# Generation of Subpicosecond X-ray Pulses in Storage Rings

A. Zholents, LBNL, Berkeley, U.S.A.

Charles Shank

In 1981 Charles Shank and coworkers at Bell Labs redefined the term "ultrafast" when they reported the demonstration of subpicosecond pulses from colliding-pulse dye laser [1]

1) R.L. Fork, B.I. Greene, C.V. Shank, *Appl. Phys. Lett.*, 38, 671(1981).

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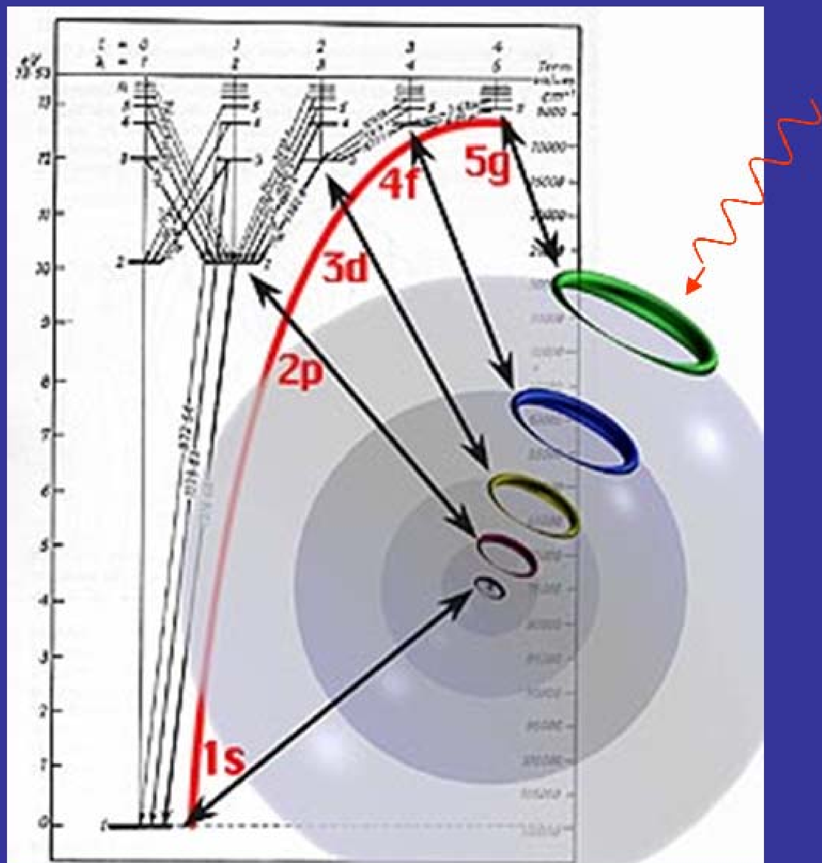


**Probe valence electrons with optical laser**

1) R.L. Fork, B.I. Greene, C.V. Shank, *Appl. Phys. Lett.*, 38, 671(1981).

Charles Shank

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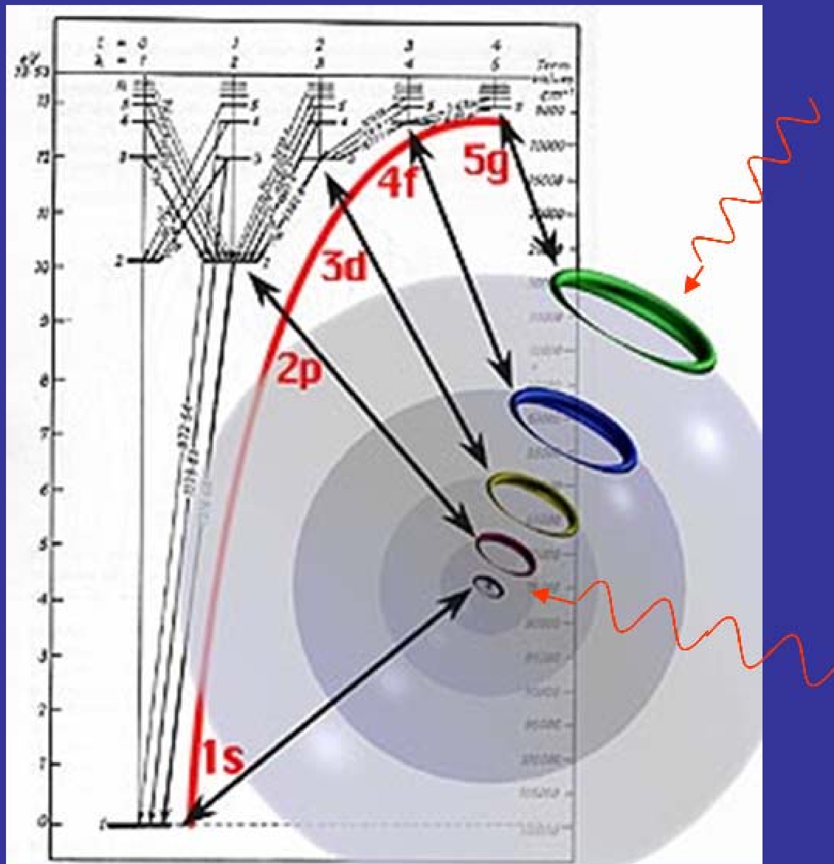
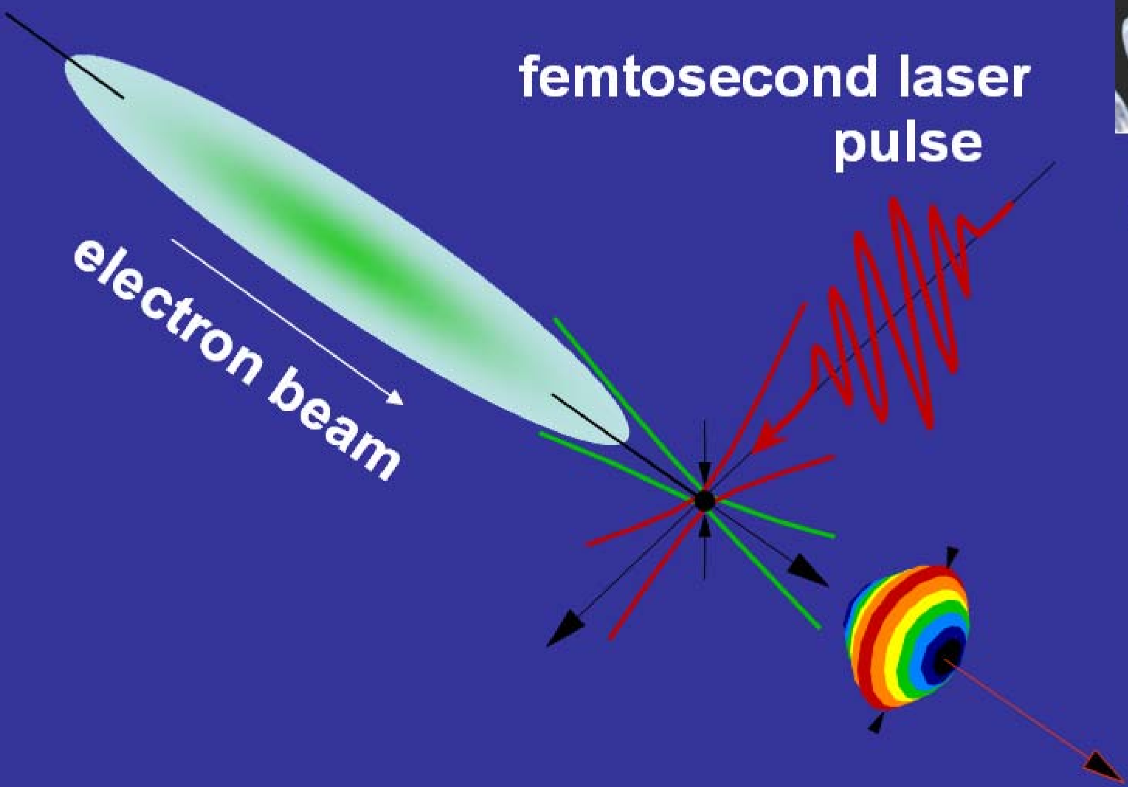



**Probe valence electrons with optical laser**

**Probe core electrons with x-rays**


1) R.L. Fork, B.I. Greene, C.V. Shank, *Appl. Phys. Lett.*, 38, 671(1981).

# 90° Thomson scattering[1]




sub-ps x-ray
x-ray pulse

300 fs
$10^5$ ph/pulse

Swapan Chattopadhyay

Kwang-Je Kim

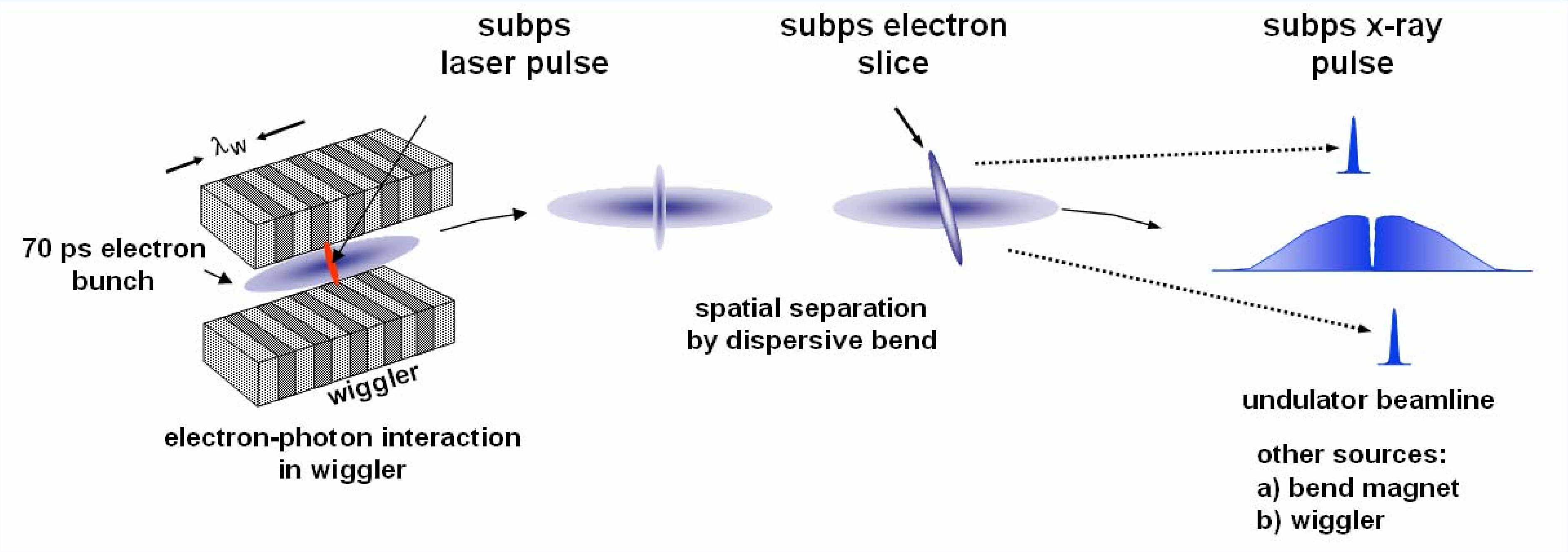

1) S. Chattopadhyay, K.-J. Kim, C. Shank, *Nucl. Instr. Meth.*, **A341,** 351 (1994).
2) R.W. Schoenlein et. al., *Science*, **274(11),** 236 (1996).

# What this experiment taught us ?

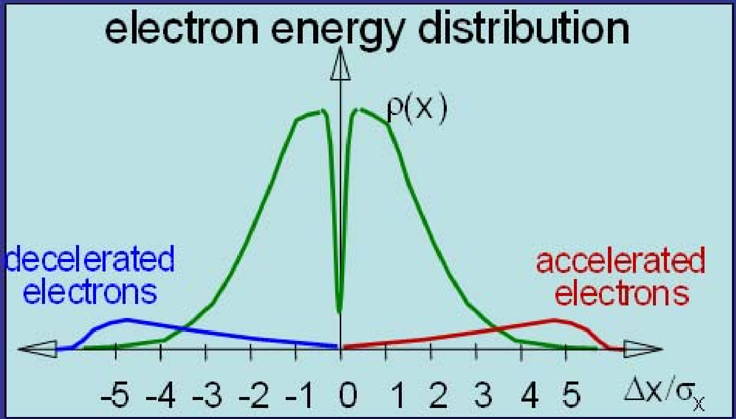
Don’t squeeze electron bunch, take part of it

Use laser pulse,  it is the shortest thing

# “Slicing” of sub-ps x-rays pulses in storage ring[1]




jitter free -
x-ray pulse is
synchronous to
the laser pulse



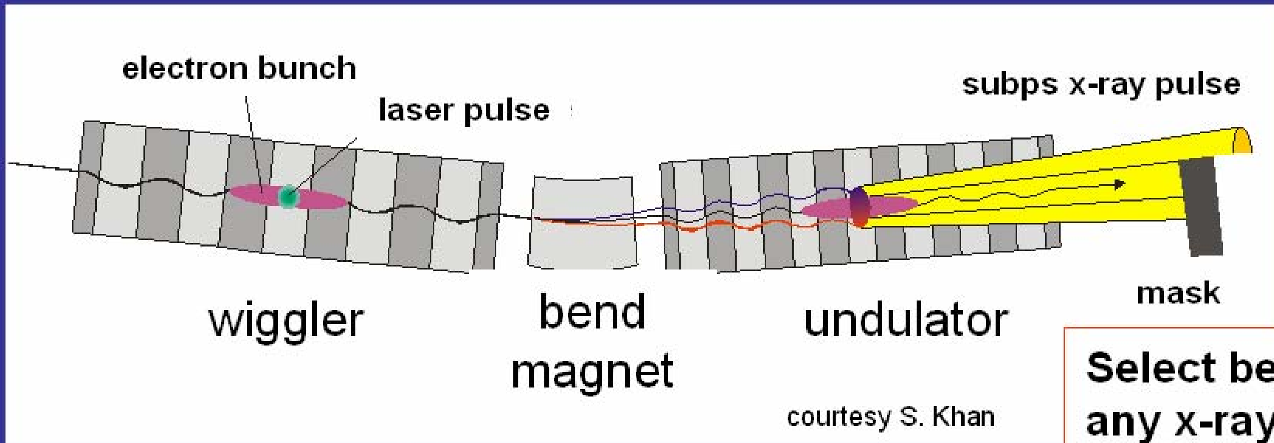

Max Zolotorev

1) A. Zholents, M. Zolotorev, *Phys. Rev. Lett.*, **76,** 912 (1996).

# Selection of sub-ps x-ray pulses[1]

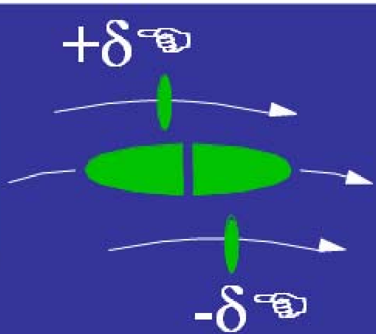
Besides coordinate, one can use other means to select subps pulse

angular selection




**Select before any x-ray optics mirror**

time-off-flight selection




spectral selection (works only with undulator source)[2]

$$\frac{\delta\omega(\varepsilon)}{\omega_0} >> \frac{1}{n\,N}$$

← Bandwidth of the undulator radiation at harmonic ***n***

1) R.W. Schoenlein, *et al., Appl. Phys.* **B71**, 1 (2000).
2) H. Padmore, private discussion

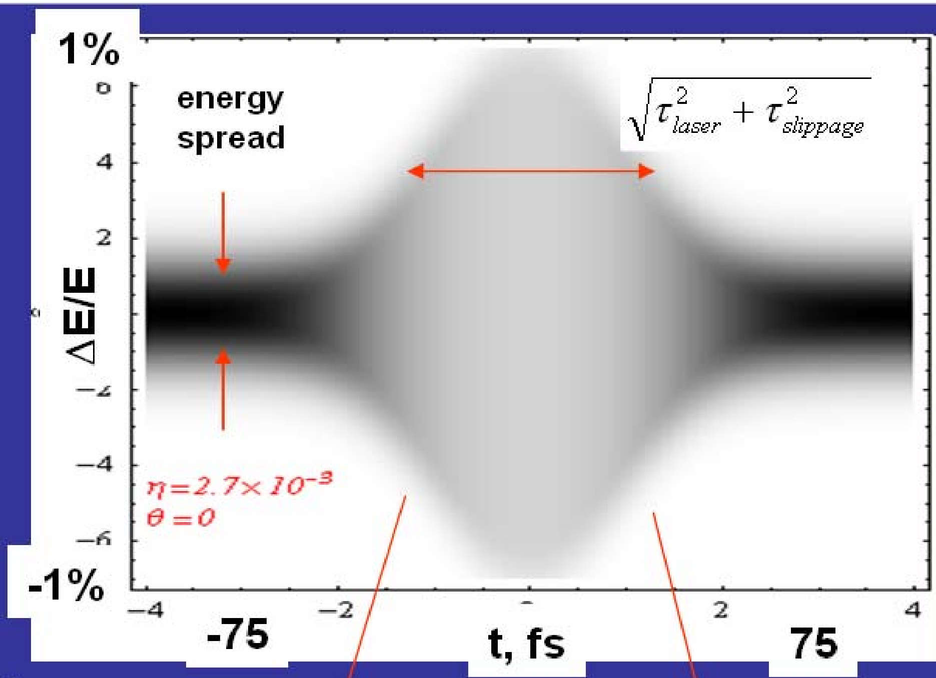
# Energy modulation

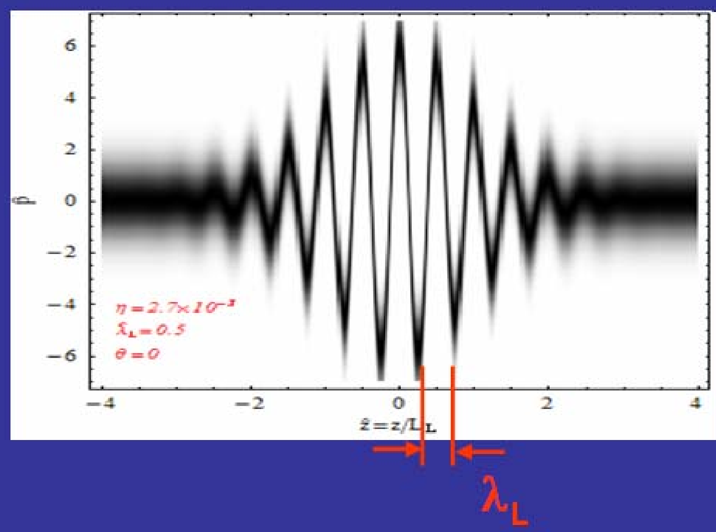
$$\Delta E(t,r) \approx \Delta E_0(r) e^{-\frac{t^2}{2\sigma_t^2}} \cos(\omega_L t)$$

$$\sigma_t = \sqrt{\tau_{laser}^2 + \tau_{slippage}^2} / 2.35$$

$$\Delta E_0(r) \approx \sqrt{\frac{10}{137} A_L \hbar\omega_L}\, e^{-\frac{r^2}{w_0^2}}$$

laser pulse energy ($A_L$) photon energy ($\hbar\omega_L$)




**What 1 mJ, 50 fs Ti:Sa laser pulse will do at:**

| | Beam energy (GeV) | Number of wiggler periods | ΔE/E (%) |
|---|---|---|---|
| ALS | 1.9 | 29 | 1.3 |
| BESSY | 1.7 | 10 | 1.0 |
| SLS | 2.4 | 17 | 0.9 |

