



Elettra Sincrotrone Trieste



# On the science with seeded FELs

***Fulvio Parmigiani***

# THE QUEST FOR SEEDED FELs

**GUEST EDITORIAL**

Seeded Free-Electron Lasers and Free-Electron Laser Applications...

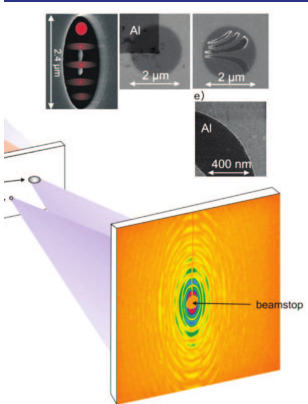
*by Fulvio Parmigiani and Daniel Ratner*

**PHYSICS TODAY 2008**

**Fleming and Ratner**

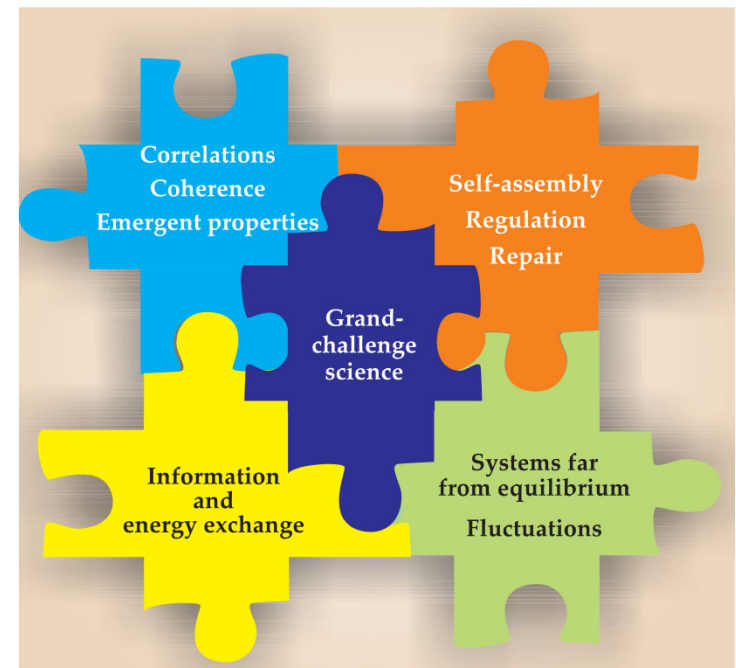
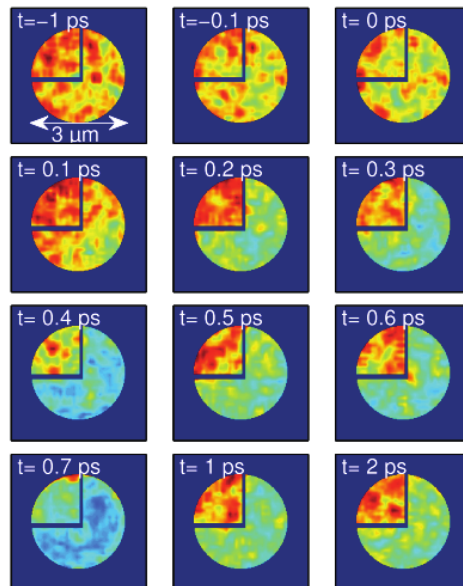
# SRN

*Synchrotron Radiation News*  
May/June 2016 • Vol. 29, No. 3



## Seeded Free-Electron Lasers

and  
Free-Electron  
Laser Applications



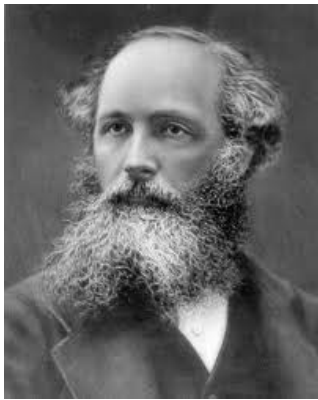
08940886 (2016) 29 (3)

# PHOTON SCIENCE WITH FELs

| Photon Science section/subsection         |                                                                        | FEL property referred to in the text |                 |                         |                      |              |                       |                        |                       |                |                       |               |               |
|-------------------------------------------|------------------------------------------------------------------------|--------------------------------------|-----------------|-------------------------|----------------------|--------------|-----------------------|------------------------|-----------------------|----------------|-----------------------|---------------|---------------|
| 6.1 Structural Biology                    | 6.1.1 Protein Structure and Dynamics                                   | ●                                    | ●               | ●                       | ●                    |              |                       | ●                      |                       |                |                       | ●             | ●             |
|                                           | 6.1.2 Live Cell Studies                                                | ●                                    | ●               |                         | ●                    | ●            | ●                     |                        |                       |                |                       |               |               |
|                                           | 6.1.3 Heterogeneous, Non-Crystalline Cell Organelle                    |                                      | ●               |                         | ●                    | ●            |                       |                        |                       |                |                       |               |               |
|                                           | 6.1.4 Viruses                                                          | ●                                    | ●               |                         | ●                    | ●            |                       |                        |                       |                |                       |               |               |
|                                           | 6.1.5 Looking to the Future                                            |                                      |                 | ●                       |                      |              |                       |                        |                       |                | ●                     |               |               |
| 6.2 Atomic, Molecular and Cluster Physics | 6.2.1 Atoms                                                            | ●                                    | ●               |                         |                      | ●            | ●                     | ●                      | ●                     | ●              |                       |               |               |
|                                           | 6.2.2 Molecules                                                        | ●                                    | ●               | ●                       |                      | ●            | ●                     | ●                      |                       | ●              |                       | ●             |               |
|                                           | 6.2.3 Clusters                                                         | ●                                    | ●               |                         | ●                    | ●            | ●                     |                        |                       |                |                       |               |               |
|                                           | 6.2.4 Future Prospects                                                 | ●                                    | ●               | ●                       | ●                    | ●            |                       | ●                      | ●                     |                | ●                     |               | ●             |
| 6.3 Photochemistry                        | 6.3.1 Molecular Photochemistry                                         | ●                                    | ●               | ●                       |                      | ●            | ●                     |                        |                       |                |                       |               |               |
|                                           | 6.3.2 Surface Photochemistry                                           | ●                                    | ●               | ●                       |                      |              |                       |                        |                       |                |                       |               |               |
| 6.4 Surfaces and Materials                | 6.4.1 Time-Resolved Photoemission                                      | ●                                    |                 | ●                       |                      |              | ●                     |                        |                       |                |                       |               |               |
|                                           | 6.4.2 Ultra-Fast Magnetization Dynamics                                | ●                                    | ●               | ●                       | ●                    |              | ●                     | ●                      | ●                     |                | ●                     | ●             |               |
|                                           | 6.4.3 Non-Equilibrium Dynamics in Strongly Correlated Electron Systems | ●                                    |                 | ●                       |                      |              | ●                     |                        |                       |                |                       |               |               |
|                                           | 6.4.4 Lattice Dynamics studied with Time-Resolved X-ray Diffraction    | ●                                    |                 | ●                       |                      |              | ●                     |                        |                       |                |                       |               |               |
|                                           | 6.4.5 Non-linear X-ray Spectroscopy                                    | ●                                    | ●               |                         | ●                    |              | ●                     | ●                      |                       |                | ●                     |               |               |
| 6.5 Shock Physics                         | 6.5.1 Background                                                       | ●                                    | ●               | ●                       | ●                    |              | ●                     | ●                      | ●                     |                |                       |               |               |
|                                           | 6.5.2 Ultimate Strength under Compression                              | ●                                    |                 | ●                       |                      |              |                       |                        |                       |                |                       |               |               |
|                                           | 6.5.3 Phase Transitions, Melting and Recrystallization                 | ●                                    |                 |                         | ●                    |              |                       |                        |                       |                |                       |               |               |
|                                           | 6.5.4 Future Experiments and Quasi-Isentropic Compression              |                                      |                 | ●                       |                      |              |                       |                        |                       |                |                       |               |               |
| 6.6 Solid Density Plasmas                 | 6.6.1 Background                                                       |                                      | ●               |                         |                      | ●            |                       |                        |                       |                |                       |               |               |
|                                           | 6.6.2 X-ray Isochoric Heating                                          | ●                                    |                 |                         | ●                    | ●            |                       |                        |                       |                |                       |               |               |
|                                           | 6.6.3 X-ray-driven Emission Spectroscopy                               | ●                                    | ●               |                         |                      |              | ●                     | ●                      |                       |                |                       |               |               |
|                                           | 6.6.4 Non-linear X-ray Processes and X-ray Scattering                  | ●                                    | ●               | ●                       |                      |              |                       | ●                      | ●                     | ●              |                       |               |               |
|                                           | 6.6.5 Summary                                                          | ●                                    | ●               | ●                       | ●                    |              |                       | ●                      | ●                     |                |                       |               |               |
| 6.7 Industrial Perspective                | 6.7.1 Materials Science and Engineering                                |                                      | ●               | ●                       | ●                    | ●            | ●                     |                        |                       |                |                       |               |               |
|                                           | 6.7.2 EUV Lithography                                                  |                                      |                 |                         | ●                    |              |                       |                        |                       | ●              |                       |               |               |
|                                           |                                                                        | 10s-fs pulses                        | Peak brightness | Multi-pulse (laser-FEL) | Transverse coherence | Pulse energy | Wavelength tunability | Longitudinal coherence | Multi-pulse (FEL-FEL) | High rep. rate | Variable Polarisation | Few-fs pulses | Sub-fs pulses |

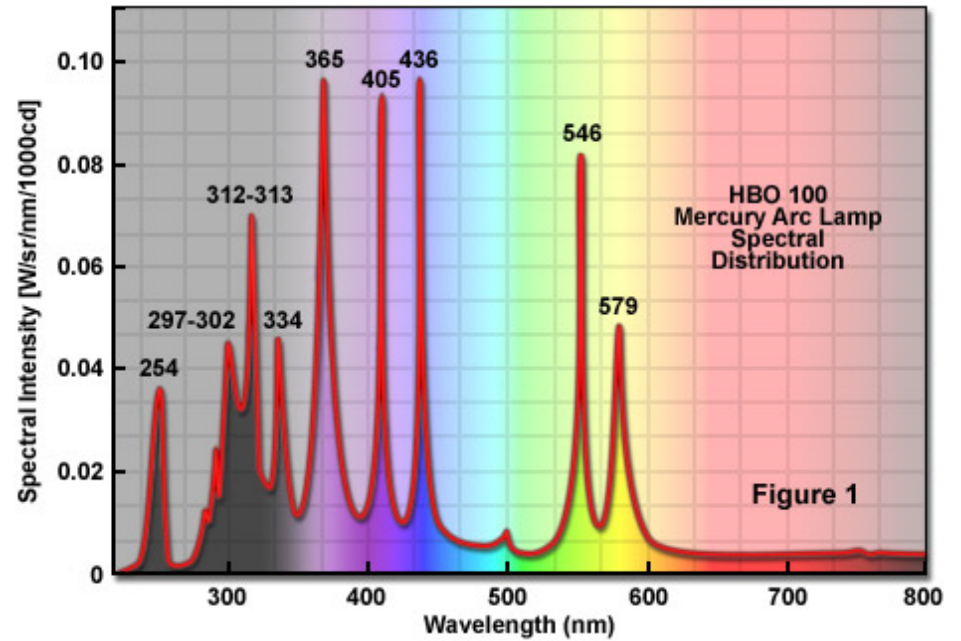
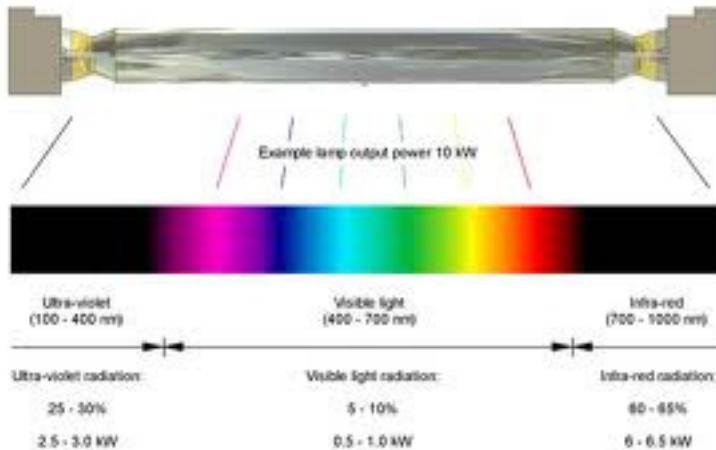
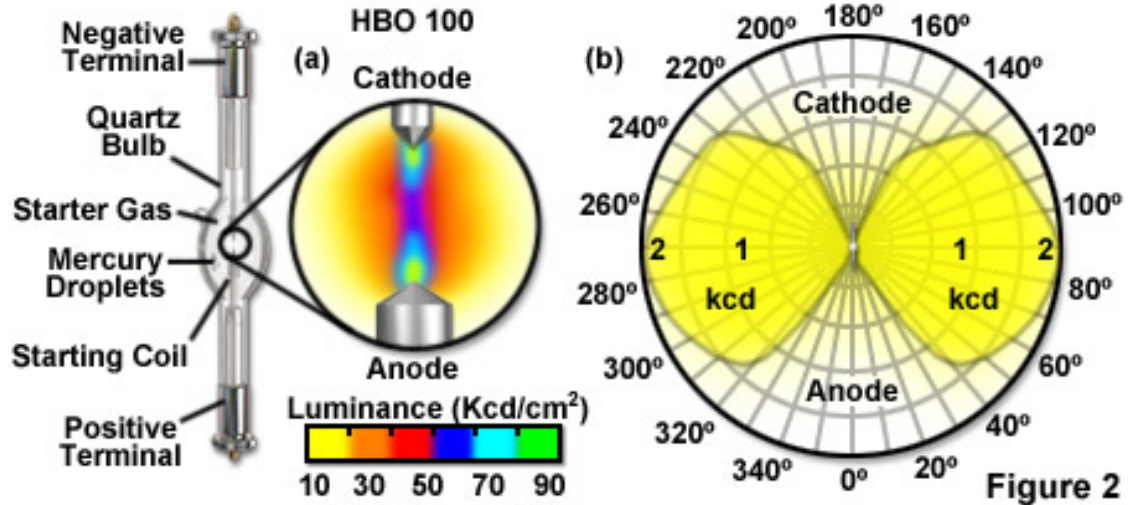
# **CONTROLLING THE LIGHT**



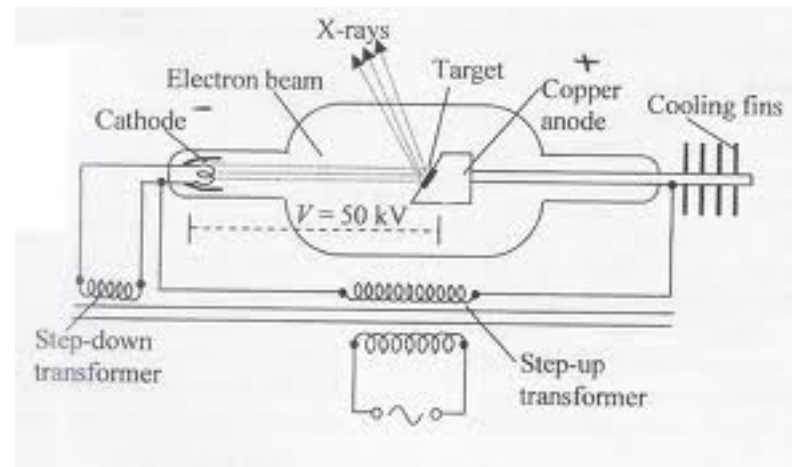


# THERMAL LIGHT

## Mercury Arc Lamp Luminance Profile and Light Flux Distribution

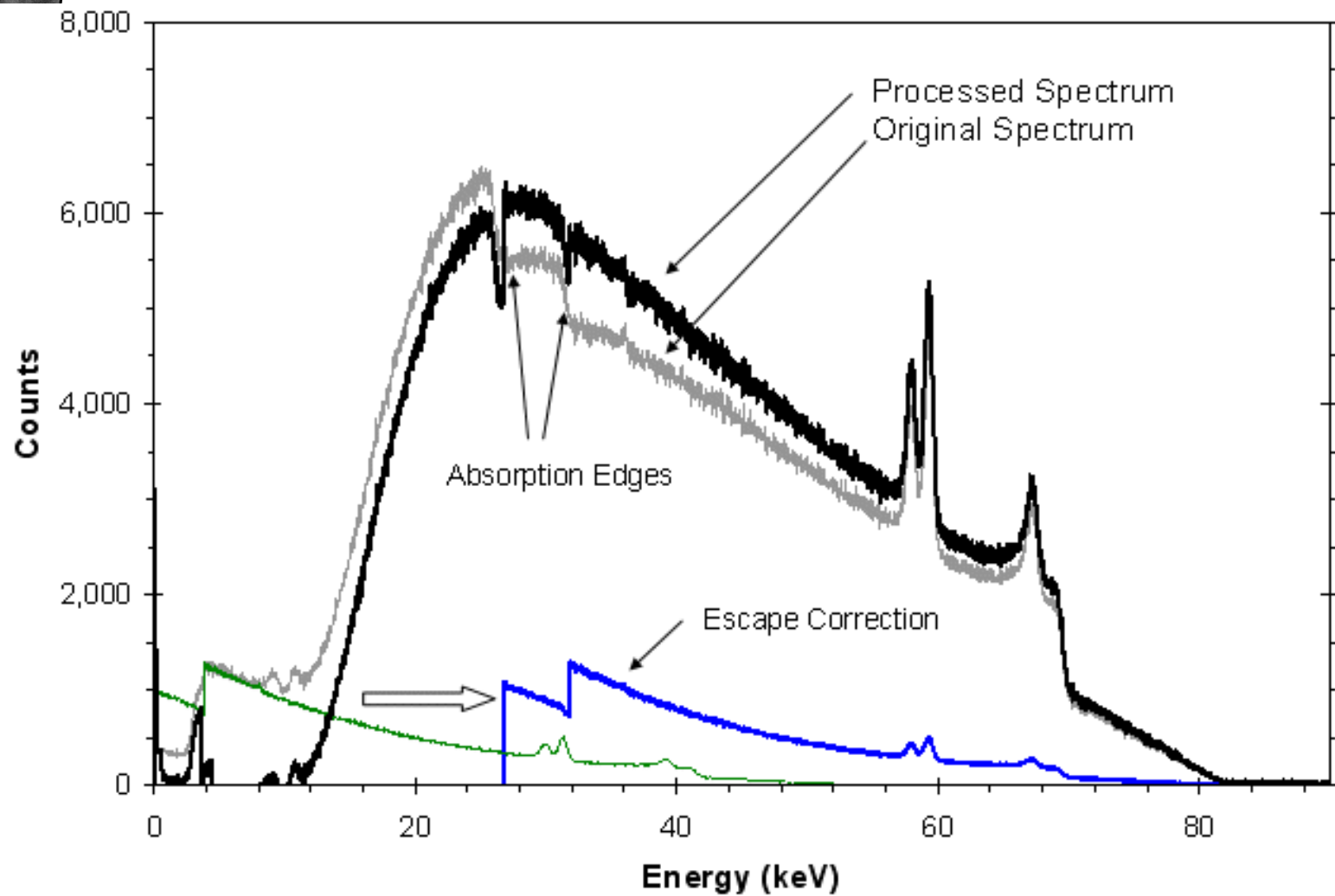


# THE BREMSSTRAHLUNG





# THE BREMSSTRAHLUNG



# THE SYNCHROTRON LIGHT

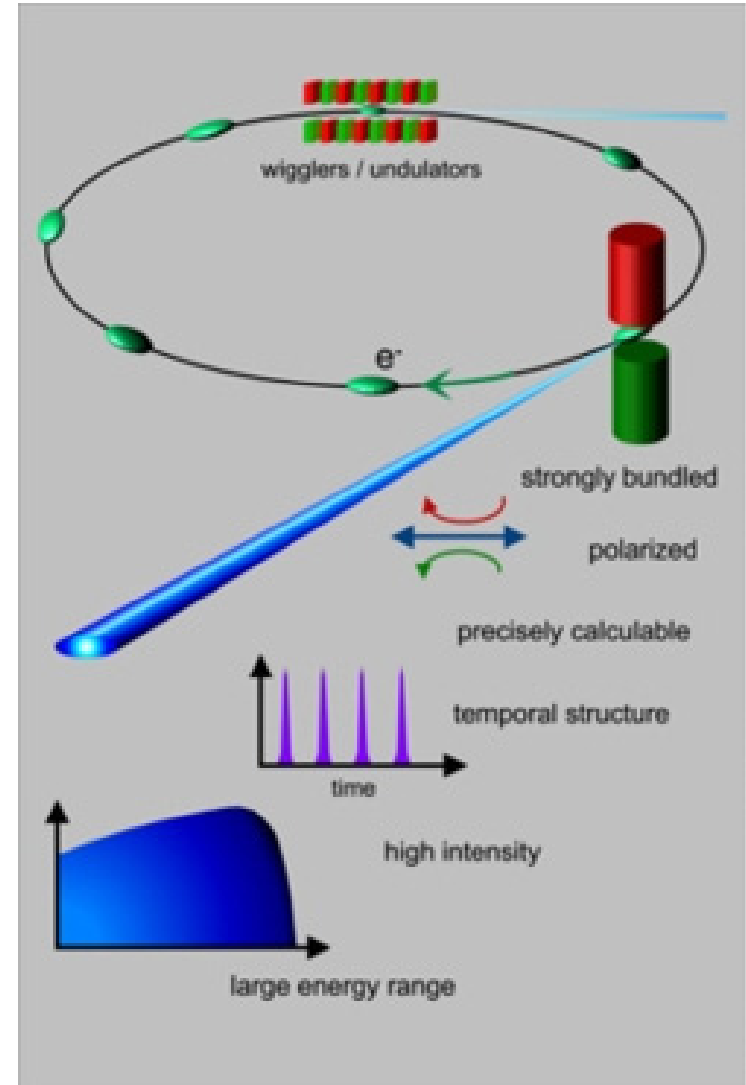
Synchrotron light occurs naturally in our Universe.

