

**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*<sup>1</sup>

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 – *Intense mid-infrared laser-cluster interactions*

Lou DiMauro – The Ohio 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 – *Mode-specific plasmonics examined using single-nanoparticle ultrafast imaging*

Ken Knappenberger – Florida State University

14:25 – *Control and measurement of attosecond pulses with two-color fields*<sup>1</sup>

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*<sup>1</sup>

Matthias Fuchs – University of Nebraska–Lincoln

16:20 – Closing remarks

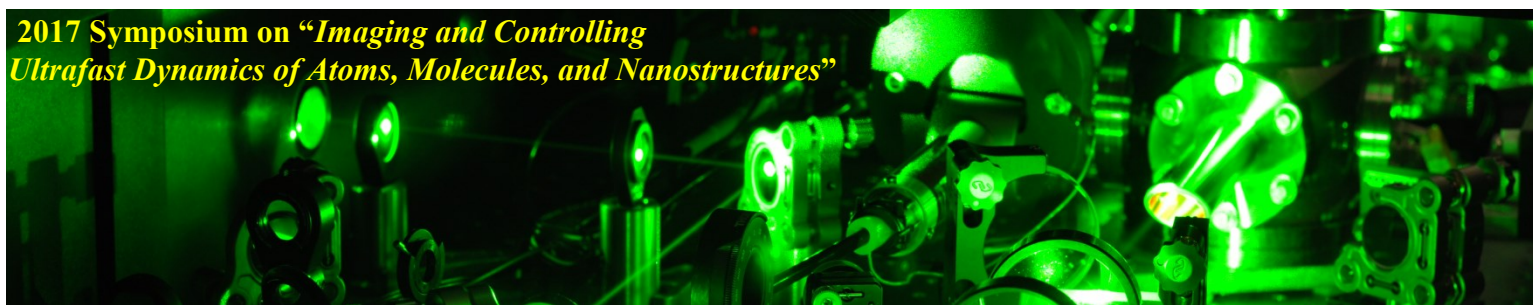
16:30 – Break

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

Room 227

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<sup>1</sup> 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.

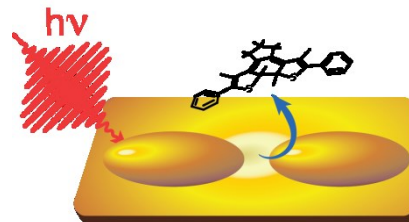
Extreme nonlinear optics in solids may provide an important source of VUV light and it is also a new diagnostic of materials. Solids can be perturbed, patterned, doped, or structured and each impresses its signature on the harmonics as they are created.

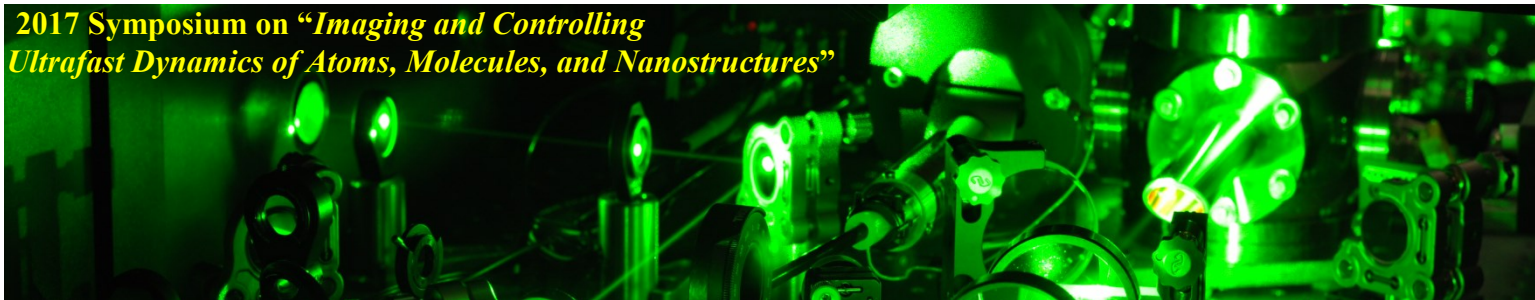
### **Nonlinear photochromic switching in the plasmonic field of a nanoparticle array**

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

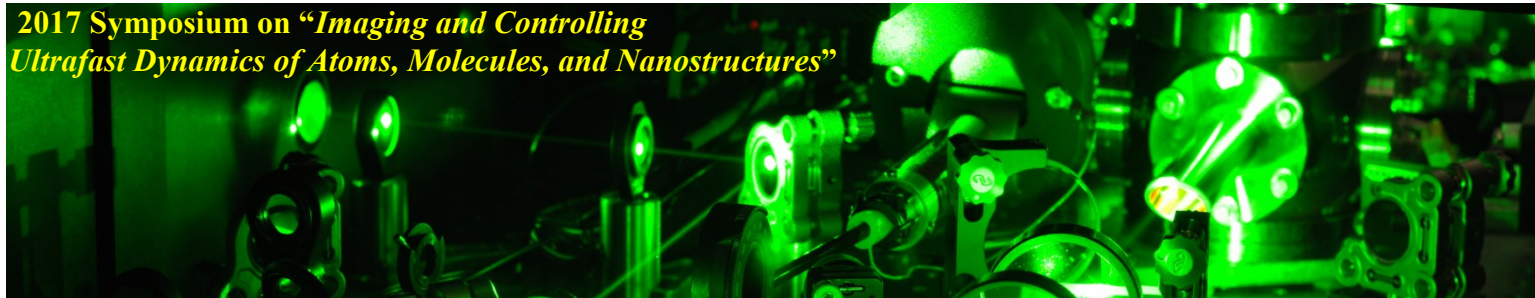
Coherent control of a strongly inhomogeneously broadened system, namely InAs self-assembled quantum dots, is presented. To circumvent the deleterious effects of the inhomogeneous broadening, which usually masks the results of coherent manipulation, pre-pulse two-dimensional coherent spectroscopy is used 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.

### **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 ( $\lambda$ ) tuning of an intense mid-infrared source to expose and distinguish different heating mechanisms. This unexplored territory of nanoplasma physics harnesses the  $\lambda^2$ -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.