# Pulse lengthening and duration of the x-ray pulse

- **It is imperative for this technique that magnetic dispersion is used to assist the selection of subps x-ray pulses**,

  i.e there must be a non achromatic lattice between modulator and radiator

- Non achromatic lattice is also a non isochronous lattice,

  i.e. $R_{51}$ or $R_{52}$ or both $\neq 0$ in addition to $R_{56} \neq 0$ ,

$$(\Delta \ell = R_{51} x + R_{52} x' + R_{56} \delta)$$

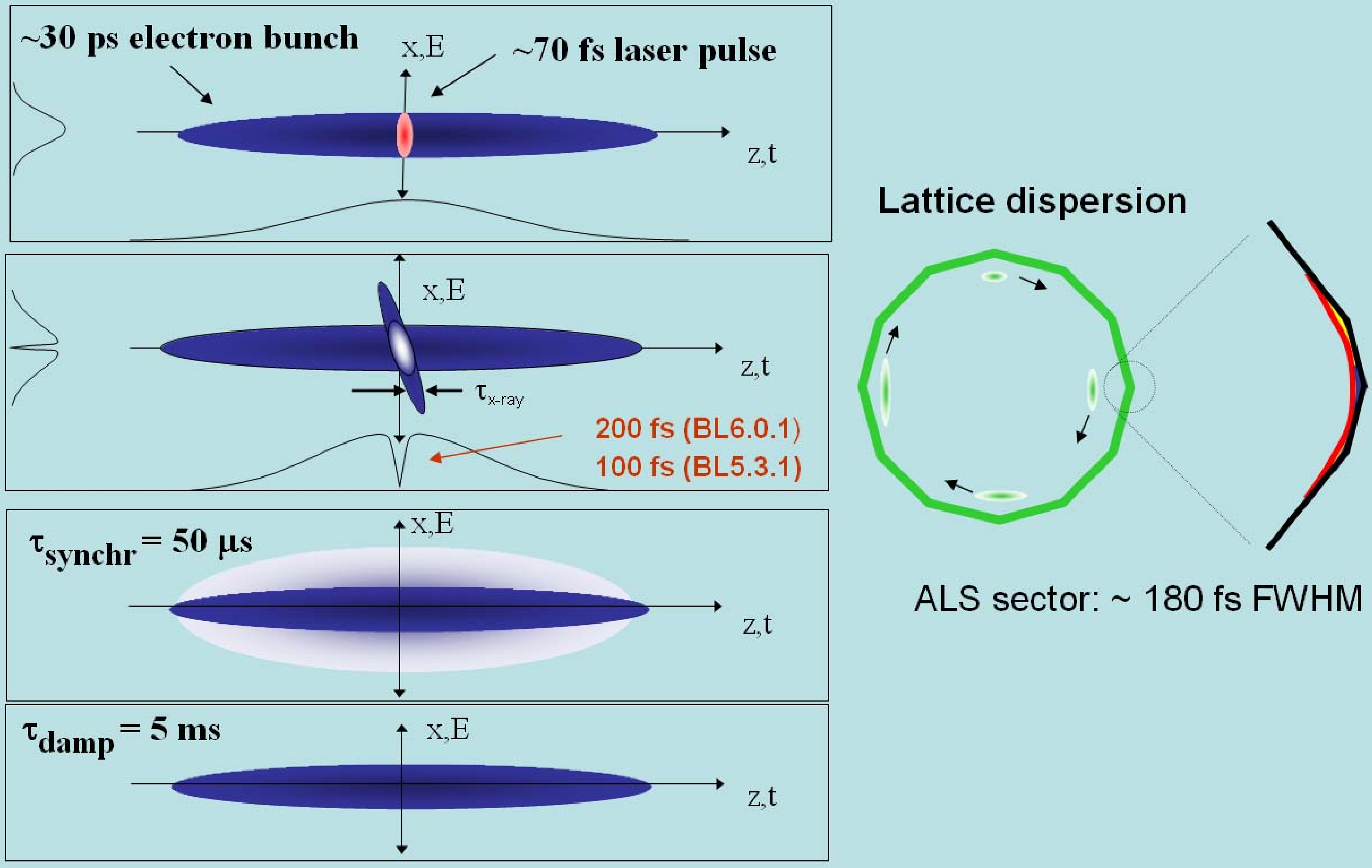
- Thus pulse widening happens when e-beam propagates from modulator to radiator

**Various contributions to the duration of the x-ray pulse**

| | Laser pulse (field), (fs) | Slippage in wiggler, (fs) | Lattice dispersion, (fs) | X-ray pulse (fs) |
|---|---|---|---|---|
| **ALS** | **100** | **70** | **180** | **~200** |
| **BESSY** | **70** | **26** | **65** | **~100** |
| **SLS** | **70** | **45** | **125** | **~150** |

BERKELEY LAB

# Evolution of Modulated Electron Beam



# Advanced Light Source (1997-2000)[1]

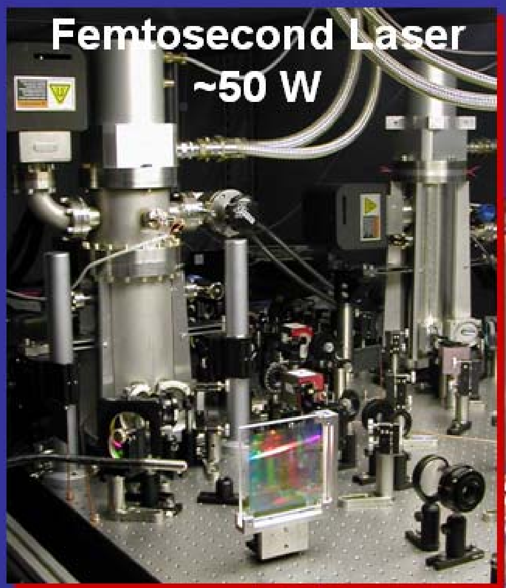

Henry Chong


Ernie Glover

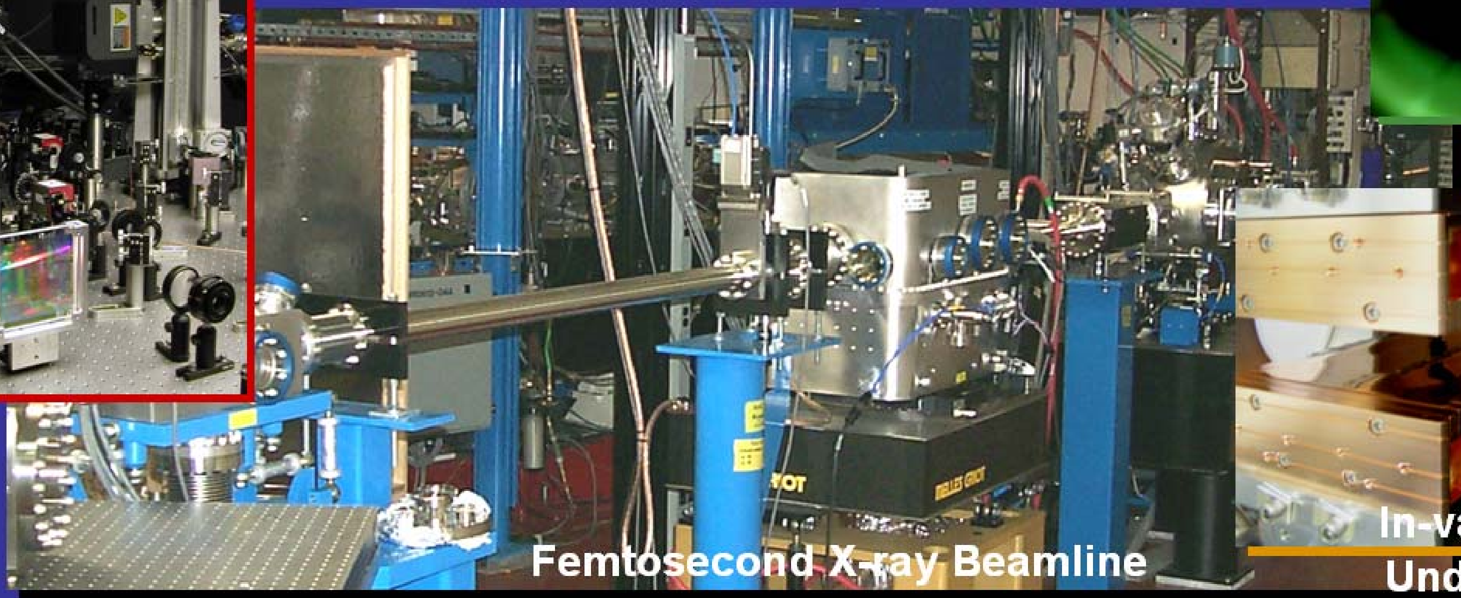

Phil Heimann

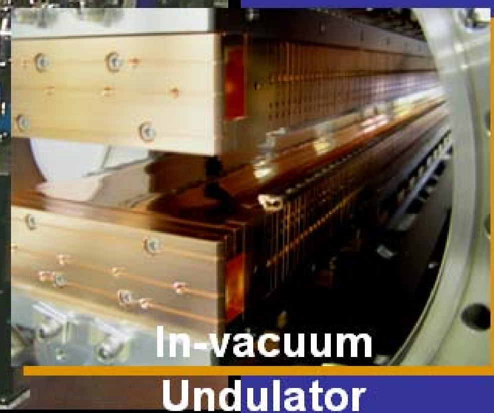

Bob Schoenlein


Max Zolotorev



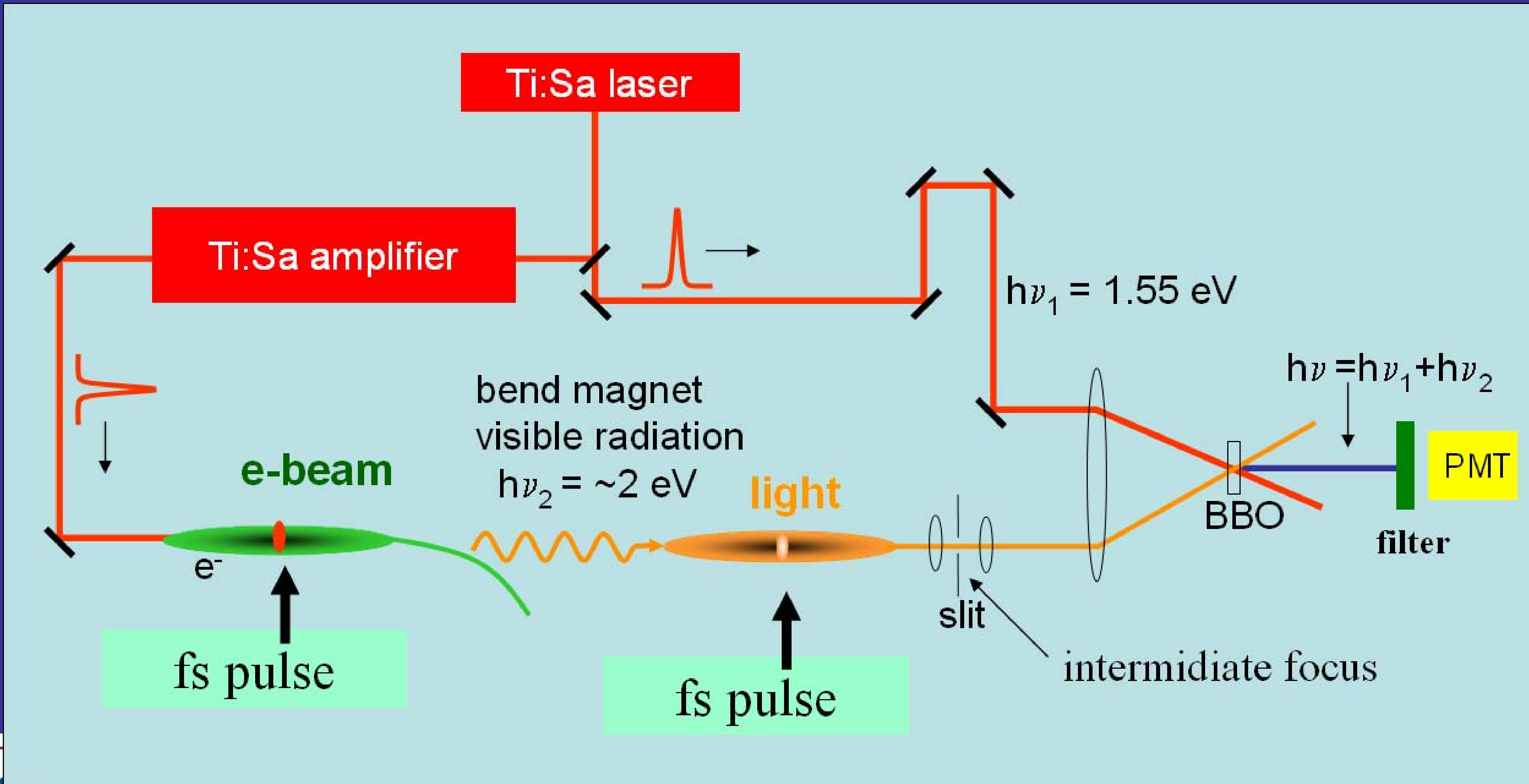

1) R.W.Schoenlein *et al* , Science, March 24, (2000)

# Diagnostics

**Measurement of a short pulse of synchrotron radiation via cross-correlation with a short laser pulse**



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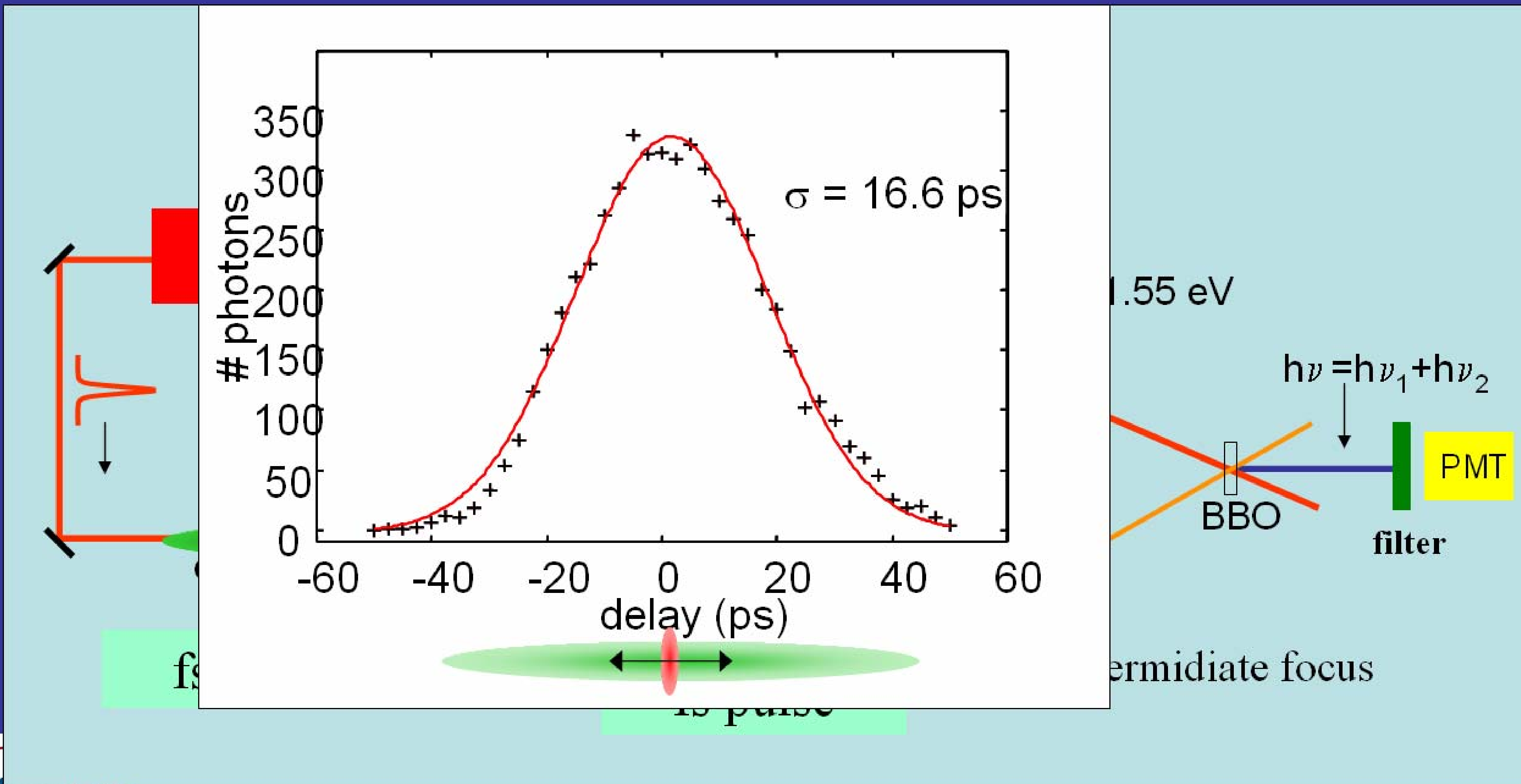
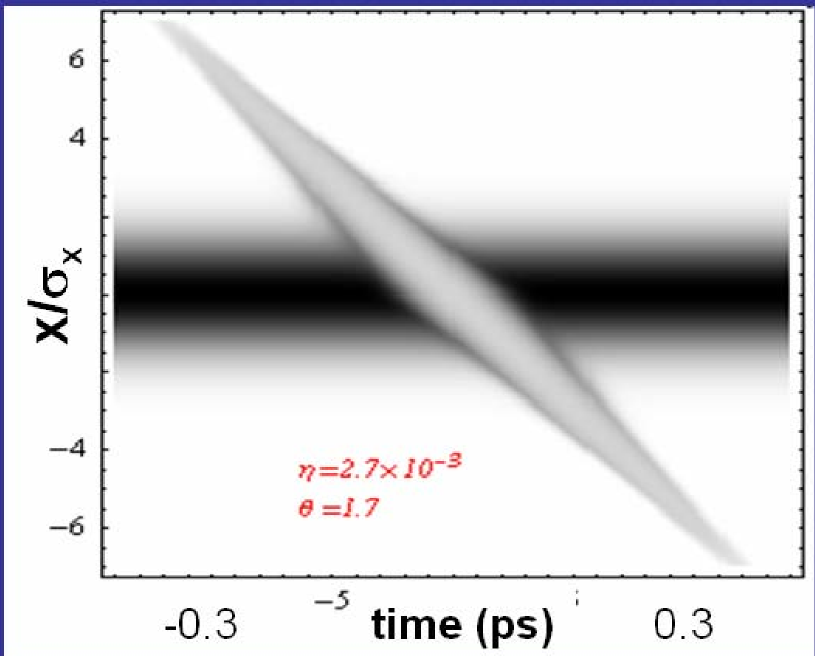
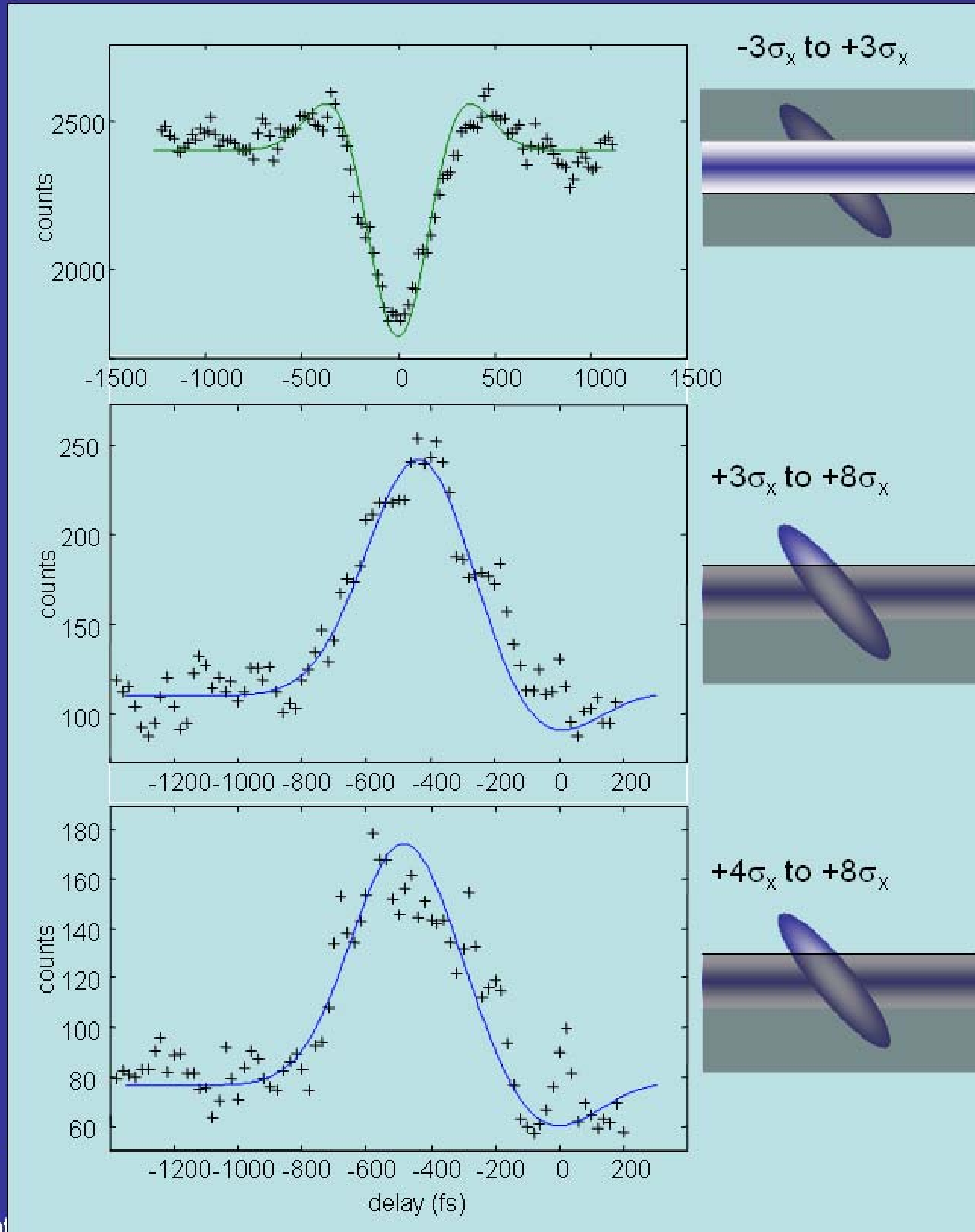