**ACCELERATOR-BASED SYNCHROTRON LIGHT WAS SEEN FOR THE FIRST TIME AT THE GENERAL ELECTRIC RESEARCH LABORATORY IN THE USA IN 1947 IN A TYPE OF ACCELERATOR KNOWN AS A SYNCHROTRON.**

**FIRST CONSIDERED A nuisance because it caused the particles to lose energy, it recognized in the 1960s AS LIGHT WITH EXCEPTIONAL PROPERTIES.**

**SYNCHROTRON RADIATION IS POLARIZED AND PULSED, AND THE FREQUENCY AND DURATION OF THE PULSES CAN BE MANIPULATED TO A CERTAIN EXTENT.**

**Worldwide there exist about 30 laboratories for the production of synchrotron radiation.**



**HIGH ENERGY  
AND  
MOMENTUM RESOLUTION**

# SOFT X-RAY EXPERIMENTS

Courtesy of Y. Zhu, BNL

## •X-ray Diffraction

**Soft X-rays (ordering)**

## •Photoelectron Spectroscopy (PES)

**Core level electron spectroscopy**

**Micro- and nano-PES**

**PhotoElectron Emission Microscopy (PEEM)**

**Angle Resolved PES (ARPES)**

**Resonant photoemission**

**Photoelectron Diffraction**

## •X-ray Absorption Spectroscopy (XAS)

**X-ray Absorption Spectroscopy (XAS)**

**X-ray Magnetic Circular Dichroism (XMCD)**

**X-ray Magnetic Linear Dichroism (XMLD)**

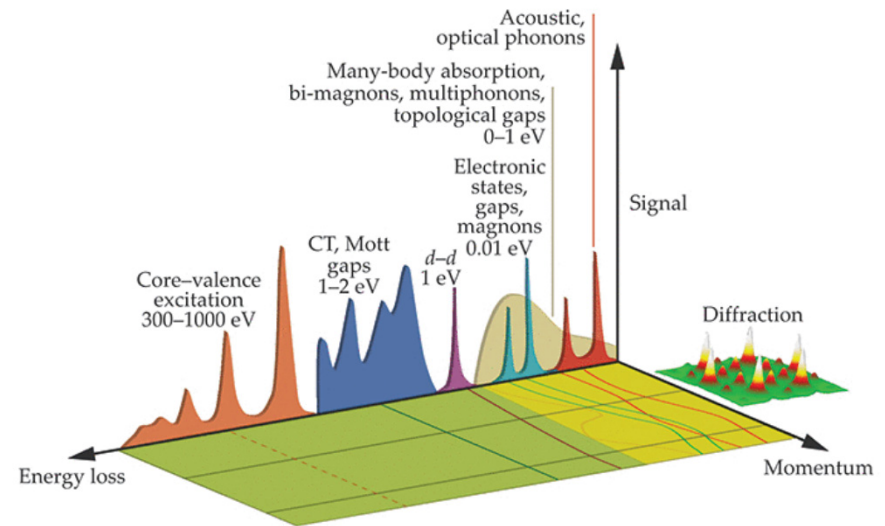
## •X-ray Emission Spectroscopy (XES)

**Resonant Inelastic X-ray Scattering (RIXS)**

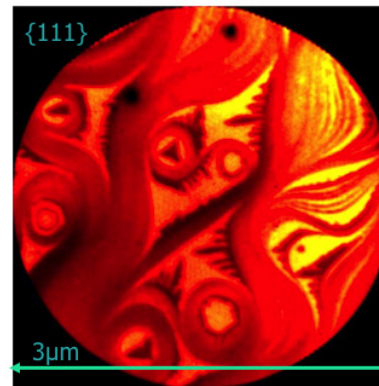
## •Soft X-ray Elastic Scattering

**Imaging**

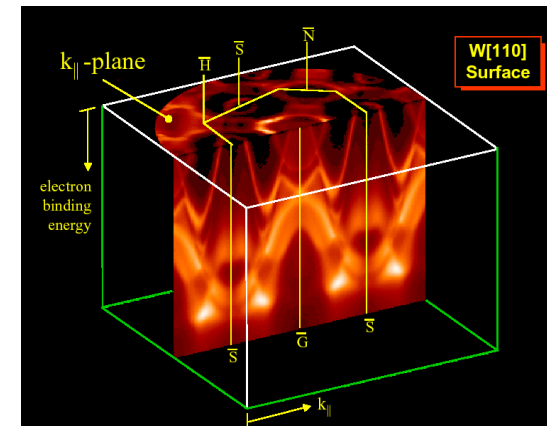
**Speckle**



Courtesy  
Nanospectroscopy Elettra



Courtesy E. Rotenberg, ALS



# **HIGH TRANSVERSAL COHERENCE**

# A SUPERMICROSCOPE WITH Å-SIZE LATERAL RESOLUTION IN MACROSCOPIC OBJECTS?

## Vascular diseases, visualization of microvasculatures.

*P. Liu, J. Sun, J. Zhao, X. Liu, X. Gu, J. Li, T. Xiao and L. X. Xu*

*[J. Synchrotron Rad. \(2010\). 17, 517-521](#)*



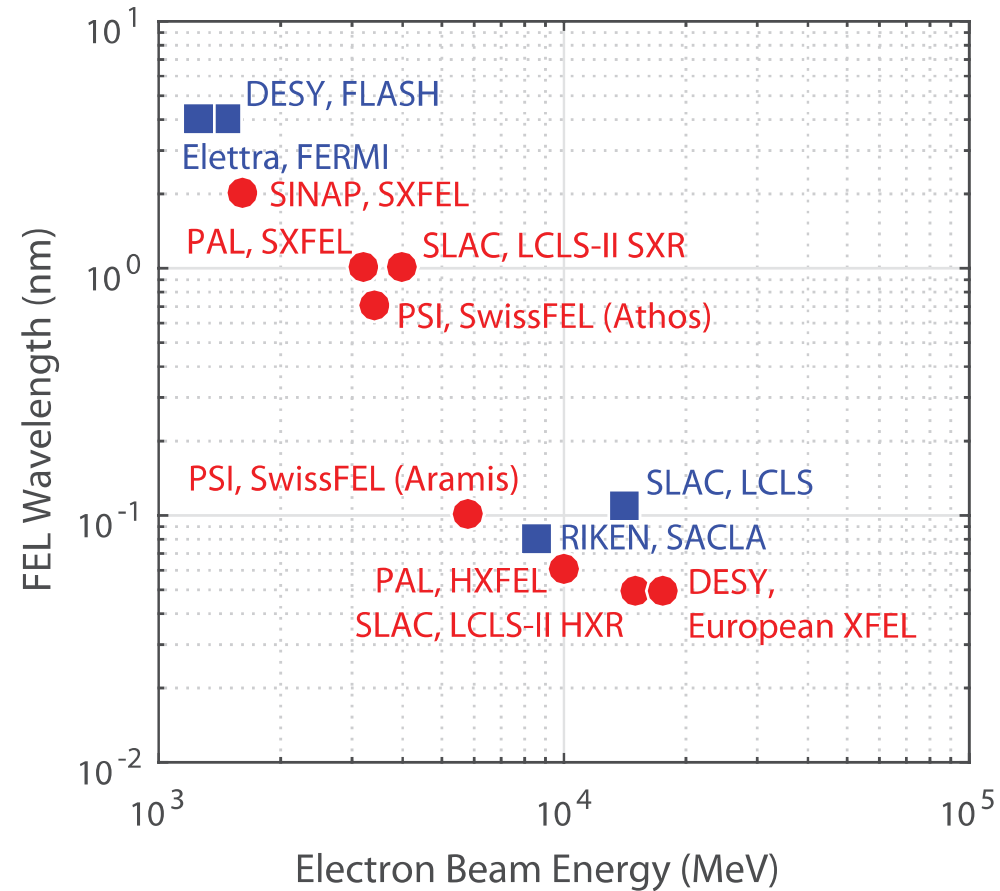
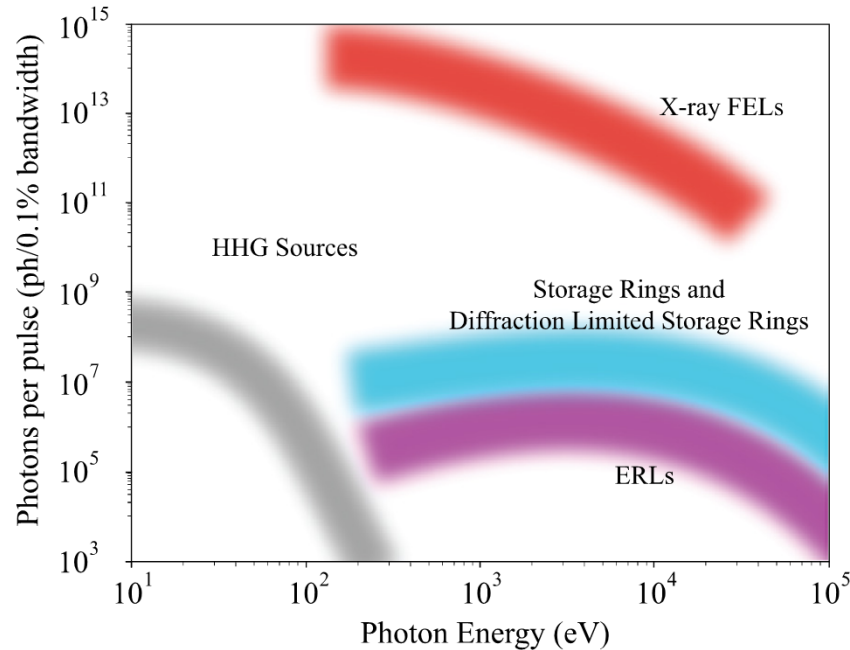


**MAX IV**

|                                                                     |                   |
|---------------------------------------------------------------------|-------------------|
| <b>Circumference (m)</b>                                            | <b>528</b>        |
| <b>Nr of straight sections</b>                                      | <b>20</b>         |
| <b>Injection full energy, top-up</b>                                |                   |
| <b>Stored current (mA)</b>                                          | <b>500</b>        |
| <b>Horizontal emittance (nm rad)</b>                                | <b>0.2 - 0.3</b>  |
| <b>Vertical emittance (nm rad)</b>                                  | <b>&lt; 0.008</b> |
| <b>Horizontal beam size (<math>\sigma</math> <math>\mu</math>m)</b> | <b>42-52</b>      |
| <b>Vertical beam size (<math>\sigma</math> <math>\mu</math>m)</b>   | <b>&lt; 6</b>     |



# RADIATIONS SURCES: AN INTERESTING COMPARISON



**IS THE RADIATION SOURCE BRIGHTNESS THE LEADING PARAMETER?**



# HOW FAR DO WE KNOW AND CONTROL THE LIGHT PROPERTIES?

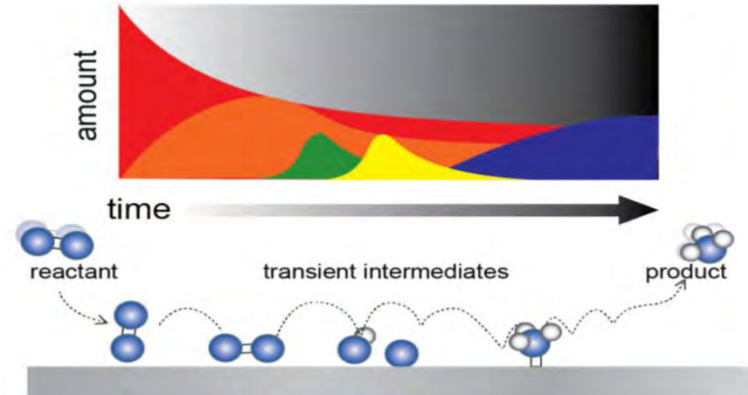
CONTROL OF COMPLEX MATERIALS  
AND CHEMICAL PROCESSES

REAL TIME EVOLUTION  
OF CHEMICAL REACTIONS, MOTION  
OF ELECTRONS AND SPIN

IMAGING AND SPECTROSCOPY  
OF INDIVIDUAL NANO-OBJECTS

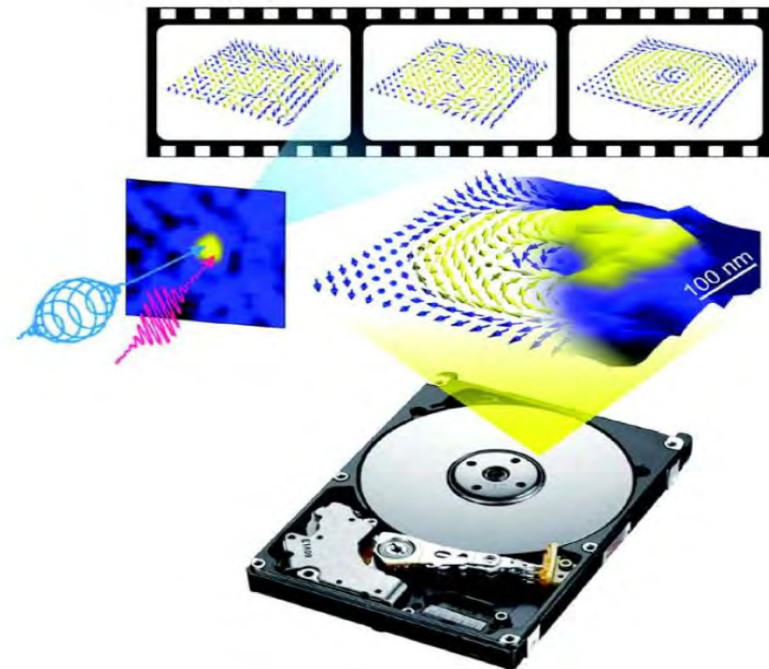
STATISTICAL LAWS OF COMPLEX  
SYSTEMS

SIMULTANEOUS ULTRASHORT  
AND ULTRAFAST MEASUREMENTS



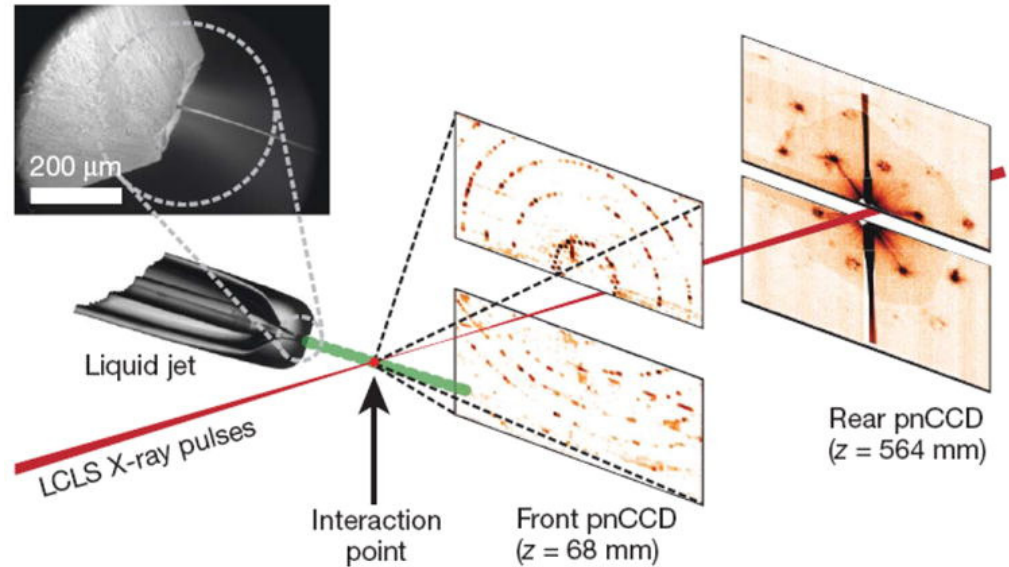
M. Dell'Angela *et al.*, *Science* 339, 1302 (2013)

- ✓ **Brightness**
- ✓ **Spectral brightness**
- ✓ **Temporal structure**
- ✓ **Polarization,**
- ✓ **Coherence,**
- ✓ **Tunability,**
- ✓ **Pulse Repetition Rate**

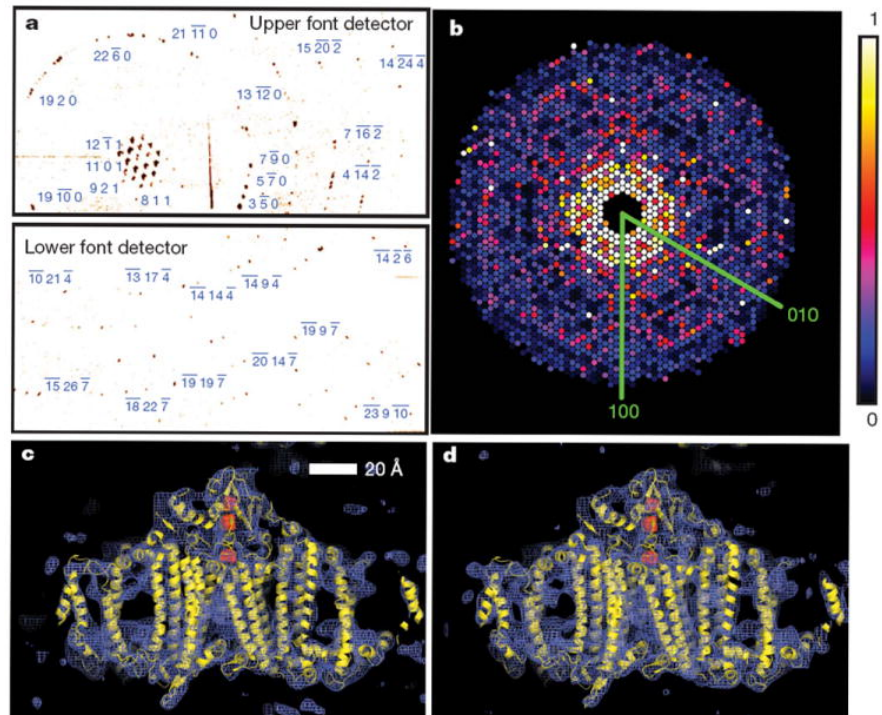


**HIGH BRILLIANCE**

# A DIFFRACTION OF NANO-OBJECTS



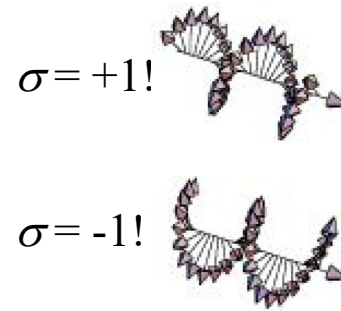
Chapman HN et al.  
Nature (2011)



# **POLARIZATION**

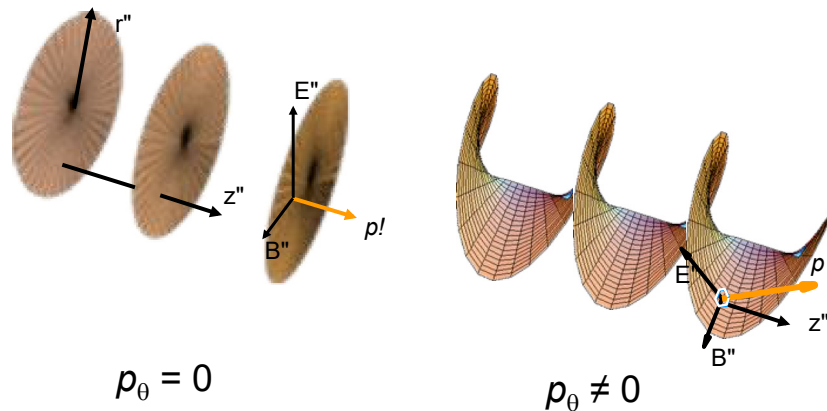
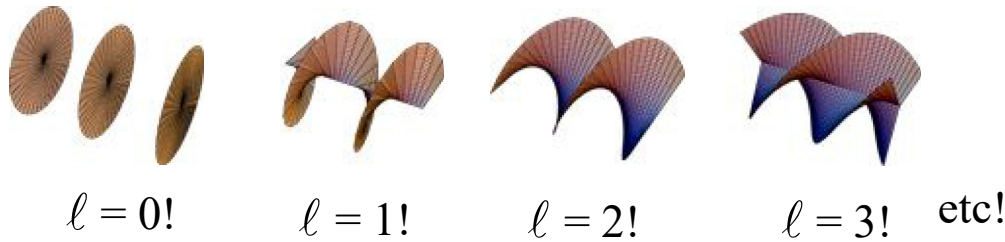
# ANGULAR MOMENTUM IN TERMS OF PHOTONS

- Spin angular momentum
  - Circular polarisation
  - $\sigma\hbar$  per photon
- Orbital angular momentum
  - Helical phasefronts
  - $\ell\hbar$  per photon

