Cryo Glow: Unveiling the Chilling Brilliance of Graphene Quantum Dots

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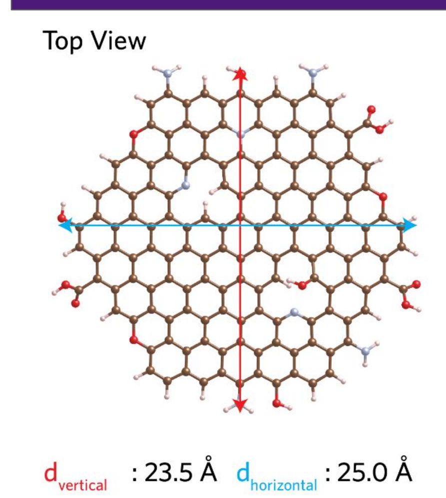
Abstract

Graphene Quantum Dots (GQDs), with their outstanding optoelectronic, chemical, and bio-compatible properties serve as versatile materials for various imaging applications. Intriguing optical properties at ultralow cryogenic temperatures have been observed in other carbon-based nanomaterials suggesting a potential for similar behavior in GQDs. This study explores GQD fluorescence across the visible and near-infrared spectral regions at temperatures ranging from ambient (300 K) down to cryogenic (76K) via experimental measurements supported by complementary DFT calculations. Our findings demonstrate a decreasing linear relationship between intensity of fluorescence temperature making GQDs a viable candidate for applications in low-temperature imaging.

Introduction



GQDs and Their Importance

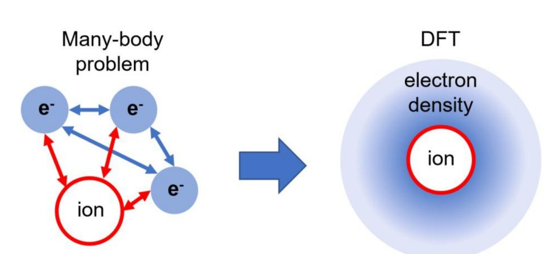
- Optoelectronic properties and NIR fluorescence offers possibilities in imaging and detection
- These properties are often masked by thermal fluctuations at higher temperatures.

Experimental Limitations:

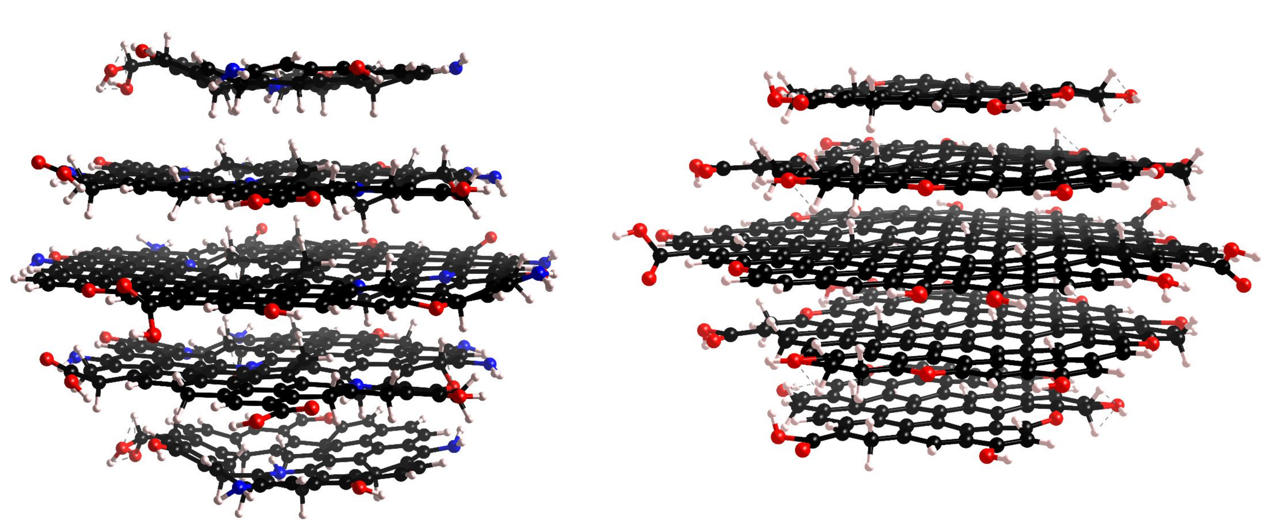
- Cryostat needs to fully depressurize to prevent condensation that influences path of light
- Location of solution must remain constant within spectrofluorometer

