



1936-47

Advanced School on Synchrotron and Free Electron Laser Sources and their Multidisciplinary Applications

7 - 25 April 2008

Two-colour and photon correlation spectroscopy

Fulvio Parmigiani University of Trieste & Sincrotrone Trieste



The VUV, XUV, and soft x-ray regions



Main parameters to be considered in radiation-matter interaction

- e.m field magnitude (linear processes vs.non-linear)
- time structure of the e.m. field (dynamics)
- transversal and longitudinal coherence (very high resolution spectroscopy and scattering)
- state of polarization (dichroic effects)



Hot Dense Matter occurs in:

- Supernova, stellar interiors, accretion disks
- Plasma devices: laser produced plasmas, Z-pinches
- Directly driven inertial fusion plasma
- Warm Dense Matter occurs in:
 - Cores of large planets
 - Systems that start solid and end as a plasma
 - X-ray driven inertial fusion implosion

Hydrogen phase diagram



R.W. Lee, LLNL

1. Boiling the Vacuum with Lasers

Spontaneous pair creation from vacuum, induced by an external field, was first proposed in the context of e⁺e⁻ pair creation in static, spatially uniform electric field [Sauter (1931); Heisenberg, Euler (1936); Schwinger (1951); ...]

One of the most intriguing non-linear phenomena in quantum field theory

- Theoretically important: beyond perturbation theory
- Eventual experimental observation: probes theory in domain of very strong fields
- Mechanism applied to many problems in contemporary physics:
 - Quantum evaporation of black holes [Hawking (1975); Damour, Ruffini (1976); . . .]
 - e^+e^- creation in vicinity of charged black holes
 - Particle production in early universe
 - Particle production in hadronic collisions [Casher, Neuberger, Nussinov (1979); ...]

A. Ringwald/DESY

[Parker (1969); . . .]

[Damour, Ruffini '75; . . .]

Time domain scale:

ps (10⁻¹² s) *collective processes (ordering) in condensed matter* (*dynamics of the magnetic* moment...)

fs (10⁻¹⁵ s) *phase transitions and VB electron dymanics*

as (10⁻¹⁸ s) *electronic dynamics in atoms and molecules*



Time scales







from the electron to the macroscopic transformation.

Depending on the system, different times-scales exist



Klaus Sokolowski-Tinten

Institut für Experimentelle Physik

Measuring Atomic Displacements (I): *Transient Debey-Waller Effect*







Simone Techert, Max-Planck Institut für biophysikalische Chemie, Abt. Spektroskopie und Photochemische Kinetik – Strukturdynamik (bio)chemischer Systeme (Göttingen)

Femtosecond Electron Diffraction Setup



J. Cao, et al., Appl. Phys. Lett. 83, 1044 (2003)

Structural Changes Probed with Diffraction



Three aspects of Bragg peak (position, intensity and width) give detailed knowledge of structure change.

Reproduced from: A. Rousse, et al., Rev. Modern Phys., 73, 17(2001)

Strongly-driven thermal melting (superheating)

3.5 ps melting time at laser fluence 70 mJ/cm² 3.5 ps > 1.5 ps e-ph thermalization (~ 1500K) predicted by TTM simulation



B. J. Siwick, et al., Science 302, 1382 (2003)

Temporal Evolution of Bragg Peaks



Coherent and in-phase motions
Amplitude is proportional the laser fluence
8 ps period and damping time constant~ 20 ps

Short pulses radiation sources

- Table-top lasers
- Free electron lasers
- Recirculating LINAC/FELs
- LINAC based sources
- Electron bunch slicing in 3rd generation synchrotrons
- Plasma sources



Zholents and Zolotorev, Phys. Rev. Lett., 76, 916,(1996).

Schoenlein et al., Science, 287, (2000)

- Coherent EUV light is generated by ionizing a gas with a fs laser
- Broad range of harmonics generated 4.5 up to 550 eV
- "Laser-like" coherent beams in EUV
 - R. Bartels et al, Science 297, 376 (2002), Nature 406, 164 (2000)













J. Zhou et al, PRL 76(5), 752-755 (1996)



Phase-matched frequency conversion in waveguides:





- Waveguide creates plane-wave geometry
- Waveguide can control the phase velocity $(v_p = \omega/k)$

$$k = \frac{2\pi}{\lambda} \left(1 + \frac{P\delta}{\lambda}(\lambda) - \frac{1}{2} \left[\frac{u\lambda}{2\pi a} \right]^2 - \frac{1}{2} \frac{N_e r_e \lambda^2}{\pi} \right)$$

vacuum gas waveguide ionization



- Repeat 200 year old experiment Young's Double Slit
 - Young, Philos. Trans. R. Soc. XCII 12, 387 (1802).
 - E. Wolf et al., JOSA 46, 895 (1957; Opt. Lett. 6, 168 (1981).



