New frontiers in Atomic, Molecular and Cluster Physics and Chemistry

ICTP, Adriatico Guesthouse
Trieste, Italy
14–15 November 2011

www.elettra.trieste.it/vFAMC

Invited Speakers
Nora Berrah (USA)
Giovanni De Ninno (Italy/ Slovenia)
Reinhard Dörner (Germany)
Tim Laarmann (Germany)
Peter Lambropoulos (Greece)
Tobias Lau (Germany)
Michael Meyer (Germany)
Catalin Miron (France)
Thomas Möller (Germany)
M Novella Piancastelli (Sweden)
Robert Richter (Italy)
Jan-Michael Rost (Germany)
Jan-Erik Rubensson (Sweden)
Giuseppe Sansone (Italy)
Toshinori Suzuki (Japan)
Simone Techert (Germany)
Kiyoshi Ueda (Japan)
Joachim Ullrich (Germany

Organizing Committee

Carlo Callegari
Kevin Prince
Frank Stienkemeier
Thomas Moeller
Maria-Novella Piancastelli
Marcello Coreno
Lorenzo Avaldi
Stefano Stranges

Program

MONDAY 14.11

8:30 8:50 Registration
8:50 9:00 Welcome speech

Chair: William M. Fawley

1. Overviews: Science and machine global perspectives

9:00 9:35 Maria Novella Piancastelli
"*Recent advances in atomic and molecular physics at the new synchrotron and free-electron laser sources*"

9:35 10:10 Jan-Michael Rost
"*Massive parallel ionization through intense short wavelength light*"

10:10 10:45 Giovanni De Ninno
"*Principles and performance of VUV/X-ray light sources*"

10:45 11:05 *COFFEE BREAK*

2. FEL Facilities

Chair: Michael Meyer

11:05 11:40 Nora Berrah
""Probing Molecules from Within using Ultra-Intense and Ultra-Fast X-Rays from the LCLS FEL""

11:40 12:15 Joachim Ullrich
"*Ultrafast Dynamics: Pump-Probe Experiments at Free Electron Lasers*"

12:15 12:50 Kiyoshi Ueda
"*FEL experiments in Japan: from EUV to X-rays*"

12:50 14:05 *LUNCH*

3. Clusters, droplets, liquids, scattering

Chair: Paolo Piseri

14:05 14:40 Simone Techert
"*Studies of the dynamics of bulk chemistry with FEL radiation - state of the art and perspectives*"

14:40 15:15 Thomas Möller
"*Ultrafast processes and imaging of clusters*"

15:15 15:50 Tim Laarmann
“Coherent superposition of two rotational quantum states in the time domain: Microwave spectroscopy without microwaves”

15:50 16:25 Tobias Lau
"X-ray magnetic circular dichroism spectroscopy of size selected cluster ions: From spin coupling and orbital quenching to magnetic phase transitions"

16:25 16:45 *COFFEE BREAK*

4. High fields

Chair: Paolo Decleva

16:45 17:20 Peter Lambropoulous
"Multiple Ionization under Strong Short Wavelength Radiation"

17:20 17:55 Giuseppe Sansone
"Molecular attosecond dynamics"

17:55 18:30 Reinhard Dörner
"Multiparticle Coincidences Studies of Photoionization of Quantum Halos"

18:30 19:30 Poster Session

19:30 *DINNER*

TUESDAY 15.11

5. Advanced applications of Synchrotron Radiation

Chair: Maria Novella Piancastelli

9:00 9:35 Jan-Erik Rubensson
"Resonant Inelastic Soft X-ray Scattering Applied to Free Atoms and Molecules"

9:35 10:10 Catalin Miron
"Electron spectroscopy – a probe for fundamental properties of isolated species"

10:10 10:30 *COFFEE BREAK*

6. Pump-probe

Chair: Marcel Drabbels

10:30 11:05 Robert Richter
"Two-color experiments at the Gasphase beamline @ ELETTRA – CW and time-resolved studies"

11:05 11:40 Toshinori Suzuki
"*Time-Energy Mapping of Photoelectron Angular Distribution*"

11:40 12:15 Michael Meyer
"*Atomic photoionization in intense XUV and optical laser fields*"

12:15 Closing speech

VISIT TO Elettra/FERMI (Schedule to be determined)

Recent advances in atomic and molecular physics at the new synchrotron and free-electron laser sources

Maria Novella Piancastelli
Uppsala University, Sweden

Recent years have witnessed a tumultuous development in the field of electronic structure and dynamics of isolated atomic and molecular species, mainly stimulated by the advent of new and more powerful light sources. I will review some of the latest breakthroughs which have taken place at the most recently operational synchrotron and free electron laser sources, together with the new research prospects now foreseeable.

