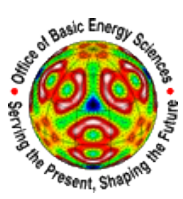


A. Zholents

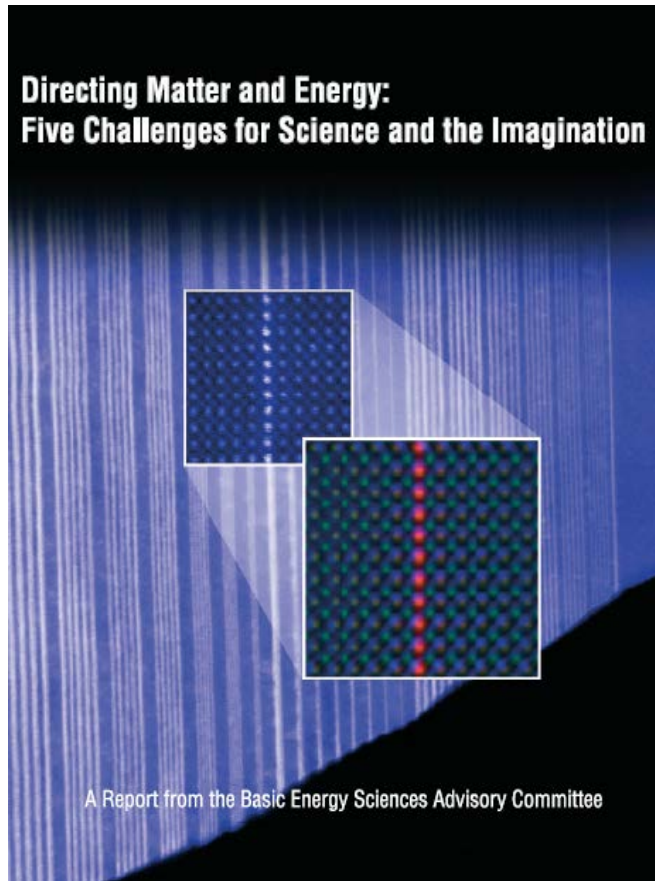
Towards a 5th Generation Light Source, October 1 -2, 2010

... 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



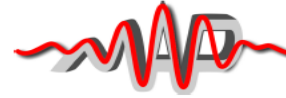
- *How do we control materials and processes at the level of electrons?*
- *How do we design and perfect atom- and energy-efficient synthesis of new forms of matter with tailored properties?*
- *How do remarkable properties of matter emerge from complex correlations of atomic and electronic constituents 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**  
and  
**Munich Centre for Advanced Photonics**

June 19, 2008

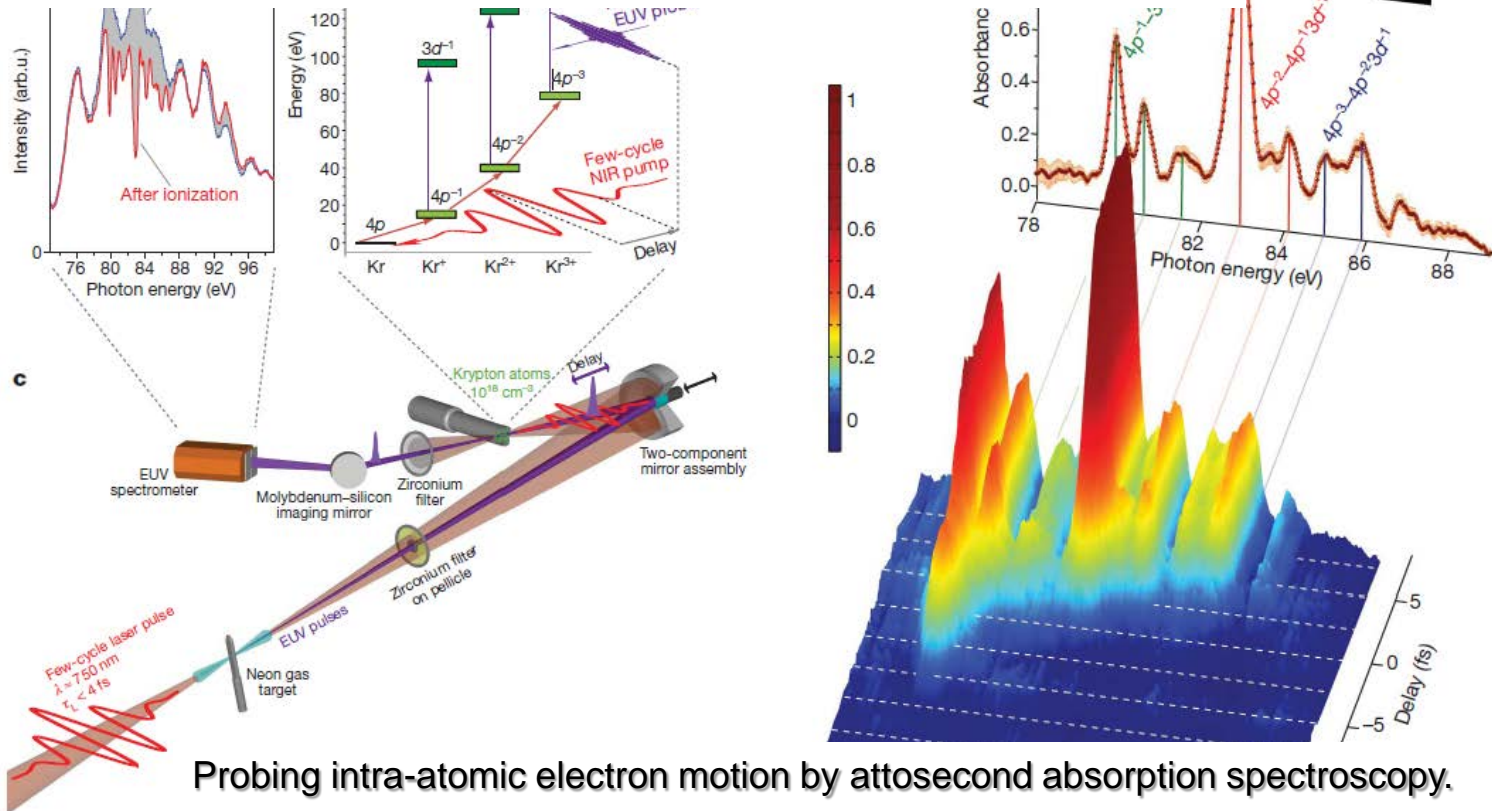


## Ultrafast Look into Atoms and Molecules

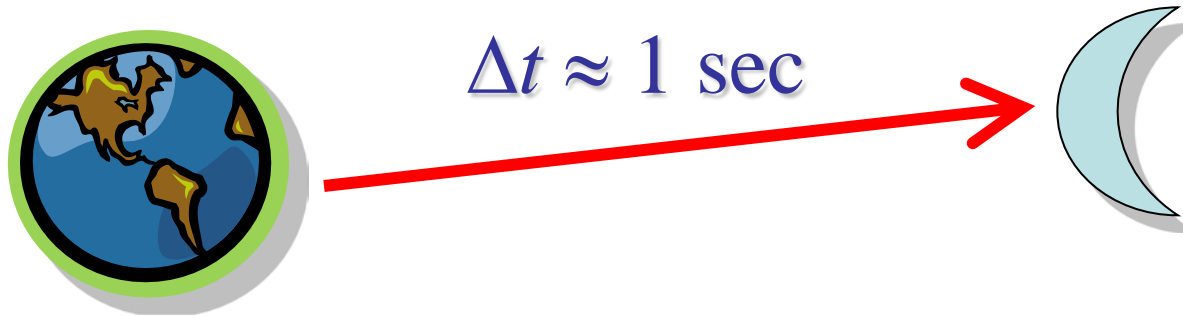
New record in ultrafast metrology  
Ludwig-Maximilians-Universität München

# 80 attoseconds !

Quantum Optics and the  
pulses lasting only 80



Probing intra-atomic electron motion by attosecond absorption spectroscopy.



$c \times 80 \text{ attoseconds} \rightarrow 24 \text{ nm} !$

$\alpha \times c \times 80 \text{ attoseconds} \rightarrow 2 \text{ \AA} !$

# 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

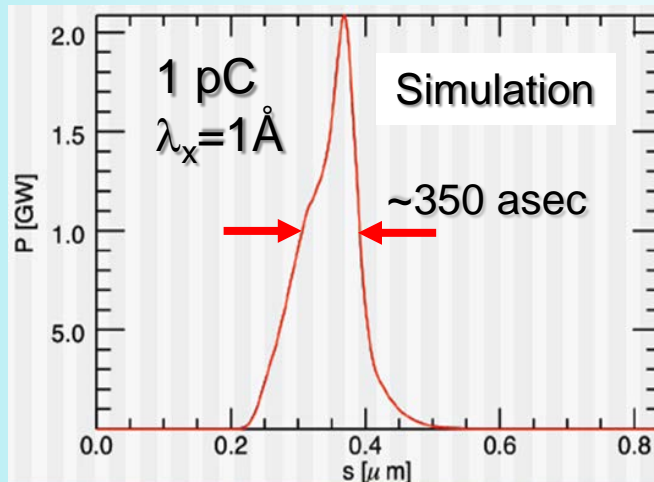




# Simple thoughts

- 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<sup>1</sup>
- Nearly FTL pulses from SASE<sup>1</sup>

$$B = \frac{2Q}{\gamma^3 \epsilon_x \epsilon_y \epsilon_z}$$



LCLS

$$Q = 1 \text{ nC}$$

$$B = 0.3 \text{ nC}/\mu\text{m}^3$$

$$\gamma \epsilon_{x,y} \approx 1 \mu\text{m}$$

$$\gamma \epsilon_z \approx \sigma_z(3 \text{ keV})/mc^2 = 6.5 \mu\text{m}$$

$$Q = 0.02 \text{ nC}$$

$$B = 4.5 \text{ nC}/\mu\text{m}^3$$

$$\gamma \epsilon_{x,y} \approx 0.15 \mu\text{m}$$

$$\gamma \epsilon_z \approx \sigma_z(1 \text{ keV})/mc^2 = 0.4 \mu\text{m}$$

P. Emma, LBNL workshop, 08/2010

Brightness defines the gain length

$$1/L_G \sim B$$

1) S. Reiche, P. Musumeci, C. Pellegrini, J.B. Rosenzweig, Nucl. Instr. and Meth. A 593 (2008) 45



# 20-pC bunch operation at LCLS<sup>1</sup>

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

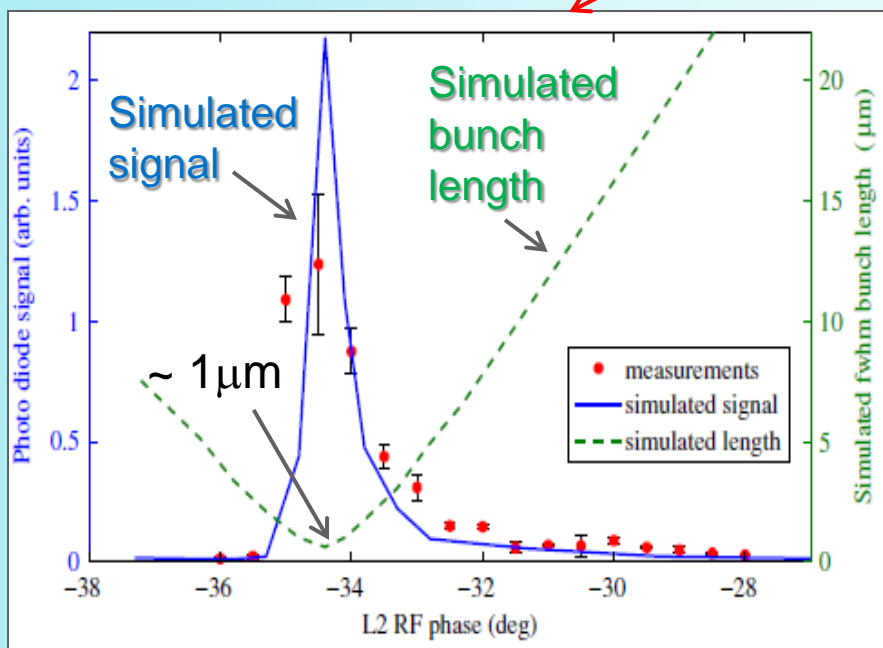
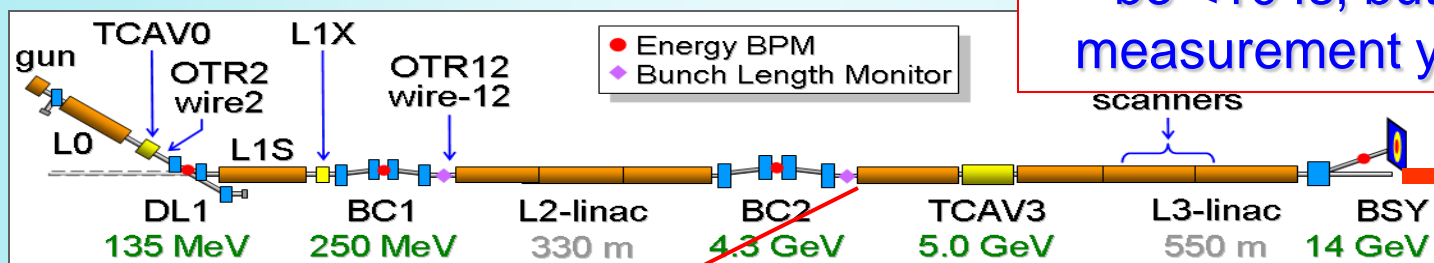
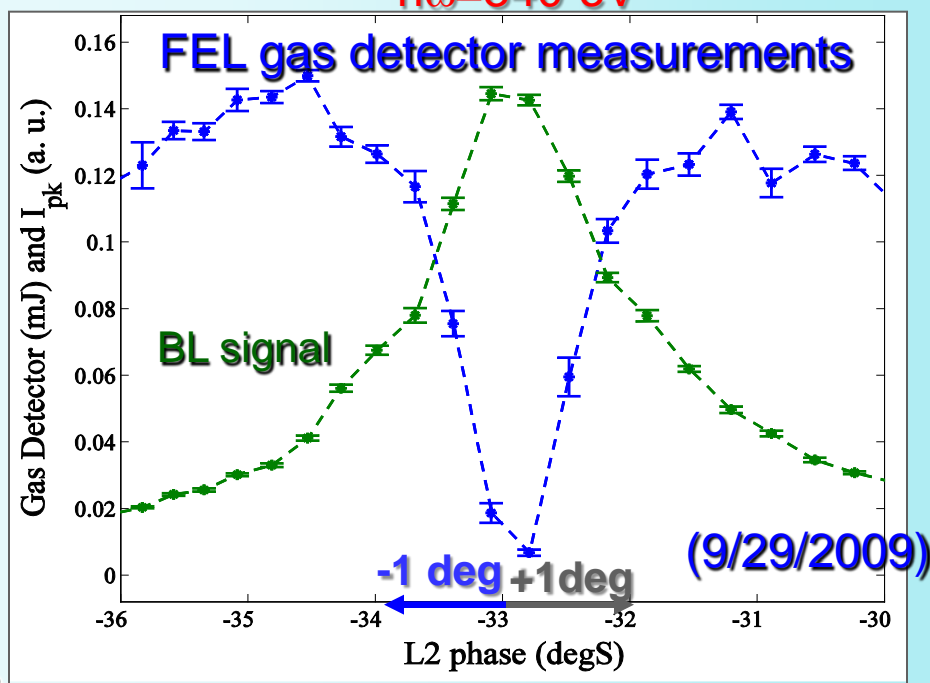


