

# Attosecond Physics – From Generation of as Pulses to Applications on Solids

Reinhard Kienberger

MPQ-Garching, Germany

The generation of ever shorter pulses is a key to exploring the dynamic behavior of matter on ever shorter time scales. Attosecond XUV pulses together with the few-cycle (few-femtosecond) laser pulses used for their generation have opened the way to the development of a technique for attosecond sampling of electrons ejected from atoms or molecules or solids [1]. This is accomplished by probing electron emission with the oscillating electric field of the few-cycle laser pulse following excitation of the atom by the synchronized sub-femtosecond XUV pulse [2]. Recently, the dynamics of the photoionization process on solids has been studied [2]. Not only that attosecond metrology now enables clocking on surface dynamics, but also the individual behaviour of electrons of different type (core electrons vs. conduction band electrons) can be resolved. Here, we measured a time delay of about 100 as on the emission of the aforementioned two types of electrons. After a general overview on attosecond physics, recent experiments towards an absolute measurement of the travel time of electron inside solids and other measurements on solid surfaces are discussed.

---

[1] A. Cavalieri et al., Nature 2007, 449, 1029.

[2] R. Kienberger et al., Nature 2004, 427, 817.

# **Laser Induced Tunneling in Less Than 12 Attoseconds: Instantaneous or Invalid Concept?**

Ursula Keller

ETH Zurich, Switzerland

It is typically assumed that electrons can escape from atoms through tunneling when exposed to strong laser fields, but the timing of the process has been controversial, and far too rapid to probe in detail. We have used attosecond angular streaking to place an upper limit of 34 attoseconds and an intensity-averaged upper limit of 12 attoseconds on the tunneling delay time in strong field ionization of a helium atom in the non-adiabatic tunneling regime. This is the fastest process that has ever been measured. To achieve this we exploit the exact timing of a close to circular polarized intense laser field in the two-cycle regime. Our experimental results give a strong indication that there is no real tunneling delay time, which is also confirmed with numerical simulations using the time-dependent Schrödinger equation. We hope that our results will shed some light on the ongoing theoretical discussion and stimulate additional discussions on strong field ionization and tunneling time. Tunneling theories are the standard approach to intense-field ionization and have successfully described high harmonic generation (HHG), quantum path interference in QPI and laser-induced electron tu

# **Generation and Application of Attosecond Pulses for Time Resolved Investigations in Atoms and Molecules.**

Giuseppe Sansone

Dipartimento di Fisica Politecnico Milano, Italy

We present results on the generation of isolated attosecond pulses by shaping the time dependent ellipticity of a few-cycle carrier-envelope phase stabilized pulse. The control of the polarization state allows to steer the electron trajectories on the attosecond timescale, determining the numbers of recolliding electron wave packets down to the single recollision regime. Using this approach, isolated attosecond pulses have been generated in Xenon around 24 eV and used to create a complex electron wave packet in Helium with bound and continuum components that can be probed by using a synchronized infrared pulse.

The polarization gating technique can also be used to generate complex attosecond waveform such as double attosecond pulses, that could be implemented in novel attosecond spectroscopic techniques (such as transient absorption attosecond spectroscopy). We will present results indicating that the relative intensity and the relative phase of the two attosecond pulses can be separately influenced, by controlling the evolution in time of the polarization state of the driving pulse.

# Temporal Characterization of Isolated Attosecond Pulse Generated with Double Optical Gating<sup>1</sup>

Hiroki Mashiko, Steve Gilbertson, Ximao Feng, He Wang, Sabih D. Khan, Michael Chini, Shouyuan Chen, Chenxia Yun, Yi Wu and Zenghu Chang

JRM Laboratory, Kansas State University, Manhattan, KS, 66506-2604, USA

Double optical gating (DOG) was demonstrated for producing isolated attosecond pulses from multi-cycle driving lasers. It combines the two-color and the polarization gating. The generated supercontinuum spectrum extends from 28 eV to 620 eV including the “water window” region and supports single 16 as pulses, below one atomic unit of time (24 as). XUV pulses with 136 as duration were generated from 9 fs lasers and 260 as from 20 fs driving lasers in argon gas. The pulse shape and phase were characterized by FROG-CRAB based on attosecond streaking. Also, we have used the isolated attosecond pulses generated with DOG in several applications. Since DOG greatly reduces the requirements on the driving laser pulse duration, we found this technique is easy to implement and delivers daily reproducible attosecond pulses.

<sup>1</sup>This work is supported by the U. S. Army Research Office under Grant No. W911NF-07-1-0475, and by the Chemical Sciences, Geosciences, and Biosciences Division, U.S. Department of Energy.

# Generation of Sub-keV Harmonics and Isolated Attosecond Pulses by an IR-OPA Source

Katsumi Midorikawa

RIKEN, Japan

We have investigated the driving wavelength scaling of high harmonics and isolated attosecond pulse generation by a high-power infrared optical parametric source. First, we show the generation of a coherent water window x-ray radiation by extending the plateau region of high-order harmonics under a neutral-medium condition. The maximum harmonic photon energy attained are 300 eV and 450 eV in Ne and He, respectively. Our proposed generation scheme, combining a 1.6  $\mu\text{m}$  laser driver and a neutral Ne gas medium, is efficient and scalable in output yields of the water window x-ray. Thus, the precept of the design parameter for a single-shot live-cell imaging by contact microscopy is presented.

We also propose and demonstrate the generation of a continuum high harmonic spectrum by mixing the infrared optical parametric output and the fundamental laser fields. The observed spectra clearly show the possibility of generating isolated attosecond pulses from multi-cycle pump pulses.

