(Supporting Information)

## Sulphur doping: a facile approach to tune the electronic structure and optical properties of graphene quantum dots

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### 1. Chemicals

Fructose ( $\geq$ 99%) was purchased from Sigma-Aldrich. Sulphuric acid (98% *wt*.) was purchased from International Laboratory, USA. These chemicals were used without further purifying.

### 2. Supplementary Figures



Figure S1. The FTIR spectra of the S-GQDs (top) and the source fructose (bottom).



Figure S2. The Raman spectra of the S-GQDs (top) and the source fructose (bottom).



**Figure S3.** (a) STEM image of the S-GQDs assembled on Cu grid coated with ultrathin amorphous carbon film. (b) Elemental C mapping (in blue) of the image shown in panel (a). (c) Elemental O mapping (in red) of the image shown in panel (a).

(d) Elemental S mapping (in green) of the image shown in panel (a).



Figure S4. The PLE spectra of the S-GQDs recorded at various  $\lambda_{Em}$  ( \* is  $\lambda_{Ex}/2$ ).

#### 3. Supplementary Table

| $\lambda_{Em}$ | $	au_1$ | $	au_2$ | $	au_3$ | $B_1$     | $B_2$    | $B_3$  | $R_1^{(a)}$ | $R_{2}^{(a)}$ | $R_3^{(a)}$ | A       | $\chi^2$ | <7>                 |
|----------------|---------|---------|---------|-----------|----------|--------|-------------|---------------|-------------|---------|----------|---------------------|
| [nm]           | [ns]    | [ns]    | [ns]    |           |          |        |             |               |             |         |          | [ns] <sup>(b)</sup> |
| 410            | 0.55    | 3.08    | 29.63   | 58703.02  | 4020.27  | 86.99  | 0.935       | 0.064         | 0.001       | 24.89   | 4.04     | 0.75                |
| 430            | 0.56    | 3.05    | 28.45   | 161000.23 | 11930.68 | 274.80 | 0.930       | 0.069         | 0.002       | 70.91   | 9.70     | 0.78                |
| 450            | 0.61    | 3.09    | 25.21   | 222435.14 | 16917.98 | 436.25 | 0.928       | 0.071         | 0.002       | 109.99  | 12.97    | 0.83                |
| 470            | 0.61    | 2.98    | 18.52   | 288853.63 | 29637.68 | 905.60 | 0.904       | 0.093         | 0.003       | 196.85  | 9.37     | 0.88                |
| 490            | 0.61    | 3.02    | 18.86   | 293968.97 | 28826.99 | 939.26 | 0.908       | 0.089         | 0.003       | 191.54  | 9.55     | 0.88                |
| 510            | 0.58    | 2.83    | 20.31   | 304790.44 | 27628.11 | 875.74 | 0.914       | 0.083         | 0.003       | 202.93  | 7.18     | 0.82                |
| 530            | 0.65    | 3.03    | 33.57   | 415088.69 | 22360.81 | 633.11 | 0.948       | 0.051         | 0.001       | 154.19  | 29.55    | 0.82                |
| 550            | 0.60    | 2.61    | 31.01   | 370878.09 | 25347.35 | 598.31 | 0.935       | 0.064         | 0.002       | 169.80  | 14.86    | 0.77                |
| 570            | 0.65    | 2.83    | 83.95   | 425290.28 | 18188.11 | 608.49 | 0.958       | 0.041         | 0.001       | -58.94  | 49.58    | 0.85                |
| 590            | 0.58    | 2.56    | 103.60  | 384996.16 | 17902.91 | 611.97 | 0.954       | 0.044         | 0.002       | -119.60 | 28.07    | 0.82                |
| 610            | 0.50    | 2.32    | 120.00  | 289038.84 | 16327.67 | 607.85 | 0.945       | 0.053         | 0.002       | -176.77 | 9.44     | 0.83                |
| 630            | 0.48    | 2.24    | 96.37   | 191806.25 | 11425.17 | 397.44 | 0.942       | 0.056         | 0.002       | -62.55  | 5.22     | 0.77                |
| 650            | 0.48    | 2.28    | 120.00  | 111100.05 | 6650.78  | 282.32 | 0.941       | 0.056         | 0.002       | -83.30  | 3.14     | 0.87                |
| 670            | 0.53    | 2.53    | 78.81   | 76635.06  | 3124.25  | 111.64 | 0.959       | 0.039         | 0.001       | -0.88   | 4.90     | 0.72                |
| 690            | 0.50    | 2.41    | 37.16   | 34843.90  | 1874.17  | 52.60  | 0.948       | 0.051         | 0.001       | 16.98   | 1.96     | 0.65                |
| 710            | 0.56    | 2.77    | 90.56   | 22224.54  | 812.62   | 39.04  | 0.963       | 0.035         | 0.002       | -5.97   | 2.72     | 0.79                |

Table S1 The fitting parameters of PL decay curves for various  $\lambda_{Em}$ 

(a) The photoluminescence decay curves were fitted to a triple-exponential function:  $I(t) = A + B_1 \cdot e^{(-t/\tau_1)} + B_2 \cdot e^{(-t/\tau_2)} + B_3 \cdot e^{(-t/\tau_3)}, R_i \text{ is the relative ratio factor which is}$ 

calculated by  $R_i = B_i / (\sum_{i=1}^3 B_i)$ .

(b)  $\tau_{\text{average}}$  is the average lifetime,  $\tau_{\text{average}}$  is calculated according to  $\tau_{\text{average}} = \sum_{i=1}^{3} R_i \cdot \tau_i$ .