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Single longitudinal mode lasing of coumarin-doped sol-gel silica laser

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Abstract. Homogeneity of sol-gel-derived silica was improved by slowing down the drying and aging rate. Single longitudinal mode lasing was achieved in sol-gel silica doped with coumarin 460 dye. The laser linewidth was 1.58 GHz and the output wavelength was at 467 nm.

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Tunable lasers in the visible to ultraviolet are of considerable interest for applications ranging from dermatology to molecular spectroscopy. In the past, the need for tunable lasers was filled by liquid-dye lasers. Recently, increased use has been made of solid-state materials as tunable laser sources. The combined use of photostable laser dyes and improved polymeric materials has made solid-state dye lasers a practical alternative to liquid-dye lasers [1, 2]. Inorganic solid matrices derived by sol-gel techniques are also attractive solid-state dye laser materials. Laser lifetimes comparable to those of polymer-dye lasers were achieved in dye-doped sol-gel materials [3]. Direct generation of UV laser output with an output wavelength as short as 342 nm was demonstrated in sol-gel silica slabs doped with UV laser dyes [4, 5].

Narrow linewidth operation is one of the important performance requirements of tunable lasers. Single longitudinal mode lasing with a linewidth of about 1.12 GHz was demonstrated for a modified poly(methyl methacrylate) (MPMMA) slab doped with rhodamine 590 (R590) dye [7]. For narrow linewidths, microscopic spatial homogeneity exemplified by the absence of refractive-index variation in the solid-state dye laser materials was considered most critical [7]. While narrow linewidth operation was achieved in high-performance dye-doped polymer, there was doubt as to the optical homogeneity of sol-gel materials, which may impair the laser spectral output performance [7–9]. Duarte et al. reported a laser linewidth of 3 GHz and output energy less than 1 mJ from R590-doped sol-gel materials in a multiple prism cavity pumped by a 160-ns liquid dye laser [10]. We recently reported narrow linewidth laser operation for three laser dyes, rhodamine 6G (R6G), coumarin 460 (C460), and exalite 377 (E377), in sol-gel silica, representing lasing in the yellow, in the blue, and in the near UV. A spectral linewidth of about 4.3 GHz (FWHM) was observed for C460-doped sol-gel silica [11].

In the past, most of the research into solid-state dye lasers has been focused on dyes in the yellow to red spectral range because of the compatibility of the perylene and pyrromethene dyes with solid-state hosts [1, 2]. Recently some published work appeared on solid-state dye lasers using coumarin dyes that emit in the technologically important blue spectral range [8, 9, 12]. Most of the laser experiments on coumarin-based solid-state dye lasers were conducted to study broadband laser emissions. In this work, we report single longitudinal mode lasing of C460-doped sol-gel silica with an output wavelength of 467 nm. The linewidth of the laser output was 1.58 GHz, which is a considerable improvement over our previous result of 4.3 GHz, obtained from a double grating resonator cavity [11].

The dye-doped silica samples were fabricated in house following the sol-gel process of acid catalyzed hydrolysis/polycondensation [4]. Attention must be paid to slow down the drying and aging rate to improve the optical quality of the samples. This we achieved by completely sealing the curvettes during gelling, drying and aging. Sol-gel silica samples of high optical homogeneity were obtained. The initial solutions were typically composed of 15 ml of tetraethoxysilane (TEOS), 11 ml of ethyl alcohol, 9 ml of water, 10 ml of formamide, and 1.5 ml of hydrochloric acid serving as catalyst. Dyes were added to the initial solutions under magnetic stirring until the desired molar concentrations were reached. For all laser experiments presented here, a molar concentration of 2×10^{-3} was used. The organic dyes and chemicals used in these experiments were procured from Exciton and Aldrich, respectively. Maintained in a thermostat at 60 °C, the chemical mix forms a gel in hours. After aging and drying in sealed acrylic cuvettes for about two weeks, the silica gels solidify and can be readily removed for laser experiments. The silica samples typically measured 6 mm(w)×6 mm(h)×12 mm(l) and were visually of good surface finish with end faces appearing to be plane parallel. They showed excellent optical transmission deep into the UV (below 200 nm) [5]. Figure 1 shows the digitized photo-

