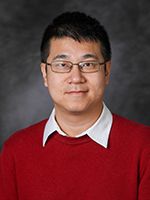
VISTA Seminar

Seminar 21

*In memory of Prof. Tao Yu (University of North Dakota)*



July 28, 2021

9:30 – 11:30 am EDT / 1:30 – 3:30 pm GMT / 3:30 pm – 5:50 pm Paris

TOC:

1. Presenter 1: Prof. George Schatz, Northwestern University, USA…..…. page 2
2. Presenter 2: Dr. Mohammed Jabed, University of North Dakota, USA….page 3
3. Presenter 3: Dr. David Lingerfelt, Oak Ridge National Laboratory, USA. page 4
4. How to connect………………………………………………………..…. page 5

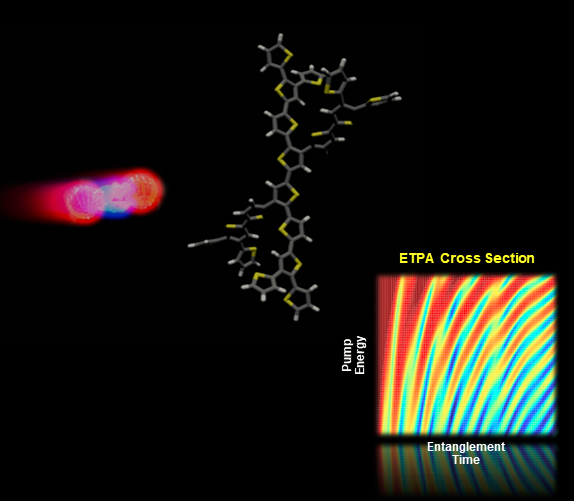
**I. Self-assembly of peptide amphiphiles**

**II. Two-photon absorption with entangled photons**

George C. Schatz

*Northwestern University, Evanston IL 60208 USA*

*Email:* [*g-schatz@northwestern.edu*](mailto:g-schatz@northwestern.edu)

The talk will provide a brief overview of my studies with Tao Yu of the self-assembly of peptide amphiphiles into micelles and fibers, that have provided a number of exciting applications to mechanically switchable materials and in biomedicine. Then I will switch to recent work in my group concerned with the development of electronic structure methods for describing the absorption of two photons that can either be unentangled or entangled. Here we connect with experiments in the Goodson group at Michigan by showing how to calculate the two-photon cross sections using density functional theory involving a second linear response formalism. The results demonstrate dramatic enhancements in cross sections that arise from entanglements, and they also demonstrate novel capabilities for probing intermediate states using entangled photons.

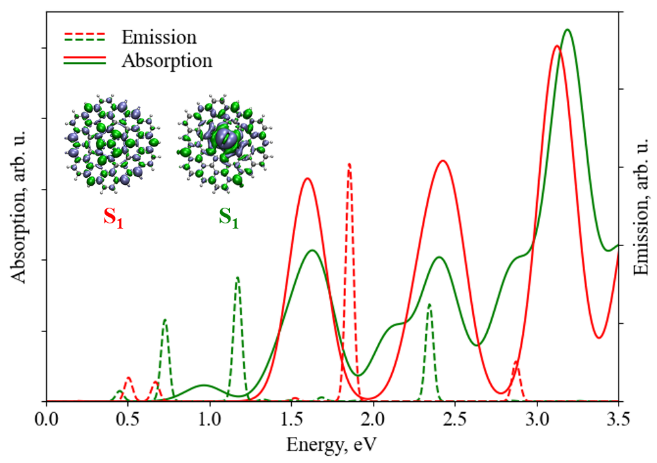
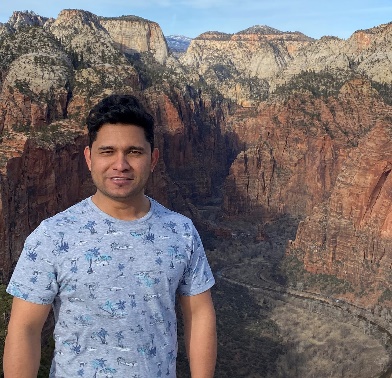
**Understanding the Optoelectronic Properties of the N-doped Graphene QDs**