### **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.

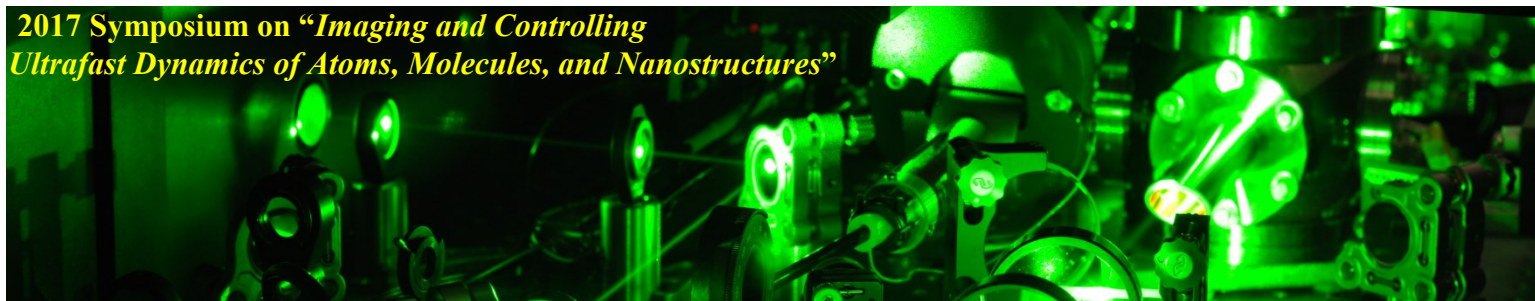
### **Mode-specific plasmonics examined using single-nanoparticle ultrafast imaging**

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-photonics-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.





### **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.

**11. Near-Gaussian spatial mode from an optically pumped acetylene-filled hollow-core fiber laser in the mid-IR**

Neda Dadashzadeh, Manasadevi P. Thirugnanasambandam, H.W. Kushan Weerasinghe, Benoit Debord, Matthieu Chafer, Frederic Gerome, Fetah Benabid, Brian R. Washburn, and Kristan L. Corwin

We report improvements in characteristics of a 3  $\mu\text{m}$  OPA pumped acetylene-filled hollow-core optical fiber laser, which are important for power scaling. The laser exhibits near-diffraction limited beam quality in the 3  $\mu\text{m}$  region with  $M^2 = 1.15 \pm 0.02$  measured at high pulse energy, and the highest mid-IR pulse energy from this laser system of 1.4  $\mu\text{J}$  is reported. Furthermore, the effects of output saturation with pump pulse energy are reduced by using longer fibers with low loss, and the laser shot-to-shot stability is improved as evident from smaller measured fluctuations compared to our previous reports. Finally, the slope efficiency is shown to be nearly independent of gas pressure, encouraging for further output power increases once higher pump pulse energies become available.

This work is supported by Air Force Office of Scientific Research (AFOSR) through grant FA9550-14-1-0024 and Air Force Research Laboratory (AFRL) under agreement number FA9451-17-2-0011. We acknowledge support from "Agence Nationale de la Recherche (ANR)" through grants PHOTOSYNTH and  $\Sigma$ \_LIM Labex Chaire and by "la région Limousin".

**12. A new phase retrieval algorithm for characterization of broadband single attosecond pulses**

Xi Zhao, Hui Wei, Yan Wu, and Chii-Dong Lin

Recent progress in high-order harmonic generation with few-cycle mid-infrared wavelength lasers has pushed light pulses into the water window region and beyond. These pulses have the bandwidth to support single attosecond pulses down to a few tens of attoseconds. Here we report a phase-retrieval method using the standard photoelectron streaking technique where an attosecond pulse is converted into its electron replica through photoionization of atoms in the presence of a time-delayed infrared laser. The iterative algorithm allows accurate reconstruction of the spectral phase of light pulses, from the extreme-ultraviolet (XUV) to soft X-rays, with pulse durations from hundreds down to a few tens of attoseconds. At the same time, the streaking laser fields can also be accurately retrieved.

This research was supported in part by the Chemical Sciences, Geosciences and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy, under Grant No. DE-FG02-86ER13491

**13. Extracting the wave-packet phase in high-order harmonic generation with a homodyne interferometer**

Jan Tross, Georgios Kolliopoulos, and Carlos Trallero-Herrero

A novel self-referencing XUV interferometer is used as a tool of extreme sensitivity to below attosecond stability. Two spatially distinct high-order harmonic sources are created with control on their relative brightness. The radiations from these two sources interfere in the far field providing a highly versatile XUV interferometer. With this tool, we investigate the dependence of the phase of higher order harmonics on the driving field intensity. Our results are compared with theoretical and experimental reports in the existing scientific literature. The error estimates are improved and help to draw a clear picture of the intensity dependent atomic dipole phase in the process of high-order harmonic generation, as expected from the three-step model. However, we observe differences from the strong field approximation: low order harmonics with photon energies below or near the ionization potential show an opposite dependence on the intensity.

Supported by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy.

**14. Retrieval of target structure information from laser-induced photoelectrons by few-cycle bicircular laser fields**

Van-Hung Hoang, Van-Hoang Le, Chii-Dong Lin, and Anh-Thu Le

We show that high-energy photoelectron momentum spectra for atoms in few-cycle bicircular laser pulses can be used to extract accurate elastic scattering differential cross sections of the target ion with free electrons. We find that the retrieval range for a scattering angle with bicircular pulses is wider than with linearly polarized pulses, although the retrieval method has to be modified to account for different returning directions of the electron in the continuum. This result can be used to extend the range of applicability of ultrafast imaging techniques such as laser-induced electron diffraction and for the accurate characterization of laser pulses.

This material is based on work supported in part by the NSF-ESPCoR Award No. IIA-1430493 and in part by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy under Grant No. DE-FG02-86ER13491

**15. Sub-cycle phase mapping of recollision dynamics**

Georgios Kolliopoulos, Jan Tross, and Carlos Trallero-Herrero

We study high harmonic generation in a two-color laser field by measuring its phase and amplitude. In an interferometric setup, we are able to measure the evolution of the harmonic phase in reference to higher harmonics generated from an identical single color field without the second harmonic present. By adding a second-harmonic field to the driving laser field of the one HHG source, we modify the spatiotemporal structure of the emanating harmonic beam; a direct comparison can be made with the unmodified harmonic beam coming out from the other source via their interference in the far field. As a function of the relative delay between the two fields, as well as the relative orientation of their polarizations, the interference structures of the odd harmonics are strongly modulated. Such a manipulation maps the subcycle dynamics of a recolliding electron to the phase domain.

