

**Multi-layered Graphene Quantum Dots derived Photodegradation mechanism of
Methylene Blue**

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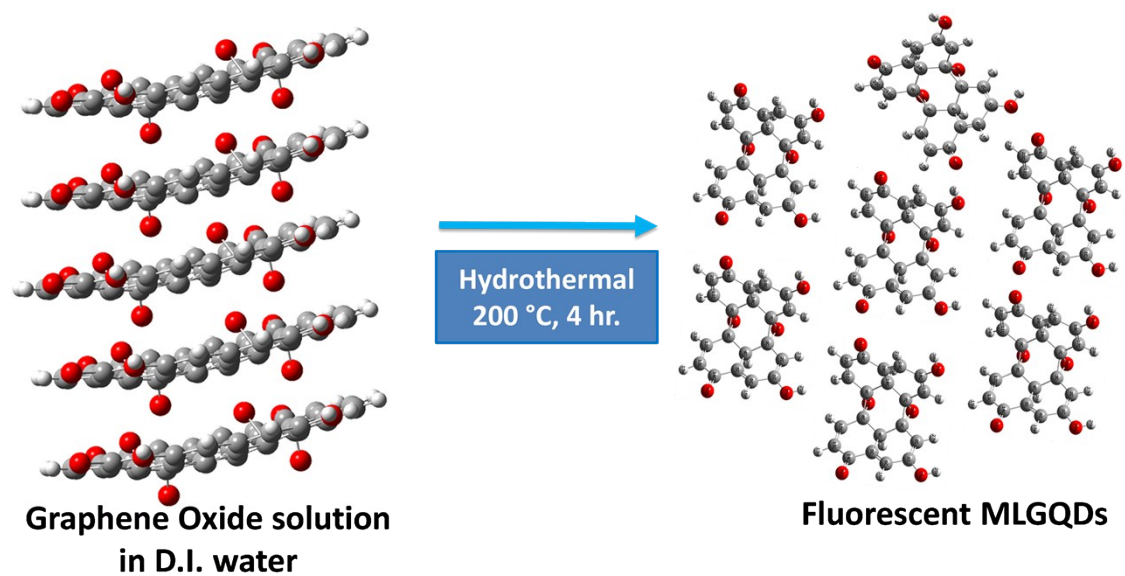


Figure S 1. Schematic for synthesis of MLGQDs by hydrothermal disintegration of GO

