High order operation of distributed feedback dye-doped sol-gel silica laser

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High order Bragg scattering ($M = 2$ and $M = 3$) operations of the distributed feedback dye doped sol-gel silica lasers are achieved using the second harmonic output of a Nd:YAG laser as the pump. Laser linewidth of less than 0.06 nm and conversion efficiency of 0.7% for $M = 3$ and 11% for $M = 2$ are measured. Wavelength tuning of about 15 nm around the emission center has been realized by varying the intersection angle.

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Solid state dye laser has been an area of considerable research interest in recent years. Previous works on solid state dye laser have focussed on broadband laser emission. More recently, narrow linewidth/single longitudinal mode laser action has been achieved in solid state dye lasers using different host materials[1−3]. Distributed feedback dye lasers (DFDL) generate short pulses with narrow linewidth and thus become attractive compact coherent light sources. The output wavelength of the DFDL follows the Bragg scattering condition of $\lambda_L = 2\eta\Lambda/M$,where η is the refractive index of the gain medium at λ_L, Λ is the period of the gain modulation structure (the fringe pattern) and M is the Bragg scattering order. For a DFDL pumped by another laser with wavelength λ_P, the modulation period Λ is given as $\Lambda = \lambda_P/(2\sin(\theta))$, where θ is half of the pump beam intersection angle at the gain medium. In other word, $\lambda_L = \eta\lambda_P/(M\sin(\theta))$. Tuning of the λ_L can be achieved by varying the intersection angle. As a beamsplitter, the use of a holographic grating considerably reduced the coherence requirement on the pump laser, making possible of broadband excimer lasers, dye lasers as effective pump sources. Several groups have investigated the lasing properties of dye-doped polymer with distributed feedback[4,5]. Our previous works have developed distributed feedback lasers emitted in the red, blue and near UV spectral ranges using dye-doped sol-gel silica slab as the gain medium[6,7].

Theoretically, high order ($M \geq 2$) distributed feedback (DFB) laser is capable of selecting the optimal pump wavelength according to the Bragg scattering condition, it makes possible of obtaining DFB lasing from lots of laser dyes where first-order Bragg scattering condition can not be satisfied. On the other hand, since the modulation period is a multiple of one-half of the oscillation wavelength for high order operation (instead of one-half wavelength for $M = 1$), the resolution required for producing the periodic gain modulation structure on the gain medium is relaxed. High order operation is thus of interest, especially in the visible and near UV regions. Since the initial demonstration of DFB dye lasers, high order operation of DFB laser was considered to be difficult because of weak Bragg scattering coupling[8]. In this paper, we report the achievements of high order operation of the DFB dye-doped sol-gel silica lasers.

In the experiments, only R6G dye was used as the laser active dopant in all sol-gel silica samples used. The samples were fabricated following the low temperature (less than 100 °C) sol-gel techniques with tetraethoxysilane (TEOS) as the silica precursor. The initial solutions were typically composed of 8 ml TEOS, 7 ml ethanol, 5 ml water, with 0.8 ml HCl as the catalyst. R6G was added into the solutions till the desired concentration reached. The mixture was filled into acrylic cuvetters and maintained in a thermostat at 60 °C for two weeks. The final dimensions of the sol-gel slabs were 4.5(L)×2(W)×10(H) mm^3.

The optical arrangement in the DFB dye-doped sol-gel silica laser experiments followed closely that of Bor[9], see Fig. 1. A holographic grating of 1800 lines/mm served as a beamsplitter allows lasers of low spatial coherence to be used as the pump source. In this regard, an unseeded Nd:YAG laser (Continuum Surelite II) operating at the second harmonic was used as the pump source in this paper. Incident on the grating, the pump beam from the pump laser was diffracted into ±1 orders of approximately equal intensities. The diffracted beams were redirected to combine at the sol-gel slab at an intersection angle of 2θ. The Bragg diffraction order was determined from the intersection angle and the wavelength of the pump laser. The surface plane of the sol-gel slab roughly corresponded to the focal plane of the focussing cylindrical lens. The typical focussed beam size at the sol-gel silica slab was 5×0.4 mm^2. The sol-gel silica slab was tilted at a small angle to prevent parasitic feedback.

