



AFRL-AFOSR-VA-TR-2015-0407

HARNESSING SOLAR POWER NOVEL STRATEGIES FOR RATIONAL DESIGN OF PHOTOCATALYSTS AND PHOTOVOLTAIC MATERIALS

**Gregory Engel
UNIVERSITY OF CHICAGO THE**

**09/01/2015
Final Report**

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AFOSR FA9550-10-1-0028

**Harnessing Solar Power:
Novel Strategies for Rational Design of Photocatalysts
and Photovoltaic Materials**

Gregory S. Engel
Department of Chemistry
James Franck Institute
The University of Chicago
Chicago, IL USA

Period of Performance: December 15, 2009 to May 31, 2015

Abstract:

The goal of this effort was to develop the spectroscopic infrastructure to be able to probe non-Born-Oppenheimer couplings near conical intersections in the condensed phase. This effort required 2D electronic spectroscopy to be extended to broader bandwidth, higher sensitivity and improved resolution of coherent dynamics to detect phase. We invented a new approach to 2D spectroscopy to address these issues: Gradient-Assisted Photon Echo Spectroscopy (GRAPES). This approach allowed improved bandwidth due to purely reflective optics, improved sensitivity due to elimination of laser power fluctuations in the indirect domain, and improved phase resolution. We applied this spectroscopy to a range of synthetic and natural photosynthetic light harvesting systems to examine the system-bath coupling (electronic-vibrational couplings) using coherences as a detailed probe of the environment.

The heart of the strategy for design of novel photocatalysts involved a detailed understanding of how electronic states interact with their environment. Detecting and interpreting this coupling occupied most of our effort during this program. In addition to technical advances in our spectroscopy, we have been able to develop models that incorporate vibronic coupling (or system-bath coupling) as parameters and use these models to help quantify the degree of vibronic mixing in molecular systems. Separately, in an effort to understand dephasing dynamics, we developed the first time-resolved 2D electronic chiral spectroscopy to probe wavefunction collapse and angular momentum of the initial wavepacket. Together these measurements demonstrate an intricate and tunable coupling between electronic and vibrational states and point to a mechanism to test theories of Berry phase near conical intersections.

In multidimensional spectroscopy, coherences among excited states report on interactions between electronic states and their environment. Mixed electronic-vibrational (vibronic) resonance has been proposed as a possible physical origin for the prolonged lifetimes of coherences observed in some systems, and recent observations confirm the existence of vibronic coupling in both model systems and photosynthetic complexes. We hypothesized that this same coupling is responsible for steering electronic wavepackets through conical intersections. We attempted to take a comprehensive spectroscopic strategy: on one hand, we sought to extend femtosecond stimulated Raman to allow correlation mapping while on the other hand, we created new ultra-sensitive measures of 2D electronic spectroscopy to directly measure excited state coherences. The FSR strategy met with many technical hurdles and remains to be fully implemented. The largest issue was getting enough power from our laser system into the pre-resonant Raman beam. With regard to the direct 2D spectroscopic measurements, the GRAPES system provided an enormous boost in signal-to-noise (up to 12,000 on a standard 2D spectrum) and permitted rapid acquisition of the spectra. This approach became the workhorse for most of our work.

Scientifically, we seek to understand how the dynamics of electronic states, to include both energy transfer and photochemical reactivity, depends on the vibrational bath surrounding the electronic wavepacket. Establishing control over this vibronic coupling within model systems will permit unambiguous identification of detailed molecular design principles that control the interplay between vibronic coupling and energy transfer. During the course of this grant, we investigated molecular systems such as PM650 that show purely vibrational dynamics, photosynthetic light harvesting complexes that show dissipative energy transfer dynamics, and nanoscale systems that show complex and rapid dephasing dynamics. In each case, we investigated coherence dynamics in time-domain spectroscopy to provide a fine probe of how a system interacts with its surroundings. The analysis of these systems has allowed us to begin to develop microscopic design principles regarding both the signals and coupling, but we have not yet been able to gain sufficient control to develop and design novel photocatalysts.

