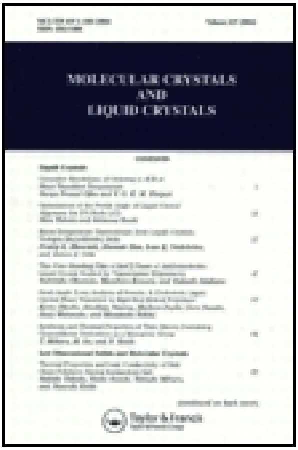

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Development of Integrated Tunable Laser System for Laser Spectroscopy

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DEVELOPMENT OF INTEGRATED TUNABLE LASER SYSTEM FOR LASER SPECTROSCOPY

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Laser systems on a chip have been developed for laser spectroscopic applications. The organic laser dyes were doped into polymer matrices and optical-waveguided laser cavities were fabricated using spin-coating and lithography techniques. Novel spectroscopic application was also demonstrated using laser array chip with multiple wavelength operation.

1. INTRODUCTION

Tunable lasers are one of the most important tool of laser spectroscopy. Recently many solid-state tunable devices, such as Ti:sapphire lasers and optical parametric oscillators, have been developed and their performance has been improved considerably as compared to the traditionally employed liquid dye lasers. However, still the whole system is complicated, bulky, expensive and maintenance-free operation is difficult. These problems are limiting the wide range industrial applications of laser spectroscopy.

As an alternative, we are studying about integrated tunable lasers, where laser material, tuning element, optical cavity and pumping source can be integrated on a planar waveguide. Though it is not easy to realize totally integrated tunable lasers presently, a very compact laser should be possible using several technique such as optical waveguide, photo-pumped laser and distributed feedback (DFB) technique. Since it makes a tunable laser as a robust device, better stability will also be expected. A DFB that is often

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employed in inorganic semiconductor lasers, was introduced on a optical waveguide made of an organic material in this work. Previously, the DFB laser action can be obtained by the interference of two pumping beams in the active medium. This dynamic DFB laser (termed holographic DFB laser) was demonstrated on dye lasers by different research groups [1–7].

We have also reported about holographically pumped DFB lasers with dynamic grating formed on dye-doped plastic waveguides [8]. Narrow-band tunable laser action was obtained at various wavelengths in the visible region by pumping with a frequency doubled or tripled Nd:YAG laser. Single-chip ultrashort pulse generation in combination with an integrated pulse compressor was also demonstrated [9,10]. In general, the solid-state plastic dye lasers have a limited durability problem, and some approaches were tried to extend its durability [11–13]. Our approach of waveguided DFB laser action demonstrates very low laser threshold, narrow spectral width and relatively better durability. The integration of DFB structure can also be used in realizing a mirror-less laser cavity on a plastic film that can be fabricated easily by a spin-coating and a subsequent photofabrication technique.

In this summary, an integrated laser system based on all-plastic DFB dye laser waveguide will be reported. The multi-wavelength laser waveguide array was introduced, and the evaluations of the developed dye laser waveguides are described [14,15]. Finally, the spectroscopic applications will be introduced [16].

2. CONCEPTS OF INTEGRATED TUNABLE LASER

Figure 1 shows a conceptual schematic of the integrated tunable laser system. The laser cavity is a light waveguide made of laser medium, and a tunable element, cavity mirrors and a frequency-doubling element are also integrated on the waveguide. The operation wavelength can be tuned electrically. Its fixed optical alignment can achieve good stability, easy alignment and maintenance-free operation because of no optical alignment. Actually, dye-doped polymer films spin-coated and then wet-etched are used as the optical laser waveguides in this work, and the DFB structure can be formed on the waveguides by fabricating periodical structure. The periodical structure is formed by exposing with interfered coherent ultraviolet (UV) beams, and the periodical coefficient Λ is given by

$$m\frac{\lambda}{2n_{\mathrm{eff}}} = \frac{\lambda_p}{2\sin\theta} = \Lambda,$$

where, θ is the incidence angle of the interfering UV beams, λ is the laser output wavelength corresponding to a Bragg wavelength, λ_P is