Our first attempt was to achieve DFB R6G dye-doped sol-gel silica laser action for Bragg scattering order of $M = 3$ pumped by 532 nm wavelength. Using a slab sample with 2×10^{-4} M/l initial concentration, as the pump energy increased more than 100 μJ, wavelength tuning in yellow optical region was realized in the third-order ($M = 3$) by varying the pump beams intersection angle, more than 15 nm wavelength tuning range around the emission center was obtained. The experimental wavelength tuning data of $M = 2$ and $M = 3$ were compared with the theoretical tuning curves in Fig. 2. In the figure, the dots represent the measured results, and the solid lines are the theoretical prediction. The refractive index for our sol-gel silica samples was determined separately by the Brewster angle method. The wavelength tuning

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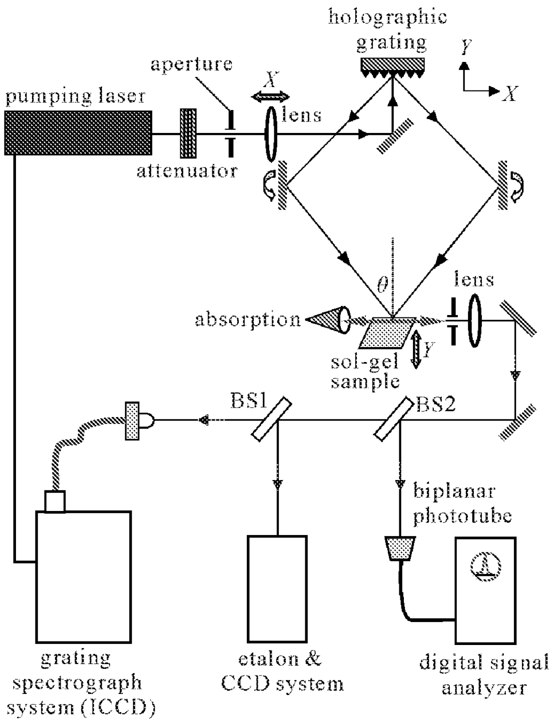

Fig. 1. The experimental setup of dye-doped sol-gel silica distributed feedback laser.

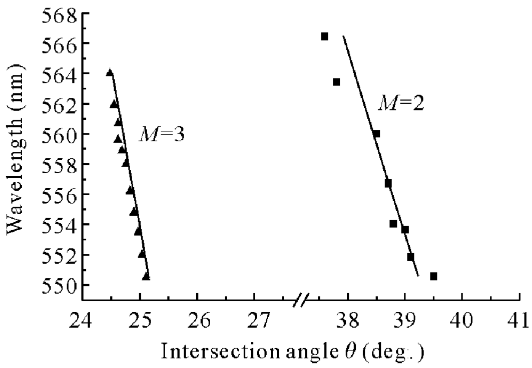

Fig. 2. Laser output wavelength λ_L as a function of θ for R6G doped sol-gel silica DFB laser, pumping source wavelength was 532 nm. The points are experimental data, solid lines are the theoretical curves (triangle for the third-order, square for the second-order).

rate $d\lambda_L/d\theta$ of a DFB solid state dye laser increased with the Bragg scattering order M. This was also verified when the third harmonic output of the YAG laser was used as the pump source to generate first-, second-, and third-order Bragg scattering operations of DFB laser in the same arrangement. The results showed that the wavelength tuning range decreased when M increased; and the threshold pump energy increased significantly with M increased. Meanwhile, due to the seriously photo-degradation of the R6G dye molecules caused by intense UV excitation, the operation lifetime of the third order DFB laser action was shorter and the wavelength tuning range was narrower.

The laser emission spectral linewidth of the third order

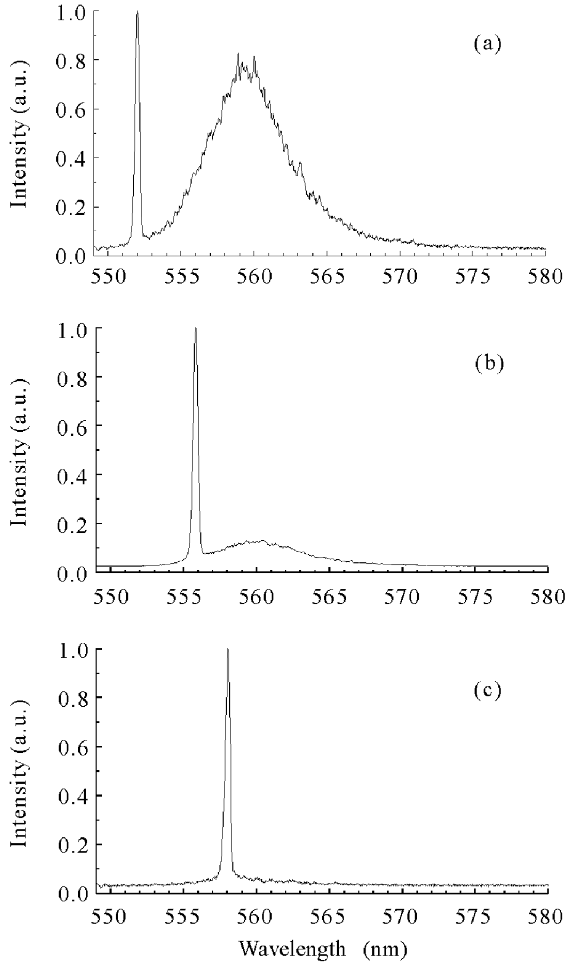

Fig. 3. $M = 3$ order DFB lasing spectrum detected by the grating spectrograph/array detector system.