Modeling Experiment Computationally

- Working with bigger structures such as 100-1000 atoms, geometrical optimizations tools can be put into use
- When needed optical characterization in comparatively large models, Gaussian is a versatile tool for an experimentalist
- DFT and VASP are there to provide the deepest and most comprehensive results by conducting ab-initio quantum mechanical simulations

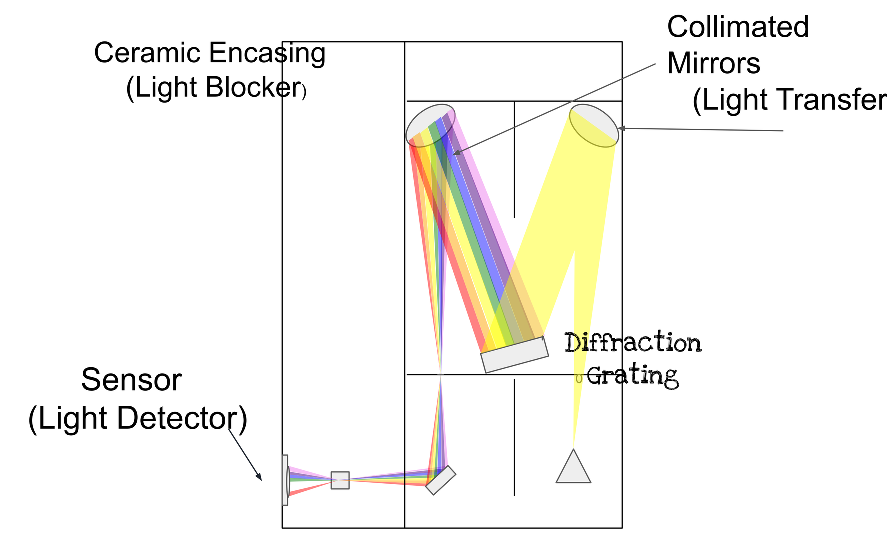


Molecular Structures of GQDs

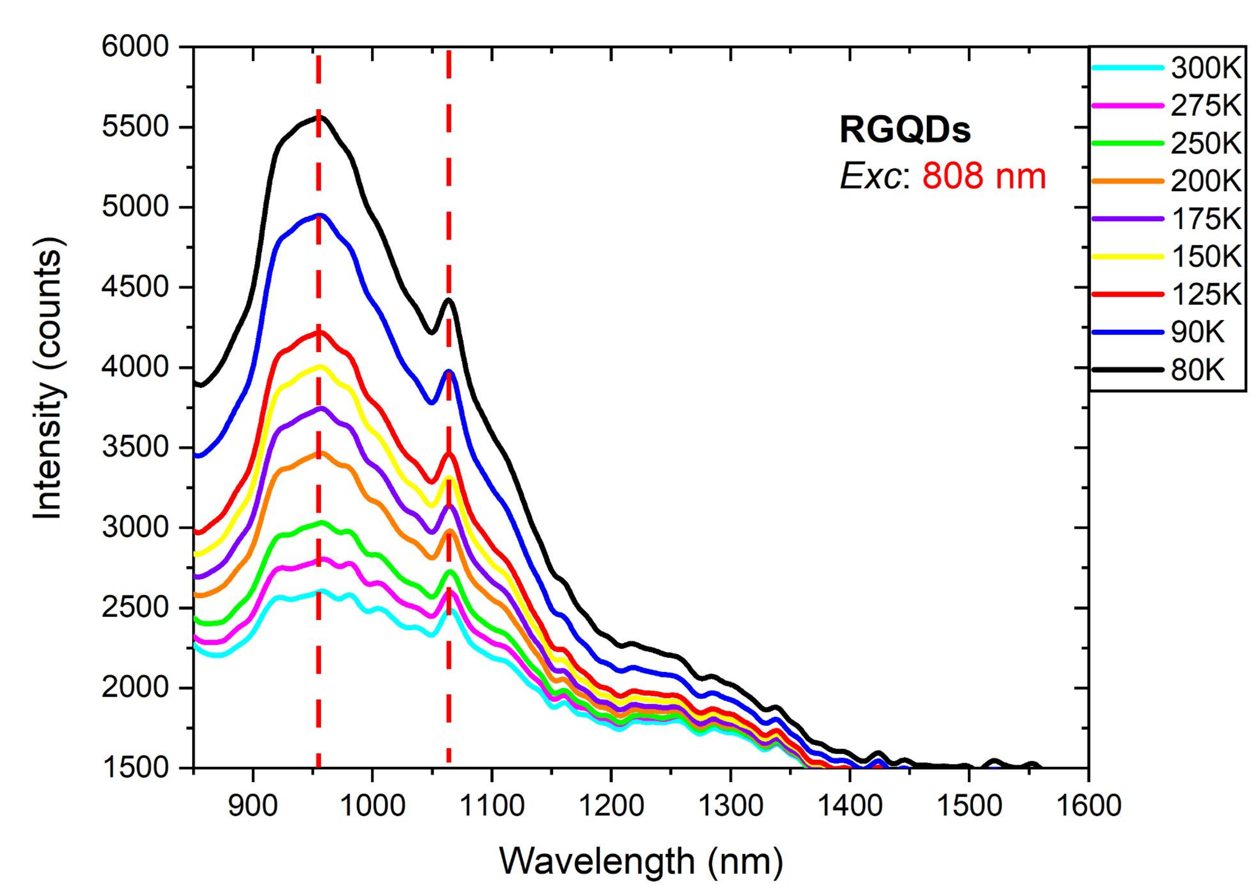


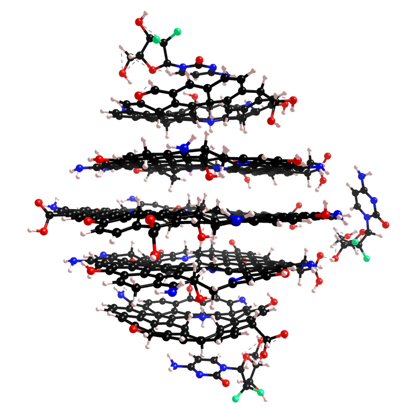
Process of Spectrofluorometry

How Does It Work?



