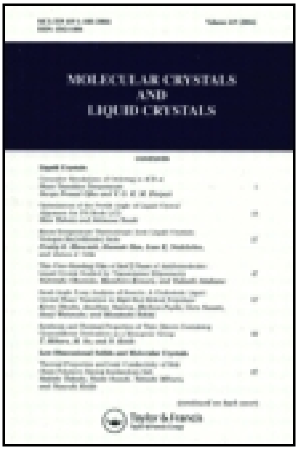

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Distributed Feedback Waveguide Laser of Organic Nano-compound Material

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The laser action was firstly demonstrated using 1-cyano-trans-1,2-bis-(4′-methyl-biphenyl)ethylene (CN-MBE), that enhances its fluorescence from organic nanoparticle. The CN-MBE is not fluorescent and has enhanced fluorescence in aggregated nanoparticle state. CN-MBE doped PMMA film showed fluorescence over 400 ~ 600 nm and spectral narrowed emission at 430 nm under pumping with a nanosecond pulsed laser at wavelength of 355 nm. CN-MBE:PMMA DFB laser waveguides were successfully fabricated, and lasing wavelength was over 430 to 490 nm. CN-MBE:PMMA DFB lasers showed laser threshold of $73\,\mu J/cm^2$ *at the waveguide size of* $15\,mm \times 100\,\mu m \times 5\,\mu m$*. The output spectra had bandwidth as narrow as 60 pm FWHM, and suppressed fluorescence background of less than 0.5%. The maximum peak power and average power were estimated to be 5 W and* $0.35\,\mu W$*, respectively. The durability was as long as* 1.7×10^4 *shots/waveguide.*

Keywords: distributed feedback waveguide laser; durability; laser power; organic nanoparticle

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1. INTRODUCTION

The plastic dye laser has been studied by many research groups [1–3] and a waveguided and/or a distributed feedback (DFB) solid-state dye laser have also been studied [4–8] to simplify the tunable laser system and to decrease laser threshold energy. We have studied optically pumped waveguide-DFB film dye laser as an integratable tunable laser. The laser-integration technique is important to extending the applicable region of the laser aided spectroscopic applications. We developed distributed-feedback waveguide laser based on acrylic copolymer with laser dye doping, and demonstrated conversion efficiency as high as 10% [9], laser threshold less than μJ, wide-tunability [10] extended durability [11], tunable random medium lasers [9], and fiber top laser scheme [12].

Subsequently, we are studying about the integration technique of film lasers, and considering about integrated spectroscopic devices. A multicolor laser array can be made using the integratable lasers and it can cover the spectral range from 400 to 1100 nm by using a frequency-doubled and -tripled Nd:YAG laser pulses. However, organic laser dye has a problem of degradation, especially in the solid-state matrix as under the UV pumping. In the case of green-laser pumping, typical DFB laser waveguide could perform the life-durability as long as several million shots in repetition rate of 100 pps [11]. However, the traditional laser dyes for blue, such as LD473 or LD466, represented short life of fewer than one hundred shots in the polymeric film and UV pumping.

In recent years, various kinds of novel luminescent organic molecule have been developed for applications in organic light emission devices. The optical properties related to the electro-luminescence (EL) performances of such molecules have been investigated and improved during the course of their development processes. On the other hand, trials on organic laser diodes have been reported; therefore the lasing properties of novel molecules also get important. We already reported the laser action of the novel organic compounds with a 2,7-bis-biphenyl-4-y1-dihexylfluorene as a first order DFB plastic blue-violet dye lasers [13].

In this summary, laser properties of organic compounds of 1-cyano-trans-1,2-bis-(4′-methylbiphenyl)ethylene (CN-MBE) were clarified and first-order-distributed-feedback (DFB) plastic blue dye lasers were demonstrated. The CN-MBE has no fluorescent in solution (molecular state) but has enhanced fluorescence in nanoparticles of J-aggregation. This is the first report of fabricated-DFB laser doped with CN-MBE compound. Furthermore, the performances of the fabricated laser were investigated using the DFB waveguide dye laser. Results showed a laser threshold of as low as 1.1 μJ and a lifetime of 17,000 shots from a laser cavity size of $15\,\text{mm} \times 100\,\mu\text{m} \times 3 \sim 5\,\mu\text{m}$.