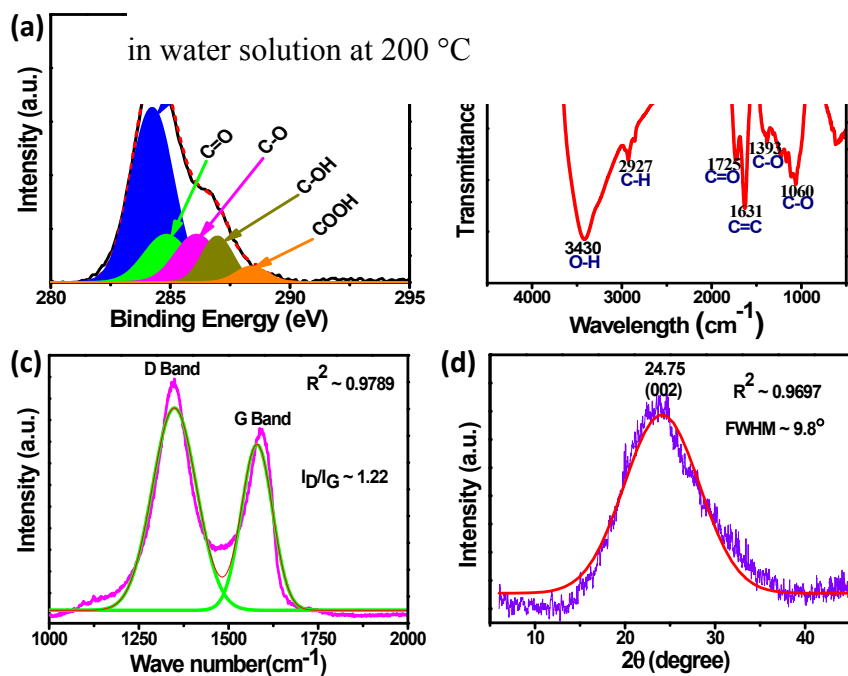


Figure S 2. (a) XPS spectra of MLGQDs, (b) FTIR spectra of MLGQDs, (c) Raman spectrum of MLGQDs, and (d) X-ray diffraction pattern of the MLGQDs.

AC impedance Spectrum:

To study about the separation of electrons and holes, the AC impedance measurement of ITO and MLGQDs electrodes in PBS solution (pH 7) containing 5 mM of $[\text{Fe}(\text{CN})_6]^{3-/4-}$ was done using three electrode system with Ag/AgCl as reference, platinum as counter and MLGQDs/ITO as working electrode at the frequency range varying from 0.01 Hz to 10 kHz as shown in **Figure S2** (see in supplementary information). The values of charge transfer resistance (R_{CT}) were recorded 133.5 ohm at open circuit voltage. A single semicircle at the high frequency region and a straight line at the low frequency region indicate a mixed charge transfer and charge diffusion process.^{S1} The R_{CT} values of the MLGQDs electrode was much smaller than that of the ITO electrode, which illustrates that MLGQDs lead to a much lower charge transport resistance and much higher separation efficiency of electrons and holes. The high separations of charge promote the enhancement of photocatalytic activity of MLGQDs.

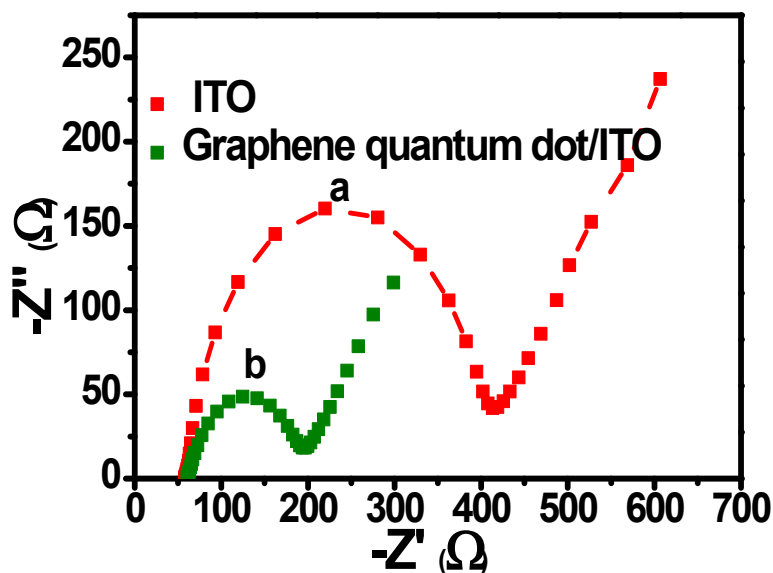


Figure S 3. AC impedance Spectra of as synthesized MLGQDs, the EIS measurements were performed in the presence of a PBS solution (pH 7) containing 5 mM $[\text{Fe}(\text{CN})_6]^{3-/4-}$.

