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Introducing optical resonators into polymer light-emitting electrochemical cells

Tingwu Zhang, Tomo Sakanoue and Taishi Takenobu

Department of Applied Physics, Nagoya University
Furo-cho Chikusa-ku Nagoya, Aichi 464-8603, Japan
Phone: +81-52-789-5165 E-mail: zhang.tingwu@e.mbox.nagoya-u.ac.jp

Abstract

2nd-order distributed feedback (DFB) resonator was introduced to the emitting layer of light-emitting electrochemical cells (LECs) that consist of a film blend of a light-emitting polymer and an ionic liquid. The lasing characteristics under optical pumping and the emission characteristics under electrical pumping were investigated for the potential use of LECs with DFB resonators for future electrically pumped polymer lasers.

1. Introduction

Organic semiconductors including semiconducting polymers have been investigated intensively because of their excellent optoelectronic properties. Since these materials exhibit lasing in thin films by optical pumping, there is a strong expectation of lasing by electrical pumping; however, this has become one of the most difficult issues in the field of organic semiconductor devices as a lot of issues have been found. For example, it is necessary to achieve high exciton density enough for light amplification. Although it is rather easy to achieve this by optical pumping, it is difficult by electrical pumping since the optical losses under high current density are found to be huge due to the exciton annihilation by charge carrier, triplet exciton and heat. Thus, it is important to minimize these losses and maximize the optical gain. One of the promising solutions for this issue is to introduce optical resonators into organic films. Distributed feedback (DFB) resonator, in which polymer thin film is prepared on diffraction grating structure (Fig. 1), is intensively studied as one of the feasible resonators for lowering the lasing threshold [1]. However, there is an issue for electrical pumping; ~100-nm-sized corrugated structure of DFB resonator is too large for introducing to the conventional organic light-emitting diodes (OLEDs) as the thickness of the emitting layers are typically ~100 nm scale.

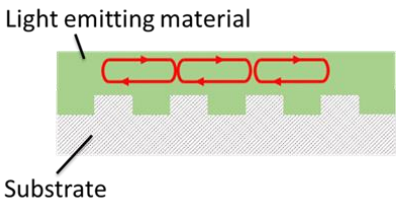

Fig.1 Structure of the DFB resonator incorporated in polymer thin films.

To address this difficulty, in this work, we employed an alternative light emitting device of light-emitting electrochemical cells (LECs) to excite DFB structure by electrical pumping. One of the advantages of LEC is flexibility for device designing: not only the sandwich structure like OLEDs, but also planar structure can be designed [2]. This allowed us to introduce DFB structures easily for electrical pumping. Furthermore, we have already reported that LECs achieved >1 kA/cm^2 of high current density that is important for achieving high exciton density [3]. Therefore, we think that LECs have advantages both for introducing DFB resonators and for electrical pumping. Here, DFB resonators were introduced to an emitting layer of a LEC, a film blend of a light-emitting polymer of poly(9, 9-dioctylfluorene-alt-bithiophene) (F8T2) and an ionic liquid of triheyltetradecylphosphonium bis (trifluoromethylsulfonyl) imide (P_{66614}-TFSI), and their lasing characteristics were investigated by optical pumping. The structure was further excited electrically and the electroluminescent (EL) spectrum was investigated.

2. Experiments

DFB structures were introduced by spin-coating F8T2 or F8T2:P_{66614}-TFSI blend solutions on a diffraction grating prepared on a quartz substrate with 340 nm periodic corrugated structure. The samples were optically pumped by a pulsed dye laser having 460 nm of wavelength that was pumped with N_2 gas laser. The light emission perpendicular to the substrate was collected and the photoluminescence (PL) and lasing properties were investigated. For electrical excitation, Au electrodes were deposited on a DFB structure consists of F8T2:P_{66614}-TFSI blend (Fig.2), of which the blend ratio was controlled to be F8T2:P_{66614}-TFSI = 15:1.

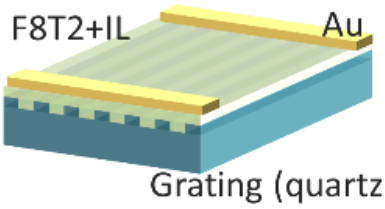

Fig. 2 Device structure of the LEC with DFB resonator.

2. Results and Discussion

In order to check whether the grating structure can work as a DFB resonator with F8T2, we first prepared a DFB laser with neat F8T2 film. As shown in Fig.3 (a), by exciting the DFB structures strongly by optical pumping, we observed clear lasing characteristics with a feature of DFB resonators: the lasing wavelength and the lasing thresholds were varied with the film thickness. The thickness dependence of lasing wavelength are explained by the Bragg's

equations:

$$m\lambda_m = 2n_{eff}\Lambda$$

in which m is the order of the DFB resonator, n_{eff} is the effective refractive index, and Λ is the period of the DFB structure. The lasing wavelength is proportional to n_{eff}, which is determined by the volume fraction of the materials composed of the periodic structure, so that the thick film of 90 nm gives the lasing with the longest wavelength, while the thickness of 55 nm gave the shortest wavelength. More importantly, as shown in Table I, the lasing threshold was very sensitive to the film thicknesses; lasing thresholds for 60-nm- and 75-nm-thick films were 3.9 μJ/cm^2, while that of 55 nm and 90 nm were 35 μJ/cm^2. Because the intrinsic PL peak of F8T2 film is approximately 550 nm, these results indicate that there are strong relationship between lasing threshold and F8T2 intrinsic peak wavelength (550 nm). Simply, the cavity mode close to 550 nm, such as 545 nm of 60-nm thickness and 557 nm of 75-nm thickness films, leads to higher PLQE, resulting in smaller lasing threshold. As the lasing wavelength can be controlled by the film thickness, we can conclude that precise control of film thickness is necessary to minimize lasing threshold.

