## <sup>1</sup> Electronic Supplementary Information

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## 3 Preparation

Bulk g-C<sub>3</sub>N<sub>4</sub> was obtained by heating thiourea (Aldrich, 99%) at 550 °C for 2h 4 [1]. The  $g-C_3N_4$  nanosheets were synthesized by thermal oxidation method [2], in 5 which grinded bulk g- $C_3N_4$  powders were placed in an open ceramic container and 6 was heated at 500 °C for 2 h with a ramp rate of 2 °C/min. Then, g-C<sub>3</sub>N<sub>4</sub> nanosheets 7 (0.05 g) were oxidized in concentrated H<sub>2</sub>SO<sub>4</sub> (10 mL) and HNO<sub>3</sub> (30 mL) for 16 h 8 9 under mild ultrasonication (500 W, 40 kHz). A clear solution was formed, which was then diluted with deionized (DI) water (200 mL) to produce a colloidal suspension. 10 CNNRs were obtained after the colloidal suspension was filtered through a 0.45-um 11 12 microporous membrane to remove the acids. The obtained CNNRs were carefully redispersed in 16 mL DI water under ultrasonication. The suspension was transferred to 13 a poly(tetrafluoroethylene) (Teflon)-lined autoclave (20 mL) and heated at 200 °C for 14 15 10 h. After cooling to room temperature, the final product of yellowish CNQDs solution (~0.14 g/L) was obtained after the obtained solution was filtered through a 16 0.22-µm microporous membrane. 17

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## 19 Characterizations

20 TEM images of the samples were observed on FEI Tecnai G2 Spirit transmission
21 electron microscopy. HRTEM of the CNQDs was obtained on a FEI Tecnai F20
22 transmission electron microscopy operated at 200 kV. FT-IR spectra were recorded on

1 a Nicolet Nexus 670 FT-IR spectrometer at a resolution of 4 cm<sup>-1</sup>. UV-vis absorption 2 of the samples were obtained using a Varian Cary 500 UV-vis spectrophotometer equipped with a Labsphere diffuse reflectance accessory. Powder samples were 3 prepared by dripping 5 and 10 mL CNQDs solution into 50 mg g-C<sub>3</sub>N<sub>4</sub> under 4 ultrasonication followed by freeze drying, and were denoted as CN-5 and CN-10, 5 respectively. XPS spectra were analyzed by Perkin-Elmer PHI 5000 X-ray 6 photoelectron spectroscopy. All the binding energy values were calibrated by using 7 C1s = 284.6eV as a reference. The PL study was carried out on a Hitachi F-4500 8 fluorescence spectrophotometer. 9

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## 11 Photocatalytic H<sub>2</sub> production

The photocatalytic H<sub>2</sub> evolution experiments were carried out in a Pyrex reaction cell 12 connected to a closed gas circulation and evacuation system. 50 mg of photocatalyst 13 were dispersed in 100 mL of aqueous solution containing 10 vol% triethanolamine as 14 15 a sacrificial agent. Appropriate amount of the as-prepared CNQDs were added to the suspension. The suspension was purged with argon to remove dissolved air before 16 irradiation. 1% Pt was loaded onto the samples by photoreduction of H<sub>2</sub>PtCl<sub>6</sub> [3]. The 17 solution was irradiated by a 300 W xenon lamp equipped with a 420 nm or 600 nm 18 cut-off filter. The amount of hydrogen generated from photocatalytic water splitting 19 was measured by Techcomp GC7900 gas chromatography with TCD detector and a 20 21 capillary column (molecular sieve 5 Å). High purity nitrogen gas was used as a carrier 22 gas.



Fig. S1. FT-IR spectra of the bulk g-C<sub>3</sub>N<sub>4</sub>, g-C<sub>3</sub>N<sub>4</sub> nanosheets, CNNRs and CNQDs.
The peak at 1639 cm<sup>-1</sup> corresponds to the typical stretching modes of CN heterocycles
[4], suggesting the triazine units remain in the CNQDs. The strong peak at 1385 cm<sup>-1</sup>
can be ascribed to stretching vibrations of –C–O in the hydrophilic carboxylate groups
[5], which makes CNQDs highly soluble in water.



Fig. S2. High-resolution XPS spectra of (a) C1s and (b) N1s obtained from CNQDs. 9 The C1s spectra can be deconvoluted into four component peaks. The peaks located at 10 285.5 and 286.1 eV reflect different bonding structures of the C-N bonds 11 corresponding to the N-sp<sup>2</sup>C and N-sp<sup>3</sup>C bonds, respectively. The other two peaks at 12 287.2 eV and 289.8 eV are attributed to the C-O and C=O bonds, respectively [6]. 13 The N1s spectra can also be deconvoluted into four component peaks. The peaks at 14 399.8, 400.9, and 401.7 eV respectively corresponds to C-N-C, N-(C)<sub>3</sub>, and N-H 15 groups, which are typically found in g-C<sub>3</sub>N<sub>4</sub> triazine units. The peak at 406.6 eV is 16 attributed to  $-NO_2$  that are derived from the oxidation of amino groups [7]. The 17 atomic percentage of N in the CNQDs is about 19.94%. The above results reveal that 18 the CNQDs preserve the basic triazine structure of g-C<sub>3</sub>N<sub>4</sub>, while surface enriched 19 with amino-functional and carbonyl-functional groups 20



8 Fig. S3. (a) PL (at 360 nm excitation) spectra of the CNQDs obtained from different
9 hydrothermal temperature. (b) The upconversion PLE spectrum with the detection
10 wavelength of 440 nm.

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8 Fig. S4. (a) TEM and (b) HRTEM images of CN-10 loading with Pt after the
9 photocatalytic H<sub>2</sub> evolution experiments.







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