

Improvement of device performances in TADF OLEDs and organic semiconductor lasers

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論 文 内 容 の 要 旨

In this thesis, I studied on different aspects that are essential for an improvement of the performances of TADF OLEDs and organic semiconductor lasers. In particular, the objectives of this thesis were: (i) to investigate the role of the molecular structure on the horizontal molecular orientation of TADF emitters in vapor-deposited thin films and to examine its impact on the OLED performances, (ii) to study the photophysical properties of novel BF₂ curcuminoid derivatives and to evaluate their potential for high efficiency TADF NIR OLEDs, (iii) to characterize the ASE properties of these curcuminoid derivatives and to demonstrate the possibility to observe light amplification due to stimulated emission in TADF materials, and (iv) to demonstrate the lowest ASE threshold reported so far in small molecule/oligomer based thin films.

In Chapter 2, I investigated the molecular orientation and the TADF properties of four novel carbazole-based emitters with different shapes and degrees of planarity in vapor-deposited thin films. Molecular orientation of the TADF molecules in neat film and in blend was investigated using VASE and angle dependent PL measurements. The results demonstrated that the most planar molecules present the best horizontal molecular orientation. The photophysical and electroluminescence properties of these TADF molecules were then investigated to examine the role of the molecular orientation on the TADF OLED performances. The most efficient device was obtained with the TADF emitter exhibiting a nearly perfect horizontal orientation of the emitting dipoles in the blend films. The maximum EQE value measured in this device was found to be 15.4%, which is about 1.75 times larger than the theoretical value determined using a random distribution of emitting dipoles. Overall, this work provides useful insights into the role of the TADF emitter structure on the molecular orientation and the electroluminescence properties of vapor-deposited TADF OLEDs. These results should serve as guidelines in the near future to develop novel TADF molecules allaying excellent photophysical properties with perfect horizontal molecular orientation.

In Chapter 3, I focused on BF₂ curcuminoid derivatives containing triphenylamine donor groups

(TPA-CBD(mH), TPA-CBD(mBu), TPA-CBD(mEster) and TPA-CBD(TPA-CBD)) and clarified unprecedented PL characteristics in the NIR region. While the molecules do not present the usual disconnected donor-acceptor structures, those organic dyes showed TADF behavior. In particular, I evidenced that both monomeric and dimeric molecules in the CBP doped films showed efficient NIR light with TADF activity. Furthermore, because of their high ground state dipole moment and their CT characteristics, those dyes showed PL that are solvatochromic in CBP doped films, allowing the fluorescence maximum from 700 to 790 nm by increasing the dopant. In addition, the high PLQY of 65% was obtained in the NIR region. Such high PLQY associated to TADF process allowed the fabrication of NIR OLEDs with EQE reaching to 9.7% with the emission maximum at 739 nm (for 6wt%-TPA-CBD(mEster):CBP), and 5.1% at 758 nm and 3.1% at 773 nm (for 2wt% and 10wt%, TPA-CBD(mTPA-CBD):CBP, respectively). Such high EQE represents the highest efficiencies ever reported for NIR-OLEDs using organic molecules.

In Chapter 4, I studied the ASE properties of two novel BF₂ curcuminoid derivatives, namely TPA-CBD(mEster) and TPA-CBD(mTPA-CBD), in spin-coated thin films. These two TADF NIR-emitting compounds were blended into a CBP host at different doping concentrations. In good consistency with the behavior of the steady-state PL spectra observed in Chapter 3, the films exhibited ASE activity with an ASE peak redshifted from 740 to 860 nm as the doping concentration was gradually increased from 4wt% to 60wt%. In addition, I found that the ASE thresholds decreased from 91 to 14.8 $\mu\text{J}/\text{cm}^2$ with the doping concentration, which could be explained by the concentration dependence of the PLQY values. Overall, this investigation has led to (i) the first demonstration of light amplification due to stimulated emission in BF₂ curcuminoid derivatives, (ii) the first demonstration of NIR ASE in an organic semiconductor thin films not based on lanthanide complexes, (iii) the demonstration that lasing can be achieved using TADF emitters, suggesting a new possible way to harvest all singlet and triplet excitons for light amplification in future electrically-pumped organic laser devices. In the near future, further studies involving CW ASE/lasing experiments should be carried out to see if triplets can be harvested in these systems to contribute to light amplification.

In Chapter 5, I studied the photophysical, ASE and electroluminescence properties of a blue-emitting solution-processable octafluorene derivative in spin-coated films. The neat film did not show any concentration quenching of the PL emission and was found to exhibit a PLQY of 87% together with a large radiative decay rate of $1.7 \times 10^9 \text{ s}^{-1}$. In addition, I examined the thickness dependence of the ASE properties of the octafluorene neat films. The results demonstrated that the lowest threshold of 90 nJ/cm^2 was achieved for a film thickness of 260 nm. Such a value corresponds to the lowest ASE threshold ever reported in small molecule/oligomer based thin films. Low-threshold mixed order organic DFB lasers based on octafluorene neat films and fluorescent OLEDs with a maximum EQE value of 4.4% are then fabricated, demonstrating that this octafluorene derivative is an excellent and very promising candidate for organic semiconductor laser applications. Future works should be carried out in the near future to explore the possibilities to achieve continuous-wave lasing in octafluorene DFB lasers and to inject high current densities into octafluorene based OLEDs via a more appropriate and improved device architecture.