Organic Polymer DBR laser by Softlithography

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We fabricated the distributed Bragg reflector (DBR) laser that consisted of the grating of 600 nm in period on the surface of the charge transport polymer film doped with a low threshold laser dye by using a soft lithography technique. The 600-nm-period grating acted as the fourth order diffraction DBR for the laser oscillation wavelength. The laser oscillation of FWHM of 0.028 nm and the Q-value of approximately 9200 were observed from the device by optical pumping. The laser oscillation threshold obtained from the input-output characteristics of the device was 37.6 µJ/cm².

Keywords: DFB laser, softlithography, charge transport polymer, DBR laser

1. Introduction

In recent years, many researchers' attention is centering to large-area processability and easy customization by chemical modification of organic electronic materials and the heat of the researches regarding those materials is rising. Among those, the organic semiconductor laser is one of the most aggressive research subjects. By using an organic single crystal, the first organic solid laser oscillation operated by electric currents has been reported by Schön et al.1 In their report, they employed double sets of field effect transistor electrodes for carrier injections in order to control the injection balance of hole and electron and also used cleavage faces of an organic crystal as reflectors of a Fabry-Perot resonator. As a result of such an ingenious device design, they accomplished such a heart-stirring enterprise. Their result is very important in the point that opened up the new phase in organic electronics by announcing potentials of the organic semiconductors.

However, it is also the goal of the ultimate to materialize the same achievement with amorphous organic solid-state lasers will provide flexibility, lower threshold, high efficiency, wide-range wavelength tunability, surface light emissivity, and so on.

However, the end-edge face organic thin film does not become in an accurate plane like the cleavage face of an inorganic semiconductor crystal, because the organic active layers formed by spin-coat method and vacuum evaporation are amorphous. In addition, the refractive index of organic materials is small in comparison with inorganic materials. From such a reason, that the end edge faces of an organic emitting layer work as ineffective reflectors of a Fabry-Perot resonator.

Thereupon, the proposal, in which distributed feedback (DFB) or distributed Bragg reflector (DBR) were used as a resonator for organic solid-state lasers, was done from before.2,6) Also, the laser oscillation by optical pumping was observed from an organic DFB laser that consists of the second order grating of small coupling coefficients and a laser dye of a low threshold.7) It has an advantage that preparation is easy in comparison with the first order grating with the same Bragg wavelength. Also, an organic DFB laser that does not accompany a structural change has been materialized by using the technology of Plano Light-wave Circuit.8) Furthermore, a
characteristic emission spectrum was observed from an organic light emitting diode (OLED) with a grating in the emitting layer even in a current drive device.9) In this time, we report that a laser oscillation is observed from an optical pumped organic DBR laser formed by softlithography10) on the surface of a laser dye doped charge transport polymer film. The common characteristic of a DBR and DFB lasers is that the light of Bragg wavelength of the grating is amplified only. The difference of DBR and DFB lasers is that the active region is separated from the grating region in DBR lasers, while the grating exists in the whole active range in DFB lasers. In the case of current operations in the future, the inhomogeneity of thickness of active layer that originate on the unevenness of grating exert bad influence on the stability of the device, when there is grating in the active region like DFB lasers. On the other hand, in DBR lasers, charge carriers can be injected homogeneously, because the active range is flat. Therefore, we expect that the performance of the OLED as an active region can be maximized by using DBR laser diode structures.

2. Experiment

2.1 Organic Materials

An active layer of the device was consisted of a semiconducting polymer material and a low-threshold laser dye. CTP-5 (FUJI XEROX) that exhibited an excellent hall transport ability caused by its TPD framework was used as a binder polymer. 1,4-Bis (2-((N, N-di (p-tolyl) amino) phenyl) vinyl) benzene (laser dye A, Tokyo Kasei Kogyo) that has an extremely low gain-narrowing threshold was doped to CTP-5 as a laser dye at a mixing ratio of 10:1 in weight. CTP-5 works not only as a binder polymer but also as an energy transfer host to laser dye A, because CTP-5 also has bright luminescence at a wavelength region around 410 nm and the luminescence is well overlapped to the absorption of laser dye A. CTP-5 has a large absorption coefficient at a optical pumping wavelength of 337 nm. This type energy transfer host-guest system is one of promising structures for efficient and low-threshold lasers because of its large effective Stokes shift consisted of the energy host absorption and the guest luminescence, the inhibition of concentration quenching, and so on.11) The structures of and notations for the chemicals used in this study are shown in Fig. 1.

2.2 Fabrication of Polymer DBR Laser

2.2.1 Softlithography

The preparation procedure of the polymer DBR laser device by softlithography is shown below. First of all, the original mold for the laser device was formed on a 100-nm-thick electron beam (EB) resist layer prepared on a Si wafer substrate by EB drawing. Next, degassed elastomer that was mixed well with a regulated catalyst was casted onto the original mold having minute resonator structure obtained by EB drawing. The casted elastomer spontaneously spreads to the whole substrate and it exactly covers the EB drawn pattern. Next, this wafer was heated for 15 minutes in an oven of which temperature was set at 150 °C. Then, the
elastomer was solidified and minute resonator structure was transferred to the contacted face to the original mold on the Si substrate, and we were able to obtained the elastic mold. Next, this elastomer mold was contacted to the surface of a prepared CPT-5 film with laser dye A on a glass substrate. The structure was transferred onto the surface of the polymer active layer by a thermal treatment at 150 °C, which was higher than the glass transition temperature of CTP-5. The elastomer mold was peeled away from the CTP-5 film after cooling down to room temperature. Then, we can obtain a precise replica of the original mold. The aforementioned preparation scheme is shown in Fig. 2.

