

## Supporting Information

### **Self-Assembled Organic Hexagonal Micro-prisms With High Second Harmonic Generation Efficiency For Multifunctional Photonic Devices**

Haihua Zhang,<sup>a</sup> Qing Liao,\*<sup>a</sup> Xuedong Wang,<sup>b</sup> Zhenzhen Xu,<sup>a</sup> Yishi Wu,<sup>b</sup> Hongbing Fu\*<sup>ab</sup>

<sup>a</sup> Beijing Key Laboratory for Optical Materials and Photonic Devices, Department of Chemistry, Capital Normal University, Beijing 100190, P. R. China

<sup>b</sup> Beijing National Laboratory for Molecular Sciences (BNLMS), Institute of chemistry, Chinese Academy of Sciences, Beijing 100190, P. R. China

## Experimental Section

### 1. Materials

The compound of 3-methyl-4-methoxy-4'-nitrostilbene (MMONS) was synthesized according to literatures. The solvents of tetrahydrofuran (THF, HPLC grade) and methanol (HPLC grade) were purchased from Beijing Chemical Agent Ltd., China. Ultrapure water with a resistivity of  $18.2 \text{ M}\Omega\cdot\text{cm}^{-1}$ , produced by using a Milli-Q apparatus (Millipore), were used in all experiments.

### 2. Preparation of organic micro-prisms.

In brief, a stock solution of MMONS (12.5mM) in tetrahydrofuran (THF) was pre-prepared. Then 50 $\mu\text{L}$  of this solution was rapidly injected into 1.0mL mixture of deionized water and methanol (volume ratio, v: v = 2:3) at room temperature under shaking. After aging at room temperature for 3 h, large amount of MMONS HMPs were obtained. Finally, precipitate was centrifugally separated from the colloidal suspension and washed twice using water prior to vacuum drying.

### 3. Structural characterization of MMONS HMPs.

The morphologies and sizes of the sample were examined using field emission scanning electron microscopy (FESEM, Hitachi S-4300) at acceleration voltages of 10-15 kV. Prior to analysis, the samples were coated with a thin platinum layer using an Edwards Sputter Coater. TEM images were obtained by a JEOL JEM-1011 transmission electron microscopy (TEM). One drop of the as-prepared colloidal dispersion was deposited on a carbon-coated copper grid, and dried under high vacuum. TEM measurement was performed at room temperature at an accelerating voltage of 100 kV.

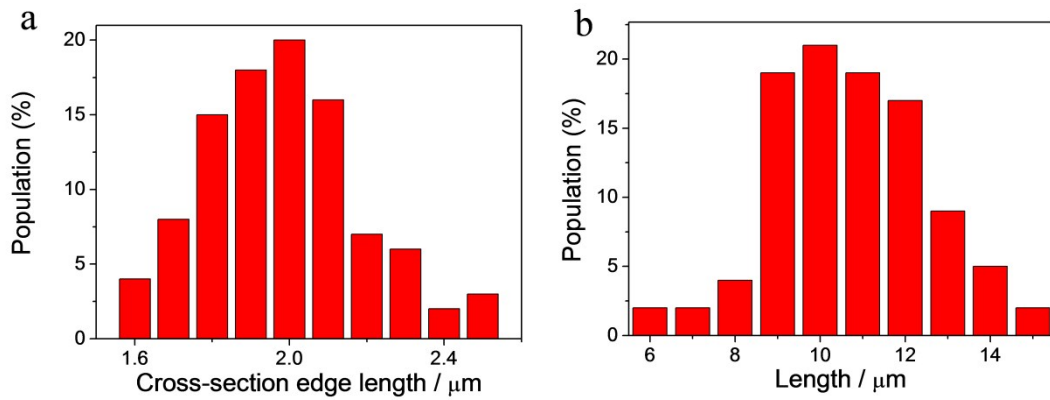
Fluorescence images of DP micro-ribbons in Figure S5 were recorded using an Olympus research inverted system microscope (FV1000-IX81, Tokyo, Japan) equipped with a charge couple device (CCD, Olympus DP71, Tokyo, Japan) camera. The excitation source is a Xenon lamp equipped with a band-pass filter (330~380 nm for UV-light, 460-490 nm for blue plight). The samples were prepared by placing a drop of dispersion onto a cleaned quartz slide.