This work was supported by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy under Grant No. DE-FG02-86ER13491.

**16. Two-color control over attosecond atomic and molecular processes**

T. Severt, J. Tross, G. Kolliopoulos, P. Timilisina, S. Buczek, C. Trallero-Herrero, B. Berry, M. Zohrabi, P. Feizollah, B. Jochim, Kanaka Raju P., J. McKenna, B. Gaire, K.D. Carnes, G.S.J. Armstrong, D. Ursrey, J.V. Hernández, F. Anis, B.D. Esry, and I. Ben-Itzhak

Two-color laser fields with a well-defined relative phase can be used to coherently control high-order harmonic generation (HHG) as well as nuclear and electronic dynamics on attosecond time-scales. Experimentally, we investigate how to improve the efficiency of HHG driven by two-color 800/400-nm laser fields. By tailoring the electric field to optimize short trajectory electron contributions, we observe about an order of magnitude enhancement, as predicted by theory [1]. In addition, we explore the dissociation of a  $D_2^+$  beam induced by an 800/400-nm ultrafast laser field. In particular, the relative phase between the fields is scanned to control the deuteron emission direction with respect to the laser polarization. By comparing the phase shifts of the spatial asymmetry between the  $v=7$  and  $v=8$  vibrational states of the  $1\sigma_g$  ground electronic state, we observe a 53-as delay.

[1] Cheng Jin, Gouli Wang, Hui Wei, Anh-Thu Le, and C.D. Lin, Nat. Comm. **5**, 4003 (2014)

Supported by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy. The HHG-enhancement project is also supported by the National Science Foundation under Award No. IIA-1430493.

**17. Enhancing high-order-harmonic generation by time delays between two-color, few-cycle pulses**

Dian Peng, Liang-Wen Pi, M.V. Frolov, and Anthony F. Starace

Use of time delays in high-order-harmonic generation (HHG) driven by intense two-color, few-cycle pulses is investigated in order to determine means of optimizing HHG intensities and plateau cutoff energies. Based upon numerical solutions of the time-dependent Schrödinger equation for the H atom as well as analytical analyses, we show that introducing a time delay between the two-color, few-cycle pulses can result in an enhancement of the intensity of the HHG spectrum by an order of magnitude (or more) at the cost of a reduction in the HHG plateau cutoff energy. Results for both positive and negative time delays as well as various pulse carrier-envelope phases are investigated and discussed.

Our TDSE and time-frequency analysis calculations were supported in part by NSF Grant No. PHYS-1505492 and were carried out at the Holland Computing Center of the University of Nebraska-Lincoln (UNL). A collaborative visit to UNL by M.V.F. and our analytic analyses were supported in part by NSF EPSCoR IRR Track II Research Award No. 1430519. The research of M.V.F. is supported in part by the Russian Science Foundation through Grant No. 15-12-10033.

**18. Multistart spiral electron matter-wave vortices in multiphoton ionization by time-delayed circularly polarized UV pulses**

J.M. Ngoko Djiokap, A.V. Meremianin, N.L. Manakov, S.X. Hu, L.B. Madsen, and Anthony F. Starace

The novel phenomenon of multistart spiral electron matter-wave vortices is predicted for the momentum distribution in the polarization plane following multiphoton single ionization of the helium atom by a pair of time-delayed, circularly polarized ultrashort laser pulses. These multistart electron vortex patterns are found to be sensitive to the carrier frequency, handedness, time delay and relative phase of the two pulses. Our numerical predictions are based on solving the six-dimensional two-electron, time-dependent Schrödinger equation; and they are interpreted analytically using perturbation theory. Our prediction of electron vortices has been recently observed experimentally [Phys. Rev. Lett. **118**, 053003 (2017)] in multiphoton ionization of potassium atoms by time-delayed oppositely circularly polarized femtosecond pulses.

This work was supported in part by U.S. Department of Energy (DOE), Office of Science, Basic Energy Sciences (BES), under Award No. DE-FG03-96ER14646, and in part by NSF EPSCoR IRR Track II Research Award No. 1430519.

**19. Enhanced ionization observed in pump-probe studies of fast ion beams**

Ben Berry, M. Zohrabi, T. Severt, Bethany Jochim, Peyman Feizollah, Kanaka Raju P., Jyoti Rajput, Youliang Yu, K.D. Carnes, B.D. Esry, and I. Ben-Itzhak

We have conducted a first-of-its-kind NIR-pump—NIR-probe measurement on a few-keV  $\text{HD}^+$  beam target in which the first pulse initiates the dissociation of  $\text{HD}^+$  and the second pulse ionizes the molecule during its dissociation. An enhancement in the ionization yield of the dissociating wave packet was observed at large time delays, specifically around 200 fs which we estimate to correspond to an internuclear separation of about 60 a.u. This enhancement is surprising given the extremely weak interaction between the ionic and neutral fragments at this large distance. To further explore this unexpected enhanced ionization we have recently conducted similar measurements on an  $\text{Ar}_2^+$  beam, for which the dissociating wave packet is expected to be much slower.

This work is supported by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy. K.R.P acknowledges support by the National Science Foundation under Award No. IIA-1430493



## 20. Photodetachment microscopy in time-dependent fields

Harindranath Ambalampitiya and Ilya Fabrikant

Photodetachment microscopy studies spatial distribution of photodetached electrons in static electric fields. The theory of this phenomenon was recently extended [1] to time-dependent electric fields in the terahertz range. In the present paper we use the semiclassical propagator (time-dependent Green's function) for investigation of temporal and spatial interference of classical electron trajectories in ac fields within a broad frequency range from radio to terahertz frequencies. The chosen length scale corresponds to the geometry of a traditional photodetachment microscopy experiment [2]. The propagator approach allows us to treat singularities in temporal and spatial distributions due to bifurcations, when the trajectories emerge in pairs from the complex space-time domain.