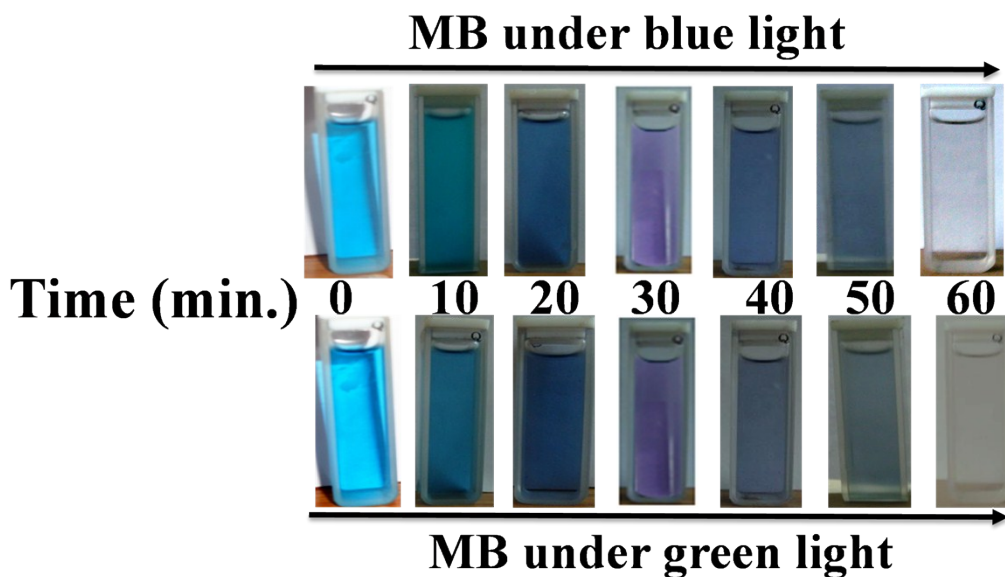


Figure S 4. Photographic image of MLGQDs-MB mixture before and after light exposure shows the color change from blue into colorless during the photodegradation of MB.

Time dependent absorbance spectrum of MB under Green and Blue light:

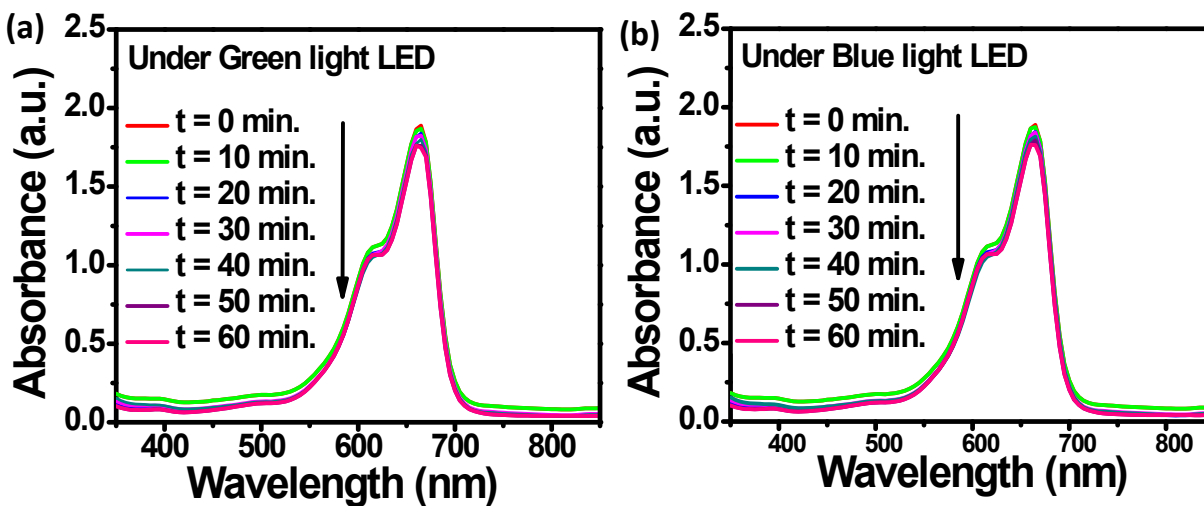


Figure S5. Time dependent absorbance of MB under green light (a) and blue light (b).