Massive parallel ionization through intense short wavelength light

Jan M Rost
Max Planck Institute for the Physics of Complex Systems, Dresden

We investigate multiphoton absorption by photons with frequencies in the XUV regime and higher.
Here, nonlinear photon absorption proceeds via many electrons absorbing one photon each which gives rise to a new kind of many electron dynamics in extended systems such as clusters and biomolecules. It does not occur with traditional intense infrared light, since there nonlinear photoabsorption typically couples many photons to only a few electrons.

Principles and performance of VUV/X-ray light sources

G. De Ninno
Sincrotrone Trieste and University of Nova Gorica

We review the working principle, the performance and the perspectives of light sources built in the aim of generating VUV/X-ray radiation with laser-like properties (high brilliance, sub-picosecond pulse duration, transverse and longitudinal coherence, good shot-to-shot reproducibility). We will in particular consider the case of synchrotrons and free-electron lasers, and carry out a comparison between free-electron lasers based on self-amplified spontaneous emission and on high-gain harmonic generation.

Probing Molecules from Within using Ultra-Intense and Ultra-Fast X-Rays from the LCLS FEL

Nora Berrah,
Physics Department, Western Michigan University

The response of molecular systems to the ultra-intense, femtosecond x-ray radiation from the hard x-ray FEL, the Linac Coherent Light Source (LCLS), was investigated. Sequential multiphoton ionization, frustrated absorption [1] and double core hole production mechanisms [2,3] will be presented.

*[1] M. Hoener et al., Phys. Rev. Lett. **104**, 253002 (2010)*
*[2] L. Fang et al., Phys. Rev. Lett **105**, 083005 (2010).*
[3] N. Berrah L. Fang, B. Murphy, T. Osipov, K. Ueda, E. Kukk, R. Feifel, P. van der Meulen, P. Salén, H. T. Schmidt, R. D. Thomas, M. Larsson, R. Richter, K. C. Prince, J. D. Bozek, C. Bostedt, S. Wada, M. Piancastelli, M. Tashiro, M. Ehara, PNAS, (in press 2011)

Ultrafast Dynamics: Pump-Probe Experiments at Free Electron Lasers

J. Ullrich
Department for Experimental Many-Particle Quantum Dynamics
Max-Planck Institute for Nuclear Physics, D-69117 Heidelberg, Germany

One of the most exciting opportunities opened by Free Electron Lasers (FEL) is the feasibility of performing, for the first time, pump-probe experiments in the VUV, EUV and X-ray wavelength regimes with femtosecond time resolution or even below. Here, a first light pulse (IR, Vis, EUV, EUV or X-ray) initiates dynamics, like a chemical reaction, a phase transition, spin-, orbital, or charge-density waves in solids and a second pulse, impinging at a variable but well-defined time delay, probes the motion.