# Diagnostics (2)

## ALS bend magnet beamline

### Electron Density Distribution



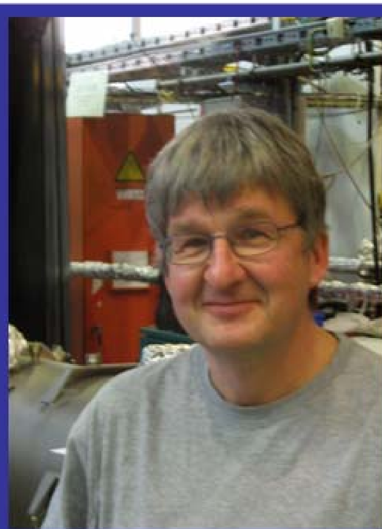




**Experimental data pointed to modulation amplitude of 6.4 MeV (expected ~ 10 MeV)**

# BESSY [1]

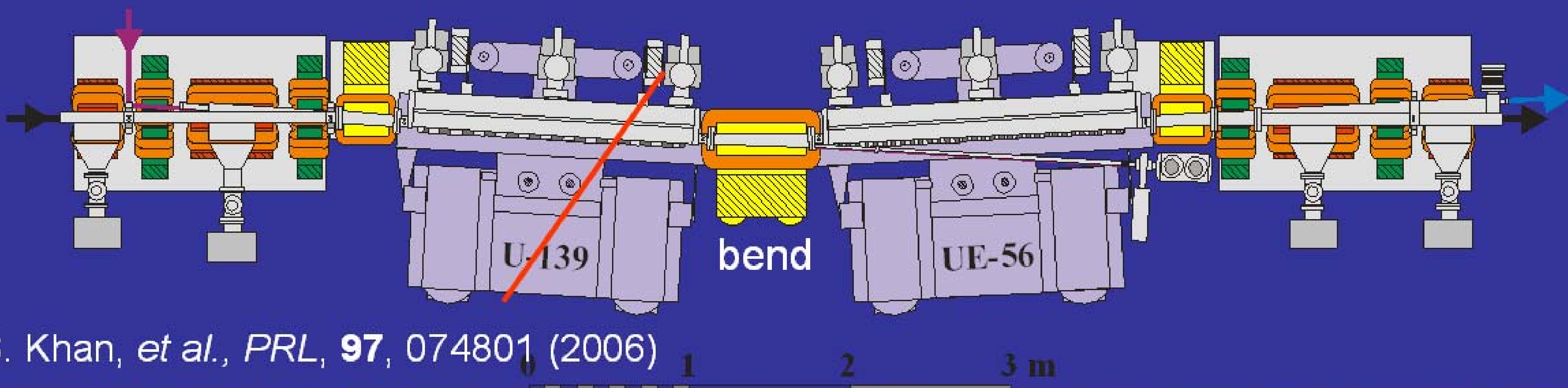
Karsten Holldack

Shaukat Khan

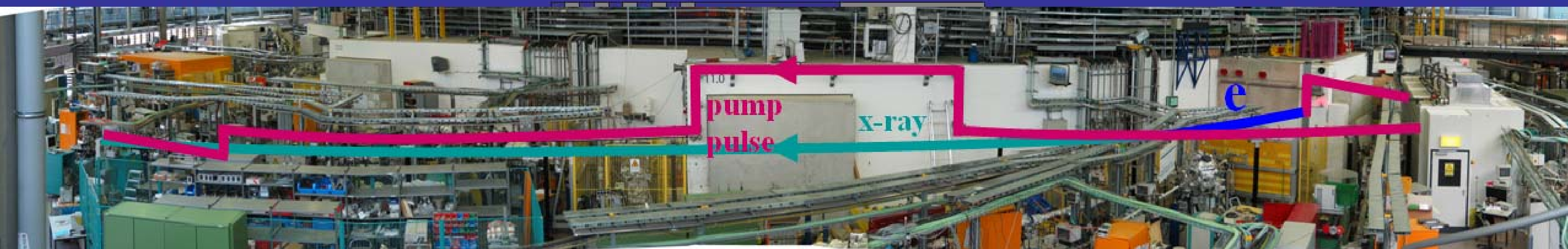


Rolf Mitzner

Torsten Quast

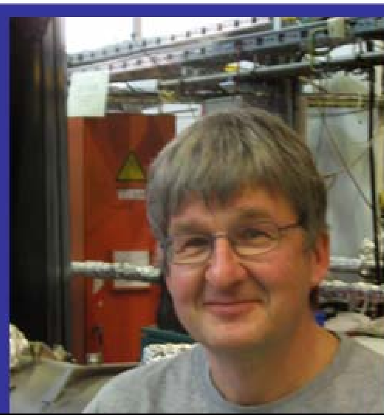



1) S. Khan, *et al.*, *PRL*, **97**, 074801 (2006)



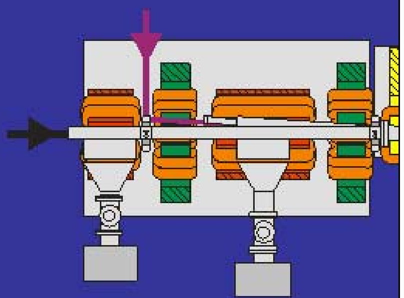
# BESSY [1]

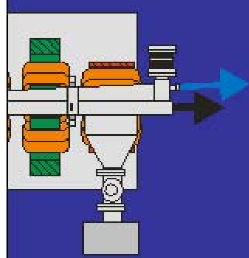
T. Quast et al., “Upgrade of BESSY fs  facility”, THPLS015

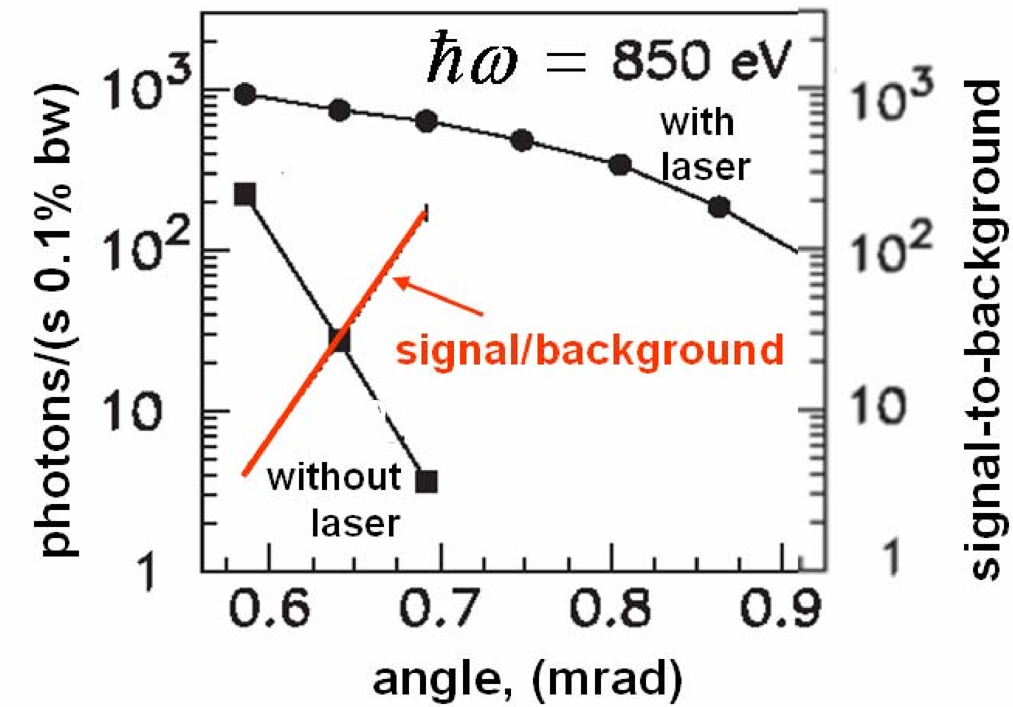



Karsten Holldack

n Quast

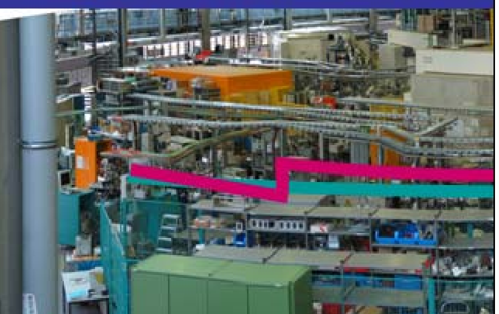







**Detected photon rate per 0.1% bandwidth versus cutoff angle**

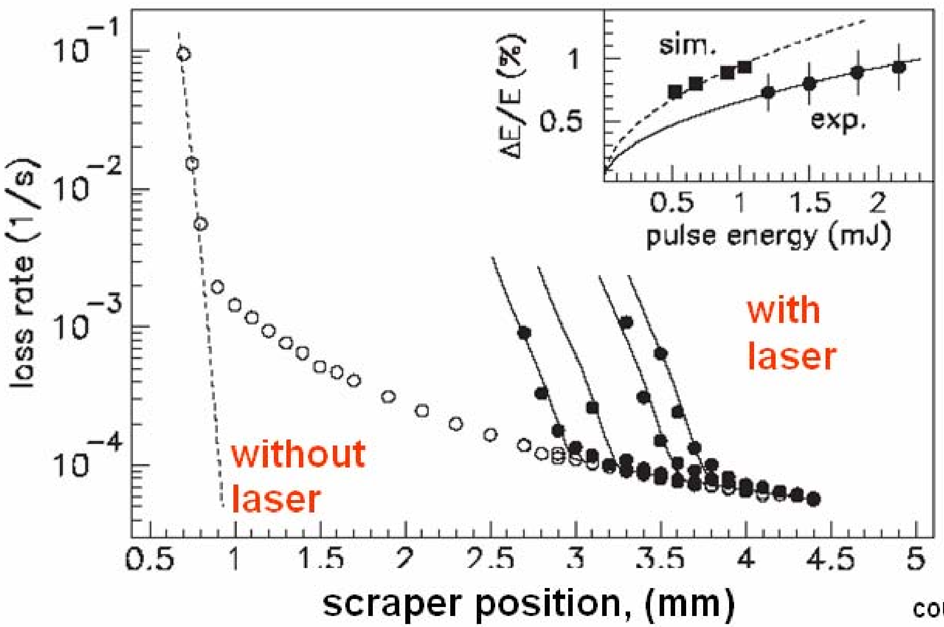
1) S. Khan, *et al.*, *PRL*, 9



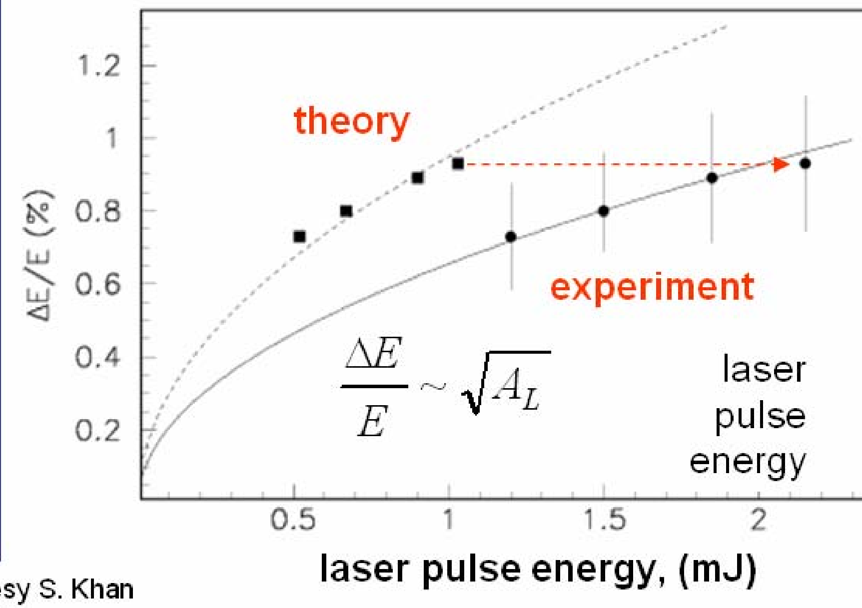
# Diagnostics (3)

Deducing energy modulation amplitude from beam lifetime measurements using scraper in dispersive location (BESSY)

Electron loss rate versus scraper position




Energy modulation amplitude versus laser pulse energy



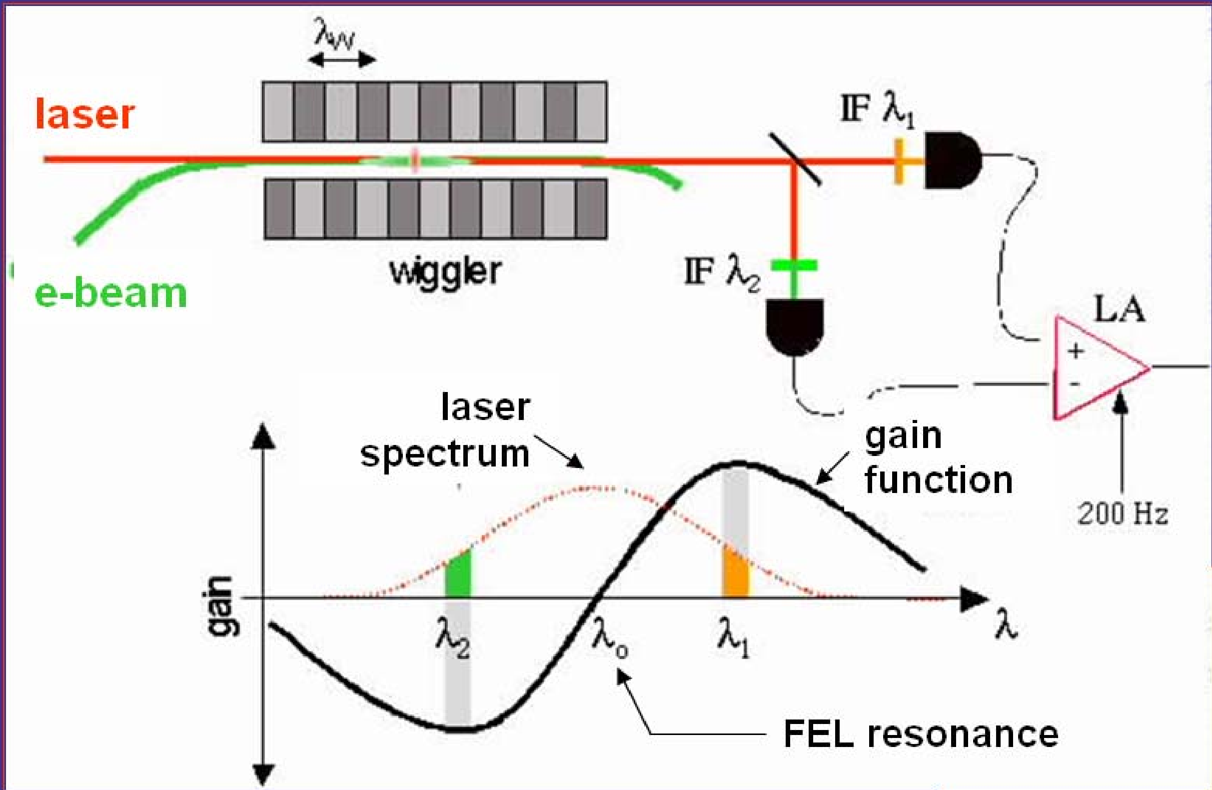

courtesy S. Khan

Approximately two times more laser pulse energy was used than it was predicted (same problem at ALS)

Not explained so far !