2. OPTICAL PROPERTIES OF CN-MBE IN PMMA FILM

Figure 1 shows the structural formula of the CN-MBE compounds used in this work. CN-MBE shows weak fluorescent in free-molecular state such as THF solution, but enhanced it in nanoparticle state such as water-THF mixture [14]. The fluorescence from the CN-MBE nanoparticle lies at about $450 \sim 550\,\text{nm}$. We had to dope the CN-MBE molecules into a plastic film to fabricate solid-state DFB laser. So, we confirmed first the fluorescent properties of CN-MBA doped film. We adopted poly(methyl methacrylate) (PMMA) as a matrix of the CN-MBE and intense fluorescence could be obtained from CN-MBE:PMMA.

The film fabrication is as the followings:

1. The CN-MBE was mixed into MMA monomer.
2. The mixture was radically polymerized and spin-coated just before completion of the polymerization.
3. Post annealing finished the polymerization of the film.

The CN-MBE:MMA soltion of CN-MBE shows very weak fluorescent like THF solution. However, after polymerization, we could see intense fluorescence from the CN-MBE:PMMA bulk and its film, where $x = 0.02$.

The solid and dashed lines in Figure 2 show the fluorescence and absorption spectra of CN-MBE:PMMA film at a concentration of 21 mM, respectively. The absorption crosssections was approximately 4.4×10^{-17} at a wavelength of 355 nm, and it seemed suitable for 355 nm pumping. The fluorescence was over $400 \sim 600\,\text{nm}$. Though there is slight difference on the fluorescence spectra between the PMMA film and THF/water [14], the CN-MBE seemed to aggregate successfully in the PMMA because of the enhanced fluorescence.

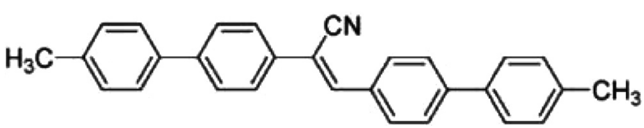

FIGURE 1 CN-MBE molecule.

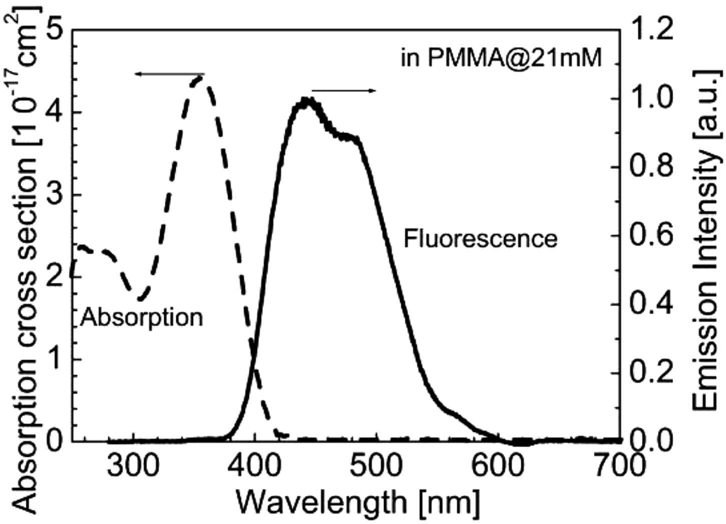

FIGURE 2 Absorption (dashed) and fluorescence (solid) profiles of CN-MBE doped PMMA film.

Figure 3 shows the results of fluorescence lifetime measurements. The filled and open circles are the fluorescence profiles of the solution and film, respectively. The lifetime of 0.36 ns in the solution extended

