Efficient tunable solid state laser around 630 nm using sulforhodamine 640 doped silica-gel

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Efficient tunable solid-state laser near 630 nm using sulforhodamine 640-doped silica gel

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Lasing is observed near 630 nm in a sulforhodamine 640-doped silica gel pumped by frequency-doubled radiation from a Q-switched Nd:YAG laser. A conversion efficiency of 20% is obtained. The laser wavelength is tunable over 40 nm.

Considerable effort has recently been directed toward obtaining new laser media for tunable solid-state lasers. Rare-earth ions provide a set of discrete laser wavelengths in the near infrared. Generally these lasers are not tunable but can be used as pump sources for optical parametric oscillators and color-center lasers.[1,2] High-efficiency lasing has been demonstrated in transition-metal media.[3,4] However, crystal growth requires specialized equipment, and production costs remain relatively high.

Recent developments in solgel techniques offer promising prospects for introducing dyes into inorganic matrices. Solgel methods enable single- or multiple-component glasses to be prepared at temperatures much lower than the fusion temperature. The classic process of preparing SiO_2 glass involves hydrolysis and polycondensation of alkoholates, e.g., tetramethoxysilane [$Si(OCH_3)_4$; TMOS] or tetraethoxysilane [$Si(OC_2H_5)_4$; TEOS]. This process leads to a gel that can be dried and sintered to solid glass. It is possible to incorporate an organic dye into the starting mixture before it gels. The resulting doped gel can be dried at a low temperature (60°C) to produce an amorphous material (xerogel) with extremely fine pores (nanometer size) that do not impair the optical properties of the material. The incorporated dye is uniformly embedded in the resulting inorganic host matrix. The inclusion of various dyes by the solgel method has already been described, and the fluorescence properties of some materials have been evaluated.[5–8] The resulting photostability of the dye is enhanced in the inorganic host matrix and surpasses that of organic media such as polymethylmethacrylates. Recently low-efficiency laser action was reported in transparent alumina–gel films doped with a laser dye.[9] In this Letter high-efficiency tunable laser action is demonstrated in a silica gel doped with sulforhodamine 640.

The starting solution for the silica gel was prepared classically[10] by mixing together 15 cm^3 of TMOS, 12.5 cm^3 of methylalcohol, and 12.5 cm^3 of formamide (used as a drying control chemical additive with 2 cm^3

of concentrated HNO_2). After magnetic stirring at 25°C, 18.3 cm^3 of water was added, and stirring was continued for 10 min. The resulting [H_2O]/[TMOS] molar ratio was 10. Finally, 10 mg of sulforhodamine 640 was added, and the stirring was continued for another 5 min. The resulting mixture was poured into polystyrene tubes that were then hermetically sealed. Geling took place after 1 h at 60°C. The gel was then aged at 60°C for 24 h in the sealed tubes, then dried at 60°C for 4 days in open tubes. The doping level in the resulting dried sample was $C = 1.72 \times 10^{-3}$ (grams of sulforhodamine 640/grams of SiO_2). The apparent density $\rho_a = 1.38$ g/cm^3 of the dry gel indicates that $c = 3.9 \times 10^{-3}$ M/liter.

The samples were cut with a diamond saw in 5-mm-thick disks and were polished on both faces.

The absorption spectrum of our sample shows a broad absorption band extending from 400 to 600 nm. Figure 1 shows the edge of the absorption band. The absorption is too large at less than 580 nm to be accurately measured. Nevertheless the absorption curve shows that all classical pulsed lasers (doubled
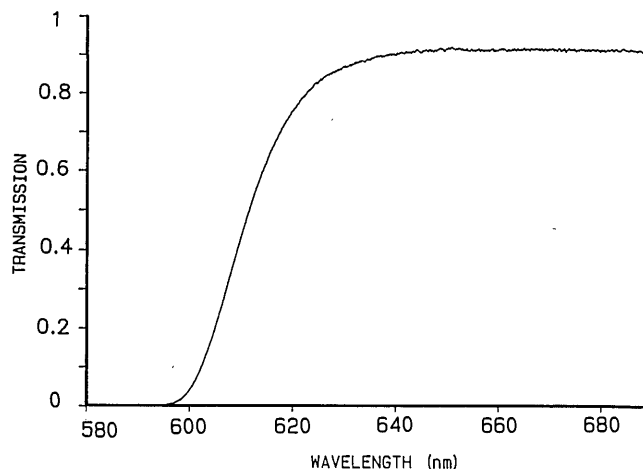

