Molecular doped, color-tunable, high-mobility, emissive, organic semiconductors for light-emitting transistors

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Developing high-mobility emissive organic semiconductors with tunable colors is crucial for organic light-emitting transistors (OLETs), a pivotal component of integrated optoelectronic devices, but remains a great challenge. Here, we demonstrate a series of color-tunable, high-mobility, emissive, organic semiconductors via molecular doping with a high-mobility organic semiconductor, 2,6-diphenylanthracene, as the host. The well-matched molecular structures and sizes with efficient energy transfer between the host and guest enable the intrinsically high charge transport with tunable colors. High mobility with the highest value >2 cm² V⁻¹ s⁻¹ and strong emission with photoluminescence quantum yield >15.8% are obtained for these molecular-doped organic semiconductors. Last, a large color gamut for constructed OLETs is up to 59% National Television System Committee standard, meanwhile with an extremely high current density approaching 326.4 kA cm⁻², showing great potential for full-color smart display, organic electrically pumped lasers and other related logic circuitries.

INTRODUCTION

Organic light-emitting transistors (OLETs) combining the light-emitting function of organic light-emitting diodes (OLEDs) and the current modulation (and amplification) function of organic field-effect transistors (OFETs) in a single device have great potential for integrated optoelectronics, smart display technologies, and organic electrically pumped lasers (1–6). The open-ended light-emitting properties of the active layer of unencapsulated OLETs are accessible for the investigation of optical and electronic fundamental physical processes in organic semiconductors, providing powerful insights into electroluminescence device physics (7–12). In addition, the position of the recombination region in the channel of ambipolar OLETs can be shifted away from electrodes under the regulation of bias voltage, thus avoiding metal-induced optical loss and exciton quenching for much higher device performances (13–19). Recently, Samori and colleagues (20) developed an optically switchable OLET by ingeniously combining light-emitting polymers and photochromic molecules, opening intriguing perspectives for the applications of OLETs in unique, optically gated, highly integrated full-color displays and active optical memory. Besides the efforts dedicated to the multifunctional applications of OLETs, developing high-mobility emissive organic semiconductors with tunable colors is crucial for advancing these potential technologies, but it remains a great challenge. Some impressive advances have been achieved in the integration of high charge transport and efficient light emission in organic semiconductors by molecular design innovation (21–28). However, the optoelectronic properties of these materials are still relatively low, and the modulation of their emissive colors is very difficult and usually at the expense of the transport properties. Therefore, integrating high charge carrier mobility and strong emission in an organic semiconductor with tunable colors is not only urgently needed but also a long-standing problem in organic optoelectronics.

Here, we demonstrate a series of color-tunable, high-mobility, emissive, organic semiconductors via molecular doping with a blue-emission, high-mobility, organic semiconductor 2,6-diphenylanthracene (DPA) as the host and tetracene (Tc) or pentacene (Pen) as the guest. The well-matched molecular structures and sizes, as well as efficient energy transfer between the host and guest, enable the intrinsically high charge transport with tunable colors of molecular doped materials. Five colored, high-mobility, organic semiconductors from blue-to-red including the host molecule are prepared with the highest mobility of more than 2 cm² V⁻¹ s⁻¹ and photoluminescence quantum yield (PLQY) > 15.8%. Last, OLETs based on these molecular doped materials are constructed with a color gamut up to 59% National Television System Committee (NTSC) standard and 83% standard Red Green Blue, and a high current density approaching 326.4 kA cm⁻². This work opens a avenue for developing color-tunable, high-mobility, emissive, organic semiconductors toward high-performance OLETs, thereby rendering their great promise for various technology applications including full-color displays, organic electrically pumped lasers, and other related integrated optoelectronic devices and circuits.

