Electronic Supplementary Information

Realization of lasing emission from graphene quantum dots using titanium dioxide nanoparticles as light scatterers

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1. Experimental Results

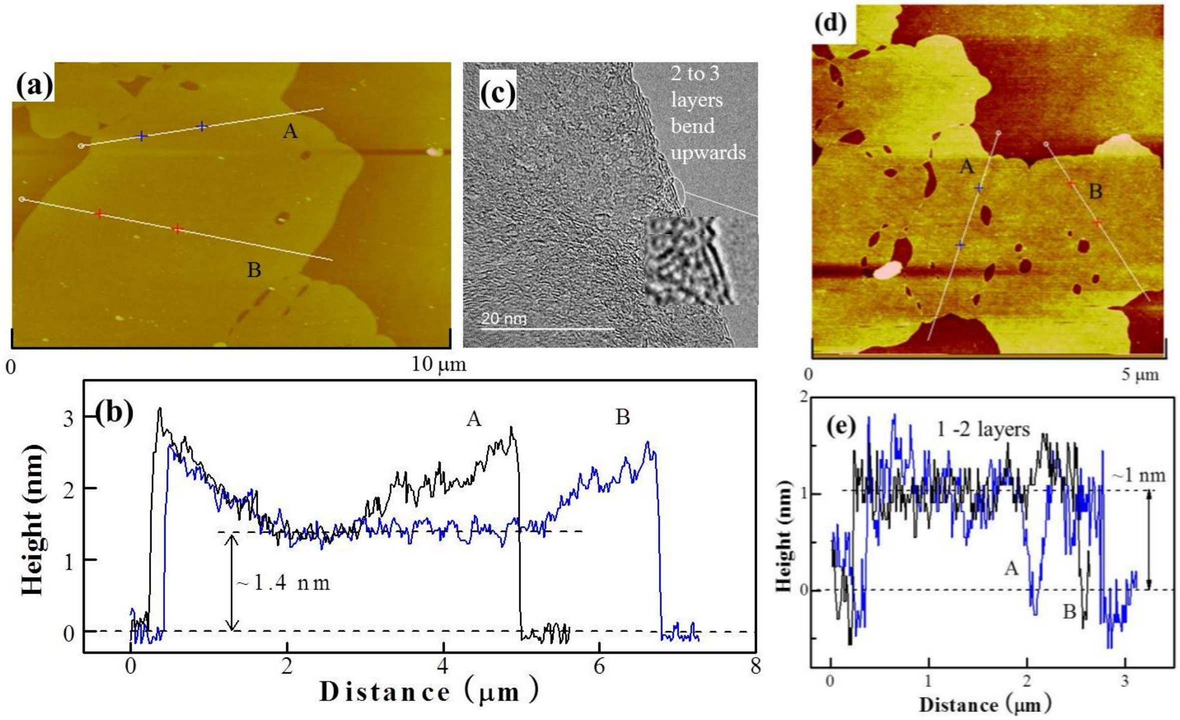


Figure S1a

Figure S1a shows an atomic force microscopy (AFM) image for a ~8×8 μm^2 graphene sheet obtained from the oxidation and reduction processes of graphite. The height profiles of two paths, *A* and *B*, of the graphene sheet were measured and the corresponding results are given in Figure S1b. It is noted that the edges of the graphene sheets were bent upwards. This can be confirmed by the high resolution -transmission electron microscopy (HR-TEM) image as shown in Figure S1c. 2 to 3 layers of carbon atoms can be observed from the TEM image of the graphene sheet as the number of bright lines at the edge represents the number of layers.[S1] Furthermore, the thickness at the centre of the graphene sheets shows to have a constant height of ~1.4 nm. Hence, it is expected that the graphene sheet has 2 to 3 layers of carbon atoms. Figure S1d shows the AFM image of a ~5×5 μm^2 graphene sheet and the corresponding height profiles are given in Figure S1e. It is noted that no bending of edges is observed and the thickness of the graphene sheet is around ~1 nm (i.e., single to bilayer graphene). These are the typical thickness of graphene sheets that can be obtained from the proposed fabrication technique.

