Coherent x-ray sources and applications

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About myself

About myself

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- 1. Goal: Unveiling the dynamics of dense matter
- 2. High brightness coherent sources
- X-ray Free Electron Lasers
- High Harmonic Generation
- 3. Advances in ultrafast imaging and dynamics
- High resolution imaging: Holography
- High resolution imaging: CDI
- Warm dense matter: Pump and probe

Betatron radiation > Félicie Plasma-based X-ray lasers > old me M. Fajardo - Sesimbra CAS - 2019

21st century challenge: Capturing the dynamics of matter

Warm Dense Matter , between cold solid and classical plasma, is difficult to model

- Giant planets
- White dwarfs
- Inertial Fusion

Physical limit: why do we need X-rays (0)

Physical limit: why do we need X-rays (1)

Physical limit: why do we need X-rays (2)

Timescales of laser pulses and related phenomena

(age of the Universe $\approx 0.43 \times 10^{18}$ s)

Physical limit: why do we need
 coherent X-rays (3)
 $\int_{\mathbb{R}^{rd} A^{th}}$ Gen Synchrotrons: 100's ns **coherent** X-rays (3)

10¹² X-ray photons are needed to take this picture

A revolution in structural biology

Neutze, Nature 406, 752 (2000)

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A revolution in structural biology

Siebert et al, Nature, February 2011

Single-crystal nanocrystal structure determination Single-shot Mimivirus et al, Nature, February 2011

The ideal X-ray source

Hard wavelength Ultra-short Coherent Bright

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The 21st century has seen the birth of X-ray Free Electron Lasers

FLASH, XUV (DESY, Germany): 2003 LCLS (SLAC, USA): 2009 FERMI(ELETTRA, Italy): 2011 SACLA (Japan): 2012 XFEL (DESY): 2017

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Experiments performed in large scale facilities

Map of scientific collaborations from 2005 to 2009 . Reaucheuse @ Science-Metrix, Inc

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Our team has been working on development of brighter X-ray lasers

Table-top Ultra **Intense Xuv Sources**

We proposed using HHG to seed an XRL, overcoming noise

Zeitoun et al, Nature 2004 : first demo of seeded X-ray laser E Oliva, M Fajardo et al, Nature Photonics 2012: proposal for CPA with plasma amplifier Zeitoun, Fajardo & Lambert, Nature Photonics 2010: seeded FELs

廿 $+ \times$ s

FERMI: a seeded Free electron laser

- Bunch of electrons accelerated
- Modulation in energy along the bunch
- Energy modulation is translated into electron density modulation
- Forming of micro bunching and coherent emission in undulator

Seeded FEL pulse duration

In standard seeded FEL operation the FEL pulse duration and seed pulse duration are correlated by

$$
\Delta t_{FEL} = n^{-1/3} \Delta t_{seed}
$$

Using a chirped seed increases the FEL bandwidth.

$$
\Delta t_{FEL}^{CPA} = n^{-2/3} \Delta t_{seed}^{FT}
$$

- XUV compressor for achieving a FT limited FEL pulse
- **Implantation of chirped-pulse amplification at an FEL**
- Experiment to demonstrate the two different regimes of operating an FEL with XUV compressor

Free electron laser - Results

FERMI
@elettra

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ARTICLE

Received 17 Sep 2013 | Accepted 4 Mar 2014 | Published 30 Apr 2014

DOI: 10.1038/ncomms4539

Generation of 10^{20} W cm $^{-2}$ hard X-ray laser pulses with two-stage reflective focusing system

Hidekazu Mimura^{1,*}, Hirokatsu Yumoto^{2,*}, Satoshi Matsuyama^{3,*}, Takahisa Koyama², Kensuke Tono², Yuichi Inubushi⁴, Tadashi Togashi², Takahiro Sato⁴, Jangwoo Kim³, Ryosuke Fukui³, Yasuhisa Sano³, Makina Yabashi⁴, Haruhiko Ohashi^{2,4}, Tetsuya Ishikawa⁴ & Kazuto Yamauchi³

> Nanofocusing of X-ray free-electron laser using wavefront-corrected multilayer focusing mirrors

