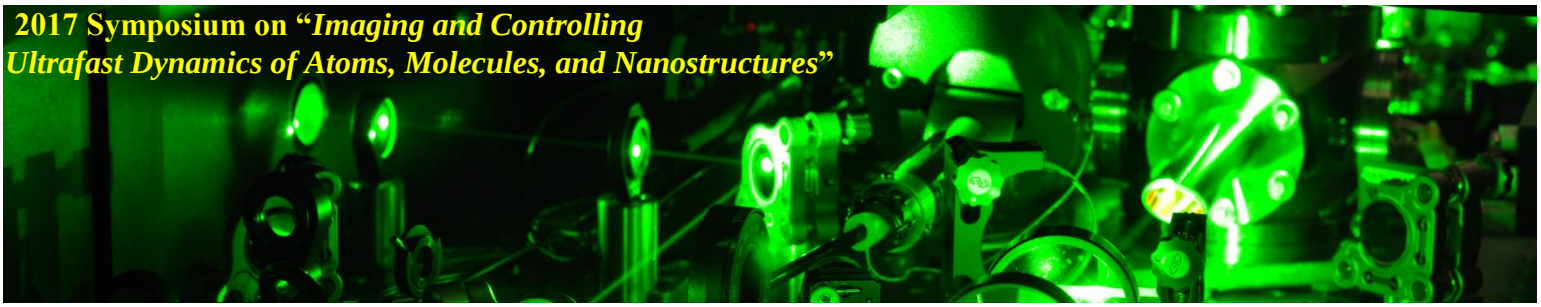


Friday, March 31, 2017 KS-NE Symposium on
"Imaging and Controlling Ultrafast Dynamics of Atoms, Molecules, and Nanostructures"

8:00 – Registration — All talks in the Big 12 Room, Student Union, KSU

8:30 – Welcome – Beth Montelone – Senior Associate Vice President for Research, KSU

Plenary Talk — **Chair:** Anthony Starace (UNL)

8:45 – *Linking high harmonics from solids and gases*
Paul Corkum – University of Ottawa and National Research Council (NRC) of Canada

Session I — **Chairs:** Herman Batelaan (UNL) & Uwe Thumm (KSU)

9:40 – *Nonlinear photochromic switching in the plasmonic field of a nanoparticle array*[1]
Chris Elles – University of Kansas

10:05 – Coffee Break — Flint Hills Room

10:30 – *Coherent control of the exciton/biexciton system in a quantum dot ensemble*
Steve Cundiff – University of Michigan

11:10 – *Mode-specific plasmonics examined using single-nanoparticle ultrafast imaging*
Ken Knappenberger – Florida State University

11:50 – *High harmonic generation in solids: dynamics of multilevel adiabatic states spanning the band structure*
Mette Gaarde – Louisiana State University

12:30 – Lunch Break — Flint Hills Room

Session II — **Chairs:** Martin Centurion (UNL) & Vinod Kumarappan (KSU)

13:45 – *Intense mid-infrared laser-cluster interactions*
Lou DiMauro – The Ohio State University

14:25 – *Control and measurement of attosecond pulses with two-color fields*[1]
Carlos Trallero-Herrero – Kansas State University

14:50 – Coffee Break — Flint Hills Room

15:15 – *Deep inner-orbital ionization of diatomic molecules by strong laser fields*
George Gibson – University of Connecticut

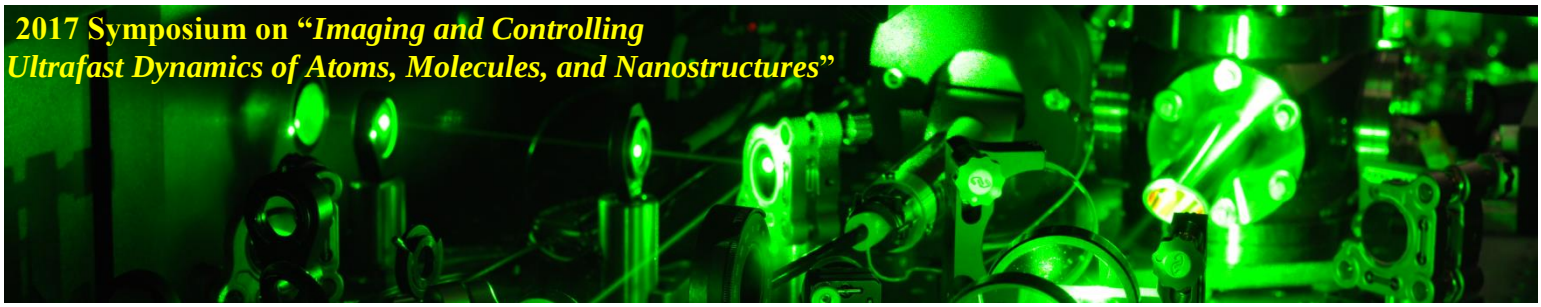
15:55 – *State-of-the-art and next-generation sources for ultrafast hyperspectral imaging*[1]
Matthias Fuchs – University of Nebraska–Lincoln