Photo-diode signal on OTR screen after BC2

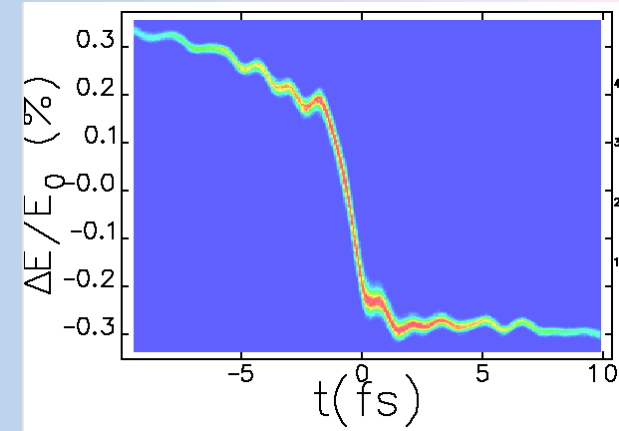
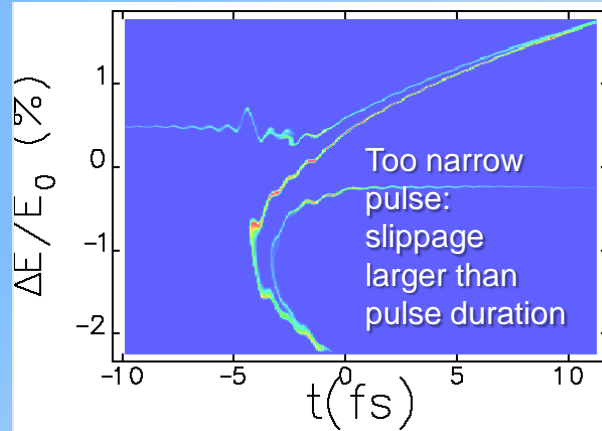
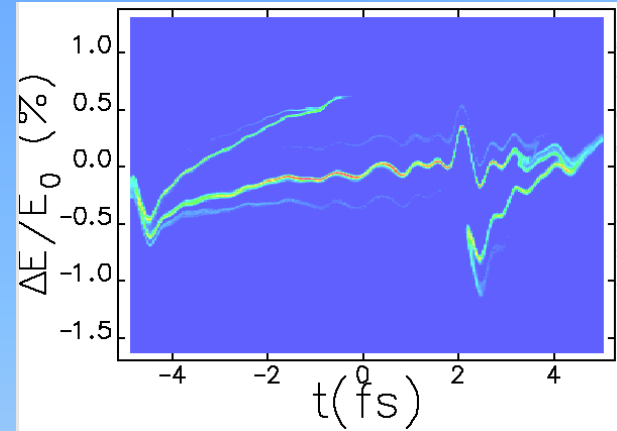


1) Y. Ding et. al, PRL 2009

# Soft x-rays at 1.5 nm (simulations for LCLS)<sup>1</sup>

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

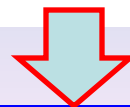
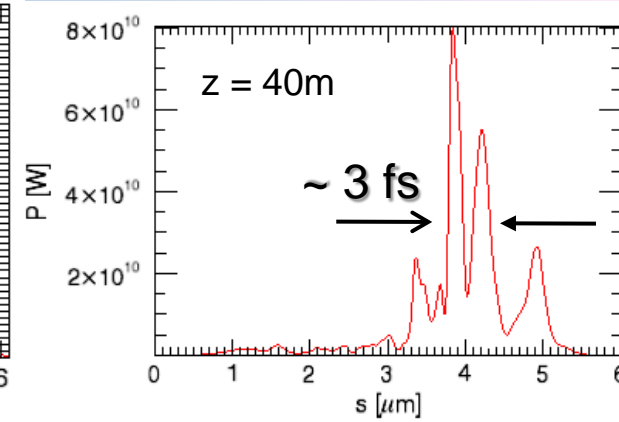
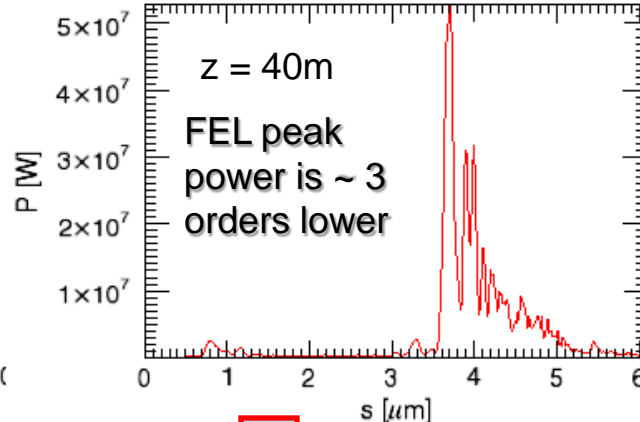
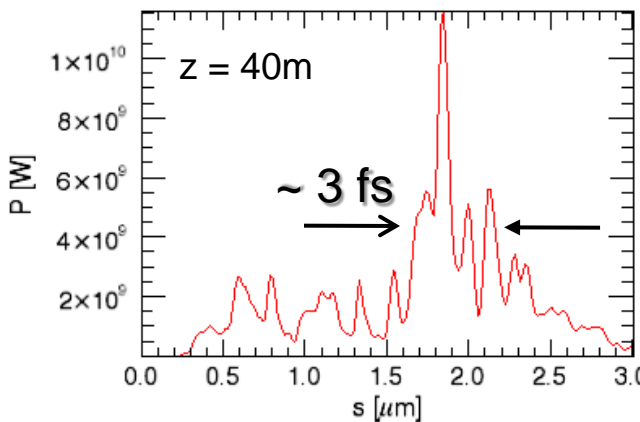
At undulator entrance, 4.3 GeV, Laser heater off



Under-compression: +1 deg off

Full-compression

Over-compression: -1 deg off

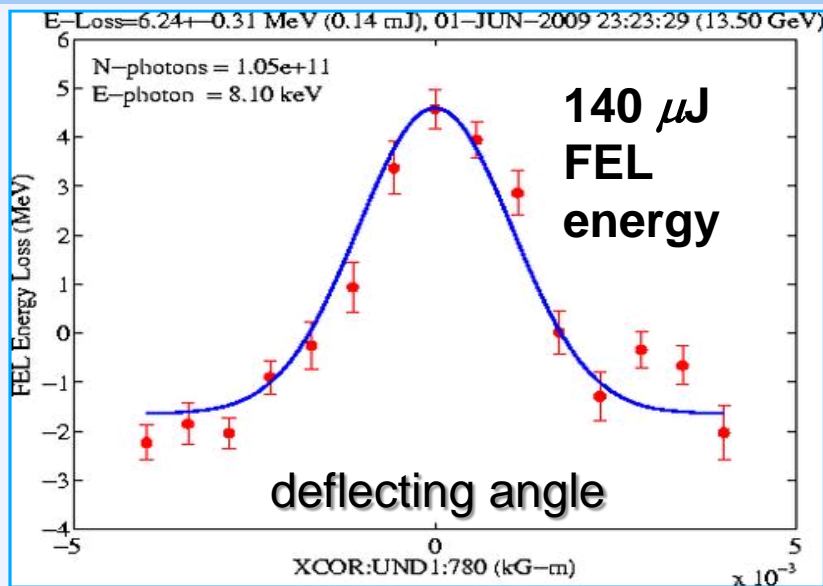
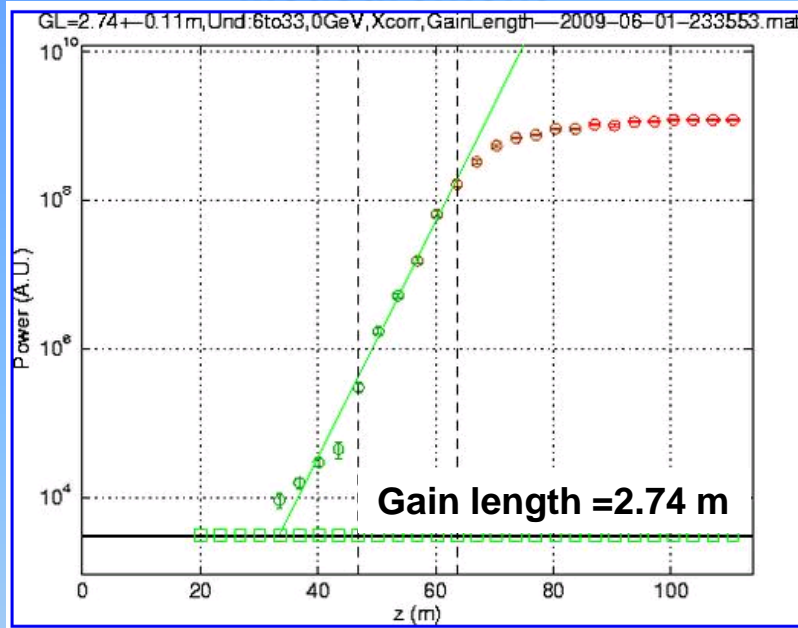


Actual measurements qualitatively confirm simulations.

Direct measurement of ultra-short x-ray pulse duration remains to be difficult.

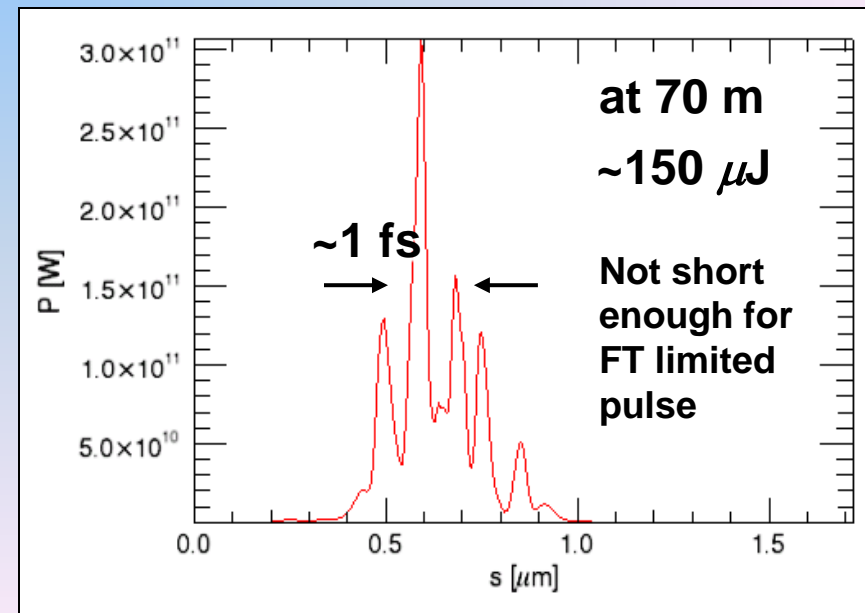
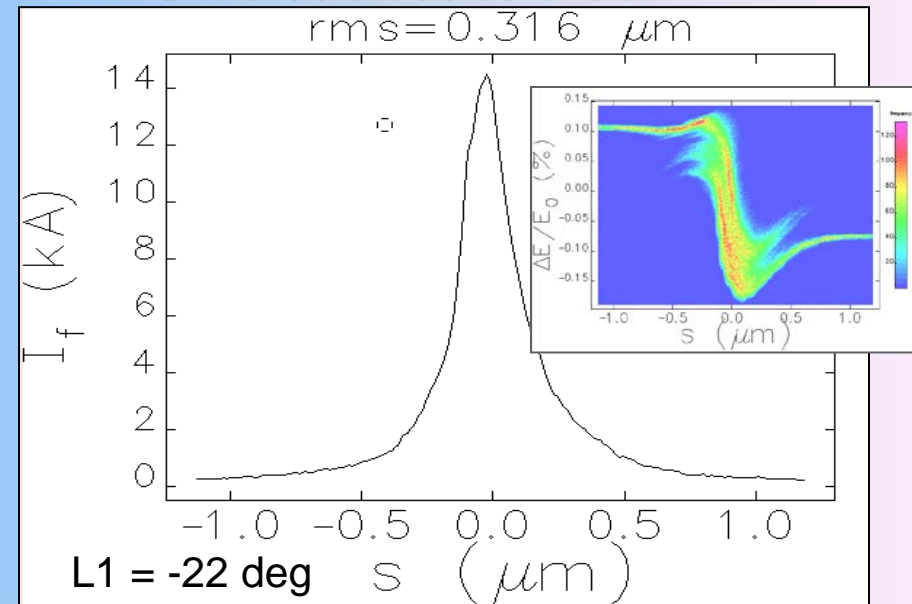
# At 1.5 Å FEL performs well at full compression (slippage just right)