### 4. SHG measurement of MMONS HMPs.

MMONS HMPs was investigated at room temperature in air by a home-made optical microscopy equipped with a  $50 \times 0.9 \text{ NA}$  excitation objective. The excitation laser pulses (800-1200 nm) for the SHG experiment were supplied by an optical parametric amplifier (OPA-

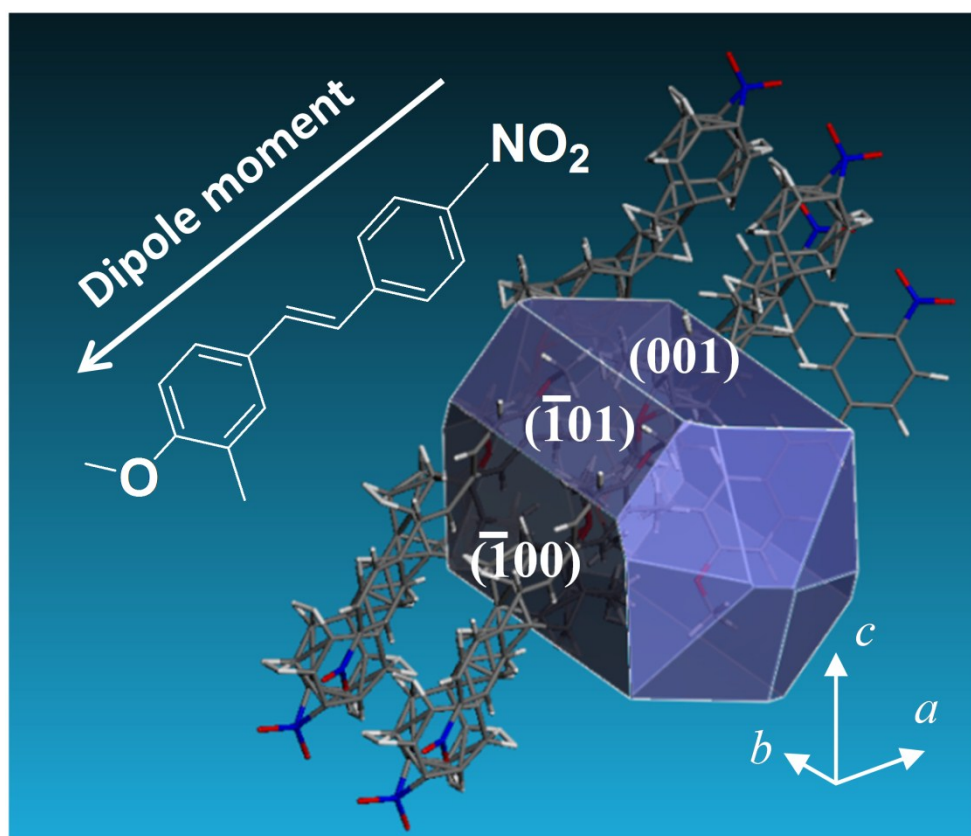
800CF, Spectra Physics), which was pumped by the output from a regenerative amplifier (Spitfire, Spectra Physics). The output laser pulse was then focused to a 1.5- $\mu\text{m}$ -diameter spot to pump the selected individual HMP. The micro-area photoluminescence ( $\mu\text{-PL}$ ) images were recorded by using a CCD (DVC-1412AM high-resolution digital camera) in a reflective mode. Then  $\mu\text{-PL}$  spectra were collected underneath by using another  $50 \times 0.9$  NA objective that was mounted a 3D movable stage. Finally the collected  $\mu\text{-PL}$  was coupled to an optical fiber and detected using a liquid-nitrogen-cooled CCD (SPEC-10:100BR, Roper Scientific) attached to a polychromator (Spectropro-550i, Acton). The spectral resolution is 0.1 nm. If necessary, we could record the spatially resolved PL spectra along the body of the selected MMONS HMP with a spatial resolution about 1  $\mu\text{m}$ .