Jabed A. Mahammed1, Julia Zhao1, Dmitri Kilin2, Tao Yu1

1.*Department of Chemistry, University of North Dakota, Grand Forks, ND 58202*

*Email:* [*mohammed.jabed@und.edu*](mailto:mohammed.jabed@und.edu)

2.*Department of Chemistry, North Dakota State University, Fargo, ND 58105*



Graphene quantum dots (GQDs) are promising in bioimaging and sensing applications due to its biocompatibility and tunability of photophysical properties. Despite the open bandgap and promising optical properties of GQD, the realization of practical devices of GQD is challenging due to its edge effect,1 low quantum yield2, and lack of control on the photoluminescence mechanism3. N-doping would be a promising strategy to modulate the electronic and structural functionalities and improving the photoluminescence properties. Nitrogen can be incorporated in conformations, and we are focusing on graphitic N-doping and atomic N chemisorption on the QDs.

In the first part of my talk, I will discuss the electronic and absorption properties of N-doped GQDs, calculated using the DFT and TDDFT methods. I will show how N-doping and edge modification modulate the hybridization and electronic transitions. The lowest energy transition is more charge transfer in nature with increasing the C/edge-O ratio.4 In the second part of my talk, I will discuss the photoexcited electron-hole relaxation dynamics calculated using time-dependent density matrix methodology and non-adiabatic coupling used as parameters to the Redfield theory, providing hot carriers’ relaxation pathways. We also simulate the time resolve emission spectra. In contrast to N chemisorption, graphitic N-doping redshifts the bandgap, but edge oxidation creates localized states and decreases hot electron-hole relaxation rates.

**References:**

1. Das, S. K.; Luk, C. M.; Martin, W. E.; Tang, L.; Kim, D. Y.; Lau, S. P.; Richards, C. I. Size and Dopant Dependent Single Particle Fluorescence Properties of Graphene Quantum Dots. *J. Phys. Chem. C* **2015**, *119*, 17988-17994.
2. Li, L.; Wu, G.; Yang, G.; Peng, J.; Zhao, J.; Zhu, J.-J. Focusing on luminescent graphene quantum dots: current status and future perspectives. *Nanoscale* **2013**, *5*, 4015-4039.
3. Zhu, S.; Zhang, J.; Liu, X.; Li, B.; Wang, X.; Tang, S.; Meng, Q.; Li, Y.; Shi, C.; Hu, R.; Yang, B. Graphene quantum dots with controllable surface oxidation, tunable fluorescence and up-conversion emission. *RSC Adv.* **2012**, *2*, 2717-2720
4. Jabed, M. A., Zhao, J., Kilin, D., & Yu, T. Understanding of Light Absorption Properties of the N-Doped Graphene Oxide Quantum Dot with TD-DFT. J *Phys. Chem. C* **2021**,125,27, 14979-14990

**Excited State and Nonadiabatic Reaction Pathways Involving Single-Atom Impurities in Graphene**

David B. Lingerfelt\*, Anthony Yoshimura\*\*\*, Panchapakesan Ganesh\*, Jacek Jakowski\*\*, Bobby Sumpter\*

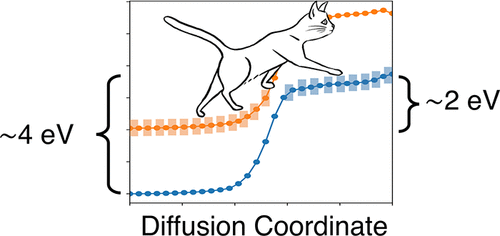
*\* Nanomaterials Theory Institute, Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, United States*