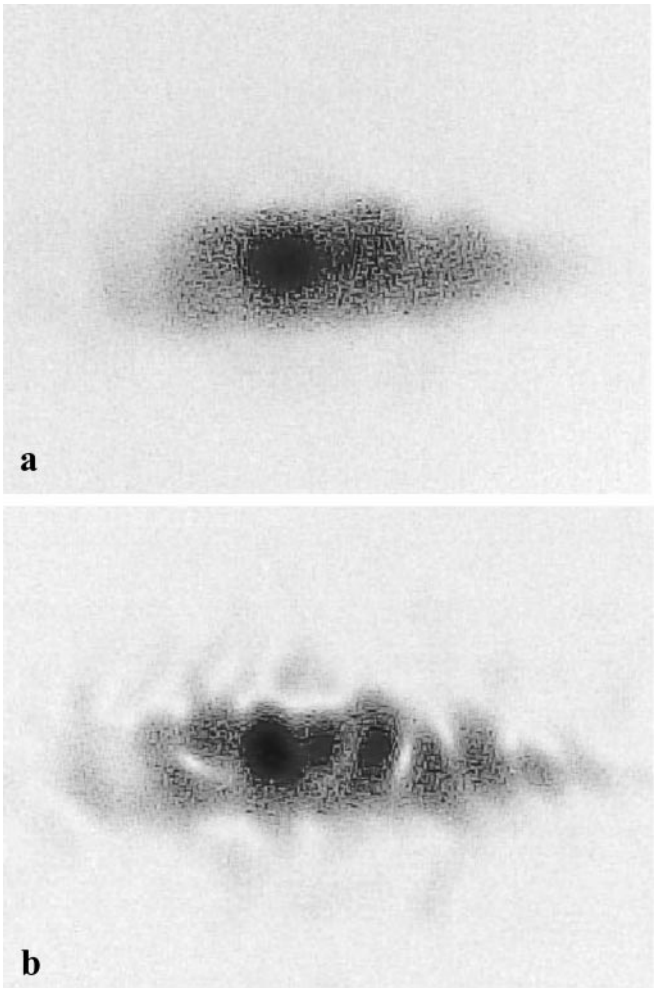

Fig. 1a,b. Transmitted beam profile of a He-Ne laser. **a** Sol-gel sample aged for 14 days. **b** Sol-gel sample aged for 3 days

graphic images of the beam profiles of a He-Ne laser after the beam traversed 6-mm-thick undoped sol-gel silica samples that dried and aged in 14 days (Fig. 1a) and in 3 days (Fig. 1b). The sample dried and aged for 14 days clearly has superior optical quality, judging from the transmitted beam profiles. Single longitudinal mode lasing was achieved in the slowly dried samples. By contrast, the narrowest linewidth that we could achieve in the samples that dried and aged in 3 days was about 3 GHz.

Single longitudinal mode lasing was demonstrated by incorporating an etalon inside a Littrow grating resonator cavity. The intra-cavity etalon was used because of the simplicity in aligning and setting up the narrow linewidth cavity. A more compact cavity arrangement such as the multiple prisms setup [6] may also be used. The experimental layout is shown in Fig. 2. The Littrow grating (Richardson Gratings) was a holographic grating with 2400 lines/mm. It has a high modulation depth for the grooves to have a maximum efficiency of 85% near 400 nm. The etalon (CVI Corp.) has a free spectral range (FSR) of 102 GHz, a finesse of 28.3, and a reflectivity of 90% at 470 nm. Single longitudinal mode lasing was not possible when the etalon reflectivity was lowered to 40%. A flat mirror of 10% reflectivity at 480 nm was used as the output coupler. The cavity length was 6.5 cm, and the mode separation between longitudinal modes was 2.3 GHz. Selected spherical and cylindrical lenses were employed for collimation and focusing purposes.

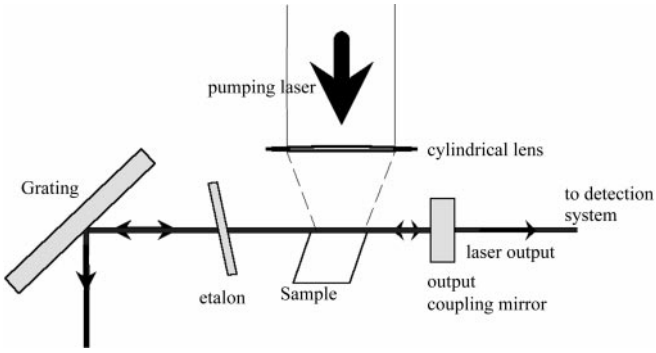

Fig. 2. Experimental layout of the single longitudinal mode laser experiments

The pump source was a lamp-pumped Nd:YAG laser (Continuum Surelite II). The third harmonic output at 355 nm was used to excite the dye-doped samples. The pump energy was typically 1–2 mJ per pulse. The pump laser pulses were about 3.5 ns. The spectral output from the sol-gel lasers was detected and analyzed by a home-built etalon/CCD system. The fixed-air-gap etalon of the home-built system has a FSR of 30 GHz and a finesse of 20. The spectral resolution of the etalon/CCD system was determined by a cw He-Ne laser to be 1.5 GHz. Output wavelengths of the sol-gel lasers can be monitored by a 0.3-m-focal-length spectrograph/array detector system. The laser emission spectrum from the coumarin-doped sample is shown in Fig. 3. The spectral resolution of the spectrograph/array detector system is about 0.8 nm. A fast sub-nanosecond phototube (Hamamatsu R1193-U) in combination with a wide-bandwidth transient digitizer was used to measure the waveforms of the sol-gel laser pulses.