We then carried out the optical excitation on the active layer of LECs, i.e., a film blend of F8T2 and P_{66614}-TFSI having 90 nm of film thickness. We found that even with the ionic liquid in the F8T2, lasing behavior was obtained (Fig.3). This suggests that the ionic liquid is not a cause of optical loss. However, we found the lasing wavelength was different from that in the neat films. This is probably because the refractive index of the film blend is smaller than the neat film of F8T2; the smaller n_{eff} makes the resonance wavelength blue shift as derived from Bragg equation.

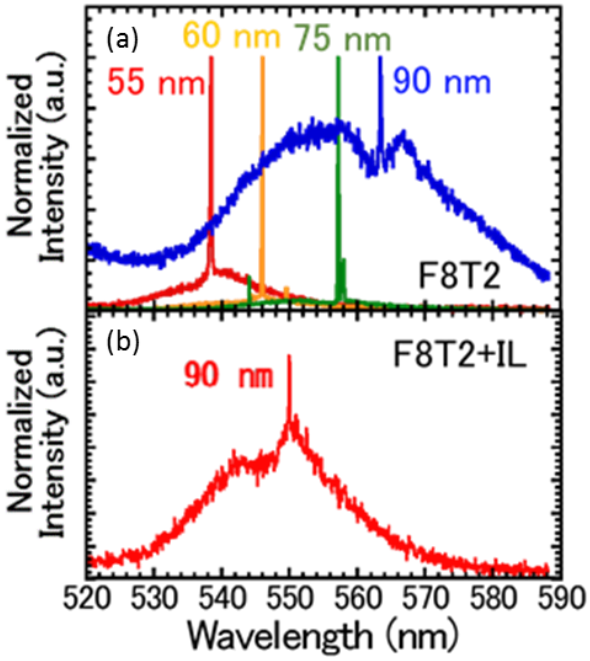

Fig3. Lasing spectrum from (a) neat F8T2 films with various film thickness and (b) film blend of F8T2 and ionic liquid.

Table I Thickness dependence of lasing characteristics.

Film Thickness (nm)	55	60	75	90
Lasing Threshold (μJ/cm^2)	35	3.9	3.9	35
Lasing Wavelength (nm)	538	545	557	563

Finally, we excited the DFB structure by electrical pumping by applying the voltage to the Au contacts. We successfully observed EL spectrum modified by DFB resonator (Fig.4). The linewidth of EL spectrum with a DFB resonator was narrower than that of the PL spectrum from a film blend without DFB resonator. This indicates that light confinement effect was successfully achieved by the DFB resonator, indicating the resonator also worked well by electrical pumping. Considering the result that optically pumped lasing from the film blend was achieved, electrically pumped lasing can be expected by injecting high career density into the LECs with DFB structure.

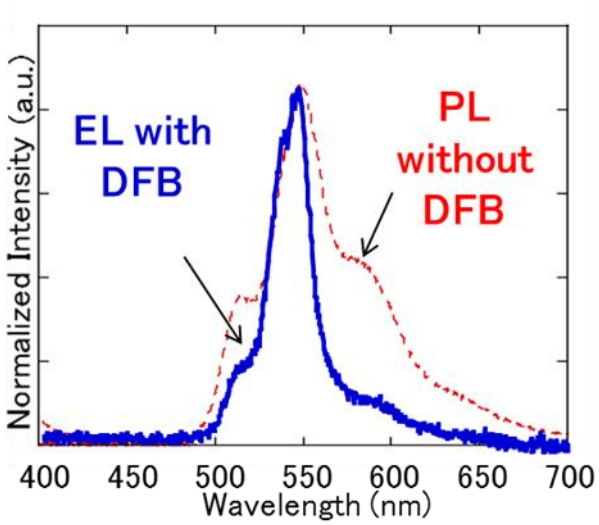

Fig4. EL spectrum of LEC with a DFB resonator. PL spectrum without DFB is also shown for comparison.

3. Conclusions

Optically pumped DFB laser was achieved in thin film of F8T2 and film blend of F8T2:P_{66614}-TFSI. The control of the film thickness is very important for lowering lasing threshold. The DFB resonator was further contributed for narrowing the emission spectrum by electrical pumping. We think that these demonstrations of the optically pumped DFB laser from the active layer blend of the LEC and the light confinement effect under electrical pumping indicates the potential advantage of using LECs with DFB resonators for future electrically pumped polymer laser. The high current density injection into the LEC with optimized DFB structure will be further considered.

References

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[3] T. Sakanoue et al., Adv. Mater, (2017), published online