## Measurement



J. Frish et. al., talk at FEL'09

## Simulation at 13.6 GeV

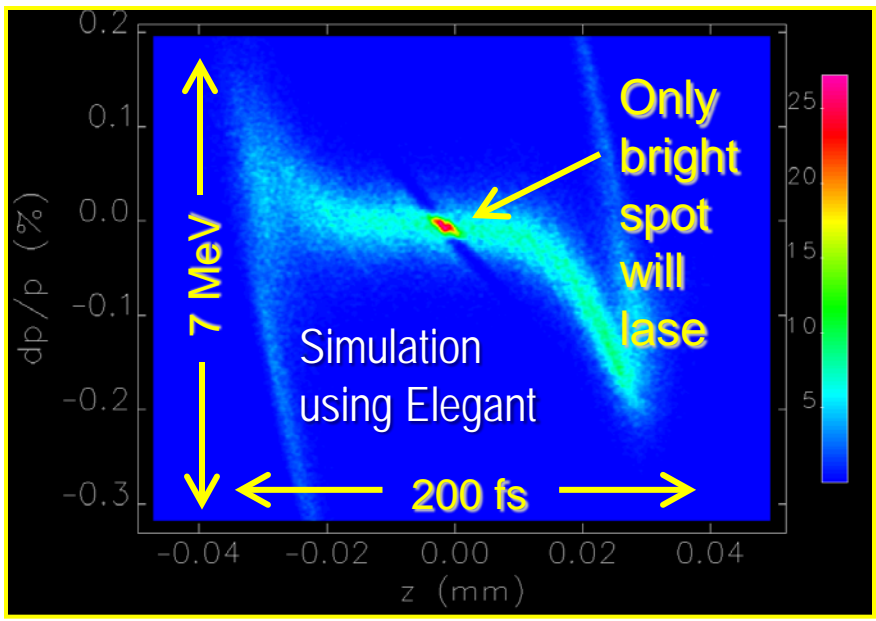
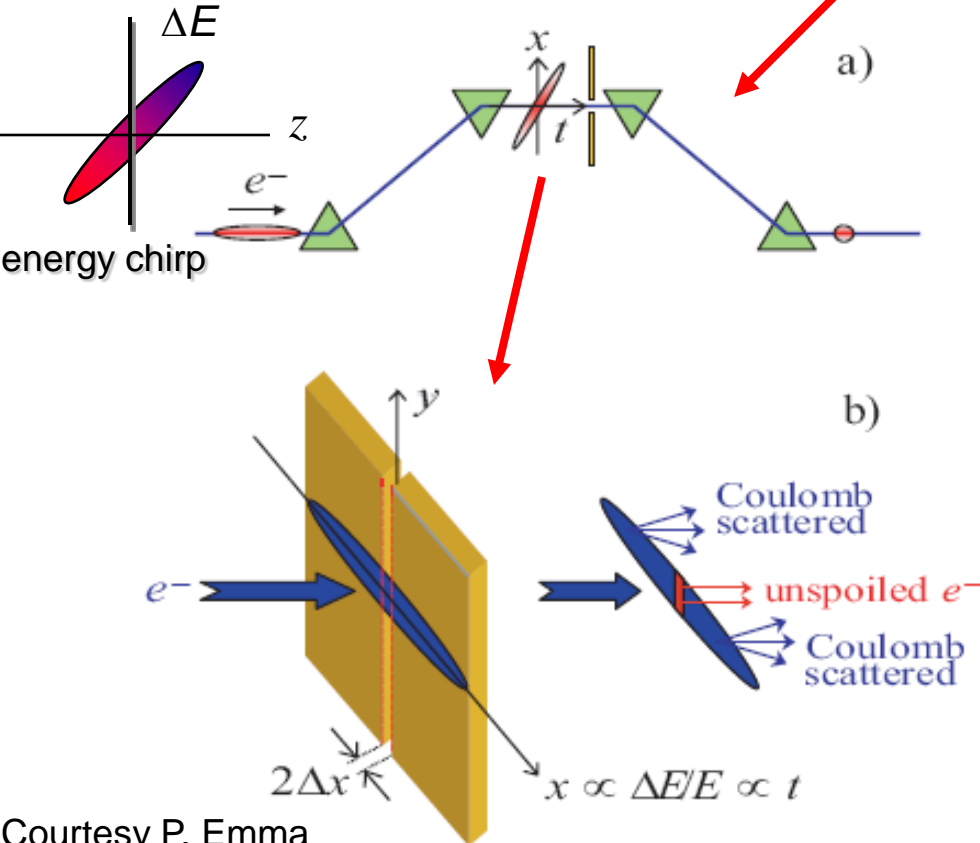
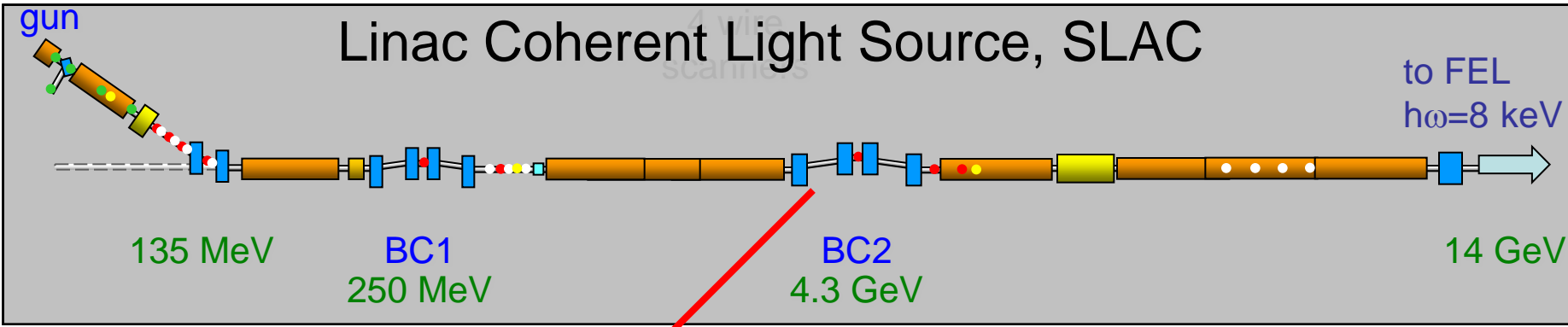


Y. Ding, LBNL workshop, 08/2010

# 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

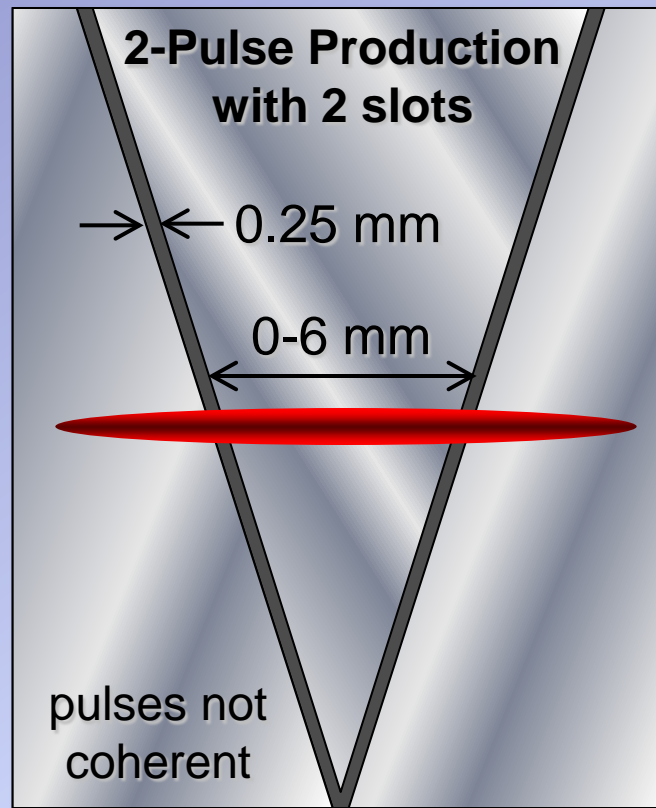
## Linac Coherent Light Source, SLAC



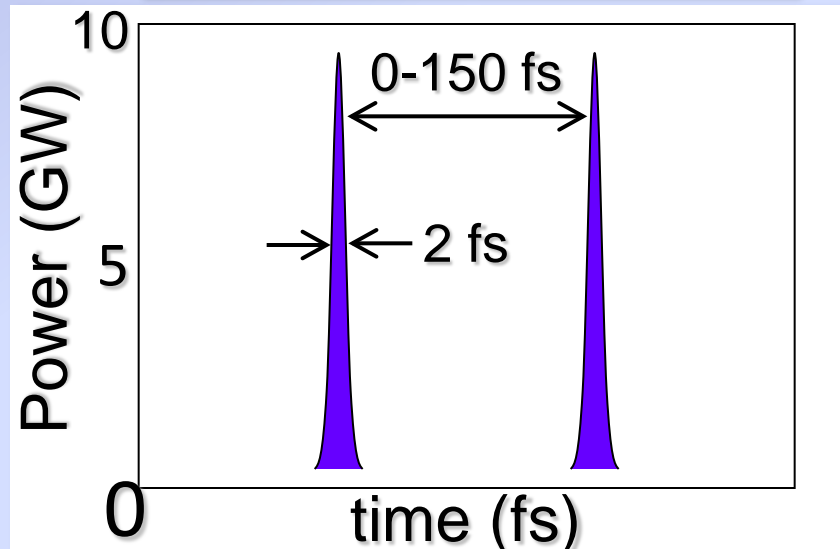
X-ray pulse width ~ 400 asec  
 Bunch arrival time jitter ~ 50 fs  
 ~ 10<sup>9</sup> photons/pulse -> 7 GW

Courtesy P. Emma

# Double X-Ray Pulses from a Double-Slotted Foil



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



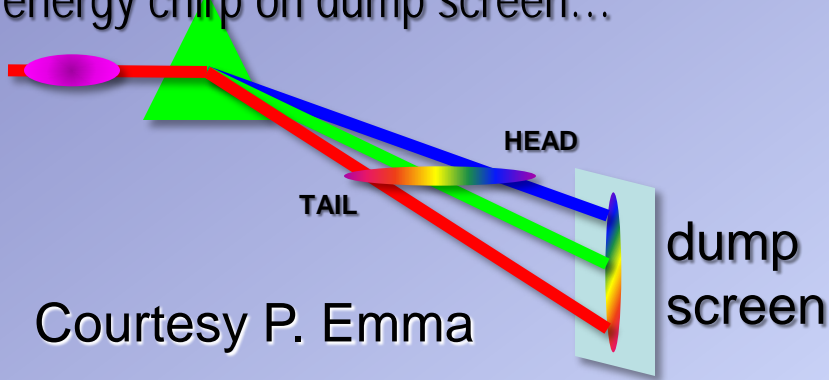
Precise controlled  
time delay between  
x-ray pump and x-  
ray probe pulses

Courtesy P. Emma

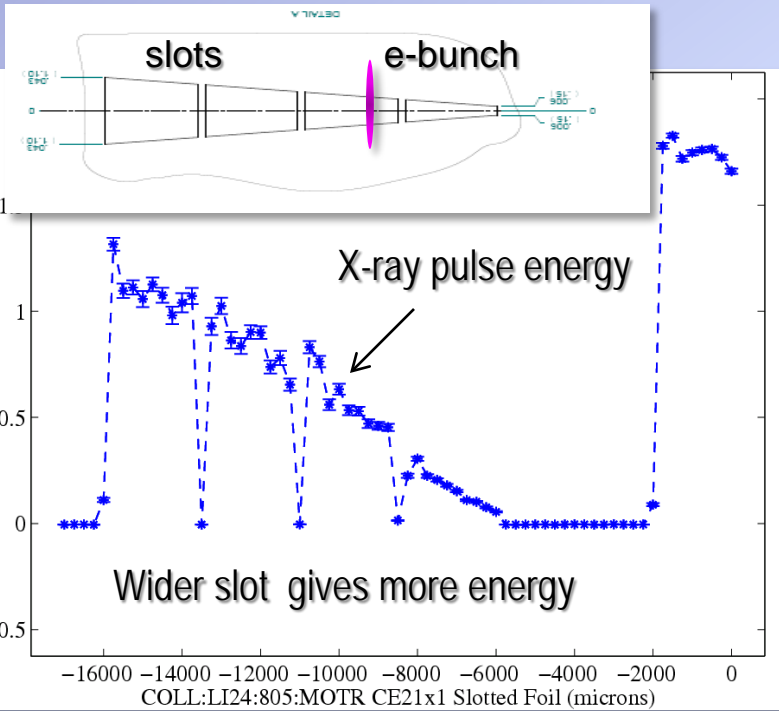
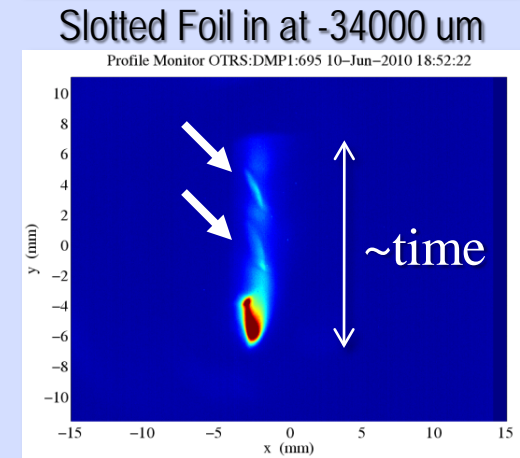
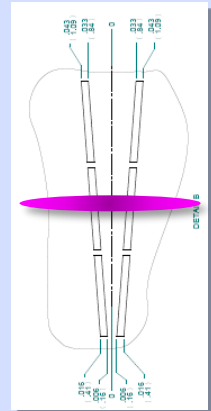
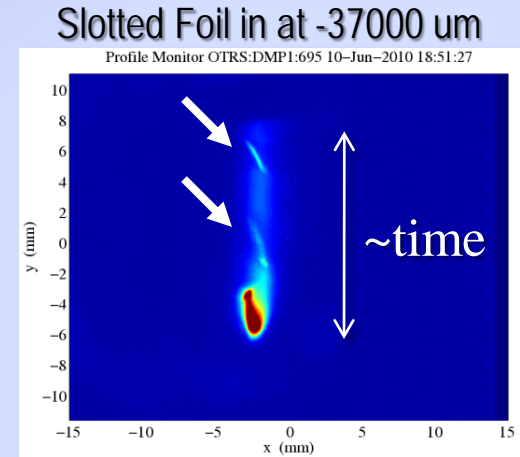
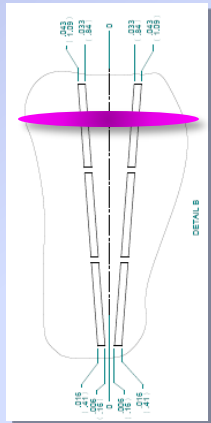
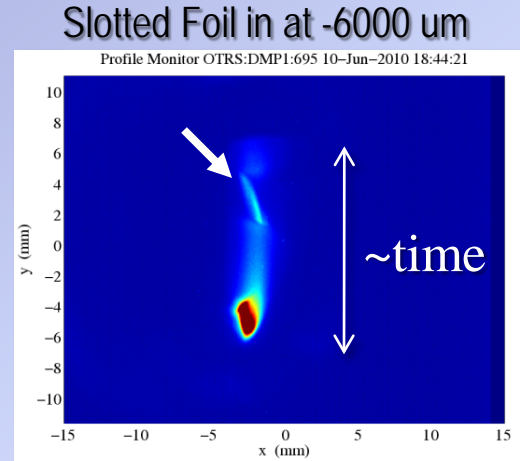
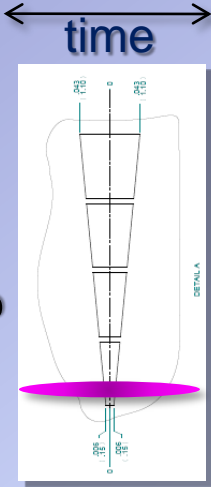


# Initial results (very preliminary), June 17, 2010

Over-compressed bunch introduces e<sup>-</sup> energy chirp on dump screen...