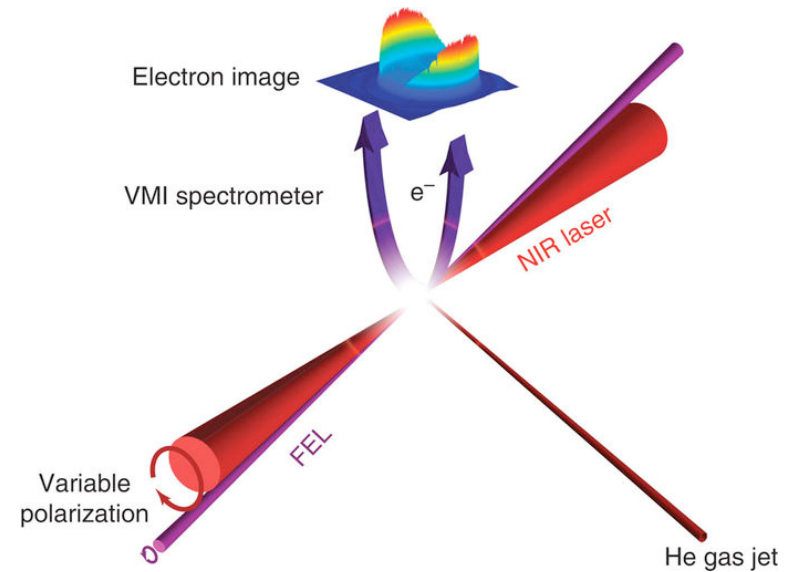
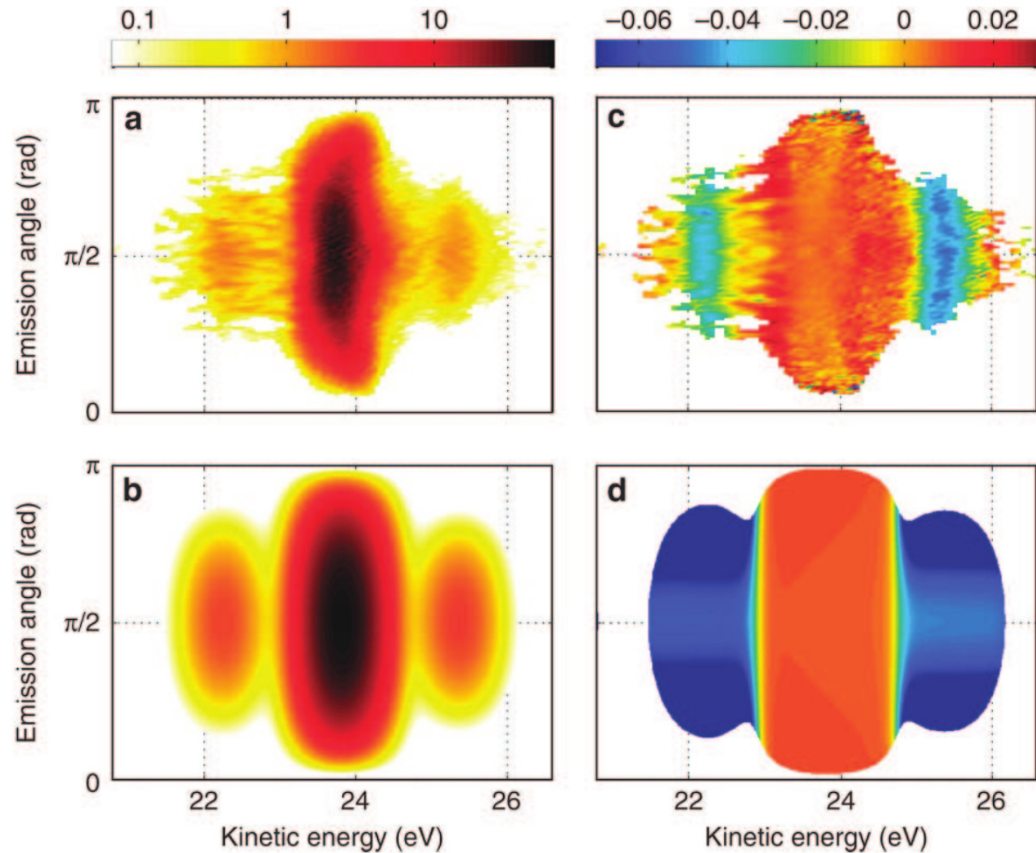


**Intrinsic angular momentum of the photon formally equivalent to the particle spin**

**Adapted from Miles Padgett**



# CONTROLLING THE POLARIZATION



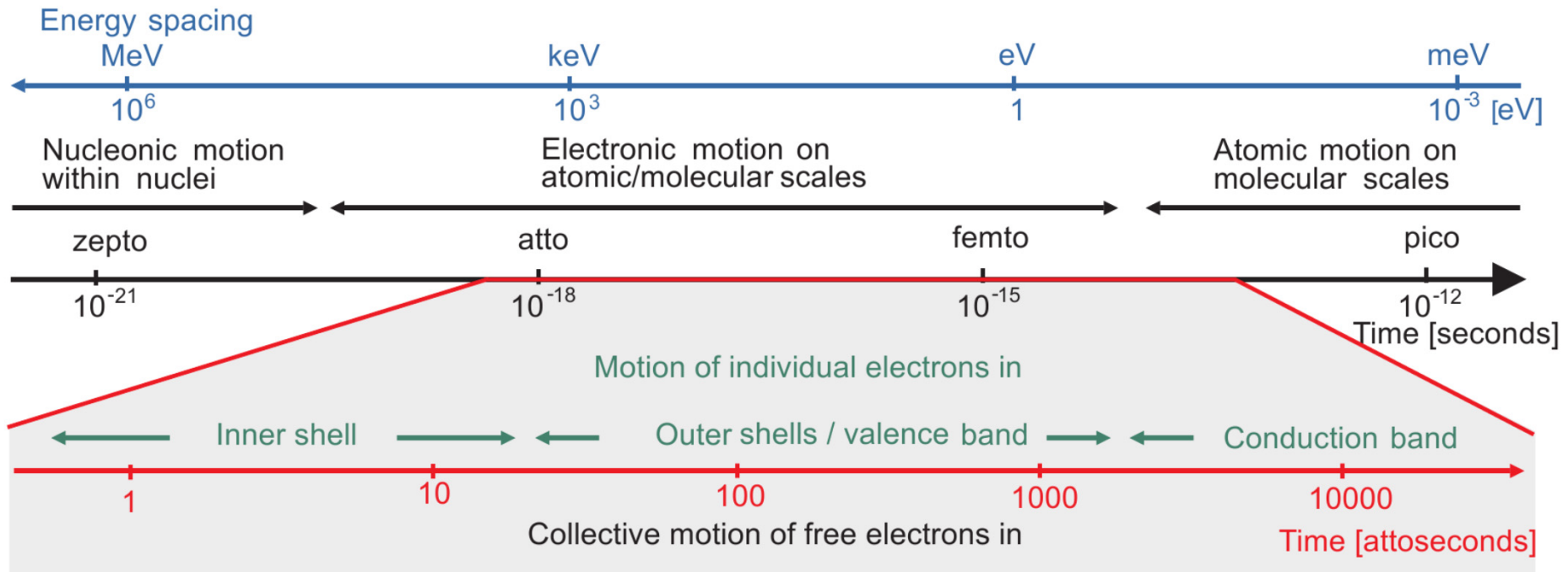
**Angle- and energy-resolved electron spectra and circular dichroism in He 1 s photoionization at low-NIR laser intensity**

Reproduced from T. Mazza et al., Nature Commun. 5, 3648 (2014)

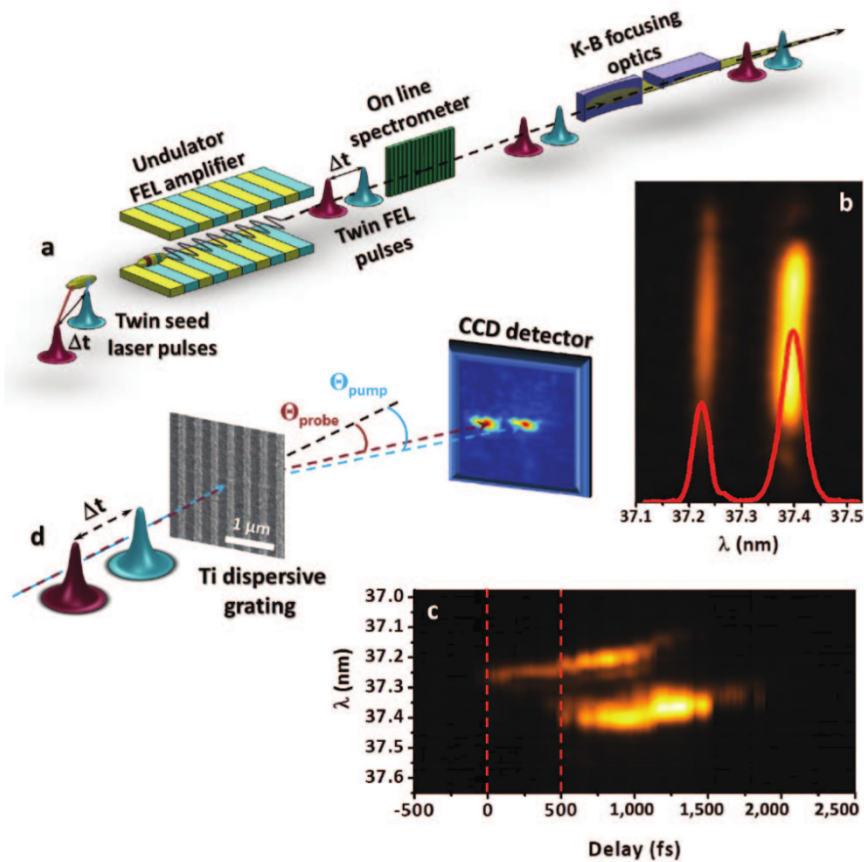


**TIME DOMAIN**

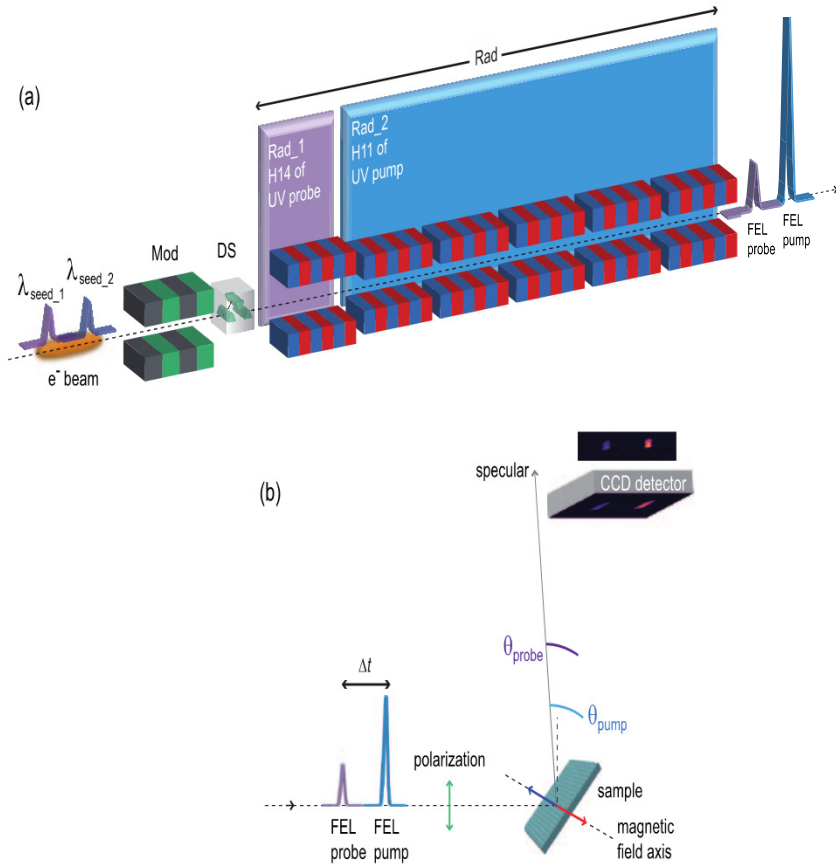
# A GLANCE TO THE TIME SCALES



# TWINED FEL PULSES FROM TWIN SEEDING LASER PULSES

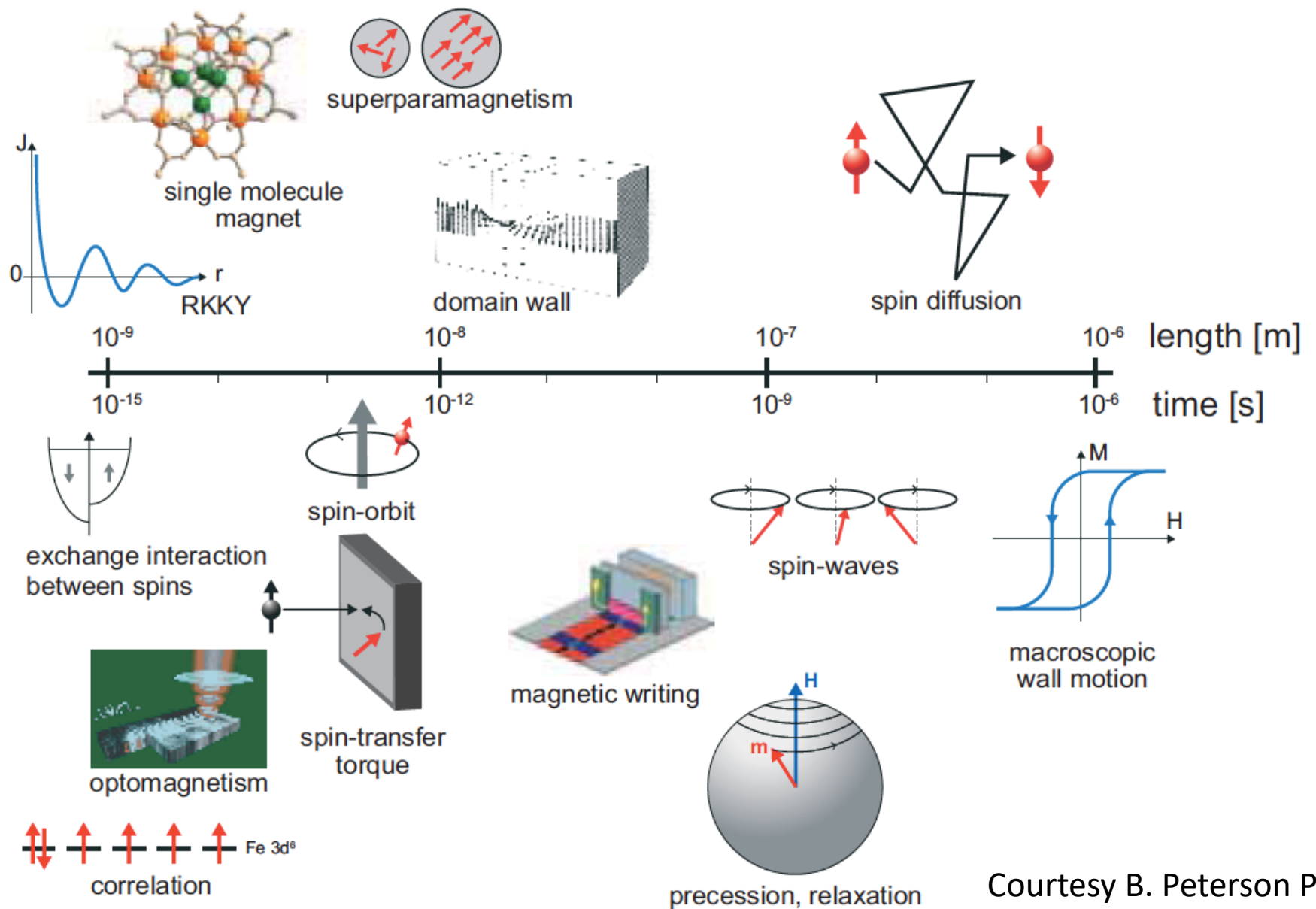


Reproduced from E. Allaria et al., Nat. Commun. 4, 3476 (2013)



Reproduced from E. Ferrari et al., Nat. Commun. 7, 10343 (2016)

# TIME SCALE OF MAGNETIC DYNAMICS



Courtesy B. Peterson PSI

# ULTRA FAST DEMAGNETIZATION DYNAMICS

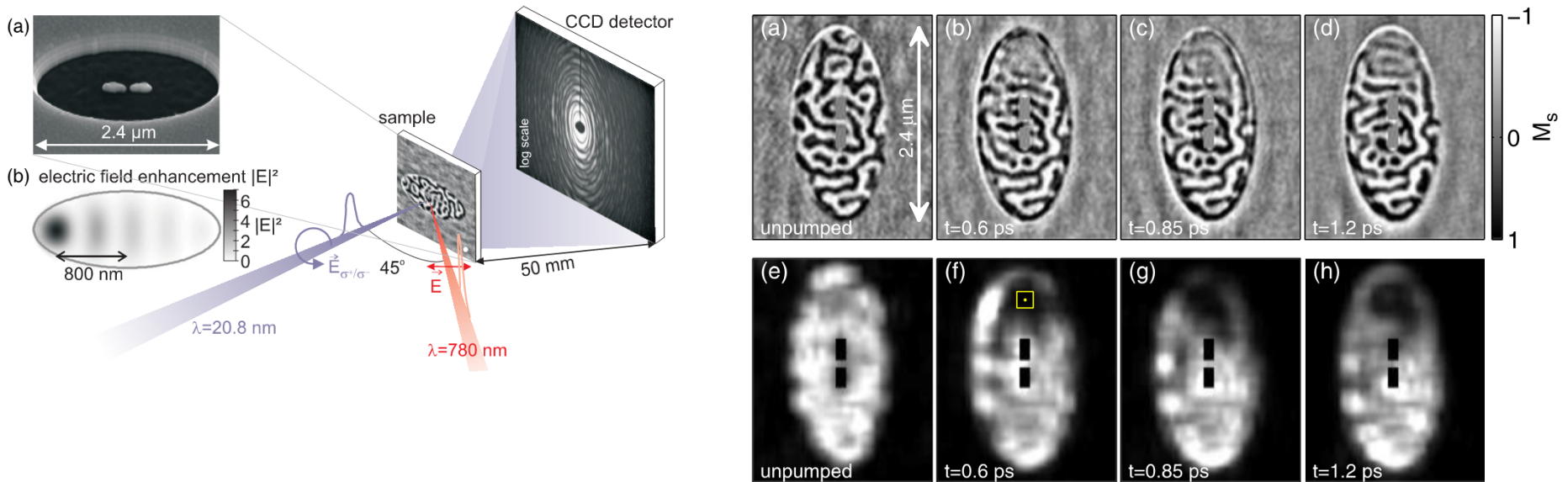
PRL 112, 217203 (2014)

PHYSICAL REVIEW LETTERS

week ending  
30 MAY 2014

## Imaging Ultrafast Demagnetization Dynamics after a Spatially Localized Optical Excitation

C. von Korff Schmising,<sup>1,\*</sup> B. Pfau,<sup>1,†</sup> M. Schneider,<sup>1</sup> C. M. Günther,<sup>1</sup> M. Giovannella,<sup>1,2</sup> J. Perron,<sup>3,4</sup> B. Vodungbo,<sup>3,4</sup>  
L. Müller,<sup>5</sup> F. Capotondi,<sup>6</sup> E. Pedersoli,<sup>6</sup> N. Mahne,<sup>6</sup> J. Lüning,<sup>3,4</sup> and S. Eisebitt<sup>1,7,8</sup>

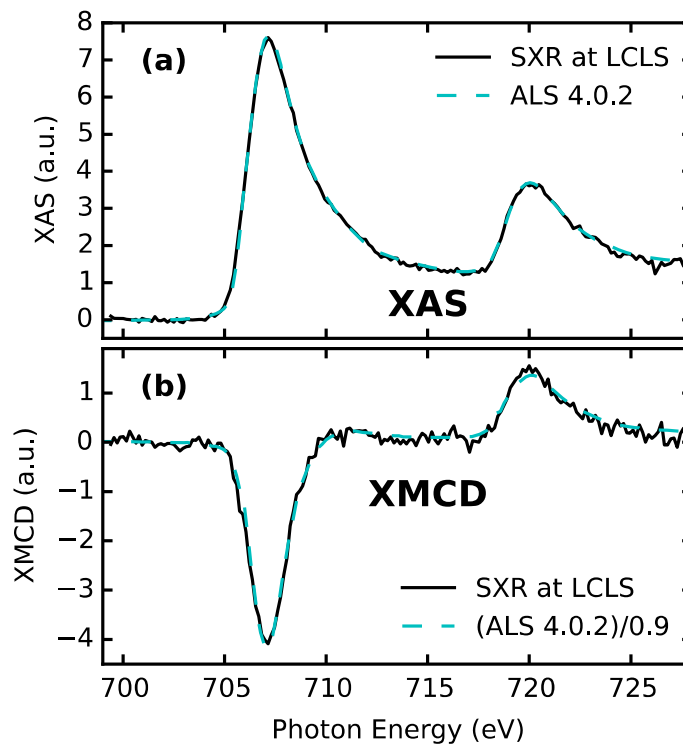
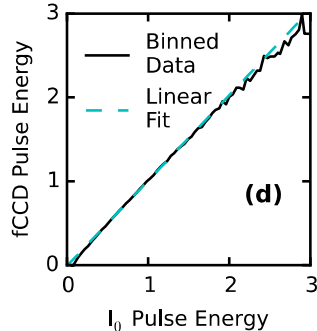
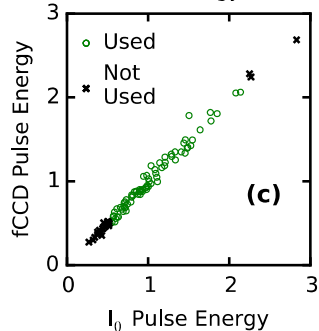
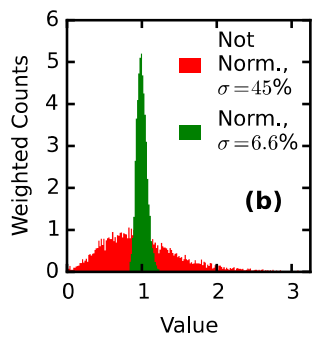
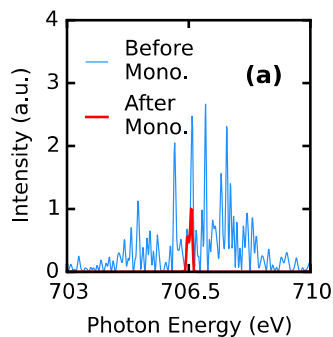
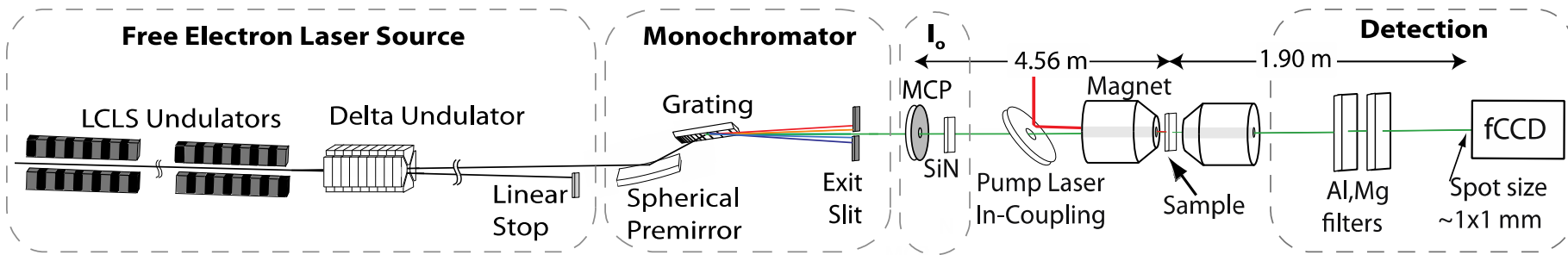


Ultrashort, coherent x-ray pulses of a free-electron laser are used to holographically image the magnetization dynamics within a magnetic domain pattern after creation of a localized excitation via an optical standing wave. We observe a spatially confined reduction of the magnetization within a couple of hundred femtoseconds followed by its slower recovery. Additionally, the experimental results show evidence of a spatial evolution of magnetization, which we attribute to ultrafast transport of nonequilibrium spin-polarized electrons for early times and to a fluence-dependent remagnetization rate for later times.

