Exciton funneling in light-harvesting organic semiconductor microcrystals for wavelength-tunable lasers

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Organic solid-state lasers are essential for various photonic applications, yet current-driven lasing remains a great challenge. Charge transfer (CT) complexes formed with p-/n-type organic semiconductors show great potential in electrically pumped lasers, but it is still difficult to achieve population inversion owing to substantial nonradiative loss from delocalized CT states. Here, we demonstrate the lasing action of CT complexes based on exciton funneling in p-type organic microcrystals with n-type doping. The CT complexes with narrow bandgap were locally formed and surrounded by the hosts with high-lying energy levels, which behave as artificial light-harvesting systems. Excitation light energy captured by the hosts was delivered to the CT complexes, functioning as exciton funnels to benefit lasing actions. The lasing wavelength of such composite microcrystals was further modulated by varying the degree of CT. The results offer a comprehensive understanding of exciton funneling in light-harvesting systems for the development of high-performance organic lasing devices.

INTRODUCTION

Organic semiconductor lasers that can deliver intense coherent light across the full visible spectrum have been intensively pursued during the past decade (1–6) for their practical applications that range from multiband communication to full-color laser displays (7–11). Nevertheless, electrically pumped organic lasers remain a grand challenge (12) and would mark a technological advance, with application potential rivaling that of organic light-emitting diodes (13–15). The main stumbling blocks behind this challenge are the unbalanced carrier mobility and extra triplet losses under electrical injection (16). Charge transfer (CT) complexes formed with p-/n-type semiconductors are considered promising candidates for electrically driven lasing because of their balanced ambipolar (17) and lower triplet loss (18, 19). However, in organic CT complexes with a fixed stoichiometry, strong CT transitions usually lead to delocalized CT states, which dissociate into free carriers and thus produce a nonradiative deactivation channel to depopulate the CT excitons (20, 21), making it difficult to achieve population inversion.

Introducing a small amount of electron acceptor/donor into the electron donor/acceptor host matrix would enable the formation of localized CT complexes (pink spheres, Fig. 1A) (22, 23). These CT complexes with narrow bandgap (24) are surrounded by the host matrix (gray sphere, Fig. 1A) with high-lying energy levels, which can serve as artificial light-harvesting antennas (25, 26). Excitation light energy can be harvested by the antennas to produce excitons (blue circles, Fig. 1B) and then transferred downstream to the acceptors (red circles, Fig. 1B) via energy transfer (27–29). This functions as exciton funnels (30) to concentrate excitons to the narrow bandgap CT complexes, benefiting the population inversion for stimulated emission (Fig. 1B) (31, 32). As a result, the controllable construction of light-harvesting systems with efficient exciton funneling is a promising method for the buildup of proper energy-level structures toward the stimulated emission of organic CT complexes.

RESULTS

Cyano-substituted oligo-(α-phenylenevinylene)-1,4-bis(R-cyano-4-diphenylaminostyryl)-2,5-diphenylbenzene (referred to as OPV; Fig. 2A and figs. S1 and S2) and fullerene (C₆₀; Fig. 2A) were selected as scaffolds to fabricate light-harvesting microcrystals for the following reasons. (i) OPV is a typical p-type semiconductor with high optical gain (33), and C₆₀ is a typical n-type semiconductor with high electron mobilities as well as large absorption cross sections (34). Introducing C₆₀ into OPV is beneficial for achieving enhanced light-harvesting ability and balanced carrier mobility. (ii) The CT interaction between OPV and C₆₀ (Fig. 2A) might contribute to the formation of localized CT complexes (C₆₀-OPV) with narrowed bandgap (23, 24), which are expected to function as the acceptors in light-harvesting systems (26).
OPV microcrystals doped with controllable amount of C\textsubscript{60} were synthesized through a liquid-phase self-assembly method (see Materials and Methods for preparation details). The doping concentration of C\textsubscript{60} was determined with a standard curve method on the basis of the Lambert-Beer's law (see Materials and Methods for the determination of doping concentrations; fig. S3) (35). From the predicted growth of OPV crystal (fig. S4), we can see that the strong intermolecular \(\pi-\pi\) interaction facilitates the crystal to form a one-dimensional structure, which is confirmed by the scanning electron microscopy (SEM) images (Fig. 2B). The pure OPV microcrystal has a well-defined wire-like morphology with smooth and flat surfaces. Upon the introduction of C\textsubscript{60}, the resulting microcrystals (C\textsubscript{60}@OPV) share a similar morphology (Fig. 2C), which indicates that the introduction of C\textsubscript{60} molecules does not significantly affect the growth of OPV microcrystals.