- R. Bartels et al, Science 297, 376 (2002)
- R. Bartels et al, Opt. Lett. 27, 707 (2002)





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Matias Bargheer, N. Zhavoronkov, Y. Gritsai, M. Wörner, T. Elsaesser

K_{α} line radiation:

	K_{α} energy/ eV	K_{α} flux / 4π *s
Ga	9.2	$4.5 ext{x} 10^{10}$
Cu	8.2	3.9x10 ¹⁰
Ni	7.5	6.7x10 ¹⁰

- Plasma generated in 15 μm copper
- X-ray focus 15 µm
- Estimated pulse duration: 200 fs

Brilliance = 4 x 10⁷ / (mrad²*mm²s*0.1%BW)

According to specs	Flux on sample / s	Focus / µm
Multilayer mirror (Osmic)	$1.4 x 10^{6}$	30
HOPG reflector (IfG)	5x10 ⁷	200



Klaus Sokolowski-Tinten

Institut für Experimentelle Physik







Spontaneous Undulator Radiation



 $\frac{\Delta \lambda}{\lambda_s} = \frac{1}{N_w}$

Amplification of External Wave



At resonance, while traversing one period of the undulator, the electron falls one radiation wavelength behind the EM-wave


















	Units	SASE 1
Wavelength*	Å	1-5
Peak power	GW	37
Average power	W	210
Photon beam size (FWHM)**	$\mu\mathrm{m}$	100
Photon beam divergence (FWHM)***	μ rad	0.8
Bandwidth (FWHM)	%	0.08
Coherence time	\mathbf{fs}	0.3
Pulse duration (FWHM)	fs	100
Min. pulse separation****	ns	93
Max. number of pulses per train****	Ħ	11500
Repetition rate****	Hz	5
Number of photons per pulse	#	$1.8 imes10^{12}$
Average flux of photons	#/sec	$1.0 imes10^{17}$
Peak brilliance	B^{*****}	$8.7 imes10^{33}$
Average brilliance	B^{*****}	$4.9 imes10^{25}$

*Parameters are given for the shortest wavelength.

** Value at the exit of the undulator.

*** Far field divergence.

**** Values determined by the time structure of the electron beam in the accelerator. The average parameters for the SASE-1 FEL are given for the ultimate case when only this beamline is in operation. ***** In units of photons/(sec $\cdot mrad^2 \cdot mm^2 \cdot 0.1\%$ bandwidth).







Spectrum of the BESSY SASE-FEL for a lasing photon energy of $\hbar \omega = 1 \text{ keV}$ compared to the BESSY II performance. BESSY II: single bunch operation with 10 mA of average beam current at E = 1.7 GeV. BESSY SASE-FEL: I = 5 kA, E = 2.25 GeV and a planar undulator with $\lambda_u = 2.75$ cm and N = 1450 periods. Spectra are calculated with a transmission efficiency of the monochromator of 5%.







Top: Basic scheme of a two-stage FEL [16] providing full longitudinal and transverse coherent light, see text for details. Bottom: GENESIS simulation of the two-stage FEL employing a 3 kW seed in the second undulator.



First Ultraviolet High-Gain Harmonic-Generation Free-Electron Laser

L. H. Yu,* L. DiMauro, A. Doyuran, W. S. Graves,[†] E. D. Johnson, R. Heese, S. Krinsky, H. Loos, J. B. Murphy, G. Rakowsky, J. Rose, T. Shaftan, B. Sheehy, J. Skaritka, X. J. Wang, and Z. Wu National Synchrotron Light Source, Brookhaven National Laboratory, Upton, New York 11973, USA

(Received 25 March 2003; published 14 August 2003)

We report the first experimental results on a high-gain harmonic-generation (HGHG) free-electron laser (FEL) operating in the ultraviolet. An 800 nm seed from a Ti:sapphire laser has been used to produce saturated amplified radiation at the 266 nm third harmonic. The results confirm the predictions for HGHG FEL operation: stable central wavelength, narrow bandwidth, and small pulse-energy fluctuation.