Near Infrared Temperature-Dependent Fluorescence

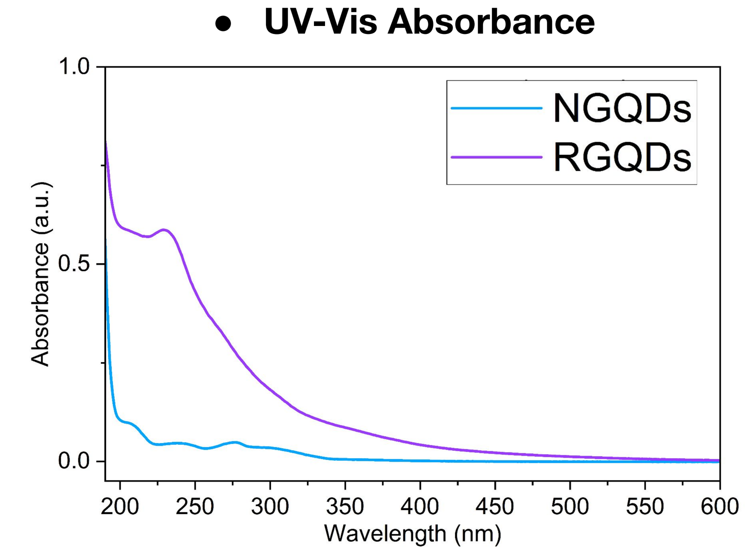




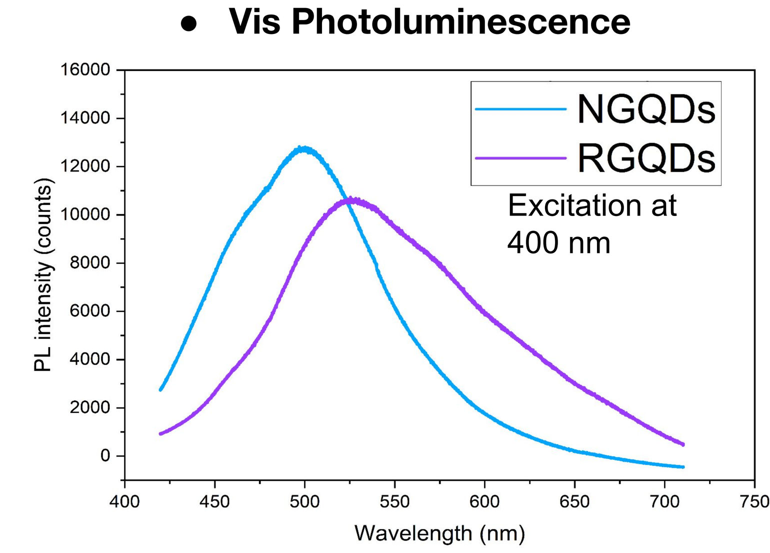
RGQD-Near IR

Experimental Characterization of GQDs

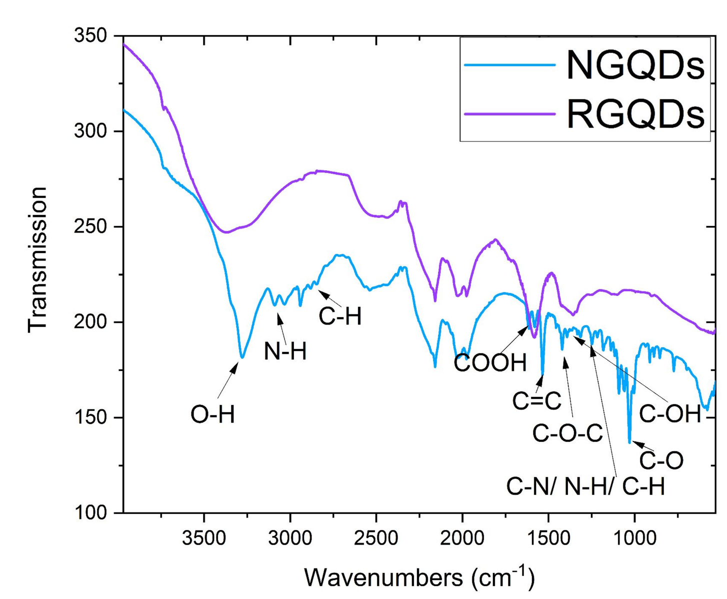
- UV-Vis Absorbance



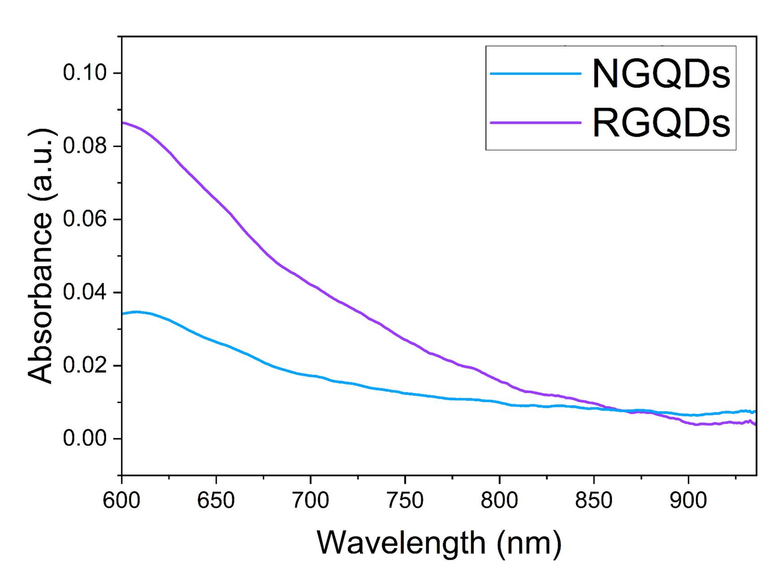