The crystal structures of the microwires were characterized using transmission electron microscopy (TEM; Fig. 2, D and E), selected-area electron diffraction (SAED; Fig. 2, F and G), and x-ray diffraction (XRD; fig. S5). OPV and C\textsubscript{60}@OPV microcrystals exhibit similar SAED patterns and XRD peaks, showing that the C\textsubscript{60}@OPV microcrystals retain the original crystal structure of OPV after the introduction of C\textsubscript{60}. Further analyses of the TEM and SAED patterns reveal that each wire is a high-quality single crystal growing along the [100] direction, which strongly suggests that the C\textsubscript{60}@OPV microwires are mostly made up of OPV, with C\textsubscript{60} doped in its matrices (see the confocal images of the C\textsubscript{60}@OPV microwire in fig. S6). The intermolecular interaction between C\textsubscript{60} and OPV molecules in C\textsubscript{60}@OPV microcrystals was investigated by analyzing micro-Raman spectra of the pure OPV, pure C\textsubscript{60}, and C\textsubscript{60}@OPV microwires. As displayed in the purple box in Fig. 2H, the Raman spectrum of the C\textsubscript{60}@OPV microwire maintains the characteristic peaks of the OPV. Meanwhile, the Raman peak near 250 cm\(^{-1}\) in the C\textsubscript{60}@OPV microwire, which is identified as the squashing mode (Hg) of C\textsubscript{60}, shows

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**Fig. 1. Illustration for exciton funneling in the light-harvesting systems.** (A) Schematics of light harvesting, where the wide-bandgap hosts (gray spheres) act as antenna and locally formed CT complexes with narrow bandgap (pink spheres) act as acceptor. (B) Diagram for exciton funneling in light-harvesting systems. The black arrows represent the exciton transfer process.

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**Fig. 2. Preparation and structural characterization of C\textsubscript{60}@OPV light-harvesting microcrystals.** (A) Chemical structures of OPV and C\textsubscript{60}, and molecular structure of the resulting CT complex. (B and C) SEM images of typical OPV and C\textsubscript{60}@OPV microwires. Scale bars, 5 \(\mu\)m (B) and 2 \(\mu\)m (C). (D and E) TEM images of individual OPV and C\textsubscript{60}@OPV microwires. Scale bars, 2 \(\mu\)m. (F and G) SAED patterns of the corresponding microwires in (D) and (E). Scale bars, 2 1/nm. (H) Raman spectra of individual OPV, C\textsubscript{60}, and C\textsubscript{60}@OPV microwires. a.u., arbitrary units. The C\textsubscript{60} doping concentration of the C\textsubscript{60}@OPV microcrystals used for these characterizations is 5.6 mol \%.
a downfield shift of 12 cm$^{-1}$ compared to that of pure C$_{60}$ microwire (blue box, Fig. 2H), implying the CT interaction between C$_{60}$ and OPV molecules (36).