**TUNABILITY**



# ULTRAFAST XMCD



**FEL PULSES  
STABILITY  
(BANDWIDTH-INTENSITY-POINTING)**

# M-edge RIXS @ FERMI

Martina Dell'Angela

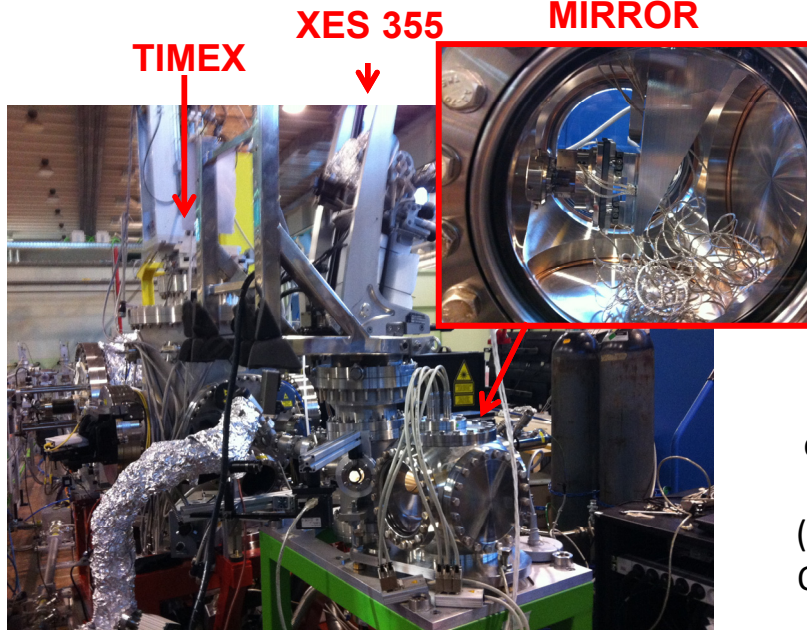
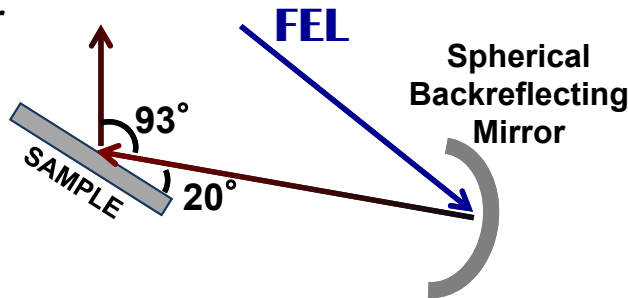
in collaboration with

W. Wurth (UNI HH/CFEL) and S. Bajit (CFEL/DESY)

TIMEX, MagneDyn and T-ReX teams

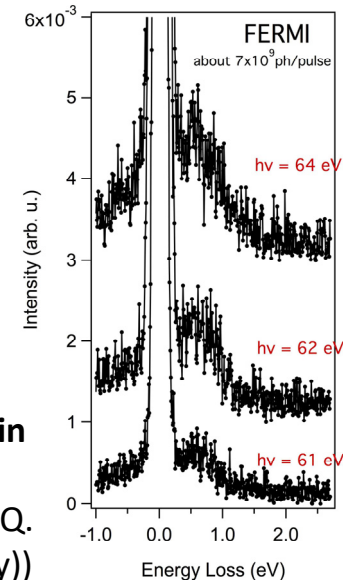
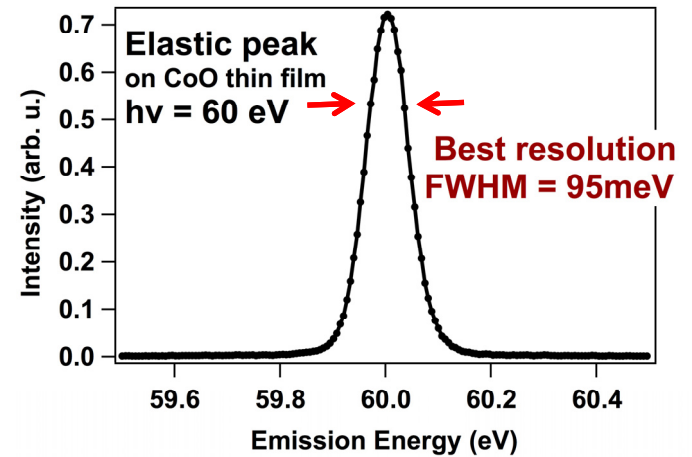
X-ray Spectrometer  
XES 355

TIMEX

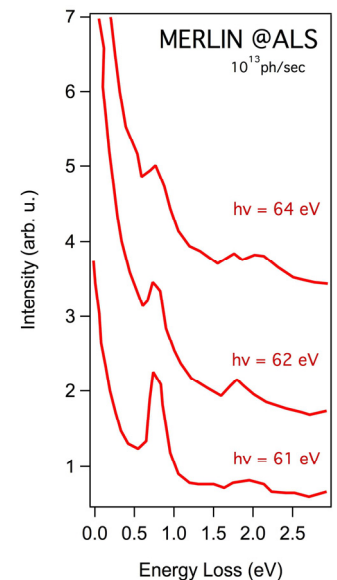


dd- excitations in  
CoO  
(sample from Z. Q.  
Qiu (UC Berkeley))

Resolution



FERMI Nov 2014



Wray et al.  
PRB 88, 035105 (2013)

# **SHAPING THE FEL PULSES**

# TOWARD ATTOSECOND X-RAY PULSES

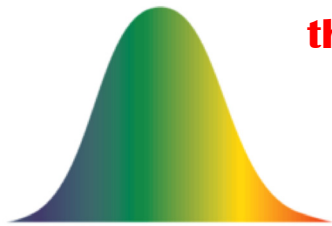
Courtesy : (A.L. Cavalieri)

Chirped seeding

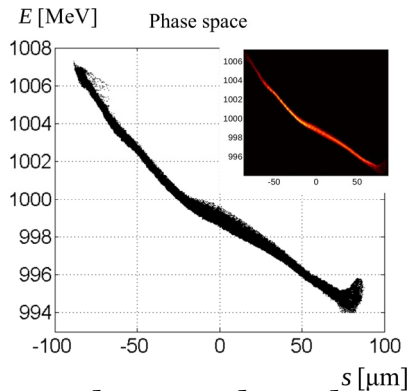
X-ray pulse compression

Compressed pulse diagnostics

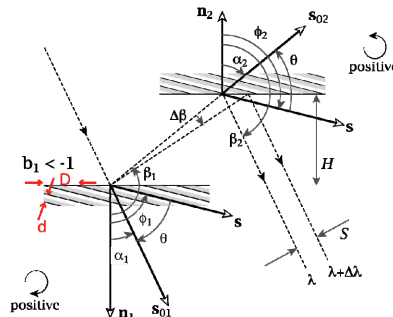
Compressor based on asymmetric-cut multi-layer coatings that are currently being developed at CFEL



optical seed

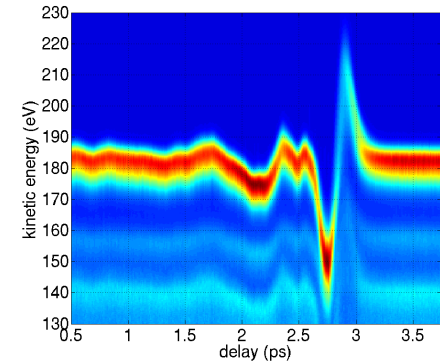


electron bunch



**broadband X-ray emission and compression**

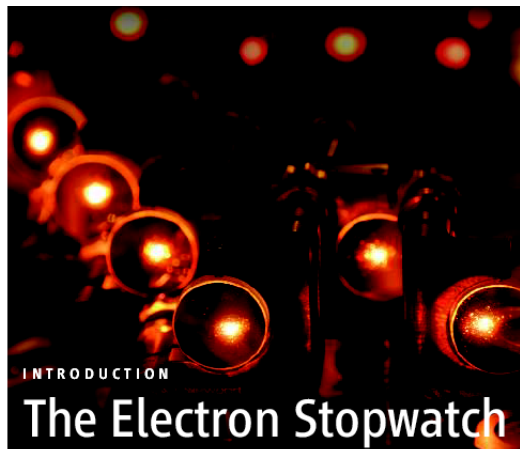
(S. Bajt *et al. J. Opt. Soc. Am. A* (2012))



**full temporal characterization by photoelectron streaking**

(Grguraš I. *et al. Nature Photonics* (2012))

- **1% BW at 200eV → ~1 fs transform limited pulse duration**
- **Constant pulse energy → 100-fold increase in peak power**



## Attosecond Spectroscopy

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#### Reviews

- 766 The Future of  
Attosecond Spectroscopy  
P. H. Bucksbaum
- 769 Attosecond Control  
and Measurement:  
Lightwave Electronics  
E. Goulielmakis *et al.*
- 775 Harnessing Attosecond  
Science in the Quest  
for Coherent X-rays  
H. Kapteyn *et al.*

THE PRECISION ATTAINABLE IN TIMING EVENTS ONCE DEPENDED ON HOW fast a human being could press the button on a stopwatch. More recently, pulsed laser sources have taken the place of those hand-held devices for measuring the fastest phenomena. The technology for tracking the time scale of nuclear motion in free molecules and solids was limited by the duration of a single cycle of visible light: approximately 0.00000000000001 second, or 1 femtosecond. Electrons move even faster than that, and for a long time, scientists could only watch their rearrangements as an indistinct blur. Over the past several years, however, laser technology has crossed the threshold into the attosecond regime (a thousandth of a femtosecond). This series of three Reviews highlights the methods underlying this advance and the scientific prospects they have enabled.

Bucksbaum (p. 766) lays out the essential physics of high harmonic generation, a technique whereby an intense laser field pulls an atomic electron away from the nucleus like a loaded slingshot and then sends it careening back, giving rise to the emission of an attosecond light pulse. The Review also describes in general terms what events such light pulses can be used to track, ranging from electron rearrangements in chemical bonding to conduction dynamics in metallic solids.

Goulielmakis *et al.* (p. 769) take a more in-depth look at the laser techniques that create and detect attosecond pulses. Their Review also details the prospects not only of passively probing electron motions, but of actively manipulating and controlling them.

In keeping with the uncertainty principle, compressing a light pulse's duration must also broaden its spectral bandwidth. Thus, attosecond pulses extend into the x-ray region of the electromagnetic spectrum. Kapteyn *et al.* (p. 775) describe efforts to harness this feature of the technology in diffraction and imaging experiments, which would otherwise depend on much more elaborate x-ray generation apparatus.

Optical technology continues to evolve. It seems that just as events at the atomic scale are at last observed with precision, they bring into view a new series of blurs, previously unappreciated. Then the quest begins for an even faster stopwatch.

— IAN OSBORNE AND JAKE YESTON

## Harnessing Attosecond Science in the Quest for Coherent X-rays

Henry Kapteyn, Oren Cohen, Ivan Christov, Margaret Murnane\*

Modern laser technology has revolutionized the sensitivity and precision of spectroscopy by providing coherent light in a spectrum spanning the infrared, visible, and ultraviolet wavelength regimes. However, the generation of shorter-wavelength coherent pulses in the x-ray region has proven much more challenging. The recent emergence of high harmonic generation techniques opens the door to this possibility. Here we review the new science that is enabled by an ability to manipulate and control electrons on attosecond time scales, ranging from new tabletop sources of coherent x-rays to an ability to follow complex electron dynamics in molecules and materials. We also explore the implications of these advances for the future of molecular structural characterization schemes that currently rely so heavily on scattering from incoherent x-ray sources.

## Soft X-ray–Driven Femtosecond Molecular Dynamics

Etienne Gagnon,<sup>1</sup> Predrag Ranitovic,<sup>2</sup> Xiao-Min Tong,<sup>3</sup> C. L. Cocke,<sup>2</sup> Margaret M. Murnane,<sup>1</sup> Henry C. Kapteyn,<sup>1</sup> Arvinder S. Sandhu<sup>1\*</sup>

## Attosecond electron wave packet interferometry

T. REMETTER<sup>1</sup>, P. JOHNSON<sup>1</sup>, J. MAURITSSON<sup>2</sup>, K. VARJÚ<sup>1</sup>, Y. NI<sup>3</sup>, F. LÉPINE<sup>3</sup>, E. GUSTAFSSON<sup>1</sup>, M. KLING<sup>3</sup>, J. KHAN<sup>3</sup>, R. LÓPEZ-MARTENS<sup>4</sup>, K. J. SCHAFER<sup>2</sup>, M. J. J. VRAKKING<sup>3</sup> AND A. L'HUILLIER<sup>1\*</sup>

<sup>1</sup>Department of Physics, Lund University, PO Box 118, SE-221 00 Lund, Sweden

<sup>2</sup>Department of Physics and Astronomy, Louisiana State University, Baton Rouge, Louisiana 70803-4001, USA

<sup>3</sup>FOM-Institute AMOLF, Kruislaan 407, 1098 SJ Amsterdam, The Netherlands

<sup>4</sup>Laboratoire d'Optique Appliquée, Ecole Nationale Supérieure des Techniques Avancées (ENSTA) - Ecole Polytechnique CNRS UMR 7639, 91761 Palaiseau Cedex, France

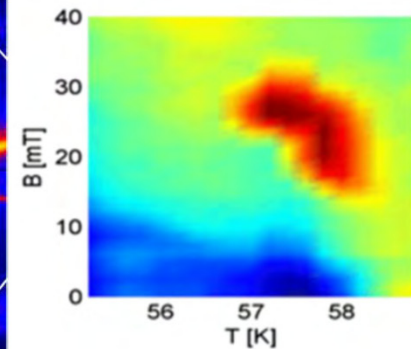
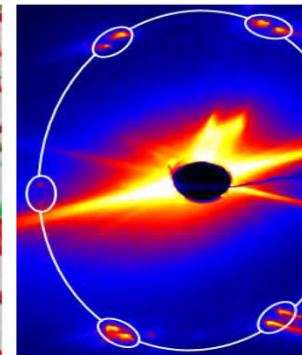
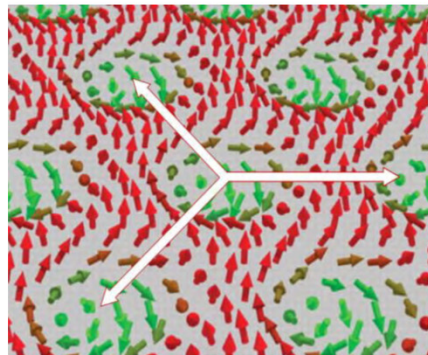
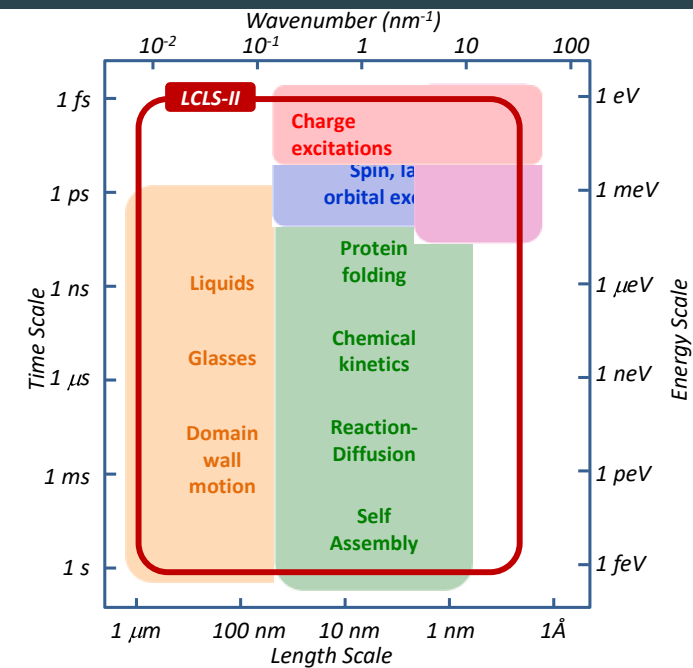
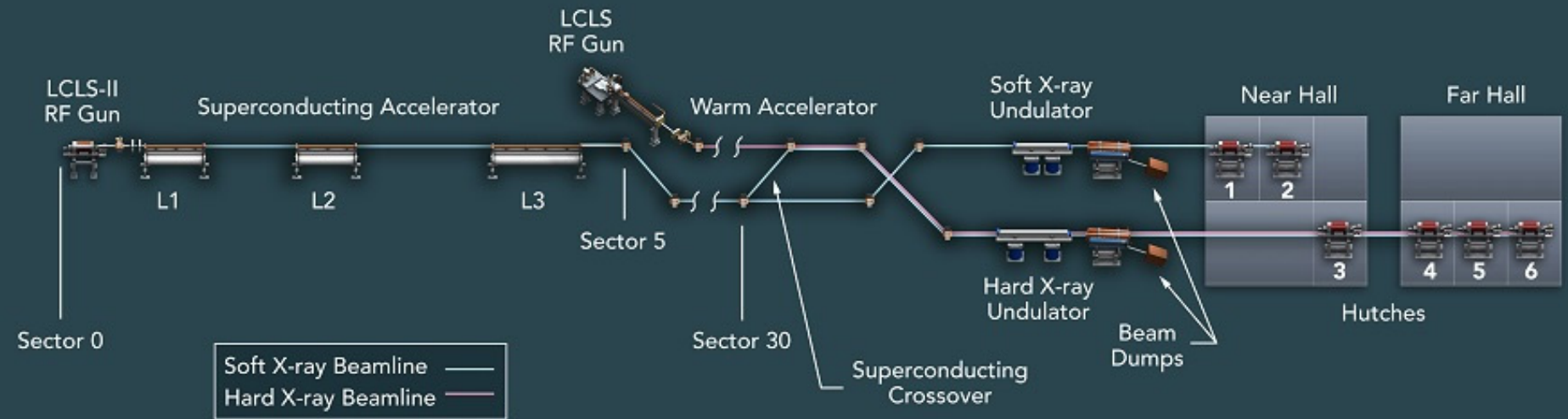
\*e-mail: anne.lhuillier@fysik.lth.se



