Investigating Modulation of Graphene Oxide Fluorescence via External Electric Fields

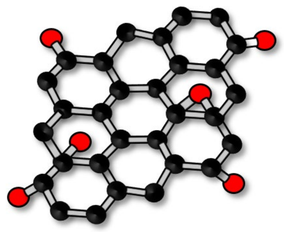
Bong Han Lee [1], Alina Valimukhametova [1], Ryan Conor [1], Thomas Paz [2], Fabian Grote [3], Anton V. Naumov [1]

[1]Department of Physics and Astronomy, Texas Christian University, Fort Worth, USA, [2] 36th Infantry Division Headquarters, Texas Army National Guard, Austin, Texas, USA, [3] Institute of Chemistry and Biochemistry, Free University of Berlin, Berlin, Germany

Abstract

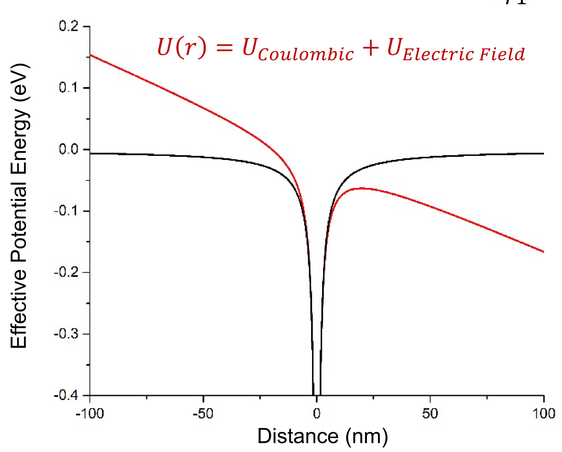
With the advent of graphene, there has been an interest in utilizing this material and its derivative, graphene oxide (GO) for novel applications in nanodevices such as bio and gas sensors, solid state supercapacitors and solar cells. Although GO exhibits lower conductivity and structural stability, it possesses an energy band gap that enables fluorescence emission in the visible/near infrared leading to a plethora of optoelectronic applications. In order to allow fine-tuning of its optical properties in the device geometry, new physical techniques are required that unlike existing chemical approaches yield substantial alteration of GO structure. Such desired new technique is one that is electronically-controlled and lead to reversible changes in GO optoelectronic properties. In this work, we for the first time investigate the methods to controllably alter the optical response of GO with the electric field and provide theoretical modelling of the electric field-induced changes. Field-dependent GO emission is studied in bulk GO/PVP films with up to 6% reversible decrease under 1.6 V/μm electric fields. On an individual flake level, a more substantial over 50% quenching is achieved for select GO flakes in polymeric matrix between interdigitated microelectrodes subject to two orders of magnitude higher fields. This effect is modelled on a single exciton level by utilizing WKB approximation for electron escape form the exciton potential well. In an aqueous suspension at low fields GO flakes exhibit electrophoretic migration indicating a degree of charge separation and a possibility of manipulating GO materials on a single-flake level to assemble electric field-controlled microelectronics. As a result of this work, we suggest the potential of varying the optical and electronic properties of GO via the electric field for the advancement and control over its optoelectronic device applications.

Introduction



- GO is a derivative of graphene.
- It possess oxygen functional groups.
- These groups localize regions of sp^2 carbon resulting in a band gap.
- This quantum confinement effect yields GO's fluorescence.

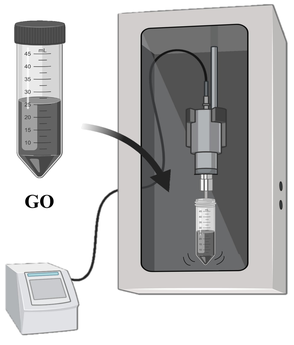
- **Schrödinger Equation:** $-\frac{\hbar^2}{2m}\frac{d^2\psi}{dx^2} + V(x)\psi = E\psi$
- **WKB Approximation:** $T \cong e^{-2\gamma};\ \gamma \equiv \frac{1}{\hbar}\int_{r1}^{r2}|p(r)|dr$



- Calculated potential well of the exciton without (**black line**) and with (**red line**) applied electric field. Using the WKB approximation, the transmission probability is ~5.2% for the bulk GO sample.

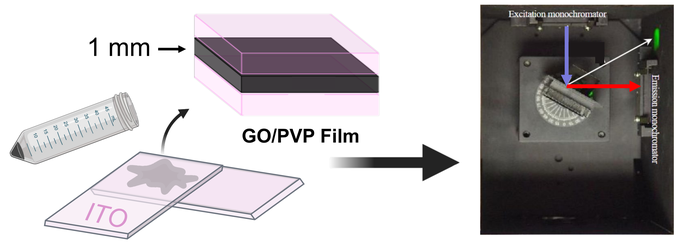
Materials and Methods

1 Tip Sonication

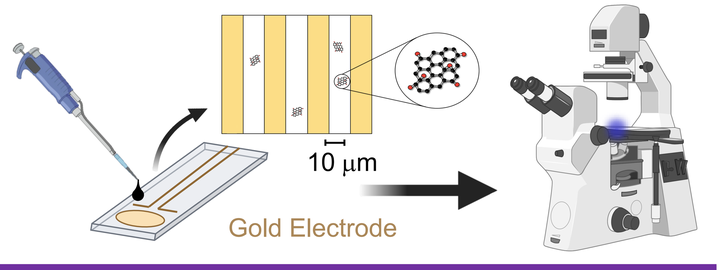


- GO is in the μm size range.
- Tip sonication was used to disperse and reduce agglomerated GO.