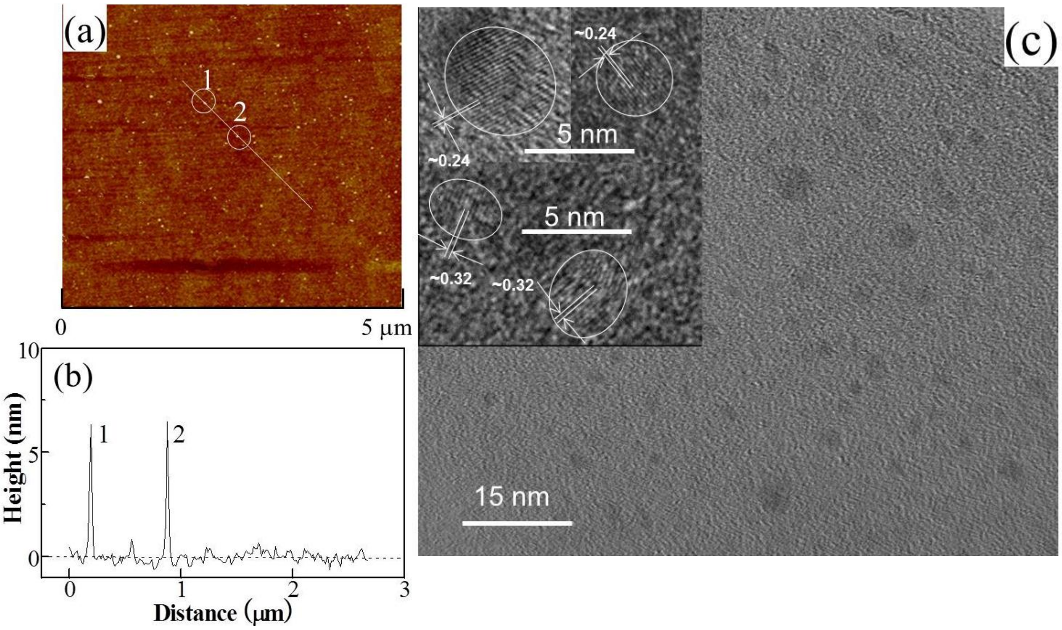
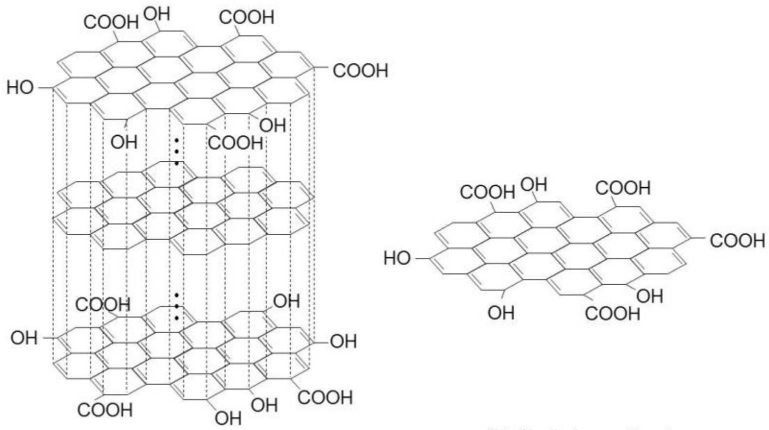


Figure S2

Figures S2a and b show an AFM image and the height profile respectively of C-dots dispersed on Si substrate. The height of the C-dots is larger than 5 nm which indicated that the C-dots have few tenths of layers of carbon atoms. Figure S2c reveals the TEM images of the as-prepared C-dots with ethanol as the solvent. These C-dots have lattice spacing of either ~0.24 or ~0.32 nm. If the C-dots have a column structure, the lattice spacing of ~0.24 nm can be considered as the distance between carbon atoms on the surface coplanar of the column structure. The lattice spacing of ~0.32 nm can be attributed to the interlayer distance of graphene sheets. Hence, it is believed that the C-dots have a column layer structure like graphite with mean diameter and height of ~3.5 and ~5 nm respectively. [S2] Figure S3 shows the schematic diagrams of a (a) C-dot and (b) GQD.