[1] B. C. Yang and F. Robicheaux, Phys. Rev. A **92**, 063410 (2015)

[2] C. Blondel, C. Delsart, and F. Dulieu, Phys. Rev. Lett. **77**, 3755 (1996)

This work is supported by the National Science Foundation under Award No. IIA-1430519.

## 21. Strong field studies of $F_2^-$ dissociation and photodetachment

Ben Berry, Bethany Jochim, T. Severt, Peyman Feizollah, Kanaka Raju P., K.D. Carnes, B.D. Esry, and I. Ben-Itzhak

While molecular anions have long been used as tools to investigate molecular dynamics, very few studies focus on their behavior in a strong field. In this work, we explore the strong field dissociation and photodetachment of  $F_2^-$  under a variety of laser conditions. The use of a keV beam enables the measurement of all molecular fragments except electrons, and we obtain the full 3D momentum of breakup using a coincidence momentum imaging technique. The measurement of the final products of photodetachment (namely  $F_2$  and  $F + F$ ) facilitates the understanding of possible photodetachment mechanisms. In addition, we identify dissociation pathways and use the measured kinetic energy release (KER) to evaluate the initial rovibrational population of the anion.

This work is supported by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy. K.R.P acknowledges support by the National Science Foundation under Award No. IIA-1430493.

## 22. XUV pump and near-infrared probe studies of $CO_2$ dissociation dynamics

Kanaka Raju P., Xiang Li, Y. Malakar, B. Kaderiya, W.L. Pearson, Wei Cao, P. Trapp, D. Trabert, F. Wilhelm, D. Rolles, I. Ben-Itzhak, and A. Rudenko

Ultrafast dynamics of the  $CO_2$  molecule have recently been studied by employing a pump-probe technique using broadband ultrashort XUV-pump pulses containing the 11th to 17th harmonics of a near-infrared laser (NIR) [1]. Here, we present the results of a complementary experiment employing narrowband ( $\sim 100$  fs), single harmonic (11th or 13th) pulses to excite molecular wave packets to specific states of  $CO_2^+$ , which are probed by NIR-induced dissociation. We measure energy- and angle-resolved yields of all charged reaction fragments as a function of XUV-NIR delay using a reaction microscope. In particular, the delay dependence of  $O^+$  and  $CO^+$  ion production for parallel and perpendicular NIR and XUV polarizations are contrasted with the data obtained by Timmers *et al.* [1] using ultrashort broadband train of harmonics.

[1] H. Timmers *et al.*, Phys. Rev. Lett. **113**, 113003 (2004)

This project is supported by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy. K.R.P. and W.L.P. acknowledge support by the National Science Foundation under Award No. IIA-1430493. P.T. and F.W. were supported by the DAAD RISE-Worldwide program.

**23. Ultrafast dynamics in photoionized CO<sub>2</sub> studied by high repetition rate setup for time resolved XUV-IR experiments employing ion and electron momentum imaging**

S.J. Robatjazi, S. Pathak, W.L. Pearson, Kanaka Raju P., J. Powell, X. Li, B. Kaderiya, I. Ben-Itzhak, D. Rolles, and A. Rudenko

We present the results of a pump-probe experiment studying ultrafast dynamics of photo-ionized CO<sub>2</sub> molecule using a combination of a 10 kHz high-harmonic generation source with a velocity map imaging spectrometer. The HHG source is capable of delivering XUV radiation of less than 30 fs pulse duration at the photon energies up to 100 eV. For the CO<sub>2</sub> experiment, the multi-harmonics pulse containing 11th to 33th harmonics of a 790 nm NIR laser, is used to excite molecular wave packets in CO<sub>2</sub><sup>+</sup>, which are then probed by near-infrared-induced dissociation. The measured delay dependent yield of CO<sup>+</sup> ion exhibits an oscillatory structure with a period of ~3.2 ps, the origin of which remains unclear. We compare our results to the data reported in [1] for a shorter harmonics train, and to our results obtained with isolated 11th or 13th harmonics.

[1] H. Timmers *et al.*, Phys. Rev. Lett. **113**, 113003 (2014)

This project is supported in part by National Science Foundation (NSF-EPSCOR) Award No. IIA-1430493 and in part by the Chemical Science, Geosciences, and Bioscience Division, Office of Basic Energy Science, Office of Science, U.S. Department of Energy. K.R.P. and W.L.P. thank NSF-EPSCOR for their support.

**24. Imaging dynamics of metastable molecules produced by intense, ultrafast laser pulses**

Bethany Jochim, T. Severt, Reid Erdwien, Y. Malakar, Peyman Feizollah, Ben Berry, Jyoti Rajput, B. Kaderiya, Farzaneh Ziaee, Kanaka Raju P., M. Zohrabi, U. Ablikim, W.L. Pearson, K.D. Carnes, D. Rolles, A. Rudenko, B.D. Esry, and I. Ben-Itzhak

Interactions of molecules with intense, ultrafast laser pulses or fast charged particles can readily produce metastable molecular ions. The lifetimes of these transient systems can range from picoseconds to even seconds [1]. Molecular dissociation dynamics involving metastable states are studied employing coincidence momentum imaging. In one case, the metastable molecules typically survive 100's of nanoseconds to a few microseconds, a significant fraction of their flight time to the detector. The lifetimes of the metastable states involved and the complete 3-D momenta of the fragments are retrieved. In the second case, for which we examine sequential fragmentation of triatomic molecules, the lifetimes of the intermediate metastable molecules are typically picoseconds. We demonstrate an approach to disentangle sequential from concerted fragmentation in any spectrum, thereby facilitating deeper understanding of these processes.

[1] D. Mathur, Phys. Rep. **391**, 1 (2004); Ibid. **225**, 193 (1993)

This work is supported by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy. K.R.P acknowledges support from the National Science Foundation under Award No. IIA-1430493.

