Indonesian Journal of Physics
Vol 21 No. 3, July 2010

Application of Hybrid Polymer as Laser Devices Based on 2D Grating with Square Structure

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Received 4 March 2010, Revised 30 March 2010, Accepted 29 June 2010

Abstract

Two dimensional gratings have been fabricated by laser interference method on hybrid polymer films doped with 4-dicyanmethylene-2-methyl-6-(p-dimethyl-aminostyryl)-4H-Pyran (DCM). The gel precursor of hybrid polymer was synthesized by sol-gel route using 3-(trimethoxysilyl)-propyl-methacrylate (TMSPMA). The film was exposed by laser interference of Nd-YAG laser yield square lattice of 2D-grating with periodicities 391 nm and 405 nm. The characteristic of lasing was investigated using strip-line excitation light of second harmonic generation (SHG) of Nd-YAG laser (λ = 532 nm). The lasing wavelengths are observed at (588 nm $\pm$ 2) and (606 nm $\pm$ 2 nm) for grating period of 391 nm and 405 nm, respectively. The spectral width of lasing is about 2 nm at pumping power 6.6 mJ/pulse.cm^2.

Keywords: *2D grating, Square structure, Hybrid polymer, Laser.*

1. Introduction

Lasers are devices that amplify or increase the intensity of light to produce a beam in certain directional and high intensity that typically has a very pure frequency or wavelength. The typical laser device consists of an amplifying or gain medium, a pumping source to input energy into the device, and an optical cavity or mirror arrangement that reflects the beam of light back and forth through the gain medium for further amplification.

A new class of laser systems occurs when the dielectric microstructure responsible for light scattering consists of a periodic array of identical unit cells. Photonic crystals are periodic dielectric structures exhibiting strong Bragg scattering as well as microscopic resonance scattering of electromagnetic waves. Under suitable circumstances with respect to material composition, topology, and lattice symmetry, a forbidden frequency range may be created. A forbidden frequency range is known as the photonic band gap. Photonic band gap able to modify the density of electromagnetic modes and enhance spontaneous emission, therefore photonic crystal can be used as laser resonator[1,2]. There are two types of lasers associated with photonic crystals. The first is the band edge micro laser, in which the light emission occurs at the photonic band edge. The second is the defect mode micro laser, which utilizes a localized state defect mode as a laser cavity[3].

M. Maier *et al.*[4] have reported fabrication and analysis of laser resonator with two dimensional distributed feedbacks from photonic crystal. The gain medium consists of 2-(4-biphenylyl)-5-(4-tert-butylphenyl)-1,3,4-oxadiazole host doped with Coumarin 490 and DCM and is deposited on patterned Si/SiO_2 substrate. Two dimensional structure of Si/SiO_2 substrate was fabricated using electron beam lithography.

Recently, hybrid polymer as one of sol-gel materials is good candidate for optical devices. They have several advantages for application as optical devices, such as easy in synthesizing and patterning process, easy customized by add some functional material, and exhibit a good transparency in optical region[5]. Several fabrication processes for optical devices have been developed, such as photo lithography, electron beam lithography, and embossing[6,7]. A simpler method for fabrication the two dimensional structure from hybrid polymer is photo-lithography using Lloyd mirror interference method[8].

In this paper we report about the lasing performance of two dimensional photonic crystal using hybrid polymers as base material. The gel precursor of hybrid polymer synthesized by sol-gel method and doped with DCM. Two dimensional micro-structures was fabricated using Lloyd mirror interference method. The photo polymerization process of precursor hybrid polymer was also investigated using IR spectrophotometer.

2. Experiment

The gel precursor of hybrid polymer was synthesized by sol-gel method using TMSPMA as monomer. TMSPMA was obtained from Aldich. Chloroform ($CHCl_3$), ethanol (C_2H_5OH), and HCl (aq) were obtained from Merck. The precursor of hybrid polymer was prepared in the process including four steps, as follow:

a. TMSPMA and ethanol were mixed then stirred for 1 hour at room temperature yielding the monomer solution.
b. Deionized water was gradually added to monomer solution until water to TMSPMA volume ratio of R=2 were attained.
c. The process of condensation of TMSPMA were carried out by adding HCl. The solution was stirred (200 rpm) over night at 65^{o}C yielding the gel hybrid polymer.
d. The final step is purification process for removing undesirable reactant using chloroform.

For fabrication of two dimensional grating, a suitable photo- initiator (IRGACURE 369, Ciba) was added into precursor hybrid polymer with weight ratio 0.5%. Organic dye laser (DCM) also was added into this mixture with weight ratio 0.1%. In order to easily control the thickness of film upon substrate, the mixture was diluted using chloroform. Then the precursor films were prepared by spin coating at 500 rpm for 5s following 2000 rpm for 30s.