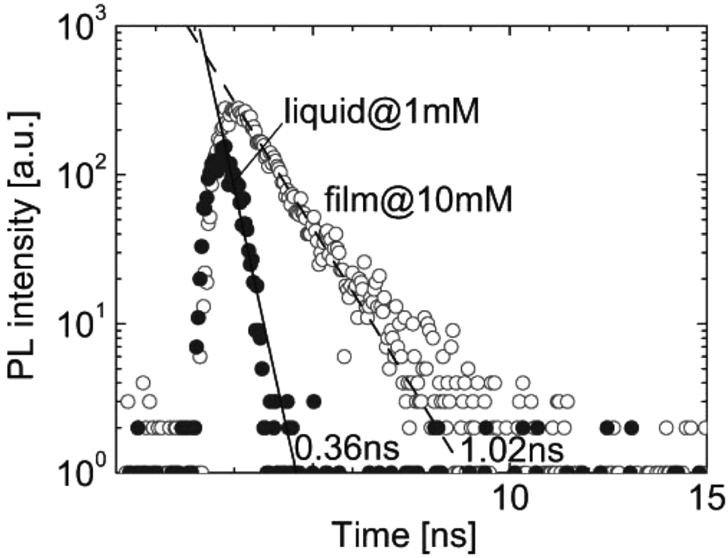

FIGURE 3 Plots of fluorescence decays of CN-MBE solution in a 1,2-dichloroethane (filled circle) and CN-MBE:PMMA (open circle).

to 1 ns in the film. The lifetimes of 1 ns are shorter than that of a rhodamine6G dye, which is a typical laser dye. The stimulated emission cross sections of the samples were also estimated using the following formula:

$$\sigma_s(\lambda) = \frac{\lambda^4 \Phi E(\lambda)}{8\pi\tau c n^2} \tag{1}$$

where $E(\lambda)$ is normalized fluorescence spectra, Φ is a quantum yield, and τ is lifetime. If the quantum yield can be assumed to be 1 here, the stimulated emission crosssection σ_{se} is expected to be $2 \times 10^{-16}\,\mathrm{cm}^2$. It is comparable to that of rhodamine6G laser dyes; therefore, laser operation can be predicted.

3. FABRICATION OF DFB LASER WAVEGUIDES

Figure 4 shows fabrication procedure of the DFB solid-state dye lasers. Core layers were PMMA doped with the sample compound, and clad

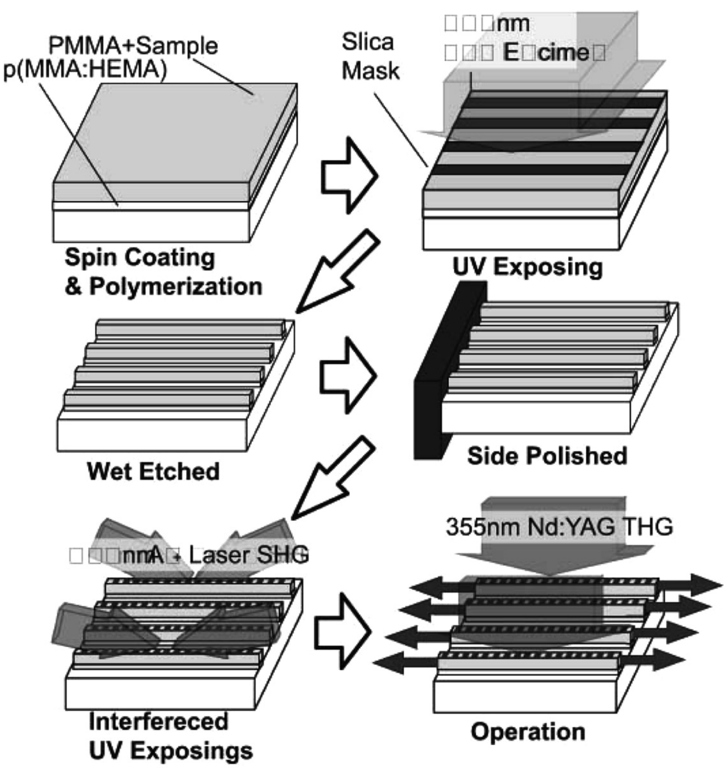

FIGURE 4 Fabrication process of array chip of DFB plastic dye lasers.

layers were also prepared if needed. First, MMA was radically polymerized at temperature of 73°C and partially polymerized. Then the sample compound was dispersed by using 1,2-dichloroethane as a delivery, since the heating can degraded the sample. Next, the mixture was spin-coated on a substrate as a core layer before completion of the polymerization. For the single mode waveguide, PMMA without the compound was and P(MMA:HEMA) with the compound were spin-coated sequentially as clad and core layer, respectively. The refractive index and propagation mode were monitored by a prism-coupler (Metricon, Model-2010) after each spin-coating.