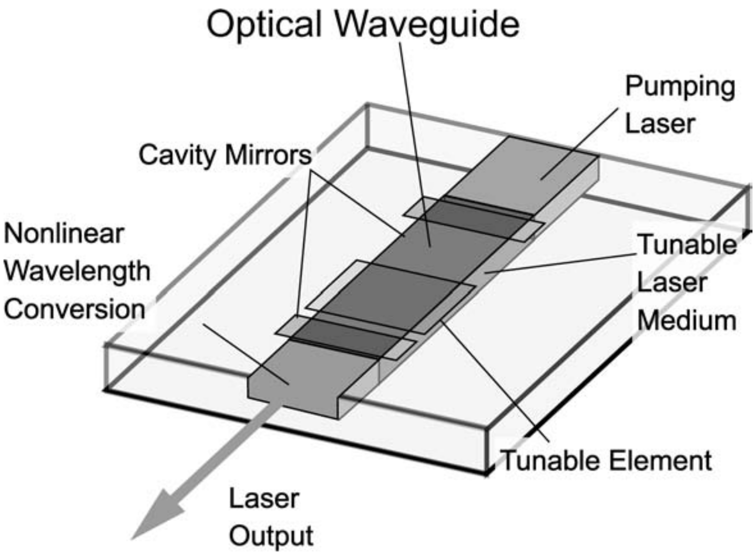

FIGURE 1 Conceptual schematic of an integrated tunable laser system on a chip.

the wavelength of the UV beams, m is an integer representing the order of Bragg reflection, and n_{eff} is the effective refractive index described below.

Our integrated dye laser has a permanent DFB structure based on periodical modulation of optical properties, such as a refractive index and/or a density of laser-gain molecules due to the UV-laser exposure. The DFB structures could be achieved even though any etching process didn't followed. After some investigations, these DFB structure was dominantly based on the refractive index modulation. The wavelength of the laser output was basically fixed because of the fixed grating pitch Λ, but it can tuned slightly (a couple of nanometers) by using a thermal control. We also demonstrated a wavelength modulation period of around 10 ms using an integrated thin-Al-heater, thus the thermal control can provide 100 Hz order modulation. In comparison with the holographic DFB laser pumped with 532 nm laser, the Bragg order of $m = 1$ ($m = 2$ for 532 nm) can decrease the laser threshold energy due to the high coupling coefficient, and it can lead the whole system compact and durable. No coherent pumping beam and no interference optical system were required, so pumping-optical system will be integrated on a same chip. Furthermore, an integrating many prefabricated DFB laser waveguides with wavelengths of $\lambda + n\Lambda\lambda (n = 0, 1, 2, \ldots)$ can achieve a digitally wavelength scanning and spatial-transformed spectroscopy as described below.

3. LASER CHIP FABRICATION

Figure 2 shows the fabricating process for the DFB laser array chip. At first, a dye-doped film was spin coated on a PMMA substrate using the pre-polymer method [17]. Depressured-distilled MMA monomer was mixed with laser dyes using a delivery such as an ethanol or a propylene carbonate (PC), then the mixture was radically polymerized by heating at a temperature of 60–75°C. An initiator of 2-2′ azobis-isobutyl-nitoryl was used to start the polymerizing. To attain single mode propagation waveguide, the refractive index modification was used in some cases, and the p(MMA:HEMA) copolymer was used in the other cases.

Before the complete polymerization, the MMA pre-polymer was spin-coated on the PMMA substrate and an active layer was formed. After spin-coating, the polymerization was completed by post heating at 75°, and planer laser waveguide was fabricated. Since the holographically pumped DFB laser operation can be obtained, the laser performance of the active medium was investigated in this step. The concentration of the laser dye can be calculated from the measured absorption coefficient and the thickness of the waveguides, and it must be as high as 10–30 mM to obtain laser operation with transverse pumping.

In the next step, multi-stripped waveguides structure was fabricated by using an ultraviolet (UV) lithography technique. A KrCl* excimer lamp (USHIO, UER20-222, 7 mW/cm^2) was used for the exposure and a mixture of ethylicetate (1) and isoimylicetate (9) was used as an etchant. Then the output side of the waveguides were polished. Typically, 100 μm wide and 20 mm long waveguides were used in the experiments.