> S. Matsuyama⁸², T. Inoue, J. Yamada, J. Kim, H. Yumoto, Y. Inubushi, T. Osaka, I. Inoue, T. Koyama, K. Tono, H. Ohashi, M. Yabashi, T. Ishikawa & K. Yamauchi

Scientific Reports 8, Article number: 17440 (2018) | Download Citation \pm

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A tabletop source of bright coherent X-rays: High Harmonic Generation

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Perturbative Nonlinear Optics Perturbative Nonlinear Optics

$$
P = \chi^{(1)} E + \chi^{(2)} E^2 + \chi^{(3)} E^3 + ...
$$

•Accurately treated by treating the polarization as a power series in E.

•With sufficiently intense laser fields, the higher order terms give rise to Fourier components of the polarization at harmonics of the laser frequency, creating radiation at harmonics of the laser frequency.

Regimes of nonlinear optics

Brabec and Krausz, Review of Modern Physics 2000

High-Harmonic Generation

Anne L'Huillier, Philippe Balcou M. Fajardo - Sesimbra CAS - 2019

High-Harmonic Generation

single atom response

Semi-classical model: 3-step-model

HHG

single atom response

HHG

single atom response

Quantum model

Solve time-depended Schrödinger equation of an electron initially bound to an electron

$$
i\frac{d}{dt}\ket{\psi}=H\ket{\psi}-E(t)x
$$

With the Hamiltonian

$$
H=-\frac{1}{2}\nabla+V(\vec{r}),
$$

Wave-function is superposition between ground state and free electron

 $|\psi(t)\rangle = a(t) |0\rangle + |\varphi(t)\rangle$

Ion and electron act as an dipole and dipole moment is

 $\vec{d}(t) = \langle \psi(t) | \vec{x} | \psi(t) \rangle$

Source of radiation is dipole acceleration FT of dipole acceleration is HHG spectrum

Phase-matching

sources of mismatch

Dispersion of neutral gas **Dispersion of plasma**

Gouy-phase **Atomic phase** Atomic phase

initial phase of HHs depends on intensity $\Delta k_{at}(z) = \alpha_q \frac{8I_0 z}{z_r^2 \left(1 + \frac{4z^2}{z^2}\right)^2}$

Attosecond (10-18s) Pulses

Shortest Pulse measured is ~ 67 attoseconds

Few-cycle laser pulses are only a few optical periods (~µm) long

"Attosecond oscilloscope": First direct visualization of the electric field of visible light (Science, 2004)

Current records

Shortest pulse duration: 67 as Krausz group: (few fs laser) Shortest wavelength: 7Å (1.6 keV) Murnane group: $(3.9 \,\mu m \text{ laser})$ Highest photon count*: 1 μ J at 45eV Kim group: (w/2w) 1 μ J at 100eV Krausz group: (f=18m, 80mJ, 5fs) $!150 \mu J$ at FORTH - Charalambidis

**That's 109 photons/pulse, compared to 3 mJ XFEL at 1012 photons/pulse*

High Harmonic Generation: A tabletop source

Characteristics

high spatial coherence, high brightness, ultra-short pulses duration, table-top, high-repetition rate

Atomic response / typical spectrum

The VOXEL Station at IST

November 2016 December 2016

Manipulating Harmonic Properties

Novel focusing optics with gas density

We have been pursuing XUV adaptive optics for a long time

G Lambert, New Journal Physics 2009: two colour fields

G Lambert, EPL, 89 (2010) 24001: wavefront measurements

2 color generation: polarization does not follow the simplistic δ^3 model

 \cap

 -450

150

 Ω

 -150

 -100

 -50

 $\mathbf 0$

50

100

 -100

 -50

 Ω

50

100

150

Control over Harmonic Properties

In collaboration with LOA: generation of HHG with circular polarization

O. Kfir, et al, Generation of bright phase-matched circularly[polarized extreme ultraviolet high harmonics,](http://www.nature.com/nphoton/journal/v9/n2/full/nphoton.2014.293.html) *Nature Photonics* 9, 99–105 (2015)

loa

Control over polarization and ellipticity

G. Lambert et al, Nature Communications 2015

01 MAY 2015 VOL 348, ISSUE 6234

This wavefront sensor has been used routinely to improve the WF of HHG

Using closed-loop correction, we achieved diffraction limited harmonics ($\langle \lambda/14$ rms)