**Figure S1**



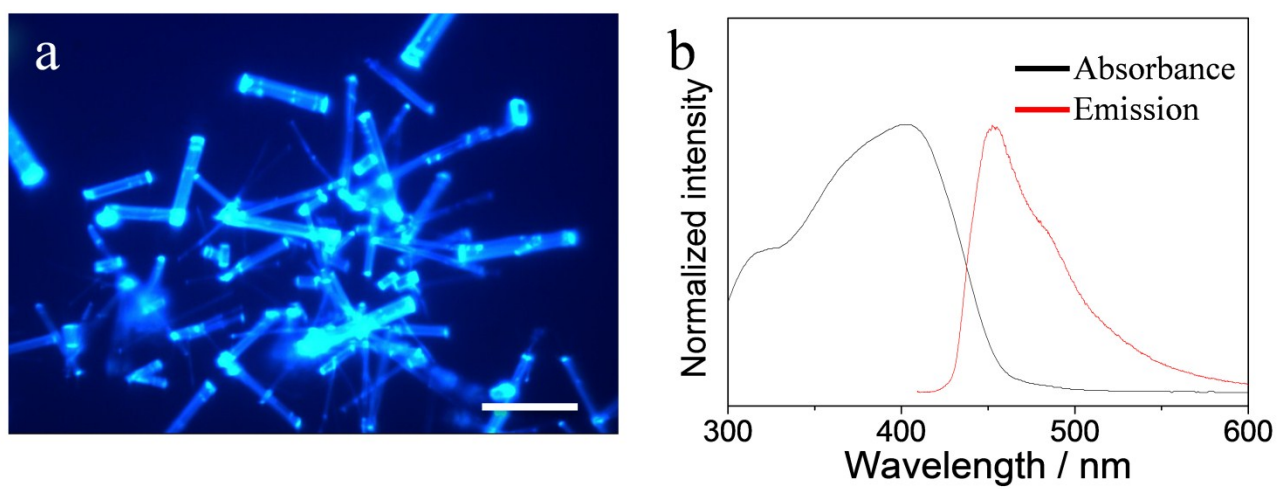
**Figure S1.** Typical size distribution accounted by 100 HMPs in once preparation.(a) Distribution of the edge length of hexagonal cross-section.(b) Distribution of length of HMPs along the axis orientation.

**Figure S2**



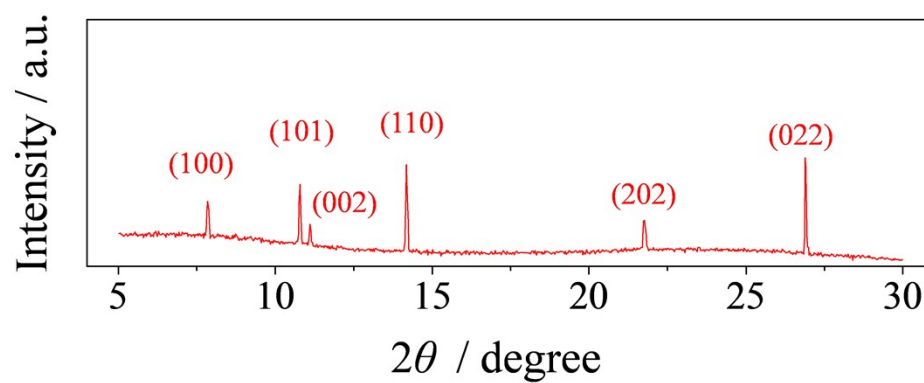
**Figure S2.** Theoretically predicted growth morphology of a single crystal of MMONS. Inset: molecular structure and dipole moment orientation of MMONS.

**Figure S3**



**Figure S3.** (a) Photoluminescence (PL) microscopy image of ensemble DP micro-ribbons on a quartz substrate excited with un-focused UV light (330–380 nm). The scale bar is 20  $\mu\text{m}$ . (b) UV–vis absorption (black) and photoluminescence (PL) (red) spectra of DP micro-ribbon.

**Figure S4**



**Figure S4.** XRD patterns of assemblies of MMONS HMPs.