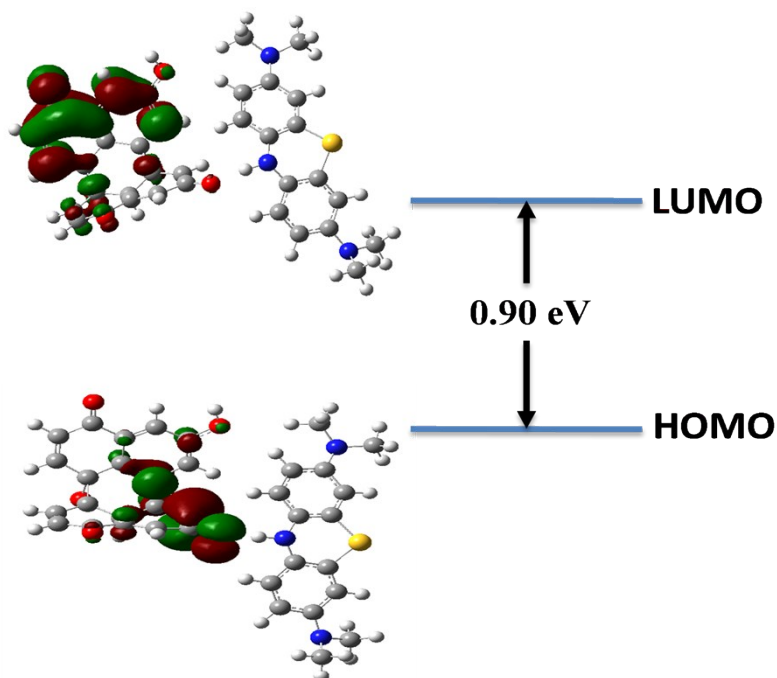
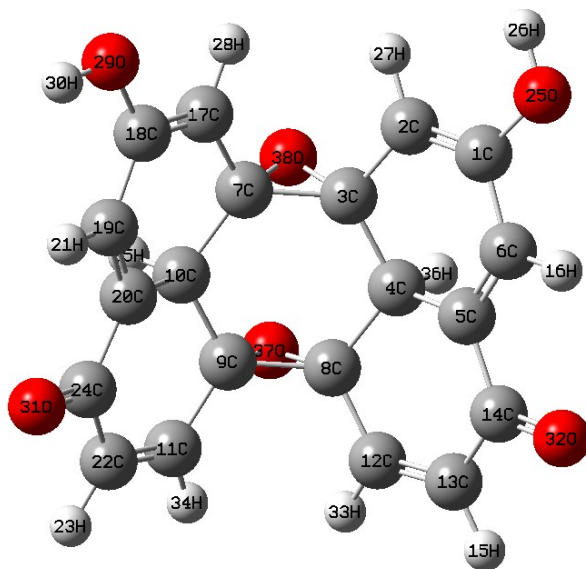
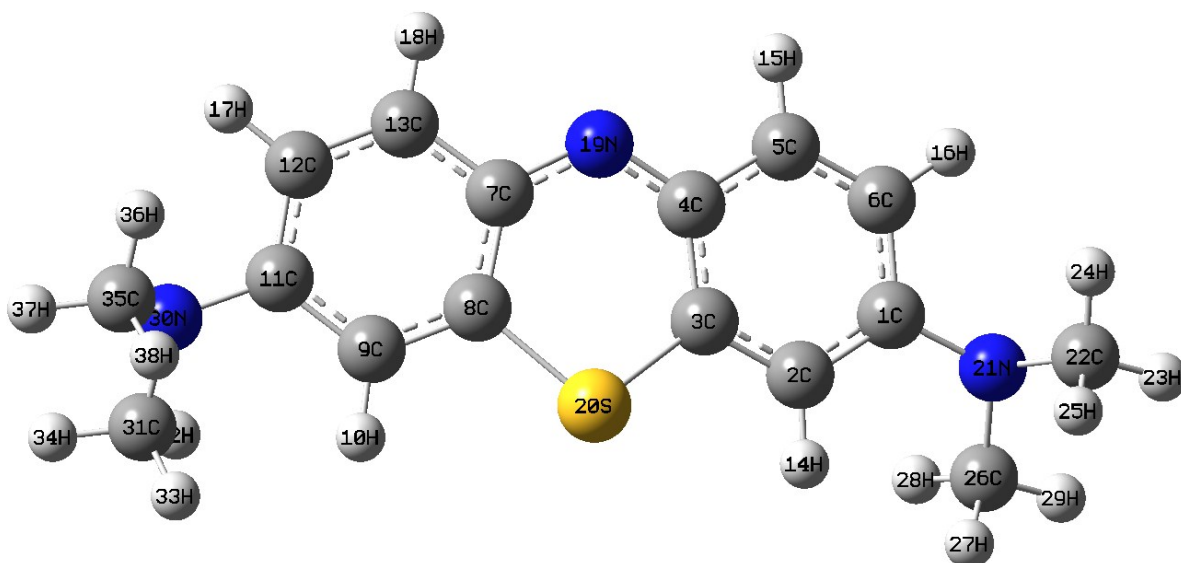


