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Harvesting triplet excitons in high mobility emissive semiconductor through triplet-triplet annihilation

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Abstract:

Organic light-emitting transistor (OLET) is an emerging integrated device which combine dual functions of switching and light-emitting, showing great potential applications in smart display and organic electrically pumped laser. Many high carrier mobility emissive semiconductors have been developed for constructing OLET. However, one issue which limits the efficiency of OLET is the insufficient utilization of 75% triplet excitons after the recombination of electrons and holes according to the spin statistics, which only 25% singlets can decay radiatively to the ground state for light emission. Herein, we demonstrated the possibility of integrating high carrier mobility and high triplet utilization property in the representative high mobility emissive 2,6-diphenylanthracene (DPA). DPA possesses triplet-triplet annihilation property which can convert two triplets to one emissive singlet. The photoluminescence emission efficiency of DPA film and external quantum efficiency of DPA film based OLET have been increased significantly by doping with a highly emissive guest coumarin 6 (C6) via energy transfer. The highest exciton utilization efficiency in OLET has been increased more than 2.4 times in C6-doped DPA film compared to that of DPA film. The concept presented here can be applied to other advanced high mobility anthracene derivatives to realize high performance and multifunctional OLET devices.

Keywords: high mobility emissive semiconductor, triplet exciton, triplet-triplet annihilation, organic light-emitting transistor

Organic light-emitting transistor (OLET) has attracted increasing attention in recent decades which combine dual abilities in electrical switching as organic field effect transistor (OFET) and light emission as organic light emitting diode (OLED) in a single device.[1-3] This unique integrated device architecture of OLET provides an ideal platform for studying the fundamental optoelectronic properties of organic semiconductors, showing great potentials in integrated electronics, smart display, organic electrically pumped lasers, *etc*.[4-8] For constructing high performance OLET, the active layer material requires to integrate high carrier mobility and strong solid-state emission properties to enable high current density and efficient electrical-to-optical conversion efficiency. Thanks to the continuous efforts made in recent years, a series of high mobility emissive organic semiconductors have emerged which greatly facilitate the development of OLET and its relative applications.[9-14] However, materials currently utilized for constructing OLET are mainly fluorescent materials, which can only harvest 25% singlet excitons generated after the recombination of electrons and holes according to the spin statistics, while the 75% triplets are wasted as nonradiative decay. Thus, harvesting triplet excitons plays an important role to improve the overall efficiency of OLET.

Until now, less attention has been paid in harvesting triplet excitons in OLET and there are only few reports of harvesting triplet excitons in OLET. Recently, Namdas *et al.*, Liu *et al.* and Meng *et al.* have carried out few pioneering works by using phosphorescent and thermally activated delayed fluorescent (TADF) materials to utilize triplet excitons in multiple-layer OLET.[15-22] The external quantum efficiency (EQE) and brightness of OLET have been improved significantly, which exhibit their promise for applications in large area display and other integrated optoelectronic devices.[22] Nevertheless, the phosphorescent materials usually contain heavy metals which caused high cost, while TADF molecules are hard to integrate the high carrier mobility and strong solid-state emission due to their inherent steric molecular structures.

From the aspects of simplifying the device architecture, single-layer OLET is more desirable for low-cost fabrication and minimizing the reduction of optical loss induced by multilayer for higher efficiencies. Therefore, apart from the high carrier mobility and strong solid-state emission properties, ideal active layer material for constructing high performance single-layer OLET also needs to have the capability of harvesting triplet excitons for light emission. Amongst several organic materials which can harvest triplets, triplet-triplet annihilation (TTA) is a process of generating one emissive singlet exciton after two triplets annihilate.[23, 24] Organic TTA materials, such as anthracene, pyrene, rubrene, and their derivatives, have been widely used in OLED for realizing highly efficient and stable electroluminescence.[25, 26] The large conjugation molecular structure of these polycyclic aromatic hydrocarbon acenes enables them to show the possibility to integrate the high carrier mobility and TTA properties in one molecule. The discovery of high carrier mobility TTA materials is essential for the construction of high performance OLET devices. Therefore, considering anthracene derivatives with high carrier mobility and emissive property developed in recent years, the study of their triplet exciton interaction mechanism and utilization efficiency is significantly meaningful.[9, 11, 12, 14, 27, 28]

