



Colloquium Announcement

of the Collaborative Research Centre 951

“Hybrid Inorganic/Organic Systems for Opto-Electronics”

Gregory D. Scholes

Department of Chemistry, Princeton University, USA

**Probing Ultrafast Chemical Dynamics
Inspired by the Rhythms of Fireflies**

Julia Stähler

Fritz Haber Institute of the Max Planck Society, Berlin, Germany

**Ultrafast quasiparticle dynamics in ZnO
and at its interface with an organic chromophore**

Time: Thursday, May 23, 2019, 3 pm c.t.

Place: Erwin-Schrödinger-Zentrum, Rudower Chaussee 26,
Room 0`119.



Collaborative Research Centre 951
Department of Physics
Humboldt-University of Berlin

Email: sfb951@physik.hu-berlin.de
Tel.: +49 30 2093 66374
www.physik.hu-berlin.de/sfb951

Partners



MAX-PLANCK-GESELLSCHAFT
FRITZ-HABER-INSTITUT



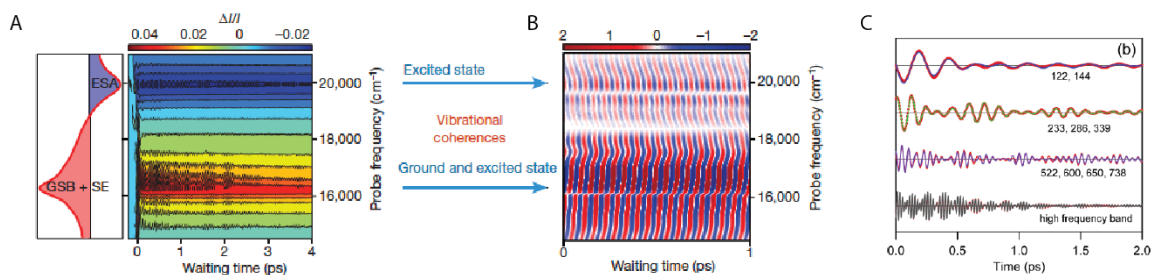
Probing Ultrafast Chemical Dynamics Inspired by the Rhythms of Fireflies

Gregory D. Scholes

Department of Chemistry, Princeton University, Princeton NJ 08544, U.S.A.

Coherence phenomena arise from interference, or the addition, of wave-like amplitudes in phase [1]. While coherence has been shown to yield transformative new ways for improving function, advances have been limited to pristine matter, as quantum coherence is considered fragile. Here I will discuss how vibrational and vibronic wavepackets entrain ensembles of molecules, like the synchronized flashing of fireflies. I will discuss how this can be used to probe mechanisms of ultrafast dynamics and how in-step vibrational motion might be employed to control function on ultrafast timescales. I will give examples that include light-harvesting in photosynthesis, energy flow in organometallic molecules that is ‘wired’ by Fermi resonance, and ultrafast electron transfer in molecular systems.

[1] Scholes, et al. “Optimal Coherence in Chemical and Biophysical Dynamics” *Nature* 543, 647–656 (2017).



Ultrafast quasiparticle dynamics in ZnO and at its interface with an organic chromophore

Lukas Gierster¹, Laura Foglia¹, Jan-Christoph Deinert¹, Sessa Vempati¹, Lea Bogner¹, Clemens Richter¹, Martin Wolf¹, Sergey Sadofev², Anton Zykov², Sylke Blumstengel², Stefan Kowarik², Yves Garmshausen², Jana Hildebrandt², Stefan Hecht², **Julia Stähler**¹

¹*Fritz Haber Institute of the Max Planck Society, Berlin, Germany*

²*Humboldt Universität zu Berlin & IRIS Adlershof, Berlin, Germany*

Due to its wide band gap and high carrier mobility, ZnO is an attractive material for light-harvesting and optoelectronic applications. Its functional efficiency, however, is strongly affected by defect-related in-gap states that open up extrinsic decay channels and modify relaxation timescales. As a consequence, almost every sample behaves differently, leading to irreproducible or even contradicting observations.

Using a complementary set of time-resolved spectroscopies that we applied to two ZnO samples of different defect density, we are able to disentangle the competing contributions of charge carriers, excitons, and defects to the non-equilibrium dynamics after photoexcitation. We find that defects affect the transient optical properties of ZnO across more than eight orders of magnitude in time, starting with photodepletion of normally occupied defect states on femtosecond timescales, followed by the competition of free exciton emission and exciton trapping at defect sites within picoseconds, photoluminescence of defect-bound and free excitons on nanosecond timescales, and deeply trapped holes with microsecond lifetimes. These findings do not only provide the first comprehensive picture of charge and exciton relaxation pathways in ZnO, but also uncover the microscopic origin of previous conflicting observations in this challenging material and thereby offer means of overcoming its difficulties.

We also characterize the ultrafast dynamics of electrons and excitons at a model organic/inorganic interface (5-phenyl-pyridine/ZnO(10-10)). In particular, we populate the LUMO of the organic molecules by two different pathways: (i) Via photoexcitation from an interfacial hybrid state just below E_F and (ii) by optical excitation of the organic dye. We thereby disentangle the two main contributions to interfacial charge separation, electronic coupling to the ZnO conduction band and electron-hole interaction in the organic molecule.