In the talk, a first series of such experiments, performed at the VUV-FEL in Hamburg, FLASH, the SCSS test facility in Japan as well as pioneering measurements at the LCLS X-ray FEL will be presented. At FLASH and SCSS the VUV-pulse has been split by a back-reflecting mirror that is cut onto two halves. One of the pulses can then be delayed by moving the two half-mirrors with respect to each other reaching sub-femtosecond accuracy. In a demonstration experiment the vibrational wave-packet motion in deuterium molecular ions with a round-trip time of about 22 fs could be traced, indicating a time-resolution of better than 10 fs. Moreover, the isomerization time in VUV-excited acetylene evolving into vinylidene cations proceeding within about 50 fs was measured for the first time, ending a 20 years controversial debate. More recently, the dynamics of a suite of other charge migration reactions was investigated via time-resolved many-particle fragment detection in a reaction microscope (REMI) watching atoms move in real-time. Using the CAMP instrument at LCLS first optical pump – X-ray probe experiments have been performed on aligned molecules, clusters and biological nano-crystals highlighting the rich future potential of these methods envisioning tracing Ångstrom spatial changes at femtosecond time resolution.
At the same time we witness tremendous technological progress at FELs: Non-linear autocorrelation traces showed sharp peaks with a FWHM in the order of the coherence length of the radiation (4 fs at FLASH and 10 fs at SCSS), explained by the statistical nature and coherence properties of the FEL pulses and pointing towards exciting possibilities to perform attosecond X-ray – X-ray pump-probe experiments at the LCLS. In addition, new machine developments, like "slotted spoilers", "longitudinal space charge amplifiers" and high-harmonic generation or lasing in FEL-excited targets nurture the expectation to create attosecond pulses that might even be synchronized with an optical laser on a sub-femtosecond time-scale, which would open the door towards keV-attosecond science.

FEL experiments in Japan: from EUV to X-rays

Kiyoshi Ueda
IMRAM, Tohoku University, Sendai 980-8577, Japan

In 2008, the SPring-8 Compact SASE Source (SCSS) test accelerator, started operation in Japan. It provides linearly polarized EUVFEL pulses in the wavelength region of 51-61 nm. We have been investigating multi-photon multiple ionization and subsequent relaxation processes in atoms, molecules, and clusters irradiated by EUVFEL pulses, using ion and electron momentum spectroscopy and pump-probe techniques. In June 2011, SCSS XFEL, nicknamed "SACLA" lased and started commissioning. Our scientific program with SACLA is largely based on research problems which constitute a bridge between atoms and small molecules and more complex systems. We plan to study, for example, light-induced phase transition in clusters and light-induced structural change of photo-reactive bio-molecules, using time-resolved coherent X-ray imaging combined with Coulomb-explosion ion imaging and photoelectron diffractions. The talk will describe current status of EUVFEL experiments as well as plans and preparations for experiments with SACLA, as well as the first commissioning experiment with SACLA.

Ultrafast processes and imaging of clusters

Thomas Möller
Institut für Optik und Atomare Physik, Technische Universität Berlin

The understanding of the interaction of high intensity, short-wavelength, short-pulse radiation with matter is essential for virtually all experiments with new superintense X-ray sources, in particular for flash imaging of nm sized particles. Clusters as a form of matter intermediate between atoms and bulk solids are ideal samples to study fundamental light – matter interaction processes. They are finite systems with the density of bulk solids allowing the investigation of inner- and interatomic phenomena.1,2,3 Very recently, initial experiments have shown that in nm-sized gas phase particles can be imaged by single shot scattering.4 Upcoming X-ray lasers will allow improving the resolution and going to smaller particles. This will open new fields in cluster and nanometer-scale science. Ultrafast electron and ion dynamics can be studied with nm spatial resolution by means of time-resolved scattering using pump-probe techniques as well as time of flight spectroscopy. The talk will give an overview of recent results obtained at the FLASH facility in Hamburg and the LCLS in Stanford.

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2 H. Wabnitz, L. Bittner, R. Döhrmann et al., Nature **420**, 482 (2002).
3 C. Bostedt, H. Thomas, M. Hoener et al., Physical Review Letters **100** (13), 133401 (2008).
4 C. Bostedt, M. Adolph, E. Eremina et al., Journal of Physics B-Atomic Molecular and Optical Physics **43** (19), 194011 (2010).

Coherent superposition of two rotational quantum states in the time domain: Microwave spectroscopy without microwaves

T. Laarmann
Photon Science, Deutsches Elektronen Synchrotron (DESY), Germany

Femtosecond laser pulses in the near-infrared spectral range were used to excite a coherent superposition of two rotational quantum states of cold carbon monoxide in a non-resonant Raman process. The associated nuclear motion is followed in time by subsequent Coulomb explosion with soft X-ray FLASH pulses at high intensity in a pump-probe scheme. The coupling of J = 0 and J = 2 states results in an asymmetry of spatial fragmentation patterns detected parallel to the laser polarization axis. The observed wave packed oscillation prevails for at least 1 nanosecond covering more than 150 recurrences without dephasing. This observation can serve as a new route to disentangle complex rotational couplings and ultrafast decoherence phenomena that occur in complex systems and environments such as doped helium droplets in real time with a spectral and temporal resolution limited by the length of the delay scan and the femtosecond pulse duration, respectively.