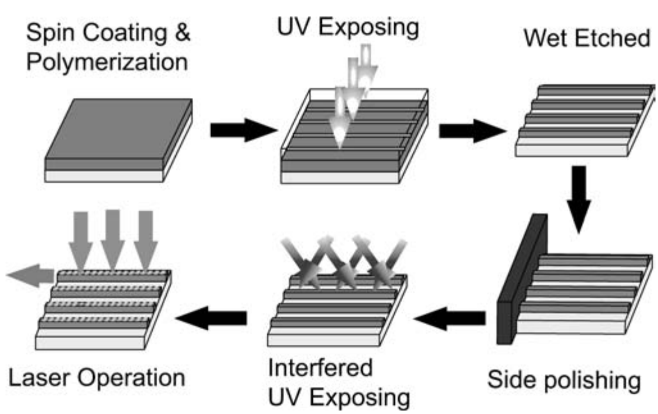

FIGURE 2 Fabrication process of array chip of DFB plastic dye lasers.

Finally, the DFB structure was written by exposing with an interfered UV beams. The frequency quadrupled (FHG, 266 nm) and doubled (SHG, 532 nm) Q-switched pulsed Nd:YAG laser (Continuum, SureliteIII-10) were used. The SHG and FHG beams were separated from fundamental laser beam with dichroic mirrors, and superposed and injected onto the waveguide with the same incidence angle. The injected beams were sheet-formed (0.5 mm × 20 mm) with two cylindrical lenses. An Al-coated mirror was attached on the waveguide film with right angle to obtain an interference of the beams. The incidence angle θ could be changed by just rotating the waveguide. The wavelength accuracy of 0.1 nm is corresponding to the angular accuracy of 0.001° of the rotation. The accuracy of 0.02% was also required on the effective refractive indices at lasing wavelength. Therefore, the monitoring using the SHG injection was very effective to determine the incidence angle.

4. LASER PERFORMANCE OF INTEGRATED LASER WAVEGUIDE

The DFB dye laser waveguides can be pumped with very simple optical system as shown in Figure 3. A frequency doubled, LD-pumped Nd:YAG microchip laser ($\lambda = 532$ nm, Nanolaser corp., PNG-002025-040) was used as a pumping source. The output pulse energy was 40 μJ and the pulse duration was 0.5 ns. The pumping beam was expanded horizontally by a Powell lens (spread angle of 45° at 532 nm) and focused vertically by a cylindrical lens ($f = 40$ mm), and injected transversely on the DFB plastic dye laser waveguide. The pumping laser size was fitted to the waveguide area. Though the laser output was obtained from both ends of the waveguide, only one end was used in the following experiments. Selection of each strip could be made by just scanning the substrate vertically without any re-alignment.

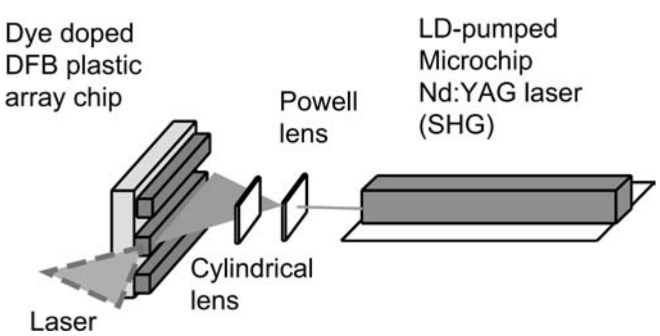

FIGURE 3 Schematic of Nd:YAG microchip pumped DFB dye laser.

The spectral coverages of fourteen different kinds of dyes or dye-mixtures were evaluated, and the results are shown in Figure 4. Since the output wavelength from each DFB waveguide was fixed, each plotted lines connect the output from different DFB waveguides that is doped with same laser dyes or dye-mixtures. Generally, the covered spectral region of the permanent DFB laser wider than that of the holographic DFB for a same laser medium. It is due the difference of the order m of the Bragg reflection and the coupling coefficient of DFB structure. For instance, the spectral coverage range of LDS722 DFB laser was about 60 nm with the this work as compared to 40 nm with the holographic DFB. Though the tunable range investigation was not finished yet for Pyrromethene 567, the demonstrated tunable region was almost covered 560–1104 nm in this results.