The Lloyd mirror interference method was used to make light patterns in sub-micrometer order on the precursor film. The two dimensional gratings can be fabricated by double exposures with rotation of precursor film between the exposure. The photo-polymerization process will occurs in irradiated area and the etching process can remove the un-irradiated part. The Lloyd mirror interferometer set up schematically shown in Figure 1[8,9]. The third harmonic generation (THG) of Nd:YAG laser (λ=355 nm) was used as curing source. Various curing powers were applied in this experiment, i.e. 120 mWatt, 200 mWatt, and 320 mWatt with shooting time ¼ second.

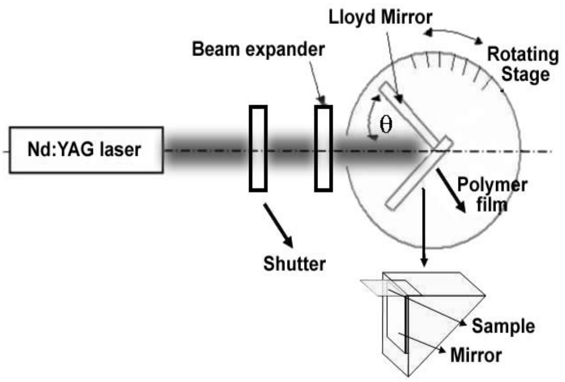

Figure 1. Lloyd mirror interference setup.

The grating period Λ corresponding to the space between fringes in the interference pattern is given by:

$$\Lambda = \frac{\lambda_C}{2\sin\theta} \quad (1)$$

Where λ_C is the curing wavelength and θ is the angle between the incident beam axis and the mirror plan.

The change of molecular structure during photo-polymerization process was observed using FTIR spectrophotometer. The formation of grating structure was investigated using the Atomic Force Microscopy (AFM). The lasing characteristic was investigated using strip-line excitation light from SHG of Nd:YAG laser (λ=532 nm).

3. Results and Discussion

The precursor hybrid polymer was prepared by the sol-gel method. The sol-gel processes consist of hydrolysis and condensation reactions of inorganic part resulting inorganic silicate network. The photo polymerization process will be constructed the cross linking in the organic side chains. The reaction scheme of the formation of inorganic and organic network is depicted in Figure 2.

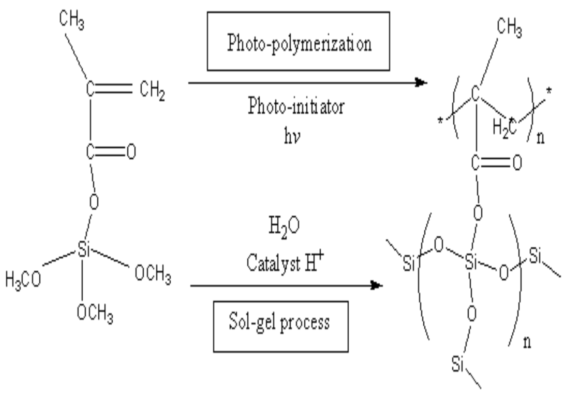

Figure 2. The formation of hybrid polymer from TMSPMA.

Figure 3 shows the changes in infra red spectrum of precursor hybrid polymer under low power UV irradiation. After irradiation or photo polymerization process, the infra red band at 1638 cm-1 was significantly increased. And the otherwise, the infra red band at 1260 was significantly decreased. They were indicating the reduction of C=C double bond and conversion into C–C bond. The process will be followed with construction of cross linking in organic part and alteration of hybrid polymer phase from gel into solid. The chemical reaction during photo-polymerization process can be seen on Figure 2.

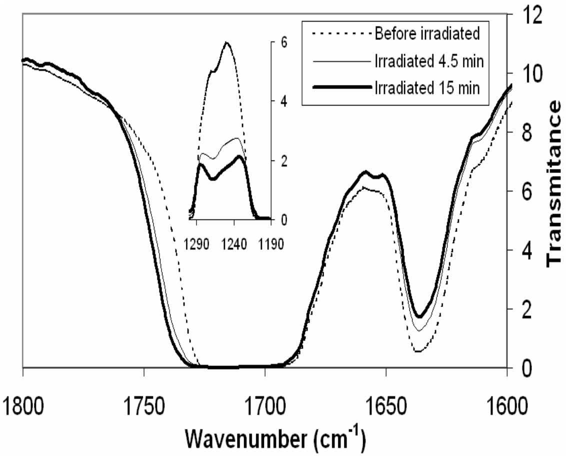

Figure 3. FTIR spectrum of precursor hybrid polymer during photo polymerization using low power UV light