- Vis Photoluminescence



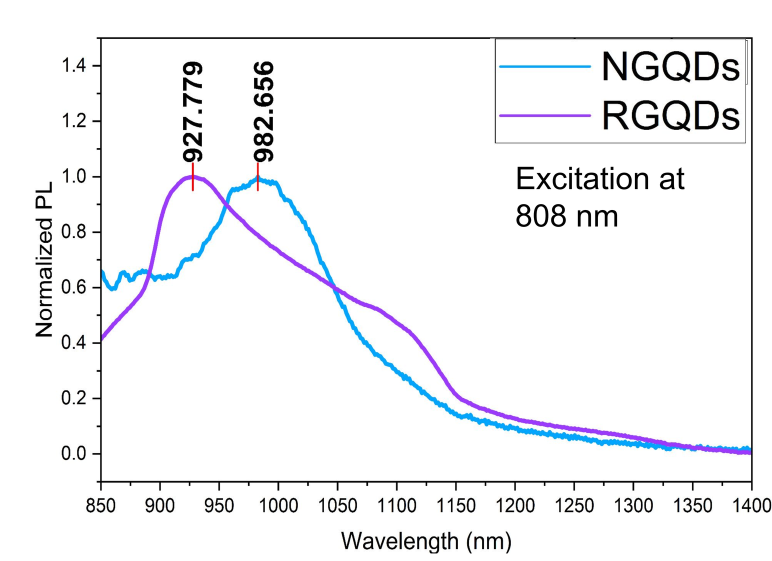
- FTIR Spectra



- Vis-NIR Absorbance

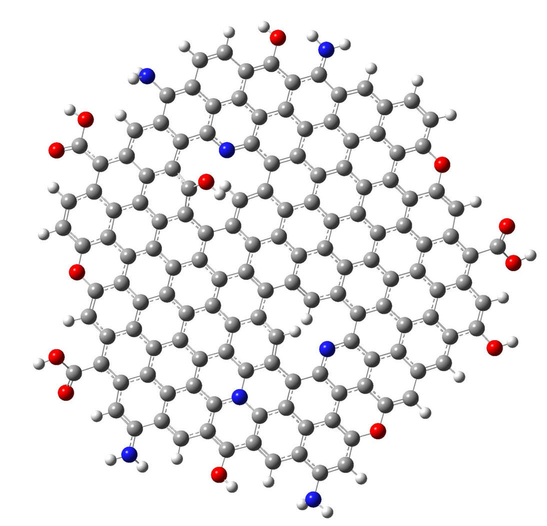


- NIR Photoluminescence



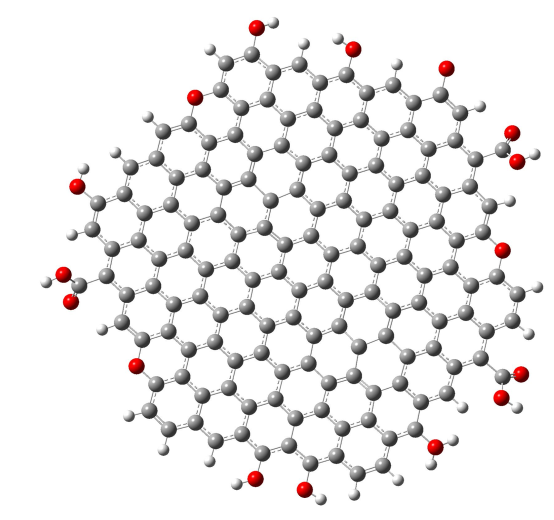
Gaussian Fluorescence of GQDs

NGQD



953 nm (983 nm)