# **Transient Absorption X-Ray Probing in the Attosecond Limit**

Stephen Leone

University of California, Berkeley and LBNL, USA

Ultrashort pulses of soft x-rays are produced by the process of high harmonic generation with a Ti:sapphire laser and used to probe atomic and molecular dynamics by pump-probe core level x-ray transient absorption spectroscopy. High field ionization of Xe atoms initiates an electron ejection process that results in orbital alignment of the resulting ion. High field ionization of molecules, such as dibromomethane and methyl iodide, produces dissociative ionization fragmentation, where Coulomb explosion and wave packet dynamics are observed by core level transitions in the soft x-ray regime. In the limit of very short driver pulses used to produce the high harmonic generation process, isolated attosecond pulses are generated and used to study ionization events. Molecular dissociative ionization pathways initiated by isolated attosecond pulses are manipulated with the high field of the few cycle driver pulses. In a collaboration with F. Krausz and E. Goulielmakis in Munich, attosecond transient absorption is demonstrated with isolated attosecond pulses in a study of high field sequential ionization of Kr atoms.

# Chemistry in Attoseconds: Ultrafast Hydrogen Migration in Hydrocarbon Molecules in Intense Laser Fields

Kaoru Yamanouchi

Department of Chemistry, School of Science, The University of Tokyo, Japan

In an ultrashort intense laser field, molecules exhibit a variety of characteristic dynamics. Among our recent experimental findings, ultrafast migration processes of hydrogen atoms within a polyatomic molecule are noteworthy [1]. The discovery of the ejection of triatomic hydrogen molecular ions,  $H_3^+$ , from methanol ( $CH_3OH$ ) was the beginning of the series of our studies on ultrafast hydrogen migration [1-4]. By introducing the coincidence momentum imaging method, we found for methanol [3] and allene ( $CH_2CCH_2$ ) [4] that the ultrafast hydrogen migration process occurs during the ultrashort laser pulse duration of 10~60 fs, indicating that hydrogen atoms move extremely rapidly. This can be regarded as an appearance of quantum mechanical nature of light-weighted hydrogen atoms. For probing this ultrafast spread of the distribution of hydrogen atoms within a molecule, it will be necessary to introduce attosecond pulses whose pulse duration is below 1 fs. The ultrafast hydrogen migration is now guiding us to the new experimental frontiers of attosecond chemistry as well as to the new theoretical frontiers beyond the BO approximation.

---

[1] Y. Furukawa, K. Hoshina, K. Yamanouchi, H. Nakano, *Chem. Phys. Lett.* **414**, 117-121 (2005).

[2] T. Okino, Y. Furukawa, P. Liu, T. Ichikawa, R. Itakura, K. Hoshina, K. Yamanouchi, H. Nakano, *Chem. Phys. Lett.* **419**, 223-227 (2006).

[3] T. Okino, Y. Furukawa, P. Liu, T. Ichikawa, R. Itakura, K. Hoshina, K. Yamanouchi, H. Nakano, *Chem. Phys. Lett.* **423**, 220-224 (2006).

[4] H. Xu, T. Okino, and K. Yamanouchi, *Chem. Phys. Lett.*, **469**, 255-260 (2009).

# Transform-limited Attosecond Pulse Generation

Chang Hee Nam<sup>1</sup>, Dong Hyuk Ko<sup>1</sup>, Juyun Park<sup>1</sup>, and Kyung Taec Kim<sup>2</sup>

<sup>1</sup>Dept. of Physics and Coherent X-ray Research Center, KAIST, Daejeon, Korea

<sup>2</sup>Advanced Photonics Research Institute, GIST, Gwangju, Korea

High harmonics can form an attosecond pulse train or an isolated single attosecond pulse. Due to intrinsic chirp structure the duration of high harmonic attosecond pulses is usually much longer than the transform-limited pulse duration. The temporal characterization of attosecond harmonic pulses by the RABITT (reconstruction of attosecond beating by interference of two-photon transition) or FROG (frequency-resolved optical gating) method also confirmed this chirped structure. We proposed the compensation of the attosecond chirp by material dispersion and showed that the positive attosecond chirp, contained in short-trajectory harmonics, can be compensated by passing the harmonic pulse through a medium with negative group delay dispersion (GDD). The experimental verification of this idea was first demonstrated using Al as the compensator by the Lund group. The application of the chirp compensation in the harmonic generation medium itself was also demonstrated by our group.

# Molecules and Attosecond Science

Paul Corkum

Joint Laboratory for Attosecond Science

University of Ottawa and the National Research Council of Canada

100 Sussex Drive

Ottawa, Canada K1A 0R6

During the past six years the minimum duration of optical (XUV) pulses has fallen from 5 femtoseconds ( $5 \times 10^{-15}$  sec) to about 100 attoseconds ( $\sim 10^{-16}$  sec)—less than the classical period of a ground-state electron in a hydrogen atom. Lasers drove this revolution by forcing electron wave packets to tunnel from the atom or molecules, move under the force of the time dependent electric field and then re-collide with their parent ions. From the ion's perspective, an attosecond electron wave packet re-collides. Attosecond XUV pulses are the byproduct of this collision.

The attosecond electron, controlled by light, is something unique in science. With wavelength  $\sim 0.5$ -3 Ångstrom, it allows us to measure spatial information -- as in a photograph. The “shutter speed” can be attoseconds.

Using  $N_2$ ,  $O_2$  and  $CO_2$  as examples, I illustrate three ways that the re-collision electron gives give us spatial images.

- **Molecular tunneling.** The lateral momentum distribution of electrons that tunnel measures a filtered projection of the momentum wave function of the ionizing orbital. Rotating the molecule, we gain all information about the orbital.
- **Laser Induced Electron Diffraction:** The re-collision electron elastically scatters from its parent molecular ion. The diffraction pattern gives the position of a molecule's atoms.
- **Orbital Tomography:** Attosecond pulses arise from the interference between the re-collision electron and the initial orbital from which it separated. The amplitude and phase of the XUV radiation contains all information to recreate the orbital image and to observe attosecond dynamics when multiple orbitals simultaneously ionize..