Peak Brightness Comparison



Brilliance — Microscopy

- X-ray microscopy
- X-ray dichroic microscopy

Coherence — **X-ray scattering**

- X-ray elastic scattering (statistical optics)
- X-ray FT interferometry

Time structure — Pump-Probe

- Time resolved core level spectroscopy
- Time resolve spectroscopy zooming in the momentum space
- TR X-ray diffraction
- Dynamics of the phase transitions

Time structure

Pump - Probe setup



$$U_e(\tau) \propto \int_{-\infty}^{\infty} I_X(t) I_{IR}(t-\tau) dt$$

Mach Zehnder type interferometer







F.Parmigiani et al.



 $H_{int} = -\frac{e}{2mc} (\mathbf{A} \cdot \mathbf{p} + \mathbf{p} \cdot \mathbf{A}) = -\frac{e}{mc} \mathbf{A} \cdot \mathbf{p}$







Time-Resolved Photoemission





Hot electron mediated surface charge transfer process on 100-500fs timescales

 Perturbation Theory gives Fermi's Golden Rule for transition probability

$$w = \frac{2\pi}{\hbar} \left| \left\langle \Psi_f \left| H_{\text{int}} \right| \Psi_i \right\rangle \right|^2 \delta(E_f - E_i - \hbar \omega)$$

For dipole allowed transitions,

$$H_{\rm int} = \frac{e}{mc} \mathbf{A} \cdot \mathbf{p}$$

Transmission absorption measurements



C.T.Chen et.al, PRL 75, 152 (1995).



FIG. 5. Normalized transmission absorption spectra through a 32-nm Fe film at normal incidence with $\mathbf{E} \perp \mathbf{M}$ and $\mathbf{E} \parallel \mathbf{M}$ are offset by 1 in β_{\perp} and β_{\parallel} , respectively. $\beta_{\perp} - \beta_{\parallel}$, multiplied by 10 and offset by 3 gives the MLD (Voigt effect).

Kortright & Kim, PRB 62, 12216 (2000).

X-ray Fluorescence, Gd 2p3d and 2p 4d





FIG. 3. Comparison of the calculated 2p3d x-ray emission spectral shape (solid line), with the $2p_{3/2}3d$ experimental results (points). The experimental curve is aligned and normalized to 1.0 at the peak position, with respect to the calculation. The theoretical MCD spectrum is given in the scale of the emission spectrum, and the experimental MCD has been normalized to it.

FIG. 4. Comparison of the calculated 2p4d x-ray emission spectral shape (dashed), with the $2p_{32}4d$ experimental results (points). The experimental curve is aligned and normalized to 1.0 at the peak position, with respect to the calculation. The solid line is the theoretical result with a reduction of the Slater integrals to 70% and a broadening as described in the text. The theoretical MCD spectrum is given in the scale of the emission spectrum, and the experimental MCD has been normalized to it.

de Groot et.al, PRB 56, 7625 (1997).

Microscopy by MXLD -MXCD: AF-FM interface coupling



F. Nolting et al., Nature 405, 767 (2000)

Some preliminary considerations

• Type of experiments

Pump-probe

Single pulse

• Energy resolution / momentum resolution:

 $\Delta E \ge h/4\pi$

 $\Delta \underline{\mathbf{p}} \ge h/4\pi$





Kinematic relations

$$\begin{split} k_{out} &= \sqrt{\frac{2m}{\hbar^2}} E_{kin} \\ k_{in} &= \sqrt{\frac{2m}{\hbar^2}} (E_{kin} + V_0) \\ k_{out,\parallel} &= k_{in,\parallel} \equiv k_{\parallel} \end{split}$$

"Snell's Law"

$$k_{\parallel} = \sin \theta_{out} \sqrt{\frac{2m}{\hbar^2} E_{kin}} = \sin \theta_{in} \sqrt{\frac{2m}{\hbar^2} (E_{kin} + V_0)}$$

Critical angle for emission

$$(\sin\theta_{out})_{\max} = \sqrt{\frac{E_{kin}}{E_{kin} + V_0}}$$





(started 2006)

Objectives

Scope of the project is to design and built an angle resolved and time resolved photoemission system with a spin detector.



Band Mapping and Fermi Contours





Damascelli, Hussain, and Shen: REVIEWS OF MODERN PHYSICS, VOLUME 75, APRIL 2003



FERMI Surface dynamics

In two recent papers has been shown that it is possible the control of the electronic phase of a manganite by mode-selective vibrational excitation [M. Rini et al., Nature **449**, 72 - 74 (2007)] and to measure the <u>ultrafast electron relaxation in superconducting $Bi_2Sr_2CaCu_2O_{8+\delta}$ by time-resolved photoelectron spectroscopy</u> [L. Perfetti et. al.,Phys. Rev. Lett. **99**, 197001 (2007)]. Our aim is to study the breathing of the Fermi surface in K₃C₆₀ (Fig 4) and similar system by exciting the structure through selective mode vibrations [Science, **300**, 2003].