The interaction between C$_{60}$ and OPV molecules generates a new CT state, which can be verified by the absorption spectra. The absorption spectrum of C$_{60}$@OPV exhibits two distinct absorption bands (Fig. 3A, red line), where abs$_1$ is obviously derived from the absorption of OPV hosts (Fig. 3A, green line). The newly emerging absorption peak (abs$_2$) is largely red-shifted compared with those of the pure components, which should be attributed to the CT transition of the ground state. Under ultraviolet excitation, the C$_{60}$@OPV microwires exhibit red emission (Fig. 3B, bottom) with a solid-state fluorescence quantum yield ($\Phi$) of $\sim$13% (fig. S7), which is in sharp contrast with the yellow emission in the pure OPV microwires (Fig. 3B, top; $\Phi'$ of $\sim$65%, as shown in fig. S7) and nonluminescence in the C$_{60}$ microwires (fig. S8). This indicates that the photoluminescence (PL) of C$_{60}$@OPV crystals originates from the new CT excited state.

Time-resolved luminescence was examined to elucidate the nature of CT state in the C$_{60}$@OPV microcrystals. Figure 3 (C and D) shows the streak camera images of individual OPV and C$_{60}$@OPV microwire, respectively, which provide a visual image of the time- and wavelength-dependent PL intensities. The spectra in the insets were obtained by integrating the corresponding streak images, where the PL spectrum of the C$_{60}$@OPV peaked at 597 nm, which is shifted bathochromically by $\sim$46 nm compared with that of pure OPV (551 nm). This implies a large change in the molecular configuration and a strong CT character of the excited state in C$_{60}$-OPV complexes. These CT complexes capture the energy from the OPV hosts, which is evidenced in fig. S9 by a longer rise time at 597 nm than that at 551 nm. The prolonged rise time indicates an efficient energy transfer from the OPV hosts to the CT complexes (37), making the C$_{60}$@OPV microcrystals act as light-harvesting systems. The efficiency of energy transfer was calculated according to $E = 1 - t_{DA}/t_{D}$ (38), where $t_{D}$ and $t_{DA}$ are the average lifetime of donor in the absence (pure OPV) and presence (C$_{60}$@OPV) of the acceptor, respectively. $t_{D}$ was determined to be 0.85 ns (fig. S10), and $t_{DA}$ was 0.25 ns (table S1). Accordingly, the efficiency was estimated to be $\sim$70.6% (table S1), which is high enough (39, 40) for effective exciton funneling and accumulation in the CT states of the light-harvesting microcrystals.

To provide an insight into the energy-level structures of the light-harvesting systems, we performed theoretical studies by calculating the highest occupied molecular orbital (HOMO) and the lowest unoccupied molecular orbital (LUMO) of OPV, C$_{60}$ molecules, and the C$_{60}$-OPV CT complex (Fig. 4A). It is found that the HOMO electron density of OPV is distributed over the entire molecule, whereas the LUMO is mostly localized on the phenylenevinylene and the adjacent cyano group. The complexation with C$_{60}$ promotes the CT from the HOMO of OPV to the LUMO of C$_{60}$, which results in a slight increase in HOMO energy and a significant decrease in LUMO.

![Fig. 3. Exciton funneling into the CT states in the organic light-harvesting microcrystals.](http://advances.sciencemag.org/)

(A) Absorption spectra of OPV (green), C$_{60}$@OPV (red) microwires, and C$_{60}$ dispersed in polymer hosts (black). a.u., arbitrary units. (B) Fluorescence microscopy images of OPV (top) and C$_{60}$@OPV (bottom) microwires. Scale bars, 20 $\mu$m. (C and D) Streak camera images and PL spectra of OPV (C) and C$_{60}$@OPV (D) microcrystals excited with a 400-nm pulsed laser ($\sim$100 fs, 1 kHz). $t_{D}$ and $t_{DA}$ are the average lifetimes of donor ($\sim$551 nm) in the absence (pure OPV) and presence (C$_{60}$@OPV) of the acceptor, respectively. The C$_{60}$ doping concentration of the C$_{60}$@OPV microcrystals used for these characterizations is 5.6 mol %.
energy for the CT complex with respect to those of OPV. Notably, the formation of the CT complex reduces the HOMO-LUMO gap from 2.65 to 1.72 eV, which is in accordance with the red shift in the absorption and emission spectra after doping with C\textsubscript{60}.