16:20 – Closing remarks

16:30 – Break

16:45 – **Poster Session** (Ends at 18:45) — Room 227

[1] EPSCoR supported project



Linking high harmonics from solids and gases

Paul B. Corkum

Professor, Joint Attosecond Science Lab, University of Ottawa and NRC

In atomic gases, multiphoton ionization creates an electron wave packet in the continuum and a time dependent current. Two sources of high harmonics arise from this fundamental process. Low harmonics of the driving field (approximately H1-H7) result from the time dependent current while the electron-ion re-collision that often follows the creation of the wave packet produces high harmonics and attosecond pulses. Each mechanism has a characteristic frequency-dependent spectral phase.

Analogous processes appear when a transparent solid is irradiated with infrared light. In solids, multiphoton excitation creates an electron in the conduction band and hole in the valence band, but now the electrons and holes move on non-parabolic bands that are characteristic of the solid. As with gases, spectral phase measurements can help us identify the mechanism(s) responsible for harmonics from solids.

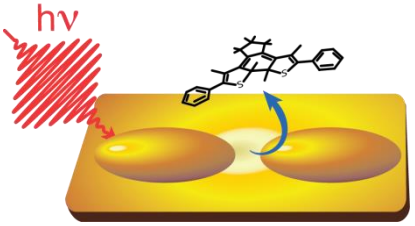
Extreme nonlinear optics in solids may provide an important source of VUV light and it is also a new diagnostic of materials. Solids can be perturbed, patterned, doped, or structured and each impresses its signature on the harmonics as they are created.

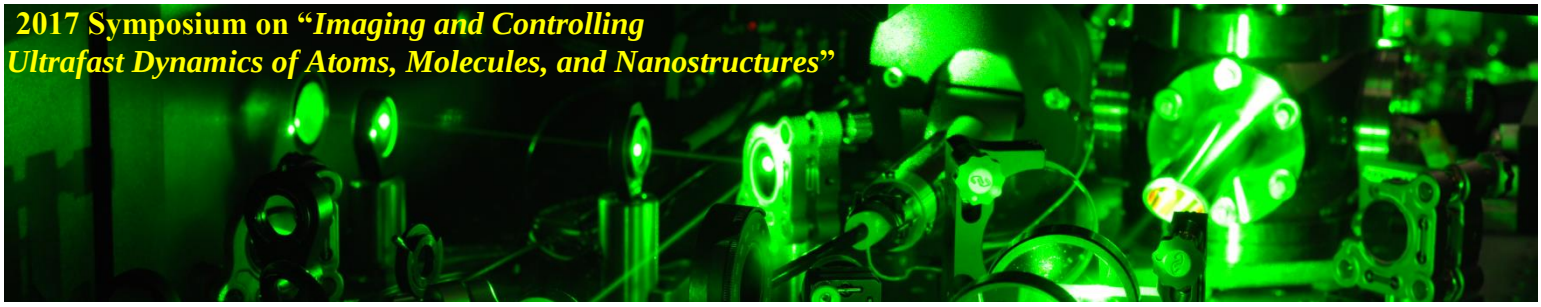
Nonlinear photochromic switching in the plasmonic field of a nanoparticle array

Christopher G. Elles

Associate Professor, Department of Chemistry, University of Kansas

We examine the non-resonant excitation of photochromic molecules in the plasmonic field of a nanoparticle array. The array was designed to enable ultrafast switching of electron diffraction, but also provides an interesting substrate to enable non-resonant optical activation of a photochromic film via two-photon excitation. Photochromic diarylethene molecules were deposited on top of the periodically ordered array of gold nanorods (170×40 nm) and then irradiated with <100 fs laser pulses. Irradiation at 800 nm drives the plasmon resonance of the nanoparticle array and induces the photochromic conversion of molecules via non-resonant two-photon excitation. Transmission measurements using broadband continuum laser pulses probe the progress of the photochemical electrocyclization reaction as molecules switch from a visible-absorbing closed-ring structure to a transparent open-ring structure. The spatial dependence of the two-photon conversion of molecules in the plasmonic near field of the array is modeled using calculated near-field intensities, and compared with similar measurements for a film of molecules on a glass substrate. The wavelength-dependent polarization in the near field of the array leads to interesting anisotropy effects in the transmission signal. The results emphasize the importance of both the spatial dependence and the anisotropy of the plasmonic fields in driving non-resonant photochromic reactions.