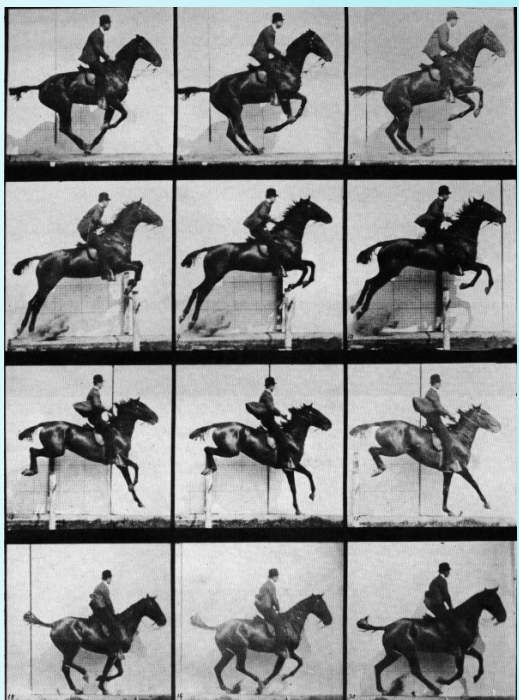
Courtesy P. Emma



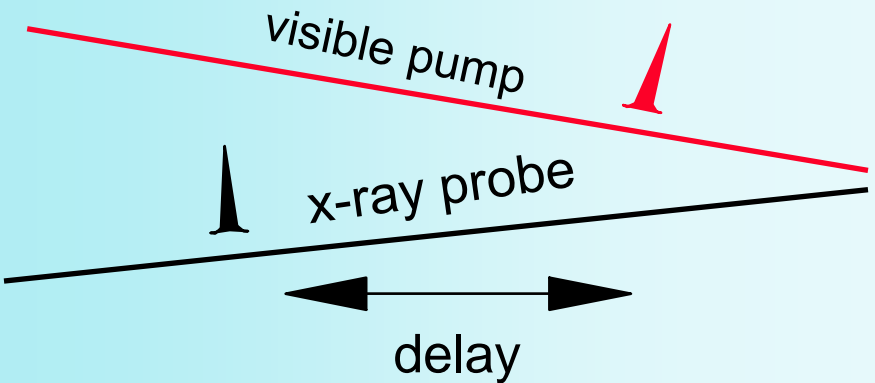
# Pump – probe studies using ultra-fast x-ray pulses



Pellet hits a strawberry



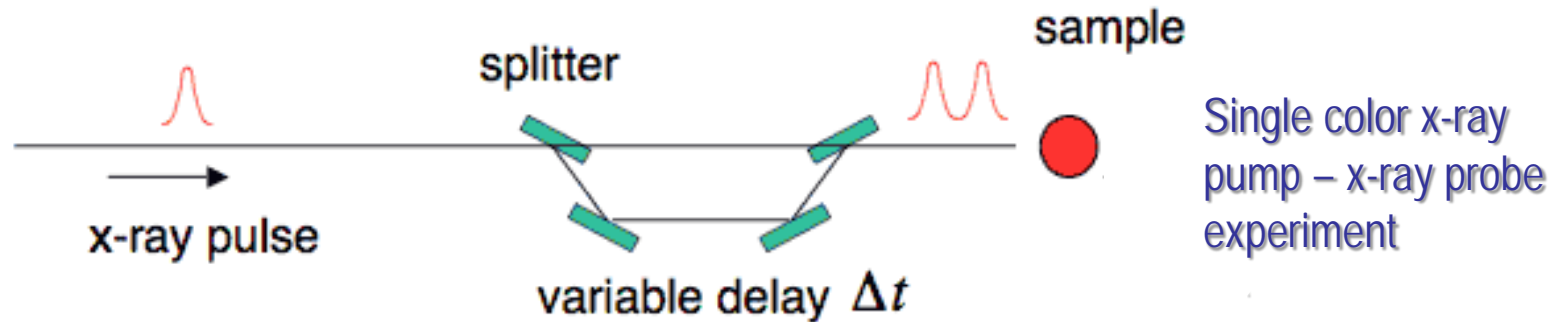
Stop-motion photography  
E. Muybridge, 1878



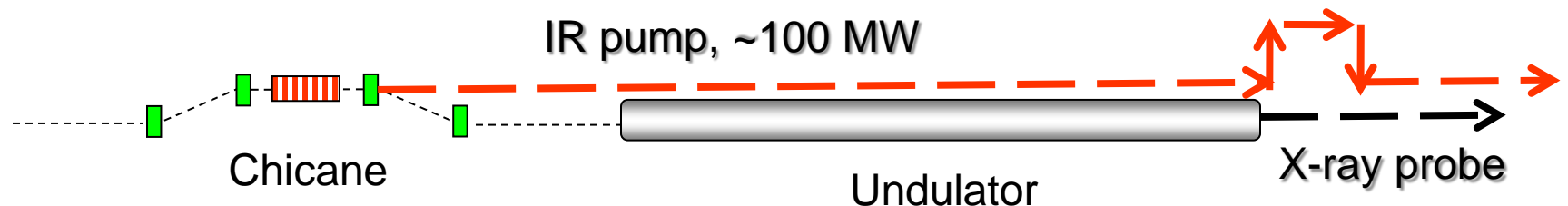


# Options for experiment utilizing synchronized pump and probe signals when electron bunch arrival time has a “large” jitter

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



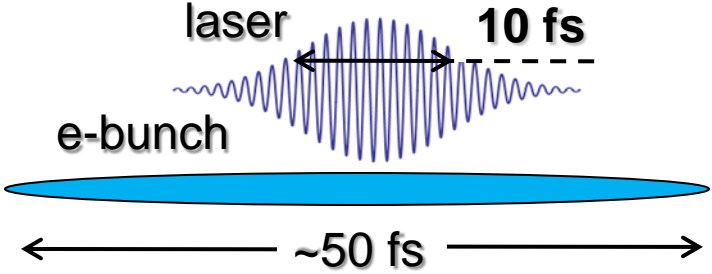
- Create pump signal using coherent undulator radiation and adjust delay<sup>1</sup> (in case of an ultra-short e-bunch,  $\sim 1$  fs)



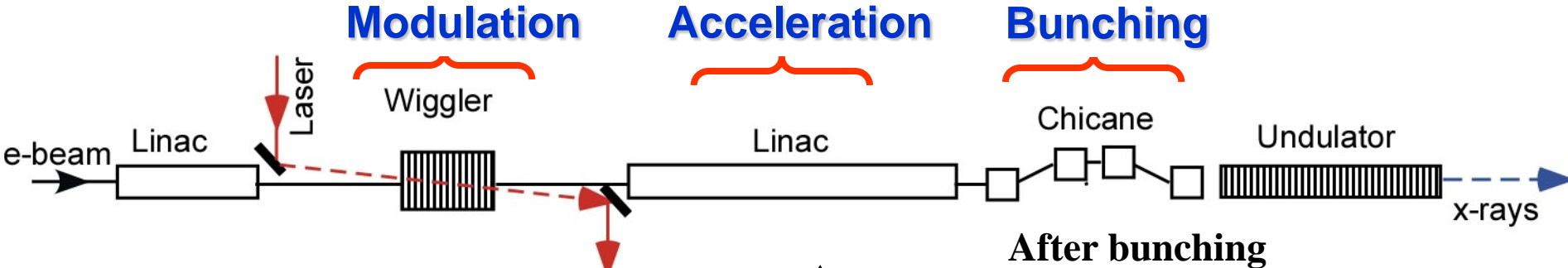
1) U. Fruhling *et al.*, *Nature Photonics*, **3**, 523(2009); also considered by UCLA group

# Precision synchronization of pump and probe pulses

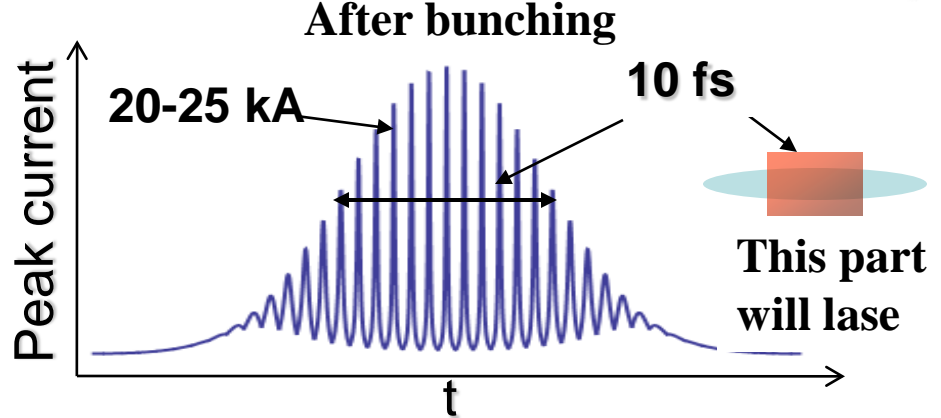
- Seeded FELs naturally possess precise synchronization
  - if electron bunch length > laser pulse + jitter



- Current-enhanced SASE FEL --> same conclusion

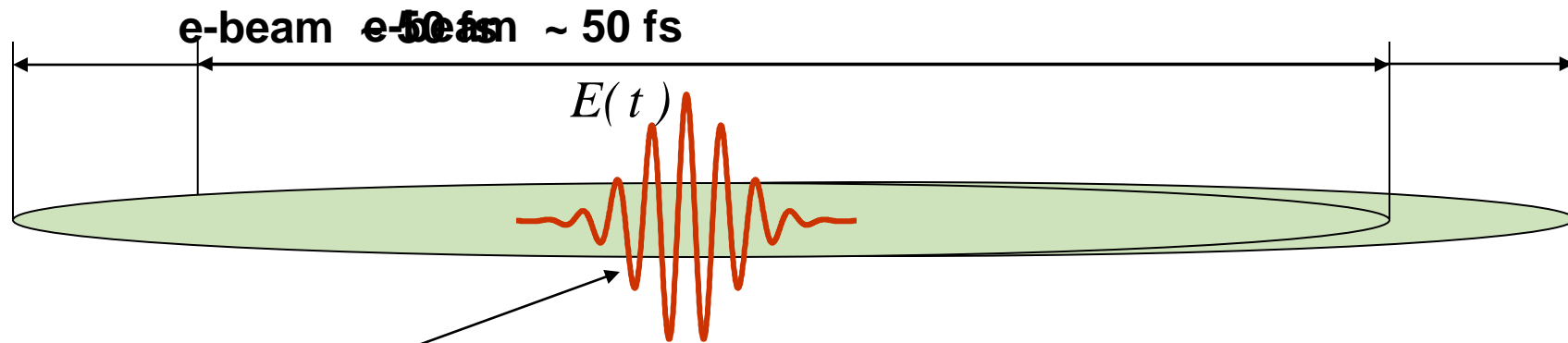


Require synchronization of the seed and pump lasers, 10 fs is state of the art \*)



\*) J. Kim et. al., Nature Photonics, 2, 733, 2008; R. Wilcox, Opt. Lett. 34, 3050, 2009

# Attosecond pulse generation via electron interaction with a few cycle carrier-envelope phase stabilized laser pulse



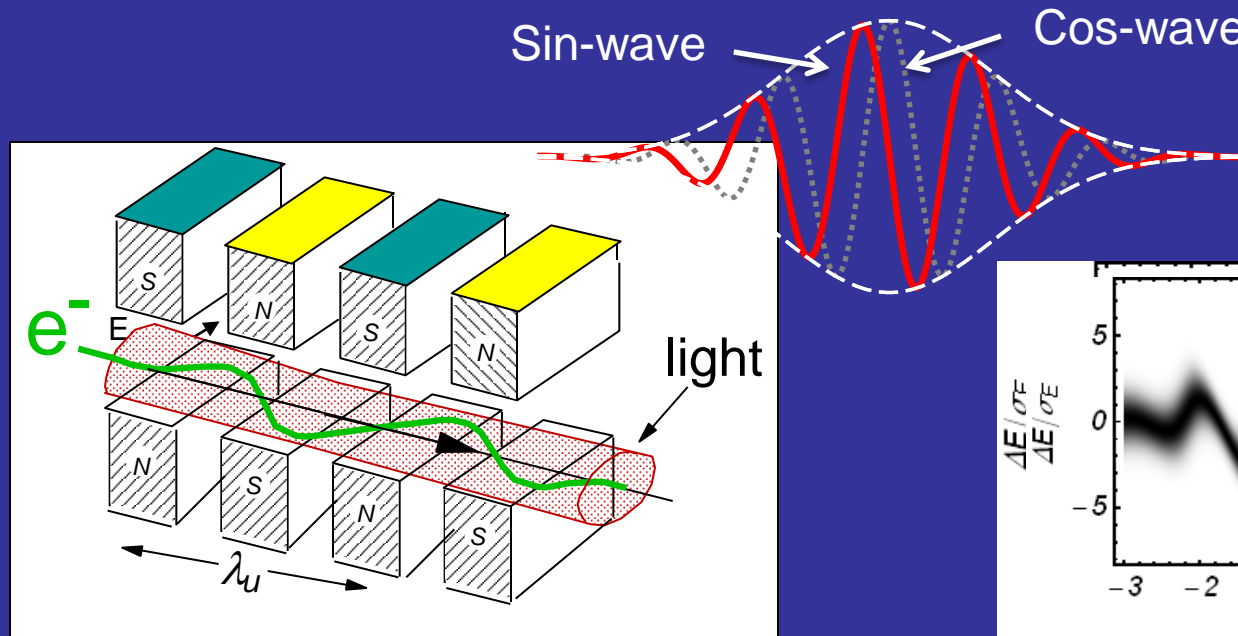
Laser pulse  $\sim 5$  fs

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

Basic idea:

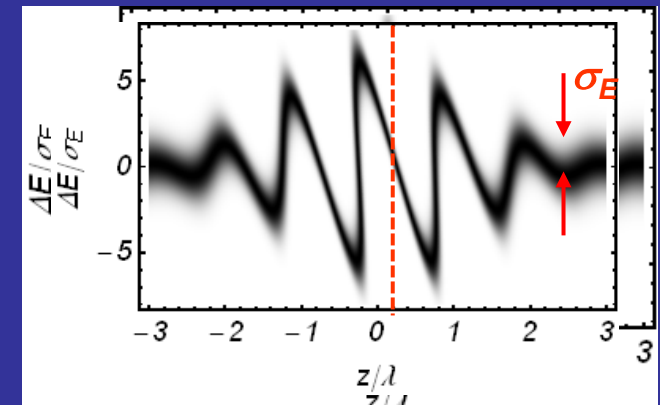
Take an ultra-short slice of electrons from a longer electron bunch to produce a dominant x-ray radiation

# Light interaction with relativistic electron



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

Electron trajectory through undulator

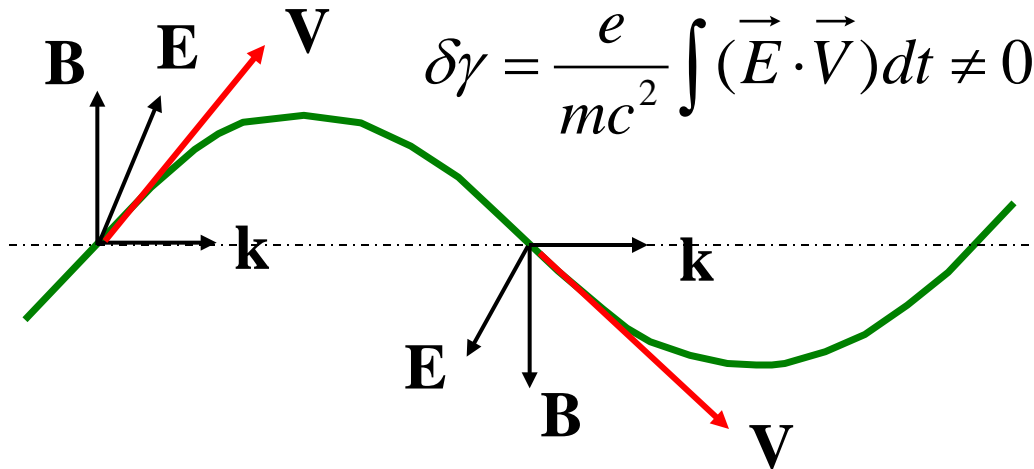


Fragment of the electron bunch

Laser wavelength

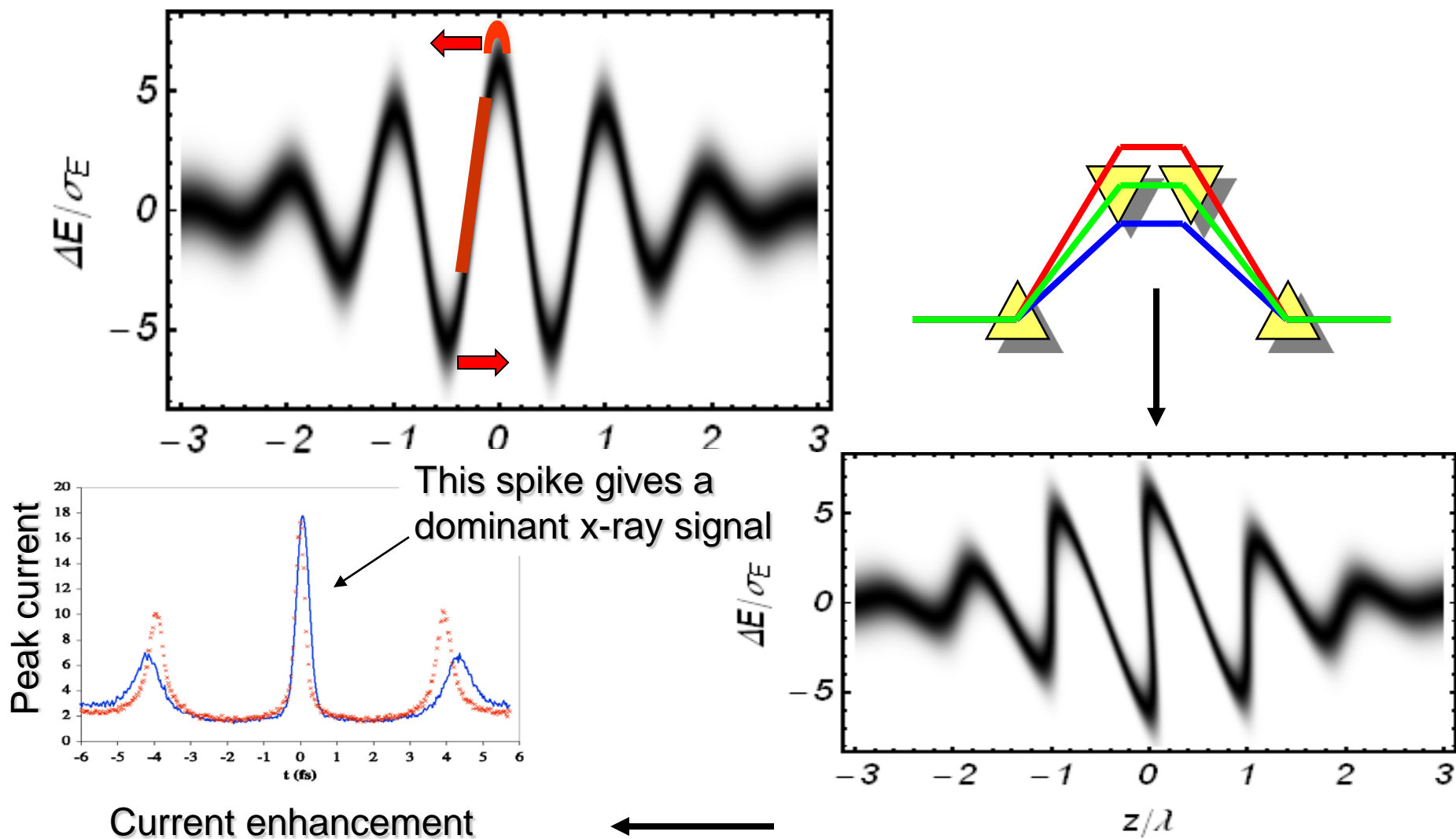
$$\lambda_L = \lambda_u (1 + K^2/2) / 2\gamma^2$$

Undulator period



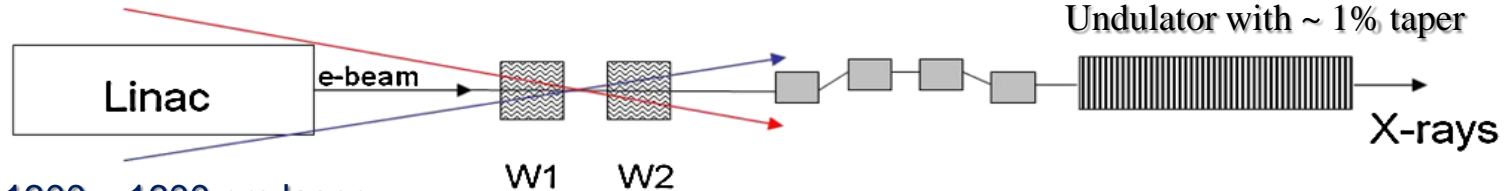
While propagating one undulator period, the electron is delayed with respect to the light on one optical wavelength

Energy modulation induced in the electron bunch during interaction with a  $\sim 1$  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

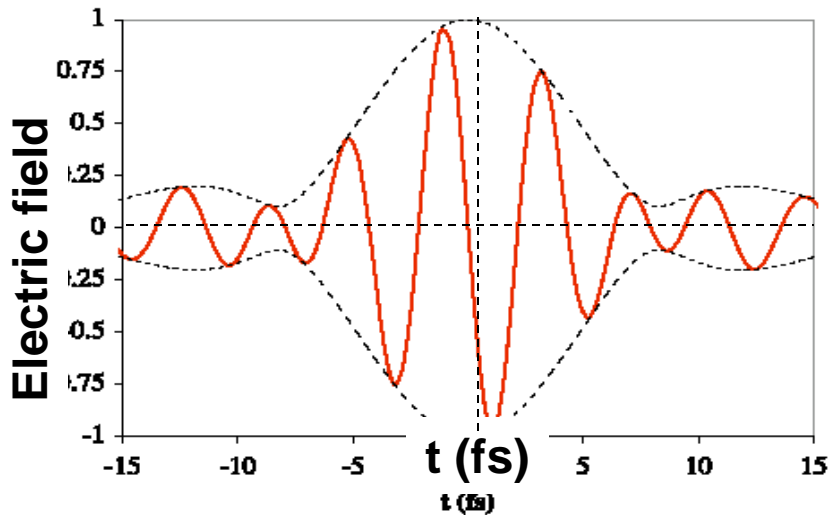


# Current enhancement method <sup>\*)</sup>

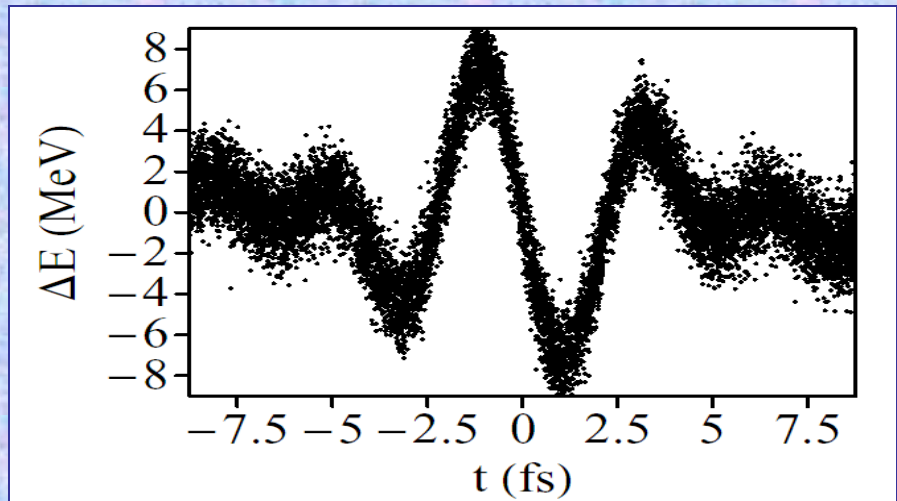
800 – 1000 nm laser;  
7.5 – 25 fs; 0.2 – 0.5 mJ



1300 – 1600 nm laser;  
10 – 45 fs; 0.07 – 0.2 mJ



Combined field of two lasers:  
increases one laser bandwidth

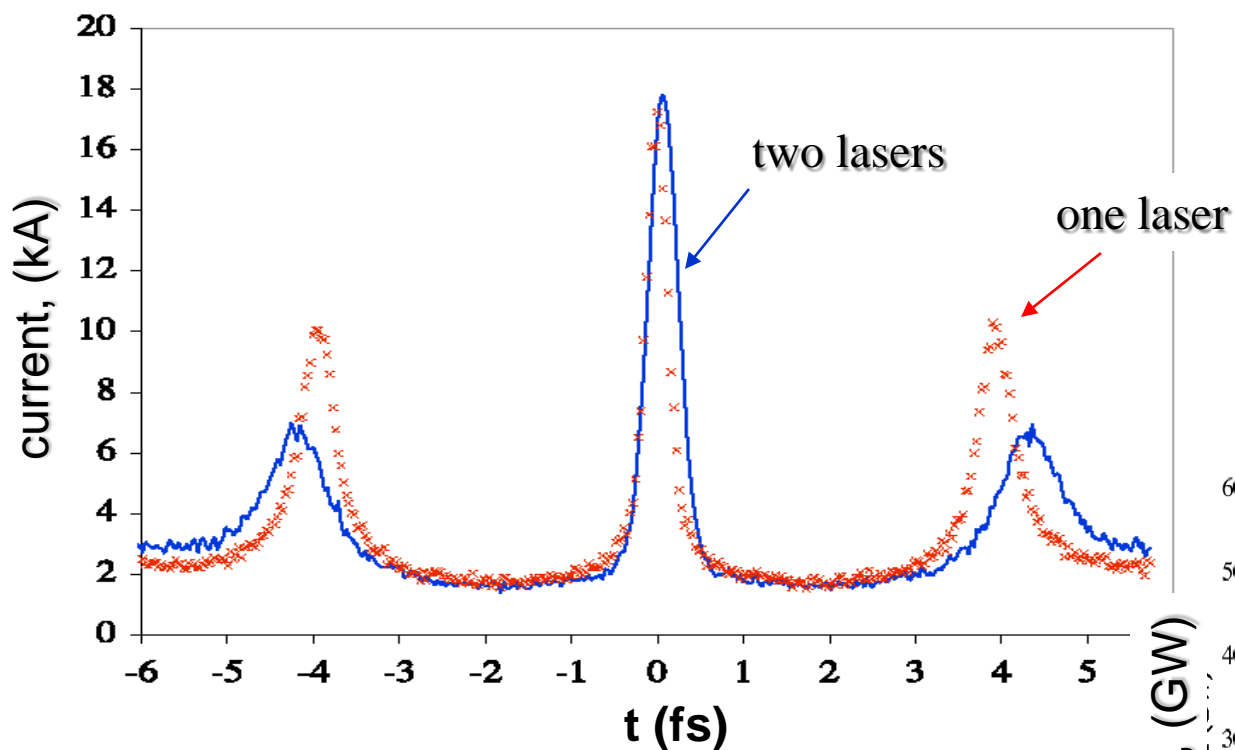


Energy modulation of electrons  
produced in interaction with two lasers

<sup>\*)</sup> Zholents, Penn, PRST-AB, 8, (2005);  
Y. Ding et al., Phys. Rev. ST-AB, 12, (2009).

