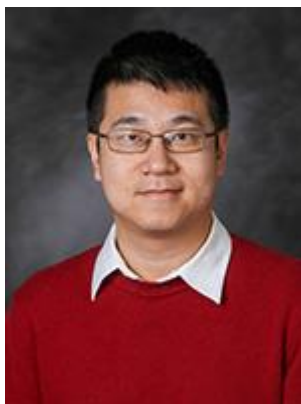


VISTA Seminar

Seminar 21

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



June 30, 2021

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

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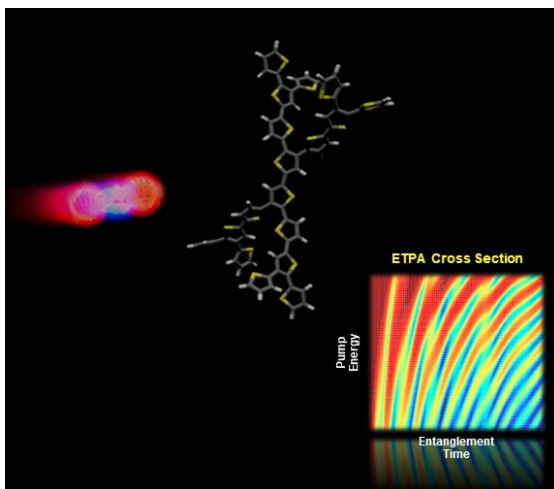
I. Self-assembly of peptide amphiphiles

II. Two-photon absorption with entangled photons

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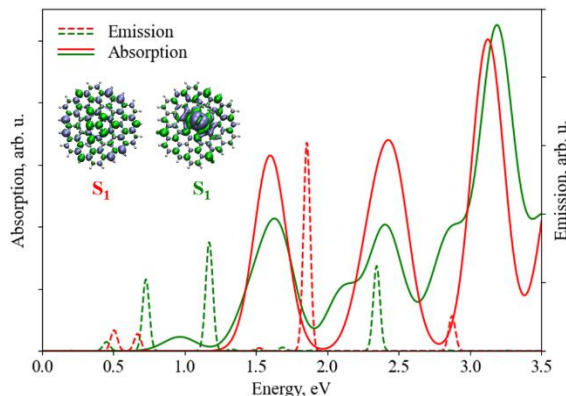
The talk will provide a brief overview of my studies with Tao Yu of the self-assembly of peptide amphiphiles into micelles and fibers, that have provided a number of exciting applications to mechanically switchable materials and in biomedicine. Then I will switch to recent work in my group concerned 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

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2.

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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

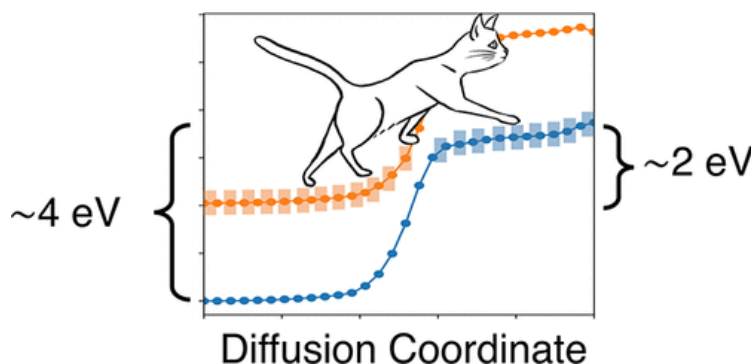
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The high-energy electron sources and aberration corrected electron optics found in modern scanning transmission electron microscopes (STEMs) can be employed not only for imaging purposes, but also to direct local chemical changes in materials. Indeed, 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)

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64.211.144.160 (Brazil)

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