In this work, one representative high mobility emissive molecule 2,6-dipenylanthracene (DPA) was selected as an example to study the triplet utilization property in high mobility organic semiconductors. The results showed that DPA has TTA property, which can convert triplets to emissive singlets through TTA. In order to improve the photoluminescence quantum yield (PLQY) of DPA film, Coumarin 6 (C6) was selected as the guest to be doped in DPA film. The PLQY was significantly improved after doping which was mainly attributed to the suitable energy level between DPA and C6, and the efficient Förster resonance energy transfer (FRET) efficiency. As a result, the EQE of OLET device based on coumarin6-doped DPA film was more than 7 times higher compared to that of DPA film. The exciton utilization efficiency was improved more than 2.4 times through molecular doping approach. The study of TTA

property in DPA has successfully confirmed the possibility of integration of high carrier mobility and triplet exciton utilization capability. Molecular doping utilized in this work also provided an efficient strategy for the realization of high performance OLET devices, which is crucial for promoting the application of OLET in other related areas.

Previously, a design concept of introducing functional groups on emissive conjugated skeleton via C-C single bond was proposed in our group for designing high mobility emissive anthracene derivatives (Figure S1).[9, 11, 14] This strategy has also been successfully used for developing high mobility lasing organic semiconductors.[29, 30] Among them, 2,6-diphenylantracene (DPA) is a representative high mobility emissive molecule (Figure 1a), which has been successfully utilized for constructing single component OLET.[6] However, the triplet exciton conversion mechanism and utilization efficiency has not been studied. Singlet and triplet energy levels of DPA were calculated based on time-dependent density functional theory (TD-DFT). The calculation results show that DPA has a singlet energy level of 3.12 eV, which is slightly lower than two times of triplet energy level (1.63 eV) (Figure 1b). Therefore, DPA can convert two triplets into one emissive singlet through TTA theoretically. Direct evidence of TTA property of DPA molecules was demonstrated by the obvious upconverted emission recorded under the sensitization of platinum octaethylporphyrin (PtOEP) both in dilute solution and films. PtOEP was selected as the triplet sensitizer due to its suitable triplet energy level of 1.93 eV (Figure 1b and 1c) compared to that of DPA and its nearly quantitively efficient intersystem crossing (ISC) to generate triplet excitons. Under optical excitation, sensitizer assisted TTA process was illustrated in Figure S2. Firstly, the sensitizer can be excited