# **PHOTON DENSITY CONTROL**

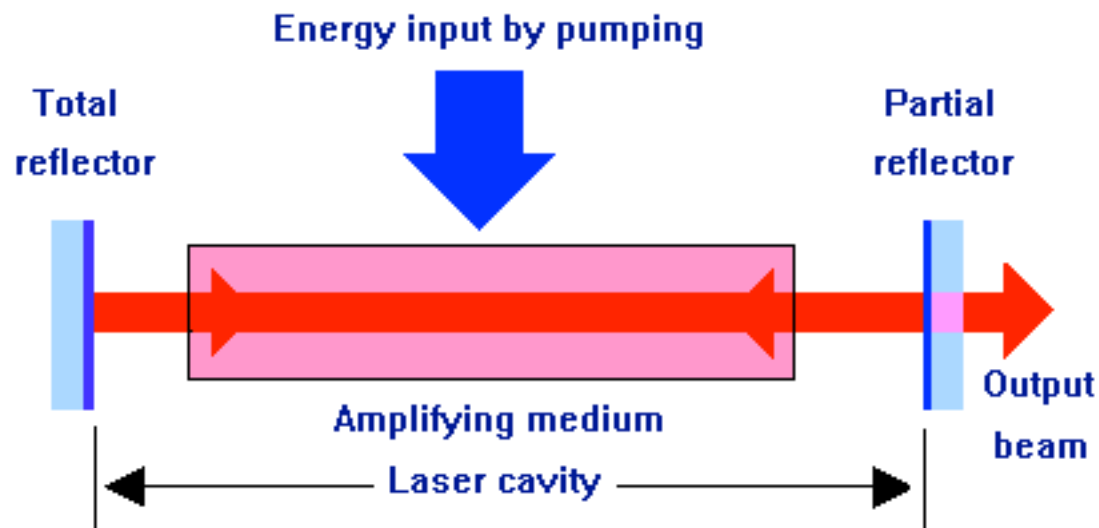
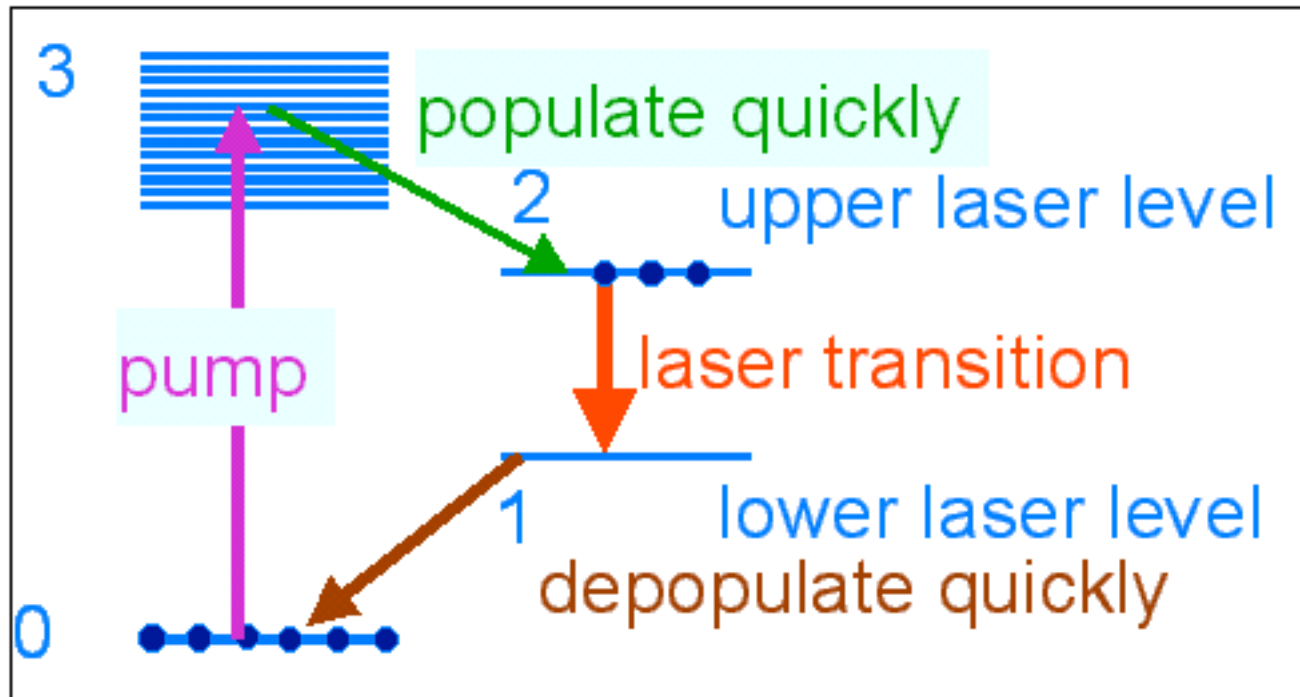
# LCLS II

## LCLS-II

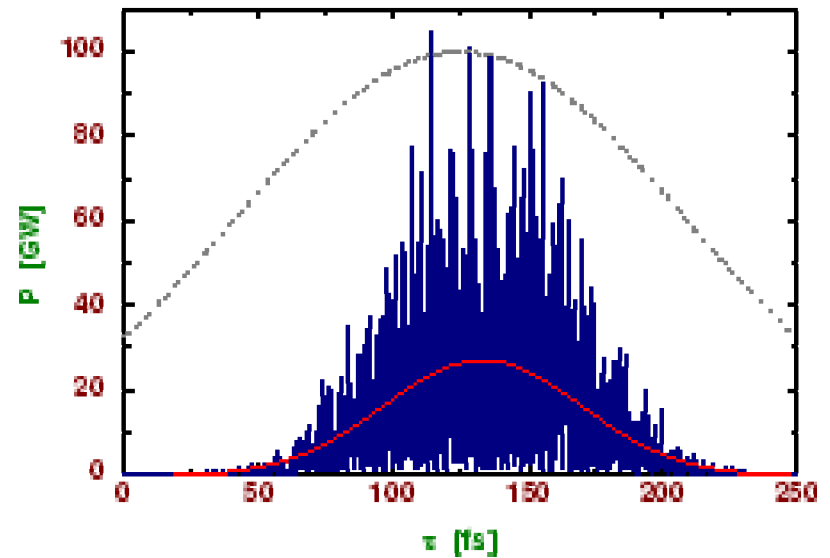
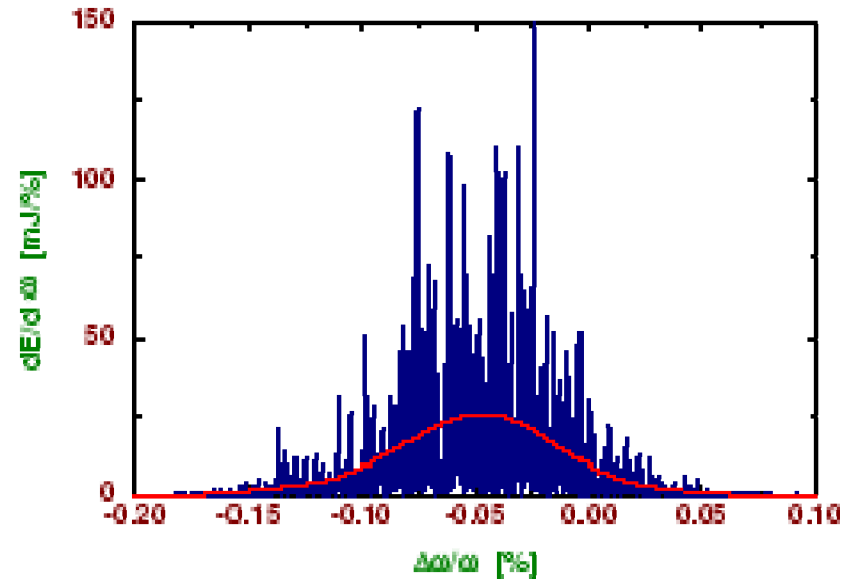
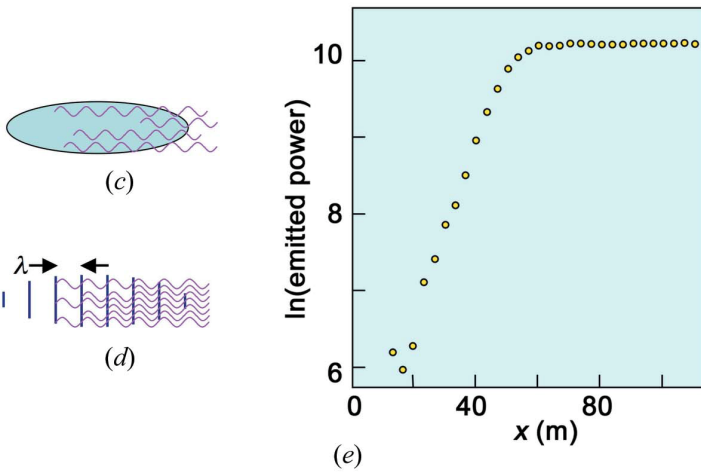
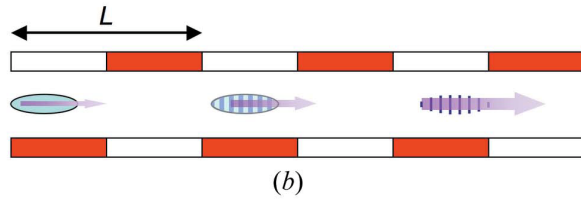
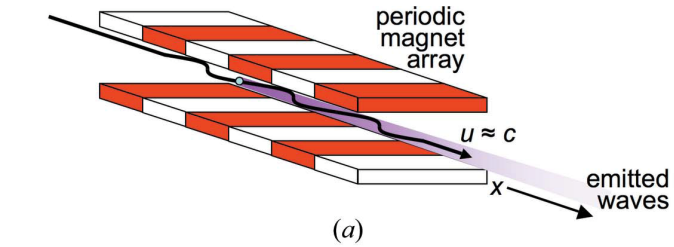


# **FULL LONGITUDINAL COHERENCE (FTL)**

# WHAT IS A LASER ?

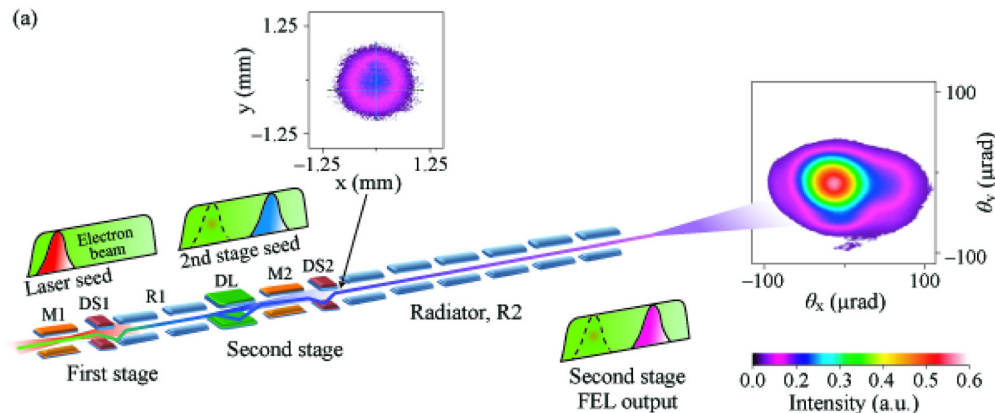
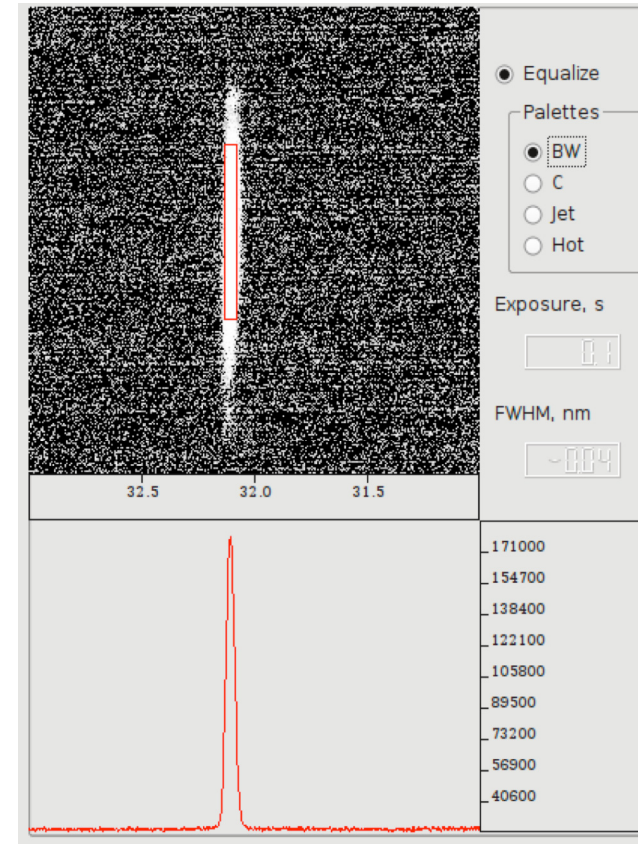
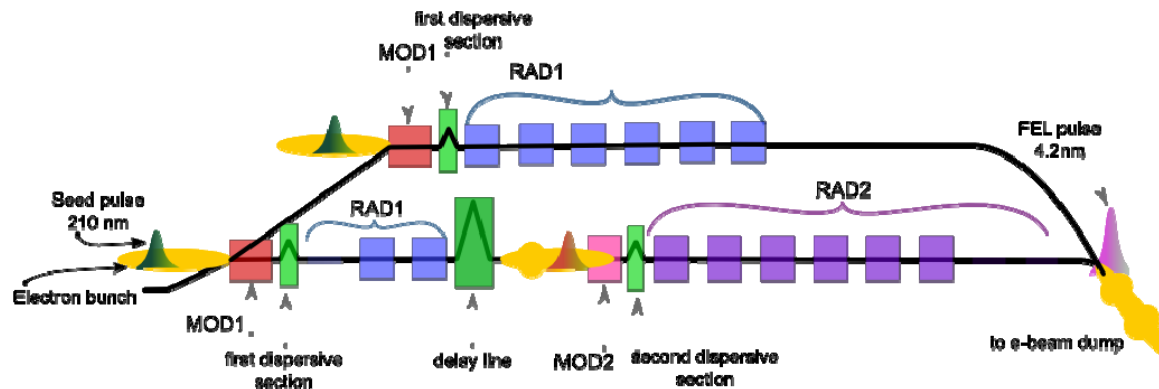
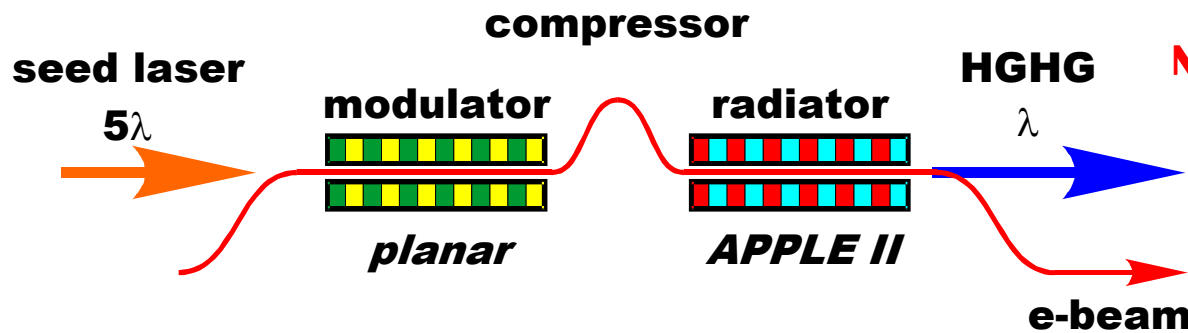


# IS A SASE FEL A LASER?



# A REAL X-RAYS LASER: EXTERNAL COHERENT SEEDING

Nature Photonics 6, 699–704 (2012)





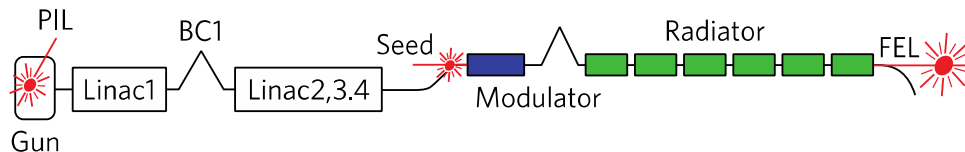
## First seeded free electron laser shines for users

Toni Feder

Citation: *Phys. Today* 65(11), 28 (2012); doi: 10.1063/PT.3.1784

## Highly coherent and stable pulses from the FERMI seeded free-electron laser in the extreme ultraviolet

E. Allaria *et al.*\*



## Two-stage seeded soft-X-ray free-electron laser

E. Allaria<sup>1</sup>, D. Castronovo<sup>1</sup>, P. Cinquegrana<sup>1</sup>, P. Craievich<sup>1†</sup>, M. Dal Forno<sup>1,2</sup>, M. B. Danailov<sup>1</sup>, G. D'Auria<sup>1</sup>, A. Demidovich<sup>1</sup>, G. De Ninno<sup>1,3</sup>, S. Di Mitri<sup>1</sup>, B. Diviacco<sup>1</sup>, W. M. Fawley<sup>1\*</sup>, M. Ferianis<sup>1</sup>, E. Ferrari<sup>1</sup>, L. Froehlich<sup>1</sup>, G. Gaio<sup>1</sup>, D. Gauthier<sup>1,3</sup>, L. Giannessi<sup>1,4\*</sup>, R. Ivanov<sup>1</sup>, B. Mahieu<sup>1,5</sup>, N. Mahne<sup>1</sup>, I. Nikolov<sup>1</sup>, F. Parmigiani<sup>1,2</sup>, G. Penco<sup>1</sup>, L. Raimondi<sup>1</sup>, C. Scafuri<sup>1</sup>, C. Serpico<sup>1</sup>, P. Sigalotti<sup>1</sup>, S. Spampinati<sup>1,3</sup>, C. Spezzani<sup>1</sup>, M. Svandrlik<sup>1</sup>, C. Svetina<sup>1,2</sup>, M. Trovo<sup>1</sup>, M. Veronese<sup>1</sup>, D. Zangrando<sup>1</sup> and M. Zangrando<sup>1,6</sup>

## First seeded free electron laser shines for users

A free electron laser (FEL) in Trieste, Italy, began producing 40- to 150-fs pulses at soft x-ray and extreme-UV wavelengths last year, and commissioning began this fall on a second FEL that will reach shorter wavelengths. Based on a conventional linac operating at 10 Hz and 1.2 GeV, the €160 million (\$210 million) FERMI@Elettra source is the first user facility to implement seeding. The result: high-gain light pulses that are coherent; are stable in intensity, photon energy, and bandwidth; and have tunable energy and polarization that can be switched from linear to circular.

In seeding, an external laser imprints its coherent field and narrow bandwidth on a relativistic electron beam as the electrons enter a series of undulators. The technique overcomes the drawbacks of relying, as most FELs do, on spontaneous electron organization. The Trieste beam shows a "spectacular degree of both transverse and longitudinal coherence and will allow experiments that could not be performed in any other manner," says MIT's William Barletta, a consultant for the FEL design team. "We can generate a detectable FEL signal at wavelengths down to about 4 nm," says Fulvio Parmigiani, coordinator of the FERMI@Elettra scientific program. But the shortest wavelength with sufficient intensity for experiments is about 20 nm.



ELETTRA-SINCROTRONE TRIESTE S.C.p.A.

The three beamlines at the Trieste FEL are dedicated to spectroscopy of gases and beams of mass-selected clusters of atoms or molecules; coherent dynamic imaging of such things as nanostructures and biological systems; and inelastic and elastic scattering experiments. Two additional beamlines for magnetic dynamics and pump-probe experiments are planned. The experimental hall is shown in the bottom photo; the top one shows an aerial view of the site.

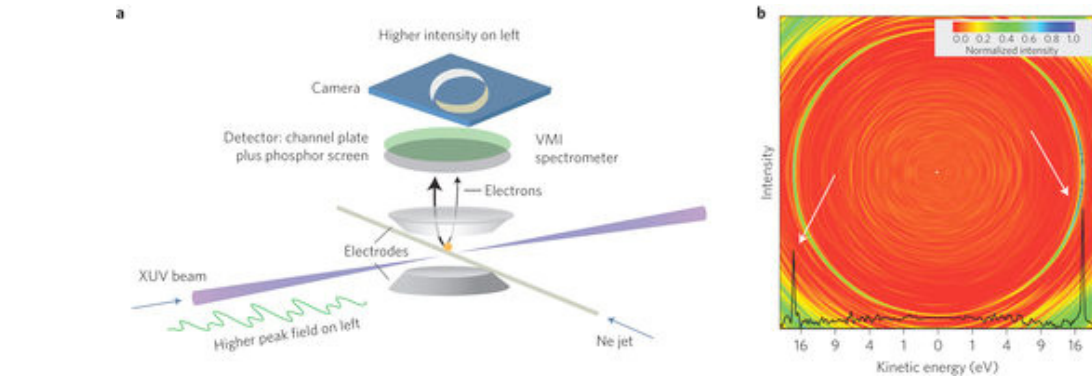


The second FEL is designed to get down to a fundamental emission of 4 nm, with harmonics providing even shorter usable wavelengths. The linac is also being upgraded to 50 Hz and 1.5 GeV next year, with hopes of going to 1.8 GeV in 2014. One aim of the upgrades is to provide sufficient intensity for experiments needing 1-nm radiation.