# Small-signal FEL gain

## Measurement of FEL gain through wiggler at ALS

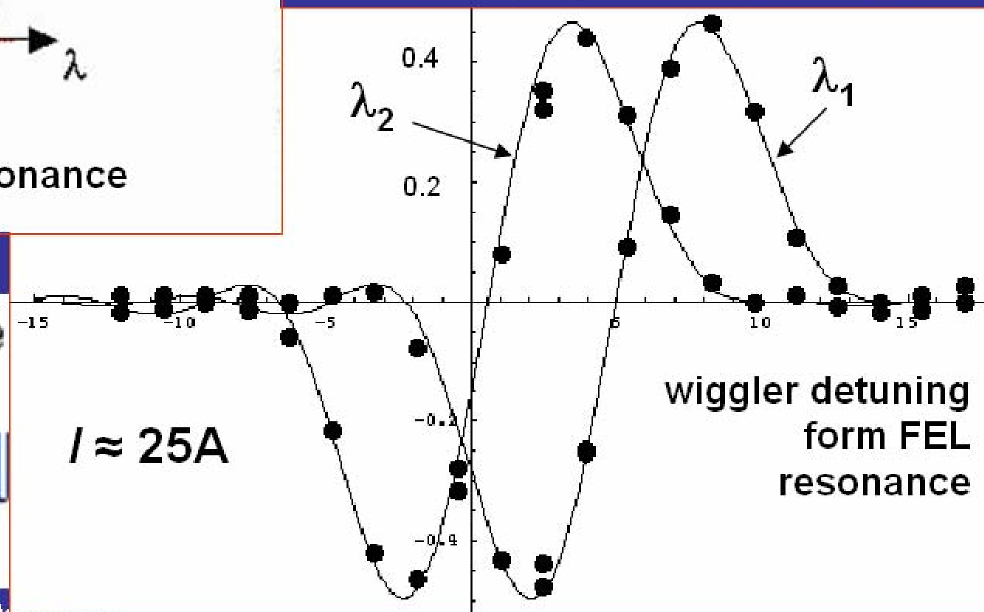


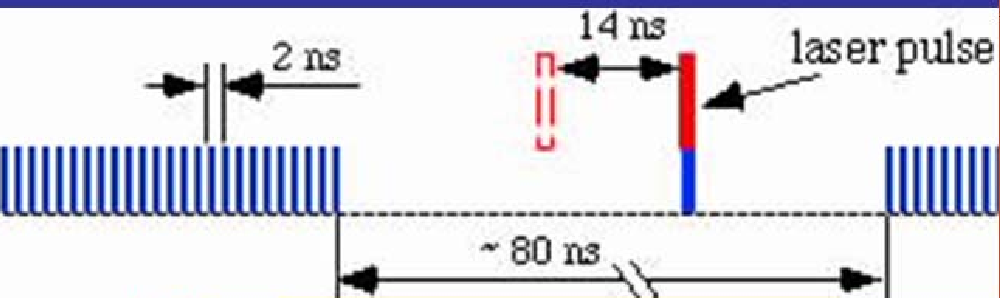

Madey's theorem

$$\text{FEL gain} \sim \frac{d}{dE}\left\langle \Delta E^2 \right\rangle$$

measured with laser oscillator

Gain, %

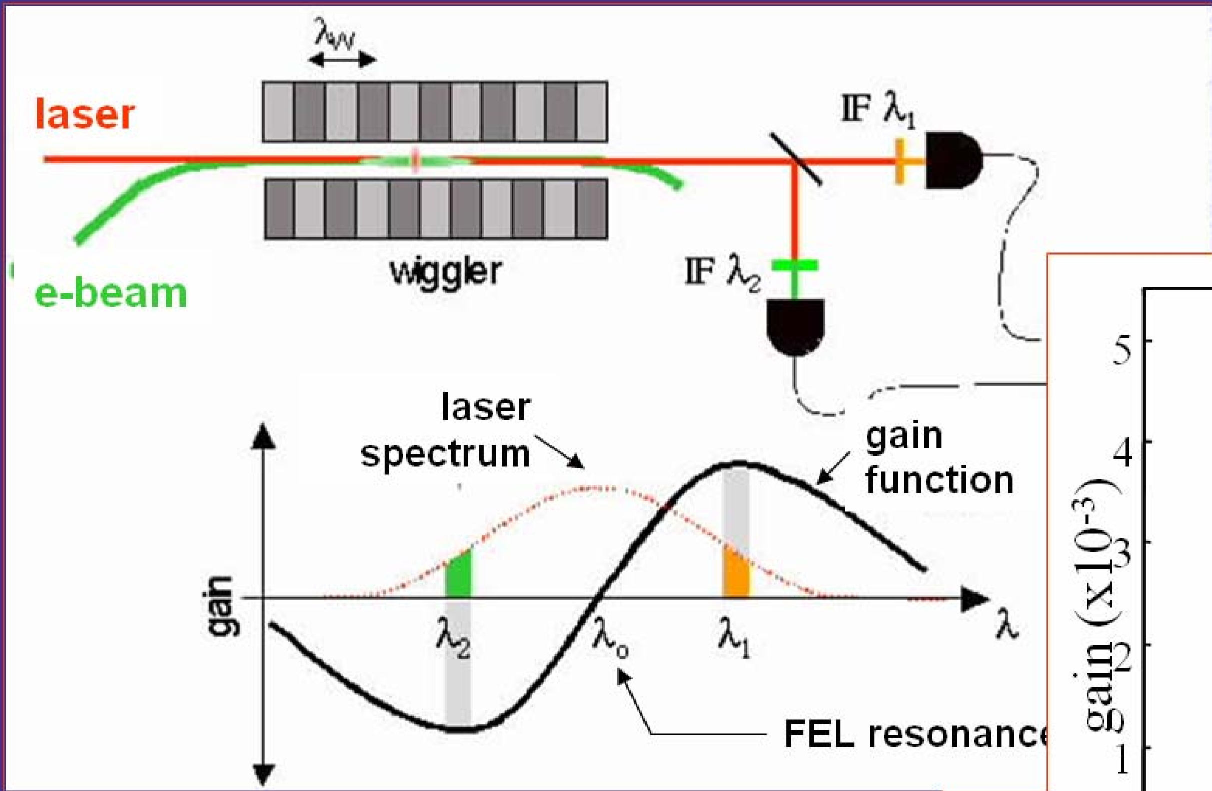



bunch train and a gap

# Small-signal FEL gain

## Measurement of FEL gain through wiggler at ALS

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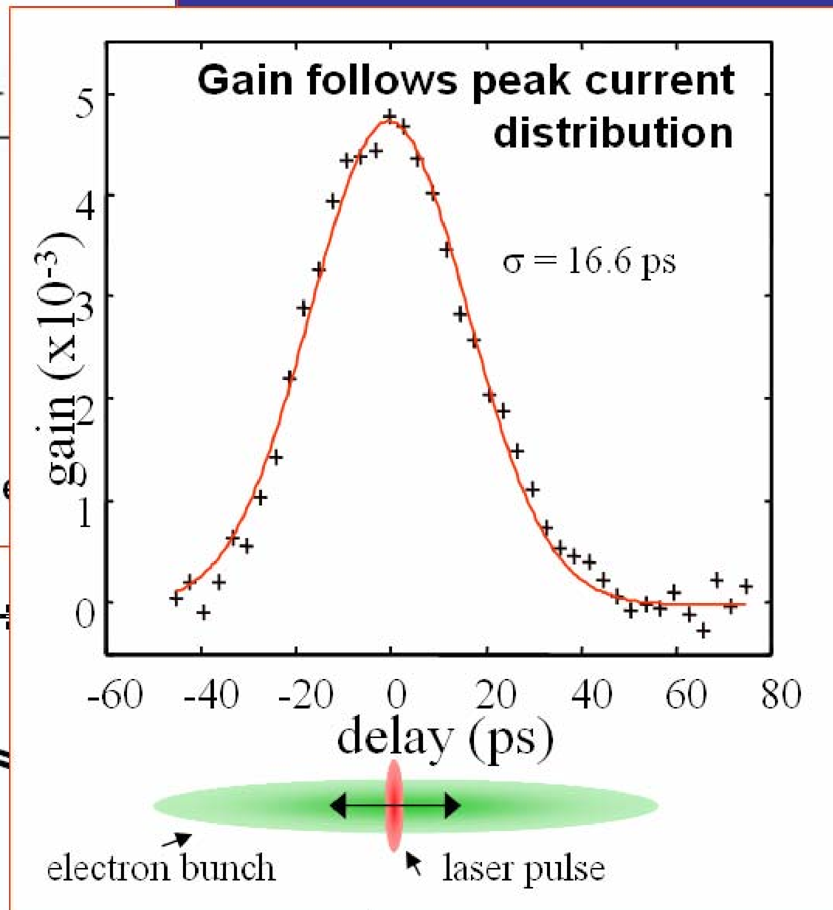


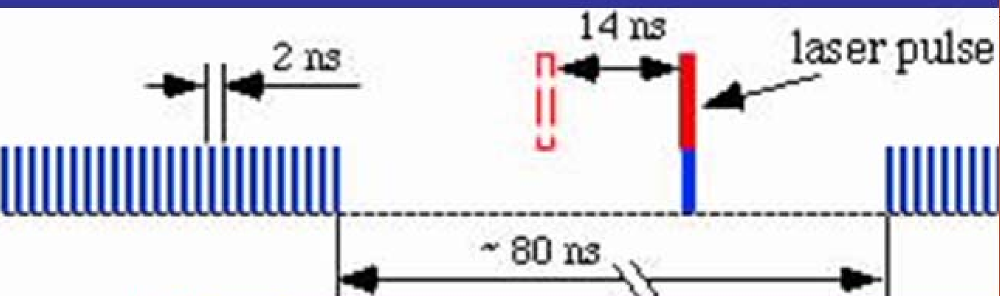


bunch train and a gap

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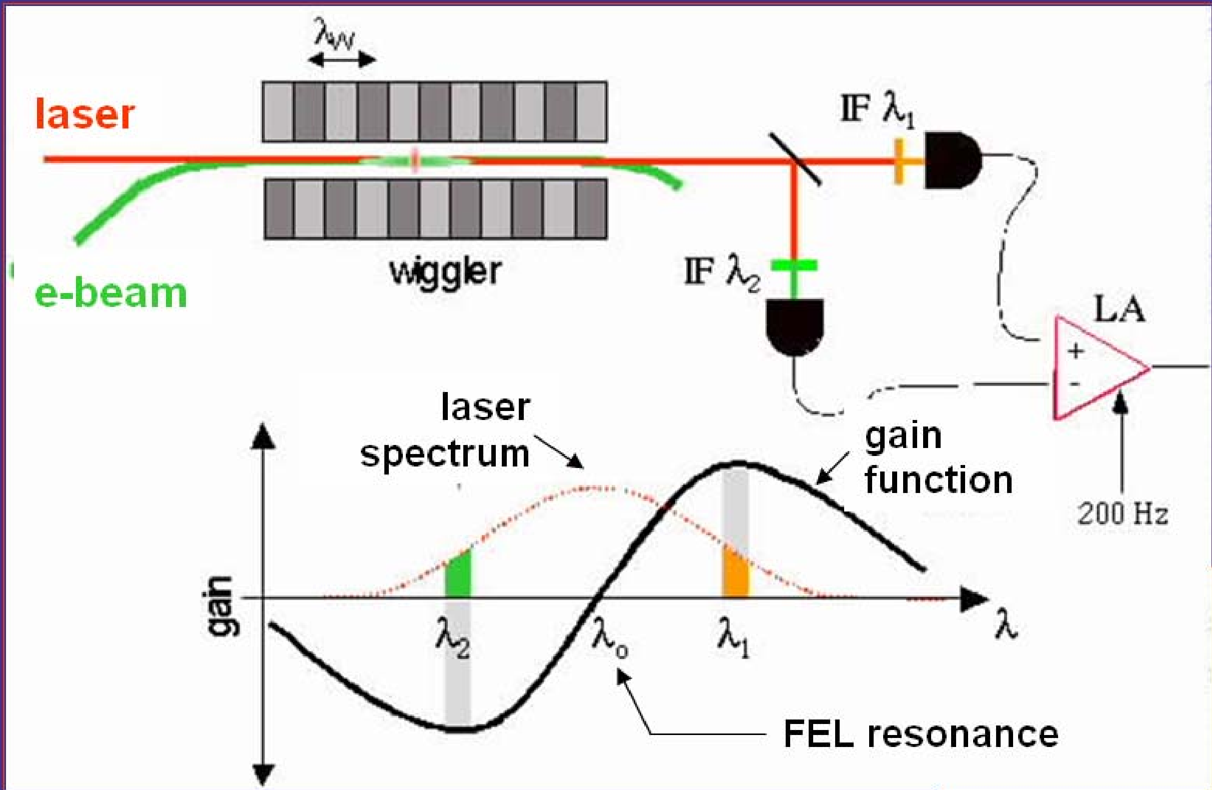
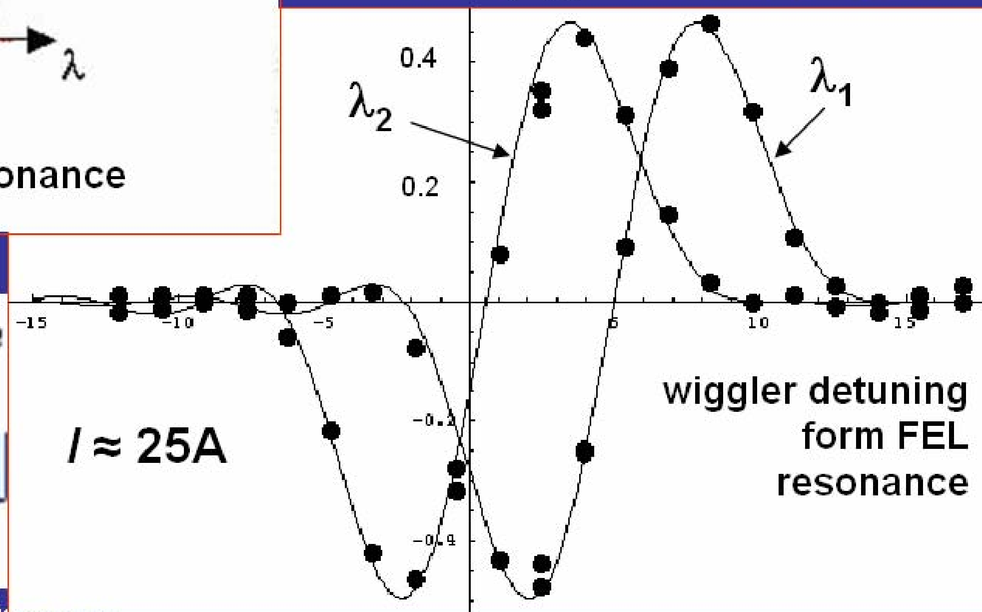


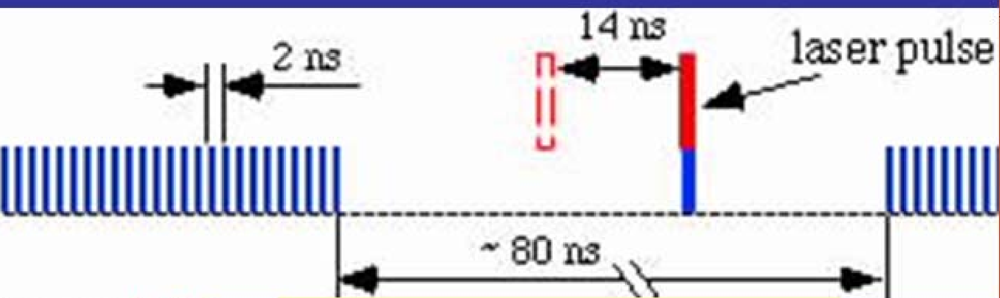

Madey's theorem

$$\text{FEL gain} \sim \frac{d}{dE}\left\langle \Delta E^2 \right\rangle$$

Gain, %

measured with laser oscillator

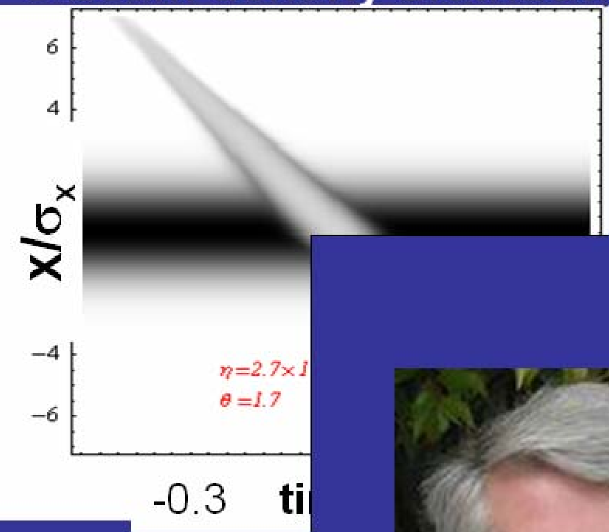



bunch train and a gap

# Subps, laser-induced coherent THz radiation[1]

e-beam density distribution



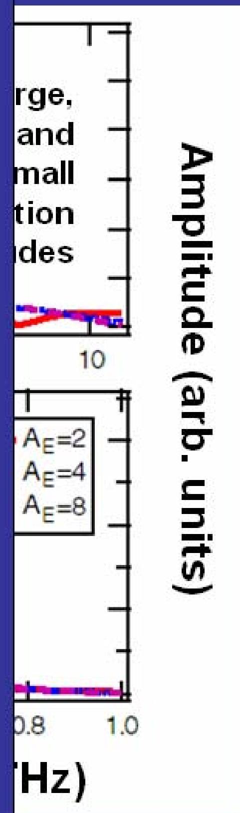

e-beam density distribution

$$I(\omega) \sim \int \rho(t) e^{i\omega t} dt$$

Michael Martin

Fernando Sannibale

John Byrd

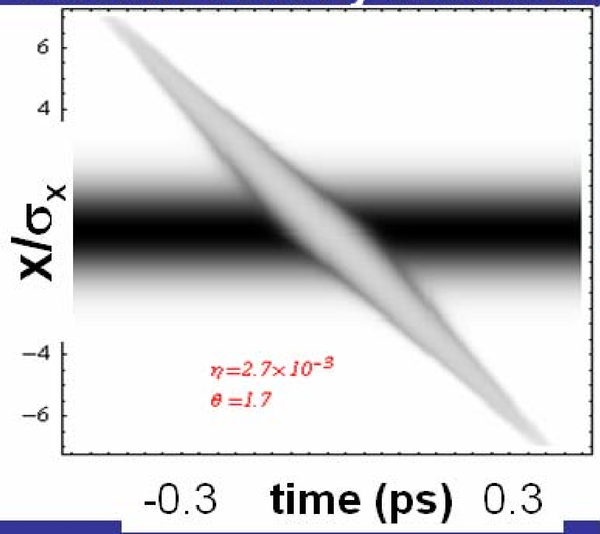



**Dip in the e**
**density dist**
**expands an**
**out as the e**
**bunch trave**
**the ring**

1) R.W.Schoenlein *et al* , *Appl. Physics*, **B71**, 1 (2000).
2) J. Byrd *et al* , *Phys. Rev. Lett.*, **96**, 164801(2006).
3) K. Holldack *et al* , *Phys. Rev. Lett.*, **96,** 054801 (2006).
4) J. Byrd *et al* , *Phys. Rev. Lett.*, **97**, 074802 (2006).