**25. Strong-field fragmentation of diiodomethane studied with time-resolved three-body Coulomb explosion**

B. Kaderiya, Y. Malakar, Kanaka Raju P., T. Severt, W.L. Pearson, F. Ziaee, K. Jensen, J. Rajput, I. Ben-Itzhak, D. Rolles, and A. Rudenko

Laser Coulomb explosion imaging (CEI) is an efficient tool for mapping time-dependent changes of molecular geometry in many light-induced bond breaking or rearrangement processes. Here we apply the three-body CEI technique to map in space and time nuclear wave packets created in strong-field ionization and dissociation of diiodomethane molecules. Analyzing coincident three-particle momentum maps in the triply ionized final state, we disentangle different ionization and break-up pathways and trace the time evolution of both bond lengths and angles for major reaction channels. By combining different representations of the three-body breakup (kinetic energy release vs. relative ion emission angle, Dalitz plots, Newton diagrams, etc.), we identify contributions due to bound and dissociating parts of the nuclear wave packet in the singly charged ionic state, observe signatures of  $I_2/I_2^+$  elimination and highlight the role of intermediate long-lived doubly charged states.

This work is supported by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy. K.R.P. and W.L.P. are supported by NSF Award No. IIA-143049, K.J. is supported by the NSF-REU grant No. PHYS-1461251.

**26. Multiphoton ionization and dissociative rearrangement processes in photoionized aniline**

Collin J. McAcy, Timothy D. Scarborough, and Cornelis J.G.J. Uiterwaal

We report on two select photodynamical processes observed in laser-ionized aniline ( $C_6H_5NH_2$ ): resonance-enhanced multiphoton ionization (REMPI), and dissociative rearrangement. Using 50-fs, 800-nm laser pulses in tandem with our unique spatially-resolved time-of-flight mass spectrometer, we record ion mass spectra and ion yields as a function of intensity without the focal volume effect. On a double-logarithmic plot of parent ion yield and laser intensity ranging from  $6 \times 10^{13}$  to  $3 \times 10^{14}$  W/cm<sup>2</sup>, we find two regions of integer slope separated by a kink: a signature of REMPI. Comparing our experimental photon energy to aniline's known excitations leads us to conclude that aniline undergoes (3 + 2) REMPI under these conditions. We also find ionic benzene in aniline's ion mass spectra, and determine that it must be the result of a molecular rearrangement which may follow dissociation of the amino group from the phenyl ring.

The authors acknowledge the Max-Planck-Institute of Quantum Optics in Germany, specifically Dr. Hartmut Schröder, for lending us the reflectron used in these experiments. This material is based upon work supported by the National Science Foundation Grant Nos. PHY-0855675 and PHY-1005071, as well as the National Science Foundation EPSCoR RII Track-2 CA Award No. IIA-1430519.

**27. Linear regression from time domain measurements to extracted observables in molecular frame: An example of SO<sub>2</sub>.**

Huynh Lam, Suresh Yarlagadda, Rajesh Kushawaha, and Vinod Kumarappan

The angle dependence of non-dissociative ionization and fragmentations of Sulfur dioxide (SO<sub>2</sub>) has been studied. We first launch a rotational wave packet by 1D impulsive alignment technique created by a linearly polarized non-resonant laser pulse, we then measure the ion yield as a function of delay between probe and aligning pulse. Finally, a fitting procedure was used to retrieve the angle dependent signals.

This work is funded by the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy. H.L., S.Y. and V.K. are partly supported by the National Science Foundation under Award No. IIA-1430493.

**28. Laser-induced electron diffraction with aligned molecules and two-color fields: A progress report**

Huynh Lam, Suresh Yarlagadda, Rajesh Kushawaha, and Vinod Kumarappan

Laser induced electron diffraction (LIED) can image molecular structure with picometer spatial and few-fs temporal resolution by using the electrons generated by an ultrafast laser from the molecule itself for a diffraction measurement. This method has the capability to image molecules undergoing ultrafast structural changes following photo-excitation. We would like to apply this technique on adiabatically aligned molecules to obtain detailed structural information, and probe by a synthesized two-colors field to enhance rescattering efficiency.

This work is supported in large part by the National Science Foundation under Award No. IIA-1430493. The HITS laser and OPA were funded mainly by NSF MRI and AFOSR DURIP grant. Part of the VMI setup was built with funds from the Chemical Sciences, Geosciences, and Biosciences Division, Office of Basic Energy Sciences, Office of Science, U.S. Department of Energy. The same DOE grant supported R.K. and partly supported H.L., S.Y. and V.K.

**29. Ultrafast dynamics of diiodotetrafluoroethane and simulated structural retrieval of trifluoroiodomethane with ultrafast electron diffraction**

Kyle Wilkin, Jie Yang, Ryan Coffee, James Cryan, Markus Guehr, Kareem Hegazy, Renkai Li, Michael Minitti, Pedro Nunes, Xiaozhe Shen, Thomas Wolf, Xijie Wang, and Martin Centurion

We examine the photo-induced dynamics of diiodotetrafluoroethane ( $C_2F_4I_2$ ) and its transient state ( $C_2F_4I$ ) as it dissociates to ( $C_2F_4$ ) using the 3.7 MeV ultrafast electron diffraction (UED) setup at SLAC. The structure of the intermediate state has been measured by the Zewail group with a resolution of 5 ps. Our resolution of 200 fs allows us to capture the coherent nuclear motion during the reaction. In addition, we discuss simulated structural retrieval of aligned Trifluoroiodomethane ( $CF_3I$ ) using two methods. The first method is a direct transform of the two dimensional intensity image of simulated electron diffraction. The second uses iterative phase retrieval techniques to access the full structure of the molecule. The strengths and weaknesses of both methods are discussed.

The experimental work was supported by the U.S. Department of Energy Office of Science, Office of Basic Energy Sciences under Award Number DE-SC0014170. The theoretical work was supported by the National Science Foundation EPSCoR RII Track-2 CA Award No. IIA-1430519.

**30. Generation and measurement of bright ultrafast electron pulses for pump-probe gas phase diffractive imaging**

Omid Zandi, Kyle J. Wilkin, Yanwei Xiong, and Martin Centurion

We have developed a table-top electron gun that delivers femtosecond electron pulses with a high beam current. The photoemitted electron pulses are compressed by a radio-frequency electric field to sub 400 fs. With this new setup, we have reduced the time it takes to capture high quality diffraction patterns from one hour to 20 seconds, while also improving on the temporal resolution by more than a factor of two. We have fabricated a laser activated streak camera to evaluate the electron pulse duration in situ that has a resolution of about 200 fs. The setup is also equipped with a tilted front laser pump that is velocity matched to the electrons, which reduces the temporal blurring due to velocity mismatch to less than 100 fs.