Publications from this effort:

P.D. Dahlberg, G.J. Norris, C. Wang, S. Viswanathan, V.P. Singh, and G.S. Engel, “Coherences Observed in vivo in Photosynthetic Bacteria Using Two-Dimensional Electronic Spectroscopy” *J Chem Phys* (accepted) 2015

V. P. Singh, M. Westberg, C. Wang, P. D. Dahlberg, T. Gellen, A. T. Gardiner, R. J. Cogdell and G. S. Engel, “Towards quantification of vibronic coupling in photosynthetic antenna complexes” *J. Chem. Phys.* 142, 212446 2015.

J.R. Caram, H. Zheng, P.D. Dahlberg, B.S. Rolczynski, G.B. Griffin, A.F. Fidler, D.S. Dolzhenkov, D.V. Talapin, and G.S. Engel, “Persistent Inter-Excitonic Quantum Coherence in CdSe Quantum Dots” *J. Phys. Chem. Lett.* 5, 196-204, 2014

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- G.B. Griffin, S. Itthuria, A. Linkin, D.V. Talapin, and G.S. Engel, "Two Dimensional Electronic Spectroscopy of CdSe Nanoparticles at Very Low Pulse Power" *J. Chem. Phys.* 138, 014705 2013.
- E. Harel, S.M. Rupich, R.D. Schaller, D.V. Tapalin, and G.S. Engel, "Measurement of Electronic Splitting in PbS Quantum Dots by Two-Dimensional Nonlinear Spectroscopy" *Phys Rev B*, 86 075412 2012.
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Changes in Period of Performance:

At AFOSR's suggestion, we agreed to

- a. extend Option 3 from 12 months to 17 months with a Period of Performance to 31 May 14.
- b. as a result, Option 4 POP will be from 01 Jun 14 to 31 May 15.

1.

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The University of Chicago

Grant/Contract Title**The full title of the funded effort.**

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Grant/Contract Number**AFOSR assigned control number. It must begin with "FA9550" or "F49620" or "FA2386".**

FA9550-10-1-0028

Principal Investigator Name**The full name of the principal investigator on the grant or contract.**

Gregory S. Engel

Program Manager**The AFOSR Program Manager currently assigned to the award**

Michael Berman

Reporting Period Start Date

12/15/09

Reporting Period End Date

05/31/2015

Abstract

The goal of this effort was to develop the spectroscopic infrastructure to be able to probe non-Born-Oppenheimer couplings near conical intersections in the condensed phase. This effort required 2D electronic spectroscopy to be extended to broader bandwidth, higher sensitivity and improved resolution of coherent dynamics to detect phase. We invented a new approach to 2D spectroscopy to address these issues: Gradient-Assisted Photon Echo Spectroscopy (GRAPES). This approach allowed improved bandwidth due to purely reflective optics, improved sensitivity due to elimination of laser power fluctuations in the indirect domain, and improved phase resolution. We applied this spectroscopy to a range of synthetic and natural photosynthetic light harvesting systems to examine the system-bath coupling (electronic-vibrational couplings) using coherences as a detailed probe of the environment.

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Archival Publications (published) during reporting period:

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Changes in research objectives (if any):

N/A

Change in AFOSR Program Manager, if any:

N/A

Extensions granted or milestones slipped, if any:

At AFOSR's suggestion, we agreed to

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b. as a result, Option 4 POP will be from 01 Jun 14 to 31 May 15.

AFOSR LRIR Number

LRIR Title

Reporting Period

Laboratory Task Manager

Program Officer

Research Objectives

Technical Summary

Funding Summary by Cost Category (by FY, \$K)

	Starting FY	FY+1	FY+2
Salary			
Equipment/Facilities			
Supplies			
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