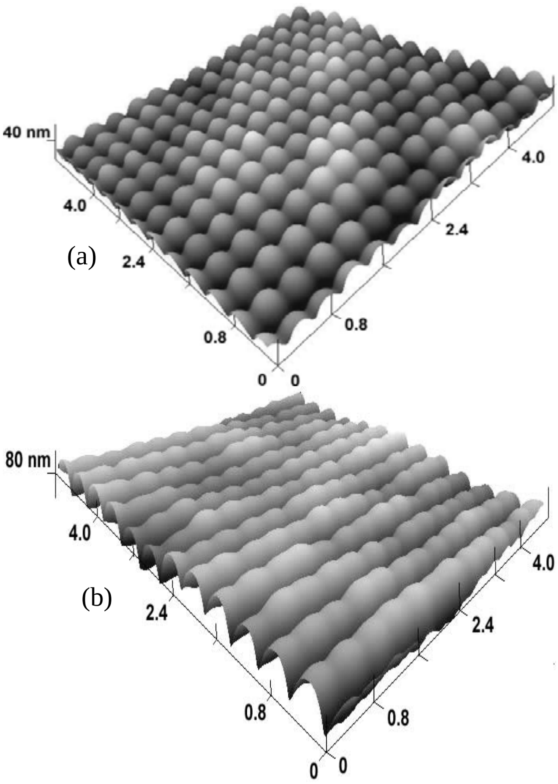

Figure 4. AFM surface profile of two dimensional grating, (a) Irradiation power: 200 mWatt; (b) Irradiation power: 320 mW.

Two dimensional gratings were fabricated from hybrid polymer incorporated with organic dye laser DCM using Lloyd mirror interference method as shown in Figure 4. The photo-polymerization processes dependent on the dose of laser and the irradiation time. In this experiment, the dose of laser was varied in three kinds, i.e. 120 mWatt, 200 mWatt, and 320 mWatt with same irradiation time at ¼ second. Fabrication the gratings at the dose of laser 120 mWatt did not generate. It caused by the photo-polymerization process did not occur when the dose of laser less than the threshold. In this case, the cross linking in organic side chain could not formatted. The film of precursor hybrid polymer remained in the gel phase and dissolved when etching process was conducted. Whereas if the dose of laser much more than the threshold, the grating will formed in one shoot, and the second shoot did not affect on grating formation as shown in Figure 4(b).

Figure 4(a) shows the AFM image of two dimensional grating that was fabricated at appropriate dose. The grating structures were prepared at various incident angle of the interference laser beam. In principle, the dependence of grating period (Λ) on the incident angle (θ) can be estimated as shown in Equation (1). The choosing of incident angle must match with wavelength of band-gap of 2D photonic crystal that corresponds to the lasing wavelength[1].

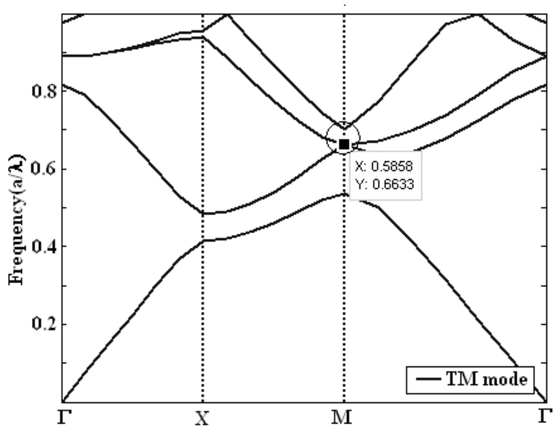

Figure 5. Two dimensional band structure in Γ-M direction

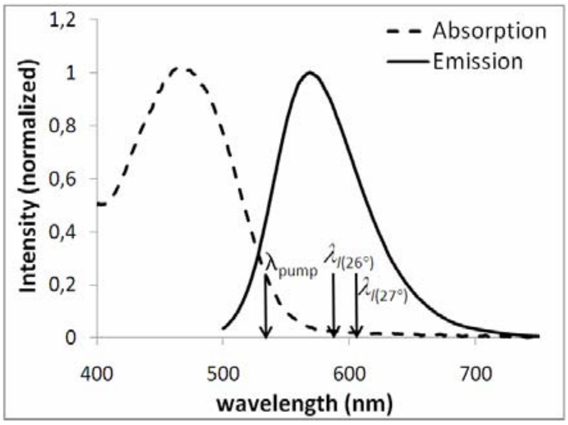

Figure 6. Emission and Absorption of Hybrid polymer doped with DCM

Figure 5 shows the photonic band structure of TM mode for 2D surface-grating with square symmetry that was calculated within the effective 2D model. The parameter structures were assumed as follow: n_{rod} =1.5, n_{hole} =1, and r_{rod} =0.25$\boldsymbol{a}$ where $\boldsymbol{a}$ is lattice constant. In this case, the complete band gap can't find for this parameter structure. The gaps only arise in certain direction as pseudo gap.[11]

When the beam of light inside to the photonic crystal, the characteristics of wave propagation will change. Around the eigen mode of photonic crystal, the group velocity will decrease and lead to increase the concentration of propagation modes. Exactly on the eigen band, the group velocity is equal to zero and then resulted the process of distributed feedback. Hence, the stimulated emission will increase at the photonic band edge when the group velocity equal to zero[12)].