The experimental and theoretical results help us draw an energy diagram of exciton evolution in such a light-harvesting system. As exhibited in Fig. 4B, the pump energy was first harvested by the OPV hosts and subsequently captured by the CT complexes through efficient energy transfer. This energy transfer concentrates the excitons into the CT complexes, which behave as exciton funnels to favor the population inversion for the lasing behavior of the CT states. Moreover, the emission wavelength of the light-harvesting microcrystals was adjusted from 551 to 597 nm through varied doping concentrations of C\textsubscript{60} (Fig. 4C and fig. S11). The wavelength shift is ascribed to the high tunability of the energy levels upon regulating the degrees of CT (fig. S12) (23), which hold great promise for tailoring the output wavelength of the microscale lasers over a wide range.

Optically pumped lasing measurements were carried out on a micro-PL system (fig. S13). Figure 5A shows the optical images and corresponding emission spectra of an individual C\textsubscript{60}@OPV microwire excited with increasing laser powers. At low pump fluence ($P < 65 \mu J/cm^2$), the C\textsubscript{60}@OPV microwire exhibits a broad spontaneous emission. At higher pump fluence ($P \geq 65 \mu J/cm^2$), the emission spectrum develops as a set of sharp peaks at ~607 nm, which indicates the stimulated emission. The emission intensity exhibits an S-shaped nonlinear dependence on the pump fluence (Fig. 5B), which demonstrates the transition from spontaneous emission ($P < 65 \mu J/cm^2$) via amplified spontaneous emission ($65 \mu J/cm^2 < P < 70 \mu J/cm^2$) to full lasing oscillation ($P > 70 \mu J/cm^2$). The occurrence of lasing in the C\textsubscript{60}@OPV microwire is further verified by time-resolved PL measurements at different pump intensities (fig. S14) (41). The C\textsubscript{60}@OPV microwire exhibits a typical characteristic of Fabry-Pérot resonators, with the laser emission mainly coupling out from the end facets (inset in Fig. 5A), which is validated by the simulated result of electric field distribution (fig. S15) and the linear relationship between $\lambda^2/2\Delta\lambda$ and the length of the microwires (fig. S16).

As was discussed above, the emission from CT complexes might be tailored controllably by doping different amounts of C\textsubscript{60} into the OPV hosts, which provides the feasibility of tuning the gain region in the microcrystals. When the doping concentration of C\textsubscript{60} increases from 0 to 5.6 mol %, the lasing color of C\textsubscript{60}@OPV microcrystals was adjusted from green (556 nm) to yellow (582 nm) and finally to red (607 nm) (Fig. 5C), which, in turn, undoubtedly demonstrates the high tunability of CT states in the constructed

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**Fig. 4. Theoretical investigation of the CT processes in light-harvesting systems.** (A) Molecular orbital diagrams of OPV, C\textsubscript{60}, and CT complex calculated by density function theory. (B) Schematic of efficient exciton funneling for the formation, accumulation, and radiative deactivation of CT excitons in the C\textsubscript{60}@OPV light-harvesting systems. (C) Maximum emission wavelength of microcrystals versus C\textsubscript{60} doping concentration.
light-harvesting microcrystals. More experimental evidence of the lasing actions in other microcrystals with varied doping concentrations can be found in figs. S17 and S18. The similar lasing thresholds of C60@OPV microwires with different doping concentrations (fig. S19) further demonstrate that exciton funneling facilitates the buildup of population inversion for lasing emission (15, 32). The chromaticities of laser outputs from the doped OPV microcrystals were calculated and marked on the CIE1931 chromaticity diagram (fig. S20). Any lasing color on the dashed line can be obtained by precisely controlling the C60 doping concentration. Furthermore, benefiting from the introduction of C60, the C60@OPV microwire exhibits some ambipolar characteristics (fig. S21) (42). This ambipolar transport behavior, together with the lasing emission from CT complexes, represents a key step toward the ultimate goal of organic laser diodes.