# Imaging orbitals from the attosecond emission of aligned molecules

Pacal Salieres

CEA-Saclay, (France)

A strong laser field interacting with molecules drives ultrafast intra-molecular electron wavepackets, resulting in the emission of attosecond XUV bursts. In linear molecules, the interaction of the laser-driven electron wavepacket with the core leads to quantum interferences in the recombination step. We characterized the attosecond emission from transiently-aligned CO<sub>2</sub> molecules and demonstrated that these interferences can be finely controlled by changing the electron recollision angle. Our control of the interference results in an attosecond pulse shaping [Boutu et al., Nature Physics 2008]. Moreover our measurements give direct access to the transition dipole matrix elements between the continuum states and the molecular orbitals involved in the emission process. When the HOMO gives the dominant contribution, a tomographic reconstruction is possible within a plane wave approximation for the recolliding electron. We performed a fully experimental reconstruction of the N<sub>2</sub> HOMO, where the polarization-resolved harmonic emission was characterized in amplitude and phase. This reconstruction exhibits Angström spatial resolution, mainly limited by the accessible spectral range.

# Probing Fixed-in-Space Molecular Structures With High Harmonic Generation<sup>1</sup>

Anh-Thu Le

Department of Physics, Kansas State University, Manhattan, KS 66506, USA

The recently developed quantitative rescattering theory (QRS) gives a simple and accurate description for high-order harmonic generation (HHG) from atoms or aligned molecules. According to the QRS, HHG spectra can be expressed as a product of a returning electron wave packet and the photo-recombination differential cross section of the *laser-free* continuum electron back to the initial bound state. This factorization opens possibilities for a relatively simple method for retrieval of the molecular frame photoelectron angular distributions (MFPAD) and the phase of the transition dipole with HHG from aligned molecules. Using the QRS combined with accurate photoionization transition dipole moments for fixed-in-space molecules obtained from state-of-the-art molecular photoionization calculations, we show that both the magnitude and phase of the high-order harmonics observed in recent experiments on aligned CO<sub>2</sub>, O<sub>2</sub> and N<sub>2</sub> can be quantitatively reproduced. Furthermore, we show that the contribution from multiple molecular orbitals, the polarization state and ellipticity of the harmonics can be all studied quantitatively for aligned molecules.

---

<sup>1</sup> This work was done in collaboration with C.D. Lin, R.R. Lucchese, T. Morishita, S. Tonzani and M.T. Lee and was supported by the US DOE.

# Optimising Fields for HHG

R.Torres, T.Siegel, L.Brugnera, F.Frank, L.Chipperfield, D.Hoffmann, J.W.G. Tisch and  
J.P.Marangos

Blackett Laboratory, Imperial College, London SW7 2AZ

We present recent work that goes beyond simply using single frequency fields at 800nm for HHG and instead try to approach more optimum conditions for HHG in various circumstances.

First we present recent experimental results investigating molecular HHG using 1300nm, where the longer wavelength has permitted us to study the harmonic spectrum over a larger photon energy range than possible at 800nm. Measurements on the spectrum and alignment dependence in CO<sub>2</sub>, N<sub>2</sub>, N<sub>2</sub>O and C<sub>2</sub>H<sub>2</sub> will be shown illustrating that new information is available by using longer wavelength fields. Next we will discuss the use of two colour fields of either commensurate (e.g. 1300nm/650nm) or incommensurate frequencies (e.g. 1300nm/800nm) to enhance HHG efficiency in atoms. In both case significant efficiency gains are observed through the use of two colour fields up to photon energies ~100eV. Finally we will discuss our recent theoretical treatment of the best waveform for HHG and present practical routes to approaching this ideal.

# Coherent frequency combs and spectroscopy – from far IR to XUV

Jun Ye

JILA, National Institute of Standards and Technology and University of Colorado,  
440 UCB, Boulder CO 80309-0440 USA

Phase-stabilized optical frequency combs can now maintain long-term ( $> 1$  s) temporal phase coherence across the entire visible spectrum. This unprecedented spectral and temporal precision of optical fields has profoundly changed optical frequency metrology and ultrafast science, with breakthrough developments in optical atomic clocks, optical frequency synthesis, united time-frequency spectroscopy, high-resolution quantum control, coherent pulse synthesis and amplification, and control of sub-femtosecond electron dynamics in atoms and molecules.

Beyond the visible spectrum, development of frequency combs in the far infrared opens the door to a wealth of fundamental molecular transitions, allowing direct frequency comb spectroscopy to simultaneously provide ultrahigh detection sensitivity, massively parallel monitoring channels, broad spectral coverage, and real-time and quantitative measurement capabilities.

The other exciting frontier naturally turns to the production of VUV and XUV frequency combs; laser-like sources with a high degree of temporal coherence in the vacuum and extreme ultraviolet spectral regions. This can be viewed as a development of the attosecond spectroscopy in the frequency domain. A strong motivation is to extend high precision laser spectroscopy and high-resolution quantum control techniques to VUV and XUV, which would allow measurement of many ground state transitions of atoms and molecules with revolutionary precision. These experiments might allow for some of the most stringent tests of quantum electrodynamics. Meanwhile, the production of VUV and XUV frequency combs provides an important platform to study extreme nonlinear dynamics with unprecedented precision. We will report the first detection of comb structure in high harmonic generations.

# **Octave-Spanning Ti:sapphire Lasers and Carrier-Envelope Phase Control**

Franz X. Kärtner

Department of Electrical Engineering and Computer Science  
and Research Laboratory of Electronics,  
Massachusetts Institute of Technology  
Cambridge, MA 02139

We discuss the progress made towards octave-spanning Ti:sapphire lasers generating carrier-envelope phase controlled sub-two cycle pulses directly from the laser. The physical origin of the carrier-envelope phase dynamics of these lasers as well as its limits on the carrier-envelope phase noise ultimately achievable will be discussed both by numerical modeling as well as experimental results.