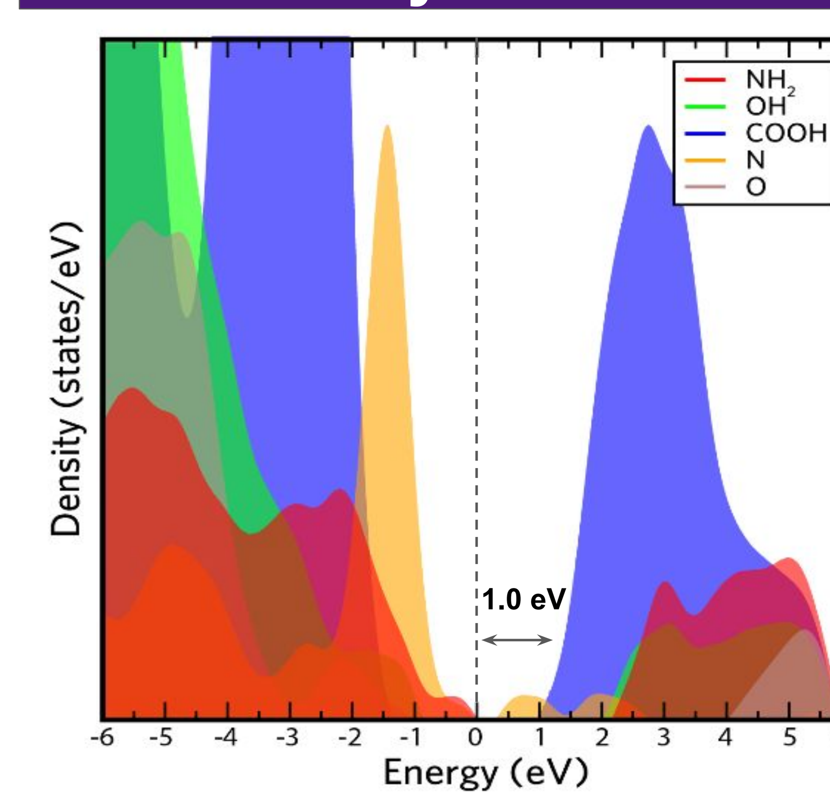
RGQD

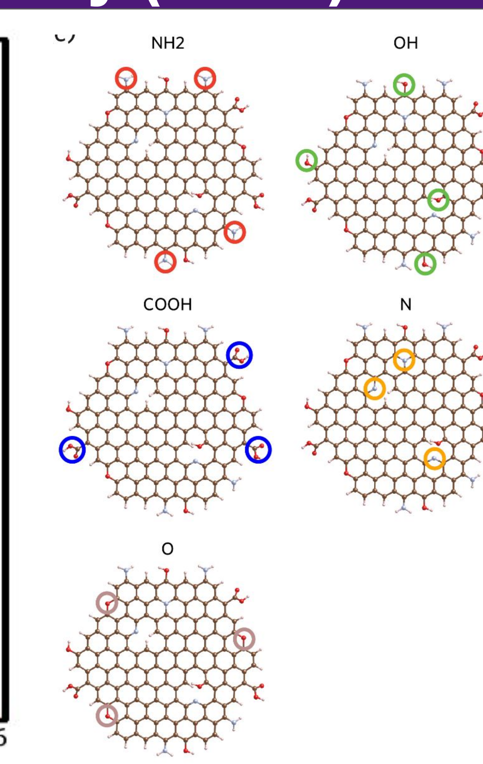


900 nm (928 nm)

- Fluorescence of GQDs were determined by Hartree-Fock 3-21G using Gaussian (Experimental results are highlighted in orange)

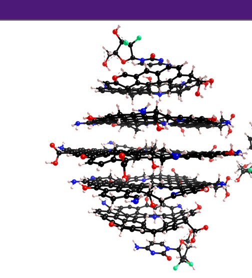
Density Function Theory (VASP)



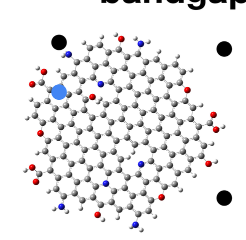


- Functional group decomposed density of states of NGQD was obtained by using DFT implemented in Vinenna ab-inito Simulation Package (VASP)

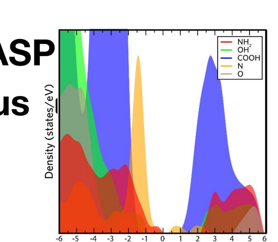
Conclusion



- Varying temperatures will cause a significant quenching in GQD fluorescence in both the visible and near-infrared regions, attributable to electron-phonon coupling changes, quantum yield variations, and bandgap alterations.



- Fluorescence Calculations performed on Gaussian with H-F 3-21G with N-GQDs reveals peaks that are reasonably close to our experimental results at 953 nm and R-GQDs at 900 nm
- RGQD's integrated density is proportional to temperature, and thereby its optical properties remain similar with minimal thermal fluctuation.



- Temperature Dependent Fluorescence obtained by DFT and VASP exhibits the most sophisticated calculations such as allowing us decompose our structure's electronic properties by functional group.
- Drawback of VASP is its lack of creating realistic systems for an experimentalist and high computational cost.

References

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