Narrow linewidth lasing was readily achieved for both liquid dye and dye-doped sol-gel silica by using the Littrow grating cavity incorporated with the etalon. The FWHH linewidth for liquid-dye laser was 1.52 GHz (Fig. 4a) and that of the sol-gel silica was 1.58 GHz (Fig. 4b). The lasers were operating at a single longitudinal mode as the mode separation of the resonator cavity was 2.3 GHz. Figure 5 shows the single-shot synchronized waveforms of the 355-nm pump laser pulse and the 467-nm sol-gel laser pulse. The two pulses

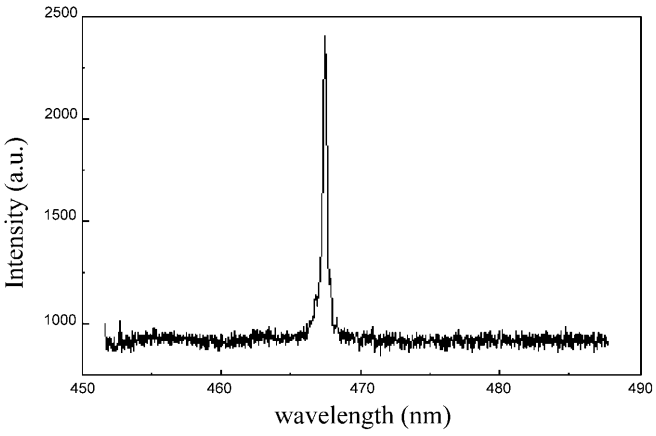

Fig. 3. Laser emission spectrum from an C460-doped sample detected by the array detector/grating spectrograph system

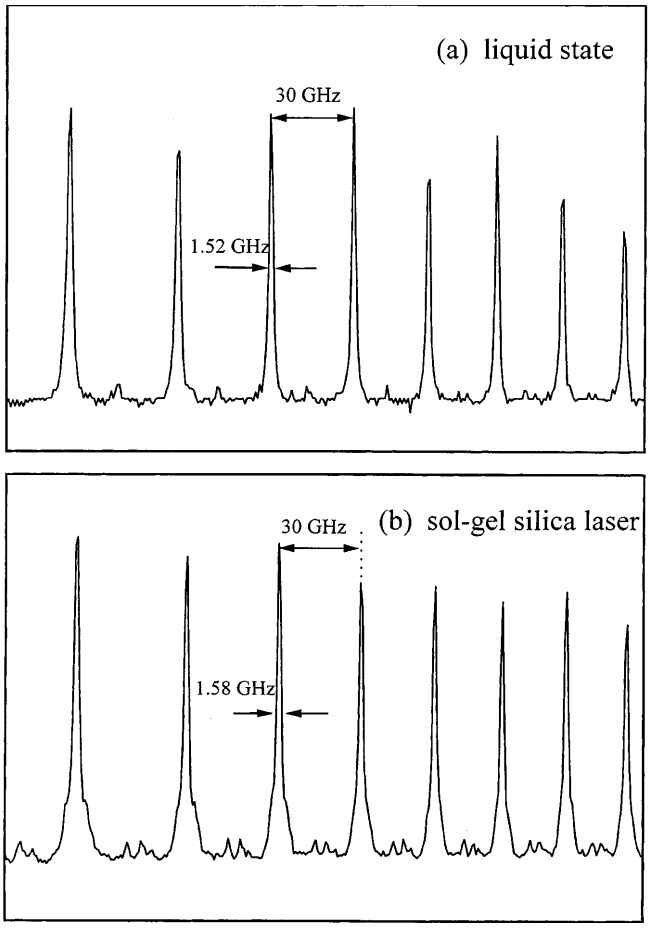

Fig. 4a,b. Linewidth analysis of the sol-gel dye laser output as recorded by the CCD after transmitting the etalon. **a** C460 in ethanol. **b** C460-doped sol-gel silica

reach their maxima simultaneously. But the duration of the sol-gel laser pulse at 1 ns is narrower. The oscillatory waveform indicative of multi-mode lasing observed in our previous double-grating cavity experiments [11] is absent. The lack of modulation in the laser waveform confirms single longitudinal mode lasing [6]. The laser output at various pump energies was also measured (Fig. 6). The threshold pump energy was 120 μJ and the slope efficiency was about 3%. Such low lasing efficiency is a result of the high reflectivity of the intra-cavity etalon. An optimized resonator cavity should be able to improve the slope efficiency considerably.