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Wavefront aberrations

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VOXEL station experiment July 2018

Wavefront aberrations

Wavefront aberrations

The future is bright for HHG

Saule et al, Nature Communications 2019

[High-flux ultrafast extreme-ultraviolet photoemission spectroscopy at 18.4 MHz p](https://www.nature.com/articles/s41467-019-08367-y)ulse repetition rate

OAM is being explored as well (see work by Fabien Queré's group)

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Gabor Holography (in-line)

 $|U_{\rm O}+U_{\rm R}|^2=U_{\rm O}U_{\rm R}^*+|U_{\rm R}|^2+|U_{\rm O}|^2+U_{\rm O}^*U_{\rm R}$

We did 3D "microscopy" in the XUV

Gabor inline digital holography

Achieved resolution: 800 nm in 2D, 140x magnification Depth of field: limited by Numerical Aperture: >100µm *A. S. Morlens et al, Opt. Letters 2006*

Aberrations at the source degrade the image Cleaning up reconstruction

 $C_2^2 = 0.5$.

 $C_2^2 = -5.$

 $C_2^2 = 2$.

Jji

Fourier Holography gives better resolution

Benefits: increased numerical aperture

 $d \sim \lambda/NA$

Resolution limit \sim size of pinhole for reference wave

Drawbacks:

- No depth it's 2D
- Balancing the two waves

Applications: Holography

Multicolor / Attosecond holography

• Demonstrating FTH with several discrete wavelengths HHG pulses can produce several discrete HH orders or a continuous XUV spectrum with attosecond duration

Ultra-fast dynamic processes can be imaged

Broad bandwidth or several discrete wavelengths requires careful target design.

Goals / Experimental setup

Applications: Holography - Results

Published: Fourier transform holography with high harmonic spectra for attosecond imaging applications." Optics Letters 40.13, 3205-3208 (2015)

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Lenslens imaging 1: Crystallography

X-ray crystallography uses the known relationship between the far-field and the object

A plane wave incident on an object is scattered. Scattered waves interfere

$$
F(\mathbf{k}) = \int_{-\infty}^{\infty} f(\mathbf{x}) \exp(2\pi i \mathbf{k} \cdot \mathbf{x}) d\mathbf{x}
$$

Bragg peaks 28 Nobel prizes using this technique

Lenslens imaging 2: Coherent Diffraction Imaging

The Fourier Transform encodes the object

But one can only measure the Intensity: the phase is lost

Solution: a Phase Guessing algorithm

Review: Coherent lensless X-ray imaging [Henry N. Chapm](http://www.nature.com/nphoton/journal/v4/n12/full/nphoton.2010.240.html%23auth-1)an [& Keith A. Nug](http://www.nature.com/nphoton/journal/v4/n12/full/nphoton.2010.240.html%23auth-2)ent Nature Photonics 4, 833–839 (2010)

Lenslens imaging 2: Coherent Diffraction Imaging

Sample: 50 nm gold spheres

Reconstruction: Full wave

Formalism **The solution**

The solution is at the intersection of two constraints spaces

Formalism **Iterative algorithm**

Recent demonstrations

PHOTONS, ATOMES ET MOLÉCULES

Coherent diffractive imaging with ultrafast coherent soft X-ray sources

Soft X-ray Free Electron Laser Chapman et al., Nature Phys. 2006

Soft X-ray High harmonic source Sandberg et al., PRL 2007, PNAS 2008

Þ **Hour acquisition time:100000 shots!**

LETTERS REVIEW

week ending 31 AUGUST 2007

order that can result in narrower bandwidths at shorter wavelengths have recently been demonstrated [23-25]. Such improvements will extend the ultimate resolution to tens of nm. As the laser repetition rates are increased from 3 kHz to tens of kHz, the soft-x-ray flux will be simultaneously increased, and acquisition time will be dramatically reduced from hours to minutes. Also, as computing

SERVICE DES PHOTONS, ATOMES ET MOLÉCULES

Coherent diffraction imaging

We have used HHG to make single-shot diffraction imaging

Coherent diffraction in the XUV achieved 62 nm, 20 fs resolution

ET MOLÉCULES

Laser energy: 35 mJ 5.5 m focal length lens 10 cm, 2 mbar argon gas cell

 $0:6 \mu J$ in XUV 500 µrad divergence, 92% coherence.