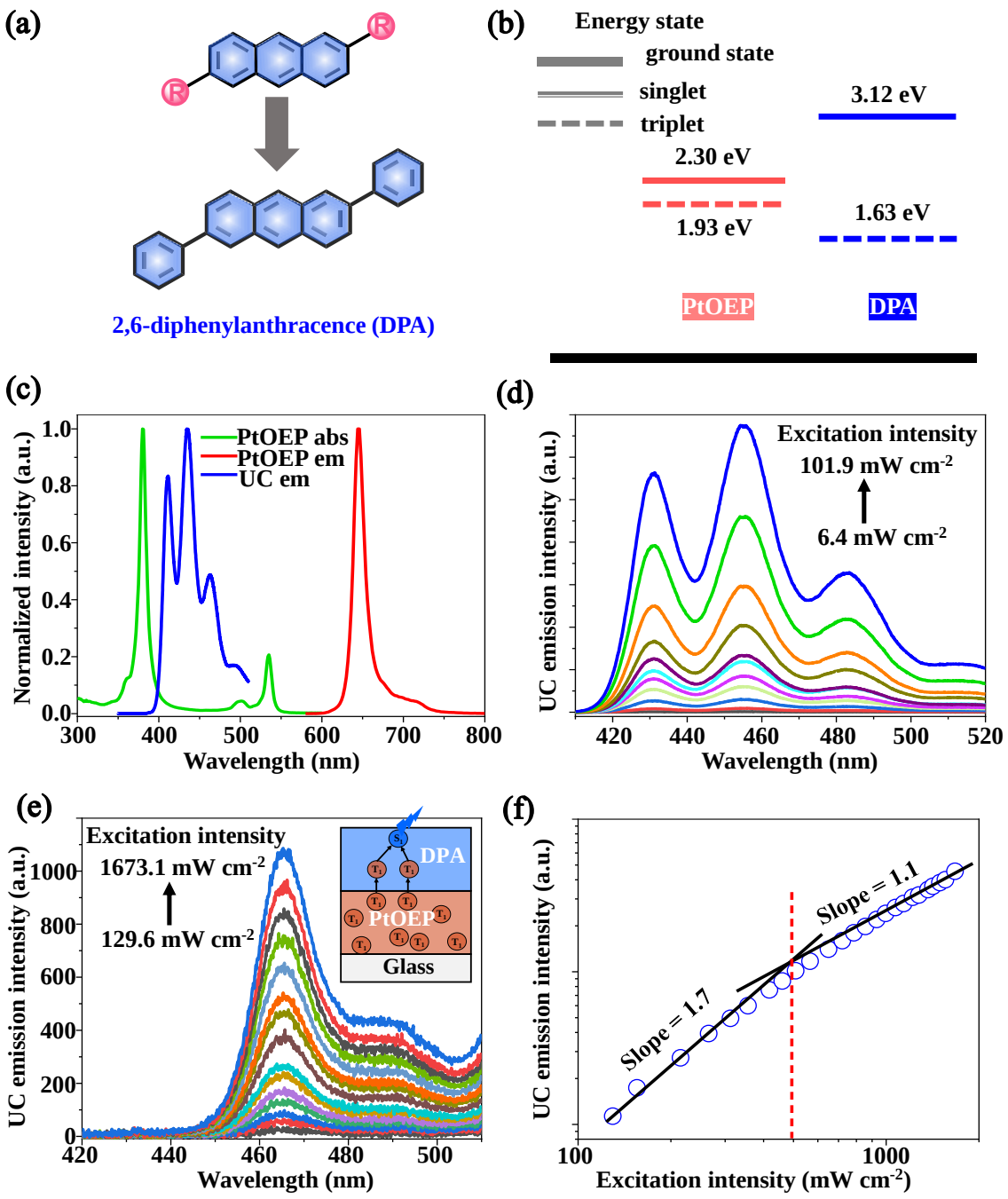

Figure 1. (a) Molecular structure of high mobility emissive semiconductor DPA. (b) Energy diagram of triplet sensitizer (PtOEP) and DPA; (c) Steady-state absorption and emission spctra of PtOEP in dilute THF solution, the upconverted emssion from DPA under the sensitization of PtOEP; (d) Upconverted emission spectra of DPA in PtOEP/DPA mixed solution in terms of excitation intensity at 532 nm; (e) Upconverted emission spectra of DPA in PtOEP/DPA deposite film in terms of excitation intensity at 532 nm; (f) Non-linear property of DPA in PtOEP/DPA deposite film under optical excitation.

to its singlet excited-state under optical excitation, consequently, triplet excited-state is generated quickly through efficient ISC. The energy in triplet excited-state sensitizer is transferred to an emitter molecule through triplet-triplet energy transfer (TTET) to form triplet species. Following, two or more triplets in emitters can interact to produce one emitter in its ground state and one in the singlet excited-state. Finally, the singlet excited-state of the emitter decays radiatively back to its ground state by producing a photon with higher energy than those initially absorbed by the sensitizer molecule. As shown in Figure 1c, by exciting the Q-band of PtOEP at 532 nm, blue upconverted emission through TTA was observed from DPA. The sensitized TTA upconverted emission intensity from DPA increased as increasing the excitation power intensity at 532 nm both in dilute solution and thin film (Figure 1d and 1e). Meanwhile, the sensitized TTA upconverted emission intensity from DPA exhibited representative non-linear property in terms of the excitation intensity (Figure 1f and Figure S3).[24, 25] These results successfully confirmed the integration of high carrier mobility and TTA property in DPA molecule.