These experiments were supported by the U.S. Department of Energy Office of Science, Basic Energy Sciences under Award Number DE-SC0014170 and the development of the streak camera was supported by the U.S. Air Force Office of Scientific Research, Ultrashort Pulse Laser-Matter Interactions program under Award Number FA9550-12-1-0149.



### 31. Development of a laser-plasma accelerator for ultrafast electron diffraction

Ping Zhang, Yunhao Fan, Matthew S. Robinson, Martin Centurion, and Matthias Fuchs

Progress in laser wake field acceleration (LWFA) has made it possible to develop a new electron source for the ultrafast electron diffraction (UED). This novel source has the potential to become an indispensable tool for studying atomic structural dynamics on the femtosecond time scale and sub-Ångström spatial resolution. By using the Archimedes laser at UNL (800 nm, 100 mJ and 35 fs), we have generated electron beams of the central kinetic energy of ~0.5 MeV. With a new technology in the novel supersonic gas jet design, we are working on the development of a novel electron source of the peak kinetic energy ~1 MeV and 100 pC/per shot and application of the source in first UED experiment.

This work is supported by the National Science Foundation EPSCoR RII Track-2 CA Award No. IIA-1430519.

### 32. Source comparison of short wavelength ultrafast X-ray pulses

Rafal Rakowski, Ping Zhang, Brendan Kettle, Donald Umstadter, Artem Rudenko, and Matthias Fuchs

Short-wavelength X-ray photons with energies on order of 100 keV have interesting characteristics, such as comparably low absorption, high spatial resolution and the ability to access inner-shell states of high-Z atoms. This makes them attractive for many applications, ranging from studies of bulk materials, scattering experiments that can explore a large fraction of reciprocal space in a single shot, to imaging of solids and liquids at high pressures or in extreme conditions. Ultrashort, femtosecond soft to hard X-ray pulses allow the direct imaging of atomic dynamics simultaneously on both its natural time and length scales. However, so far studies of atomic dynamics using high-energy (~100 keV) X-rays have been extremely limited due to the lack of sources that can generate sufficiently short (femtosecond) pulses in this regime. Here, we present first diffraction experiments using a novel ultrashort all-laser based Thomson backscattering source with a photon energy of ~70 keV. The source is based on inverse Compton scattering (Thomson backscattering) of optical photons from relativistic electron beams generated by a laser-wakefield accelerator. We compare this source to a laser-driven betatron source that holds the promise to generate significantly higher photon flux in a photon energy range of 1-30 keV. The ultimate goal of the research is to study relaxation dynamics of molecules containing a high-Z atom after inner-shell ionization.

This work was supported by the National Science Foundation EPSCoR RII Track-2 CA Award No. IIA-1430519 and No. IIA-1430493.

### 33. Generating long wave infrared few cycle pulses for use in strong field science

Derrek J. Wilson, Adam M. Summers, Stefan Zigo, Brandin Davis, Jeff Powell, Seyyed Robotjazi, Artem Rudenko, and Carlos Trallero-Herrero

We demonstrate a technique of difference frequency generation (DFG) which is capable of generating fields with carrier wavelengths ranging from 5.5  $\mu\text{m}$  to 8.5  $\mu\text{m}$  and a peak power at the Gigawatt level. This source is capable of multiply ionizing atomic and molecular targets from their ground states. Xe charge states up to  $\text{Xe}^{3+}$  are observed using an ion time-of-flight apparatus. These results open the door to a new wavelength region to study strong-field science.

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**34. Wavelength scaling of strong field effects in GaAs**

Derrek J. Wilson, Adam M. Summers, Giriraj Jnawali, C.M. Sorensen, J.H. Edgar, and Carlos Trallero-Herrero

We measure the electron excitation rate in the strong field limit for GaAs by observing the absorption rate of a laser. Using an optical parametric amplifier (OPA), we are able to scan the wavelength across a full octave of the NIR. In particular, the intensity dependence shows that the excitation rate can be saturated along directions of the lattice with higher density of states. This saturation effect is more dramatic as the wavelength increases.

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**35. Blending teaching and research: Low-cost surface-enhanced Raman scattering to probe the reaction dynamics of a molecular photoswitch**

Jordan H. Mantha and Christopher G. Elles

Surface-enhanced Raman scattering (SERS) is well suited as an example of a modern spectroscopic tool that has wide application, but commercial instruments are often cost-prohibitive for small or primarily undergraduate institutions. A low-cost SERS instrument and educational modules that can be used in undergraduate analytical chemistry, physical chemistry, and molecular/optical physics courses are being developed. This surface-enhanced Raman system is also being investigated as a probe of the reaction dynamics of diarylethene photoswitches in plasmonic fields.

This work has been supported by the National Science Foundation under Award No. IIA-1430493 and U.S. Department of Education under Award No. P031F140031

**36. Comparative time-resolved photoelectron spectroscopy from Cu(100) and Cu(111) surfaces**

M.J. Ambrosio and Uwe Thumm

We study the effects of initial-state modeling and Fresnel reflection of the incident IR pulse on time-resolved IR-laser-assisted XUV photoelectron spectroscopy of Cu(100) and Cu(111) surfaces. These effects are shown to strongly influence the photoelectron spectra, and thus need to be incorporated in theoretical calculations. Comparative RABBITT phases are discussed as a way of eliminating the phase shifts introduced by the unknown high harmonic phases of the XUV pulse train.

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**37. Streaked photoemission from polycrystalline Au**

M.J. Ambrosio and U. Thumm

We present streaking traces for polycrystalline gold, generated by single-photon photoelectron emission in attosecond XUV pulses and an assisting non-ionizing time-delayed IR pulse. Our model is first verified with available experimental data from Mg(0001) for photon energies of 118 eV, and then applied to polycrystalline gold for photon energies of 93 eV and 20 eV. We analyze the effects of Fresnel reflection of the incident IR pulse and final state approximations on the center of energy and higher moments of the streaking traces.