X-ray magnetic circular dichroism spectroscopy of size selected cluster ions: From spin coupling and orbital quenching to magnetic phase transitions

Tobias Lau
Institut für Methoden und Instrumentierung der Synchrotronstrahlung, Helmholtz-Zentrum Berlin für Materialien und Energie GmbH

X-ray spectroscopy of size-selected atomic, molecular, and cluster ions is challenging because of ultralow target densities. A viable approach is to perform X-ray spectroscopy in ion trap experiments [1–5] with sensitivity in the femtomol or 10^{-6} monolayer equivalent range.

To study spin and orbital contributions to magnetic moments of size-selected ions via X-ray magnetic circular dichroism (XMCD) spectroscopy, we have upgraded our linear ion trap setup [1–5] with liquid helium cooling and a 5 T magnetic field. This setup allows us to explore magnetic coupling of model systems in the molecular limit, where we could demonstrate ferromagnetically coupled local high spin states in the archetypical bulk antiferromagnets chromium and manganese. Vice versa, we observed antiferromagnetic alignment of the central atom to the outer shell in iron, the archetypical bulk ferromagnet. In small iron clusters, we also followed the rapid quenching of the orbital magnetic moment [6].

[1] J. T. Lau *et al*., Phys. Rev. Lett. **101**, 153401 (2008).
[2] J. T. Lau *et al*., Phys. Rev. B **79**, 241102(R) (2009).
[3] J. T. Lau *et al*., Phys. Rev. A **79**, 053201 (2009).
[4] K. Hirsch *et al*., J. Phys. B: At. Mol. Opt. Phys. **42**, 154029 (2009).
[5] J. T. Lau *et al*., J. Chem. Phys. **134**, 041102 (2011).
[6] M.Niemeyer *et al*., submitted for publication

Multiple Ionization under Strong Short Wavelength Radiation

P. Lambropoulos
IESL-FORTH and Physics Dept. University of Crete, Greece

Strong, short wavelength radiation, of pulse duration in the range of few to tens of femtoseconds obtained in present day Free Electron Lasers (FEL) has ushered in a new era in laser- matter interactions. With photon energies ranging from the XUV to hard X-rays, various layers of new processes and phenomena are expected to emerge.
Although the hard X-ray machines have become operational only recently, in the XUV to soft X-ray range, a substantial body of experimental and theoretical work has already produced a variety of results, posing at the same time a number of questions for the future. Owing to the high intensity, the common feature of those processes is the non-linear dependence on the intensity of the radiation and the production of multiply ionized species, occasionally of quite high ionization stage. After a broad overview of the landscape of existing data and general understanding, I outline the underlying mechanisms and their dependence on the photon energy range, the present theoretical framework, as well as an attempt to identify crucial areas of experimental and theoretical investigations necessary for further progress.

Molecular attosecond dynamics

G. Sansone[1,2]

[1]*Dipartimento di Fisica Politecnico, Piazza Leonardo da Vinci 32 20133 Milano Italy*
[2]*Max Planck Institut für Kernphysik, Saupfercheckweg 1, D-69117 Heidelberg Germany*

The generation and characterization of single attosecond pulses have been achieved through several efforts in the field of ultrafast intense laser sources over the last 20 years [1] and through theoretical developments on the interaction of intense light pulses with atomic and molecular systems [2]. The duration of attosecond pulses is rapidly approaching the atomic unit of time [3] that represents, in the classical description of the atomic model, the natural time scale of the electronic motion; also in quantum mechanics the attosecond regime is the relevant time domain for electrons as the inverse of the energy spacing among electronic levels, (that determines the time constant for non-stationary states), lies typically in this range. First applications of such pulses have been mainly focused on simple atoms or molecules to validate new experimental approaches and to gain first information on electron-electron correlation.
Experimental and theoretical results on the ultrafast dynamics initiated by single attosecond pulses in helium and H_2 and D_2 will be shown.
In hydrogen several states of the neutral molecule (autoionizing states) or of the molecular ion ($1s\sigma_g$ and $2p\sigma_u$) can be accessed due to the large bandwidth of the attosecond pulses. The electron dynamics can be probed and controlled using a synchronized infrared few-cycle pulse [4].
New directions for the investigation of attosecond dynamics in more complex molecular systems will be discussed. In particular the experimental challenges related to the complete characterization of complex dynamics involving electron and nuclear degree of freedom will be analyzed.