The integration of high carrier mobility, emissive and TTA properties in DPA makes it a suitable alternative for constructing single layer OLET. According to the EQE equation (EQE = $\eta_{ph} \times \gamma \times \Phi \times \eta_{ex}$) of OLET, the device efficiency is ralated to the light out-coupling efficiency (η_{ph}), the ratio of electrons to holes (γ), the PLQY (Φ) and the utilization efficiency of excitons of the emissive layer (η_{ex}).[3, 31] Similar to OLED devices, 25% singlet and 75% triplets are generated after electron and hole recombination in OLET based on the spin statistics. Therefore, the efficiency of OLETs is highly related to the utilization efficiency of non-radiative triplet excitons and PLQY of the active layer. However, the PLQY of DPA film is relatively low, which cannot satisfy the requirements for high performance OLET. To address this issue, an approach used in OLED can be leant that using TTA material as the host and doped with a highly emissive guest material, which is called molecular doping.[32-34] Molecular doping is an efficient strategy to integrate outstanding optoelectrical properties in one system.[35-39] Doping

a highly emissive guest into a high mobility TTA material has its intrisic advantages, such as increasing the triplet utilization efficiency and PLQY of active layer, improving the device stability and overall efficiency.

Herein, to further increase the PLQY of high mobility emissive DPA and harvest the triplet exciton utilization efficiency, a host-guest system was prepared for constructing high performance OLET. The energy transfer and triplet exciton harvesting process are illustrated in Figure 2a. Firstly, charge carriers are transported in high mobility host DPA and subsequently singlets and triplets are generated in DPA after the electron and hole recombination. Triplets in DPA can be converted to singlets through TTA. Following, the singlet excitons are transfered to a highly emissive guest molecule to emit light with a high PLQY through FRET. Accordingly, C6 was selected as the guest to be doped in DPA films due to its highly emissive and amplified spontaneous emission (ASE) properties.[40, 41] C6-doped DPA films were prepared with different doping concentrations from 10% to 2% via deposition under vacumm, the preparation details are shown in Experimental Section and Table S1. The PL emission of C6-doped DPA films are changing from blue to green from the doping ratio of 2% to 10% (Figure 2b), which is due to the different FRET efficiency at different doping concentrations. The crystallinity of these C6-doped DPA films is getting more obvious from the doping ratio of 2% to 10% (Figure 2c and Figure S4). This is mainly attributed to the decreasing deposition rate of DPA from 2.5 Å/s to 0.5 Å/s (Table S1).