**DISCUSSION**

We report exciton funneling and stimulated emission in light-harvesting organic semiconductor microcrystals. CT complexes with narrow bandgap are surrounded by the host molecules with high-lying energy levels, which serve as exciton funnels to harvest the energy and concentrate excitons into the CT states, and thus promote the accumulation of population in CT complexes for effective lasing. In addition, the emission of light-harvesting microcrystals was modulated by varying the degree of CT, which was used to build wide wavelength-tunable microlasers. The results not only provide an insight into the exciton funneling in light-harvesting systems but also offer a promising route to developing organic materials with high optical gain and balanced carrier mobilities for achieving electrically driven lasers toward full-color laser displays.

**MATERIALS AND METHODS**

**Synthesis of model compound**

The model compound OPV used in this work was synthesized via Knoevenagel condensation reaction (fig. S1). The starting reagents, 2,5-dibromobenzene-1,4-dicarbaldehyde and 4-(diphenylamino) benzaldehyde, were purchased from J&K Chemical Ltd. (Beijing, China) and purified by recrystallization. Phenylboronic acid, tetrakis(triphenylphosphine)palladium, potassium tert-butoxide, tetra- butyl ammonium hydroxide, and tosylmethyl isocyanide were purchased from Sigma-Aldrich and used without further treatment. Organic solvents—toluene, dichloromethane, chloroform, tetrahydrofuran, and methanol—were dried and distilled before use.

**Preparation of light-harvesting microcrystals**

The light-harvesting microcrystals were prepared with solvent evaporation–induced self-assembly. In a typical preparation, 30 µl of 2 mM OPV in chlorobenzene was first mixed with 30 µl of C60 chlorobenzene solution to form a homogeneous mixture, which was then placed in a 50-ml closed beaker containing 3 ml of cyclohexane anti-solvent. In the whole procedures, a solvent atmosphere was kept and the evaporation speed of the solvent (i.e., the saturation degree of the solution) can be well controlled. A large number of doped microcrystals were finally obtained after 12 to 24 hours. The C60 doping concentrations in OPV were adjusted by changing the concentrations of C60 chlorobenzene solution from 0, 1 mM, 2 mM,
The C$_{60}$ doping concentration in the light-harvesting microcrystals was measured. As the Lambert-Beer’s law was obeyed for both compounds, redissolved in chlorobenzene, of which absorption spectra were calculated by solving a system of equations.

The 1H nuclear magnetic resonance (NMR) spectrum of OPV in CDCl$_3$ was determined on a Bruker AVIII 500 MHz NMR spectrometer. The morphology of the as-prepared microwires was characterized by SEM (Hitachi SU8010) and TEM (JEOL JEM-1011). The crystal structure of the microcrystals was examined by SAED (JEOL JEM-1011) and XRD (Japan Rigaku D/max-2500) with Cu Kα radiation (λ = 0.154 nm). Confocal images were recorded with a laser confocal fluorescence microscope (Olympus FV1000-IX81) equipped with a 405-nm laser. The field-effect transistor characteristics were measured at room temperature in air on a Keithley 4200-SCS and micro-manipulator 6150 probe station, and the carrier mobility ($\mu$) was extracted from the saturation region through the equation of $I_{DS} = (W/2L)C_i(V_G - V_T)^2$, where $I_{DS}$ is the drain current; $L$ and $W$ are the channel length and width of devices, respectively; $C_i$ is the capacitance of the gate dielectric capacitance per unit area; and $V_T$ and $V_G$ are the threshold voltage and gate voltage, respectively.