# **Carrier Envelope Phase Stabilized Sub-5fs Laser for Attosecond Pulse Generation.**

Zhiyi Wei, Jiangfeng Zhu, Qiang Du, Hainian Han, Hao Teng, Wei Zhang and  
Chenxia Yun

Beijing National Laboratory for Condensed Matter Physics, Institute of Physics,  
Chinese Academy of Sciences, Beijing 100080, China

The generation of single attosecond laser pulses has opened up the new era for research the electron dynamics in attosecond time scale, in general, intense few-cycle optical pulses was used to drive the gas jet to generate high-order harmonic wave for attosecond laser, it has been well known that field reproducibility in each laser shot is predominant for isolated attosecond pulses generation. In this presentation, we report the carrier-envelope phase controlled 5fs laser at repetition rate of 1kHz through hollow-core capillary for spectral broadening, white-light continuum was first generated by injecting the 25 fs amplified pulses with pulse energy of 800  $\mu\text{J}$  into a hollow fiber filled with rare gas at high pressure. By dispersion compensation with a set of chirped mirrors, the shortest pulses are measured to be 4.6fs which is less than two optical cycles, and the pulse energy is up to 400  $\mu\text{J}$ . Following, we carry out carrier-envelope phase stabilization of the few-cycle optical pulses. The carrier-envelope offset frequency of the seed oscillator is fixed at a quarter of the repetition frequency of the oscillator, so every fourth pulse has the same carrier-envelope phase. The phase drift during the amplification stage is monitored by spectral interferometry between an octave spanning white-light continuum generated in a sapphire plate and its second harmonic. Carrier-envelope phase slip is extracted from the interference through Fourier transform and is stabilized by a phase lock loop. The jitter is found to be less than 53 mrad (root-mean-square, RMS). This laser system with carrier-envelope phase stabilized few-cycle pulses enables us to produce coherent soft X-ray emission and open the way to generate single attosecond pulses.

# Current Progress in XUV frequency combs

Akira Ozawa

Max Planck Institute of Quantum Mechanics, Germany

In the field of high precision spectroscopy, frequency combs have been an indispensable tool for phase coherently linking the frequency of laser with the radio frequency domain. This enables direct counting of optical frequencies. The measured transition frequency in simple atomic system like the hydrogen 1S-2S transition, allows for precise tests of bound state quantum electrodynamics (QED). So far, the applications of frequency combs have been limited to wavelengths longer than the visible region. This is due to limitations from available mode-locked lasers and possible frequency conversions. For precision spectroscopy of charged systems like  $\text{He}^+$ , which is more sensitive to higher-order QED corrections, a frequency comb will be required in extreme ultra-violet (XUV). In order to obtain a high power frequency comb in the XUV region, cavity assisted high harmonics generation is expected to be a promising tool. In this talk, our recent work and future plans towards high precision spectroscopy with XUV frequency combs will be presented. In addition, recent investigations on several possible techniques to further improve the output power of XUV frequency combs, such as Non-Collinear High Harmonics Generation (NCHHG) and high repetition rate phase-stable amplifiers, will be discussed.

# **Quantum Control with Shaped Pulses**

Yaron Silberberg

Weizmann Institute of Science, Rehovot, Israel

Shaping of femtosecond pulses has been the main tool in the field of quantum coherent control. In this talk I shall review the main achievements of this field over the last decade and the challenges it faces as it moves to intense fields and into attosecond interactions.

# Manipulating the Motion of Large Molecules: Translation, Rotation, and Conformer Selection

Jochen Küpper

Fritz-Haber-Institut der MPG, Berlin, Germany

Large molecules have complex potential-energy surfaces with many local minima. They exhibit multiple stereo-isomers, even at the very low temperatures of  $\sim 1$  K in a molecular beam. We have developed methods to manipulate the motion of large, complex molecules and to select their quantum states.<sup>1</sup> We have exploited this state-selectivity to spatially separate individual conformers (structural isomers) of complex molecules<sup>2</sup> and to demonstrate unprecedented degrees of laser alignment and mixed-field orientation of these molecules.<sup>3</sup>

Such clean, well-defined samples would strongly benefit or simply allow novel experiments with complex molecules, for instance, femto-second pump-probe measurements, X-ray or electron diffraction in the gas-phase, high-harmonic generation, or tomographic reconstructions of molecular orbitals. These samples would also be very advantageous for metrology applications, such as, for example, matter-wave interferometry or the search for electroweak interactions in chiral molecules. Moreover, these samples provide an extreme level of control for stereo-dynamically controlled reaction dynamics of complex molecules. In this presentation, I will describe and compare the manipulation methods employed and our respective results. In addition, I will discuss the prospects of imaging experiments using the upcoming X-ray free-electron lasers.

---

<sup>1</sup> Wohlfart et al. *Phys. Rev. A* **77**, 031404(R) (2008)

<sup>2</sup> Filsinger et al. *Phys. Rev. Lett.* **100**, 133003 (2008)

<sup>3</sup> Holmegaard et al. *Phys. Rev. Lett.* **102**, 023001 (2009);  
Filsinger et al. arXiv:physics 0903.5413

# Ultra-relativistic Electron Acceleration by a High-power Ultrafast Laser

Donald Umstadter

University of Nebraska, Lincoln, USA

Diocles, a recently commissioned laser system at the University of Nebraska, Lincoln (UNL) has demonstrated exceptional performance characteristics, including 30-fs pulse duration, 100-TW peak power, diffraction-limited focusing, and energy stability of only a few percent. In initial experiments, the laser was used as the driver for a wakefield accelerator, to accelerate a beam of electrons to energies up to 0.8 GeV, with angular divergence of  $< 2$ -mrad. In this case, a high-intensity laser pulse ( $> 10^{19}$  W/cm<sup>2</sup>) was relativistically self-guided through plasma a distance  $> 5$ mm, corresponding to over 8 Rayleigh ranges. When these electrons underwent betatron oscillations in the plasma channel, a beam of hard x-rays, with fs pulse duration, was also generated. We will discuss the laser's architecture and performance, as well as plans to upgrade it to the petawatt peak power level. Details of the experiments and a comparison of their results to theory and simulation will also be presented.