The electron beam drawing apparatus (BEAM DRAW™, Tokyo Technology) and diluted ZEP-520 (ZEON) with o-dichlorobenzene at 2 times was used for the fabrication of the original mold. We used SILPOT® 184 W/C (DOW CORENING) for the preparation of the elastic mold.

2.2.2 Device assembling

The schematic illustration of the laser device that we made in this study is shown in Fig. 3. The procedure of the device preparation is as follows. At the beginning, the laser dye doped polymer film was prepared on glass substrate at the film thickness becomes about 130 nm by a spin-coating method. The film was deposited from the dichloromethane solution (solid content: 3 wt%) of CPT-5 and laser dye A at the mixing ratio of 10:1 in weight. Next, two 1.0-mm-width gratings of each length of 1.0 mm and 0.5 mm was formed as DBRs by softlithography in both ends of the flat range of the length of about 1.0 mm on the CPT-5 film doped with laser dye A. The period and depth of the grating are 600 nm and approximately 50 nm, respectively. The grating of 600 nm period feedbacks lights of oscillation wavelength by a fourth order diffraction condition. At last, we cut the glass substrate at near the end edge of DBR gratings and made it a measurement sample.

2.3 Measurement of polymer DBR laser characteristics

A nitrogen gas laser (MSG-800, Laser Teknik Berlin) that generated a 500-ps-width optical pulse at a repetition rate of 10 Hz was used for the optical pumping of the device. The excitation light from the nitrogen gas laser was shaped to a rectangular of 1 mm X 5 mm in area by convex lenses and cylindrical lens, and irradiated vertically to the device surface. Furthermore, the excitation light was only exposed to the active region by covering the grating part with a shadow mask. We used an...
optical density (OD) filter (ODC-WTO5, Asahi Spectra) in order to adjust the excitation energy. The emitted light was corrected at the device end edge, and then the light was analyzed with a computerized spectroscope (MS7504, SOLAR TII) with a charge coupled device camera (DU420-OE, Andor). High and normal resolution gratings, whose groove lines are respectively 1800 and 300 line/mm, were used. Their wavelength resolving powers are approximately 0.03 and 0.2 nm, respectively. We used a power meter (PEM100, Laser Teknik Berlin) in order to calibrate excitation energy. The setup of the optics system used for the spectroscopic measurement is shown in Fig. 4. We also confirmed that emission light was assigned to either TE or TM. The polarization directions of TE and TM modes were orthogonal each other. Therefore, the devices emission spectra that respectively have parallel or vertical polarization directions to the substrate face were measured through a polarizer.

3. Result and Discussion

The emission spectrum from the device is shown in Fig. 5. The extremely sharp peak that was not observed from the part without the resonator was observed at 509 nm from the part with the resonator that consists of a pair of gratings as DBRs. Even in the input-output characteristics of the device that was shown in Fig. 6, a kink point of the inclination of the increase of peak intensity was observed. Therefore, we are able to consider that the peak observed here was caused by a laser oscillation. The laser oscillation threshold of this polymer DBR laser that was obtained from the input-output characteristics in Fig. 6 was 37.6 µJ/cm². The laser oscillation threshold of the device was calculated from the point, where the regression line of the plots of the peak intensity of the sharp emission band meets to the axis of a peak intensity of 0.

A precise laser emission spectrum measured by using the high-resolution spectrooscope is shown into Fig. 7. As shown in Fig. 7, many laser lines were observed around 509 nm. Full width at half maximum (FWHM) of a laser line and the averaged line interval are respectively 0.028 nm (0.13 eV) and 0.12 nm. From this FWHM, the Q-value is calculated with approximately 9200. As also shown in the figure, the extent width of the longitudinal lights was approximately 0.5 nm, namely, 19.5 cm⁻¹ in wavenumber. The extent width in wavenumber is probably equal to the stop-bandwidth (Δν) of the...
DBR. From this value, we solved that the coupling coefficient ($\kappa$) of the DBR used in this study was $1.0 \times 10^{-2}$ $\mu m^{-1}$ from the following equation, $^{12}$

$$\kappa = \frac{\pi n_{\text{eff}} \Delta V}{c}$$ (1)

The spectra of the laser that observed through a polarizer are shown in Fig. 8. The intensity ratio $I_{II}/I_{II}$ of the both polarized spectrum, namely, parallel and perpendicular to the device substrate, was about 100. Furthermore, the positions of the peaks of both two polarized spectra were completely same. In addition, there is some errors in accuracy of the polarizer rotation. Therefore, in fact, it is conceivable that each peak observed in both polarized spectra is assigned to the same transversal mode. From over, we identified the transversal mode of the laser observed in this study is TE that has the oscillation of a parallel electric field to the device substrate.

4. Conclusion

We fabricated the polymer laser that consists of a pair of DBR as reflectors on the laser dye doped charge transport polymer layer by soft lithography. And we observed laser oscillation from the device. The lasing threshold value of this laser device was 37.6 $\mu J/cm^2$. From FWHM of 0.028 nm, the Q-value of our device was approximately 9200. From the emission spectrum measurement with a polarizer, the laser emission observed from our device was assigned to a TE mode.

Unlike DFB laser structure, as for DBR laser structure the active region is flat. Therefore, the charge carrier can be injected homogeneously at the time of the current operation. Therefore, the performance of an OLED as an active region can be almost maximized. Also, with different from DFB laser, the photon density in the active region becomes homogeneous in DBR lasers, because the active region is separated from the feedback region. Therefore, the DBR type device can be probably operated with high energy efficiency.

Soft lithography used in this study needs not to use etching and physical shaving and other severe processing methods. Therefore, the calm processing suited for the organic polymer is possible. Therefore, this is also the effective method for fabricating a laser light source on polymer light circuits.
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