(a) Carbon nanodot

(b) Single-layer Graphene Quantum Dot

Figure S3

Figure S4a compares the absorption spectra of GQDs and C-dots dispersed inside ethanol. It is observed that C-dots have higher absorption loss throughout the entire UV and visible regimes. The photoluminescence (PL) spectra of GQDs and C-dots, which were studied with the same excitation power and wavelength of 266 nm, are also given in the inset of Figure S4a. It is noted that GQDs have higher emission intensity than that of C-dots.

Figure S4b plots the peak output intensity versus L_S of C-dots excited at different pump power, P. The width of the pump strip used in this experiment was set to about 0.05 cm. By fitting eqn (1) with the measured data (i.e., the solid lines), G at the peak wavelength can be deduced. Top left-hand-corner of Figure S4b shows the estimated value of G versus P for C-dots and the corresponding optical gain per pump power, $\partial G/\partial P$, is found to be ~1.6 cm^{-1}kW^{-1}. This value of $\partial G/\partial P$ is about 5 times lower than that obtained from GQDs.

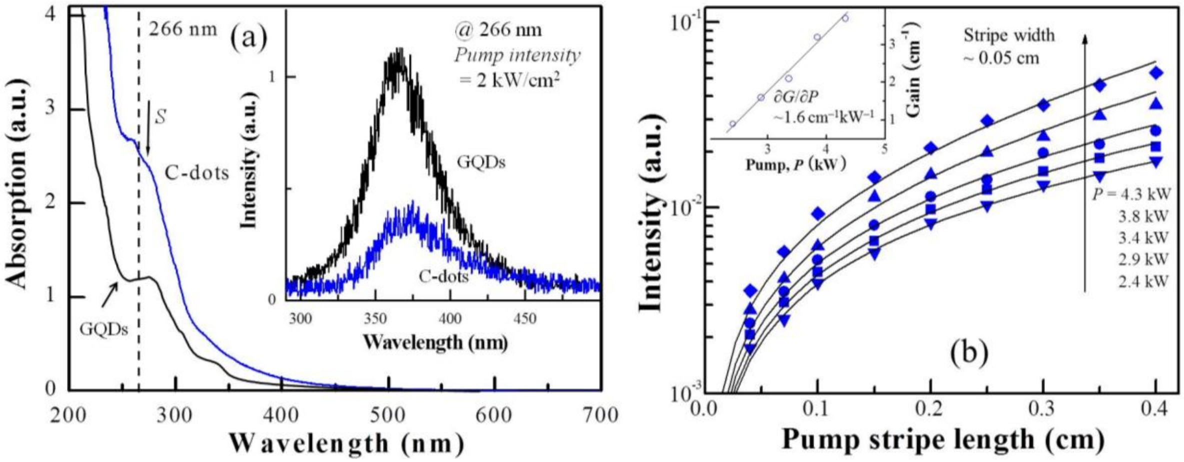


Figure S4

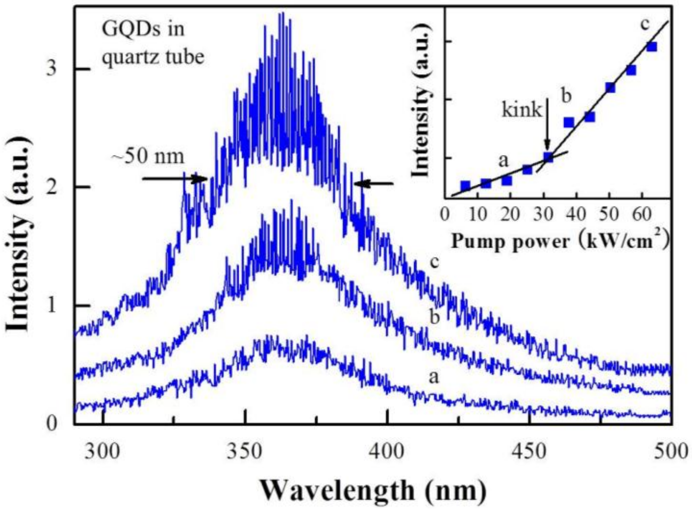


Figure S5

Figure S5 shows the emission spectra and light-light curve of the GQDs Fabry Perot laser of length L = 5 mm without TiO_2 nanoparticles. The laser cavity is under optical excitation by a 266 nm pump beam. Lasing characteristics can be verified from the measurement because 1) a kink (i.e., threshold) is exhibited in the light-light curve, and 2) sharp modes are excited from the emission spectra for pump power larger than the threshold. However, we cannot resolve the mode spacing from the measured spectra. As we know that mode spacing, $\Delta\lambda$, can be related to the emission wavelength, λ, cavity length, L, and the refractive index of the laser cavity, n, using the following equation:

$$\Delta\lambda = \frac{\lambda^2}{2Ln} \qquad \text{(S1)}$$

If L = 0.5 cm, λ = 366 nm and n = 1.65, $\Delta\lambda$ can be calculated to be ~8×10^{-3} nm or ~0.08 Å. This is too small to be resolved by the available monochromator (i.e., resolution is > 1 Å). Therefore, the lasing modes observed from the emission spectra are not truly reflected the modal characteristics of the laser.

Figure S6 compares the light-light curves of the GQDs Fabry Perot laser (with quartz tube length L = 5 mm and without TiO_2 nanoparticles) and C-dots laser given in ref [S3]. It is noted that the lasing threshold, P_{th}, of GQDs and C-dots lasers is about 30 and 180 kW/cm^2 respectively. The threshold is reduced by almost an order of magnitude for using GQDs as the gain medium. In addition, the external power efficiency (i.e., the slope of light-light curve above threshold) of GQDs laser has improved by about 400% over the C-dots laser. Hence, it is believed that GQDs are more suitable to be applied as gain media than that of C-dots. The realization of GQDs may lead to the development of practical lasers using carbon as the gain media.

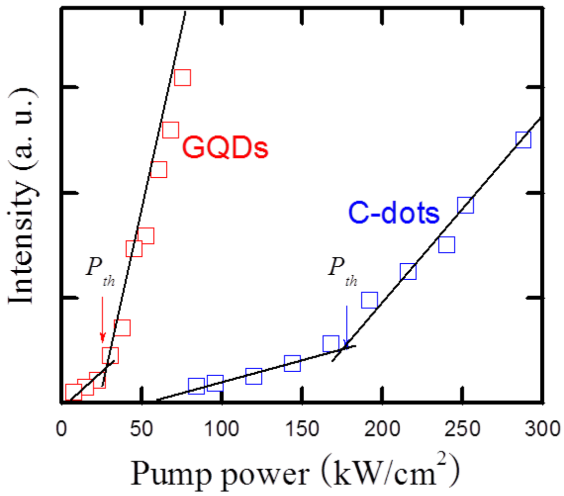


Figure S6

References

[S1] Baker, S. N.; Baker, G. A. Luminescent carbon nanodots: Emergent nanolights. *Angw. Chem. Int. Ed.* **2010**, *49*, 6726-6744.

[S2] Liu, Y.; Liu, C.Y.; Zhang, Z. Y. Synthesis and surface photochemistry of graphitized carbon quantum dots. *J. Colloid Interface Sci.* **2011**, *356*, 416-421.

[S3] Zhang, W. F.; Zhu, H.; Yu, S. F.; Yang, H. Y. Observation of lasing emission from carbon nanodots in organic solvents. Adv. Mater. 2012, 24, 2263-2267.