A. Ravasio et al, Phys. Rev. Letters 2009

Experimental Setup @ CEA- Saclay

Experimental Data

Computed phase stereo lensless X-ray imaging

Computed phase stereo lensless X-ray imaging : synchrotron data

Novel developments – promising perspectives for tabletop coherent imaging

Optical Coherence Tomography

Silvio Fuchs, Martin Wünsche, Jan Nathanael, Johann J. Abel, Christian Rödel, Julius Biedermann, Julius Reinhard, Uwe Hübner, Gerhard G. Paulus, "Optical coherence tomography with nanoscale axial resolution using a laser-driven high-harmonic source," Optica **4,** 903-906 (2017);

[https://www.osapublishing.org/optica/abstract.cfm?uri=optica-](%20%20https:/www.osapublishing.org/optica/abstract.cfm?uri=optica-4-8-903)4-8-903

Novel developments – promising perspectives for tabletop coherent imaging

X-ray Ptychography Review: F Pfeiffer, Nature 2018

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Conclusions and perspectives

What are the material and transport properties of Warm Dense Matter

First studies

Isochoric heating, isentropic release Aluminum and Aluminum A. Levy, et al, Phys. Plasmas 22, 030703 (2015) Time and space resolved interferometry of freely expanding Ag thin foil irradiated by the LCLS Front - experiment Х, ф **Back experiment** Front - Hydro code (ESTHER) Back - Hydro code (ESTHER) 18 16 Electron temperature (eV) Confirmation of 14 scenario 12 Hydrodynamic 10 codes describe recorded expansion 4.5 10¹⁵ 5 10¹⁵ 5.5 10¹⁵ 6 10¹⁵ 6.5 10¹⁵ 72

X-ray Irradiance (W/cm²)

Pumping solid density plasmas with XFELs

Transient absorption of Warm Dense Aluminum

- Al thin foil
- Heated to 6 eV at solid density
- Fermi energy 13 eV

The answer from QMD

DFT calculations using VASP: Density of states, electron occupation and transition probabilities

Self emission spectrum shows signatures of degeneracy

VOXEL Station a platform for WDM studies

WDM studies at VOXEL

IR case

- provides valuable data on ultrafast transition to plasma
- can DFT get it right?

XFEL case

- energy transfer from core electrons to outer electron system: delay?
- electron relaxation timescales in HED plasmas? 78

XUV KB grazing optics for pump probe studies

9

Gareth Williams

Optical properties DFT calculations Solid titanium

Conclusion

High brightness coherent sources:

- X-ray Free Electron Lasers Record intensities in X-rays due to extreme brightness
- High Harmonic Generation Record short pulse duration, tabletop Advances in ultrafast imaging and dynamics
- High resolution imaging: Holography & CDI: Unprecedented spatial and temporal resolution
- Warm dense matter: Pump and probe starting to unveil dynamics

XUV team@GOLP

Thomas Wodzinski

Marta Fajardo* Gareth Williams

... EN FAIT, NON, C'EST PAS VRAI INFINI. IL NE S'ARRETE PAS COM AU BOUT D'UN MÊTRE

DE PWS, UN FAISCEAU LASER EST INVISIBLE. SAUF S'IL RENCONTRI OBSTACLE (FUMÉE, POUSSIÈRE...)

Mukhtar Hussain | Patricia Estrela | Filipa Ribeiro

2019 Trainees José Figueiredo Fernando Lima Robin Sureau

Joana Duarte

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MINISTÉRIO DA CIÊNCIA, TECNOLOGIA E ENSINO SUPERIOR

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