Coherent control of the exciton/biexciton system in a quantum dot ensemble

Steven Cundiff

Harrison M. Randall Collegiate Professor of Physics and Professor of Electrical Engineering and Computer Science, University of Michigan

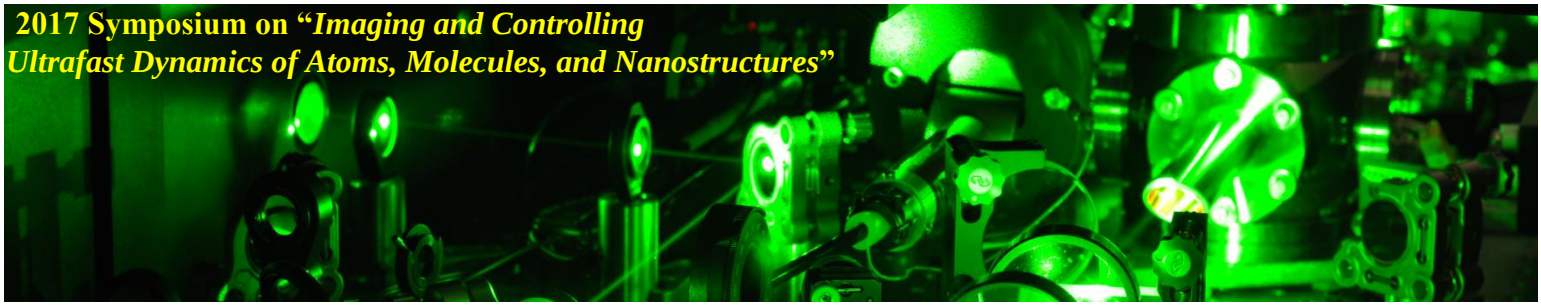
Coherent control of a strongly inhomogeneously broadened system, namely InAs self-assembled quantum dots, is presented. To circumvent the deleterious effects of the inhomogeneous broadening, which usually masks the results of coherent manipulation, pre-pulse two-dimensional coherent spectroscopy is used to provide a size-selective readout of the ground, exciton and biexciton states. Pre-pulse polarization dependent measurements confirm the behavior expected from selection rules. All measured spectra can be excellently reproduced by solving the optical Bloch equations for a 4-level system.

Mode-specific plasmonics examined using single-nanoparticle ultrafast imaging

Kenneth L. Knappenberger, Jr.

Associate Professor, Department of Chemistry and Biochemistry, Florida State University

I will describe recent advances in understanding the influence of nanoscale structure on plasmon-mediated electron dynamics. Steady-state extinction spectra of plasmonic nanoparticle networks are accurately described using hybridization models reminiscent of molecular orbitals. We have extended these molecular-based descriptions to account for nanoparticle electron dynamics by quantifying the coherence dephasing times of collective inter-particle plasmon modes of single nanostructures. In particular, we demonstrate that interference between plasmon modes of different angular momenta leads to increased coherence times. These observations are consistent with a model based on superpositions of molecular-like electronic states. These fundamental studies are important for understanding the structure-photonic-function relationship of plasmonic nanoparticles. This is because the spectroscopically determined coherence times reflect mode quality factors, which determine achievable amplification factors of optical signals. These new insights are made possible by recent advances in single-nanoparticle/molecule spectroscopy based on interferometric nonlinear optical detection. I will describe how the generation of sequences of phase-locked femtosecond laser pulses (33mrad phase stability) and their integration to an optical microscope were critical for this research.



High harmonic generation in solids: dynamics of multilevel adiabatic states spanning the band structure

Mette Gaarde

Professor of Physics, Department of Physics & Astronomy, Louisiana State University

We investigate high harmonic generation in a solid, modeled as a multilevel system dressed by a strong infrared laser field. We show that when the multilevel system originates from the Bloch states at the gamma-point of the band structure, the laser-dressed states map out the band structure away from the gamma-point as the laser field increases. We demonstrate that the cutoff energies and the relative strengths of the multiple plateaus that emerge in the harmonic spectrum can be understood both qualitatively and quantitatively by considering the dynamics of the laser-dressed system. Such a model was recently used to interpret the multiple plateaus observed experimentally in harmonic spectra generated by solid argon and krypton.