References

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[2] M. B. Gaarde, J. L. Tate and K. J. Schafer, *J. Phys. B: At. Mol. Opt. Phys.* **41**, 132001 (2008).
[3] H. Mashiko *et al.*, *Opt. Lett.* **34**, 3337 (2009).
[4] G. Sansone *et al., Nature* **465**, 763 (2010).

Multiparticle Coincidences Studies of Photoionization of Quantum Halos

Reinhard Dörner
Frankfurt University, Germany

We will discuss recent experiment on the most "quantum" of all molcules, the Helium Dimer. This super dilute system shows an extreme internuclear distance of 52Angstram and is bound by only neV.
We show how a single photon can lead to double ionization of the two distant centers.

Resonant Inelastic Soft X-ray Scattering Applied to Free Atoms and Molecules

Jan-Erik Rubensson
Department of Physics and Astronomy, Uppsala University, Sweden

Resonant inelastic X-ray scattering (RIXS) reflects fine details in electronic structure and dynamics. The process is site specific on the atomic length scale (sub-nanometer) and time specific on the timescale for nuclear and electronic rearrangements (femto- to attoseconds). Consequently, RIXS spectroscopy has a tremendous potential in atomic and molecular, chemical and condensed matter physics. RIXS techniques have, however, suffered from the lack of adequate radiation sources. In practice this has limited the spectral quality and only a fraction of the inherent advantages have been exploited.

Here RIXS spectra of free molecules (O_2 and CO_2) with an energy resolution ($E/\Delta E \sim 10000$) that allows for separation of individual vibrational excitations [1] are presented. This opens a wealth of new possibilities, provides detailed information about ultrafast dynamics, and facilitates accurate mapping of the final state potential surfaces.

The measurements were made with the SAXES spectrometer [2] at the ADRESS beamline [3] at the Swiss Light Source of the Paul Scherrer Institut, using a gas/liquid cell with an ultrathin membrane. The data is discussed in terms of *ab-initio* multimode scattering calculations.

RIXS opportunities at soft X-ray free-electron-lasers are briefly discussed, with emphasis on non-linear processes such as multi-photon excitations, stimulated RIXS and four-wave mixing [4].

References

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[2] G. Ghiringhelli, et al., Rev. Sci. Instrum. 77, 113108 (2006).
[3] V. N. Strocov, et al., J. Synch. Rad. 17, 631 (2010).
[4] Y. Sun et al., PRA 81, 013812 (2010).

Electron spectroscopy – a probe for fundamental properties of isolated species

Catalin Miron
Synchrotron SOLEIL, l'Orme des Merisiers, France

Since many years, electron spectroscopy is an excellent analytical tool to characterize material's chemical composition or degree of oxidation [1]. When performed at ultra-high resolution inner-shell spectroscopies provide an accurate probe of ultrafast (fs) decay dynamics, in particular for isolated species [2]. Completely new scientific opportunities are being offered by the bright and highly monochromatic x-ray beams coupled to state-of-the-art instrumentation available at the newest facilities, such as the PLEIADES [3] beamline at SOLEIL (France), operated as a user facility since March 2010.

Selected examples will be shown from a panel where high-resolution spectroscopies have been employed to investigate fundamental properties of matter, such as the Vibrational Scattering Anisotropy (**VSA**) [4], the Auger-Doppler effect using circularly polarized light [5], or the rotational Doppler broadening of molecular electron spectra [6].