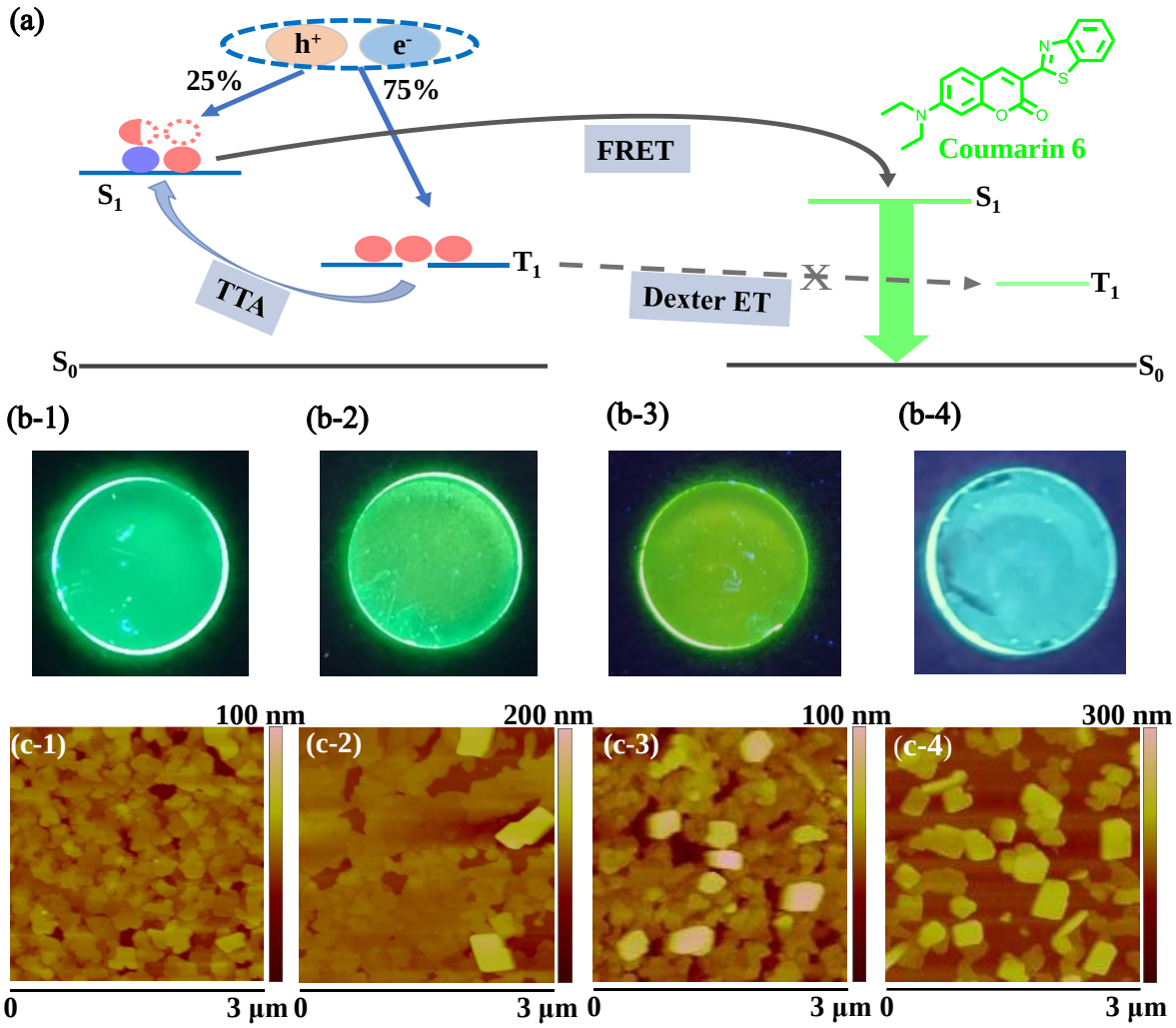

Figure 2. (a) Schematic illustration of triplet harvesting and FRET energy transfer processes with molecular structure of C6. Fluorescent pictures of C6-doped DPA films at doping concentrations of (b-1) 2%, (b-2) 5%, (b-3) 7%, (b-4) 10%. AFM images of C6-doped DPA films on quartz substrate with different ratios, (c-1) 2% doping, (c-2) 5% doping, (c-3) 7% doping, (c-4) 10% doping.

As shown in Figure 3a, C6 shows a suitable singlet energy level with DPA according to the large overlap of its absoprtion spectrum with the emission spectra of DPA, which enables the efficient FRET from DPA to C6. Interestingly, the normalized emission intensity of DPA dropped dramatically by decreasing the doped C6 concentration with the appearance of dopant C6 emission (Figure 3b). This is mainly due to the molecular aggregation of C6 at high doping