($M = 3$) DFB laser was analyzed by a Burleigh WA 4500 Wavemeter fitted with a Fizeau and a Fabry-Perot etalons for coarse and fine spectral measurements. The third order DFB laser output typically has an emission spectral linewidth of the order of 0.06 nm over the entire tuning range, the same as that obtained in the second order DFB sol-gel silica laser. The narrowest linewidth of 0.045 nm was measured while pumped near the threshold. The results showed that the emission spectral linewidth of the solid state DFB dye laser was independent of the Bragg scattering order and was primarily governed by the holographic grating and the pump beams quality. The emission linewidth broadened with increased pump energy, more than 0.1 nm linewidth output was also observed as the pump energy increased significantly. The relationship of the emission linewidth and the pump energy was in good agreement with the prediction of coupled-wave theory. For the third-order output, the amplified spontaneous emission (ASE) background became comparable to that of the DFB dye laser intensity because of weak Bragg scattering feedback. Figure 3 shows the emission spectra of the third order DFB R6G doped sol-gel silica laser output recorded by a spectrograph/array detector system. The ASE intensity could be as strong as the laser when the DFB laser was tuned

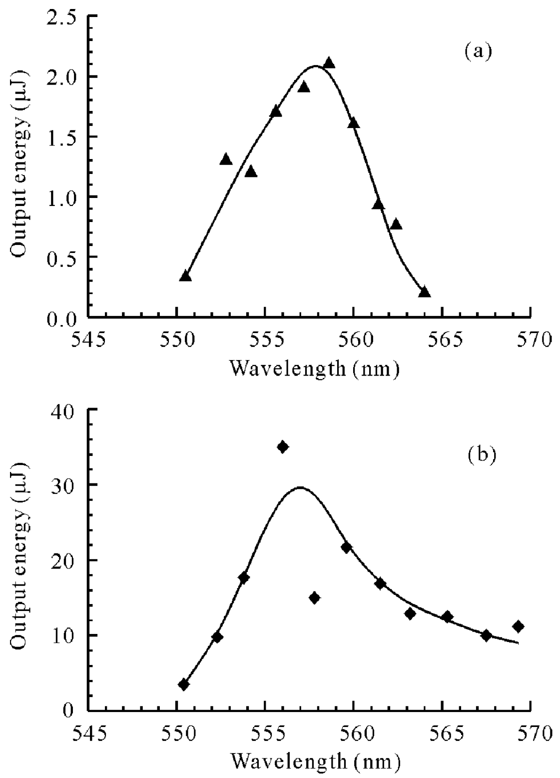

Fig. 4. Laser output energy as a function of the emission wavelength. Pumping energy 310 μJ, pumping wavelength 532 nm. (a) $M = 3$, (b) $M = 2$.

to the side wings of the gain profile (Fig. 3(a)). The laser intensity increased quickly as the lasing wavelength was turned near to the center of the gain profile (Fig. 3(b)). Once the output wavelength was centered at the gain profile, the ASE was ignorable compared with the laser pulse output (Fig. 3(c)).

For a typical pump energy of 310 μJ at 532 nm wavelength, the maximum conversion efficiency of the third order ($M = 3$) DFB laser was 0.7%, but was approximately 11% for $M = 2$ at the center emission wavelength. Figure 4(a) displays the experimentally measured laser output energy as a function of the output wavelength from the third-order DFB R6G dye doped sol-gel silica laser pumped at 310 μJ. Figure 4 (b) was obtained for the case of $M = 2$. When we raised the pump energy to 710 μJ, the conversion efficiency of $M = 3$ DFB laser increased to 1.6%. Since the spurious feedback was easily induced in this case due to high pumping energy input, the sample slab must be tilted at a sizable angle with respect to the optical axis. The threshold pump energy required for the high order action ($M = 2$ and $M = 3$) was also measured. For $M = 2$ and $M = 3$ DFB solid state dye laser at an initial concentration of 2×10^{-4} Molar, the threshold pump energy was typically 75 and 100 μJ, respectively. The ratio of the threshold pump energy for second and third orders was 3:4. This value was roughly in agreement with the theoretical prediction obtained from the coupled-wave model based on the coupling constant κ_M, of about 7:12.

Under a pumping energy level of 310 μJ (about 3 times over the threshold energy) at a repetition rate of 1 Hz, more than 6000 shots were obtained before the DFB laser output energy drop to 50% of the initial output energy value.

As conclusions, higher-order Bragg scattering operation of DFB dye-doped sol-gel silica lasers have been demonstrated. Narrow linewidth of the order of 0.06 nm has been obtained for the $M = 2$ and $M = 3$ cases. Wavelength tuning of 15 nm around the center wavelength has been achieved.

For higher-order operation, the optimal pump wavelength can be chosen experimentally corresponding to the laser dye used, the energy conversion efficiency can still keep considerably high, and the operation lifetime can also be improved due to reducing the possibility of photo-degradation of the dye molecules.

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