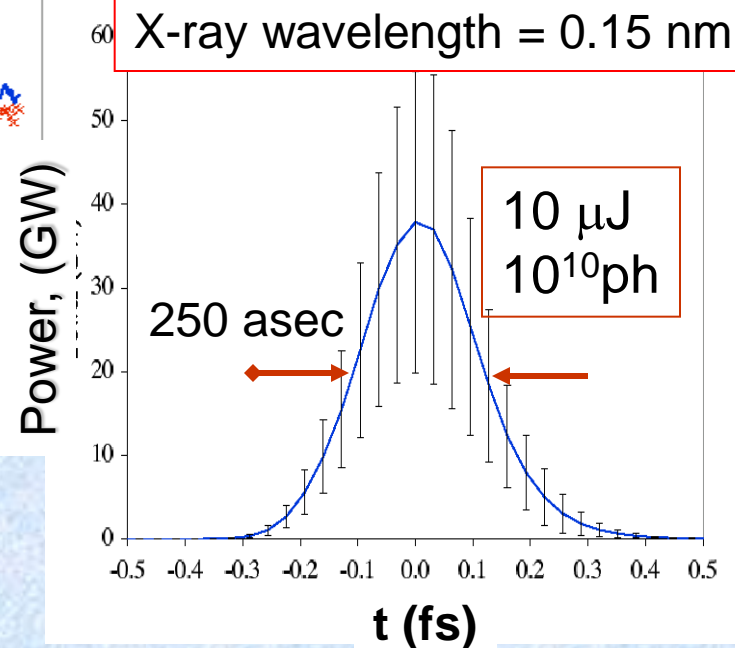


# Current enhancement method (2)



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

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



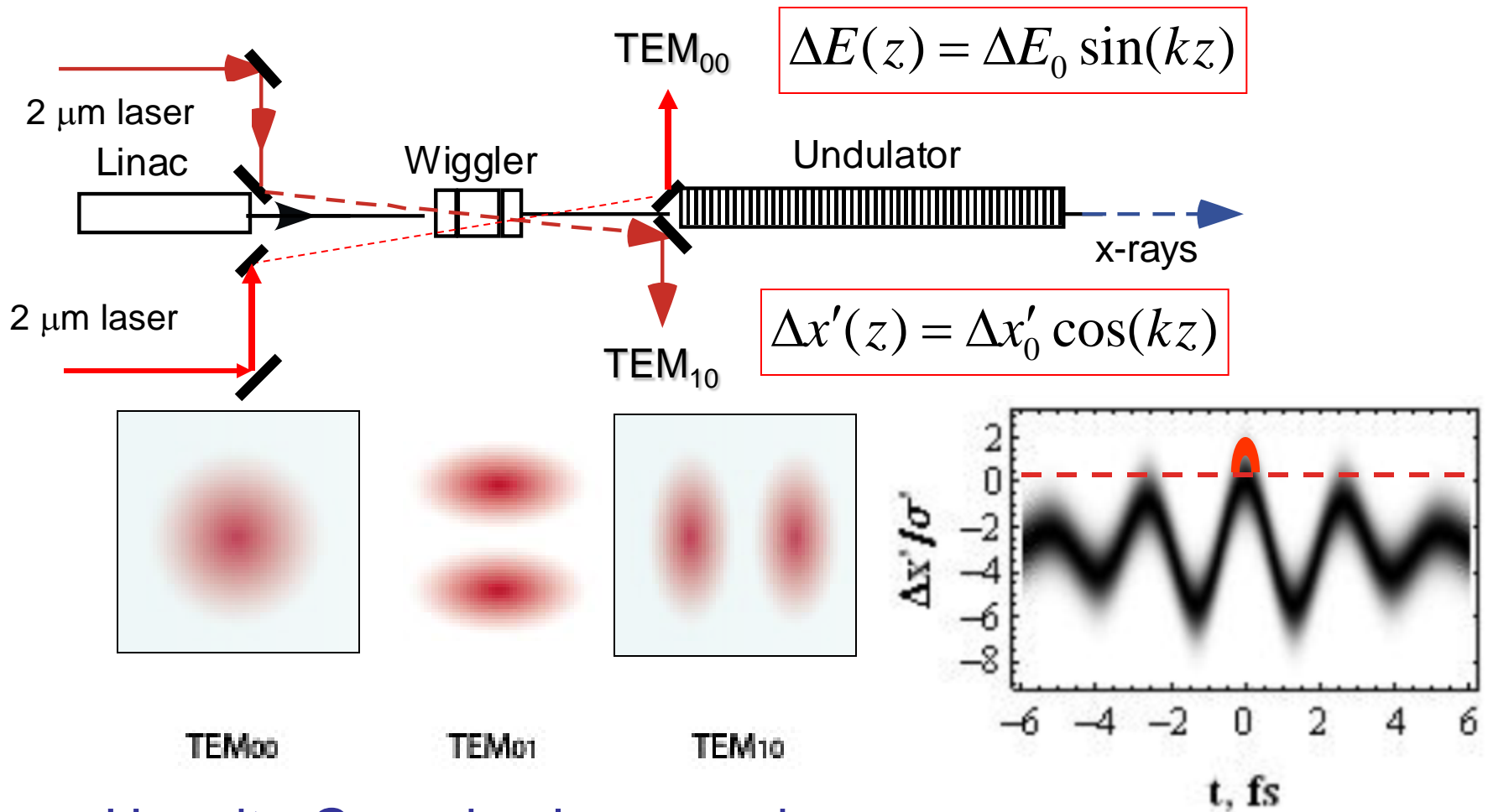
**Nearly Fourier transform  
limited pulse**



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

- [1] A. A. Zholents and W. M. Fawley, Phys. Rev. Lett. 92, 224801 (2004).
- [2] E.L. Saldin, E.A. Schneidmiller, M.V. Yurkov, Opt. Commun. 237,153 (2004).
- [3] E.L. Saldin, E.A. Schneidmiller, M.V. Yurkov, Opt. Commun. 239,161 (2004).
- [4] A. A. Zholents and G. Penn, Phys. Rev. ST-AB 8, 050704 (2005).
- [5] E.L. Saldin, E.A. Schneidmiller, M.V. Yurkov, Phys. Rev. ST-AB 9, 050702 (2006).
- [6] A. A. Zholents and M.S. Zolotarev, New J. Phys. 10, 025005 (2008).
- [7] W.M. Fawley, Nucl. Inst. and Meth. A 593, 111(2008).
- [8] Y. Ding, Z. Huang, D. Rather, P. Bucksbaum, H. Maerdji, Phys. Rev. ST-AB 12, 060703 (2009).
- [9] D. Xiang, Z. Huang, G. Stupakov, Phys. Rev. ST-AB 12, 060701 (2009).
- [10] A. A. Zholents and G. Penn, Nucl. Inst. and Meth. A 612, 254(2010).

# Selection of attosecond x-ray pulses via angular modulation of electrons<sup>\*)</sup>

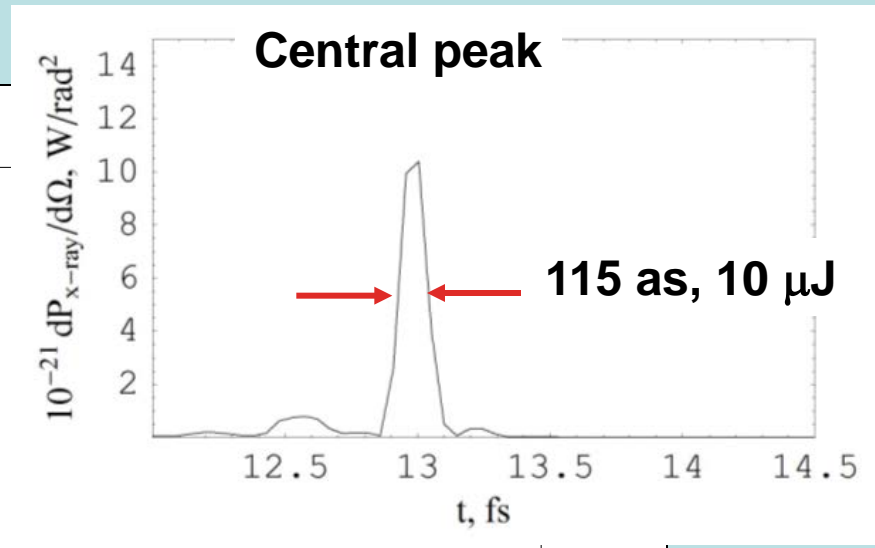
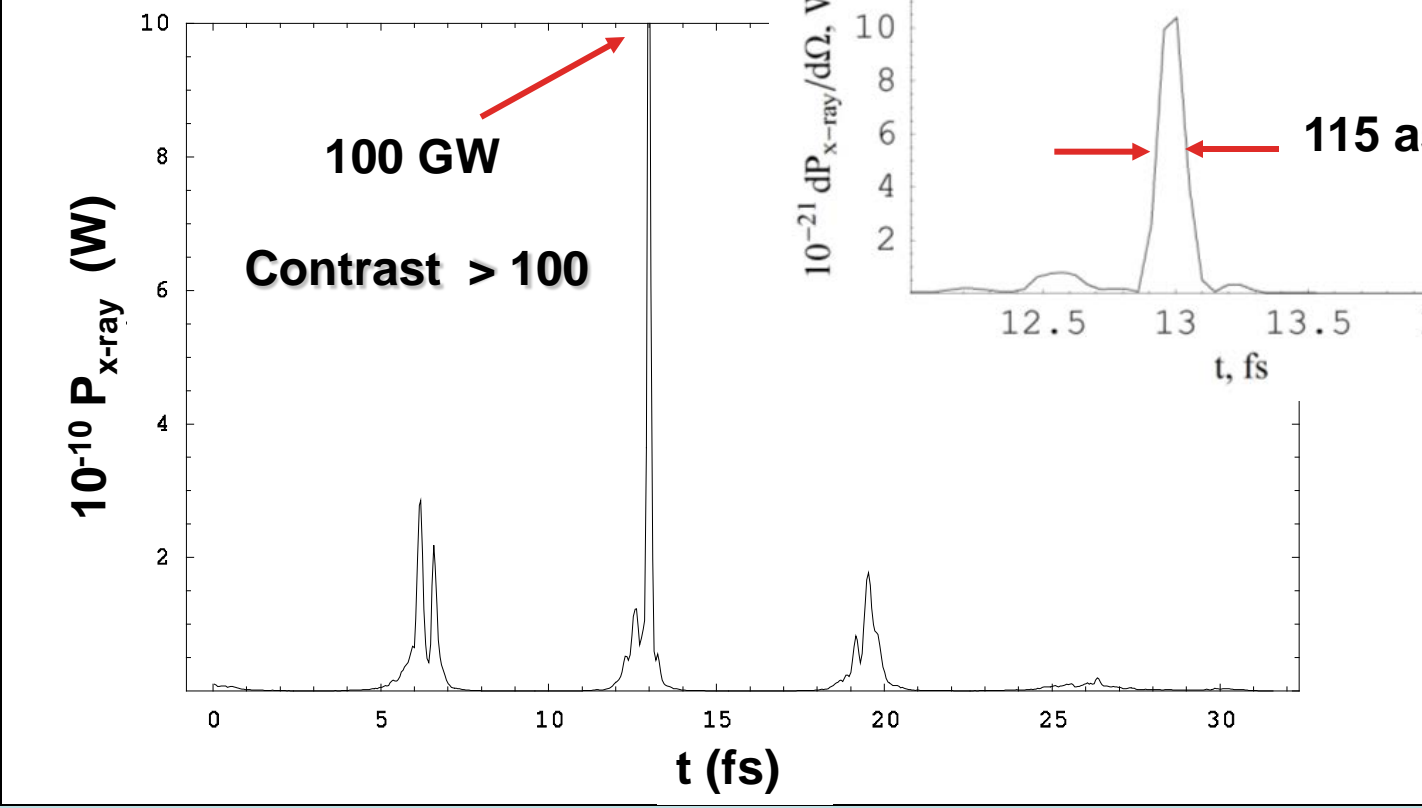


## Hermite-Gaussian laser modes

<sup>\*)</sup> Zholents, Zolotarev, New Journal of Physics, 10, 025005 (2008).

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

X-ray wavelength = 0.15 nm

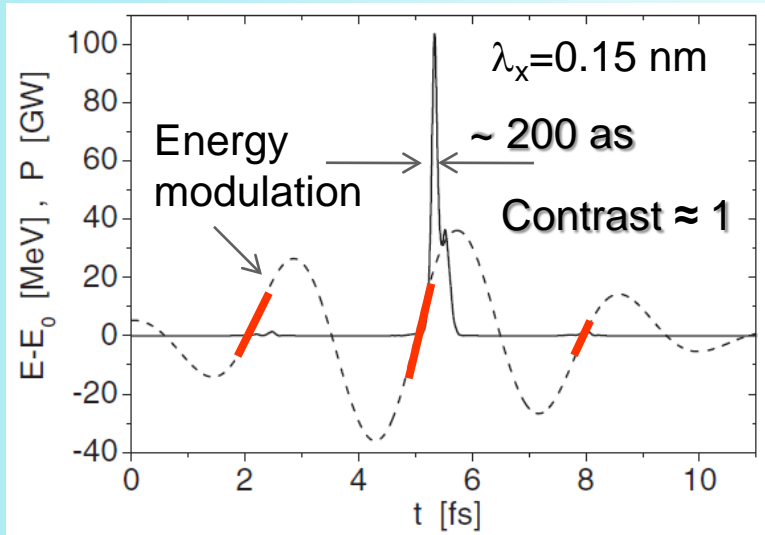


X-ray peak power as a function of time

# Tapered undulator method\*

## Hard x-rays

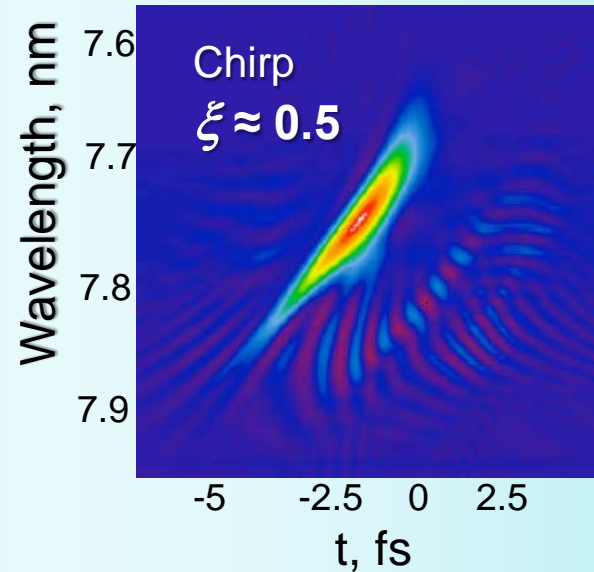
Energy chirp is compensated by the undulator taper in the central slice



$$\frac{d \ln K}{dz} = -\frac{\lambda_x}{\lambda_u} \frac{1 + K^2 / 2}{K^2 / 2} \frac{d \ln \gamma}{cdt}$$