# Subps, laser-induced coherent THz radiation[1]

**e-beam density distribution**

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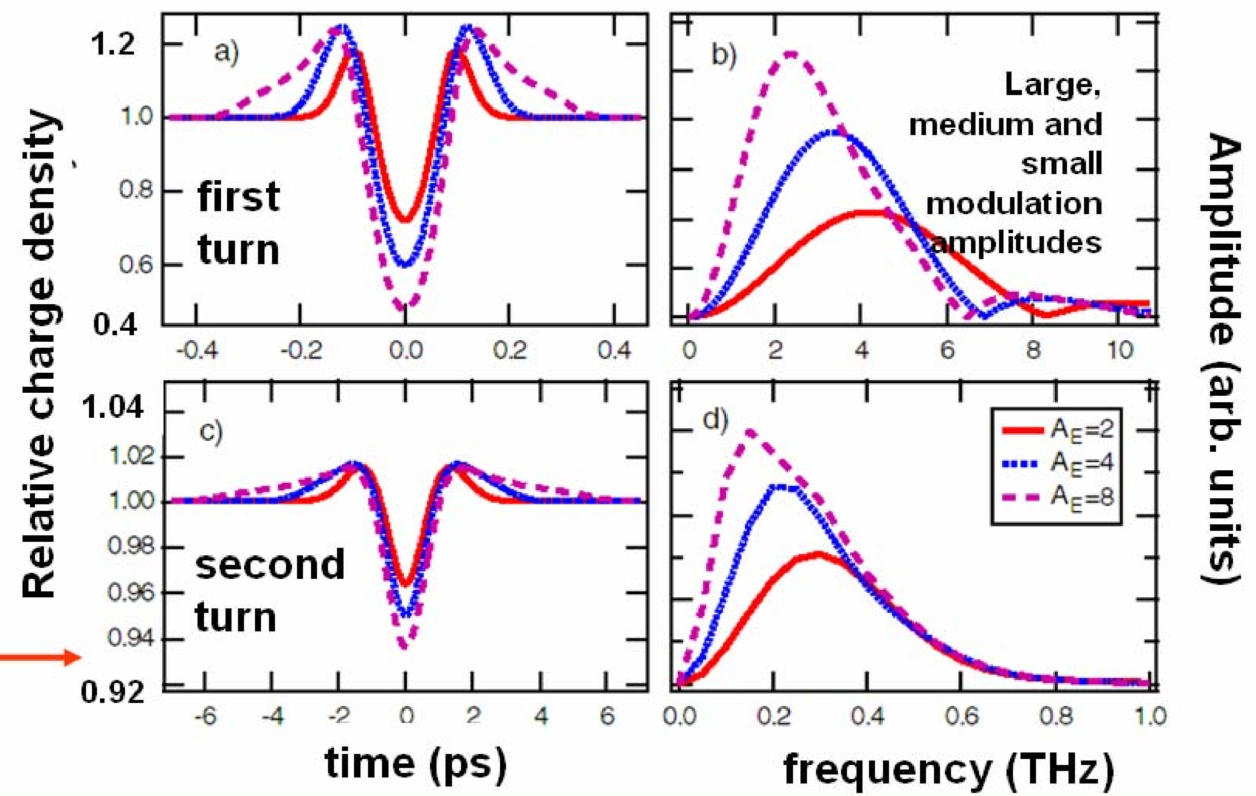
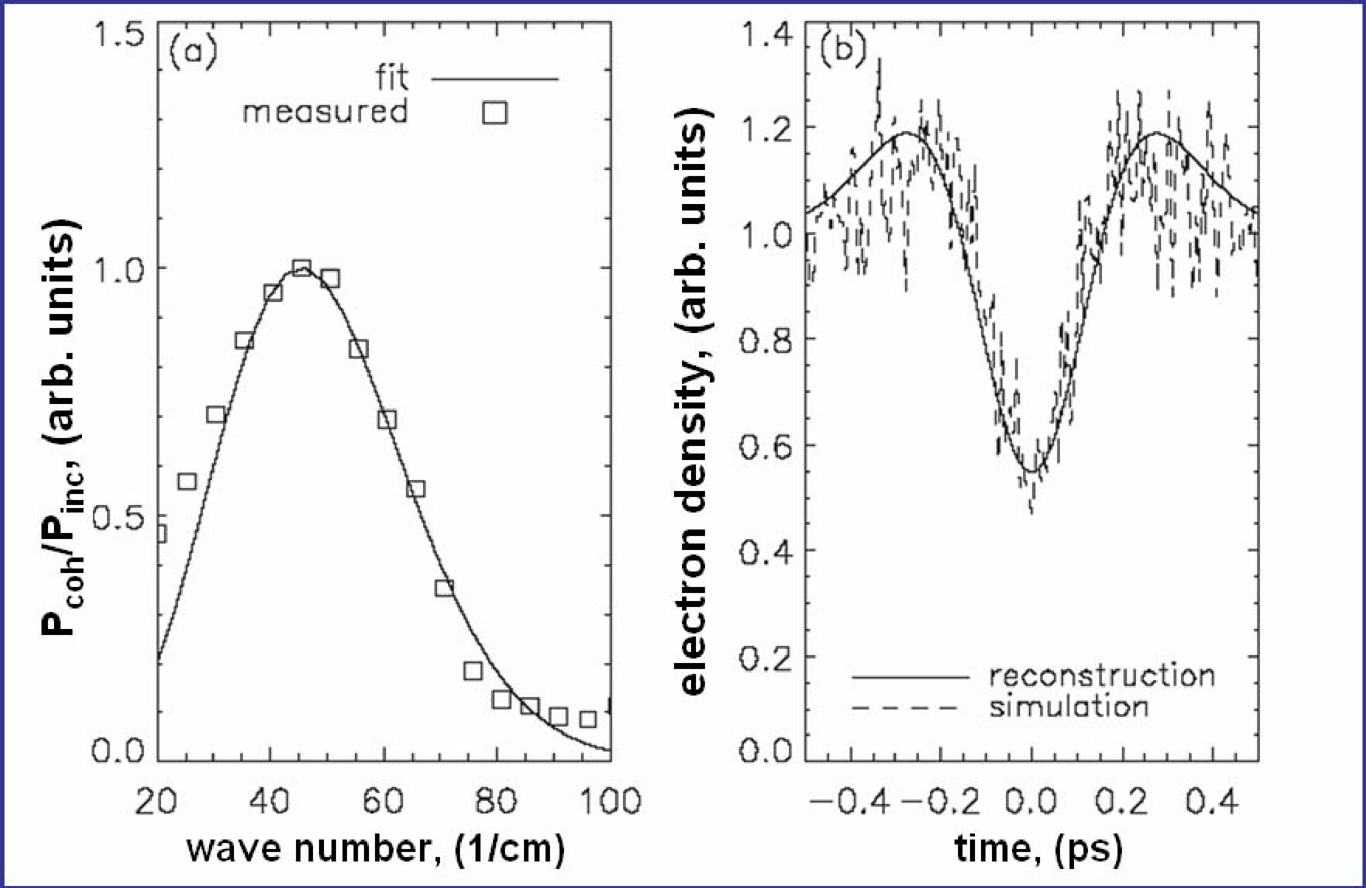



**Dip in the electron density distribution expands and dries out as the electron bunch travels along the ring**


1) R.W.Schoenlein *et al* , *Appl. Physics*, **B71**, 1 (2000).
2) J. Byrd *et al* , *Phys. Rev. Lett.*, **96**, 164801(2006).
3) K. Holldack *et al* , *Phys. Rev. Lett.*, **96,** 054801 (2006).
4) J. Byrd *et al* , *Phys. Rev. Lett.*, **97**, 074802 (2006).

# Dip reconstruction from THz spectra[1]




**THz signal is now routinely used for tuning of laser e-beam interaction and to maintain it with a feedback on mirrors – BESSY, ALS, SLS**


1) K. Holldack *et al* , *Phys. Rev. Lett.*, **96,** 054801 (2006).

# Swiss Light Source [1]

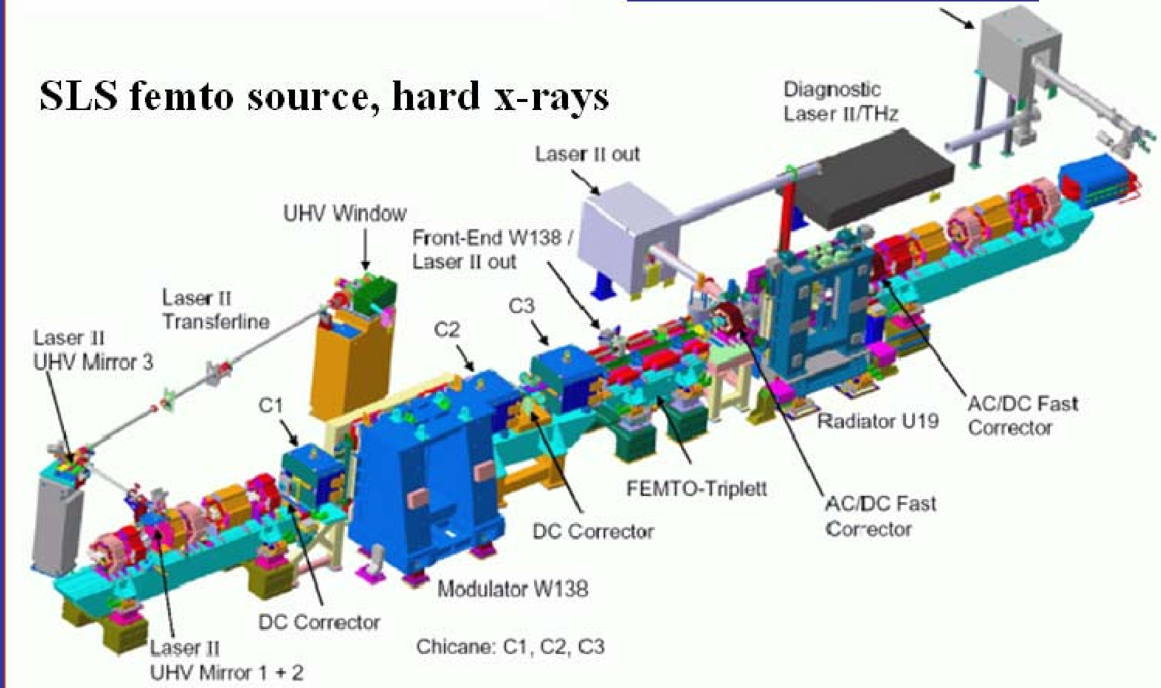
Rafael Abela

Paul Beud

Gerhard Ingold

Steven Johnson

Volker Shlott




Andreas Streun

1) A. Streun *et al* , *Proc. EPAC06,* THPLS062, Edinburg, (2006).

# Toward sub-fs x-ray pulses[1]

We propose to use electron interaction with the laser pulse in $TEM_{01}$ mode (polarized in the vertical plane):

$$E_y = E_0 \frac{y}{w_0} e^{-\frac{\tau^2}{4\sigma_\tau^2}} e^{-\frac{r^2}{w_0^2}} \sin(k(z-ct)+\psi)$$

to get an energy gain (variable with *y* , *i.e.* $\Delta E(y)$ )

and, according to Panofsky-Wentzel theorem :

$$\frac{\partial \Delta y'}{\partial s} = \frac{\partial}{\partial y}\left(\frac{\Delta E(y)}{E}\right)$$

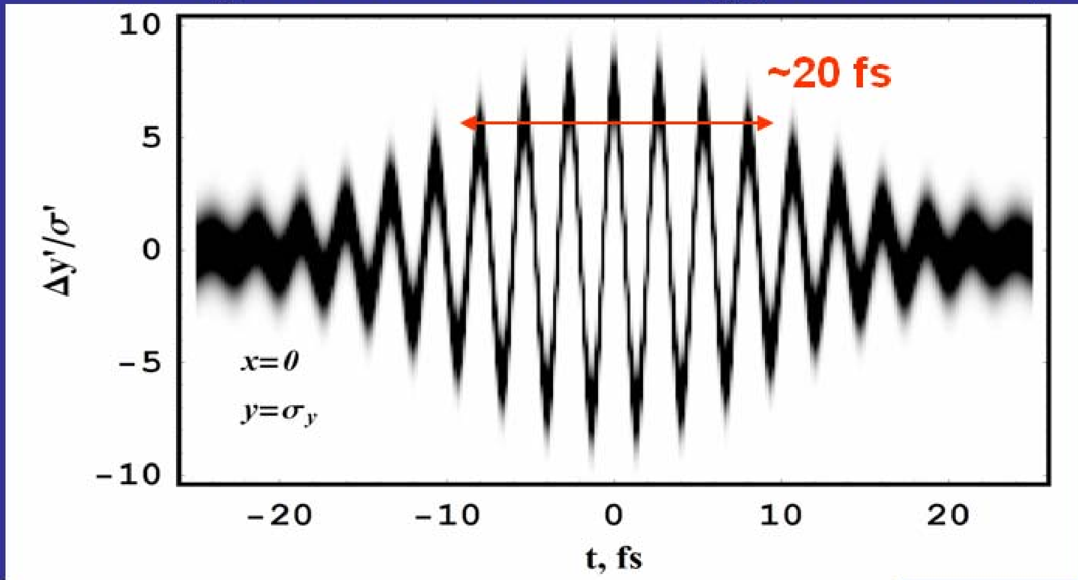
a vertical kick $\Delta y'$

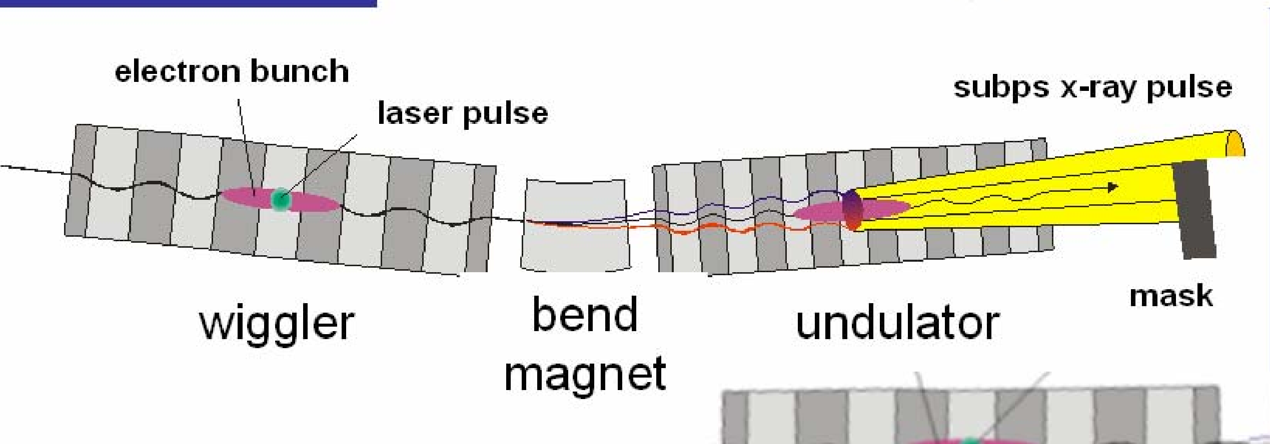

1) A. Zholents, M. Zolotorev, CBP Tech Note-372, (2007), submit. to New Journal of Physics.

2) A. Zholents, M. Zolotorev, LBNL-62750, (2007)

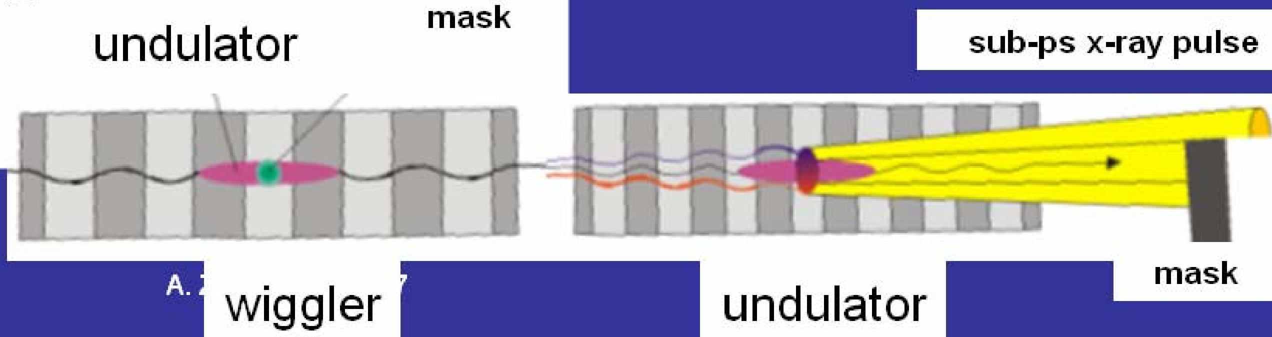
# Toward sub-fs x-ray pulses (2)

Example of angular modulation produced by 5 mJ, 15 fs laser pulse in $TEM_{01}$ mode interacting with electrons in a wiggler with five periods




Difficult because electrons emit into a relatively large angle



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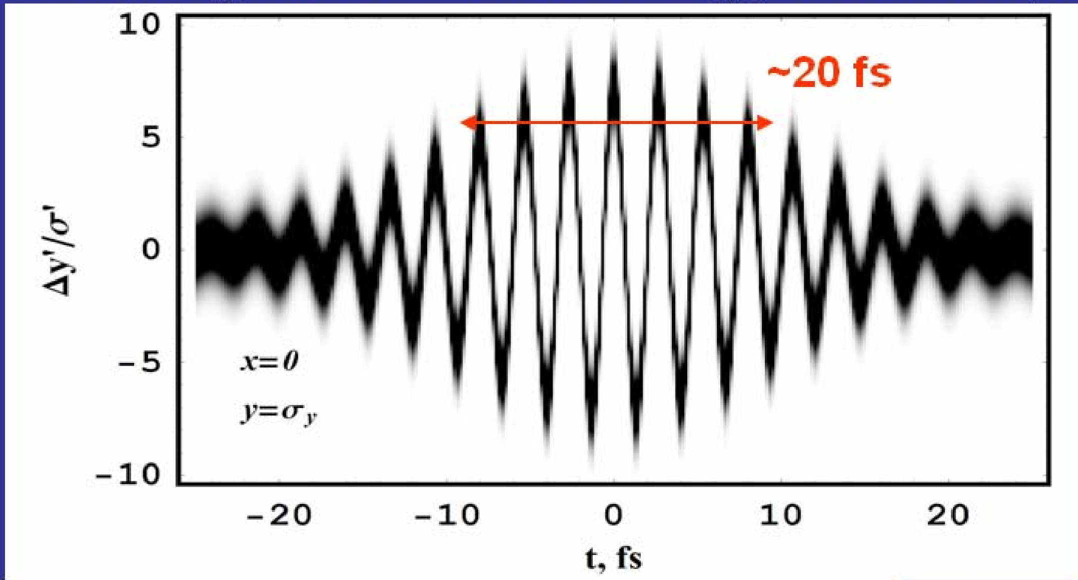
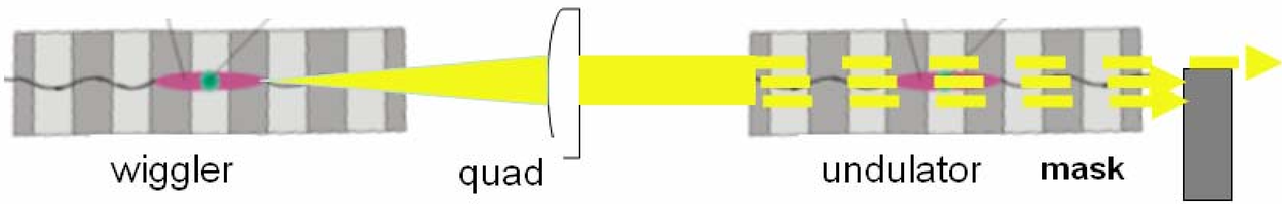
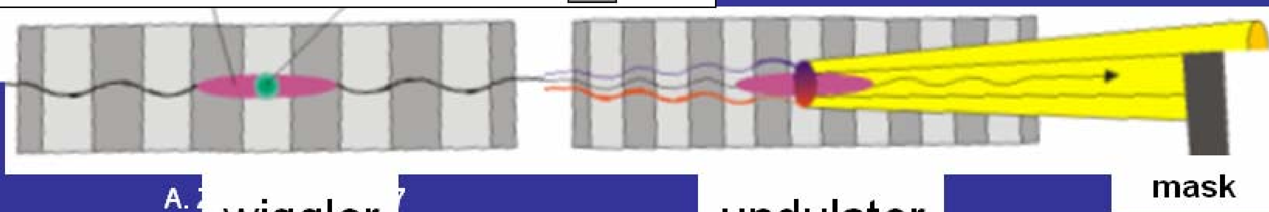
Example of angular modulation produced by 5 mJ, 15 fs laser pulse in $TEM_{01}$ mode interacting with electrons in a wiggler with five periods