Subsequently, spectral narrowing properties were also evaluated for several kinds of dyes by using a spectrometer (resolution is 0.1 nm) and a solid etalon (free spectral range = 1 cm^{-1} @ 500 nm). The spectral width of 36 pm was calculated from a fringe pattern of the laser doped with Rhodamine 640 for example. The spectral width of 30 and 27 pm were also obtained from DCM and LDS722, respectively. From the calculation of the output spectral based on the coupling wave theory [18], the stop band of the DFB structure was estimated as 15 pm from the measured coupling coefficient of 1.74 cm^{-1} of an usual DFB dye lasers. The split spectrum, such as shown by a semiconductor laser with uniform DFB, could not be resolved because it is too narrow.

An investigation about input/output characteristics of the DFB lasers doped with Rhodamine 590 obtained that the laser threshold energies of

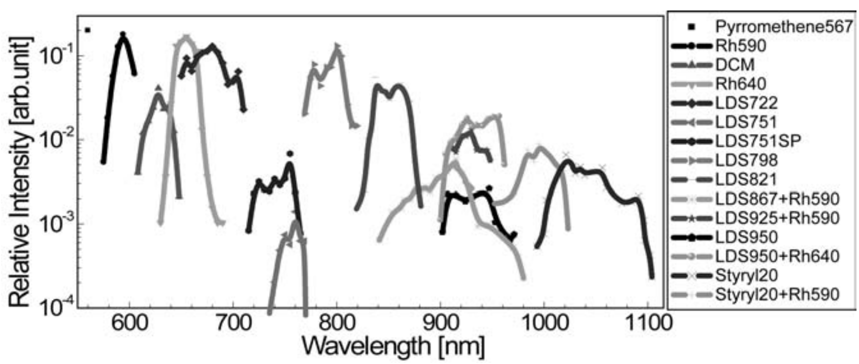

FIGURE 4 Tuning characteristics of prefabricated DFB dye laser using 10 different dopants of laser dyes and dye mixtures. Only laser operation was confirmed at Pyrromethene 567, LDS867 and LDS925.

0.52 μJ(37 μJ/cm^2) and the slope efficiencies of 0.98%. The threshold was almost 10% of the threshold of 6 μJ in the case of 6 ns pulse pumping (flashlamp-pumped Nd:YAG laser; Surelite III), and only less than 1% of that from the holographically pumped DFB (57 μJ).

Durabilities of the DFB waveguides were also evaluated. The lifetime of 2.8 million shots were obtained as a number of shots corresponding to decrease in the output down to a half of its initial value, where the waveguide was doped with Rhodamine 590 at 15.9 mM, pumping energy was 1.3 μJ, and the waveguide size was 4.5 μm(T) × 15 mm(L) × 100 μm(W). Rhodamine 640 and DCM laser dyes also showed the lifetime over million shots. In the case of Rhodamine 640, the lifetime of more than 5 million was observed with the pumping energy of 3 μJ, which corresponds to 14 hours at the repetition rate of 100 pps. These results show that the DFB dye laser has enough enough durability for a period of spectroscopic experiment. Hence the lifetime can be practically extended 100 times by fabricating 100 waveguides on the same substrate and rapidly exchanging the waveguide without any other alignment.

5. SPECTROSCOPIC APPLICATIONS

As described above, one of the advantages of our laser is that several tunable waveguided DFB lasers of different wavelength can be integrated on a plastic chip. It can propose a spectroscopic application with a novel concept, that is labeled "digital spectroscopy" in which the laser array chip recorded at scanned wavelengths like $\lambda + n\Delta\lambda$, while the traditional scheme with a wavelength scanning is termed as "analog spectroscopy".