# Attosecond Pulse Trains: Measurement, Control and Applications

L'Huillier, Anne

Lund University, Sweden

This presentation will review the physics of attosecond pulse generation and some of its applications.

When atoms are exposed to intense laser radiation, electrons in the ground state may tunnel ionize, acquire energy from the field, and recombine, leading to the generation of attosecond pulses with broad bandwidth. When this process is repeated many times, the emitted radiation takes the form of a frequency comb, with peaks at odd harmonics of the laser field. The first part of this presentation will describe some of the attosecond tools that are being developed ranging from single attosecond pulses to pulse trains with one or two pulses per laser cycle and the techniques used to characterize them.

One of the most interesting properties of attosecond pulses is that their short pulse duration allows us to measure the phase of an unknown wave function or wave packet by pump-probe interferometric processes, giving us access to the temporal dynamics of the process that led to this

# Attosecond Time-resolved Electron Dynamics in Molecules

Marc Vrakking

FOM Institute for Atomic and Molecular Science (AMOLF)  
Kruislaan 407, 1098 SJ Amsterdam, The Netherlands

In the last few years, first experiments have been performed where attosecond laser pulses have been used to study time-resolved electron dynamics. So far, these experiments have been two-color XUV+IR experiments, where attosecond time-resolution was achieved by attosecond IR streaking (appropriate for continuum electron dynamics) and by use of the sub-cycle time-dependence of IR (multi-photon) ionization. We have recently introduced a new method that is particularly suitable for studies of attosecond dynamics in molecules, namely a measurement of an asymmetry in the ejection of charged fragments that is indicative of the localization of electrons in the molecule at the time of dissociation. The method was demonstrated in a pump-probe experiment on the H<sub>2</sub> molecule.

# **Control of Electron Dynamics in Light Systems Using Attosecond XUV and Femtosecond IR Radiation**

Predrag Ranitovic

Kansas State University, USA

Inducing a chemical process and controlling its outcome in real time is a central theme of attosecond science. Understanding the interaction of attosecond pulses with an atomic system fully accessible to the best available theoretical calculations is essential for the progress of this science. In this work we characterize some underlying processes behind the infrared-assisted ionization of helium by XUV radiation. We show that, if used in the form of an attosecond pulse train (APT) which has good resolution in both energy and time, processes are seen which could not be observed without high resolution in both domains simultaneously. We show that resonant absorption is important in the excitation of the helium and that small changes in the energies of the harmonics which comprise the APT can result in large changes in the ionization process. Good agreement between the data and theoretical calculations is found. With the help of this agreement, ionization pathways for the infrared-assisted ionization of helium by XUV attosecond pulses have been identified and simple model interpretations have been developed which should be of general applicability to more complex systems.

# **High Harmonics and Attosecond Pulses as Diagnostics of Intensity Spikes in Laser Filamentation.**

Mette Gaarde

Louisiana State University, USA

We show that the laser intensity in an ultrashort pulse filament can exceed the clamping intensity by more than a factor of three over several cm of propagation. We have performed fully non-adiabatic calculations of filamentation self-compression and the generation of coherent XUV radiation via HHG in argon at atmospheric pressure. We show that both the high-intensity laser pulse and the XUV radiation can be coupled out of the filament via a short pressure gradient, and that the XUV light emerges from the truncated filament as an isolated self-focused attosecond pulse with a peak intensity approaching  $10^{11}$  W/cm<sup>2</sup>. We discuss that the XUV yield presents an excellent diagnostic of the intensity spikes because of its extremely nonlinear dependence on peak intensity.

# Recent Advances in Ultra Intense Laser Plasma Interactions

David Neely

Central Laser Facility, Rutherford Appleton Laboratory, STFC, Didcot,  
Oxfordshire, OX11 0QX, UK  
University of Strathclyde, Glasgow, G4 0NG, Scotland  
Photo Medical Research Centre, JAEA, Kyoto, Japan

In the field of ultra intense laser plasma interactions there have been many significant advances over the last few years. These advances are intimately linked to the growth and development of the drive lasers in terms of power, repetition rate and pulse fidelity. With many systems worldwide achieving on target intensities of  $> 10^{20} \text{ Wcm}^{-2}$ , pulse contrast has been one of the areas in which many new techniques have been tested and their influence on the plasma interaction will be reviewed.

In the fields of laser driven electron and ion acceleration, as well as increasing the maximum particle energy, experimenters have been working towards demonstrating control and stability of the beams. First experiments have been conducted where the electron and ion beams have been used as secondary sources to enable new experimental areas to be investigated. Examples where the laser driven particle beam produce secondary sources will be discussed, such as, Photon generation with electrons and plasma heating with ions to investigate warm dense matter. Secondary beams have also been used as probe sources, allowing ultra short measurements of the electric fields inside plasmas and high contrast imaging of low  $Z$  targets such as the core of a laser driven fusion capsule.

As the accessible intensity on target increases, new areas of experimental investigation are opening up. First experiments exploring the radiation pressure driven acceleration regime and the potential for testing QED are underway and the near term opportunities in this area will be discussed.

# Continuum Attosecond Electron Wavepackets

Andre Staudte

National Research Council, Ottawa, Canada

Strong field ionization produces free electron wave packets of a few hundred attosecond duration. These continuum electron wave packets are at the heart of attoscience. Using coincidence momentum spectroscopy, i.e. COLTRIMS, we study the spatial and temporal structure of these wave packets. We present experimental results that show how a two color laser field can be tailored to steer the wave packets on the attosecond time scale. The relative phase between the two frequencies can be used to determine the emission time of the wave packet. It also separates the direct electrons from re-collision electrons by their phase dependent lateral momentum. This approach permits attosecond collision experiments using multi cycle laser pulses [1].

Finally, the electron provides information about the electronic structure of its origin and therefore could be used to image orbitals with attosecond resolution.

