A. Zholents

Towards a 5th Generation Light Source, October 1-2, 2010
... all great discoveries in science come on heels of innovations in technology and experimental tools
Attosecond x-ray pulses is a powerful tool for addressing Grand Challenges in Science and BES Research Needs

- How do we control materials and processes at the level of electrons?
- How do we design and perfect atom-and energy-efficient synthesis of new forms of matter with tailored properties?
- How do remarkable properties of matter emerge from complex correlations of atomic and electronic constituents and how can we control these properties?
- Can we master energy and information on the nanoscale to create new technologies with capabilities rivaling those of living systems?
- How do we characterize and control matter away—especially very far away—from equilibrium?
Attosecond XUV pulses had been obtained

Joint PRESS RELEASE of
Max Planck Institute of Quantum Optics
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Munich Centre for Advanced Photonics

Ultrafast Look into Atoms and Molecules

New record in ultrafast met
Ludwig-Maximilians-Un

80 attoseconds !

Quantum Optics and the
pulses lasting only 80

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Probing intra-atomic electron motion by attosecond absorption spectroscopy.


, Avalon, 10/2010
Electron beam-based sources of ultrashort x-ray pulses

Where we are?

State-of-the-art for a short x-ray pulse generation using FELs

Where we are going?

Generation of attosecond x-ray pulses
Short EUV/x-ray pulses are routinely produced at FLASH (10 – 70 fs), SCSS (~ 30 fs), LCLS (<10 – 80 fs)

All future x-ray FEL projects consider ultra-short x-ray pulse capabilities
**Simple thoughts**

- Short electron bunch radiates short x-ray pulse
- For a given peak current short bunch holds a low charge
- With low charge one may expect better 6D electron bunch brightness

\[ B = \frac{2Q}{\gamma^3 \varepsilon_x \varepsilon_y \varepsilon_z} \]

- Nearly FTL pulses from SASE

<table>
<thead>
<tr>
<th>( Q = 1 \text{ nC} )</th>
<th>( B = 0.3 \text{ nC/\mu m}^3 )</th>
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<tbody>
<tr>
<td>( \gamma \varepsilon_{x,y} \approx 1 \mu m )</td>
<td>( \gamma \varepsilon_z \approx \sigma_z (3 \text{ keV})/mc^2 = 6.5 \mu m )</td>
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<table>
<thead>
<tr>
<th>( Q = 0.02 \text{ nC} )</th>
<th>( B = 4.5 \text{ nC/\mu m}^3 )</th>
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<tr>
<td>( \gamma \varepsilon_{x,y} \approx 0.15 \mu m )</td>
<td>( \gamma \varepsilon_z \approx \sigma_z (1 \text{ keV})/mc^2 = 0.4 \mu m )</td>
</tr>
</tbody>
</table>

P. Emma, LBNL workshop, 08/2010

Brightness defines the gain length \( 1/L_G \sim B \)

20-pC bunch operation at LCLS\(^1\)

X-ray pulse duration should be <10 fs, but no direct measurement yet possible

soft x-ray \(h\omega=840\) eV

Simulated signal
Simulated bunch length

Photo-diode signal on OTR screen after BC2

1) Y. Ding et. al, PRL 2009
Soft x-rays at 1.5 nm (simulations for LCLS)\(^1\)

At undulator entrance, 4.3 GeV, Laser heater off

1) Y. Ding, LBNL workshop, 08/2010

Actual measurements qualitatively confirm simulations. Direct measurement of ultra-short x-ray pulse duration remains to be difficult.
At 1.5 Å FEL performs well at full compression (slippage just right)

**Measurement**

Gain length = 2.74 m

**Simulation at 13.6 GeV**

\[ \text{rms} = 0.316 \mu \text{m} \]

\[ L_1 = -22 \text{ deg} \]

**140 \( \mu \)J FEL energy**

**Deflecting angle**

\[ E = \text{Loss} = 6.24 + 0.31 \text{ MeV (0.14 mJ), 01-JUN-2009 23:32:29 (13.50 GeV)} \]