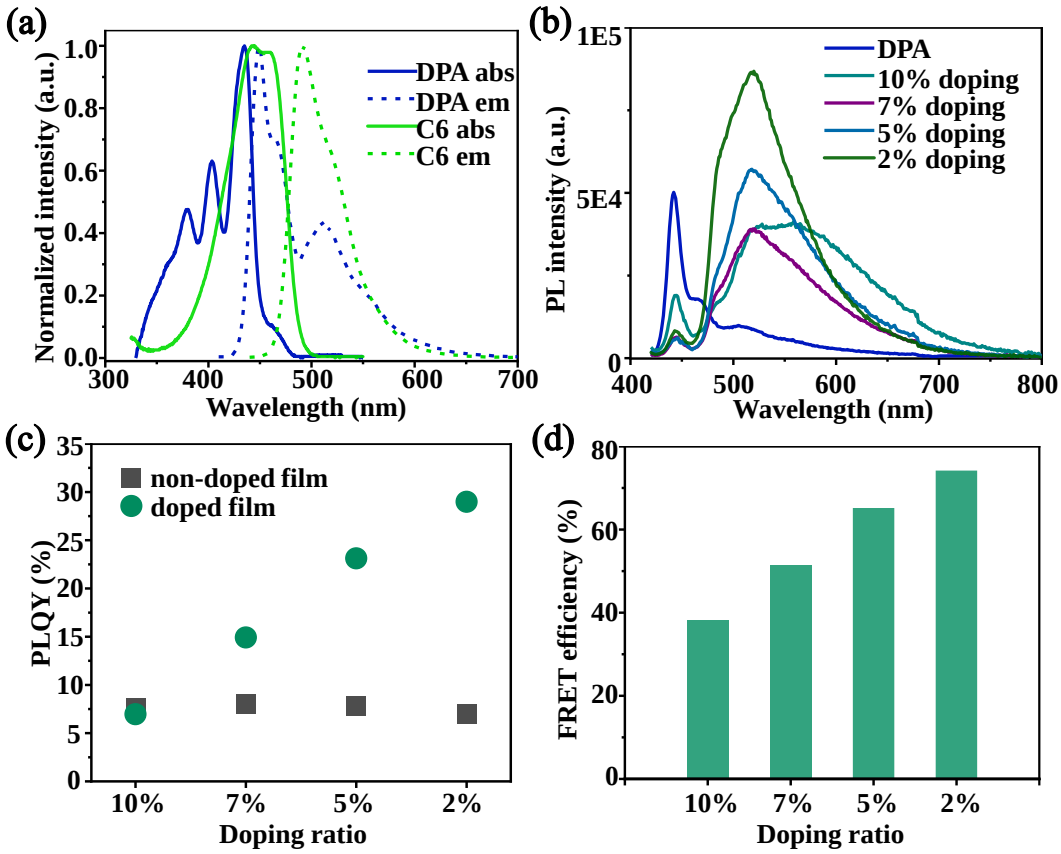

Figure 3. (a) Absorption and emission spectra of DPA film (blue) and C6 dilute solution (green). (b) PL spectra of DPA film and C6-doped DPA films at different doping ratio. (c) PLQY of DPA film and C6-doped DPA films. (d) FRET efficiency of DPA to C6 in different C6-doped DPA films.

concentration which causes the low FRET efficiency and low PLQY of C6. At the C6 doping concentration of 2%, the emission from host DPA is almost disapeared and mainly shows the green emission from C6 (Figure 3b and Figure 2b). For non-doped DPA film, the PLQY is only around 7.6% (Figure 3c). After molecular doping, C6-doped DPA film show significantly increased PLQY and the highest one reaches to 29%, which is about 4 times higher compared to that of DPA film (Figure 3c and Table S1). The highest FRET efficiency is nearly 80% at the doping ratio of 2% (Figure 3d). High PLQY of C6-doped DPA was achieved at low doping concentration which provide the advantage for retaining the high carrier mobility property in

host DPA. The successful integration of high carrier mobiliy and high PLQY in C6-doped DPA film makes it the suitable active material for constraucting high performance OLET device.

Different doping concentrations of C6-doped DPA films were used to fabricate planar OLET devices with a comparison to that of non-doped DPA based OLET. Au/MoO_3 and Ca/CsF were selected as the non-symmetric electrodes for achieving efficient electron injection (Figure 4a and Figure S5). Non-doped DPA film-based OLET device shows typical blue emission, while the 7% and 5% coumarin6-doped DPA films based OLET show green emission from C6, which further confirmed the efficient FRET from DPA to C6 (Figure 4b and 4e). In 2% C6-doped DPA film based OLET, the electroluminescence is close to white, which is an integration of blue emission from DPA and green emission from C6 (Figure 4b and 4e). However, the 10% C6-doped DPA films based OLET did not work which may be attributed to the high C6 doping concentration hindered the carrier transport ability of DPA. In this case, the electrons injected from the electrodes cannot transport for electron and hole recombination in 10% C6-doped DPA films based OLET. The typical transfer and output curves of DPA and C6-doped DPA films based OLET devices are shown in Figure 4c, 4d and Figure S6, which demonstrate the well operation of these devices. All the OLET devices show good gate tuneable property. In comparison to the EQE of non-doped DPA film based OLET, C6-doped DPA films based OLET devices show significant increase in EQE. The highest EQE was obtained in 5% C6-doped DPA film based OLET, which is more than seven times higher compared to that of non-doped DPA film based OLET (Figure 4f). Thanks to the high carrier mobility DPA host, excitons generated in host DPA after the recombination of electrons and holes were successfully harvested through energy transfer to molecularly doped C6. The efficient FRET between DPA and C6 facilitated the radiative decay of singlet excitons in OLET and suppressed the charge-carrier-exciton annihilation and triplet-singlet annihilation. To further investigate the exciton utilization efficiency in C6-doped DPA film based OLET, the comparison of PLQY of DPA film and C6-doped DPA films, and the EQE of their OLET devices are summarized in Table