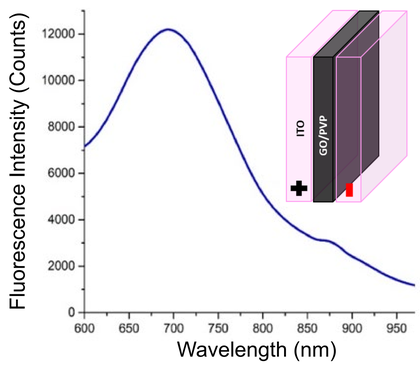
2 Bulk GO Sample Fluorescence Spectroscopy

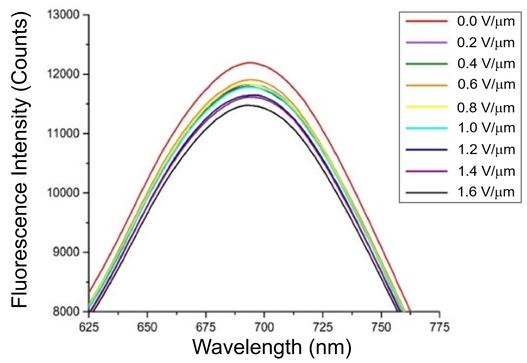


3 Single Flake GO Fluorescence Microscopy

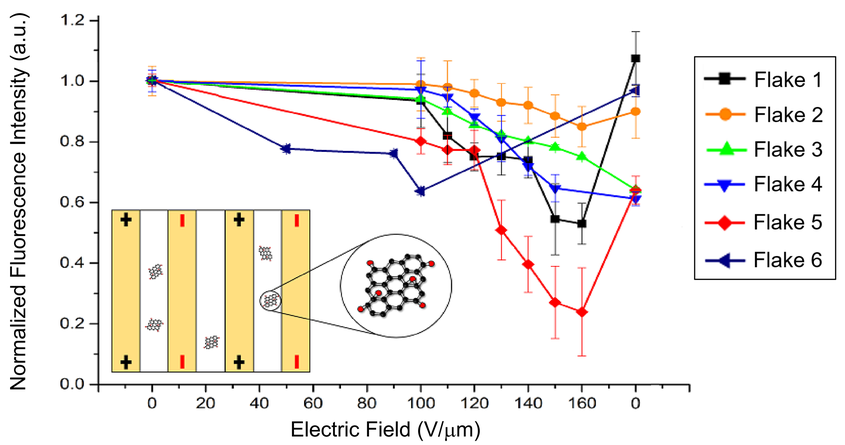


Results: Effect of Applied Electric Field onto GO



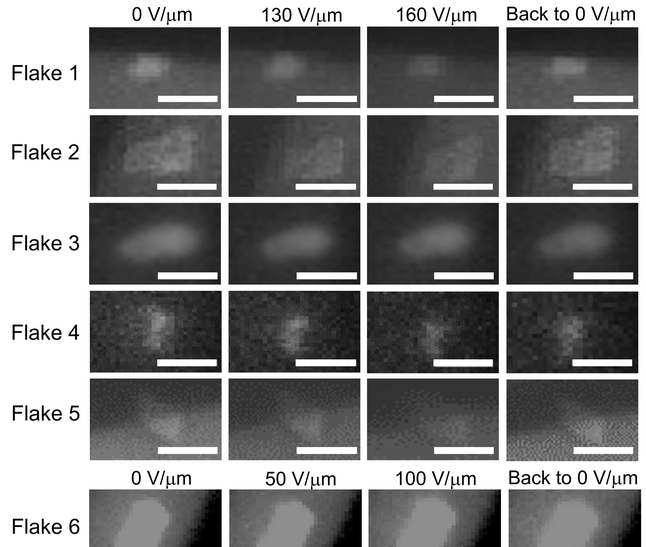


- GO/PVP film is subject to the fields of 0 - 1.6 V/μm incremented in 0.2 V/μm steps.
- Fluorescence intensity gradually decreases by up to 6 % at the maximum field strength, and shows a return.



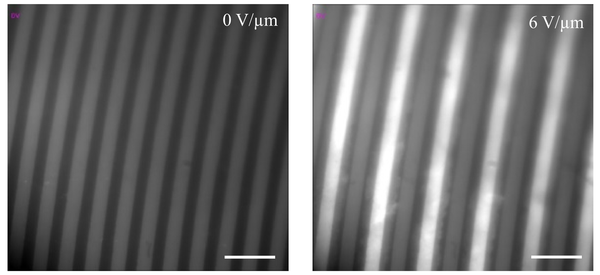
- With the application of increasing electric fields, GOs' flake fluorescence intensity gradually decreases.
- Upon removal of the field, the fluorescence intensity of a number of GO flakes become fully/partially restored.

Results: Single Flake GO Image Analysis



- Fluorescence microscopy images analysed with ImageJ.
- GO flakes 1, 2, 5 and 6 show a degree of reversible fluorescence.

Results: Electrophoretic Migration



- Electrophoretic deposition of GO in aqueous suspension shows large numbers of flakes exhibit migration to the electrodes' sides.

Conclusion and Outlook

- The transmission probability of ~5.2% is a close estimate to the experimental 6% quenching for bulk GO sample.
- Upon field removal, the fluorescence intensity of a number of GO flakes fully or partially restored, however, some have remained quenched potentially due to charge trapping or partial reduction via internal currents plausible at such field levels.
- The migration of GO flakes using electrophoresis offers the potential to form microstructures and devices from flakes on the microscale.
- Utilizing the electric fields as a mechanism to fine-tune optoelectronic properties of GO offers a degree of reversibility in device geometry.

References

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- Lee D, Seo J, Zhu X, Lee J, Shin H-J, Cole J M, Shin T, Lee J, Lee H and Su H 2013 Quantum confinement-induced tunable exciton states in graphene oxide *Scientific reports* **3** 2250