RESULTS

Molecular doping is an effective strategy for modifying the electrical and optical properties of semiconducting materials (29–36), but it is difficult to realize the simultaneous modulation for both of these properties due to the contradiction of their requirements and uncontrollable structures induced by doping process. In our studies, we choose a superior high-mobility, blue-emission, organic semiconductor, DPA, as the host with green emission Tc or red emission Pen as the guest molecules (Fig. 1A and fig. S1) based on the following considerations: (i) The similar fused-ring–conjugated molecular structures, sizes, and electron-cloud distribution of host and guest molecules facilitate the formation of doping systems with long-range
ordered structures, resulting in efficient charge transport channels, which are essential for high charge carrier mobility (Fig. 1B and figs. S2 and S3) (37–39). (ii) The large overlap between the photo-luminescence (PL) spectrum of DPA and the absorption spectra of guest molecules ensures an efficient energy transfer between host and guest (Fig. 1C), thereby enabling the modulation of the degree of Förster resonance energy transfer (40) and emission color by precisely controlling the molecular doping concentration (Fig. 1, D and E).

Appropriate doping concentration is crucial for resulting in high-quality and uniform doped crystals with tunable emission colors due to the controlled energy transfer process. Doped crystals are prepared with guest mass fractions of 2 to 8% for the Tc-doping process and 0.1 to 3% for the Pen-doping process through a physical vapor transport (PVT) method. As confirmed by fluorescence micrographs (Fig. 2, A, C, and E) and fluorescence spectroscopy (Fig. 2J), flaky DPA, Tc 8%, and Pen 3% single crystals emit bright blue, green, and red color under ultraviolet (UV) excitation, respectively. The cyan blue emission crystal (Tc 3%) and pink emission crystal (Pen 0.5%) can also be successfully prepared by decreasing the doping concentration to reduce the degree of energy transfer between the host and guest in the doped crystals (Fig. 2, B and D).

According to our results, the optimized mass fractions for Tc-doped crystals are 3 and 8% and 0.5 and 3% for Pen-doped crystals. The absorption spectrum of doped crystals shows a well-structured vibrational band of typical anthracene derivatives peaking at λ = 425, 402, and 379 nm ascribed to 0 to 0, 0 to 1, and 0 to 2 vibronic peaks, respectively (fig. S4), which is similar to that of pure DPA. The regular micrographs of obtained crystals and the typical birefringence characteristics under polarized light microscopy confirm that the prepared molecular doped crystals are high-quality organic single crystals (figs. S5 and S6). The structure of these molecular doped single crystals is proved to be well consistent with DPA (Cambridge Crystallographic Data Centre 1044209) by x-ray diffraction (XRD) (Fig. 2F) and selected-area electron diffraction (SAED) patterns (Fig. 2, G to I, and fig. S7), suggesting that molecular stacking of the host is not changed after doping. No changes in the SAED pattern are observed in different parts of the same crystal, indicating that the whole sample is a single crystal.

The decay lifetime of doped and undoped crystals at the nanosecond time level is studied using the time-correlated single-photon counting (TCSPC) system to evaluate the degree of energy transfer between the host and guest. Figure 2K shows the TCSPC decay curves and corresponding fitting results of the undoped and doped crystals at the emission wavelength of 472 nm. By fitting the decay curves, the decay lifetime (τ0) of DPA is calculated to be 8.21 ns, while τd decreased to 3.84 and 0.31 ns for Pen 0.5%, and Pen 3%, respectively, indicating that an effective energy transfer between the host and guest occurs in these molecular doped single crystals. The energy transfer rate can be calculated from the decay lifetime of DPA (τ0) and the decay lifetime after doping (τd) according to the equation: 

$$k_{ET} = \frac{1}{\tau_d} - \frac{1}{\tau_0}.$$ 

The kET calculated for Pen 0.5% and Pen
3% is $1.39 \times 10^8$ and $3.10 \times 10^9$ s$^{-1}$. Last, according to the equation, $E = k_{ET} / (1/\tau_0 + k_{ET})$, the calculated energy transfer efficiencies ($E$) of Pen 0.5% and Pen 3% are 53 and 96%, respectively. The $E$ of Tc-doped crystals cannot be calculated by the variation of the decay lifetime of DPA crystals and Tc-doped crystals at the DPA emission wavelengths because the spectra of Tc and DPA overlap considerably in the DPA emission wavelength range. The $E$ of Tc-doped crystals increased with increasing doping concentration of Tc, as evidenced by the complete disappearance of the PL peak attributed to DPA in the PL spectrum of Tc 8%. In addition, the decay lifetime of Tc-doped crystals monitored at 501 nm is prolonged from 11.46 to 12.19 ns when the doping concentration is increased (Fig. 2L). Although severe fluorescence quenching can occur because of molecular aggregation in crystalline Pen and Tc, the effective energy transfer between the host and guest and the single-molecule state of the guest molecule in doped crystals facilitate efficient fluorescence of these molecular doped single crystals. The PLQY of Tc 3%, Tc 8%, Pen 0.5%, and Pen 3% in the single crystal are approaching 37.9, 27.7, 23.9, and 15.8%, respectively, which are much higher than that of pure Tc and Pen crystals (table S1) (41–45). The value of PLQY decreases with increasing doping concentration, probably because of the fluorescence quenching caused by the aggregation of guest molecules in the high-concentration molecular doped single crystals (46–48).