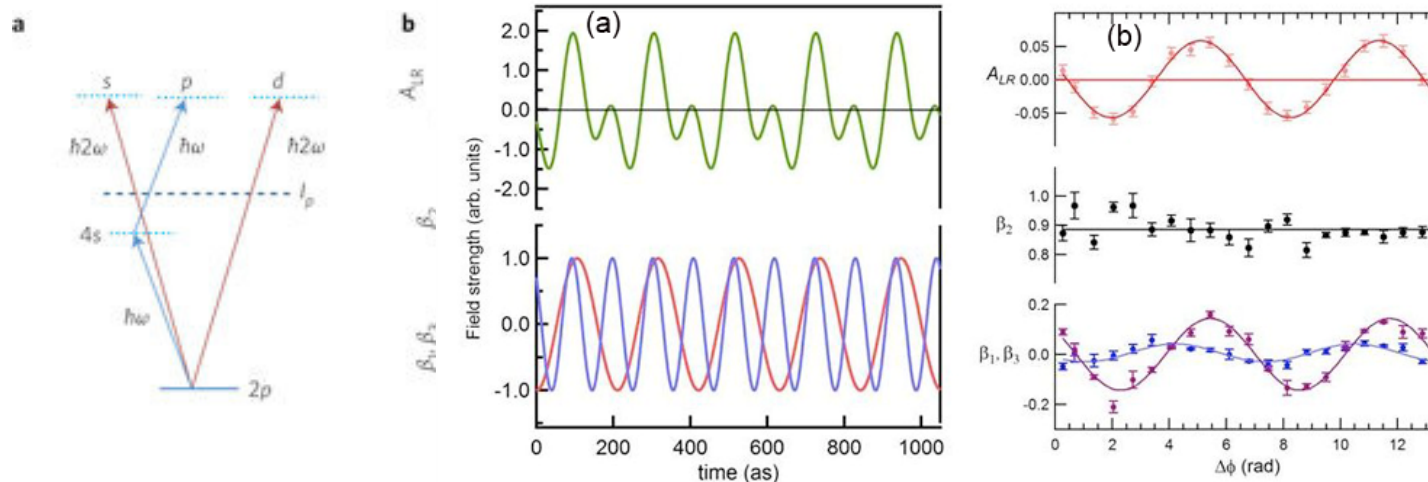
Says Barletta, "Most future machines in the soft x-ray region will have some seeded beams." The FEL in Trieste "is important to see how well seeding works." **Toni Feder**



# COHERENT CONTROL WITH A EUV FEL



Reproduced from K. Prince et al.,  
Nature Photonics 10, 176 (2016)

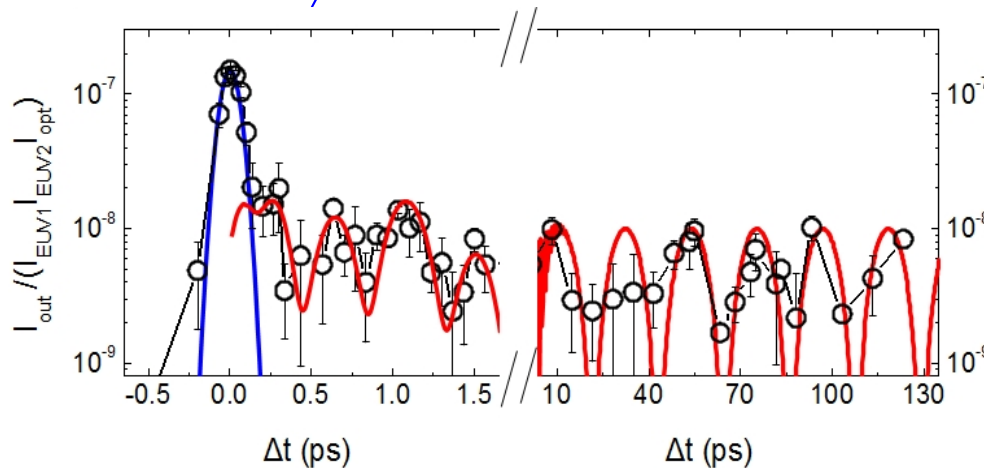
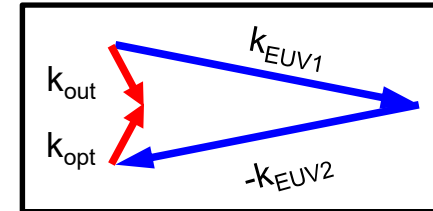
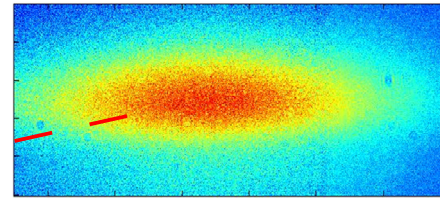
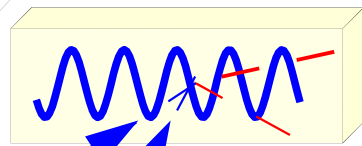


**Schematic drawing of the electric fields of the two harmonics (bottom) and of the total electric field due to the two wavelengths. (b)  $\beta$  parameters, which describe the angular distribution of the photoelectron patterns. These are the parameters of Legendre polynomials, and the even numbered parameters give rise to symmetric distributions, while the odd numbered parameters describe the asymmetry. The latter oscillate as a function of phase between the two wavelengths.**

# FEL-STIMULATED FOUR-WAVE-MIXING

**“phase matched” (3<sup>rd</sup> order) non-linear signal**

**EUV dynamic grating**

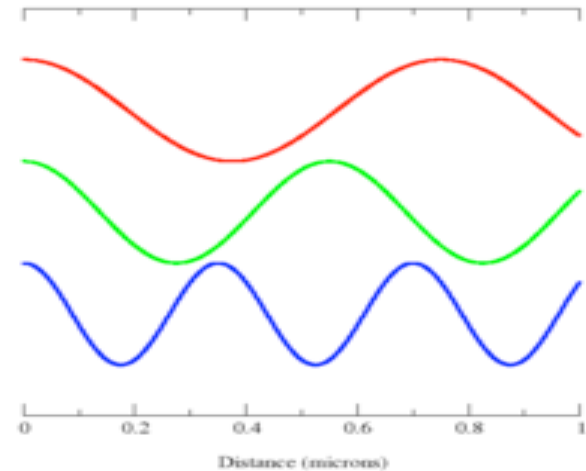
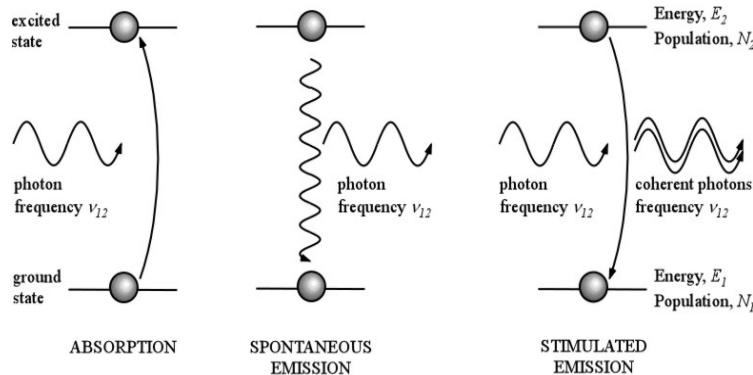
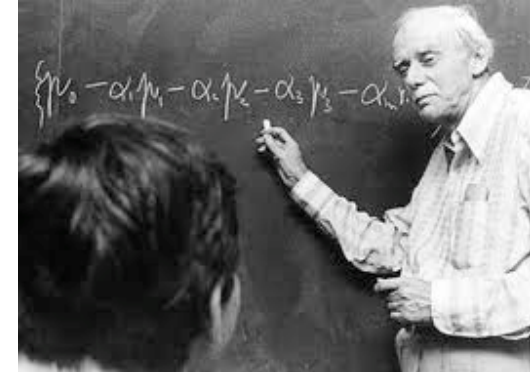
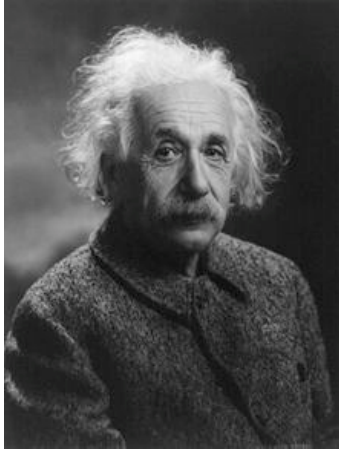


**Time-dependence!  
(impulsive stimulated  
scattering)**

**F. Bencivenga  
Nature 520, 205–208 (2015)**

**X-RAYS  
QUANTUM LIGHT  
AND  
OPTICS**

# QUANTUM OPTICS



In 1916, Einstein showed that Planck's radiation law could be derived from a semi-classical, statistical treatment of photons and atoms, which implies a relation between the rates at which atoms emit and absorb photons.

Dirac treated the interaction between a charge and an electromagnetic field as a small perturbation that induces transitions in the photon states, changing the numbers of photons in the modes, while conserving energy and momentum overall.

# WOULD BE THE X-RAY QUANTUM OPTICS

## A NEW REVOLUTION?



### Quantum optics: Controlling the light

#### 'Coherent States'

- Coherent states, or as they are sometimes called 'Glauber Coherent States' are the eigenstates of the annihilation operator

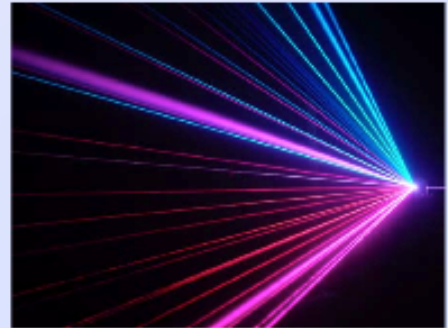
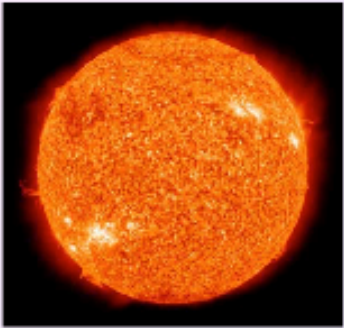
$$A|\alpha\rangle = \alpha|\alpha\rangle \quad \langle\alpha|\alpha\rangle = 1$$

Following the work of [Dirac](#) in [quantum field theory](#), [George Sudarshan](#), [Roy J. Glauber](#), and [Leonard Mandel](#) applied quantum theory to the electromagnetic field in the 1950s and 1960s to gain a more detailed understanding of photo-detection and the [statistics](#) of light.

- [coherent state](#) as a concept which addressed variations between laser light, thermal light, exotic [squeezed states](#), .
- Quantum entanglement

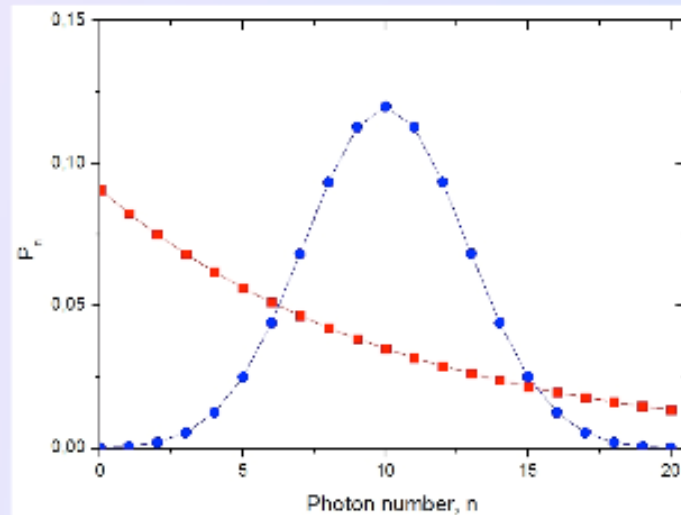
- Here  $\alpha$  can be any complex number
- i.e. there is a different coherent state for every possible choice of  $\alpha$
- (Roy Glauber, Nobel Prize for Quantum Optics Theory 2005)
- These states are not really any more 'coherent' than other pure states,
  - they do maintain their coherence in the presence of dissipation somewhat more efficiently

# QUANTUM OPTICS



$$P(n) = \frac{1}{\bar{n} + 1} \left( \frac{\bar{n}}{\bar{n} + 1} \right)^n$$

$$\Delta n = \sqrt{\bar{n} + \bar{n}^2}$$



$$P(n) = \frac{\bar{n}^n}{n!} e^{-\bar{n}}$$

$$\Delta n = \sqrt{\bar{n}}$$



# QUANTUM OPTICS

## Photon number states or Fock states

1. Introduce creation and destruction operators

$$\hat{X} = \left( \frac{\hbar}{2\omega} \right)^{1/2} [a^+ + a] \quad \hat{P} = i \left( \frac{\hbar\omega}{2} \right)^{1/2} [a^+ - a]$$

$$H = 1/2 (\hat{P}^2 + \omega^2 \hat{X}^2) = \hbar\omega (a^+ a + 1/2) = \hbar\omega (\hat{n} + 1/2)$$



V. Fock

1898-1974

2. Fock states is eigen states of energy operator.  
Therefore a photon is the single excitation of oscillator (mode)

$$E_n = \hbar\omega \left( n + \frac{1}{2} \right)$$

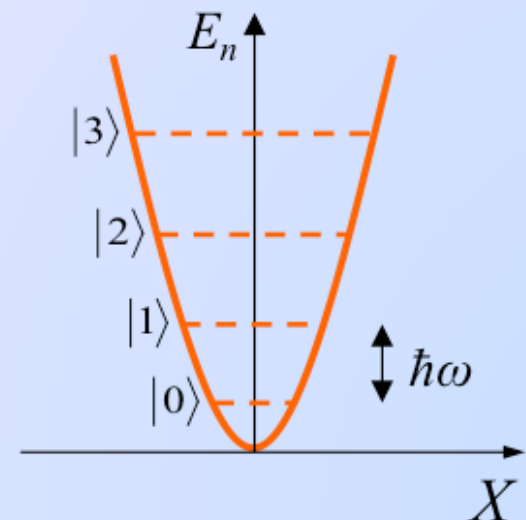
3. Simple math with operators:

$$a^+ |n\rangle = \sqrt{n+1} |n+1\rangle$$

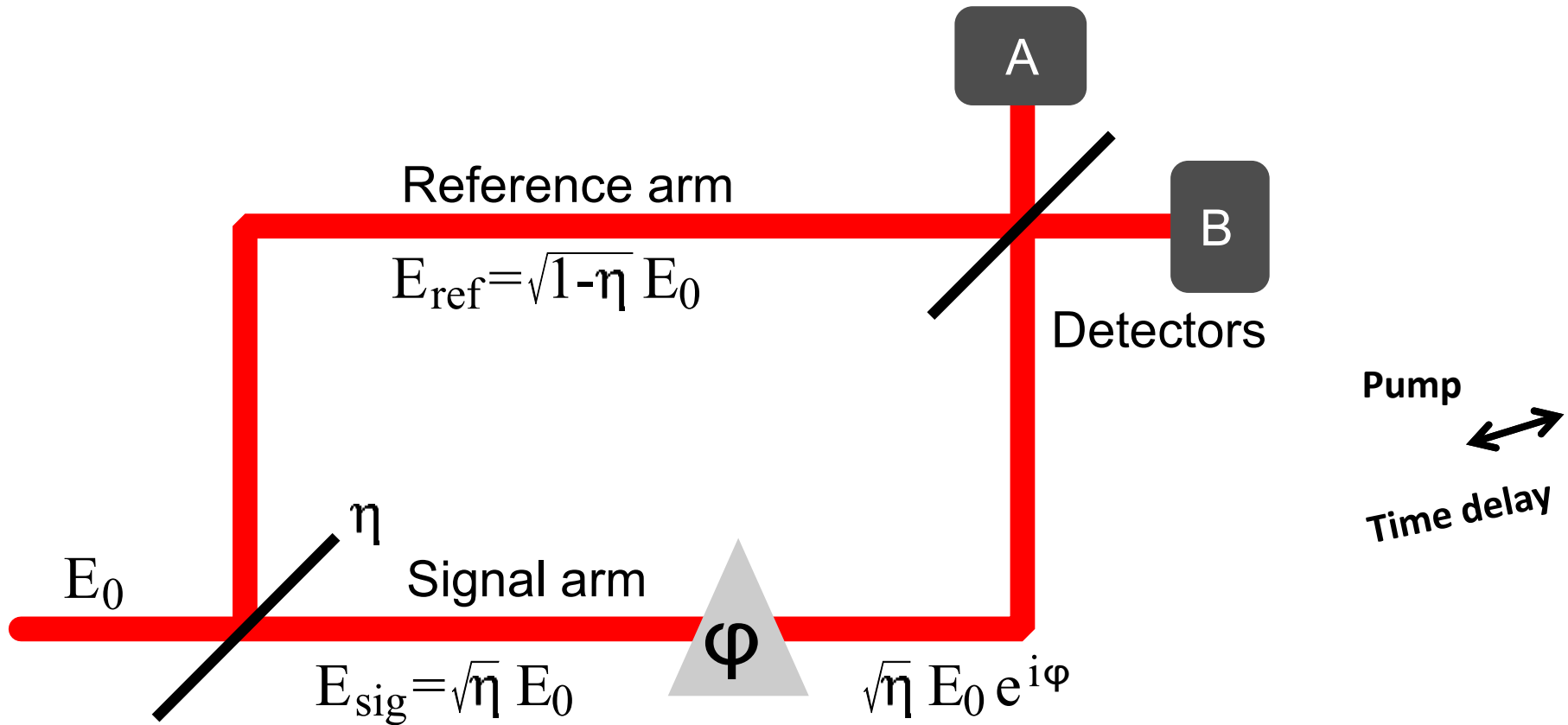
$$a |n\rangle = \sqrt{n} |n-1\rangle$$

$$a |0\rangle = |0\rangle$$

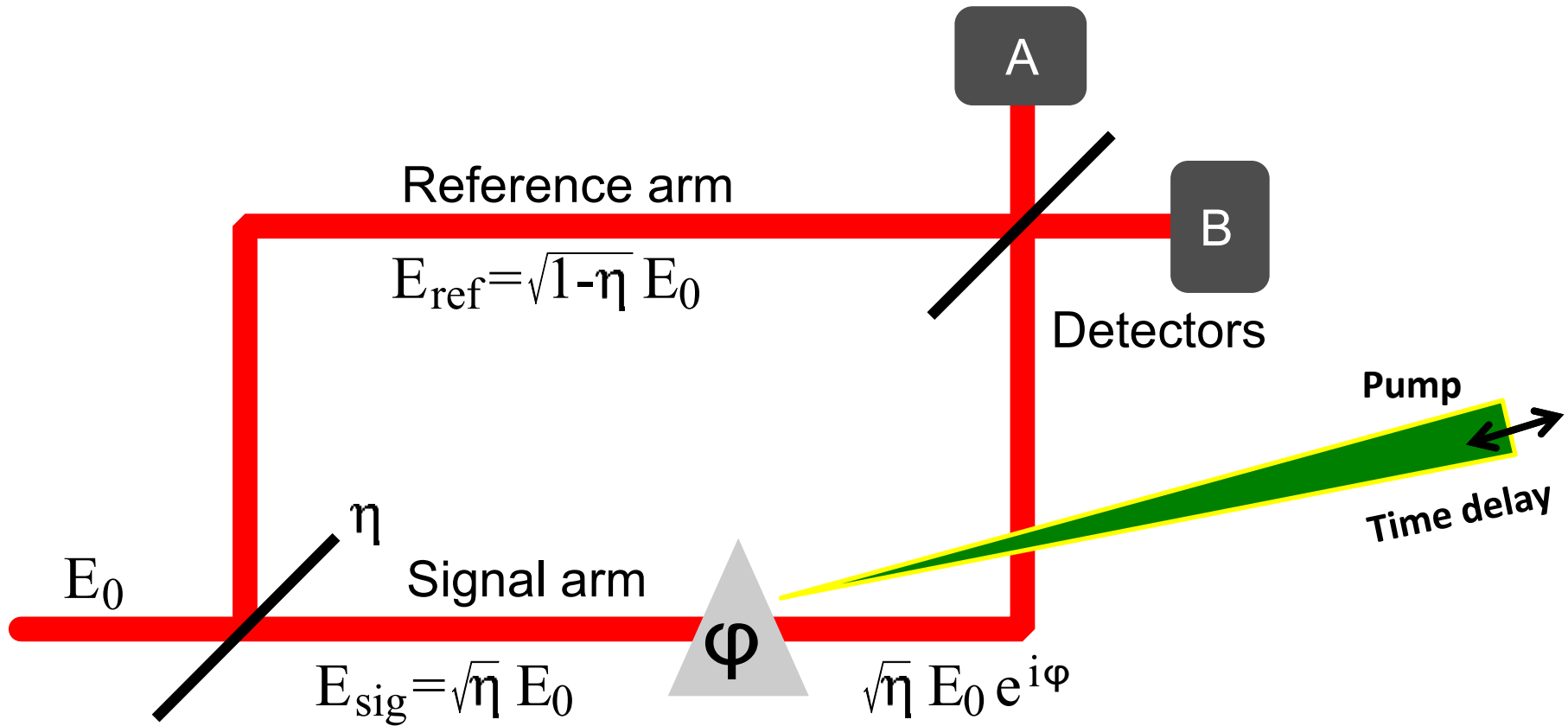
$$a^+ a |n\rangle = \sqrt{n} a^+ |n-1\rangle = n |n\rangle$$