---

[1] M. Kitzler and M. Lezius, PRL 95, 253001 (2005)

# **Relativistic High-order Harmonic and Short Pulse Generation**

Karen Hatsagortsyan

Max-Planck-Institut für Kernphysik, Heidelberg, Germany

High-order harmonic generation (HHG) with relativistically strong laser pulses employing highly charged ions is a promising path towards coherent short wavelength radiation sources. The systems generating harmonics are highly charged ions and the medium in which HHG is realized is an underdense plasma. We discuss several means of increasing the efficiency of rescattering in the relativistic regime and obtaining a strong harmonic signal. This is achieved via assisting the infrared strong laser field with a weak attosecond pulse train of XUV photons, employing strong tailored laser pulses, counterpropagating circularly polarized and equal-handed laser pulses or counterpropagating attosecond pulse trains. In order to give evidence of a macroscopic harmonic yield after propagation through plasma, we investigate the conditions for rendering quasi-phase-matching of the harmonics. High-order harmonic emission in these setups are enhanced by several orders of magnitude in comparison to the case of a single laser wave. This way hard x-ray harmonics and extremely short pulses can be feasible.

# **High Power Filament Propagation in Atmospheric Quantum Wakes**

Howard Milchberg

University of Maryland, USA

The quantum coherent rotational response of air molecules excited through filamentation in the atmosphere is shown to greatly enhance or completely extinguish follow up filaments injected at specific time delays. As well, the optical energy of the follow up pulse can exhibit strong space and time localization.

# Brilliant FEL Light: New Frontiers in AMO Research

J. Ullrich

Max-Planck-Institut für Kernphysik, Heidelberg, Germany

Upcoming 4<sup>th</sup> generation light sources, Free Electron Lasers (FEL), will provide, for the first time, intensities, coherence properties, short-time and pump-probe options in the VUV to X-ray regimes comparable to those presently realized by intense, ultra-short laser pulses in the visible. At least three completely new fields of research are expected to open up in atomic and molecular physics. First, the huge integrated radiation flux enables to investigate in unprecedented detail dilute samples, as for example positive ions up to the highest charge states, negative atomic ions, negative or positive state-prepared molecular and size-selected cluster ions. Second, the tremendous peak intensities allow investigating, for the first time, fundamental non-linear processes where few photons interact with few electrons in atoms, molecules, clusters or ions. Third, the short-time properties will enable unique time dependent experiments with any of these targets and first femtosecond VUV-VUV pump-probe measurements have been demonstrated recently. In the talk, these novel fields will be highlighted and first results of pioneer experiments at the **Free Electron Laser at Hamburg (FLASH)** [1-6] as well as at the **Spring8 Compact SASE Source (SCSS)** in Japan will be discussed. Future possibilities opened e.g. by the integration of large area imaging photon CCD detectors into reaction microscopes (REMI) [7] or by providing ultra-cold targets via a magneto-optical trap (MOT) in a REMI, the streaking of electrons and ions by overlapping phase stabilized THz radiation etc. will be envisioned.

- 
- [1] R. Moshhammer, et al., Phys. Rev. Lett. **98** (2007) 203001
  - [2] S.W. Epp, et al., Phys. Rev. Lett. **98** (2007) 183001
  - [3] H.B. Pedersen, et al., Phys. Rev. Lett. **98** (2007) 223202
  - [4] A. Rudenko, et al., Phys. Rev. Lett. **101** (2008) 073003
  - [5] Y. H. Jiang, et al., Phys. Rev. Lett. **102** (2009) 123002
  - [6] M. Kurka et al., J. Phys. B (submitted)
  - [7] J. Ullrich et al., Prog. Rep. Phys. **66** (2003) 1463

# **Multiphoton Physics in the X-ray Domain**

Robin Santra

Argonne National Laboratory / University of Chicago, USA

The Linac Coherent Light Source (LCLS), the world's first x-ray free-electron laser, will become available for experiments in the fall of 2009. Assuming LCLS design parameters for photon energies near 1 keV and focusing on processes that are expected to dominate, calculations on the prototypical neon atom will be presented. Topics that will be discussed include valence stripping; double-core-hole formation; the resonant Auger effect; and the impact of SASE-FEL pulse structure. In addition, calculations on x-ray two-photon absorption by the organic molecule para-aminophenol will be presented.

# Pushing the Temporal Resolution at an XUV-FEL: Tagging, Streaking, Seeding

Markus Drescher

University of Hamburg, Germany

The new generation of x-ray sources based on large-scale linear accelerators provides users with intense ultrashort pulses at small wavelengths. In spite of unprecedented intensities, the applicability of these pulses for studies of ultrafast dynamics is hampered by temporal properties inferior to those of HHG sources. Although the bandwidth supports few-fs or even sub-fs pulses, the noise-based origin of the amplified pulses leads to a complex amplitude and phase evolution, strongly fluctuating from shot to shot. We adapt methods similarly to those applied in attosecond metrology in order to meet this challenge. An XUV/visible cross-correlator allows to determine the arrival time of individual XUV pulses with respect to visible pulses from an external laser system. Tagging of simultaneously acquired spectra then allows to compensate the XUV-laser jitter, thereby considerably improving the temporal resolution of dynamical studies. A strong THz field is used to realize an ac-field streak camera, sampling time information of individual pulses from the XUV-FEL FLASH. As a next step, XUV pulses from HHG will be used for seeding the SASE undulator at FLASH.

# Generation of Intense Few Cycle Laser Pulses for Driving Attosecond High Harmonic Emission

Ruxin Li, Yuxin Leng, Zhinan Zeng, Peng Liu, and Zhizhan Xu

State Key Laboratory of High Field Laser Physics, Shanghai Institute of Optics and Fine Mechanics, Chinese Academy of Sciences, P.O. Box 800-211, Shanghai 201800, China

We will report the recent progress at SIOM, in the field of generation of intense ultrafast laser pulses, and high order harmonic generation from atoms and molecules.

