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PRESENTATIONS

- Beckstead, A. A.; Li, X. –B.; Zhang, Y.; Burrows, C. J.; Kohler, B. “Charge Transfer Dynamics of 5-Hydroxycytidine and 5-Hydroxyuridine Investigated by Time-Resolved IR Spectroscopy”, The 63rd Pacific Conference on Spectroscopy and Dynamics, Pacific Grove, CA, 2016 (contributed talk)
- Beckstead, A. A.; Li, X. –B.; Zhang, Y.; Burrows, C. J.; Kohler, B. “Charge Transfer Dynamics of 5-Hydroxycytidine and 5-Hydroxyuridine Investigated by Time-Resolved IR Spectroscopy”, MSU Graduate Student Summit, Bozeman, MT, 2015 (poster)
- Beckstead, A. A.; Zhang, Y.; Dood, J.; Li, X. –B.; Burrows, C. J.; Improta, R.; Kohler, B. “Monitoring the Photoinduced Electron Transfer Capability of 8-Oxo-7,8-dihydro-2'-deoxyguanosine Using Ultrafast Time-Resolved IR Spectroscopy”, The 69th Northwest Regional Meeting of the ACS, Missoula, MT, 2014 (contributed talk)
- Beckstead, A. A.; Zhang, Y.; de La Harpe, K.; Hariharan, M.; Kohler, B. “Excited-State Dynamics of Adenine DNA Systems in Non-Aqueous Deep Eutectic Solvents Revealed by Femtosecond Transient Absorption Spectroscopy” OpTeC Optical Science and Engineering Conference, Bozeman, MT, 2014 (poster)
- Beckstead, A. A.; Dood, J.; Zhang, Y.; Li, X. –B.; Burrows, C. J.; Improta, R.; Kohler, B. “Excited-State Dynamics of a Modified Monomer Nucleobase as Revealed by Femtosecond Pump-Probe Spectroscopy” OpTeC Optical Science and Engineering Conference, Bozeman, MT, 2013 (poster)



Department of Chemistry and Biochemistry

Doctor of Philosophy
in Chemistry

DISSERTATION DEFENSE

Ms. Ashley A. Beckstead

B.Sc. University of Utah, Salt Lake City, UT (2011)

Monday, May 8, 2017 – 12:30 pm

Byker Auditorium

Department of Chemistry and Biochemistry

“Excited-State Dynamics of Biological Molecules in Solution: Photoinduced Charge Transfer in Oxidatively Damaged DNA and Deactivation of Violacein in Viscous Solvents”

Graduate Committee

- Dr. Bern Kohler (Research Advisor, chemistry)
Dr. Robert A. Walker (Committee Chair, chemistry)
Dr. Patrik Callis (chemistry)
Dr. Rufus Cone (physics)

ABSTRACT

UV radiation from the sun is strongly absorbed by DNA. The resulting electronic excited states can lead to the formation of mutagenic photoproducts. Decades of research have brought the excited-state dynamics of single DNA and RNA nucleobases to light, but questions remain about the nature of excited states in DNA strands. In this thesis, I present our recent observations of photoinduced electron transfer (ET) between the oxidatively damaged bases, 8-oxo-7,8-dihydro-2'-deoxyguanosine, 5-hydroxy-2'-deoxycytidine and 5-hydroxy-2'-deoxyuridine, and adenine in three different dinucleotides. Our results reveal that charge-transfer (CT) states are formed in the dinucleotides studied on an ultrafast timescale, and the back ET process that returns the excited-state population to the ground state is very efficient. In addition to other spectroscopic observations of CT state species in DNA excited states, our results have increased understanding of the long-lived transient signals observed in DNA strands. We discuss these results in the context of a recent proposal that oxidatively damaged nucleobases may have served prebiotic redox catalytic roles.

In this thesis, I also present the results of a recent ultrafast spectroscopic investigation of violacein, a pigment isolated from Antarctic bacteria. Despite claims for the photoprotective role of the pigment, there has never been a spectroscopic analysis of excited-state deactivation in violacein. The emission spectra, fluorescence quantum yields and excited-state lifetimes of violacein in various solvents were obtained for the first time. The fluorescence quantum yield and excited-state lifetime of violacein increase as the solvent becomes more viscous, suggesting a large-scale motion mediates excited-state deactivation. These results are compared to similar observations of viscosity-dependent excited-state decay rates. We also consider the relevance of our excited-state lifetime measurements to the hypothesized UV-screening role of violacein. Together, the studies presented in this dissertation illustrate how ultrafast spectroscopic techniques can be used to unravel complex biomolecular excited-state dynamics.

BIOGRAPHICAL NOTES

Academic Preparation:

2005-2011 University of Utah, Salt Lake City, UT (Dec. 2006)
Bachelor of Science in Chemical Physics (Honors), Mathematics Minor
Thesis: High Temperature-Programmed Desorption Surface Chemistry
Advisor: Prof. Scott L. Anderson

Graduate Studies

Field of Study: physical chemistry, photochemistry

Teaching and Outreach Activities

2015 Physical Chemistry TA (Jan-May)
2015 NanoDays Outreach (Apr)
2014 Physical Chemistry TA (Jan-May)
2014 Science Olympiad Volunteer (Nov)
2013 Outreach Trip to Twin Bridges, MT (Nov)
2013 General Chemistry Lab TA (Aug-Dec)
2012 General Chemistry Lab TA (Aug-May)

Awards

2014-2017 NSF AGEP-Graduate Research Supplement
2016 NSF CHE Supplement for Professional Development
2012 Meritorious Graduate Fellowship, Montana State University

PUBLICATIONS

- Beckstead, A. A.; Zhang, Y.; Smith, H.; Bermel, E.; Skowron, D.; de La Harpe, K.; Oliver, T. A. A.; Foreman, C.; Kohler, B. "Excited-State Deactivation of the Bacterial Pigment Violacein", *in preparation*.
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