# QUANTUM OPTICS SPECTROSCOPY

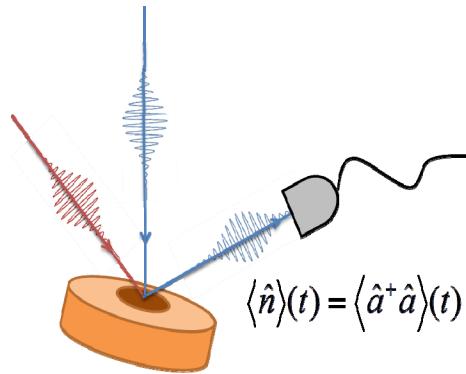


# QUANTUM OPTICS SPECTROSCOPY



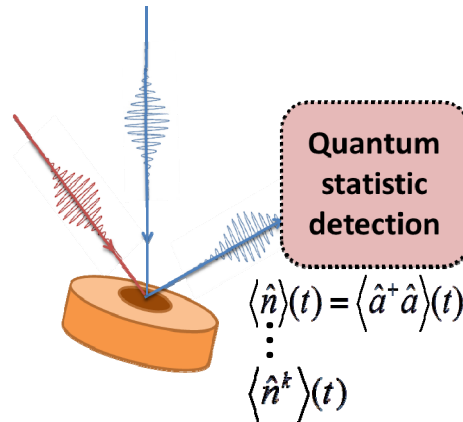
# QUANTUM OPTICS SPECTROSCOPY

## 1) Standard Pump-Probe



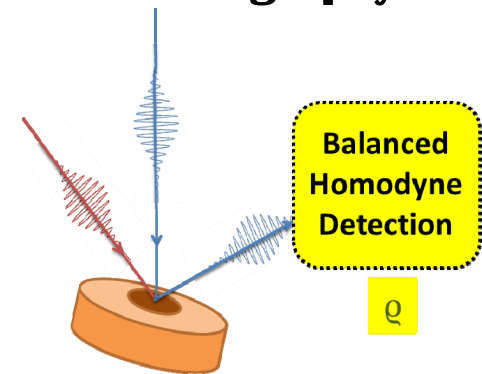
- Time domain study on collective atomic vibrations in transparent materials

## 2) Pump-Probe quantum statistic reconstruction



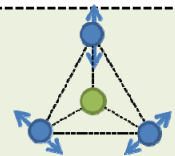
- Disclosing fluctuations of lattice atomic positions by non-equilibrium optical experiments

## 3) Pump-Probe quantum state tomography

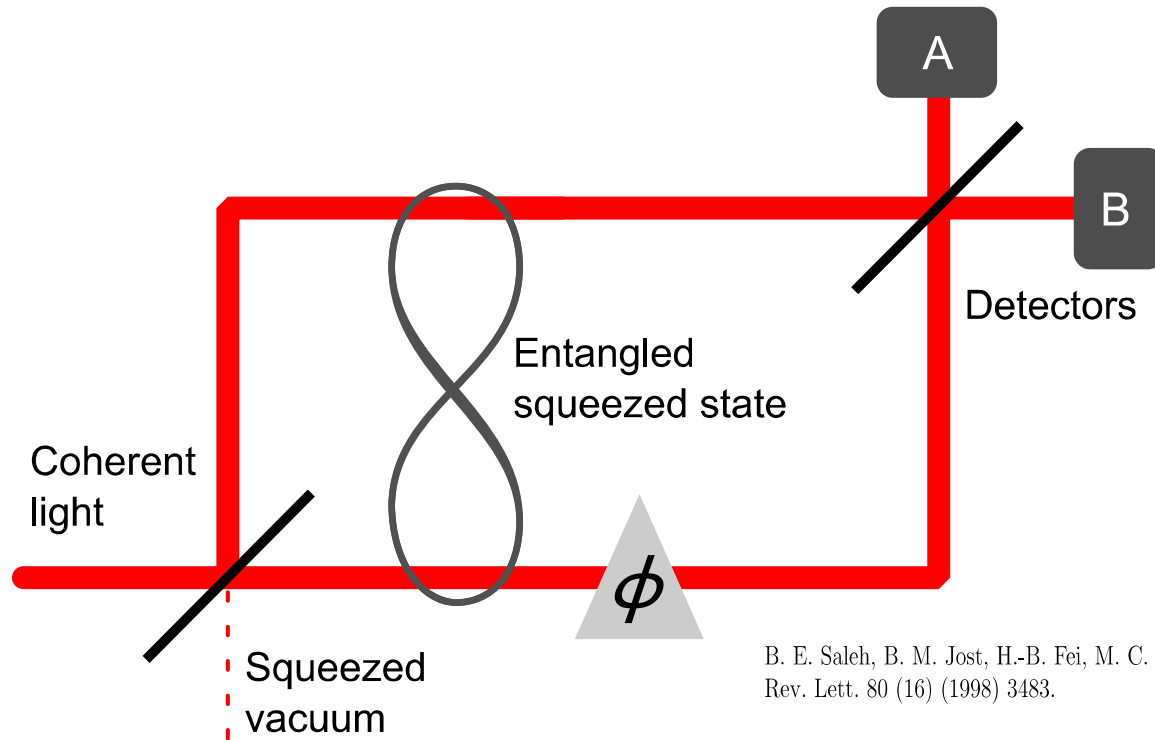


- Homodyne detection + pump probe, preliminary results

**Fluctuations of out-of-equilibrium atomic displacements**



# IS THE QFEL LIGHT SUITABLE FOR OBSERVING ENTANGLED STATES IN MATTER?



B. E. Saleh, B. M. Jost, H.-B. Fei, M. C. Teich, Entangled-photon virtual-state spectroscopy, Phys. Rev. Lett. 80 (16) (1998) 3483.

F. Wolfgramm, C. Vitelli, F. A. Beduini, N. Godbout, M. W. Mitchell, Entanglement-enhanced probing of a delicate material system, Nat. Photon. 7 (2013) 28–32.

W. Wasilewski, K. Jensen, H. Krauter, J. J. Renema, M. Balabas, E. S. Polzik, Quantum noise limited and entanglement-assisted magnetometry, Phys. Rev. Lett. 104 (13) (2010) 133601.

D. G. Monticone, K. Katamadze, P. Traina, E. Moreva, J. Forneris, I. Ruo-Berchera, P. Olivero, I. Degiovanni, G. Brida, M. Genovese, Beating the Abbe diffraction limit in confocal microscopy via nonclassical photon statistics, Phys. Rev. Lett. 113 (14) (2014) 143602.

O. Schwartz, D. Oron, Improved resolution in fluorescence microscopy using quantum correlations, Phys. Rev. A 85 (3) (2012) 33812.

C.-M. Li, N. Lambert, Y.-N. Chen, G.-Y. Chen, F. Nori, Witnessing quantum coherence: from solid-state to biological systems, Sci. Rep. 2 (2012) 885.

**PAPER**

## What defines the quantum regime of the free-electron laser?

Peter Kling<sup>1,2,4</sup>, Enno Giese<sup>2,4</sup>, Rainer Endrich<sup>1,2</sup>, Paul Preiss<sup>1,2</sup>, Roland Sauerbrey<sup>1</sup> and Wolfgang P Schleich<sup>2,3</sup>

The current development of FELs focuses on the X-ray regime of radiation as exemplified by the LCLS at SLAC in Stanford [5] or the European XFEL at DESY in Hamburg [6]. With decreasing wavelengths, the quantum mechanical recoil which is proportional to the wave number of the laser field increases and the emergence of a domain where the discreteness of the momentum does play a role for the FEL dynamics is evident. This new regime of FEL-operation, the so-called quantum regime or Quantum FEL, was theoretically predicted by Bonifacio *et al* [7]. It is expected that a Quantum FEL displays better radiation properties such as a narrower linewidth and better temporal coherence in comparison to its classical counterpart [8]. Even though an experimental realization is still far from reach, due to progress in the fields of accelerator and laser physics it might be possible within the future.

[7] Bonifacio R, Piovela N and Robb G R M 2009 *Fortschr. Phys.* **57** 1041–51

[8] Bonifacio R, Piovela N and Robb G R M 2005 *Nucl. Instrum. Methods Phys. Res. A* **543** 645–52

through the Thomas-Fermi approximation, as followed, for instance, in Ref. 2.

The purpose of this paper is to show that a new insight into the fascinating FEL dynamics can be gained by formulating the quantum mechanics of the FEL in the language of quantum field theory (QFT). I shall demonstrate that in the case of a very large number of electrons  $N$ —which is always the case in practice—the FEL dynamics is governed by the electromagnetic field self-consistently generated by  $N$  copies of electron itself. Furthermore, beyond this kind of Thomas-Fermi approximation, one can also compute the size- $O(1/\sqrt{N})$ —of the quantum fluctuations and, under certain conditions, establish a perturbative scheme for their computation. Turning to the “Thomas-Fermi” approximation, a rather new computational strategy will also be presented.

EUROPHYSICS LETTERS

*Europhys. Lett.*, **69** (1), pp. 55–60 (2005)

DOI: 10.1209/epl/i2004-10308-1

## A quantum model for collective recoil lasing

R. BONIFACIO<sup>1</sup>, M. M. COLA<sup>1</sup>, N. PIOVELLA<sup>1</sup> and G. R. M. ROBB<sup>2</sup>

\* \* \*

One of us (RB) has to acknowledge a big mistake he made almost 20 years ago when G. PREPARATA, a well-known field theorist, presented to him his general “Quantum Field Theory of a Free Electron Laser” [3]. RB did not understand this work thinking it was incorrect. On the contrary, Preparata’s theory was perfectly correct, as recognized in this letter, which is dedicated to his memory.

in terms of the operators  $a_k$  and  $a_k^\dagger$  the Hamiltonian (2.1) is easily seen to become

$$H = \frac{1}{2\bar{p}} \sum_k k^2 a_k^\dagger a_k + i \frac{\bar{p}^{1/2}}{N^{1/2}} \left[ a^\dagger \sum_k a_k a_{k-1}^\dagger - a \sum_k a_k^\dagger a_{k-1} \right] . \quad (2.4)$$

This Hamiltonian defines a  $(1+1)$ -dimensional QFT in the following way: Define the complex scalar field

$$\psi(t, \phi) = \sum_k a_k \frac{e^{ik\phi}}{(2\pi)^{1/2}} , \quad (2.5)$$

with the equal-time commutation relations

$$[\psi(t, \phi), \psi^\dagger(t, \phi')] = \delta(\phi - \phi') . \quad (2.6)$$

1 January 2005



# ***Future Scenario***

- **The way to produce fully coherent soft X-ray radiation is paved.**
- **Tunability**
- **Variable polarization**
- **Full coherence**
- **High repetition rate**

**The future scenario**

**Coherent X-ray Optics**

**Quantum X-ray optics**

**Stroboscopic phase tomography**

**An extraordinary effort is  
needed to develop a suitable  
science program**

# **ACKNOWLEDGMENT**

**I am indebt with many colleagues and friends, in particular with,**

**Colleagues at LBNL**

**W. Barletta**

**W. Fawley**

**M. Zolotorev**

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**C. Pellegrini**

**R. Schoenleine**

**LANL**

**S. Milton**

**and all the FERMI team at the Elettra Sincrotrone Trieste**

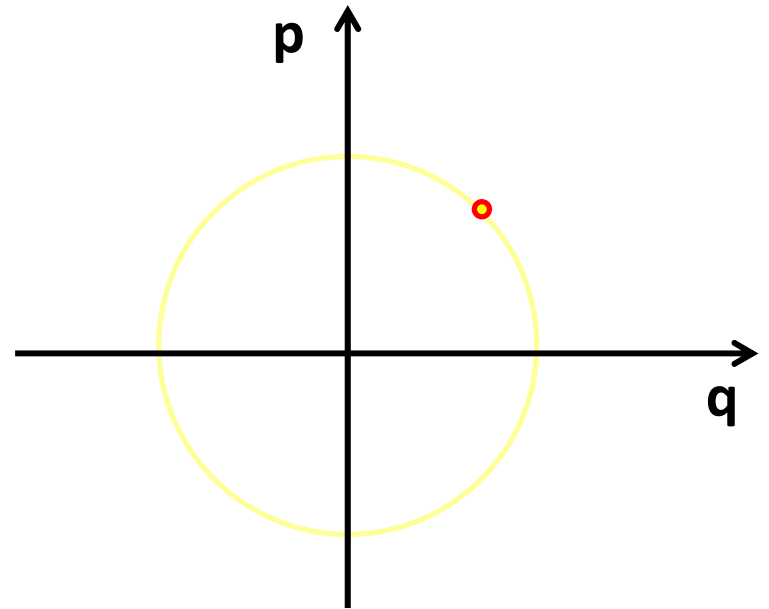
**THIS TUTORIAL IS DEDICATED TO RODOLFO BONIFACIO  
AND TO HIS SCIENTIFIC LEGACY**



# Pump&Probe quantum statistic reconstruction

## PHONONS DESCRIPTION

### Quantum harmonic oscillator



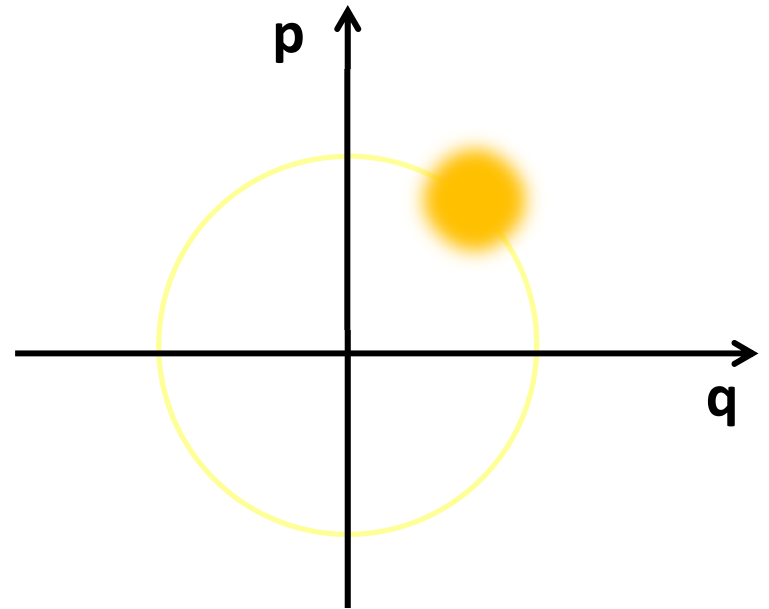
# Pump&Probe quantum statistic reconstruction

## PHONONS DESCRIPTION

### Quantum harmonic oscillator

#### Coherent state

$$\sigma_q \sigma_p = \hbar/2$$



# Pump&Probe quantum statistic reconstruction

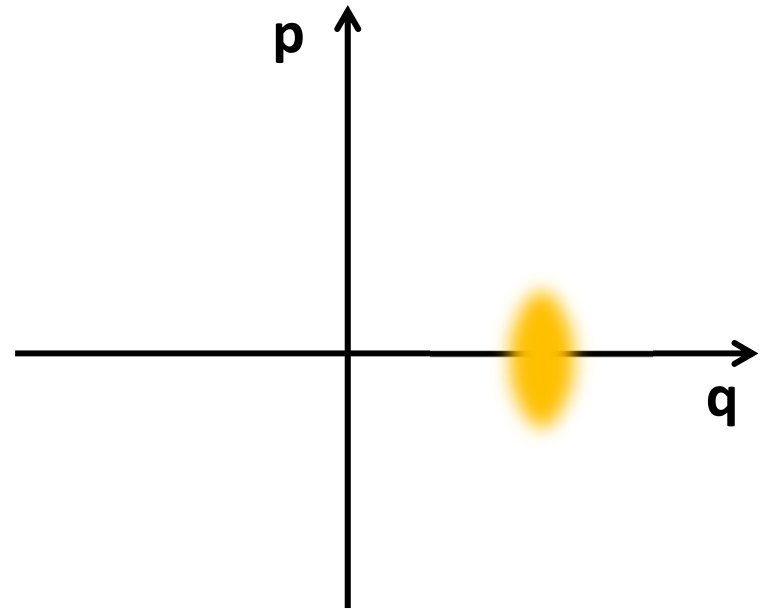
## PHONONS DESCRIPTION

### Quantum harmonic oscillator

#### Squeezed state

$$\sigma_q \sigma_p = \hbar/2$$

$$\sigma_q < \sigma_p$$



# Pump&Probe quantum statistic reconstruction

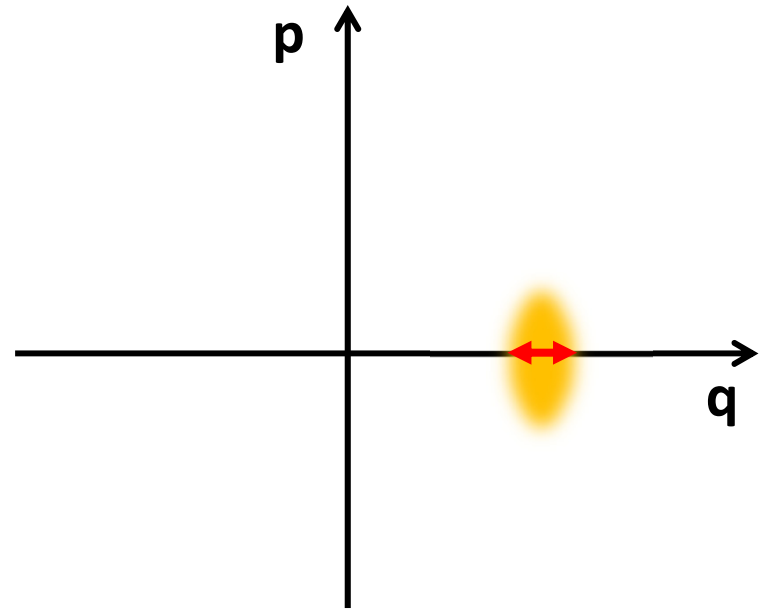
## PHONONS DESCRIPTION

### Quantum harmonic oscillator

#### Squeezed state

$$\sigma_q \sigma_p = \hbar/2$$

$$\sigma_q < \sigma_p$$





# Pump&Probe quantum statistic reconstruction

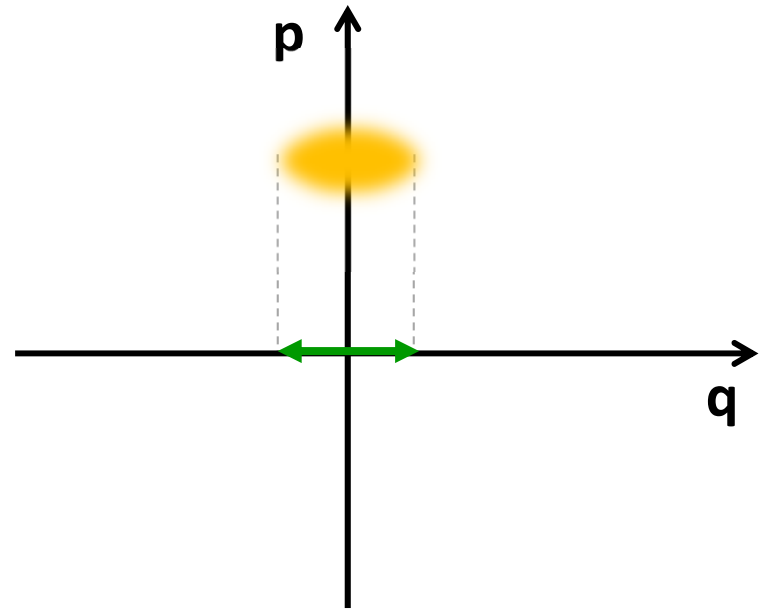
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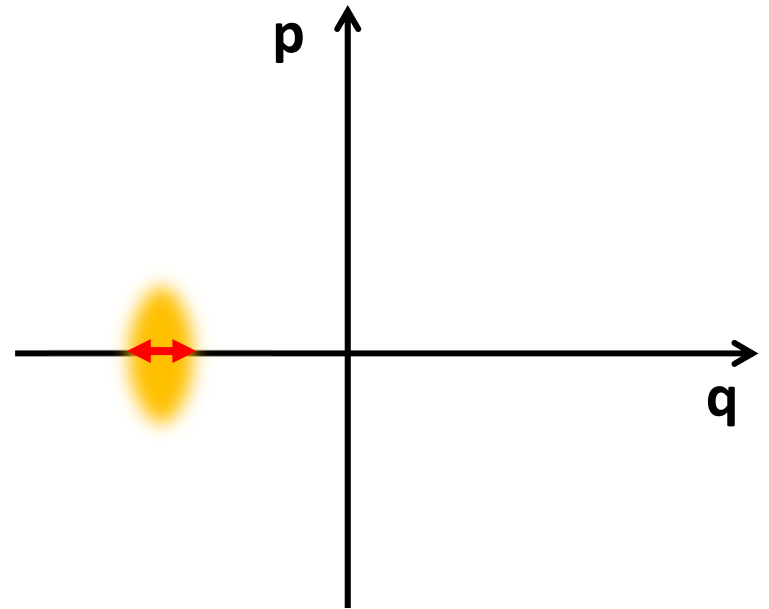
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# Pump&Probe quantum statistic reconstruction