Subsequently, stripped waveguides with size of 15 mm(L) × 100 μm(W) × 3 μm(T) were fabricated by an UV lithography (KrF Excimer laser, 248 nm) and a wet etching. Thirty independent waveguides could be fabricated with arrayed formation that has a splitting-pitch of 600 μm on a plastic chip. The DFB was written on each waveguide by two-beam-interfered exposing with a frequency-doubled Ar^+ laser (Spectra Physics, BeamLok 2060 and Wavetrain). Each DFB waveguide has the recorded Bragg grating made of refractive index modulation, so they can have individual DFB wavelengths one another. The grating pitch Λ was determined using

$$\Lambda = \frac{m\lambda}{2n_{eff}} = \frac{\lambda_e}{2\sin\theta} \tag{2}$$

where m, n_{eff}, λ_e and θ are an order number of Bragg reflection, an effective refractive index of the waveguide, the exposing UV wavelength, and the incident angle of UV beams for interfered exposure, respectively. Since the first-order ($m = 1$) Bragg grating for the DF is required for the lowest laser threshold, a fine grating of $\Lambda \approx 146 \sim 164$ nm must be fabricated using the angle θ of $48 \sim 57°$. Fabrication adopted the exposing fluences of 1 J/cm^2. The refractive index and propagation mode were monitored by a prism-coupler (Metricon, Model-2010) after each spin-coating.

4. EXPERIMENTS AND RESULTS

First, the nanosecond frequency-tripled Nd:YAG irradiates the CN-MBE:PMMA file to confirm spectral narrowing. Figure 5 shows the fluorescence spectra when the pumping fluence was increased. Increasing the fluence from 5.8 to 631 mJ/cm^2 make the spectral width changed from 100 to 12 nm FWHM. The center wavelength of the ASE is 435 nm. Since the spectral narrowing started from 45 mJ/cm^2, the ASE threshold seems around it. It is too intense for

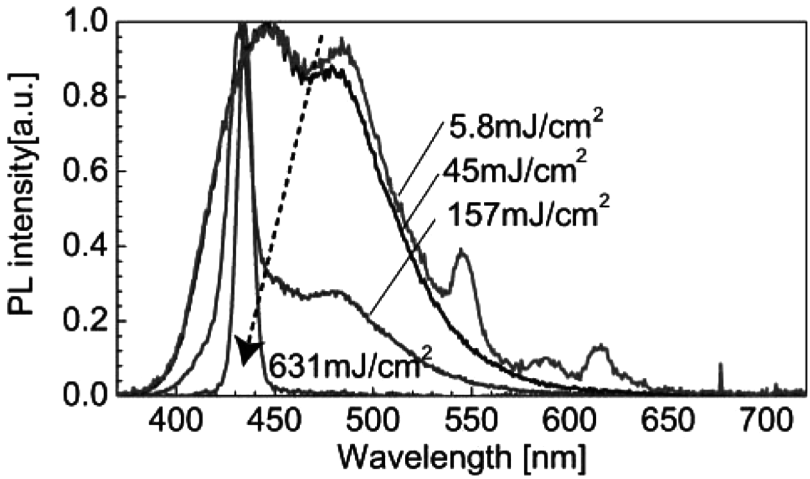

FIGURE 5 Fluorescence spectra of CN-MBE:PMMA film pumped with 355 nm UV pulse.

the organic film generally, so CN-MBE:PMMA films were also degraded rapidly with such a high fluence.

Figure 6 shows examples of the spectra of fabricated DFB waveguide laser that the molecular concentration was approximately 20 mM and film thickness was about 5 μm. The pumping source was a frequency-tripled microchip Cr:Nd:YAG laser; a output pulse energy, pulse duration and repetition rate were 16 μJ, 0.7 ns and 100 pps, respectively. The DFB waveguiding attained clear monochromatic spectrum, so the sample attained a single mode waveguiding. The center wavelength of 454 nm, represents that a fine Bragg grating with pitch of 153 nm was successfully fabricated. The bandwidth of laser output was 0.06 nm, and DFB cavity could suppress a background ASE less than 0.5% of the peak intensity. However, we could also observe double or triple modes waveguiding from some of samples, refractive index control may be needed in future.

