 1.5 GHz. Incorporating the refractive index of the prisms, the Glan–Thompson polarizer output coupler, and the gain medium yields an optical cavity length of 13.4 cm, which implies that $\Delta\nu \approx 1.12$ GHz, in agreement with temporal measurements. In the case of the HMPGI oscillator, lasing was restricted to a single longitudinal mode [see Fig. 2(b)], with $\Delta\nu$ in the subgigahertz range ($\Delta\nu \leq 500$ MHz) under reduced gain conditions.

In a direct comparison between the dispersive oscillator performances with MPMMA and ORMOSIL, it appears that, at this stage, better efficiencies and beam quality can be achieved with MPMMA-type materials. The main difference between silicate matrices and MPMMA-type matrices appears to be the superior optical homogeneity of the latter. Certainly the issue of optical homogeneity is very important to beam quality. The difference between MPMMA and present-day silicate matrices is due to the internal refractive-index inhomogeneities of the latter. These inhomogeneities can cause spatial variations in the 10–200-μm range. Consequently these spatial variations induce interference that leads to beam spatial inhomogeneities. This has been measured and quantified with the interference approach discussed by Duarte[18] and will be described in detail elsewhere. The origin of these inhomogeneities, or turbulence, is not yet well understood. However, they may be associated with the gelation process.[19]

Recently,[20] high conversion efficiencies ($>52\%$) have been achieved for broadband emission with pyrromethene dyes in hydroxypropyl acrylate–methyl methacrylate under pulsed laser exictation. This suggests good prospects for the achievement of higher pulse energies with amplification stages. Also, Pacheco *et al.*[21] have demonstrated pulse energy of 50 mJ, in a 475-ns pulse, with Coumarin 540 in PMMA under flash-lamp excitation. These figures indicate that solid-state dye gain media, such as dye-doped MPMMA, have become an attractive alternative as gain media for tunable dye lasers. Additional research on dye-doped ORMOSIL's and transparent silica gel–polymer nanocomposites[22] should open further alternatives. Indeed we have recently achieved broadband lasing with Rhodamine-doped silica gel–polymer nanocomposites.[23]

In summary, widely tunable narrow-linewidth emission ($\Delta\nu \approx 1.12$ GHz) has been demonstrated with solid-state dispersive dye-laser oscillators incorporating a Glan–Thompson polarizer output coupler. The output characteristics of these oscillators make them quite attractive for spectroscopic applications. The compact high-performance oscillators described here could be easily excited by diode-pumped solid-state lasers, thus producing compact all-solid-state dye-laser systems.

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