## Soft x-rays

Wigner transform of the on-axis far field



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

Fourier transform limited pulse  
 ~ 1.5 fs (FWHM)

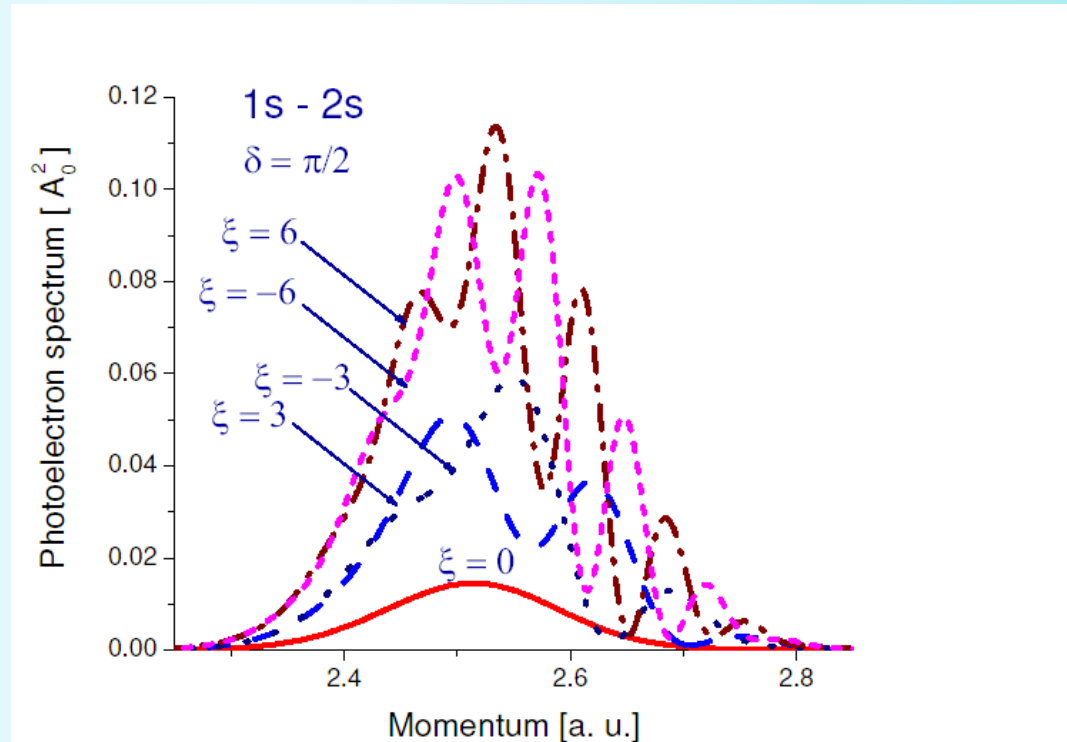
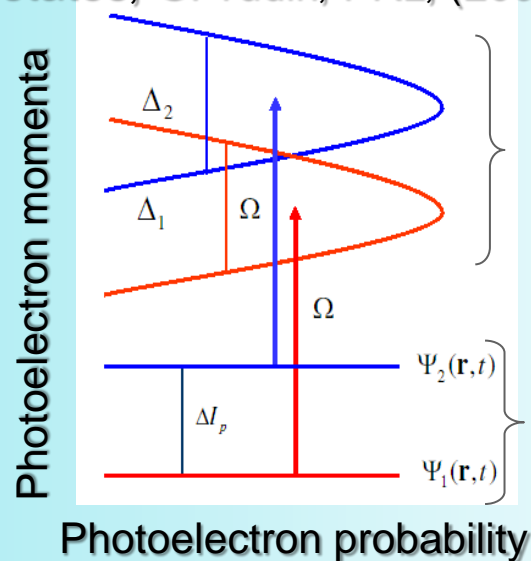
W.M. Fawley, Nucl. Inst. and Meth. A 593, 111(2008).

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

\*) E.L. Saldin, E.A. Schneidmiller, M.V. Yurkov, Phys. Rev. ST-AB 9, 050702 (2006).

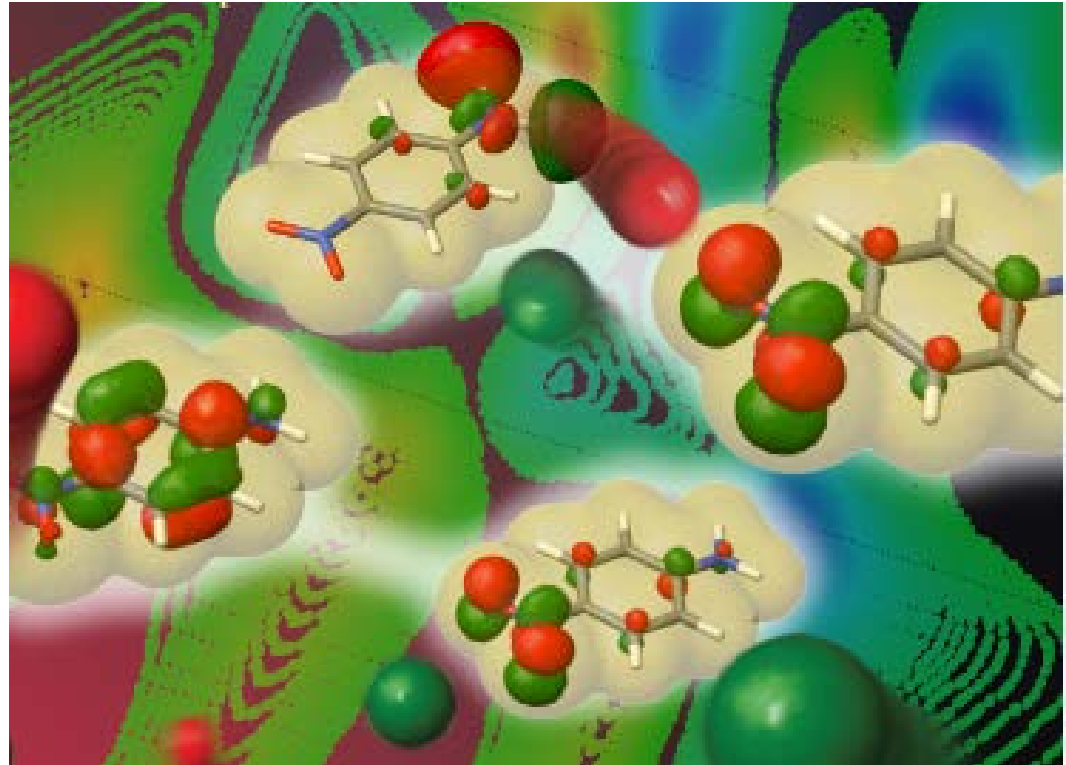
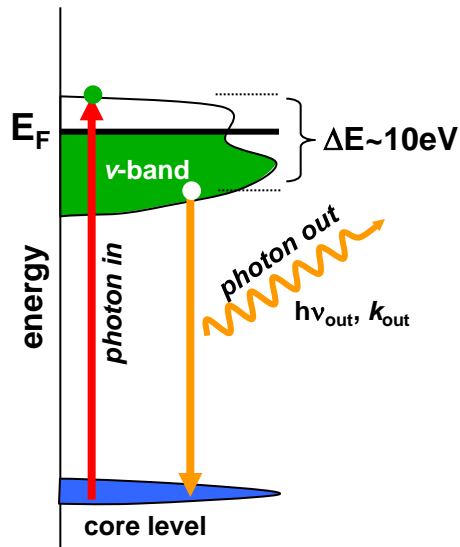
# The figure-of-merit is broad bandwidth of attosecond pulses

“Sudden” photoionization creates a coherent superposition of electronic states, G. Yudin, PRL, (2006)



# 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



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

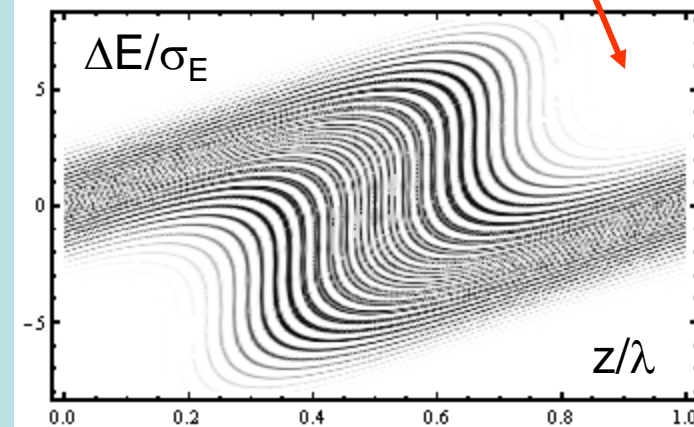
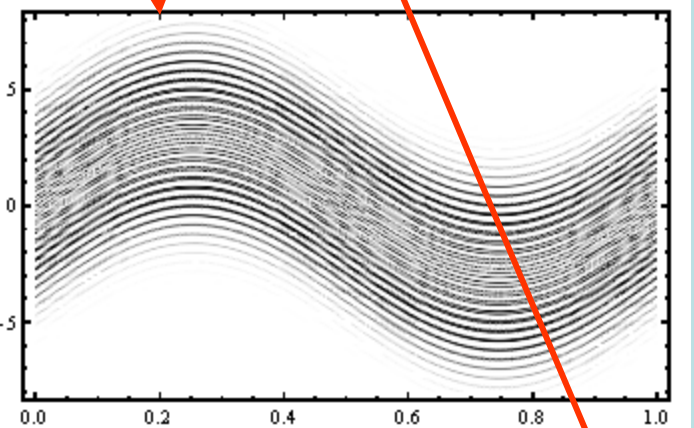
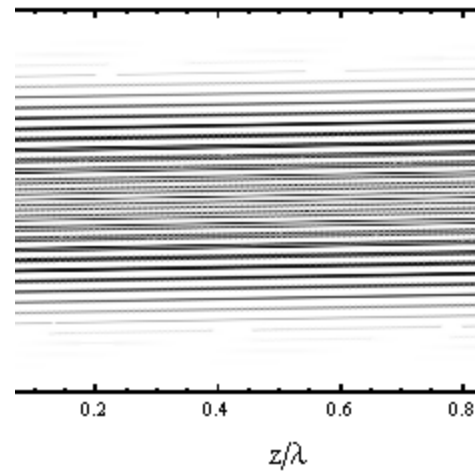
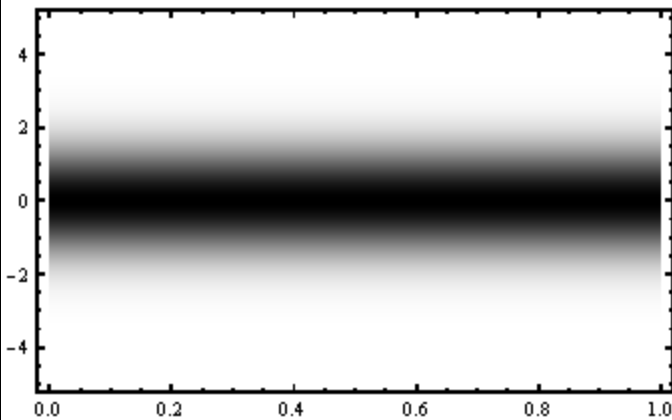
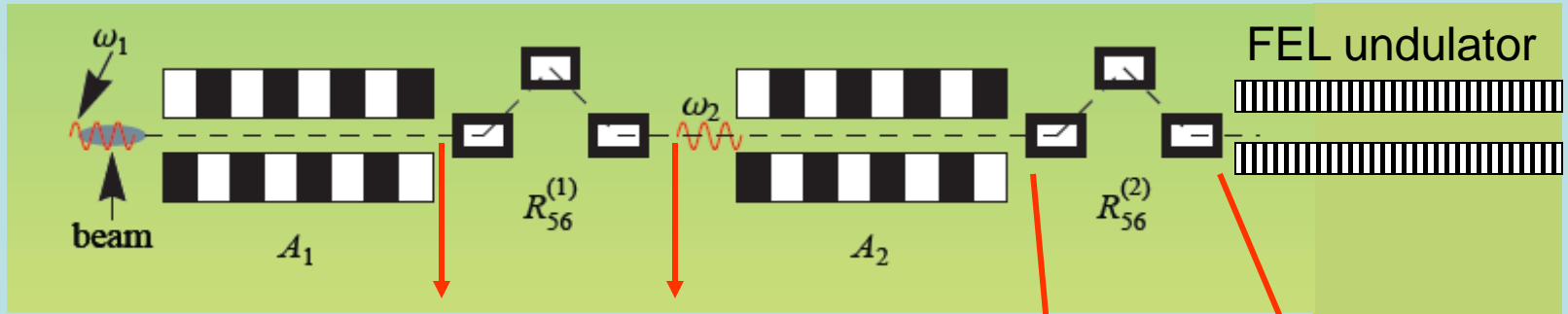
In molecules all electrons move in a combined potential of ion core and other electrons

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

\*) Schweigert, Mukamel, Phys. Rev. A 76, 012504 (2007)