The absorption and PL spectra were measured with Hitachi UH4150 and Hitachi F-7000 fluorescence spectrometers, respectively. The absolute quantum yields were measured by using a steady and transient fluorescence spectrometer (Edinburgh instruments, FLS980). The Raman spectra of individual microcrystal were conducted with a Thermo Fisher DXRxi Raman spectrometer (excitation wavelength, 780 nm; laser power, 8 mW). The time-resolved PL spectra of the microcrystals were measured with a streak camera (Hamamatsu Photonics, C10910).

Optically pumped lasing measurements were carried out on a home-built far-field micro-PL system. The excitation pulses (400 nm, ~100 fs, 1 kHz) were generated from the second harmonic of the fundamental output of a regenerative amplifier (Solstice, Spectra-Physics; 800 nm, ~100 fs, 1 kHz), which was, in turn, seeded by a mode-locked Ti:sapphire laser (Mai Tai, Spectra-Physics; 800 nm, ~100 fs, 80 MHz). The microwires on glass substrates (refractive index, ~1.5) were locally excited with a laser beam focused down to ~20 μm in diameter through an objective (Nikon CFLU Plan; 50x; numerical aperture, 0.8), with input power altered by neutral density filters. After passing through a 420-nm long-pass emission filter, the collected PL signal from the microwires was subsequently coupled to a grating spectrometer (Acton SP-2358) and recorded with a thermal electrically cooled charge-coupled device (Princeton Instruments, ProEm: 1600B).

The growth morphology of the OPV crystal was predicted on the basis of the Bravais-Friedel-Donnay-Harker model using the Materials Studio package. The electric field intensity distribution in the C$_{60}$@OPV microwire was calculated by using the commercial software COMSOL. The geometry optimizations of the molecular complex and the individual molecules were performed on Gaussian 09 software with B3LYP function and the 6-31G basis set. After the optimization by the density functional theory calculation, the electron-cloud density of HOMO and LUMO and their corresponding orbital energies were obtained. The vertical excited energies and charge difference densities were calculated with optimized geometries using CAM-B3LYP/6-31G+(d,p).

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Acknowledgments: We appreciate the discussions with B. Qiu from the Institute of Chemistry, Chinese Academy of Sciences. We thank C. Li and L. Jiang from the Institute of Chemistry, Chinese Academy of Sciences, for the measurements of FET characteristics. Funding: This work was financially supported by the Ministry of Science and Technology of China (grants 2017YFA0404502 and 2015CB932404), the National Natural Science Foundation of China (grants 21773265, 21790364, and 21533013), and Youth Innovation Promotion Association CAS (grant 2014028). Author contributions: Y.S.Z. conceived the original concept. Y.S.Z., Y.Y., and J.Y. supervised the project. K.W. designed and performed the experiments and prepared}

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the materials. K.W., Z.G., X.L., Z.Z., and H.M. performed the optical measurements. K.W., W.Z., H.S., and A.X. put forward the theoretical model and contributed to the theoretical calculations. K.W., Y.Y., J.Y., and Y.S.Z. analyzed the data and wrote the paper. All authors discussed the results and commented on the manuscript.

**Competing interests:** The authors declare that they have no competing interests.

**Data and materials availability:** All data needed to evaluate the conclusions in the paper are present in the paper and/or the Supplementary Materials. Additional data related to this paper may be requested from the authors.

**Citation:** K. Wang, Z. Gao, W. Zhang, Y. Yan, H. Song, X. Lin, Z. Zhou, H. Meng, A. Xia, J. Yao, Y. S. Zhao, Exciton funneling in light-harvesting organic semiconductor microcrystals for wavelength-tunable lasers. *Sci. Adv.* 5, eaaw2953 (2019).
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Sci Adv 5 (6), eaaw2953.
DOI: 10.1126/sciadv.aaw2953

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