 $K_{3}C_{60}$





Spin states dynamics

Objective

The goal of this project is to measure the spin resolved band dispersion and spin dynamics in solid by harmonics and high harmonics generated from a 250 kHz Ti:SA amplified source





Collaboration with 4GLS (E. Seddon and C. Cacho)



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Electron detector
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Mott polarimeter

Test of the SR-TR ToF on the Au(111) surface states



Osterwalder et al. PRB 69 pp.241401R, 2004)

aelettra



Comparison between the 1kHz and 250kHz laser system



Ultrafast coherent imaging at Fermi

Spokesperson: H. Chapman (LLNL-CA), J. Haidu (Stanford University and Uppsala University)

First EUV-FEL experiments show that structural information can be obtained before destruction



SynCro'073-7 September 2007 - Rijeka-Croatia

Ultrafast coherent imaging at Fermi

Spokesperson: H. Chapman (LLNL-CA), J. Haidu (Stanford University and Uppsala University)



H.N. Chapman et al.; Nat. Phys. 2, 839 (2006) and Physics Today, Jan 2007, pag. 19



Fig. 1. Schematic of the diffraction camera for FLASH, which uses a multilayer-coated mirror to reflect the on pattern onto a CCD. (a) The configuration shown is for the measurement of diffraction from silicon nurice test objects. (b) Plots of the reflectivity of the mirror on axis, as a function of photon energy, for mirrors made for the first harmonic (32 nm) and second harmonic (16 nm). (c) Schematic for forward scattering, and (d) schematic for pump-probe imaging where the beam is reflected back onto the sample with a normal-incidence multilayer mirror. A foil filter (green) and light shield (beige) are also shown.

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SynCro'073-7 September 2007 -Rijeka-Croatia

Defining the first experiments with FERMI@Elettra

CELLS INJECTED INTO THE FEL BEAM: 1st EXPERIMENTS ON LIVE PICOPLANKTON

J.Hajdu and H. Chapman

Procedure:

- 1. Take cells from solution
- 2. Introduce them into the beam
- 3. Hit them with the XFEL pulse and record diffraction pattern

Challenges:

- 1. Particle concentration
- 2. Keeping cells alive and in the "native" state
- 3. Diagnostics: How do we know if a hit was good?

SynCro'073-7 September 2007 -Rijeka-Croatia

FIRST FLASH DIFFRACTION IMAGE OF A LIVE PICOPLANKTON

March 2007 FLASH soft X-ray laser Hamburg, Germany

FLASH pulse length: 10 fs Wavelength: 13.5 nm

Thanks

J.Hajdu and H. Chapman





Filipe Maia, Uppsala

J. Hajdu, I. Andersson, F. Maia, M. Bogan, H. Chapman, and the imaging collaboration



FLASH-DIFFRACTIVE IMAGING - EXPECTED PRIMARY IMPACT

NON-CRYSTALLINE STRUCTURES IN THE MICROMETRE TO NANOMETRE SIZE DOMAIN

2D STRUCTURES FOR NON-REPRODUCIBLE OBJECTS

3D STRUCTURES FOR REPRODUCIBLE OBJECTS

4D STRUCTURES IN ATTOSECOND - FEMTOSECOND TIME-RESOLVED EXPERIMENTS

J.Hajdu and H. Chapman

Ultrafast processes and imaging of gas phase clusters and nanoparticles

Spokespersons: **T. Möller, C. Bostedt (TU-Berlin)** Co-proponents: P. Milani, University of MilanoJ. Hajdu, University of Stanford and University of Uppsala H.N. Chapman, LLNL, Livermore


Ultrafast processes and imaging of gas phase clusters and nanoparticles

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Clusters stay intact during exposure (30 fs), $\Delta R < 3 \text{\AA}$

February 2006

Coherent scattering on FePd magnetic stripes



Available Instruments to measure Collective Excitations



Transient Grating Spectroscopy at the Nanoscale

Liquids - Fluids

- Transition from the Hydrodynamic to the Kinetic regime in Simple liquids and fluids.
- Effect of the Local Structure on the Relaxation Dynamics of Molecular liquids and H-bonded liquids.
- Sound **Bifurcation** in Gas Mixtures
- Thermal Relaxation Dynamics in Liquid Metals.

Glasses

- Relaxational Processes in Super-Cooled liquids and their relation to the Glass Transition.
- Vibrational and Relaxational Low Temperature Properties of Fragile and Strong glasses.
- Characteristic Length of the Disorder

Surface Dynamics

- Transverse Dynamics in Liquids.
- Phase Transition in thin Films.