N-photon = 1.05e+11
E-photon = 8.10 keV

J. Frish et. al., talk at FEL’09

Y. Ding, LBNL workshop, 08/2010

Not short enough for FT limited pulse
SUB-FEMTOSECOND X-RAY PULSES USING THE SLOTTED FOIL METHOD
P. Emma, M. Cornacchia, K. Bane, Z. Huang, H. Schlarb, G. Stupakov, D. Walz, PRL, 2004

Linac Coherent Light Source, SLAC

135 MeV  BC1  250 MeV  BC2  4.3 GeV  14 GeV

gun

ΔE

energy chirp

X-ray pulse width \( \sim 400 \) asec
Bunch arrival time jitter \( \sim 50 \) fs
\( \sim 10^9 \) photons/pulse -> 7 GW

Only bright spot will lase

Simulation using Elegant

Courtesy P. Emma
Double X-Ray Pulses from a Double-Slotted Foil

**FEMTOSECOND X-RAY PULSES IN THE LCLS USING THE SLOTTED FOIL METHOD**

- Precise controlled time delay between x-ray pump and x-ray probe pulses

**Time (fs)**

- 0-150 fs
- 2 fs

**Power (GW)**

- 10
- 5
- 0

**0.25 mm**

**0-6 mm**

pulses not coherent

**2-Pulse Production with 2 slots**

Courtesy P. Emma
Initial results (very preliminary), June 17, 2010

Over-compressed bunch introduces $e^-$ energy chirp on dump screen...

Wider slot gives more energy

Courtesy P. Emma
Pump – probe studies using ultra-fast x-ray pulses

Pellet hits a strawberry

Stop-motion photography
E. Muybridge, 1878
Options for experiment utilizing synchronized pump and probe signals when electron bunch arrival time has a “large” jitter

- Use double slotted foil
- Split single x-ray pulse into two and adjust delay

Create pump signal using coherent undulator radiation and adjust delay\(^1\) (in case of an ultra-short e-bunch, \(\sim 1\) fs)

1) U. Fruhling et al., Nature photonics, 3, 523(2009); also considered by UCLA group
Seeded FELs naturally possess precise synchronization
if electron bunch length > laser pulse + jitter

**Current-enhanced SASE FEL --> same conclusion**

Require synchronization of the seed and pump lasers ¹)

¹) J. Kim et. al., *Nature Photonics*, 2, 733, 2008;
Attosecond pulse generation via electron interaction with a few cycle carrier-envelop phase stabilized laser pulse

Basic idea:
Take an ultra-short slice of electrons from a longer electron bunch to produce a dominant x-ray radiation

Small jitter in the electron bunch arrival time is not important – good for pump-probe experiments using variety of pump sources derived from initial laser signal
Electron trajectory through undulator

While propagating one undulator period, the electron is delayed with respect to the light on one optical wavelength.

\[ \delta \gamma = \frac{e}{mc^2} \int (\vec{E} \cdot \vec{V}) dt \neq 0 \]

\[ \lambda_L = \lambda_u \left(1 + \frac{K^2}{2}\right) / 2\gamma^2 \]

Light interaction with relativistic electron

Laser pulse:
1 mJ, 5-fs
at 800 nm wavelength with CEP stabilization

Fragment of the electron bunch

Undulator period
Energy modulation induced in the electron bunch during interaction with a ~1 mJ, 5 fs, 800 nm wave length laser pulse in a two period wiggler magnet with $K$ value and period length matched to FEL resonance at 800 nm.