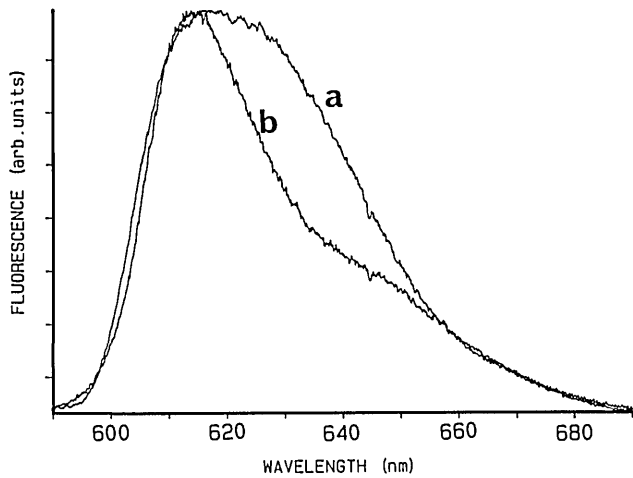Fig. 1. Transmission spectrum of the sample.

Fig. 2. Fluorescence spectra of (curve a) the sulforhoda-mine 640-doped glass and (curve b) the methanol solution of this dye.
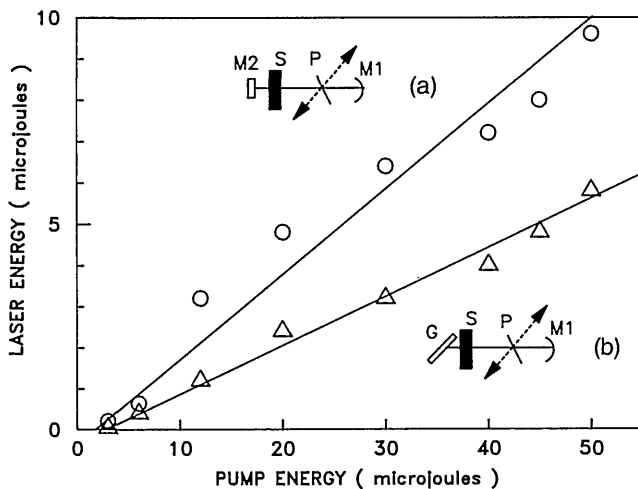

Fig. 3. Output pulse energy as a function of the pump pulse energy at 532 nm for (a) the two-mirror cavity ($\lambda = 621$ nm) and (b) the tunable cavity ($\lambda = 620$ nm). The dye-laser energy is obtained by summing the energy of the two output beams.

glass plate (P) used as a beam splitter inside the cavity yields two outputs of equal power. The uncoated sample (S) is placed near the planar mirror. The pump radiation is provided by a 6-nsec Nd:YAG laser that is frequency doubled. The Nd:YAG laser repetition rates can be varied from less than 1 Hz to up to 50 Hz. The pump pulse is focused on the sample with a 200-mm focal-length lens to produce a 100-μm beam waist. Owing to the high concentration of our sample, nearly all the pump energy is absorbed in the first millimeter. Nevertheless laser action at 621 nm is observed even for low pump powers. The threshold energy is approximately 1 μJ at 532 nm. The variation of the output energy at 621 nm versus the incident pump energy is given by the circles in Fig. 3. Even for pump energies equal to 50 times the threshold, no gain saturation is observed. The slope efficiency is approximately 20%. This value can be increased by using a more optimized cavity with a higher output-coupling value. The output pulse shape exhibits a smooth profile (FWHM of 2 nsec). Owing to the good optical quality of the sample we are able to obtain high-order transverse modes by a simple rotation of the spherical mirror (Fig. 4).