Polymers

- Structural Relaxations.
- Shear and Density Fluctuations.

Cluster and nanoparticle spectroscopy



Spokespersons: **F. Stienkemeier, B. von Issendorff** (Univ. of Freiburg-D) Co-proponents :K.Fauth (MPI- Stuttgart, D), M. Drabbels (EPFL- CH), M. Schmidt(CNRS –Orsay, Fr), U.Buck (MPI-Goettingen, D)





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Photo-induced and Magnetic Field-Dependent X-Ray Emission in Magnanites

Photo-induced and magnetic field-induced insulator-to-metal transition

Photo-induced isothermal magneto-structural phase transition and magnetoresistance

For H>5 Tesla, T< 30 K

OuickTime™ and a

Vol 449 6 September 2007 doi:10.1038/nature06119

LETTERS

nature

Control of the electronic phase of a manganite by mode-selective vibrational excitation

Matteo Rini¹, Ra'anan Tobey², Nicky Dean², Jiro Itatani^{1,3}, Yasuhide Tomioka⁴, Yoshinori Tokura^{4,5}, Robert W. Schoenlein¹ & Andrea Cavalleri^{2,6}

QuickTime[™] and a TIFF (LZW) decompressor are needed to see this picture. D Aoki *et al.* 2001 <u>Coexistence of</u> <u>superconductivity and ferromagnetism in</u> <u>URhGe</u>, *Nature* **413** 613-616

T R Kirkpatrick et al. 2001 <u>Strong enhancement</u> of superconducting *T*c in ferromagnetic phases *Phys. Rev. Lett.* **87** 127003

C Pfleiderer *et al.* 2001 <u>Coexistence of</u> <u>superconductivity and ferromagnetism in d band</u> <u>metal ZrZn2</u> *Nature* **412** 58-61

H vs T phase diagram for (H_2O) Cobaltites. Open and closed circles represent T_c and magnetic ordering Temperature



Superconductivity and Ferromagnetism

A different class?

Coexisting superconductivity and ferromagnetism due to itinerant electrons is unusual, but even among them URhGe stands out. Its surprising behaviour could help reveal the underlying physics of ferromagnetic superconductors.

Gilbert G. Lonzarich

is in the Quantum Matter Group, Department of Physics, Cavendish Laboratory, J J Thomson Avenue, Cambridge CB3 0HE, UK. e-mail: gl238@hermes.cam.ac.uk



Figure 1 Schematic phase diagrams of the novel ferromagnetic superconductors URhGe, UGe_2 and UIr, close to first-order quantum end points. **a**, For URhGe, the solid line marks the magnetic transition between two spin-polarized states in which the magnetization **M** is aligned in different directions in the orthorhombic unit cell. The easy axis of magnetization is the *c* axis. The magnetic transition is second order (continuous) above the tricritical point and first order below the tricritical point. The value of the upper critical field along the *a* axis seems to diverge as the component of the magnetic field along the *b* axis is tuned towards the first-order quantum end point. This unprecedented behaviour is the key finding of Levy and colleagues¹. **b**. The solid line in the temperature–pressure phase diagram marks a transition from a ferromagnetic state to a spin-unpolarized state (paramagnetic) in UGe₂ or UIr (metamagnetic transition lines are not shown). It is interesting that in all three itherant-electron ferromagnets superconductivity is observed only in the spin-polarized state and electron pairing is therefore expected to be in spin-triplet *p*-wave states⁵.





Joint Programme for Higher Education in Physics

University of Trieste - ICTP

The ICTP cooperates with the Faculty of Sciences of the University of Trieste in the education in physics of graduate students from Developing countries.

The cooperation concerns the "Laurea Magistralis in Fisica"

The "Laurea Magistralis in Fisica" can be compared to an advanced master in physics in the Anglo-Saxon system or to the last two years of the New European Educational System as fixed by the Bologna-Sorbonne agreement among the European Ministers of Education in the mid nineties.

ICTP announces that in 2008 a few scholarships and travel grants will be awarded to qualified, selected students from Developing countries in order to register and follow the training track in Condensed Matter Physics of the "Laurea Magistralis in Fisica" of the University of Trieste.

Each scholarship consists of: 800 Euro/month, travel grant, insurance and tuition fees.

The Programme covers two academic years

Courses are held in English

The minimum qualification for applicants is a degree equivalent to a M.Sc. (or an exceptionally good B.Sc.)

The selection of candidates will be based on their university record and academic recommendations.

More information at the web-page: http://physics.units.it/didattica03/ICTPuniv.php