To characterize the charge transport properties of these molecular doped crystals, bottom-gate top-contact OFETs based on doped crystals are fabricated on the octadecyltrichlorosilane (OTS)–modified SiO$_2$ (300 nm)/Si substrates. Schematic energy levels of doped DPA
crystals are shown in fig. S8. According to the dominant transport direction of DPA, the channel direction of the transistor is constructed along the $b$ axis of these doped crystals (Fig. 3A and fig. S9A) (21). As evidenced by the representative transfer and output curves of transistors based on these doped crystals (Fig. 3, B to G), these devices exhibit good switching functionality with a $I_{on}/I_{off}$ ratio of around $10^7$. The highest mobility values calculated from the saturation region of representative transfer curves are 3.3, 3.1, 3.2, and 2.2 cm$^2$ V$^{-1}$ s$^{-1}$ for Tc 3%, Tc 8%, Pen 0.5%, and Pen 3%, respectively. Totally, 20 devices are measured for each doped crystal, and the average mobilities for Tc 3%, Tc 8%, Pen 0.5%, and Pen 3% are 1.8, 2.1, 1.5, and 1 cm$^2$ V$^{-1}$ s$^{-1}$, respectively (Fig. 3, H to K). The mobility of Tc-doped crystals is more uniform than that of Pen-doped crystals based on the statistics of device mobility, which is probably due to the smaller molecular size of Tc beneficial for higher-quality doped DPA crystals. The above experimental data fully demonstrate that a series of high-mobility emissive organic semiconductors with tunable colors are successfully prepared by a carefully designed molecular doping strategy.

Furthermore, OLETs based on these color-tunable, high-mobility, emissive, organic crystals are constructed using an asymmetric device geometry with high work function electrode (Au/MoO$_3$ $\approx$ $-$5.6 eV) for hole injection and low work function electrode (Ca/CsF $\approx$ $-$2.5 eV) for electron injection (Fig. 4A and fig. S9B) (49, 50). A polymethyl methacrylate (PMMA) buffer layer close to 20-nm (fig. S10) thick is modified on the SiO$_2$ surface to reduce charge traps on the dielectric layer (SiO$_2$). As demonstrated by representative transfer and output curves of Tc 8%, Pen 3%, Tc 3%, and Pen 0.5% light-emitting transistors, these prepared OLET devices with asymmetric source-drain electrodes exhibit ambipolar charge transport characteristics (Fig. 4, B to G, and fig. S11). The negligible hysteresis of a Tc 8%, Pen 3%, Tc 3%, and Pen 0.5% light-emitting transistor at positive and negative source-drain voltages indicate that the PMMA buffer layer has a good ability to shield the charge traps (fig. S12). There are substantial hole and electron transport at the negative and positive gate voltages with the $I_{on}/I_{off}$ ratio of photocurrent in a range of $1.6 \times 10^1$ to $5.8 \times 10^2$ (fig. S13) and a nearly linear increase in current on the output curves corresponding to opposite charge carrier injection at high source-drain voltages. The working mechanism of OLETs can be divided into the following processes: (i) Charges (both holes and electrons) are injected into active layers from the high work function electrode and low work function electrode separately. (ii) In the operation region, holes and electrons recombine to form excitons. (iii) The high-energy excitons decay to the ground state by emitting photons as the final output. Totally, we fabricate and test more than 5, 5, 10, and 5 OLETs devices for Tc 3%, Tc 8%, Pen 0.5%, and Pen 3%, respectively. The device leakage current is in a small $10^{-10}$ A range at the gate and a source-drain voltage of $-$100 V (fig. S14). The summary of the mobility of these devices is shown in fig. S15, where both electron mobility and hole mobility are on the same order of magnitude as that of DPA-OLETs (12) and are among the highest-mobility values reported for OLETs so far in the literatures.