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**38. Nonlinear graphene metasurfaces with enhanced electromagnetic functionalities**

Boyuan Jin, Tianjing Guo, and Christos Argyropoulos

The conductivity of graphene at THz and infrared (IR) frequencies is dominated by intraband transitions and can be characterized by the Drude model. Strong surface plasmons can be excited at the surface of graphene in this frequency regime. Even stronger resonance conditions in the transmission or reflection spectrum can be obtained when graphene is patterned to periodic rectangular patches or strips forming new planar graphene metasurfaces. It is interesting that the third-order nonlinear conductivity of graphene is much larger compared to the nonlinear properties of thin metallic and dielectric layers measured at the same frequency range. We will employ the enhanced nonlinear properties of graphene metasurfaces to boost several relative weak THz and IR optical nonlinear processes, such as third-harmonic generation and four-wave mixing. In addition, we will use nonlinear graphene metasurfaces to demonstrate tunable negative refraction and coherent perfect absorption.

This work has been partially supported by the National Science Foundation under Award No. IIA-1430519.

**39. Ultrafast electron transport between atomic layers**

Yuanyuan Li, Matthew Z. Bellus, Ming Li, Xiao Cheng Zeng, and Hui Zhao

Two-dimensional materials have attracted a lot of attention recently due to their ability to be stacked via van der Waals forces into unique customizable heterostructures. In these structures, electron transport between the atomic layers is a key process for the applications in optoelectronics and electronics. Here we use two examples to show that ultrafast laser measurements can provide valuable information about this process. First, electron transport between two randomly stacked monolayers of MoSe<sub>2</sub> was time resolved by utilizing a graphene layer as a rapid carrier recombination channeling. We found a transfer time of a few picoseconds. Second, in a MoS<sub>2</sub>-ReS<sub>2</sub> structure, electrons excited in the wide-bandgap layer of MoS<sub>2</sub> transfer to the narrow-bandgap ReS<sub>2</sub> in about one picosecond. Such efficient interlayer charge transfer indicates the feasibility of developing van-der-Waals multilayer structures with novel electronic transport performance.

This work is supported by the National Science Foundation under Award No. IIA-1430493.

**40. Retrieving plasmonic field information from metallic nanospheres using attosecond photoelectron streaking spectroscopy**

Jianxiong Li, Erfan Saydanzad, and Uwe Thumm

Streaked photoemission by attosecond extreme ultraviolet (XUV) pulses into an infrared (IR) or visible streaking pulse, which allows for sub-fs-resolution study of the streaking field, holds promise of studying the plasmonic response of metallic nanoparticles to the streaking pulse. We calculated the plasmonic field induced by streaking pulses for 10 to 200 nm diameter Au, Ag and Cu nanospheres, and obtained streaked photoelectron spectra by employing our quantum-mechanical model. Our simulated spectra show significant oscillation-amplitude enhancement and phase shift for all three metals relative to the results excluding the induced plasmonic field, leading to the successful reconstruction of the plasmonic field enhancement and phase shift profile for each material.

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**41. Theoretical study of imaging the plasmonic field enhancement on the surface of the gold nanosphere by using attosecond streaking spectroscopy**

Erfan Saydanzad, Jianxiong Li, and Uwe Thumm

Attosecond time-resolved spectroscopy has been shown to be a powerful method for investigating the electronic dynamics in atoms, and this technique is now being transferred to the investigation of electronic excitations, electron propagation, and collective electronic (plasmonic) effects in near solid surfaces [1,2] and nanoparticles [1,3]. By sampling over classical photoelectron trajectories, we simulated IR-streaked XUV-photoemission spectra for gold nanospheres of 5 and 50 nm radius. Based on our numerical results, we show how spatio-temporal information of the sub-infrared-cycle plasmonic and electronic dynamics is embedded in streaked spectra.

[1] U. Thumm, Q. Liao, E.M. Bothschafter, F. Süßmann, M.F. Kling, and R. Kienberger, *Handbook of Photonics*, Vol. 1, (Wiley 2015), p. 387

[2] Q. Liao and U. Thumm, *Phys. Rev. A* **92**, 031401(R) (2015)

[3] J. Li, E. Saydanzad, and Uwe Thumm, *Phys. Rev. A* **94**, 051401(R) (2016)

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**42. X-ray imaging of ultrafast laser driven nanoplasma formation and evolution**

J. Powell, A. Summers, C. Peltz, P. Rupp, Q. Liu, I. Halfpap, E. Antonsson, K. Sander, Y. Kumagai, B. Langer, T. Osipov, D. Ray, S. Moeller, R. Coffee, G. Coslovich, M. Bucher, T. Gorkhover, B. Erk, D. Rompotis, C. Bomme, R. Boll, S. Bari, J. Correa, B. Rudek, M. Gallei, E. Rühl, C. Trallero-Herrero, D. Rolles, C. Bostedt, M. Kling, A. Rudenko, and T. Fennel

A free electron laser (FEL) is used to image and measure the dynamics of nanoscale objects with femtosecond level temporal precision. We used a dynamic X-ray imaging technique to study nanoplasmas created through the interaction of single SiO<sub>2</sub> nanoparticle with an intense ( $>10^{14}$  W/cm<sup>2</sup>) 800 nm laser. We studied this process using 800 eV, 80 fs radiation produced by the Linac Coherent Light Source (LCLS) at SLAC and also using 250 eV, 100 fs light produced by the FEL in Hamburg (FLASH) at DESY. During the nanoparticle/laser interaction, the optical laser ionizes the outer shell of the nanoparticle, leading to the formation of a nanoplasma. This process can be described as surface ablation followed by a core melt-off. Both effects are measurable as deviations from hard-sphere scattering in the X-ray diffraction images. Using a pump-probe scheme, the dynamics of the laser-driven nanoplasma can be "watched" in femtosecond resolution.

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**43. Progress report on electron emission from nanotips and gold nanowires**

Eric Jones, Gobind Basnet, Wayne Huang, Bret Flanders, and Herman Batelaan

Laser-induced field emission of electrons from tapered metallic nanotips is the current standard for ultrafast electron microscopy and diffraction. Single-crystal gold nanowires represent novel electron sources, whose growth can be tailored to suit different applications. Dynamics of electrons on the surface and in the bulk of such nanowires may be revealed through the particular emission characteristics. We present progress in fabricating improved tungsten and gold tapers. Mechanisms are studied through the electron emission dependence on laser intensity and polarization, and by timing coincidence measurements between electron and laser pulses. Considerations include thermal emission from laser heating, tip crystallinity, and trajectories of emitted electrons between the source and detector. Understanding and controlling the various features of emission from tapers will then be applied to nanowires. Fabrication of GaAs nanotips, possible sources of spin-polarized electrons, is demonstrated.