The high gain amplification in a large aperture Ti:sapphire crystal was achieved by developing a new scheme of parasitic lasing suppression, the 800nm laser output with peak power of 0.89PW and pulse width of 29fs was obtained. The generation of carrier envelope phase (CEP) stabilized intense infrared laser pulses by means of differential frequency generation and optical parametric amplification was demonstrated. Pumped by an 800nm Ti:sapphire laser, the CEP stabilized pulses are tunable from 1.2 $\mu$ m to 2.4 $\mu$ m, with the maximum output energy 1.2mJ in a 40fs pulse with the 6.8mJ pump energy. New scheme for measuring the CEP of ultrafast laser pulses was proposed.

High order harmonic generation towards attosecond XUV pulse emission in a precisely shaped laser field, either with two color or single color double driving laser pulses were intensively investigated, both theoretically and experimentally.

We investigated the high harmonic generation from aligned CO<sub>2</sub> molecules and demonstrated experimentally that the modulation inversion of harmonic yield with respect to the molecular alignment can be manipulated by tuning the intensity of driving laser pulse.

# **Coherent X-rays From Ultrafast Lasers, and Applications - Attosecond Science Meets Nonlinear Optics**

Henry C. Kapteyn

JILA and Department of Physics, University of Colorado and NIST, and NSF Engineering  
Research Center in Extreme-Ultraviolet Science and Technology  
Boulder, CO, 80309-0440, USA

The technology of bright coherent x-ray generation using tabletop-scale ultrafast lasers has opened up many new opportunities. To date, most experiments that use light from high harmonic generation have been limited to the EUV spectral region at ~50-100 eV. The grand challenge for extending bright HHG to higher energy is the development of new phase matching techniques for efficient HHG at high photon energies. The past two years have seen rapid progress in this area, essentially solving the high-harmonic phase matching problem. By employing mid-infrared driving pulses, bright phase-matched HHG should extend even into the hard x-ray region of the spectrum above 1 keV. Quasi phase matching techniques can also be implemented employing interference with counterpropagating light pulses. This coherent control process represents a useful manipulation of electronic wave function on attosecond time-scales. By combining these phase matching techniques, coherent tabletop x-ray sources with carefully engineered source properties will be possible. At the same time, the use of HHG sources to study dynamics in atomic, molecular, materials, and engineered systems has expanded rapidly. I will briefly discuss recent experiments in our group in observing molecular “radiation femtochemistry,” high-resolution imaging, ballistic heat transport in nanoscience, and ultrafast studies of magnetism dynamics.

# Phase-Resolved Attosecond Near-Threshold Photoionization of Molecular Nitrogen

Baptiste Fabre<sup>2</sup>, S. Haessler<sup>1</sup>, J. Higuët<sup>2</sup>, J. Caillat<sup>3</sup>, T. Ruchon<sup>1</sup>, P. Breger<sup>1</sup>, B. Carré<sup>1</sup>, E. Constant<sup>2</sup>, A. Maquet<sup>3</sup>, E. Mével<sup>2</sup>, P. Salières<sup>1</sup>, R. Taïeb<sup>3</sup> and Y. Mairesse<sup>2</sup>

<sup>1</sup>CEA-Saclay, IRAMIS, Service des Photons, Atomes et Molécules,  
91191 Gif-sur-Yvette, France

<sup>2</sup>CELIA, Université Bordeaux I, UMR 5107 (CNRS, Bordeaux 1, CEA), 351 Cours de la  
Libération, 33405 Talence Cedex, France

<sup>3</sup>UPMC, Université Paris 06, CNRS, UMR 7614, LCPMR, 11 rue Pierre et Marie Curie, 75231  
Paris Cedex 05, France

We photoionize nitrogen molecules with a train of extreme ultraviolet attosecond pulses together with a weak infrared field. We measure the phase of the two-color two-photon ionization transition (molecular phase) for different states of the ion. We observe a 0.9 shift for the electrons produced in the ionization channels leading to the  $X^2 \Sigma_g^+$ ;  $\nu = 1$  and  $\nu = 2$  states. We relate this phase shift to the presence of a complex resonance in the continuum. By providing both a high spectral and temporal resolution, this general approach gives access to the evolution of extremely short lived states, which is hardly accessible otherwise.

# Intense Laser-Atom Physics in the Long Wavelength Limit

L. F. DiMauro

The Ohio State University, Columbus, OH 43210 USA

Over the last fifteen years, the tailoring of a light field for manipulating the dynamics of a system at the quantum level has taken a prevalent role in modern atomic, molecular and optical physics. As first described by Keldysh [1], the ionization of an atom by an intense laser field will evolve depending upon the light characteristics and atomic binding energy. Numerous experiments have thoroughly investigated the dependence of the intensity and pulse duration on the ionization dynamics of inert gas atoms. However, exploration of the wavelength dependence has been mainly limited to wavelengths less than 1  $\mu\text{m}$ , or in the language of Keldysh to the multiphoton or mixed ionization regime. It is now technically possible to perform more thorough test, and perhaps exploit, the scaling laws at wavelengths greater than 1  $\mu\text{m}$ . In addition, excitation with mid-infrared light augments a variety of atomic systems which will tunnel ionize, as well as posing different model atomic structure, e.g. one- and two-electron like systems.

This talk will examine the implication of the strong-field scaling as it pertains to the production of high energy particles and the generation of attosecond pulses. We will interpret the intense laser-atom interaction using a semi-classical trajectory model.

---

1. L.V. Keldysh, Sov. Phys. JETP 20, 1945 (1964).

## **Ultrafast dynamics in solids and at interfaces**

*Martin Wolf*

*Freie Universität Berlin and Fritz-Haber-Institut der MPG, Berlin, Germany*

One of the basic questions in solid state physics is to understand why a material behaves like an insulator or metal. Systems with a half-filled band are usually expected to be metallic, however, may undergo a metal-to-insulator transition at low temperatures due to charge density wave (CDW) formation or pure electron correlations (Mott insulator). Electron correlations are also at the heart of solid state phenomena like superconductivity or the Kondo effect. The dynamics of the underlying elementary processes occurs on femto- to attosecond timescales, however, only few experiments provide direct access to the time evolution of the electronic structure. Similar timescales govern interface phenomena like charge transfer and relaxation of core excited adsorbates. In this talk I will present a brief overview on electron dynamics in solids and at interfaces and discuss some challenges for ultrafast spectroscopy and recent experiments in this field.