Figure 5 show an example of the spectroscopic measurements by using a multi-wavelength dye laser in combination with an array detector. All of the waveguides can be pumped simultaneously by a single laser pulse, and

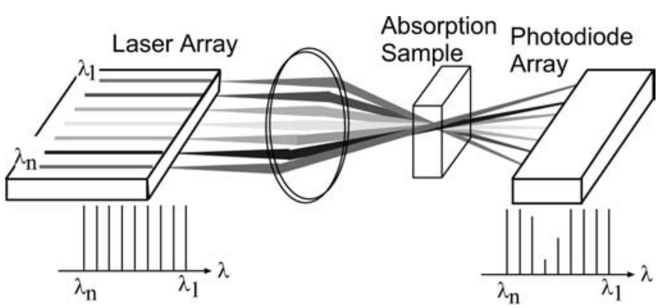

FIGURE 5 Conceptual schematic of applications of multi-wavelength waveguided dye laser array to absorption spectroscopy.

independently operating laser waveguides are formed on the single chip. By selecting different dye for each waveguide, multi-wavelength dye laser covering an arbitrarily wide spectrum can also be designed. The spectrum can be measured in the single laser shot without any wavelength scanning. It can provide a very compact, stable and cheap spectrometer, and the desired wavelength range and the wavelength intervals can be designed according to any requirements.

In the experiment, Rhodamine 590 dye with a concentration of 23 mM was used and as a typical example, absorption lines of sodium atoms were detected. Altogether 9 waveguides were fabricated with a spatial pitch of 0.5 mm and the DFB structure was written on each waveguide with a spectral pitch of $\approx$0.11 nm. The wavelength region between 589.8 nm and 590.7 nm was used to cover the primary absorption line D_1 and D_2 of the sodium atoms. The whole size of DFB laser array was 15 mm $\times$ 7 mm, thus the array could be pumped transversely with only one shot of the Nd:YAG laser pulse. This was achieved by using an optical lens system and forming a rectangular pump beam.

Figure 6(a) shows the spatial intensity distribution at the surface of the photodiode array. The dashed-line represents the spectrum without the absorption at the sodium cell and the solid line represents the spectrum with the absorption. Even though the horizontal-axis represents spatial position on the photodiode array surface, each peak position in the profile

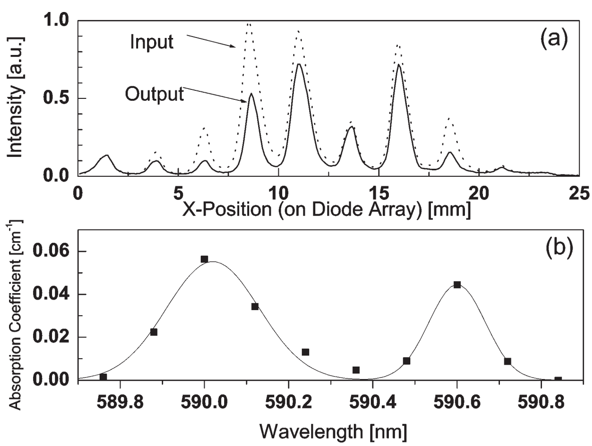

FIGURE 6 Spatial intensity profile of multi-wavelength dye laser array output through sodium vapor cell. (a) Represents a spatial profile without (input) and with (output) absorption, and (b) is digital spectral profile calculated from (a). Each peak position on the profile (a) corresponds to a separate output wavelength from each waveguide.

corresponds to a designed output wavelength from each waveguide. The intensity of the peaks at the wing of the spectrum was decreased as a result of the limited aperture size controlled by the smaller sodium cell dimension. Figure 6(b) shows the digital absorption spectrum derived from Figure 6(a). Sodium D_1 and D_2 lines were observed clearly, however the spectral width of the profile was broadened to 0.3 nm. The resolution is expected to be enhanced by improving the accuracy in a DFB fabrication. This experiment clearly demonstrated that the DFB waveguide array can be used to obtain a spectral profile without any wavelength scanning.

6. CONCLUSION

Integrated tunable laser array systems on a plastic chip have been reviewed. The waveguided distributed feedback dye laser waveguides were fabricated by a spin-coating, UV lithography, and interfered UV exposure techniques. The narrowed spectal width of 0.04 nm, the laser threshold energy of less than 1 μJ, the durability of more than 5 million shots were attained. The tunable range from 560 to 1100 nm was also demonstrated. Finally a novel scheme of spectroscopic application was also proposed and demonstrated.

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