**Fig. 3.** Device performance of field-effect transistors. (A) Schematic of the device structure of the transistors. (B and C) Representative transfer curves of transistors based on Tc 3%, Tc 8%, Pen 0.5%, and Pen 3% single crystals. (D to G) The output curves corresponding to the transfer curves in (B) and (C). (H to K) Mobility distribution based on 20 devices of Pen 0.5% (H), Pen 3% (I), Tc 3% (J), and Tc 8% (K) single-crystal transistors.
(7, 28), confirming the good integrated optoelectronic properties in these doped crystals. Noticeably, although an asymmetric device geometry is incorporated for the modulation of efficient electron and hole injection, a certain mismatch is still existing for hole and electron mobility. It is believed that a better match between the lowest unoccupied molecular orbital energy level of the materials and the work function of the thermo-evaporation electrode can be achieved by optimizing the thickness of the CsF electrode. Mismatch of energy levels and large crystal thicknesses can lead to some contact resistance. The high repeatability of the performance of these light-emitting transistors based on doped crystals demonstrates the maturity and reproducibility of the process for constructing single-crystal devices. The electroluminescence spectra and videos are collected in real-time using a spectrometer and a charge-coupled device camera, respectively. Strong and spatially controlled electroluminescence within conducting channels is demonstrated due to the inherent high-mobility emissive characteristics of these doped crystals and ambipolar charge transport characteristic of OLETs device. As evidenced by electroluminescence spectra (Fig. 4, H and I, and fig. S16) and images (figs. S17 and S18), Tc 8%, Pen 3%, Tc 3%, and Pen 0.5% light-emitting transistors emit colors, which are consistent with PL of green, red, cyan blue, and pink, respectively. The electroluminescence spectra of these devices all exhibit voltage-independent properties. Because of the inherent high charge transport properties and strong emission characteristics of these molecular doped materials, the current densities of high-performance Tc 3%, Tc 8%, Pen 0.5%, and Pen 3% light-emitting transistors (figs. S19 and S20) are extremely high current density, reaching 325.6, 146.3, 105.4, and 66.8 kA cm$^{-2}$ (fig. S21), respectively, assuming that the charge is transported within only one molecular layer (44, 51, 52). As for the planar OLETs, it is difficult to measure the exact luminous area. Here, the luminance of 10,282, 13,349, 9029, and 18,743 cd m$^{-2}$ are achieved for Tc 8%, Pen 3%, Tc 3%, and Pen 0.5% light-emitting transistors, respectively (fig. S22) with a width value of around 6 μm estimated from their emission images. Much higher luminance could be obtained if we use the 2-μm-wide value that is commonly used in literatures (16, 53). The electroluminescence parameters for our constructed OLETs, reported neat Tc-, and Pen-based OLETs are summarized in table S1. Obviously, superior performances are demonstrated for these molecular doped crystal–based OLETs due to their good optoelectronic properties simultaneously integrating high charge carrier mobility and strong emission, suggesting their great promising potential for future applications and fundamental studies.

The uniformity in mobility and high device operating stability (fig. S23) of DPA and doped crystal light-emitting transistors...
demonstrates the unique advantage of three primary color OLETs prepared from DPA and doped crystals for display panel fabrication. The Commission Internationale de l’Eclairage (CIE) coordinates of DPA, Tc 8%, Pen 3%, Tc 3%, and Pen 0.5% light-emitting transistors are (0.1617, 0.1101), (0.2750, 0.5361), (0.6535, 0.3223), (0.2819, 0.4401), and (0.5135, 0.2530), respectively. Representative three primary color OLETs based on DPA, Tc 8%, and Pen 3% single crystals are demonstrated (Fig. 5, A and B). The color triangle achieved by DPA, Tc 8%, and Pen 3% light-emitting transistors covers 59% of the NTSC standard in the CIE 1931 color space, which greatly exceeds the 45% of the NTSC standard covered by some commercial display panels, thereby rendering their great promise for integrated full-color optoelectronic devices and circuits.