**Select x-rays using coordinate separation at undulator location with $90^0$ betatron phase advance from wiggler**

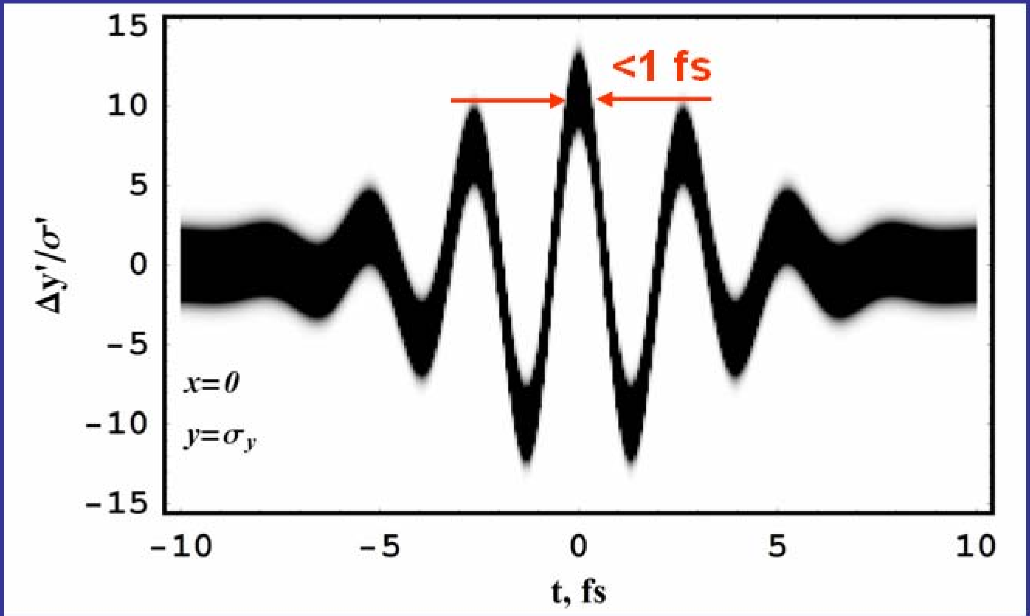



Difficult because electrons emit into a relatively large angle



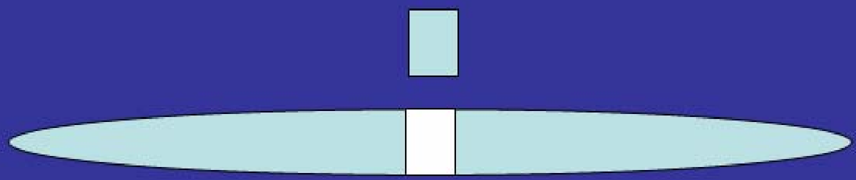
# Toward sub-fs x-ray pulses (3)

Example of angular modulation produced by 5 mJ, 5 fs laser pulse in $TEM_{01}$ mode with carrier-envelop phase stabilization interacting with electrons in a wiggler with one periods



# RF orbit deflection technique

# RF orbit deflection technique



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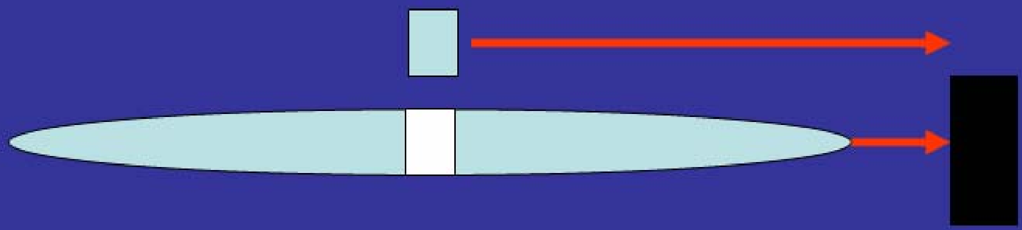

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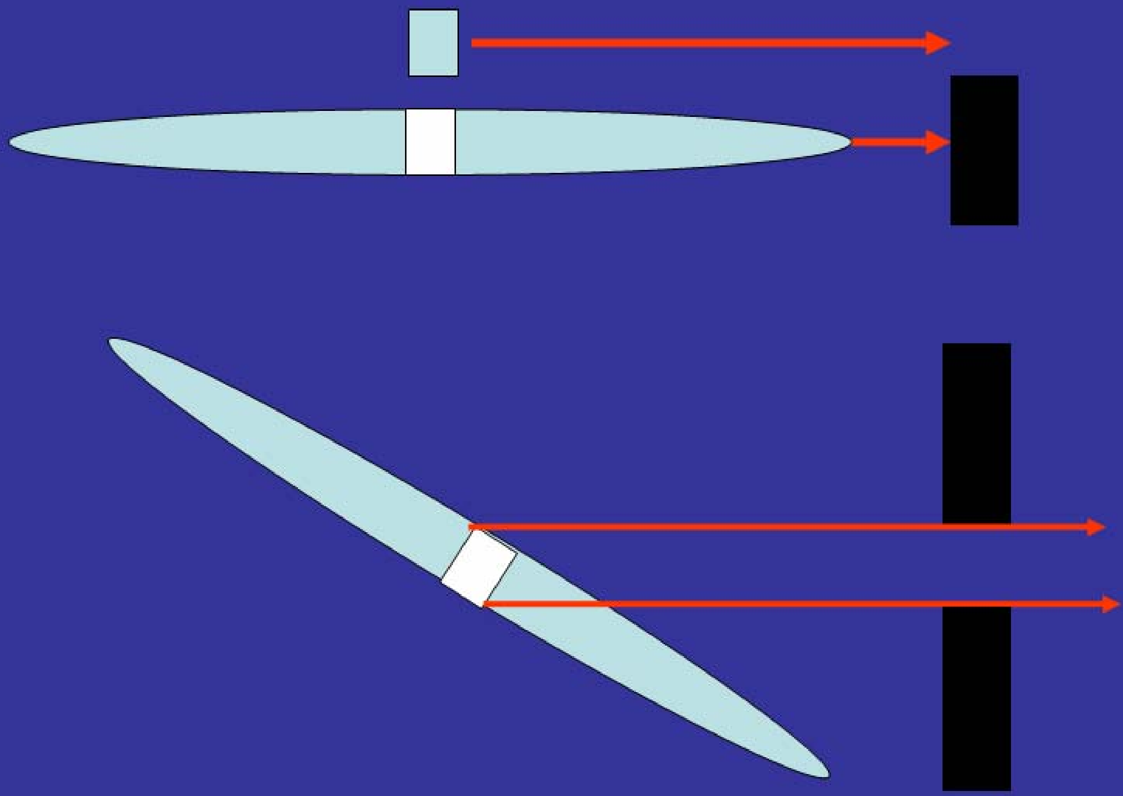
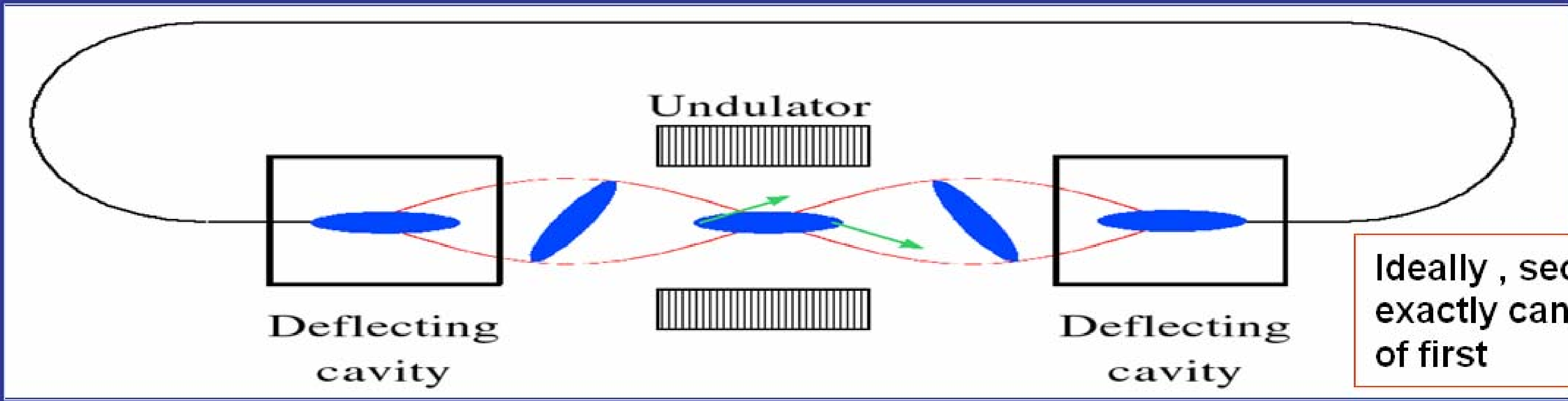


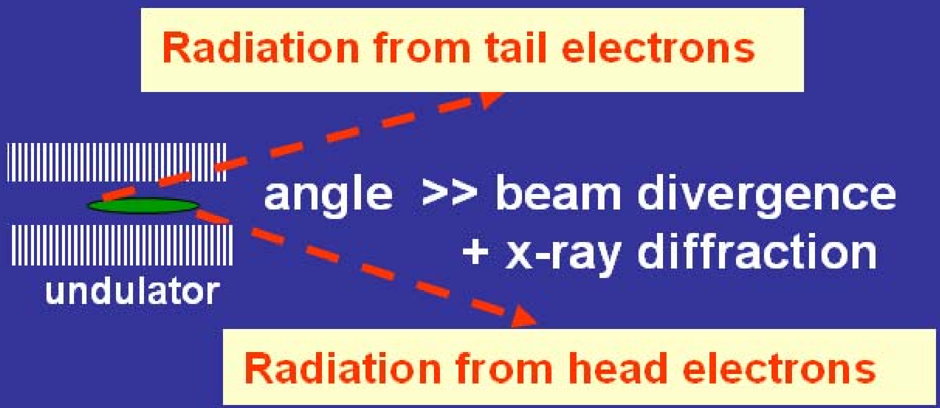
# High repetition rate source of sub-ps x-ray pulses[1]

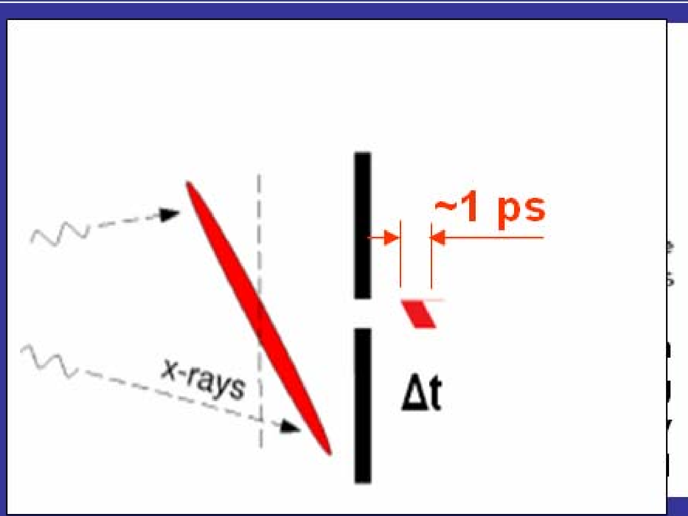
## Utilizing Compression of sub-ps x-ray pulses




Ideally , second cavity exactly cancels effect of first

Deflecting cavity delivers a time-dependent vertical kick to the beam




1) A. Zholents, P. Heimann, M. Zolotorev, J. Byrd, *NIM A*, **425,** 385 (1999).
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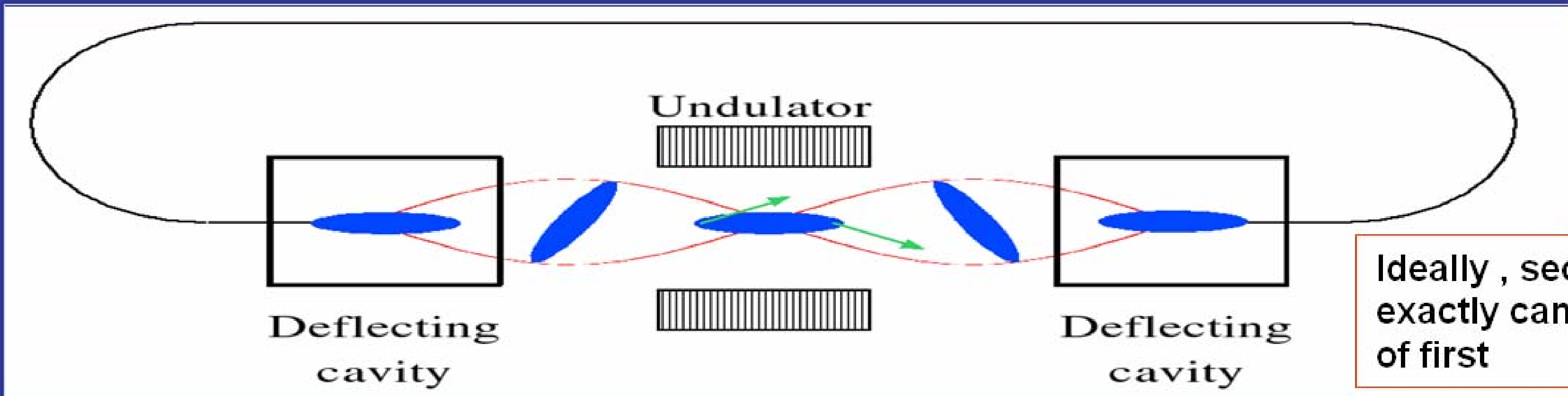


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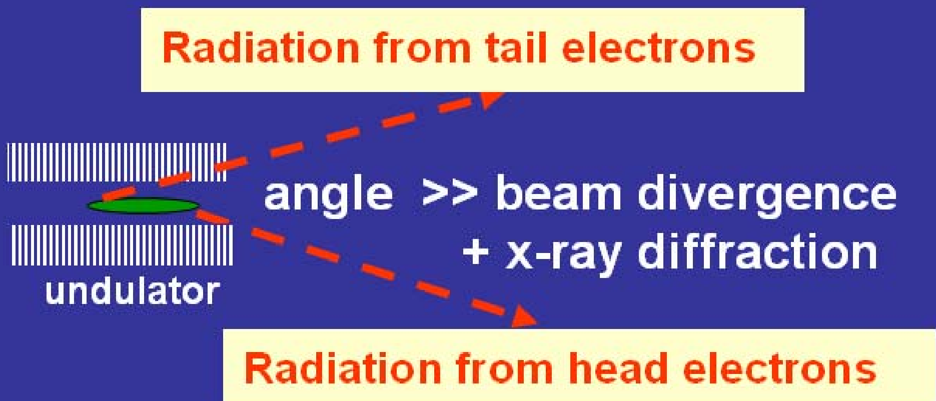


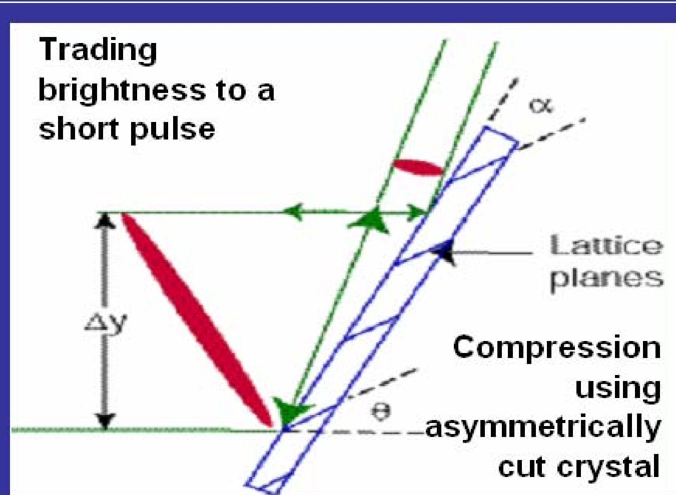


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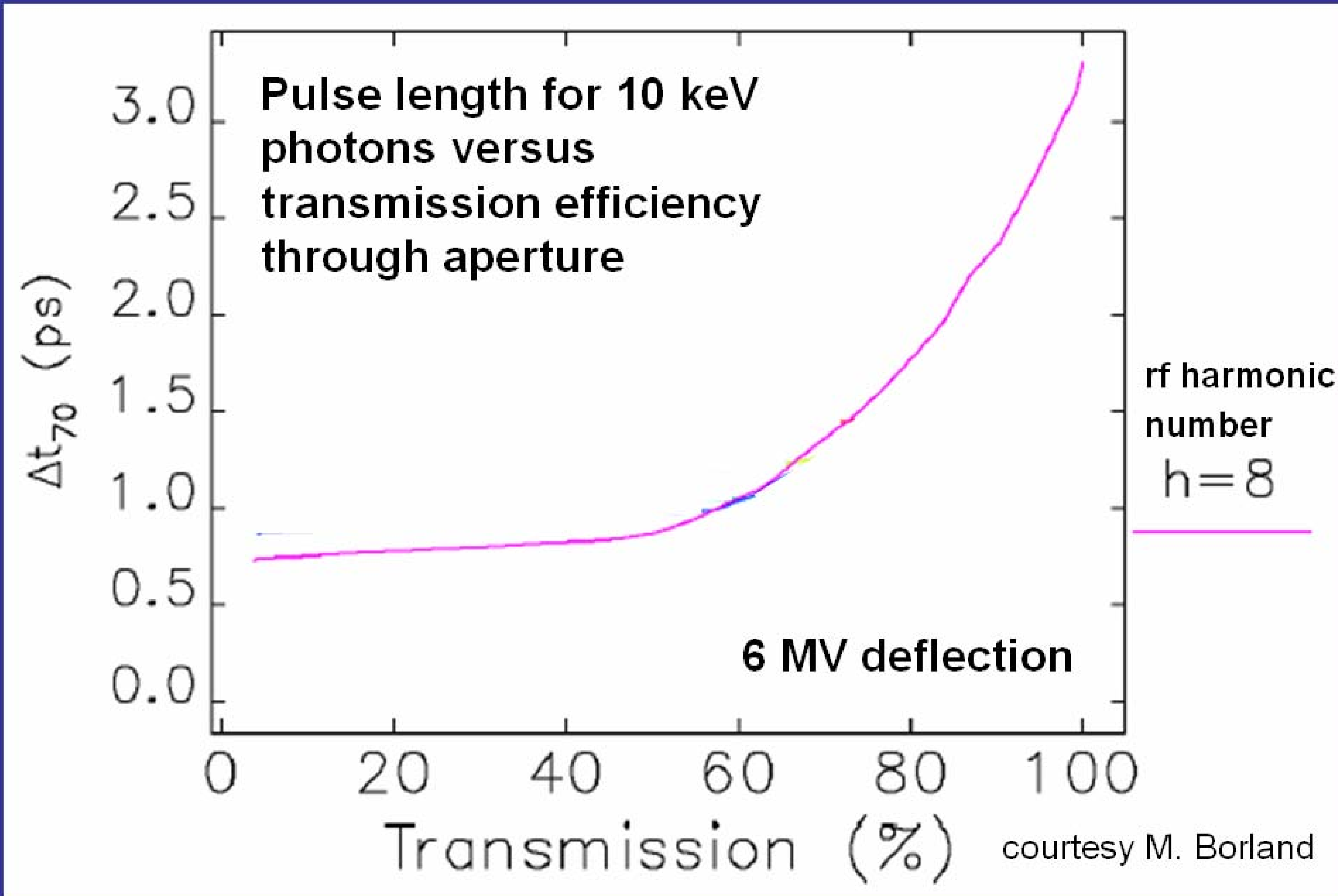
# Application to the Advanced Photon Source[1]