This spike gives a dominant signal.
Energy modulation of electrons produced in interaction with two lasers

Combined field of two lasers: increases one laser bandwidth

Energy modulation of electrons produced in interaction with two lasers

Selection of attosecond x-ray pulse: regions with higher peak current reach saturation earlier

Contrast \( \approx 1 \) (assuming 100 fs long bunch)

X-ray wavelength = 0.15 nm

Power, (GW)

10 \( \mu \)J

10\(^{10}\)ph

250 asec

Nearly Fourier transform limited pulse
Publications exploring generation of attosecond x-ray pulses using a few-cycle laser pulse with a carrier envelop phase stabilization:

Few examples
Tapered undulator method

Hard x-rays

Energy chirp is compensated by the undulator taper in the central slice

$\lambda_x = 0.15 \text{ nm}$

$\sim 200 \text{ as}$

Contrast $\approx 1$

![Energy modulation graph]

$\frac{d \ln K}{dz} = -\frac{\lambda_x}{\lambda_u} \left( 1 + \frac{K^2}{2} \right) \frac{d \ln \gamma}{c dt}$

Soft x-rays

Wigner transform of the on-axis far field

Chirp $\xi \approx 0.5$

Frequency chirp definition

$\varphi = \xi (t / \sigma_t)^2$

Fourier transform limited pulse

$\sim 1.5 \text{ fs (FWHM)}$

With two laser one can manipulate the energy chirp and, thus, the frequency chirp

The figure-of-merit is broad bandwidth of attosecond pulses

Intense attosecond x-ray pulses from FELs provide the opportunity to probe the matter on atomic scale in space and time.

Stimulated X-ray Raman spectroscopy *)
X-ray pump, X-ray probe; element specific

300 asec -> 6 eV, i.e. “sudden” excitation reveals multi-electron dynamics

Artist’s (Denis Han) view of excited electron wavepackets in molecule created by core excitation with attosecond x-ray pulses (courtesy S. Mukamel)

In molecules all electrons move in a combined potential of ion core and other electrons

Selection of attosecond x-ray pulses via angular modulation of electrons*)

$$\Delta E(z) = \Delta E_0 \sin(kz)$$

$$\Delta x'(z) = \Delta x'_0 \cos(kz)$$

Hermite-Gaussian laser modes

Combining angular and energy modulations for improved contrast of attosecond x-ray pulses.

X-ray wavelength = 0.15 nm

X-ray peak power as a function of time

100 GW

Contrast > 100

Central peak

115 as, 10 µJ ~100mJ/cm²

X-ray peak power as a function of time
Obtaining attosecond pulses at 1 nm using echo effect*)

Adding energy chirp via interaction with ultra short laser pulse; needs sub-fs synchronization


radiator has only 12 periods

x-ray pulse

20 asec
Two color attosecond pump and attosecond probe x-ray pulses*)

Fragment of the longitudinal phase space

Zoom into the main peak shows microbunching at 2.28 nm

*) Zholents, Penn, Nucl. Inst. and Meth. A 612, 254(2010).
Simulation results using 1D code and GENESIS for two color scheme

![Graph showing power distribution over time and photon energy]

- 543 eV, 210 asec FWHM
- 410 eV, 480 asec FWHM

ΔT adjustable

Molecule structure of 5-quinolinol

![Molecule structure diagram]

Simulation results using 1D code and GENESIS for two color scheme

![Graph showing power distribution over time and photon energy]

- 6.4 eV FWHM
- 7.2 eV FWHM

Simulation results using 1D code and GENESIS for two color scheme

- 543 eV, 210 asec FWHM
- 410 eV, 480 asec FWHM

ΔT adjustable

Molecule structure of 5-quinolinol

![Molecule structure diagram]
X-ray SASE FEL amplifier with mode-locking produces a train of attosecond pulses\(^1\)

\[\text{Spike FWHM \sim 23 as}\]

We are at the threshold of a new era of science, where for the first time, the new instruments, the x-ray FELs, are capable to study the matter with a single atom time and space resolution.

FLASH, SCSS and LCLS routinely work with ultra-short XUV/x-ray pulses.

Remarkably, adding attosecond capabilities to existing FELs require rather modest modifications.
Thank you for your attention