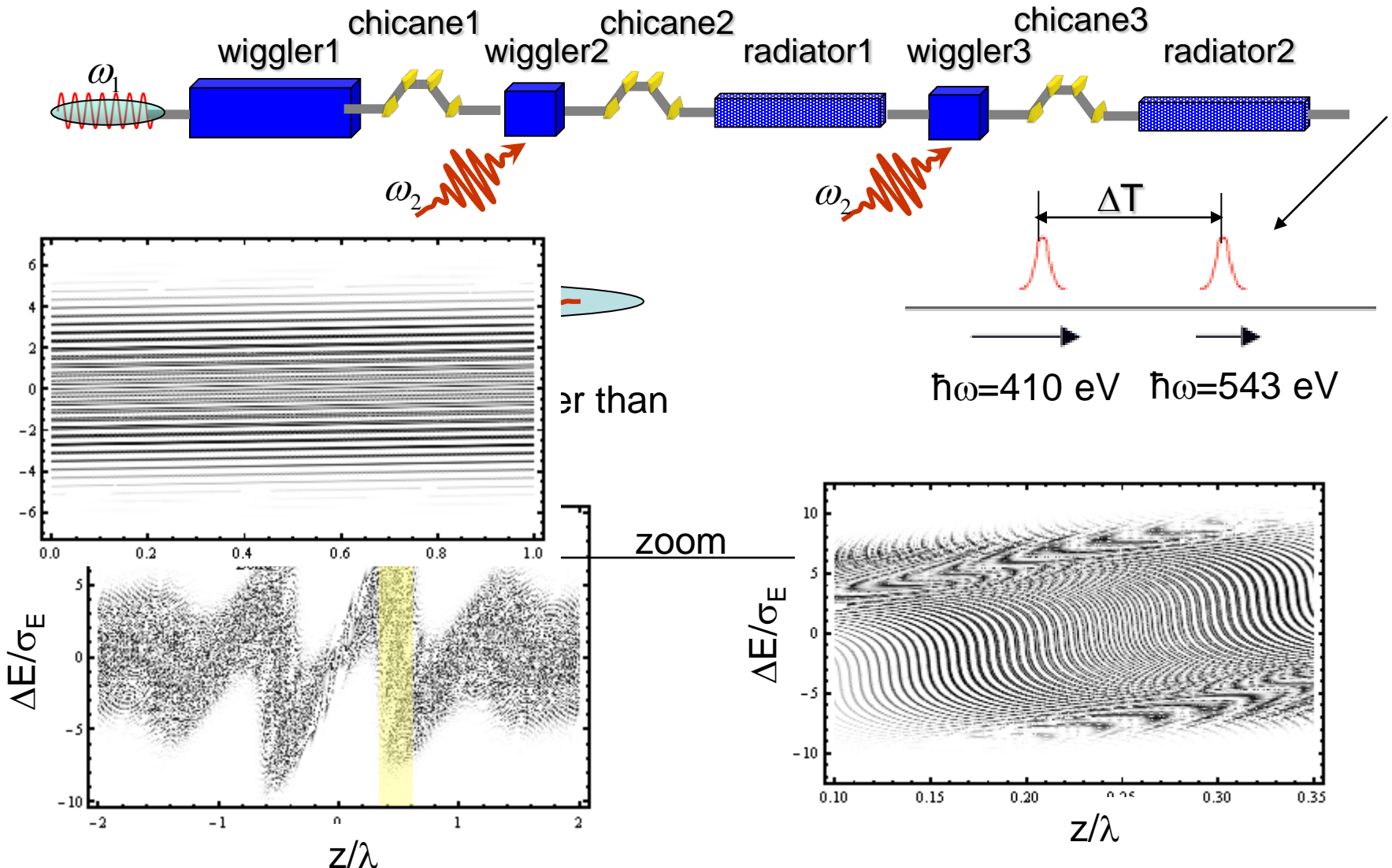
# Echo effect for harmonic generation of x-rays<sup>\*)</sup>



<sup>\*)</sup> G. Stupakov, PRL, 102, 074801(2009).

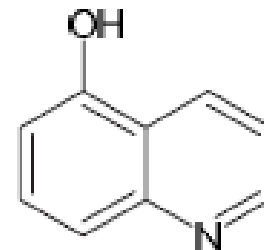
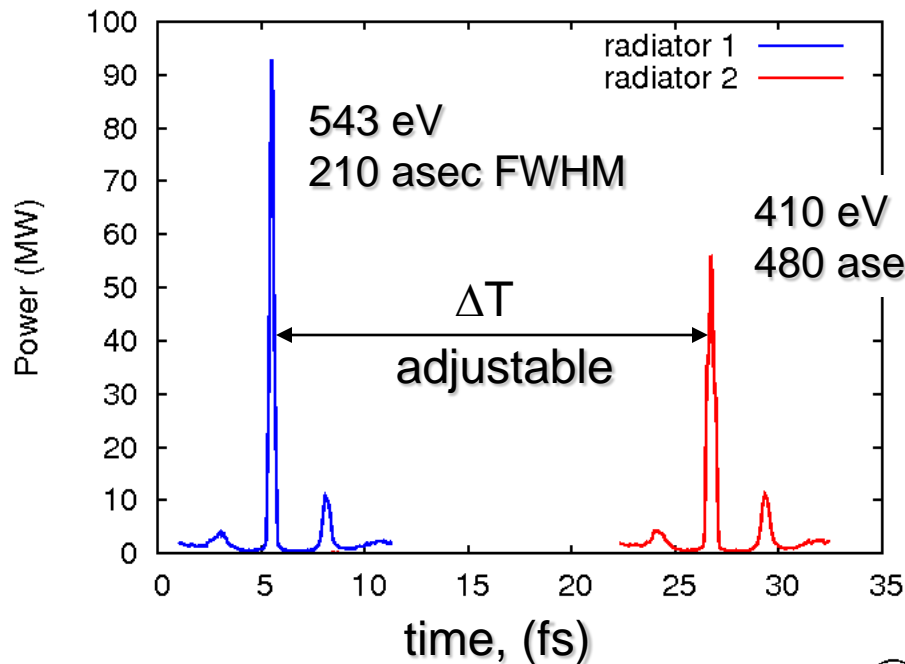


# Two color attosecond pump and attosecond probe x-ray pulses\*)

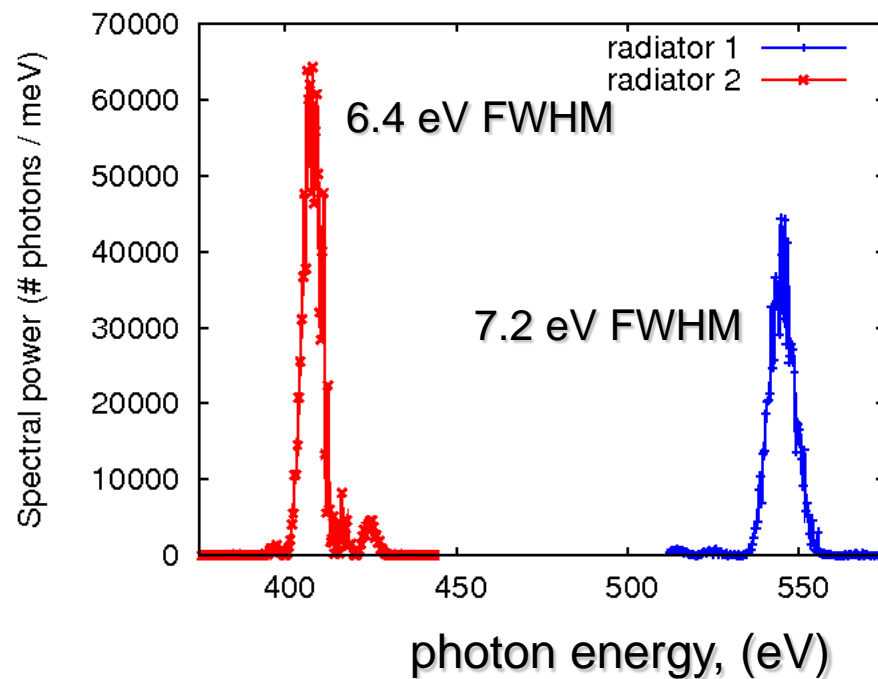


\*) Zholents, Penn, Nucl. Inst. and Meth. A 612, 254(2010).

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

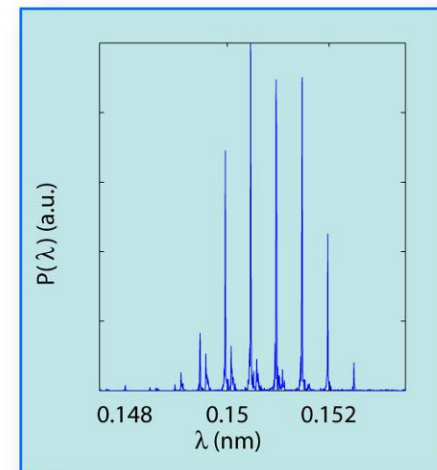
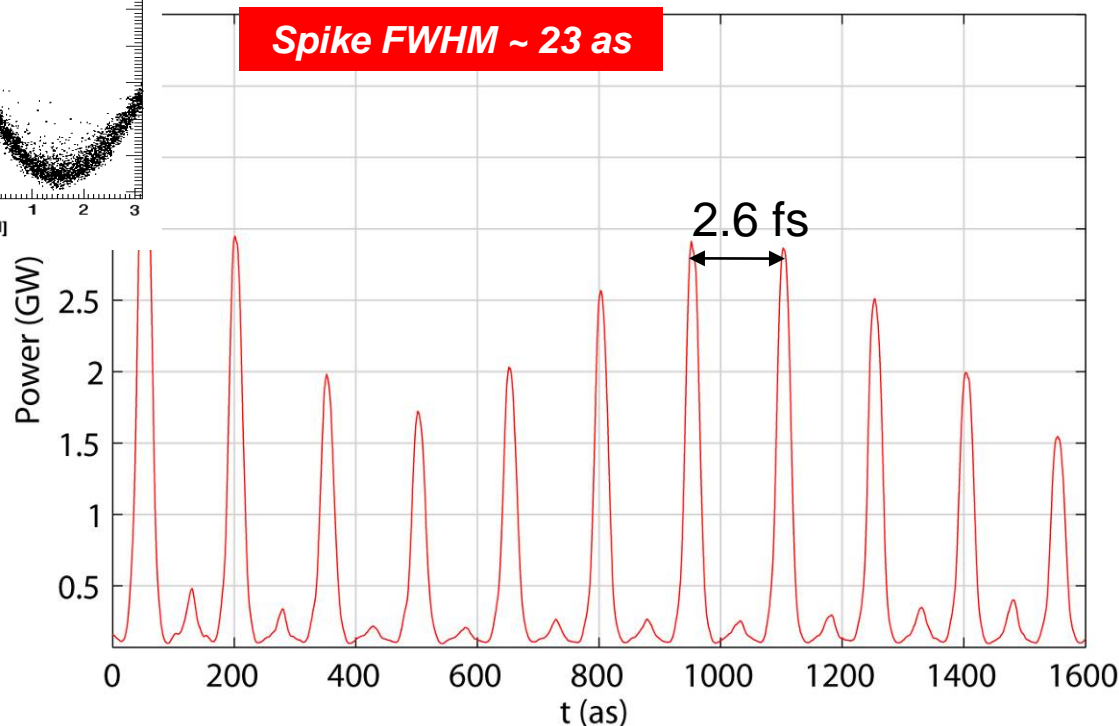
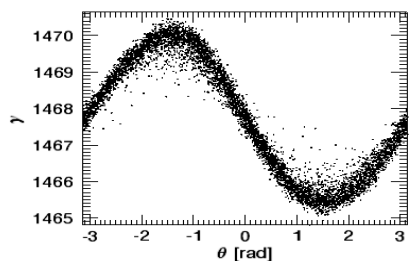
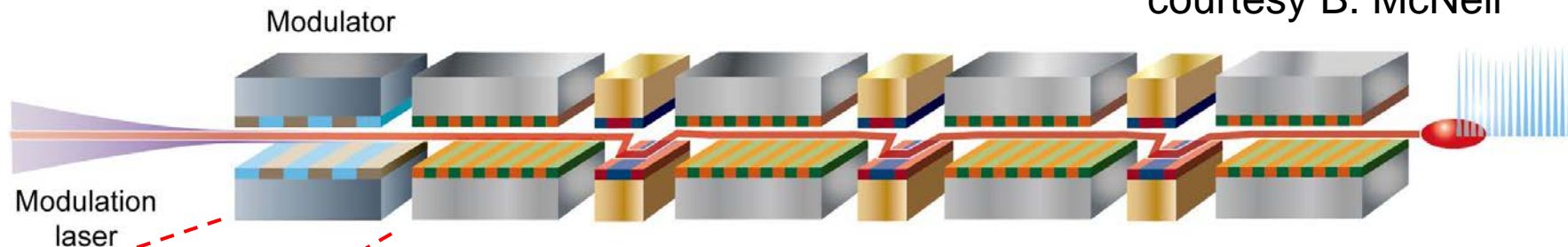


Molecule structure of 5-quinolinol



# X-ray SASE FEL amplifier with mode-locking produces a train of attosecond pulses\*

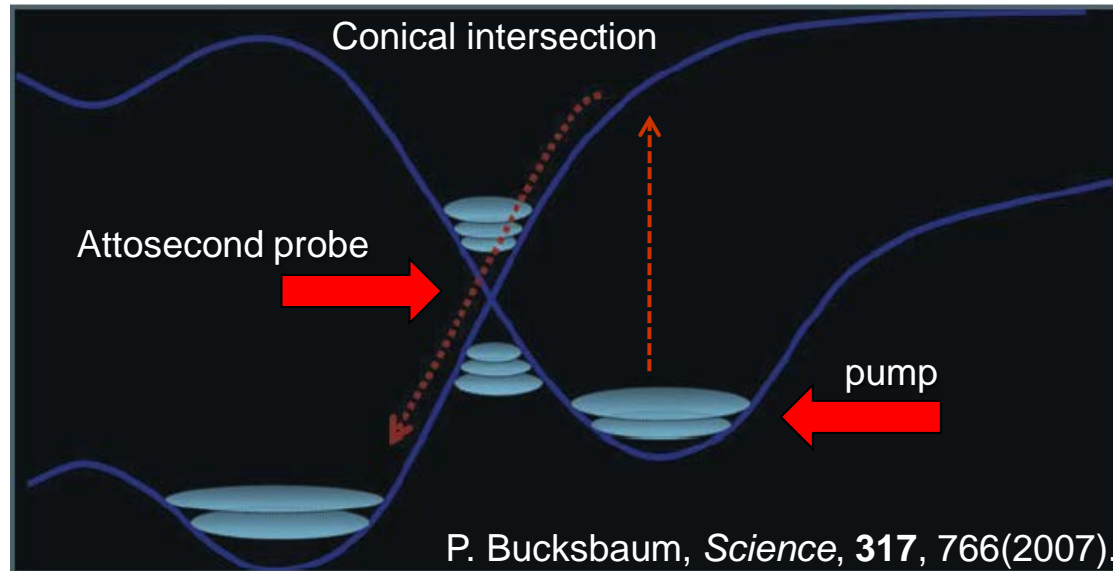
courtesy B. McNeil



\*) Thompson, McNeil, PRL, 100, 203901(2008);

Kur, Dunning, McNeil, Wurtele, Zholents, New Journal of Phys. 063012(2011).

# Summary: FELs – the light fantastic



Million times brighter  
and thousand times  
shorter than anything  
spontaneous emission  
can do

- 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. SACLA and FERMI are getting ready.
- Remarkably, adding attosecond x-ray pulse capabilities to existing FELs require rather modest modifications – in most cases a three pole wiggler and a carrier-envelope stabilized laser.

Thank you for your attention