# Intense Attosecond Pulses

P. Tzallas<sup>1</sup>, E. Benis<sup>1</sup>, E. Skatzakis<sup>1,2</sup>, Y. Nimura<sup>3</sup>, G.D. Tsakiris<sup>3</sup> and  
D. Charalambidis<sup>1,2</sup>

<sup>1</sup>FORTH-IESL, PO Box 1527, GR711 10 Heraklion, Greece

<sup>2</sup>Department of Physics, Univ. of Crete, PO Box 2208, GR71003 Heraklion, Greece

<sup>3</sup>Max-Planck-Institut für Quantenoptik, D-85748 Garching, Germany

We report on recent results in the generation of high peak power XUV radiation, depicting sub-fs temporal confinement. The research is targeting XUV intensities, high enough to induce clearly observable non-linear XUV processes. Such processes are considered as pivotal for both the temporal characterization of attosecond pulses, as well as for the investigation of ultrafast dynamics in the XUV spectral region.

The main barrier in increasing the XUV peak power is the depletion of the generating medium, due to ionization saturation. In media suffering depletion, this barrier can be partially overcome in loose-focusing geometries, thus maintaining high driving energies, while intensities remain below saturation. This approach has led to the demonstration of a number of non-linear XUV processes and applications induced by attosecond pulse trains or individual harmonics [1-5].

In non-depleting media, there is in principle no limitation on the driving intensity. A highly promising such medium is the laser driven surface plasma. Recently, surface plasma harmonics in the spectral region 12eV-21eV have been measured to have an energy content of 40μJ at the source [6]. This energy is more than one order of magnitude higher than that of harmonics emitted by ionizing gaseous media. The superposition of these harmonics has been further successfully 2<sup>nd</sup>-order autocorrelated and a sub-fs temporal confinement in the pulse train has been deduced from the measured trace [6].

Current efforts are focusing on merging the “isolated pulse” and “intense pulse” characteristics. Towards this goal the highly promising method of the Interferometric Polarization Gating (IPG) has been recently successfully implemented [7]. By this technique, utilizing many cycle, high peak power driving laser pulses, a 40eV broad super continuum, extending down to 15nm, with an energy content of 20nJ has been measured [8]. Those are parameters that will enable the observation of a two-XUV-photon transition induced by an isolated attosecond pulse.

- 
1. N. A. Papadogiannis *et al.* *Phys. Rev. Lett.* **90**, 133902 (2003)
  2. E. Benis *et al.*, *Phys. Rev. A* **74**, 051402 (R) (2006)
  3. P. Tzallas *et al.*, *Nature* **426**, 267 (2003)
  4. L. A. A. Nikolopoulos *et al.*, *Phys. Rev. Lett.* **94**, 113905 (2005)
  5. O. Faucher *et al.*, *PRA Rap. Com.* (accepted)
  6. Y. Nimura *et al.*, *Nature Physics* **5**, 124 (2009), (see also <http://www.nature.com/nphys/journal/vaop/ncurrent/abs/nphys1155.html>)
  7. P. Tzallas *et al.*, *Nature Physics* **3**, 846 (2007)
  8. P. Tzallas *et al.*, *Opt. Lett.* (in press)

# Simultaneous Excitation of Equivalent Electrons to Non-equilibrium with Attosecond Pulses

Jan M. Rost

Max Planck Institute for the Physics of Complex Systems, Dresden, Germany

Multi photon absorption is a well understood phenomenon which, in most cases, refers to the absorption of many photons by one or two electrons over the duration of the laser pulse (typically 10-100 fs). Multiphoton absorption with attosecond pulses implies that the photons are absorbed within a time, too short for the relaxation of electron dynamics. Moreover, in a cluster equivalent electrons (one from each atom) absorb almost simultaneously one photon which constitutes a new light-matter coupling mechanism. It results for clusters in an excited electron dynamics, so far never activated optically.

Time permitting another -- quite surprising -- ultrafast multi-electron process will be introduced. It can be triggered by a few seed atoms implanted in a rare gas cluster when illuminated by a standard 800nm strong laser pulse with 50 fs duration. Primary ionization of the seed atoms (with ionization potential lower than those of the cluster) again creates a strong electric field gradient which removes very quickly (on a time scale of 1 fs) many electrons bound to atoms of the cluster.

We will explain the effect which occurs for both, linear and elliptic polarization of the laser.

# Two-Dimensional Fourier Transform Spectroscopy

Steven Cundiff

JILA, National Institute of Standards and Technology and University of Colorado,

Boulder, CO 80309-0440, USA

Two dimensional Fourier transform (2DFT) spectroscopy is a powerful tool for probing complex systems. 2DFT spectroscopy requires precise control of the phases of the incident pulses and measurement of the phase of emitted signal. By taking a Fourier transform, the signal phase can be correlated with the excitation phase, allowing information about coupling between resonances to be discerned. We are applying 2DFT spectroscopy to the exciton resonances in semiconductor nanostructures. The pulse ordering and how they are scanned determines the type of information gained. When the conjugated pulse arrives first, we can determine the homogeneous and inhomogeneous widths of the excitons and biexcitons as well as provide unique insight into many-body interactions. If the conjugated pulse arrives last, we probe two-quantum coherences, which cannot be directly observed in non Fourier transform measurements. Our results show that many-body interactions dominate the non linear optical response of excitons in semiconductors. This is true for both one-quantum, where it was evident from previous measurements, and two-quantum coherences, where it was not previously appreciated.