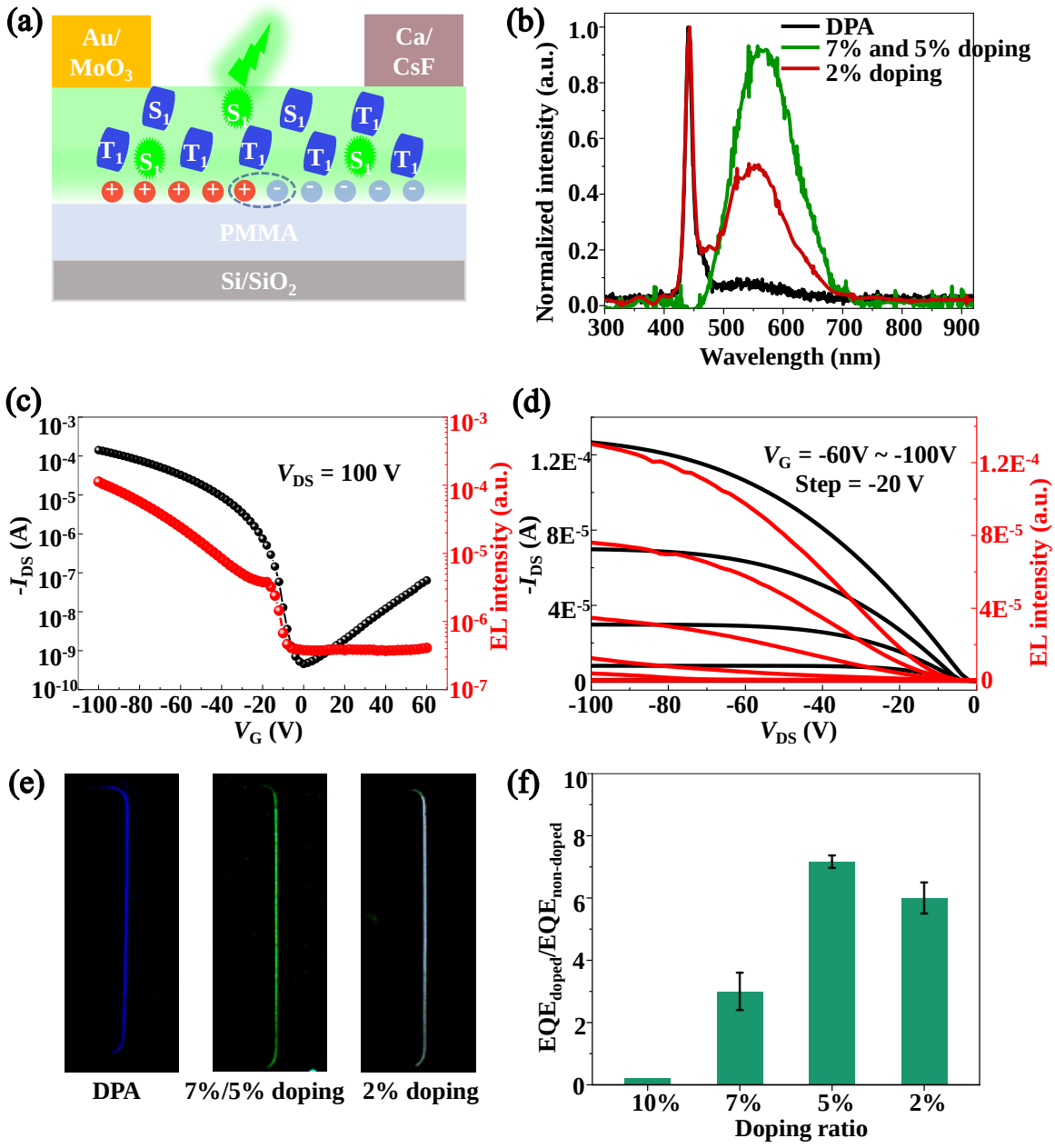

Figure 4. (a) Scheme of device architecture of OLET (S_1: first energy level singlet, T_1: first energy level tinglet), (b) EL spectra of DPA film and different C6-doped DPA films based OLET devices, (c) and (d): trnasfer and output curves of DPA film based OLET, (e) EL images of DPA film and different C6-doped DPA films based OLET devices. (f) EQE of C6-doped DPA films based OLET devices compared to that of DPA film based OLET.