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[4] C. Miron *et al*., *Phys. Rev. Lett*. **105**, 093002 (2010).
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Two-color experiments at the Gasphase beamline @ ELETTRA – CW and time-resolved studies

Robert Richter
Sincrotrone Trieste S.c.p.A.

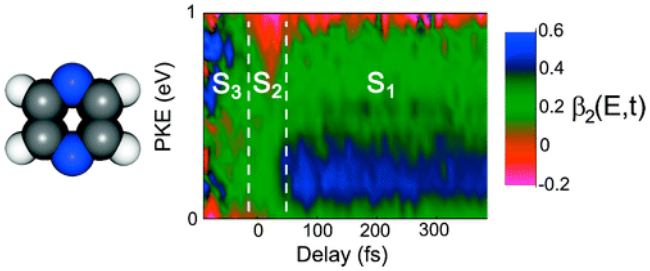
Time resolved pump-probe experiments allow the determination of the energy and lifetimes of excited states, the study of internal and intra-molecular energy distribution after a photo-absorption process or the exploration of vibrational levels out of the Franck-Condon region. In addition the interaction of laser radiation with atoms and molecules is known to change their electronic structure and give rise to new physical phenomena. The relative importance of various effects depends on the intensity of the laser field and ranges from harmonic generation, the AC Stark effect, above threshold ionization to electromagnetically induced transparency. The understanding of such effects is also important for the development of new techniques and tools in optics. If appropriate conditions can be found, laser coupling offers the possibility of shortening synchrotron light pulses (in principle) even into the fs region, without any modification to the machine. In a first approximation, the duration of the light pulse is expected to be equal to the duration of the laser pulse.

A mode-locked tunable Ti:Sapphire oscillator is synchronized with the time structure of the storage ring and can be used to study the photoionization dynamics. In multibunch operation of the ring the setup permits the direct observation of the dynamics from a few nanoseconds down to the 10's of picoseconds range. Experiments can be performed both in the frequency domain, taking advantage of the high energy resolution of the picosecond sources, and in the time domain. The characteristics of the setup are demonstrated by examining recent results on two-color ionization of noble gases and molecular nitrogen. Experiments on effects of laser-induced coupling between bound levels of noble gases on the length of the synchrotron radiation pulse will also be discussed.

Time-Energy Mapping of Photoelectron Angular Distribution

Toshinori Suzuki
Kyoto University: CREST JST: RIKEN

Time-resolved photoelectron imaging allows accurate measurements of photoelectron kinetic energy and angular distributions with light sources such as femtosecond lasers, a VUV free electron laser, and a conventional He(I) light source. In my presentation, I introduce some important technical and scientific advancement we made in the past decade, and I discuss in particular non-adiabatic dynamics of polyatomic molecules studied by time-energy mapping of photoelectron angular distribution with 22 fs time-resolution.



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Atomic photoionization in intense XUV and optical laser fields

Michael Meyer
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The combination of intense femtosecond X-ray and NIR pulses produced by Free Electron Lasers (FEL) and synchronized optical lasers, respectively, offers various new possibilities to investigate the dynamics of atomic photoionization. Some recent results obtained at the XUV-FEL FLASH in Hamburg and the first X-ray FEL, the LCLS in Stanford, will be presented.

In the experiments at FLASH, the strong dressing field ($>10^{12}$ W/cm^2) produced by the optical laser, give rise to the so-called two-color Above Threshold Ionization (ATI), which could be studied for the first time in a regime free from unwanted interference effects [1]. In addition, the NIR field can modify also the dynamics of the resonant Auger decay. For the resonant Kr 3d-5p excitation at 91.2 eV, we have investigated by electron spectroscopy the laser-induced shift of the resonance position and have observed competition between resonant and direct Auger decay caused by the ionizing of the excited 5p electron in the NIR field.

Recent experiments at LCLS have taken advantage of the very short (2-5 fs) pulse durations delivered by this FEL. The duration coincides with the lifetime of the Ne 1s core hole state and with the temporal width of one optical cycle of the 800 nm radiation from the NIR dressing laser. The analysis of the angle-resolved KLL Auger decay in atomic reveals strong interference effects, which result from the coherent emission of electrons produced during one cycle of the superimposed optical field. The experimental results are in excellent agreement with recent theoretical work [2].

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