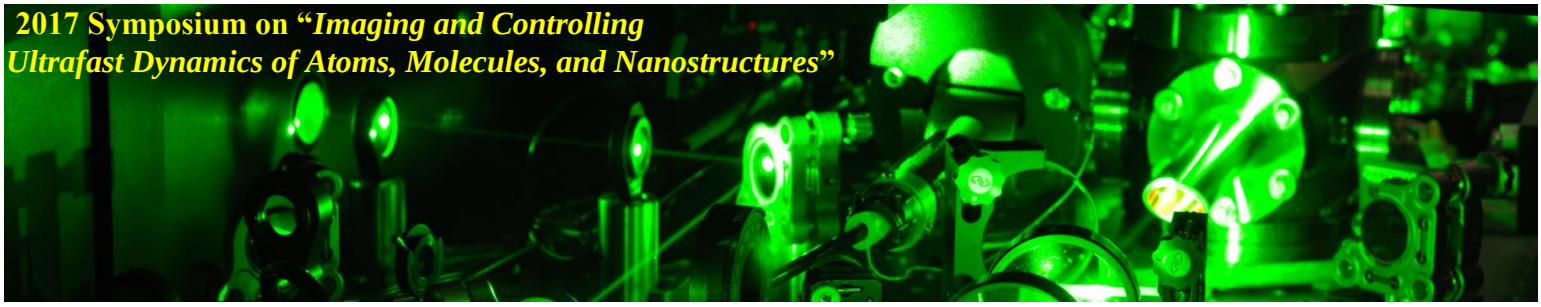
Finally, we discuss how this understanding leads to a semiclassical three-step picture in momentum space that describes the HHG process in a solid. In this picture, the delocalized electron first tunnels from the VB to the CB at the zero of the vector potential and then is accelerated on the CB as the vector potential increases and decreases through an optical half-cycle. The coherence between the VB and the CB populations leads to the emission of XUV radiation, with photon energies corresponding to the instantaneous energy difference between the VB and the CB. This means that each energy below the cutoff energy is emitted twice in each laser half-cycle.

Intense mid-infrared laser-cluster interactions

Louis F. DiMauro

Hagenlocker Chair of Physics/Professor of Physics, Department of Physics, The Ohio State University

An intense laser-plasma interaction generates bright, energetic bursts of electrons, ions and photons that have been utilized to enable a variety of applications. Nanoplasmas, formed from nanometer-sized clusters under intense field conditions, are distinguishable from conventional plasmas from bulk solids or gases since they can absorb a large amount of laser energy. During the subsequent relaxation, the field energy is released as incoherent radiation extending into the soft x-ray region or acceleration of neutral atoms, anions and cations. Several heating mechanisms have been considered in the past but restricted experimental control has been limiting. In this talk, we utilize the wavelength (λ) tuning of an intense mid-infrared source to expose and distinguish different heating mechanisms. This unexplored territory of nanoplasma physics harnesses the λ^2-scaling of the ponderomotive energy to impact the electron dynamics, which in turn has profound ramifications on the collisional heating. As the wavelength is increased we uncover a new nanoplasma mechanism based on vacuum heating while the contributions from the previously observed inverse Bremsstrahlung process diminishes. The vacuum heating is analogous to Brunel heating for planar solids but it is distinguished by sub-wavelength target size and gated ion emission. The study provides new insight into and control over the nanoplasma dynamics for refined production of high energy particles and unprecedented knowledge for guiding theories in laser interactions with complex systems.



Control and measurement of attosecond pulses with two-color fields

Carlos Trallero-Herrero

Associate Professor, Department of Physics, Kansas State University

Using two-color fields for the generation of high harmonic generation we show how attosecond pulses can be manipulated. First we show how a stable, high intensity 800 + 400 nm synthesized pulse can be used to increase the yield of XUV pulses. We then show how we can use a two-source interferometer to measure and control the phases of the train of attosecond pulses.

Deep inner-orbital ionization of diatomic molecules by strong laser fields

George Gibson

Professor, Department of Physics, University of Connecticut

While most studies of atoms and molecules in strong laser fields have focused on ionization and rescattering, evidence for direct excitation was observed in some of the earliest experiments in strong field physics, including ion, electron, and VUV spectroscopy. In this talk, I will focus on one excitation mechanism, deep inner-orbital ionization, using data from wavelength dependences, velocity-map imaging, and pump-probe spectroscopy. We find that strong field ionization of molecules is far more complicated than simple tunneling from the highest occupied molecular orbital.

State-of-the-art and next-generation sources for ultrafast hyperspectral imaging

Matthias Fuchs

Assistant Professor, Department of Physics and Astronomy, University of Nebraska–Lincoln

In this presentation, I will give an overview of ultrafast sources that are available and currently being developed at UNL. I will focus on sources capable of generating photon and electron pulses that cover an extremely large wavelength range, starting at soft X-rays down to less than 1 picometer. The expected pulse duration of these sources is only a few femtoseconds and they are temporally perfectly synchronized to a laser pulse. This makes them ideal tools for the complete investigation of ultrafast dynamics on the atomic scale.