1) M. Borland, TUPMN091, this conference
2) B. Yang, TUPMN104 , this conference

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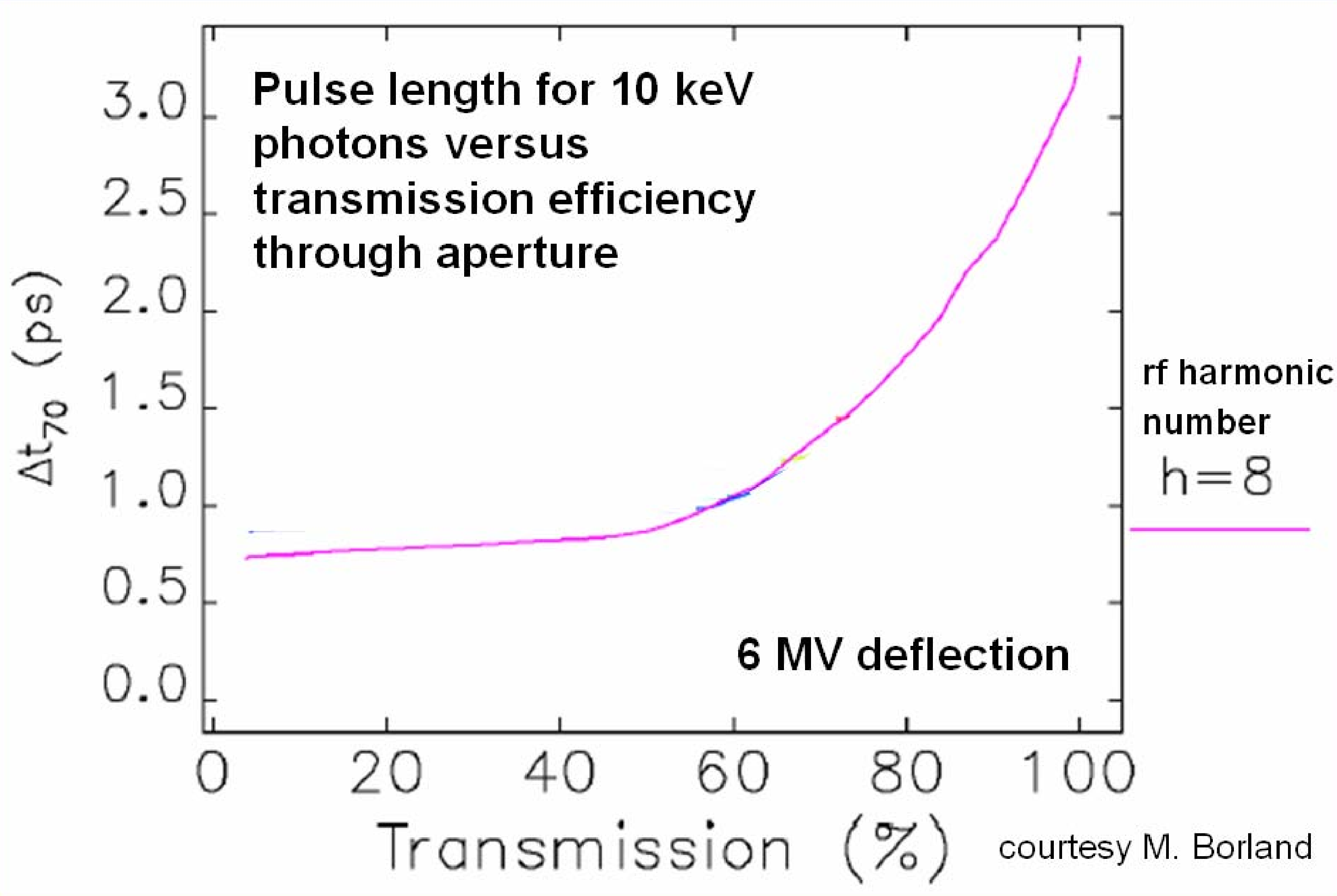


**APS**
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Pulsed rf system with 1 kHz rep. rate is planed for installation at APS in the fall 2007

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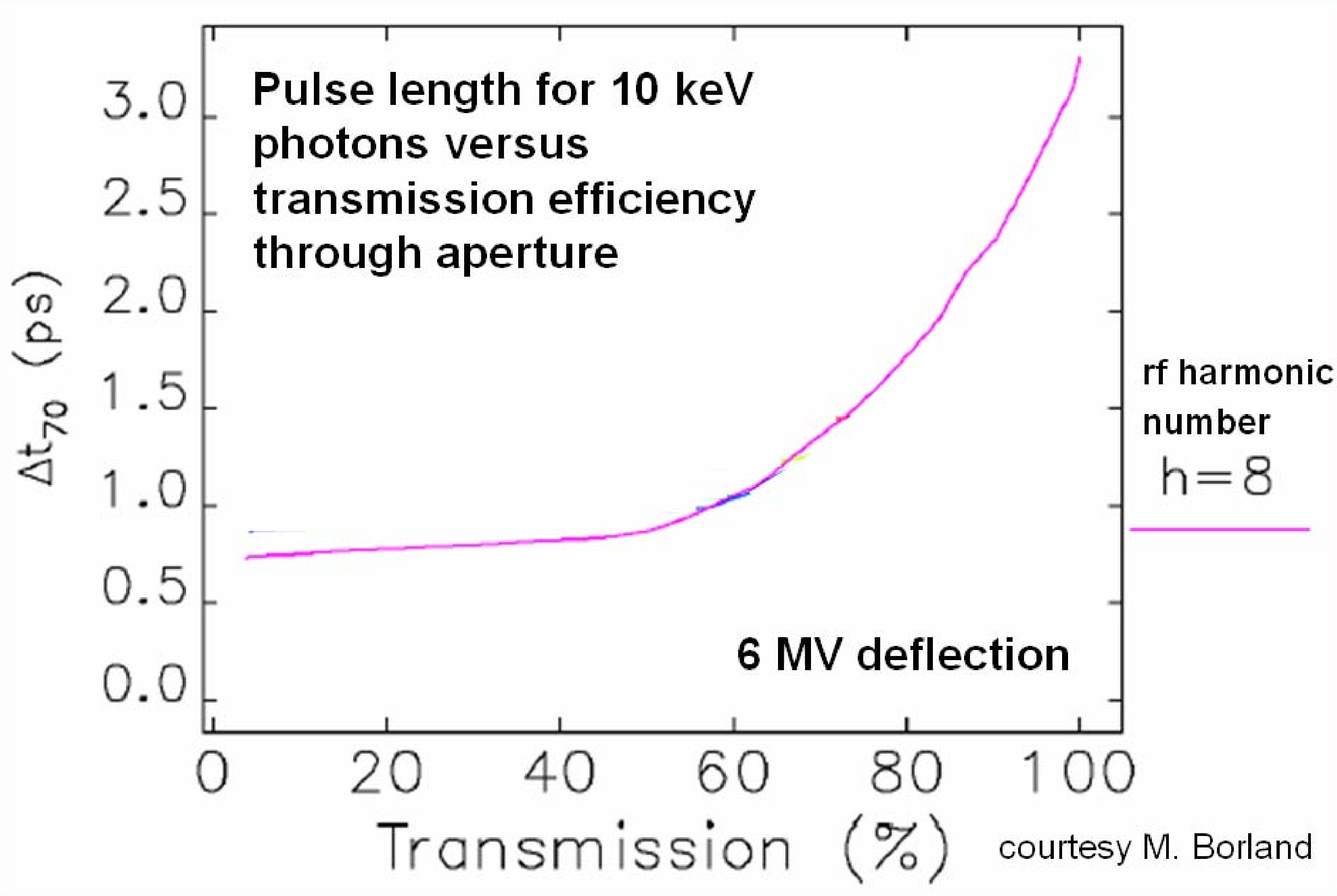


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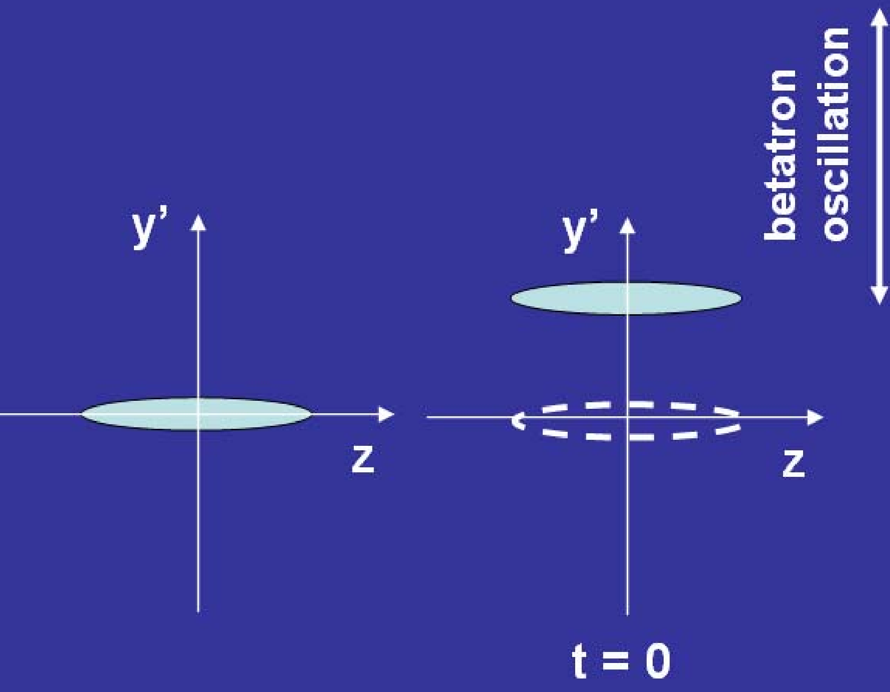
Y-C Chae et al., FRPMN105; L. Emery et al., FRPMN108

# Generation of sub-ps x-ray pulses using orbit kick[1]

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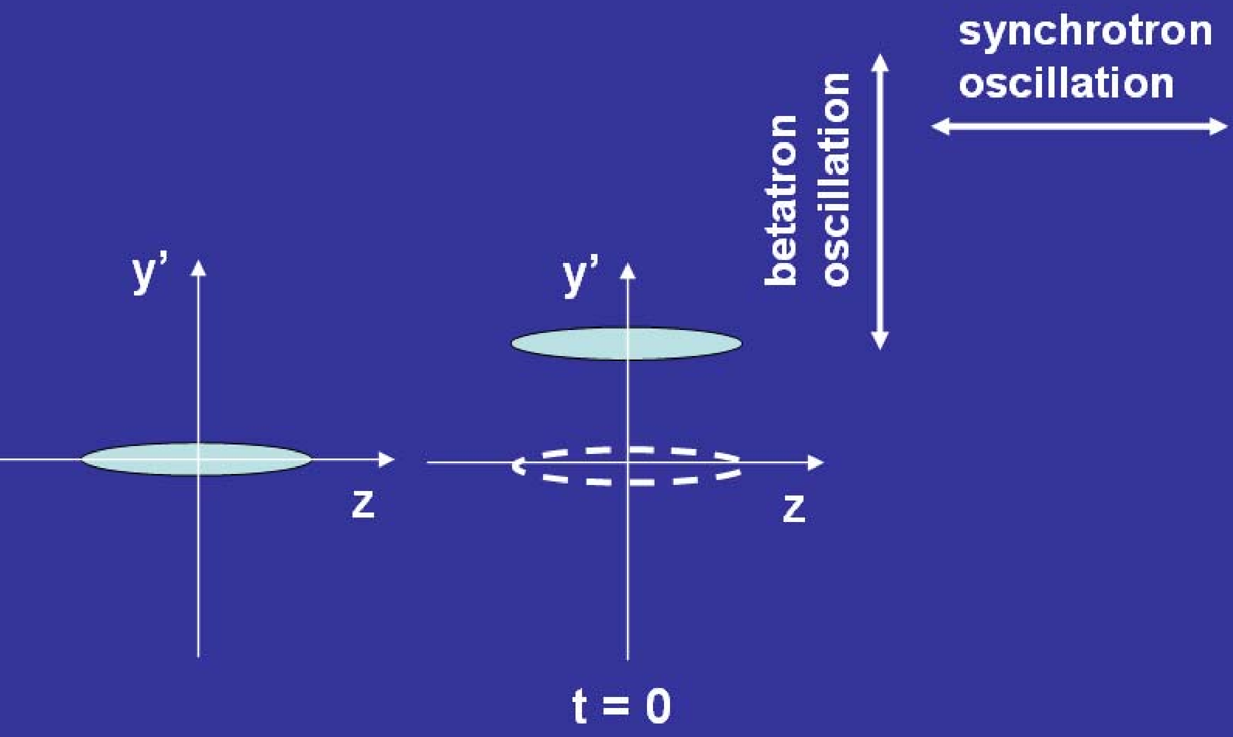


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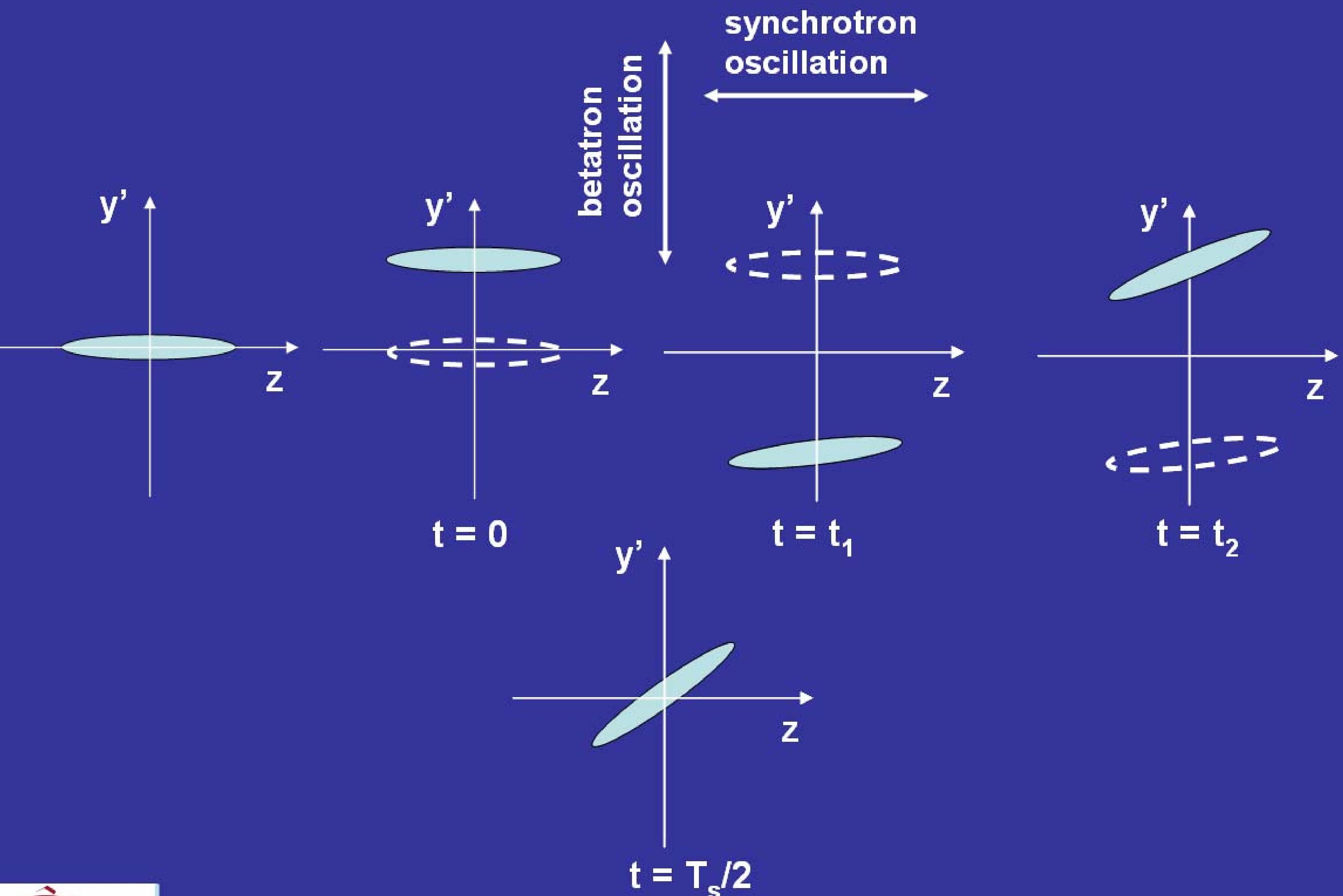


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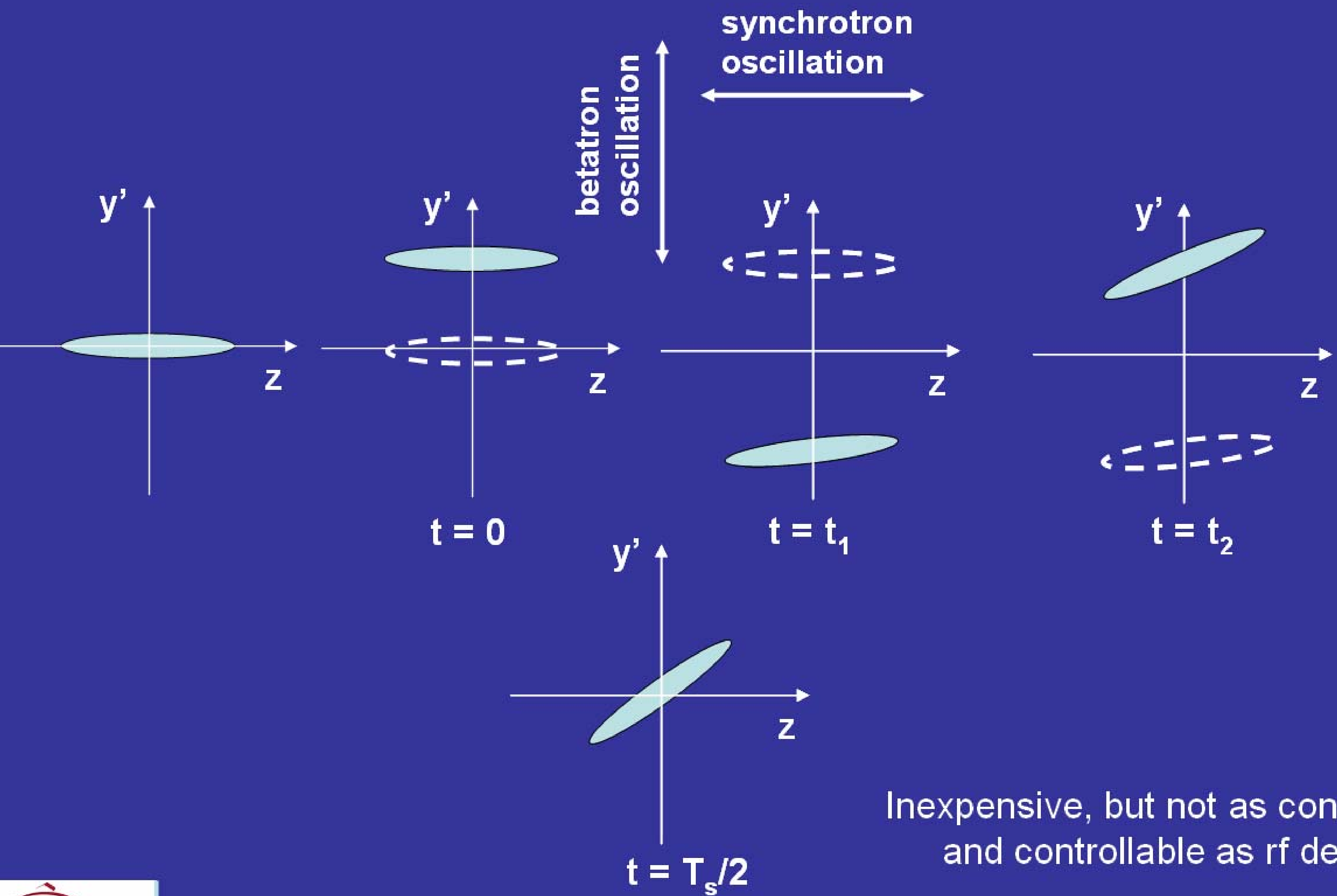


Inexpensive, but not as convenient and controllable as rf deflection

1) W. Guo et al., *Phys. Rev. ST –AB*, **10**, 020701 (2007).

