Supporting Information for

Tailoring the Structures and Photonic Properties of Low-Dimensional Organic Materials by Crystal Engineering

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Experimental Section

The preparation of 2D microsheets: In a typical preparation, a stock solution containing 1.0 mM DCF in the mixed solvent of methanol and acetone (0:1, v/v) heated at 60°C for 5 min by using an electric-heated thermostatic water bath. Then a drop of DCF solution was placed on substrates that were ultrasonically cleaned with iso-Propyl alcohol and acetone for 30 min, respectively. With the solvents evaporating, 2D microsheets were obtained finally.

The preparation of 1D mircrorods: In a typical preparation, a stock solution containing 1.0 mM DCF in the mixed solvent of methanol and acetone (4:1, v/v) heated at 60 °C for 5 min by using a electric-heated thermostatic water bath. Then a drop of DCF solution was placed on substrates that were ultrasonically cleaned with iso-Propyl alcohol and acetone for 30 min, respectively. With the solvents evaporating, 1D mircrorods were obtained finally.

Measurements: The mircrorods and microsheets were transferred onto different substrates for measurements by SEM (Hitachi, S-4800), TEM (Tecnai, F20), PL spectroscopy (Hitachi, F-4500) and X-ray diffraction (XRD, Japan Rigaku D/max-2500). The schematic demonstration of the experimental setup for optical characterization is shown in Fig. S10†. PL images and confocal images were taken with an Olympus FluoView-500 positive microscope. To measure the PL spectra of single mircrorod and microsheet, the mircrorod and microsheet were excited locally with a 445 nm argon ion laser (Spectra-Physics, Beamlok2065, 150 nW) focused down to the diffraction limit. The excitation laser was filtered with a 445 nm notch filter. The light was subsequently coupled to a grating spectrometer (Acton SP-2358) and recorded by a thermal electrically cooled CCD (Princeton Instruments, ProEm: 1600B). PL microscopy images were taken with an inverted microscope (Nikon, Ti-U).
Synthesis of DCF (1,7-dicarbazole-9-fluorenone):

Scheme S1. Synthesis route of target compound DCF

Step: 2,7-diiodofluorenone (500.6 mg, 1.2 mmol), carbazole (601.6 mg, 3.6 mmol), anhydrous potassium carbonate (2.08 g, 15.1 mmol), copper-bronze (471.0 mg, 7.4 mmol) and 18-crown-6 (107.8 mg, 0.4 mmol) were dissolved in anhydrous o-dichlorobenzene (5 mL). The brown mixture was degassed with 5 cycles vacuum-nitrogen and heated to 180 °C under nitrogen atmosphere for 48 hours. The reaction mixture was filtered over Celite and washed with dichloromethane. The solvents were removed in vacuo, and the residue was purified by flash chromatography on silica gel (petroleum ether/dichloromethane 7:3)( 582.0 mg, 95%).

Chemicals and solvents used in synthesis were reagent-grade reagents purchased from Aldrich.
Figure S1. Energy dispersive X-ray (EDX) element analysis of the DCF microsheet. The EDX microanalysis proves the presence of C, N, O elements, testifying the chemical compositions of the DCF microsheet.
Table S1. Single crystal date of DCF

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<tr>
<th>Compound</th>
<th>DCF</th>
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<tr>
<td>Empirical formula</td>
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<tr>
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Figure S2. ORTEP drawings of DCF crystal structure with 50% probability thermal ellipsoids
Figure S3. X-ray diffraction patterns of DCF 1D microrods, 2D microsheets and simulated powder pattern using Mercury software.
Figure S4. Optimized structure of DCF-CH$_3$OH complex by DFT calculation at B3LYP/6-31G(d,p) level. The calculation result indicates that the hydrogen-bonding is formed between the DCF molecule and the protic solvent molecules.
Figure S5. PL microscopy images of DCF structures: controlled synthesis of DCF structures by adjusting the volume ratio of alcohol/acetone (A = 1/4, B = 2/3, C = 3/2, D = 4/1). All scale bars are 20 μm.
Figure S6. A) The dark-field image of 1D microrod. Scale bar =10 μm. B) The dark-field image of 2D microsheet. Scale bar =20 μm.
Figure S7. A) Bright-field and PL images obtained from a single 2D sheet by exciting the sheet at different positions. Scale bar =20 μm; B) Spatially resolved PL spectra from the tip of the sheet for different separation distances between the excitation spot and tip of the sheet shown in A). C) The ratio of the intensity $I_{\text{tip}}/I_{\text{body}}$ against the distance $D$. Curves were fitted by an exponential decay function $I_{\text{tip}}/I_{\text{body}} = A \exp(-RD)$.
Figure S8. (A) The view of the unit cell of DCF crystal at [100] direction. Solid white arrows (1, 2) show the components of the transition dipole moments for the molecules in the [100] direction; (B) The view of the unit cell of DCF crystal at [010] direction. Solid white arrows (3) show the components of the transition dipole moments for the molecules in the [010] direction.
Figure S9. PL images obtained from a single sheet by exciting the left (A) and the right (B) with the same excitation power ($\lambda = 445$ nm). (C) The corresponding spatially resolved PL spectra of the four edges of the microsheet shown in (C). All scale bars = 20 μm.
Figure S10. Schematic demonstration of the experimental setup for the optical characterization. PL microscopy images were taken with an inverted microscope (Nikon, Ti-U). To measure the PL spectra of the nanowires or nanosheets, the samples were excited locally with a 445 nm argon ion laser (Spectra-Physics, Beamlok2065) focused down to the diffraction limit through an objective (Nikon CFLU Plan, 50 ×, N.A. = 0.8). The power at the input was altered by the neutral density filters. The emissions from the distal ends of nanowires and nanosheets were dispersed with a grating (150 G/mm) and recorded with a thermal-electrically cooled CCD (Princeton Instruments, ProEm: 1600B).