Subsequently, the DFB pitch varied on each laser waveguide to change the oscillating wavelength. Just exchanging the waveguide on the array, could change the lasing wavelength as shown in Figure 7. A dashed line in Figure 7 shows a spectral narrowed ASE in Figure 6. Obtained laser output coverage was from 433 to 493 nm. Though the wavelength was discrete, the output wavelength can be selected to any wavelength in the region by controlling the incidence angle θ. As an output spectrum at 434 nm shows, multiple-peak outputs spectrum because of multimode were observed in several case.

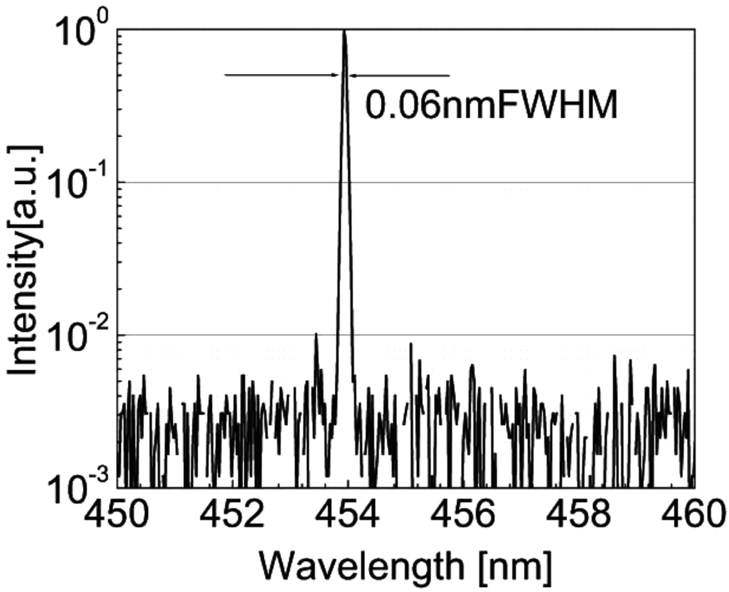

FIGURE 6 Spectral profile of DFB laser output.

But refractive control can remove this problem. We could not obtained DFB laser output in longer than 500 nm even though the fluorescence of CN-MBE:PMMA extends to 600 nm approximately. The reason has not been clear, yet, but an excitation state absorption can be guessed.

Figure 8 shows the input-output characteristic of the CN-MBE: PMMA DFB laser. The laser threshold was only 1.1 μJ (73 μJ/cm^2)

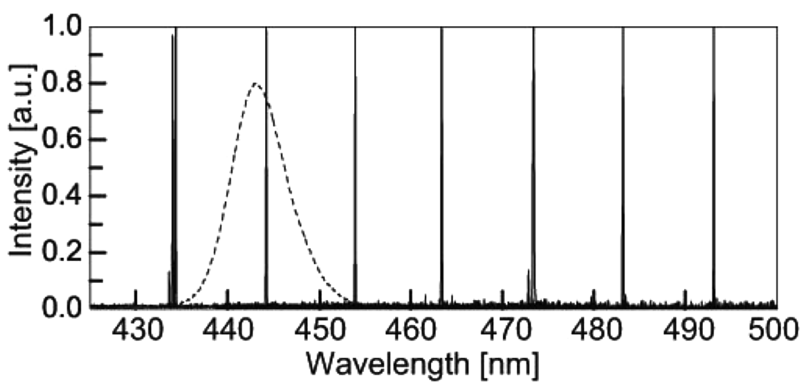

FIGURE 7 Spectral profiles of DFB laser output with varying DFB pitch. Dashed line represents spectral narrowed ASE.