S1. The light out-coupling efficiency and the ratio of electrons to holes are assumed to be same for all OLET devices since they have the same device architecture. Therefore, according to the

EQE calculation equation, the highest exciton utilization efficiency has been increased more than 2.4 times of C6-doped film than that of DPA film. This result confirmed that doping a highly emissive C6 into DPA film is an effective way for realizing the construction of high performance OLET with the exciton utilization efficiency significantly improved.

Conclusion

In summary, the possibility of integrating high mobility and high triplet utilization efficiency has been confirmed in the representative high mobility emissive DPA molecule. To further harvest the triplet excitons in DPA, C6 was doped into DPA for increasing the PLQY of DPA film and supressing annihilation between excitons. After molecular doping, the EQE of C6-doped DPA film based OLET has been increased more than seven times compared to that of DPA film based OLET. Meanwhile, the highest exciton utilization efficiency of C6-doped DPA film based OLET has been increased more than 2.4 times compared to that of DPA film. The concept provided in this work can be extended to other advanced high mobility anthracene derivatives, which provides a new approach for enriching high mobility emissive matrial systems. Moreover, more outstanding properties can also be integrated in such materials by molecular doping, such as narrow emission, high triplet utilization efficiency, lasing, ect. As such, high performance OLET devices with multifunctions are expected to construct for applications in smart display and organic electrically pumped lasers.

Experimental Section

Photophysical property measurement: Steady-state absorption spectra of solution and film samples were recorded on a Jasco V-570 spectrometer and an Analytik Jena Specord 210 spectrophotometer equipped with an integrating sphere, respectively. Photoluminescence spectra of solution and films were recorded on a Jasco FP-6600 spectrophotometer and PLQY of films were measured by using the LabSphere® integrating sphere (FluoroMax-4, HORIBA JobinYvon, PLQY software package). De-aerated DPA/PtOEP TTA upconversion sample in

THF was prepared by bubbling under Ar for 25 mins. TTA upconversion film was prepared by thermal vacumm depositing PtOEP (5 nm) and DPA (50 nm) on clean glass substrate. The TTA upconversion emission was measured under the excitation of a 532 nm CW laser with a Princeton Instruments Acton SP2500 spectrograph and a SPEC-10 liquid nitrogen-cooled CCD camera.

OLET device fdabrication and measurement: First, SiO_2 (300 nm)/Si substrates were modified by PMMA via sipin-coating its 6 mg mL^{-1} chlorobenzene solution at 4,000 r.p.m. for 60 s in the glove box and then annealed at 140 °C for 2 hours. Second, DPA and C6-doped DPA films were prepared on Si/SiO_2/PMMA substrate by thermal vacumm deposition. Then, asymmetric electrodes Au (30 nm)/MoO_3 (1 nm) and Ca (35 nm)/CsF (1 nm) were fabricated by thermal evaporation with a mask for hole injection and electron injection, respectively. The electrical and optical characterizations were performed with a semiconductor FS-PRO (Platform Design Automation, Inc.). The optical data were tested with the assist of a photomultiplier tube (H10721), which was calibrated by a standard LED device with the help of an absolute spectrum measurement system (Spectrum TEQ-EL, Ocean Optical, Inc.). AFM images were obtained by using a Digital Instruments atomic force microscope Nanoscope III in air.

Calculation: The geometry optimization of DPA was calculated by using density functional theory (DFT) within the Gaussian 16 Suite, all geometries were optimized using the B3LYP functional and the 6-311G basis set. The excited-state energy level of was calculated by using time dependent DFT (TD-DFT) based on the optimized ground state geometry.

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