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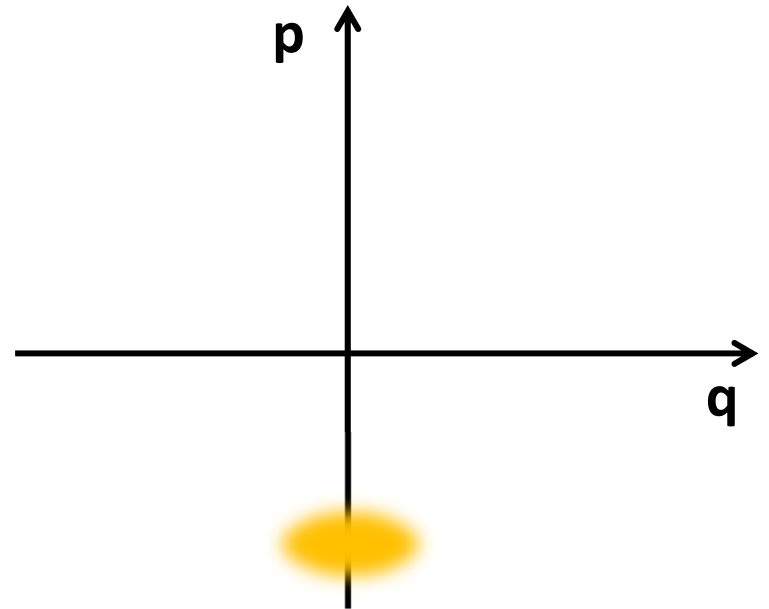
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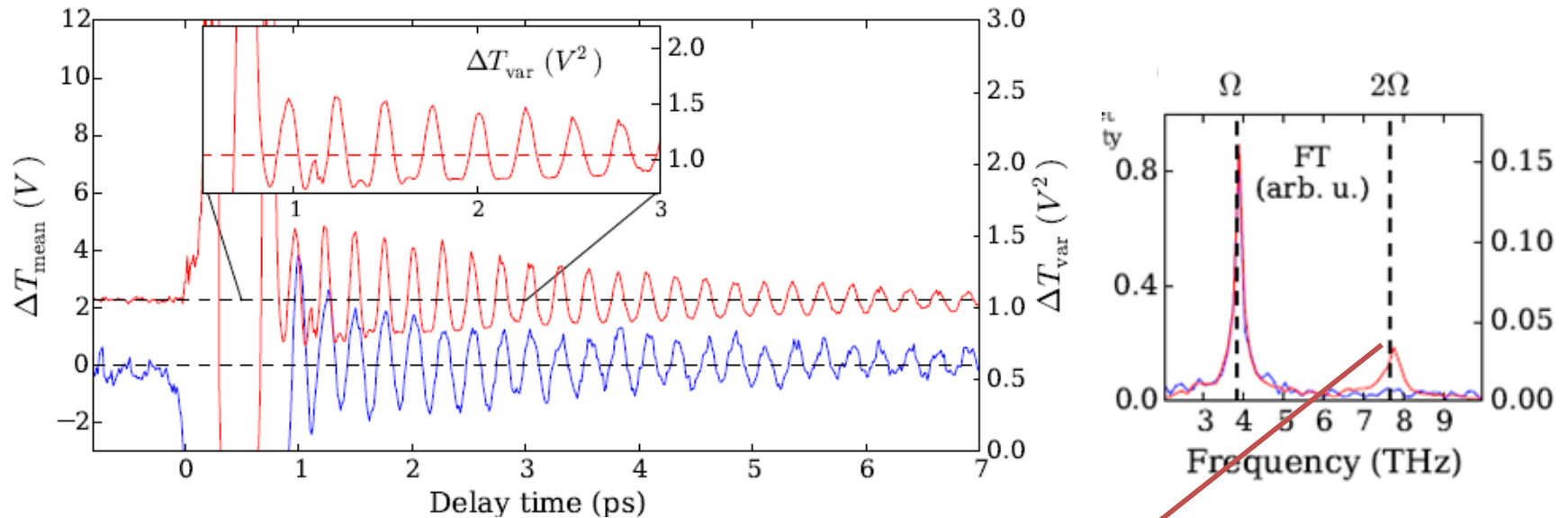
$$\omega(q) = \omega_0$$

$$\omega_\sigma = 2\omega_0$$



# Pump&Probe quantum statistic reconstruction

## EXPERIMENTAL RESULTS

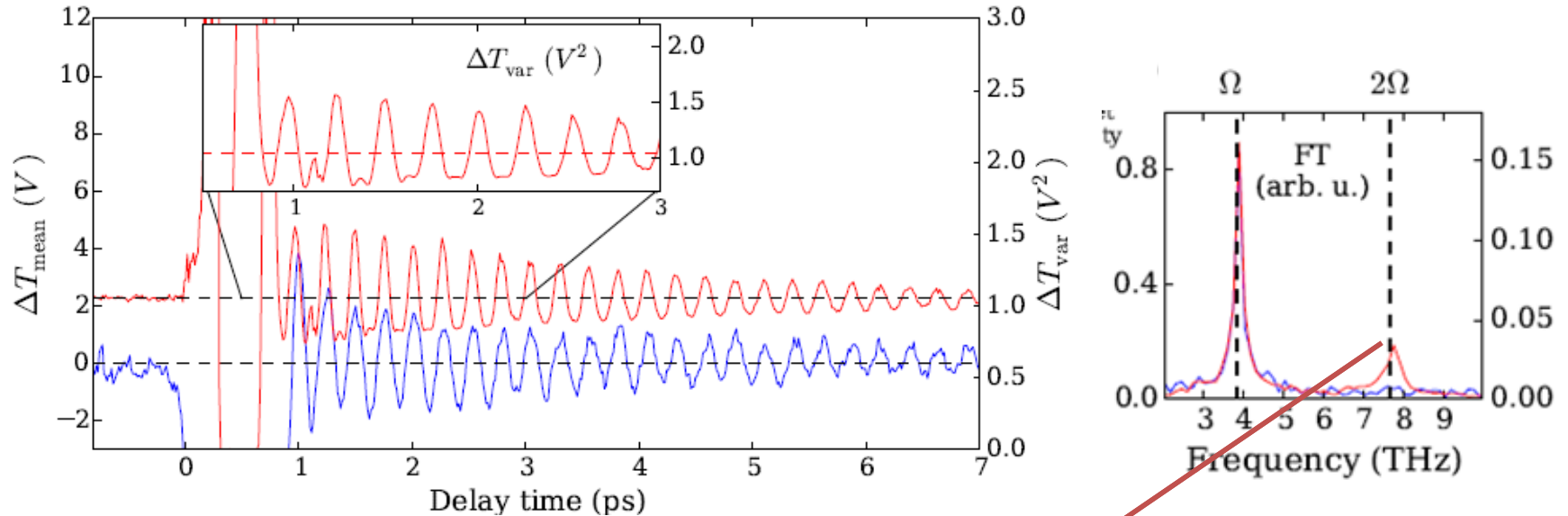


?

M. Esposito et al. (2015)  
Accepted in *Nature Communication*

# Pump&Probe quantum statistic reconstruction

## EXPERIMENTAL RESULTS



Indication  
of phonon  
squeezing

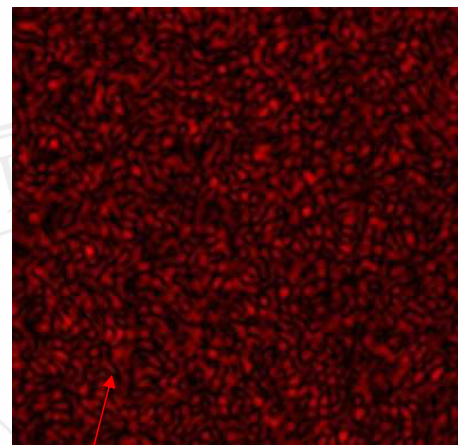
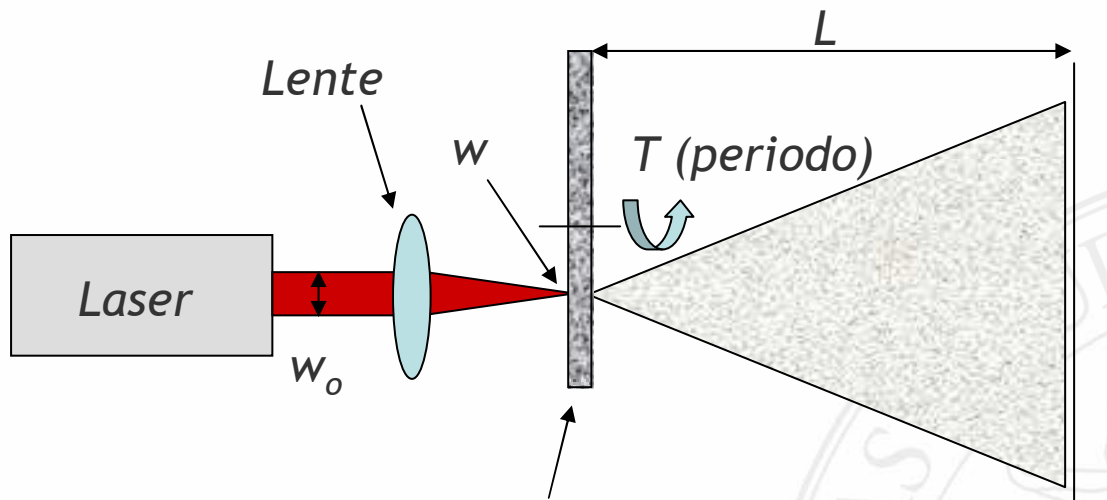
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MEASUREMENT OF THE STATISTICAL DISTRIBUTION OF GAUSSIAN AND LASER SOURCES\*

F. T. Arecchi

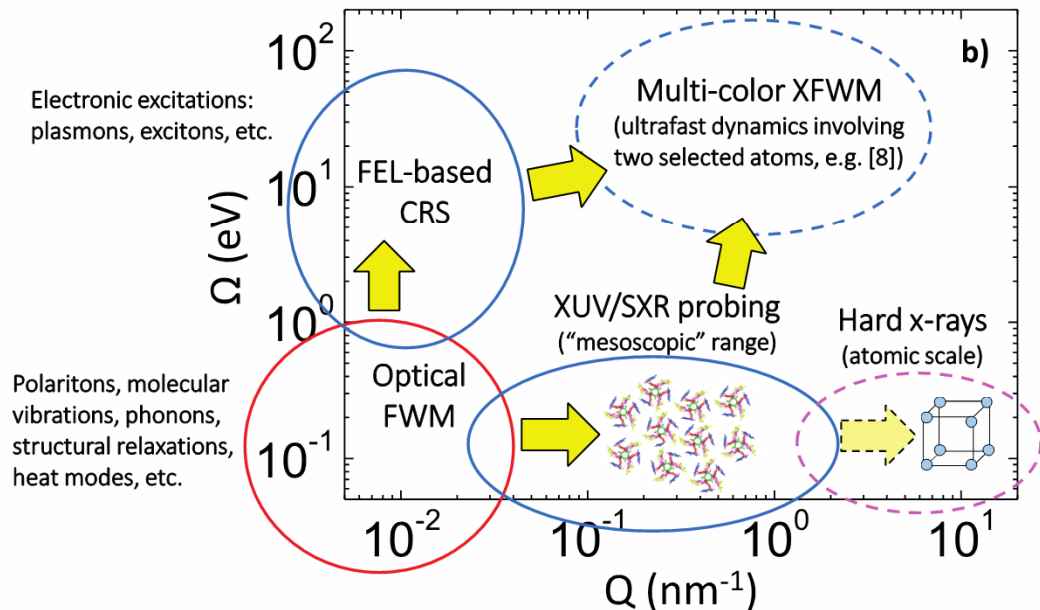
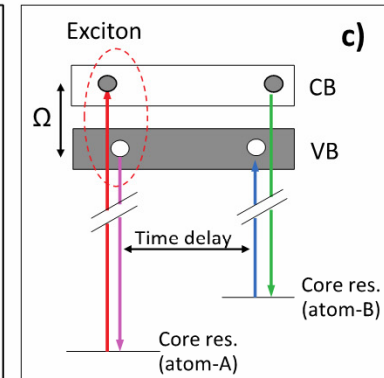
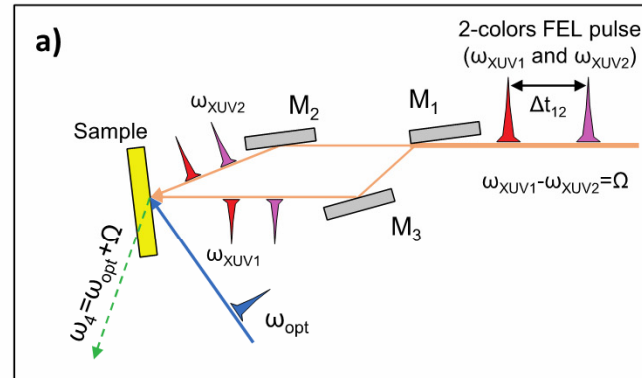
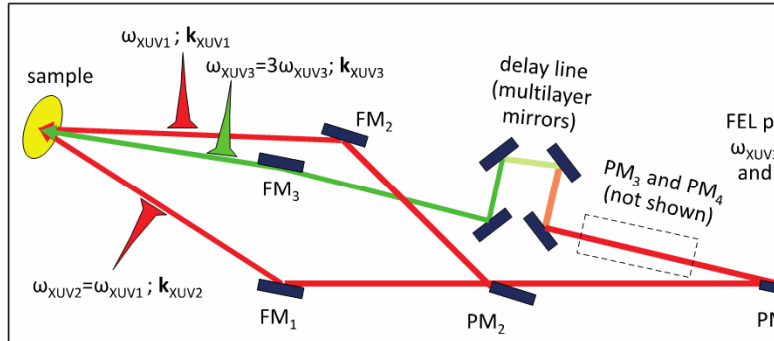
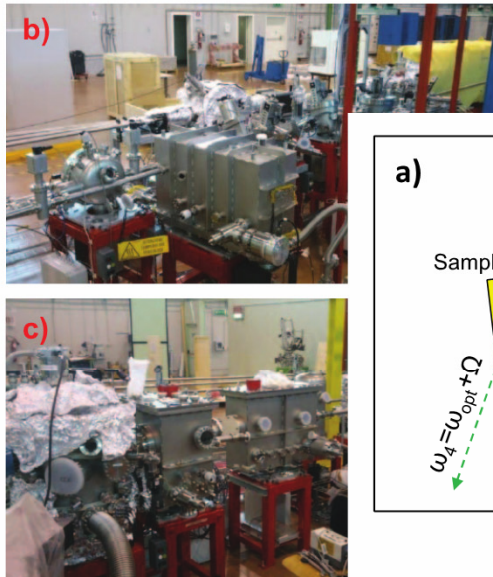
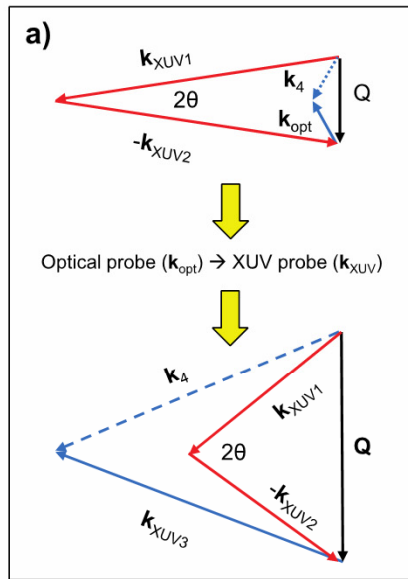
Laboratori Centro Informazioni Studi Esperienze, Milano, Italy,  
and Istituto di Fisica dell'Università, Milano, Italy

(Received 18 November 1965)



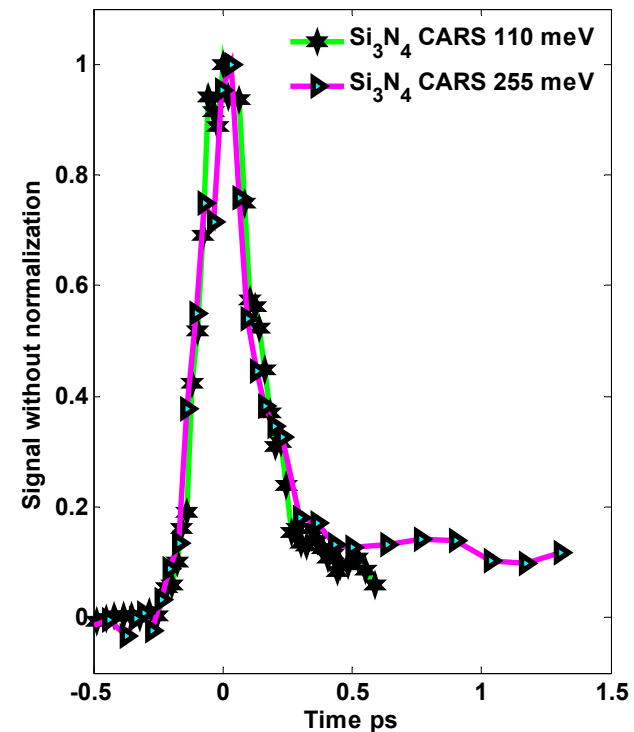
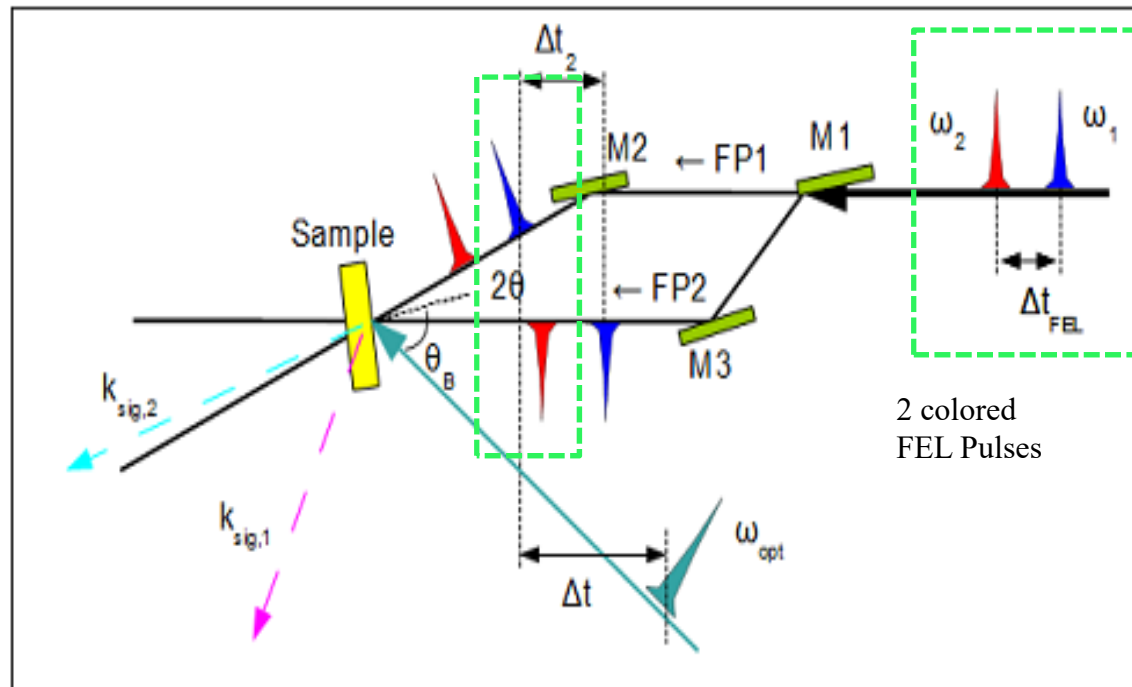
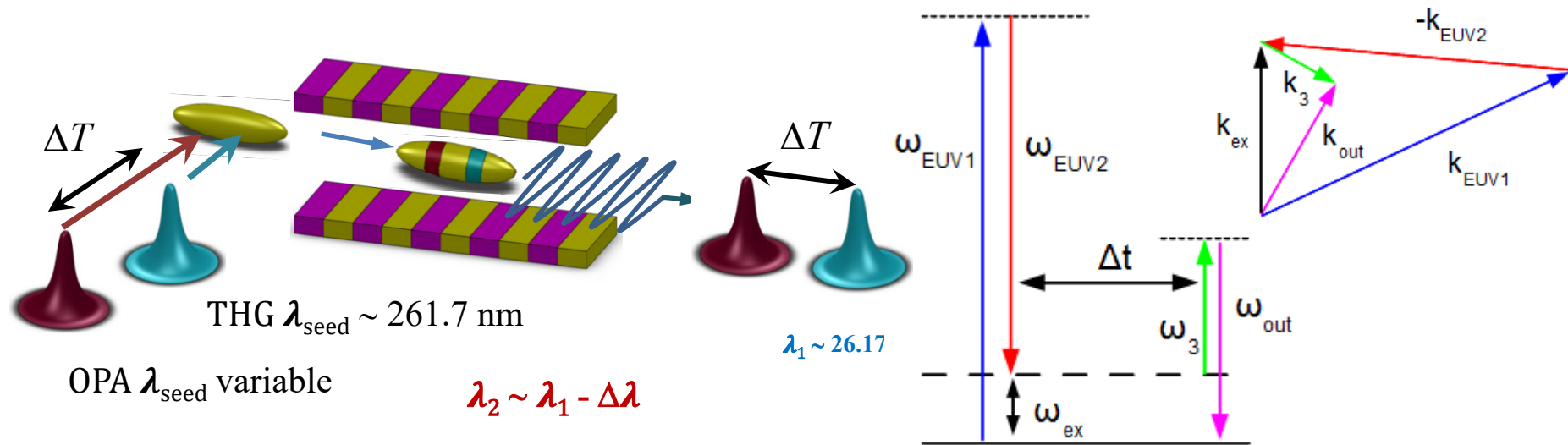
$$P(I) = \frac{1}{\bar{I}} e^{-\frac{I}{\bar{I}}}$$

# CLASSICAL TEMPORAL (LONGITUDINAL) COHERENCE



Reproduced from: Filippo Bencivena and Claudio Masciovecchio

# FERMI BASED COHERENT ANTI-STOKES RAMAN SCATTERING



Adapted from F. Bencivenga, C. Masciovecchio