

Studies on device architecture and materials system for lowering the laser threshold in organic semiconductor lasers

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論 文 名 : Studies on device architecture and materials system for lowering the laser threshold in organic semiconductor lasers

(有機半導体レーザーの低閾値化のためのデバイス構造及び材料系の検討)

区 分 : 甲

論 文 内 容 の 要 旨

Thesis summary:

Organic semiconductor lasers (OSLs) have many different applications such as spectroscopy, sensing, and data communications due to their unique properties including color tunability over visible to NIR range, low-cost fabrication, and mechanical flexibility. Since the first demonstration of lasing in organic materials in the early 60s, the OSLs have been developed significantly. However, the laser operation under a continuous-wave (CW) regime and a low current density threshold in organic semiconductor laser diodes (OSLDs) have remained a big challenge in the field of organic lasers. Despite the development of organic gain materials and optical resonators to minimize the losses and maximize the laser efficiency, the current density required for lasing is very high, resulting in few reports on the indication of OSLDs. This challenge was a trigger for this work. In this thesis, with the aim of the realization of OSLD with a low current density threshold, I studied the laser dynamics and sought feasible ways to reduce the laser threshold and increase the efficiency.

In **Chapter 1**, I explained the general introduction of the lasers and briefly described the main components of a laser system including gain medium, optical resonator, and pump excitation. In this chapter, I focused on the OSLs by explaining the history of OSL developments as well as organic gain materials, including conjugated polymers and small molecules. The light amplification properties including stimulated emission in the organic materials via ASE measurements were explained. Finally, by describing the OSLDs and their challenges, I explained the aim of this work.

In **Chapter 2**, the dynamics of an OSL/OSLD were studied to investigate the impact of various loss mechanisms on the laser threshold and slope efficiency. For this study, the coupled rate equations were numerically solved by the Euler method in a three-level energy system approximation. Different excitonic and photonic loss mechanisms were considered individually to avoid the complexity of interpreting the results due to the correlation of more than one loss mechanism at a time. The simulation results showed that excitonic loss mechanisms only increase the laser threshold due to singlet exciton quenching. In these mechanisms, slope efficiency remained unchanged. It should be noted that, only in the STA process under electrical

excitation, due to different generation mechanisms of triplet excitons compared to the optical excitation, an increase in the k_{STA} not only increases the laser threshold but also decreases the slope efficiency. On the other hand, the photonic loss processes increase the laser threshold due to the singlet exciton quenching and reduce the slope efficiency by absorbing the emitted photons. Also, the dynamics of an OSL/OSLD composed of a TADF emitter were studied based on DABNA-2 parameters to investigate the contribution of triplet excitons in the laser emission. The results confirmed the triplet exciton contribution in lasing through fast RISC with a high k_{RISC} .

In **Chapter 3**, the charge carrier injection and transport in semi-transparent OLEDs composed of DMD electrodes were studied in the standard and inverse structures. The electron injection was improved in the standard structure using a DMD cathode doped by the Cs as an n-type dopant in one of the dielectric layers. The DMD cathode was composed of Cs:MoO₃ (5 nm)/Ag (20 nm)/ MoO₃ (22 nm). Further, the DMD anode in the inverse OLED was modified by inserting a HATCN layer between Ag and MoO₃ layers to enhance the hole injection property. This Anode was composed of MoO₃ (5 nm)/HATCN (10 nm)/ Ag (20 nm)/ MoO₃ (22 nm). Based on the results, the inverse structure showed well-balanced charge carrier densities inside the emitting layer leading to high and stable EQE compared to the standard structure.

In **Chapter 4**, based on simulation results of TADF-based lasers in Chapter 2, two TADF emitters with fast k_{RISC} were selected to utilize as a laser gain medium. Despite having good TADF properties, the ASE properties were not observed in these materials. Further, to benefit good properties of these TADF emitters such as high PLQY, high k_{RISC} and short delayed lifetime which are beneficial in laser emission based on harvesting the triplet excitons, it seems promising to use them in a TAF system. I expect that, with this TAF system, both laser threshold and stability will improve. The TAF system including MCz-XT/MCz-TXT as a TADF assistant dopant and mDMCz as a fluorescent emitter in the mCBP as a host matrix was considered to investigate the ASE properties. The results showed a reduction of the ASE threshold in the proposed TAF system compared to the mDMCz:mCBP blend film from 26 $\mu\text{J cm}^{-2}$ to <10 $\mu\text{J cm}^{-2}$. This indicates the successful contribution of recycled triplet excitons to the emission by using TADF materials with fast k_{RISC} as an assistant dopant.

In **Chapter 5**, I summarized the findings of each chapter and concluded the thesis. Then, I explained my prospect for this research. The main goal of this thesis was the indication of the OSLD with a low current density threshold which is a great challenge in organic lasers. For future work, the TAF system proposed in this thesis will be optimized to maximize the contribution of triplet excitons in the laser emission by using a variety of fluorescent laser dyes with a large overlap between absorption of fluorescent dye and emission of TADF assistant dopants. Also, the inverse OLED composed of DMD will be optimized to adapt to this TAF system. Finally, I proposed to apply them in an optical resonator such as distributed feedback (DFB) or vertical-cavity surface-emitting laser (VCSEL) to indicate an OSLD.