We also investigated the tuning properties of this laser. For this experiment the plane mirror is replaced by a 600-groove/mm grating [Fig. 3(b)]. This grating (G) is used at the Littrow incidence for first-order operation. By rotating the grating we can tune the laser frequency. Figure 5 shows the tuning curve for a pump energy of 12 μJ. The laser can be tuned over 50 nm with a tuning curve slightly red shifted from that for the methanol solution of sulforhodamine 640. The output energy versus the pump energy at a fixed wavelength of 620 nm is given by the triangles in Fig. 3. The lower slope efficiency (10%) is attributed to the low diffraction efficiency (<80%) of the grating. The output pulse duration remains unchanged (~2 nsec).

All these measurements were made at a 10-Hz repetition rate. Even at this modest repetition rate we observed thermal effects in the sample. These effects
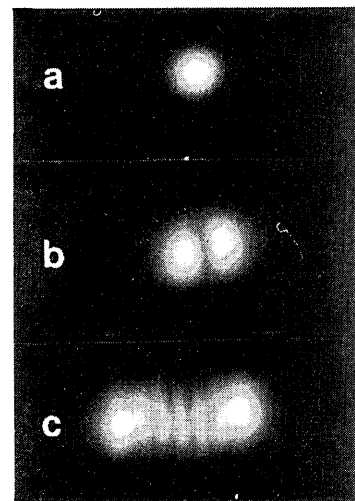
Nd:YAG, copper vapor, and even excimer and nitrogen lasers) could be used as pump sources for this sample. The fluorescence spectrum of the sulforhodamine 640-doped glass and a methanol solution of the same dye are presented in Fig. 2. In both cases the pump source is a 6-nsec pulse at 532 nm. The broadband fluorescence spectrum obtained is typical for dyes. We also observe a broadening of the fluorescence spectrum. The fluorescence decay was recorded with a fast photodiode and gives an excited-state lifetime of approximately 10 nsec. This value is approximately three times greater than that of sulforhodamine 640 dissolved in methanol. This longer lifetime can be attributed to the solid state of the sample, which reduces collisional effects.

Laser action was achieved in a two-mirror cavity [Fig. 3(a)]. Mirror M1 is a 100-mm radius-of-curvature high reflector centered at 610 nm. Mirror M2 is a planar mirror with high reflectivity. A 1-mm-thick

Fig. 4. Transverse modes of the dye-doped gel laser. a, TEM_{00}; b, TEM_{01}; c, TEM_{06}.
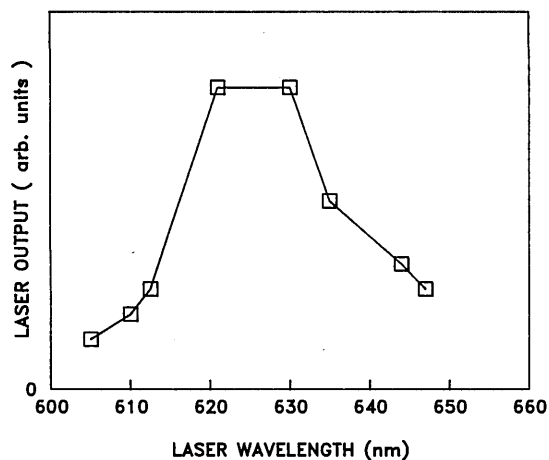
Fig. 5. Tuning curve of the sulforhodamine 640-doped gel laser at a 12-μm pump pulse energy.

are characterized by a slow decrease in the output power and a degradation of the spatial profile. We have verified that reversible photochemical effects are induced in the sample by stopping the pump radiation and waiting a few minutes. After this time the output pulse recovers its initial energy. We believe that these thermal effects are due to the high concentration of our sample.

In conclusion, we report the observation of lasing action in a solgel glass doped with sulforhodamine 640. This tunable solid-state laser may have important ap-plications in those fields that require tunable lasers with low output energies (<100 μJ). This laser has a low threshold energy and can be pumped by a small-frame laser, such as a frequency-doubled diode-pumped Nd:YAG laser. A monolithic tunable solid-state laser made with a single piece of doped gel with appropriate coatings on both faces in order to form the cavity is one feasible application. Because the number of dyes or other dopants (e.g., rare-earth ions) that can be included in solgel matrices is large, the outlook for this type of gel laser seems promising.

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