**DISCUSSION**

OLETs, as a kind of unique integrated optoelectronic device, show great promise for various miniature and integrated optoelectronic technologies, but their development seriously lags behind. One of the challenges is the limit of high-mobility emissive organic semiconductors with tunable colors due to the contradictory demands of electrical and optical properties. In this work, we present a series of color-tunable, high-mobility, emissive, organic semiconductors via molecular doping with a superior high-mobility, emissive, organic semiconductor used as the host. This strategy can be extended to more conjugated organic molecule systems with the rational selection of high-mobility emissive host and guest molecules for superior performances. Moreover, such a research concept can also provide us a possibility for the preparation of colorful high-mobility organic single crystals via the good control of doping position, which is crucial for monolithic colorful (even white) lasers and integrated optoelectronic applications (54–56). It is believed that color-tunable, high-mobility, emissive, organic semiconductors will bring a rapid development of OLETs and their related potential applications, including integrated technologies such as smart displays, organic electrically pumped lasers, and on-chip optoelectronic circuits by combining with high-resolution patterning techniques and integrating with complementary metal-oxide semiconductor electronics (20, 57–60).

**MATERIALS AND METHODS**

**Materials and instruments**

Pen and Tc powders were purchased from Sigma-Aldrich. DPA was synthesized according to the reported method (26). The UV-visible (UV-vis) absorption spectra were characterized using a UV-vis–near-infrared spectrophotometer. The crystallinity of crystals was characterized by transmission electron microscopy (JEM 1011 and 2100) and XRD (Empyrean). Optical images and PL images were characterized through a fluorescence microscope (OLYMPUS BX51). The field-effect transistors were characterized on a Keithley 4200-SCS in the atmospheric environment, and the mobility was calculated from the saturation region by using the equation of $I_{ds} = C_i \mu (W/2L) (V_G - V_T)^2$. The $W$ and $L$ are determined by the crystal used in the devices (fig. S9). All the electrical and optical tests of light-emitting transistors were performed by using the FS-380 semiconductor test system equipped with a spectrometer (Ocean Optics) and a photomultiplier tube (Hamamatsu) in the glove box.

**Preparation of molecular doped crystals**

First, Pen, Tc, and DPA powders were mixed uniformly according to the mass fraction of the guest. It should be stated that because of the poor solubility of host and guest and the low doping concentration...
of guest, it is difficult for us to determine the exact quantitative percentage of Pen and Tc in the doped crystals with a standard curve method based on Lambert-Beer’s law. The doping concentrations are referred to as the mass fraction of guest molecules in mixed powders. After mixing, the mixture was transferred to a vacuum-drying oven and dried at 80°C for 12 hours. The dried mixture was then placed in a tube furnace with a single heating zone (150°C) for the growth of DPA, Tc 3%, Tc 8%, Pen 0.5%, and Pen 3% crystals under vacuum. After 5 hours of growth, single crystals were obtained on Si/SiO₂/OTS substrate from a PMMA solution (6 mg ml⁻¹; chlorobenzene (HNO₃ : H₂O = 7:3, volume ratio), pure water, and oxygen plasma. (ii) OTS-modified layers were processed onto Si/SiO₂/OTS substrate by vacuum vapor phase method. (iii) Freshly doped crystals were grown onto the Si/SiO₂/OTS substrate according to the crystal preparation method described above. (iv) Gold electrodes were transferred to the crystal surface by probes under the microscope.

The single-crystal, light-emitting transistors were fabricated by the following steps: (i) The PMMA layers were spin-coated onto Si/SiO₂ substrate from a PMMA solution (6 mg ml⁻¹; chlorobenzene as a solvent) at 4000 rpm for 60 s in the glove box and then annealed at 90°C for 2 hours. (ii) Large-sized molecular doped crystals were transferred onto Si/SiO₂/PMMA substrate by shearing friction in the glove box. (iii) Asymmetric electrodes Au (30 nm)/MoO₃ (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 PMMA buffer layer (20 nm) has a negligible effect on the capacitance of the SiO₂ (300 nm), and thus, the capacitance used in the calculation of the mobility of OLET is still 10 nF cm⁻².

SUPPLEMENTARY MATERIALS
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