*\*\* Computational Sciences and Engineering Division Oak Ridge National Laboratory, Oak Ridge, Tennessee 37831, United States*

*\*\*\* Department of Physics, Applied Physics, and Astronomy, Rensselaer Polytechnic Institute, Troy, New York 12180, United States*

*Email:* [*DBLinger@gmail.com*](mailto:DBLinger@gmail.com)





The high-energy electron sources and aberration corrected electron optics found in modern scanning transmission electron microscopes (STEMs) can be employed not only for imaging purposes, but also to direct local chemical changes in materials. Indeed, there are now numerous examples of atomically-(or nearly atomically-)precise modifications being effectuated in graphene and other resilient materials using STEMs.[1] While the elastic collisions between high energy electrons and atomic nuclei can be understood in terms of Rutherford-like scattering cross sections, the inelastic scattering of the incident electrons by the material-bound electrons — and the coupled electronic and vibrational response of the material to the electron impact — demands an *ab initio* treatment. To this end, a straight-forward TD-DFT based approach for predicting electronic transition probabilities due to the presence of focused electron beams was devised, and applied to elucidate the electronic response of some experimentally-relevant doped graphene nanoparticles to convergent electron beam irradiation. [2,3] These results, combined with insight into the excited state potential energy surfaces, indicate the possibility for beam-induced electronic excitations to significantly modulate reactivity through both the lowering of barriers in excited states relative to the ground state, and by opening electronically nonadiabatic pathways which can impart momentum along the reaction coordinate *via* nonradiative decay of the excited state populations.

This work was performed at the Center for Nanophase Materials Sciences, a DOE Office of Science User Facility.

**References:**

[1] O. Dyck *et al, Nat. Rev. Mater.,* 4(7), 497–507 (2019)

[2] D. B. Lingerfelt, P. Ganesh, J. Jakowski, B. G. Sumpter, *J. Chem. Theory Comput.*, 16(2), 1200–1214 (2020)

[3] D.B. Lingerfelt, T. Yu, A. Yoshimura, P. Ganesh, J. Jakowski, B. G. Sumpter, *Nano Lett.*, 21(1), 236–242 (2021)

**How to connect**

Alexey Akimov is inviting you to a scheduled Zoom meeting.

Topic: VISTA, Seminar 21

Time: Jul 28, 2021 09:30 AM Eastern Time (US and Canada)

Join Zoom Meeting

https://buffalo.zoom.us/j/96738173790?pwd=bTY0YzJrTnhCb0RNOWVkRm9sYXh4dz09

Meeting ID: 967 3817 3790

Passcode: 837146

One tap mobile

+16465588656,,96738173790#,,,,\*837146# US (New York)

+13017158592,,96738173790#,,,,\*837146# US (Washington DC)

Dial by your location

+1 646 558 8656 US (New York)

+1 301 715 8592 US (Washington DC)

+1 312 626 6799 US (Chicago)

+1 253 215 8782 US (Tacoma)

+1 346 248 7799 US (Houston)

+1 669 900 9128 US (San Jose)

Meeting ID: 967 3817 3790

Passcode: 837146

Find your local number: https://buffalo.zoom.us/u/abLhgobz5Z

Join by SIP

96738173790@zoomcrc.com

Join by H.323

162.255.37.11 (US West)

162.255.36.11 (US East)

115.114.131.7 (India Mumbai)

115.114.115.7 (India Hyderabad)

213.19.144.110 (Amsterdam Netherlands)

213.244.140.110 (Germany)

103.122.166.55 (Australia Sydney)

103.122.167.55 (Australia Melbourne)

149.137.40.110 (Singapore)

64.211.144.160 (Brazil)

69.174.57.160 (Canada Toronto)

65.39.152.160 (Canada Vancouver)

207.226.132.110 (Japan Tokyo)

149.137.24.110 (Japan Osaka)

Meeting ID: 967 3817 3790

Passcode: 837146