We can assume that distributed feedback occurs at the edge of upper band or frequency at 0.663 ($\boldsymbol{a}/\lambda_l$), where $\boldsymbol{a}$ is grating-period and λ_l is lasing wavelength. In this experiment, we have fabricated grating with the incident angle of 26° and 27°, which correspond to grating period of 405 nm and 391 nm respectively. The lasing wavelengths are expected at 610 nm and 589 nm for grating period 405 nm and 391 nm respectively. Both values will be in rank of emission frequency of hybrid polymer doped with DCM as shown at Figure 6.

Figure 7 shows the lasing action of 2D structure from hybrid polymer doped with DCM. The samples were optically pumped at frequency 532 nm from SHG of Nd-YAG laser. If pumping power less than pumping threshold, then emission spectrum only amplified spontaneous emission (ASE). Increasing pumping power will sharpen the ASE. If pumping power higher than power threshold, then the lasing action will occur. For both grating-periods, the power thresholds are around 4 mJ/pulse.cm^2. We can observe the lasing action at 588 nm and 606 nm for grating period of 391 nm and 405 nm respectively as shown in Figure 7. The spectral widths (Full Width Half Maximum) are around 2 nm at pumping power 6.6 mJ/pulse.cm^2. The increasing of pumping-power will make the spectral width more sharp. If the lasing-action shoots to screen, we can observe the orange-laser. The lasing action can be observed in various directions in plane of grating sample that perpendicular to the pumping beam. The lasing actions appear at the angles of 0° till 5° with the intensities are decreasing respectively. In the rank of angles from 6° till 82°, the lasing intensities are zero. The lasing actions arise again at the angles of 83° to 90°. The complete lasing action in various directions can be seen at Figure 8.

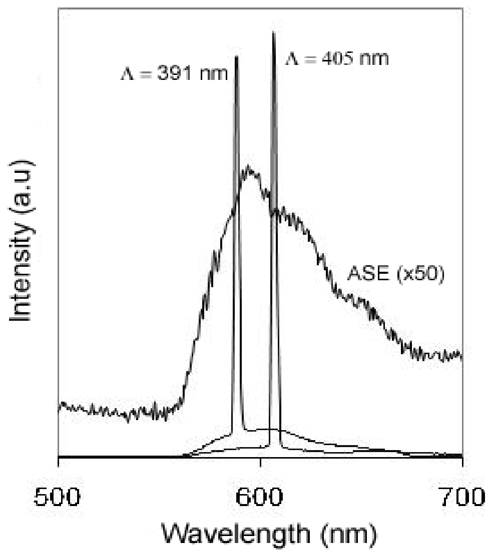

Figure 7. Lasing action in x-direction

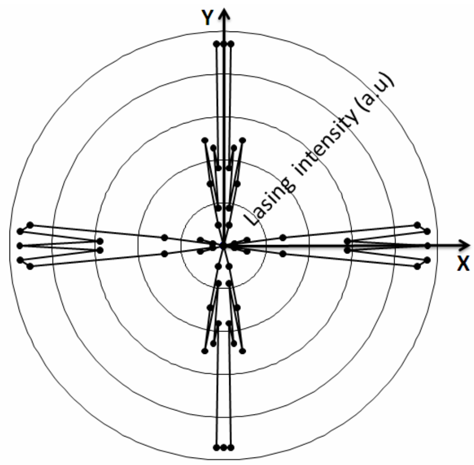

Figure 8. Lasing action in various directions

4. Conclusions

In this experiment, we have implemented a new fabrication technique for 2D surface grating using hybrid polymer incorporated with organic dye laser. The organic dye laser was mixed with precursor hybrid polymer using chemical process before fabrication of film. The compact gratings have been fabricated using Lloyd mirror interference method. The grating parameters are easy to control by setting the incidence angle of pumping beam onto the film and the curing power. We have been demonstrated the lasing action from distributed feedback of 2D photonic crystal with square structure. The experimental measurements also have been confirmed by theoretical results with agree well.

Acknowledgements

We would like to thank to Masayoshi Ojima for helpful in running AFM measurements. This work was supported by JSPS and DGHE Indonesia.

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