Figure S 6. Chemical structures and distribution of HOMO and LUMO for MLGQDs-MB complex in first energy band where minimal charge transfer within MLGQDs.

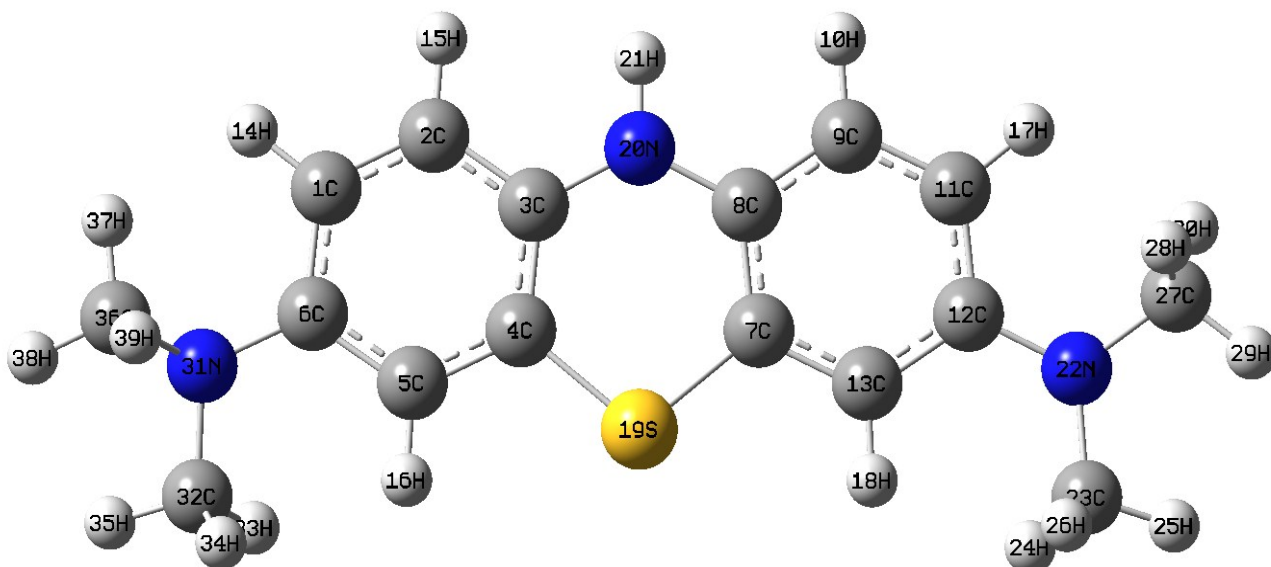
Optimize structure of MLGQDs:



Optimize spectrum of MB:



Optimize structure of LMB:



S1. Y. Liu, D.-P. Wang, Y.-X. Yu and W.-D. Zhang, *Int. J. Hydrogen Energy*, 2012, **37**, 9566–9575.