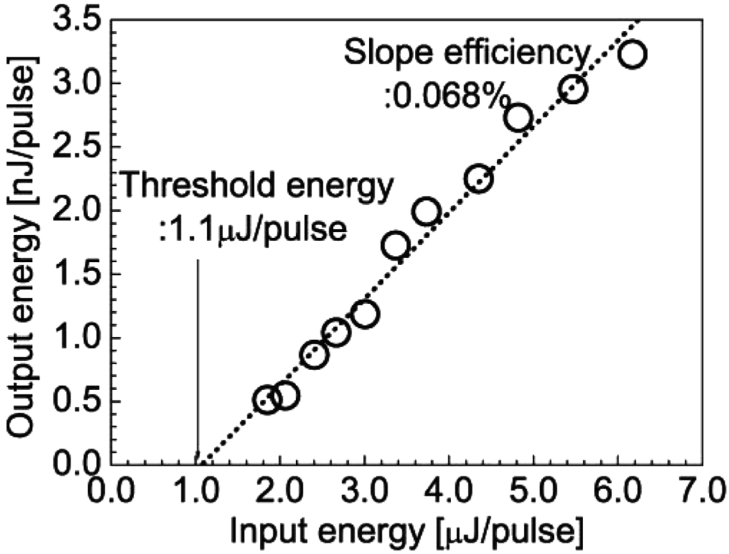

FIGURE 8 Output energy of DFB waveguide laser as a function of pumping UV pulse energy.

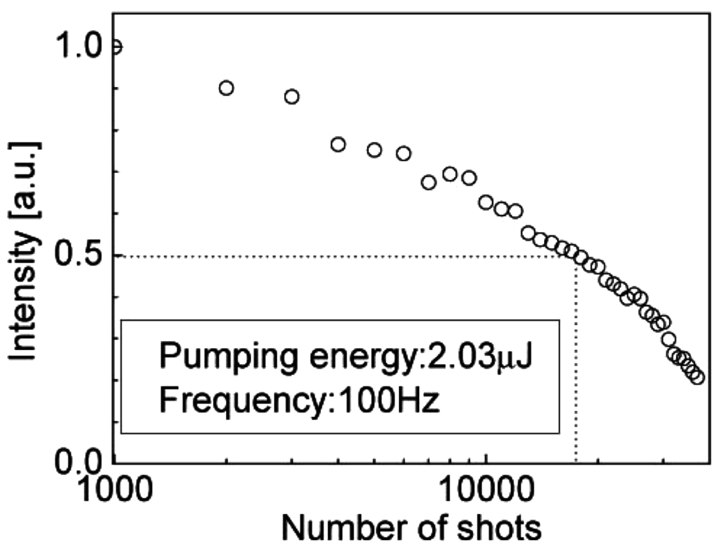

FIGURE 9 Durability of DFB waveguide laser.

for the laser medium volume of 15 mm(L) $\times$ 100 μm(W) $\times$ 5 μm(T). This value was relatively high in comparison with fluorene-based blue-violet laser [13]. An average absorption photon density of 2.6time-s;10^{17} cm^{-3} can be estimated from 1.1 μJ, so the population inversion ratio N_{S1}/N_{all} was almost 1%. A slope efficiency of 0.068% was also obtained.

Finally, laser durability against the optical pumping of the CN-MBE:PMMA DFB waveguide was examined using the same pumping source. Figure 9 shows the results, in the case of a dopant concentration of 20 mM, an input energy of 2 μJ and a waveguide thickness of 5 μm. A lifetime of about 1.7×10^4 shots was obtained, which was the longest among all the developed DFB waveguide lasers for 450 nm. For instance, we previously tried LDS473 and LDS466 in the PMMA films, only 80 and 30 shots were obtained as a lifetime. It seems because of the robust structure of the aggregated CN-MBE particles.

5. CONCLUSIONS

A novel material CN-MBE molecule was investigated as laser medium. CN-MBE can make aggregation in a spin-coated PMMA films and CN-MBE:PMMA film shows enhanced fluorescence even though the CN-MBE:MMA solution shows no fluorescence. The stripped waveguides with distributed feedback were success-fully fabricated, and DFB laser action was demonstrated for first time using CN-MBE by using frequency-tripled Cr:Nd:YAG microchip laser. The tunable range was wide as 433 $\sim$ 493 nm and laser threshold of 73 μJ/cm^2 in the 5 μm thick film. The conversion efficiency was 0.068%. The CN-MBE:PMMA shows better durability, such as the life of 1.7×10^4 shots/waveguide.

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