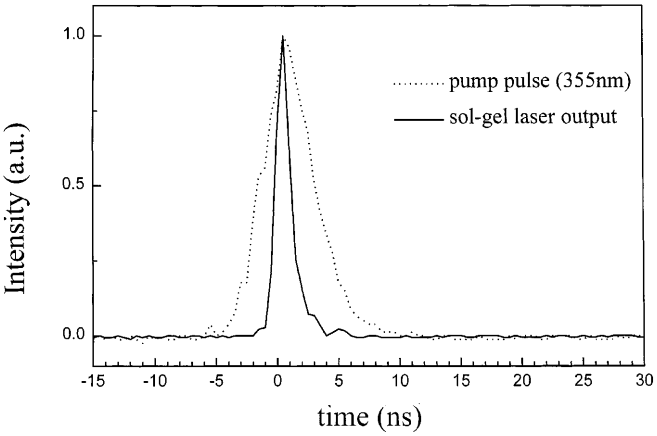

Fig. 5. Laser waveforms of 355-nm pump laser and 467-nm sol-gel laser

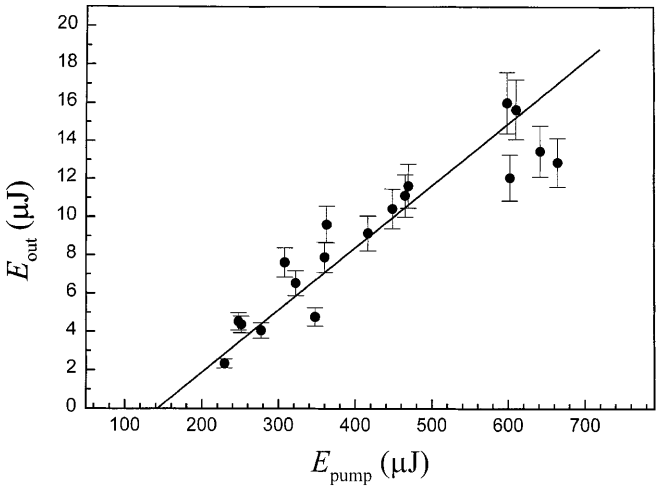

Fig. 6. Sol-gel laser output energy at 467 nm as a function of pump laser energy

In conclusion, we fabricated dye-doped sol-gel silica slabs of high optical homogeneity. Single longitudinal mode lasing of dye-doped sol-gel silica lasers was demonstrated at 467 nm by using an intra-cavity etalon in a Littrow grating resonator cavity. A slope efficiency of 3% was observed for the sol-gel laser.

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References

1. R.E. Hermes, T.H. Allik, S. Chandra, J.A. Hutchison: Appl. Phys. Lett. **63**, 877 (1993)
2. R. Sastre, A. Costela: Adv. Mater. **7**, 198 (1995)
3. M. Canva, P. Georges, J.F. Perelgritz, A. Brun, F. Chaput, J.P. Boilot: Appl. Opt. **34**, 428 (1995)
4. C. Ye, K.S. Lam, K.P. Chik, D. Lo, K.H. Wong: Appl. Phys. Lett. **69**, 3800 (1996)
5. K.S. Lam, D. Lo: Appl. Phys. B **66**, 427 (1998)
6. F.J. Duarte: Appl. Opt. **33**, 3857 (1994)
7. F.J. Duarte, A. Costela, I. Garcia-Moreno, R. Sastre, J.J. Ehrlich, T.S. Taylor: Opt. Quantum Electron. **29**, 461 (1997)
8. A. Costela, I. Garcia-Moreno, J. Barroso, R. Sastre: J. Appl. Phys. **83**, 650 (1998)
9. A. Costela, I. Garcia-Moreno, J. Barroso, R. Sastre: Appl. Phys. B **67**, 167 (1998)
10. F.J. Duarte, J.J. Ehrlich, W.E. Davenport, T.S. Taylor, J.C. McDonald: "A New Tunable Dye Laser Oscillator: A Preliminary Report" in Proceedings of The International Conference on Lasers '92, ed. by C.P. Wang (STS, McLean, VA 1993) pp. 293–296
11. D. Lo, S.K. Lam, C. Ye, K.S. Lam: Opt. Commun. **156**, 316 (1998)
12. A. Weissbeck, H. Langhoff, A. Beck: Appl. Phys. B **61**, 253 (1995)