We gratefully acknowledge support from NSF-EPSCoR NE-KS Track-II Awards numbers IIA-1430519 and IIA-1430493.

**44. Enhanced photoswitching using a gold plasmonic nanorod array**

Christopher J. Otolski, Christos Argyropoulos, and Christopher G. Elles

We examine the reactions of photochromic molecules following indirect excitation in the plasmonic field of a patterned array of gold nanorods. Incident 800-nm laser pulses induce the oscillating plasmonic field, which then excites nearby molecules through a nonlinear two-photon interaction. The enhanced electric field due to the oscillating surface plasmons assists in the non-resonant energy transfer to the molecule. When the photochromic molecule is in the near-field of the nanorod array a faster initial absorbance decay is observed, compared with a film of molecules in the absence of nanorods. The modified switching rate is due to stronger electric fields between the tips of the gold nanorods, but also depends on the spatially selective probing due to the near-field structure of the probe light. The information gathered in this study emphasizes the importance of the spatial dependence and anisotropy of the enhanced electric field in controlling the rate of conversion of photochromic molecules on a nanoplasmonic array.

This work is supported by the National Science Foundation under Award No. IIA-1430493.

**45. Highly efficient and anomalous charge transfer in van der Waals trilayer semiconductors**

Frank Ceballos, Ming-Gang Ju, Samuel D. Lane, Xiao Cheng Zeng, and Hui Zhao

Two-dimensional materials, such as graphene and monolayer transition metal dichalcogenides, allow the fabrication of multilayer structures without lattice matching restriction. A central issue in developing such artificial materials is to understand and control the interlayer electron transfer process, which plays a key role in harnessing their emergent properties. Recent studies revealed that the electron transfer in heterobilayers occurs on ultrafast time scales. However, there is still a lack of fundamental understanding on how this process can be so efficient at van der Waals interfaces. Here we show evidence suggesting the coherent nature of such interlayer electron transfer. In a trilayer of MoS<sub>2</sub>-WS<sub>2</sub>-MoSe<sub>2</sub>, electrons excited in MoSe<sub>2</sub> transfer to MoS<sub>2</sub> in about one picosecond. Surprisingly, these electrons do not populate the middle WS<sub>2</sub> layer during this process. Calculations showed the coherent nature of the charge transfer and reproduced the measured electron transfer time.

This work is supported by the National Science Foundation under Award No. IIA-1430493.

**46. Pressure broadening and frequency shifts in acetylene-filled photonic microcells**

Sajed Hosseini-Zavareh, Ryan Luder, Manasadevi Thirugnanasambandam, Brian R. Washburn, and Kristan L. Corwin

We have developed a gas filled photonic microcell from a photonic bandgap fiber that can replace typical optical frequency standard reference materials. Comparison with sub-Doppler gas-filled fiber references and analysis of pressure broadening and shift indicate accuracies of about  $\pm 2$  MHz. We have reported efficiency of the PMC measuring the butt coupling through 200 and 400  $\mu\text{m}$  multi-mode fibers. Long-term stability of the cell shows a change less than 3 Torr over seven months. Recent improvements in the process using fiber tapering allow the use of standard 126  $\mu\text{m}$  diameter FC/PC connectors and improve optical transmission.

Funding provided by the Christopher Sorensen Incubator Fund, K-State Foundation, and AFOSR

**47. Intense-field photoionization of molecules using ultrashort radiation pulses of various wavelengths**

Joshua Beck and Cornelis J. Uiterwaal

We report on the design and construction of a variable intensity attenuator for broadband, ultrafast, intense laser pulses. With the use of broadband, achromatic waveplates and Brewster windows made from UV fused silica (UVFS), sufficient attenuation over a range from 465 nm to 1650 nm can be achieved. It is found that for UVFS, Brewster's angle varies by less than 25 arcminutes over the range of interest, allowing for minimal deviations in alignment as the wavelength is changed. This, along with the use of an OPA, allows us to investigate the nature of the ionization process (tunneling, MPI). We shall also investigate the role of electronic resonances in the ionization process, and for some molecules, possible contributions from lower lying orbitals (HOMO-1, HOMO-2, etc.) to the ion yield.

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**48. Modeling of Fast Photoemission from GaAs**

E.Brunkow, N.B.Clayburn, M.Becker, E.Jones, H.Batelaan, and T.J.Gay

When two intense 10-nJ pulses of 800-nm photons cause electron emission from GaAs, the number of electrons emitted depends on the delay between the two pulses. For delays between  $\sim 200$  fs and about one nanosecond, we find that the electron emission is sub-additive, i.e., fewer electrons are emitted for two incident pulses in combination than for the sum of two single-pulse emission events. We have modeled the multi-photon ionization of GaAs by these pulses and find good agreement with the sub-additive case when we assume that the multi-photon emission occurs through resonant population of the conduction band when the first photon is absorbed.

Work supported by the National Science Foundation (NSF) Grants PHY-1505794 and PHY-1306565, and NSF EPSCoR Grant IIA-1430519.

**49. Growth and Electron Transport in Ultralong, Gold Nanoribbons**

Gobind Basnet, Krishna R. Panta, Prem S. Thapa, and Bret N. Flanders

This paper describes the electrochemical growth of branchless gold nanoribbons with approximately 40×300 nm cross sections and >100 μm lengths (giving length-to-thickness aspect ratios of >10<sup>3</sup>). These structures are useful for opto-electronic studies and as nanoscale electrodes. The 0.75–1.0 V voltage amplitude range is optimal for branchless ribbon growth. Reduced amplitudes induce no growth, possibly due to reversible redox chemistry of gold at reduced amplitudes, whereas elevated amplitudes, or excess electrical noise, induce significant side-branching. The inter-relatedness of voltage-amplitude, noise, and side-branching in electrochemical nanoribbon growth is demonstrated. The electrical resistivity of these nano-ribbons is also investigated. Their thickness (~40 nm) is comparable to the mean free path of gold (~38 nm) and, hence, the near single crystalline ribbons behave as thin metallic films whose electrical and thermal transport properties are well-described by K Fuch's theory.

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