# Summary

• The "slicing" technique for generation of sub-ps x-ray pulses in a storage ring had been successfully implemented at ALS, BESSY and SLS.

• All three facilities produced experimental results:

1) A. Cavalleri, M. Rini, H.H. Chong, S. Foumaux, T.E. Glover, P.A. Heimann, J.C. Kieffer, R. W. Schoenlien, *Phys. Rev. Lett.*, **95**, 067405 (2005)
2) A. Cavalleri, S. Wall, C. Simpson, E. Statz, D.W. Ward, K.A. Nelson, M. Rini, R. W. Schoenlien, *Nature*, **442**, 664 (2006)
3) C. Stamm, T. Kachel, N. Pontius, R. Mitzner, T. Quast, K. Holldack, S. Khan, C. Lupulescu, E.F. Aziz, M. Wietstruk, H.A. Dürr, W. Eberhardt, submitted for a publication,
4) M. Chergui, R. Abela *et al.*, submitted for a publication,

• The technique of the rf orbit deflection has been intensively studied for APS and should soon be tested there using pulsed S-band cavities

• These facilities permit us to have a glimpse of a microworld with a sub-ps resolution that will eventually be scrutinized when x-ray free electron lasers will come to life.

# Outlook

Ferenc Krausz

In 2001 Ferenc Krausz and coworkers at *Technische Universität Wien, Steacie Institute of Molecular Science, Canada, Universität Bielefeld, Germany* once again redefined the term "ultrafast" when they reported the demonstration of subfemtosecond pulses using HHG and a few-cycle laser pulse with a carrier-envelope phase stabilization[1].

1) M. Hentschel, R. Kienberger, Ch. Spielmann, G. Reider, N. Milosevic, P. Corkum, U. Heinzmann, M. Drescher, F. Krausz, " Attosecond metrology", *Nature*, **441**, 509 (2001).

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This trend will be followed by generation of subfemtosecond x-ray pulses using new x-ray free electron lasers under construction.

1) M. Hentschel, R. Kienberger, Ch. Spielmann, G. Reider, N. Milosevic, P. Corkum, U. Heinzmann, M. Drescher, F. Krausz, " Attosecond metrology", *Nature*, **441**, 509 (2001).

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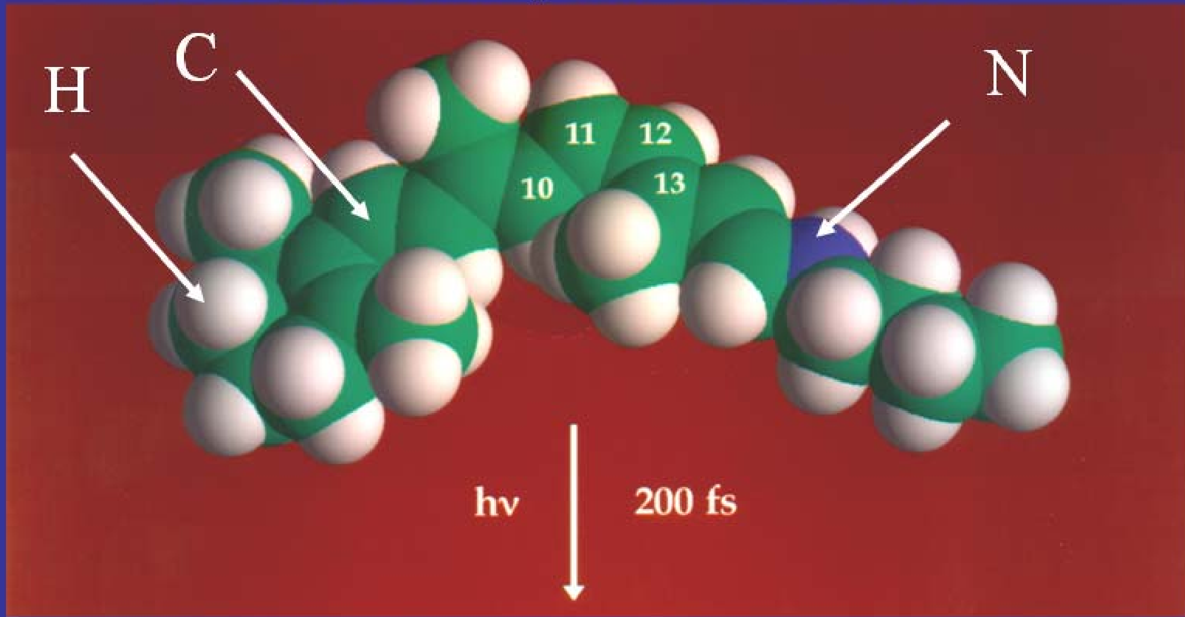
*Thank you for your attention*

*Back-up slides*

# Ultrafast processes in biology

## "The first step in vision….." [1]

Molecule of rhodopsin before …

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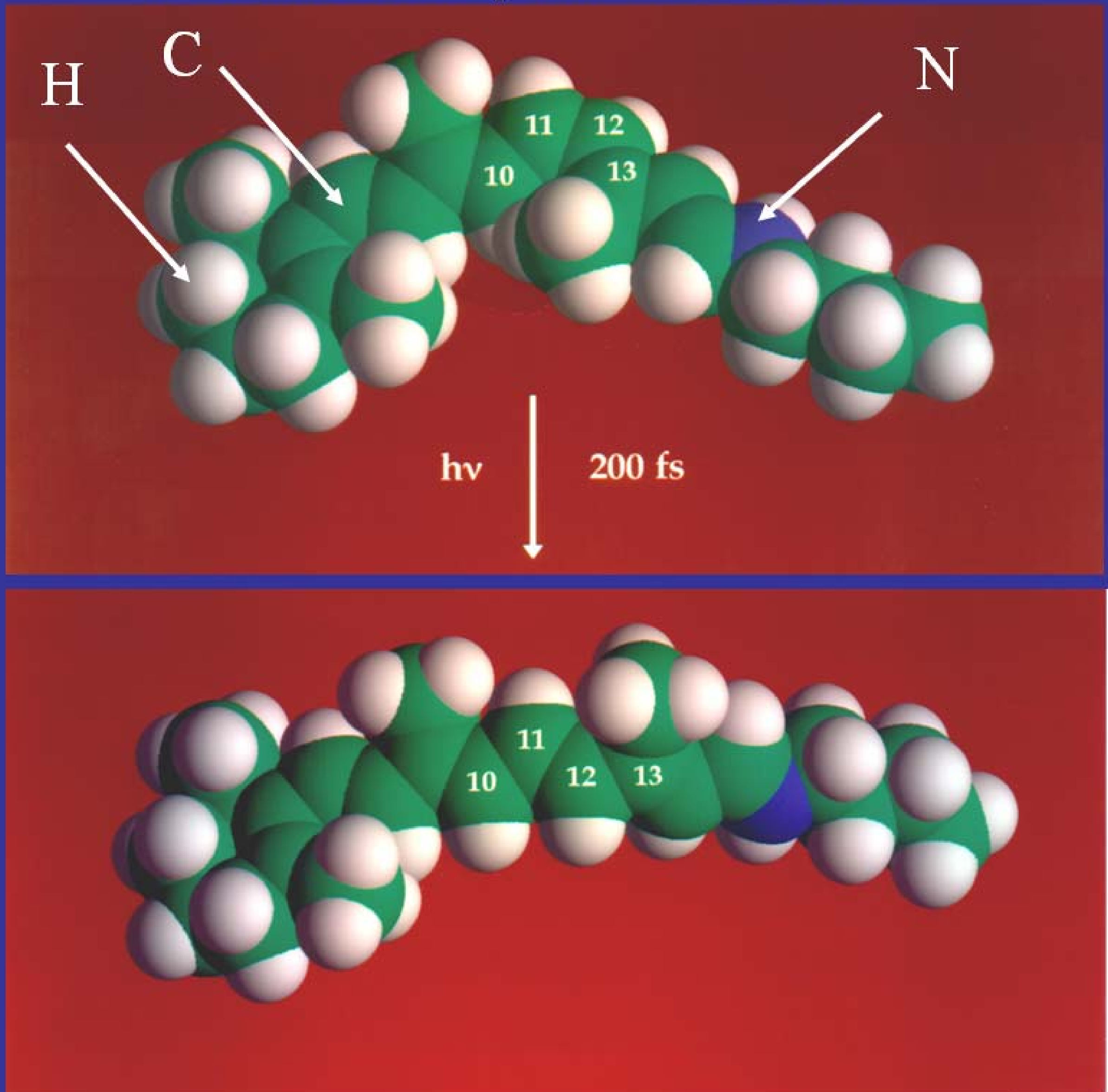
… and after absorption of a photon




1) R.W. Schoenlein, L.A. Peteanu, R.A. Mathies, C.V. Shank, Science, **254**, 412 (1991).

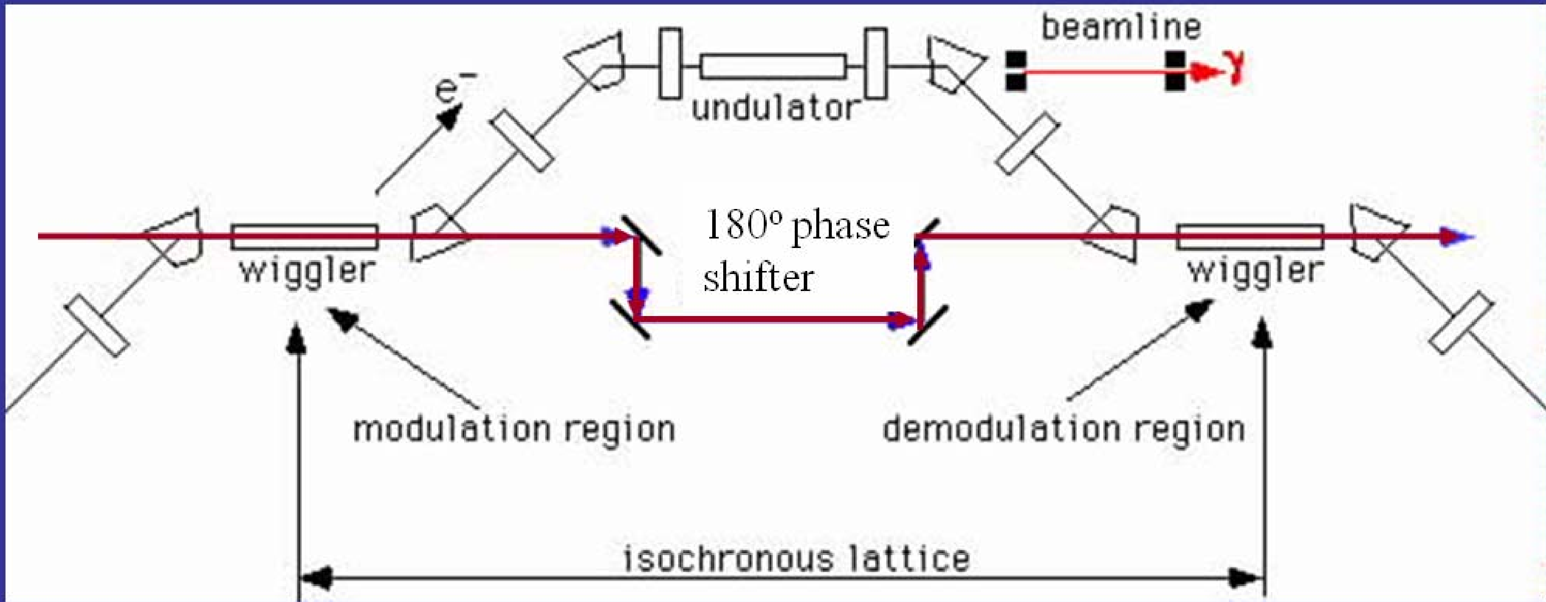
# What we may learn with sub-ps x-ray pulses?

- Phase transitions in solids
- Surface dynamics
- Making and braking of bonds during chemical reactions
- Chemical dynamics in proteins

All these processes are associated with atomic motion and have a typical time scale:

$$\tau \sim 1\text{Å}/(\text{speed of sound}) \sim 100\ \text{fs}$$

# Two wiggler method



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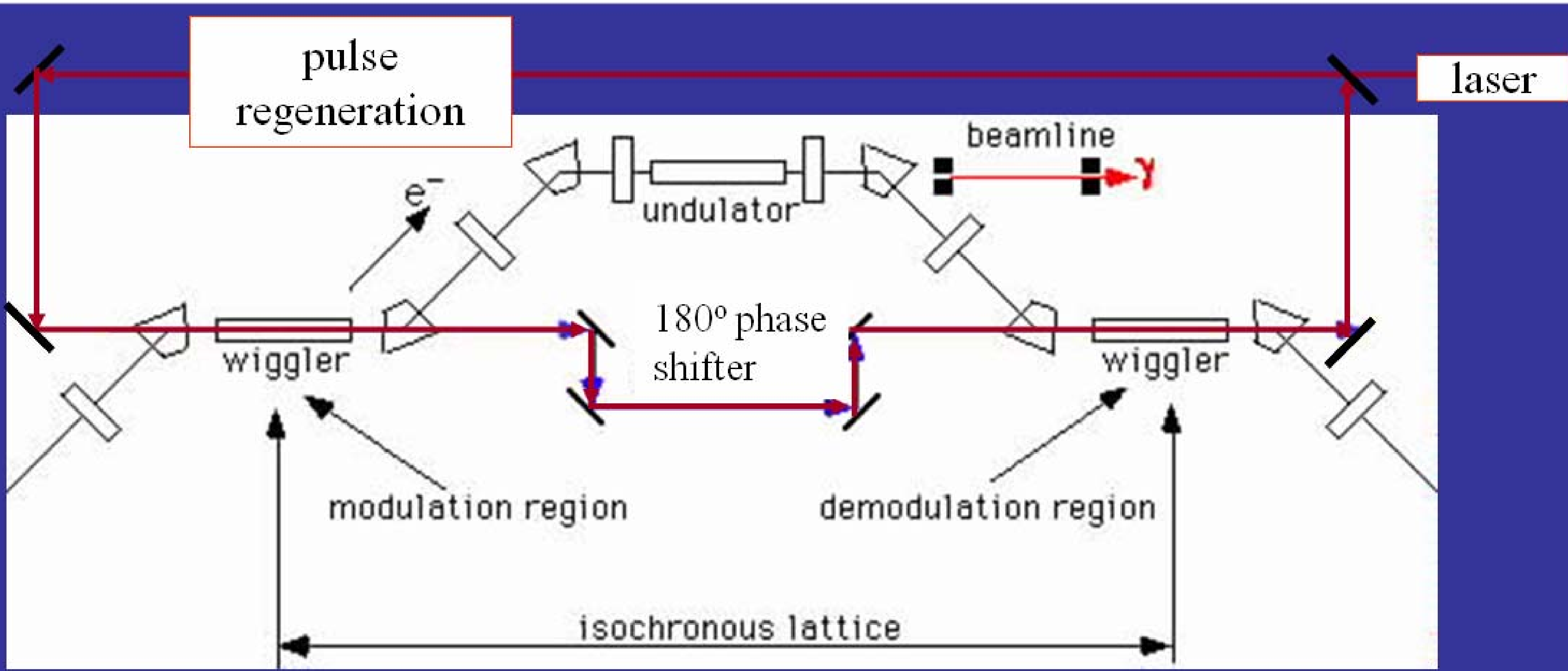

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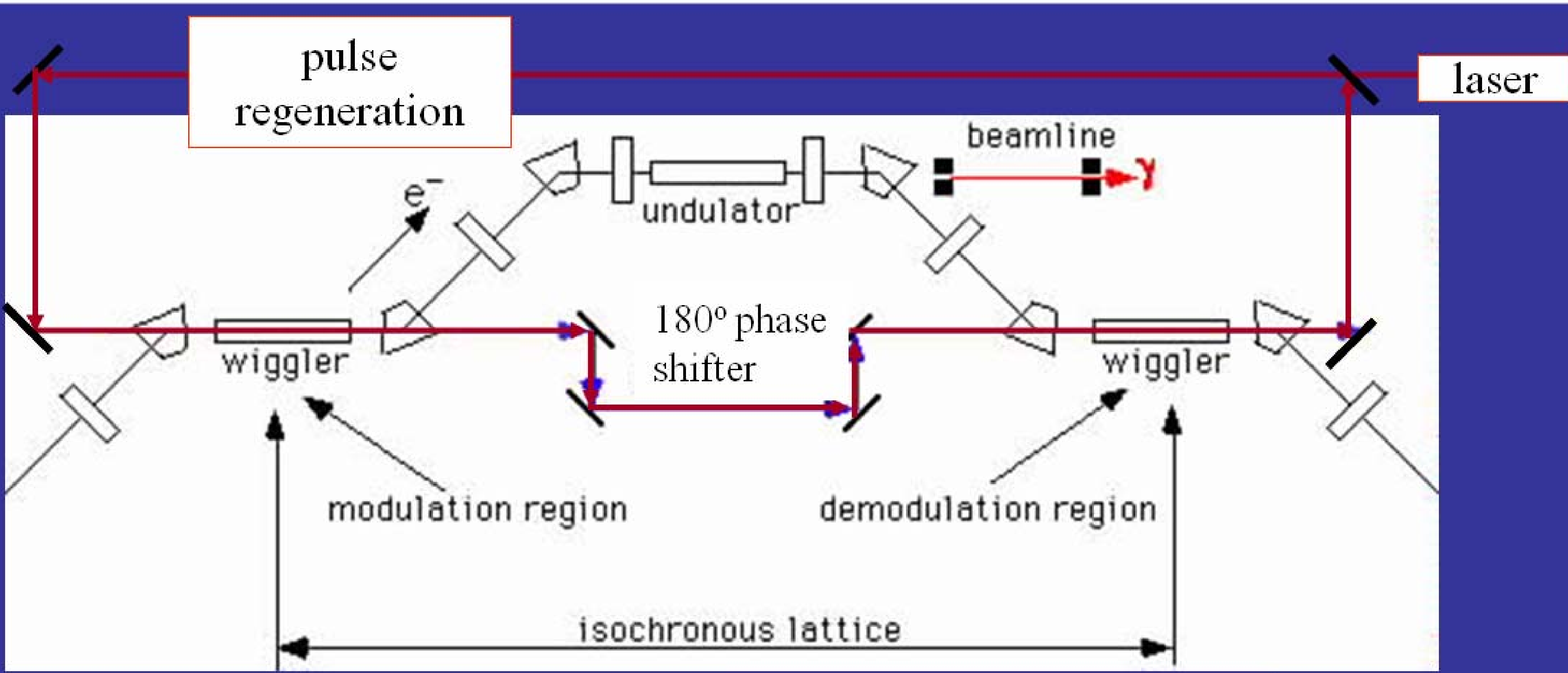
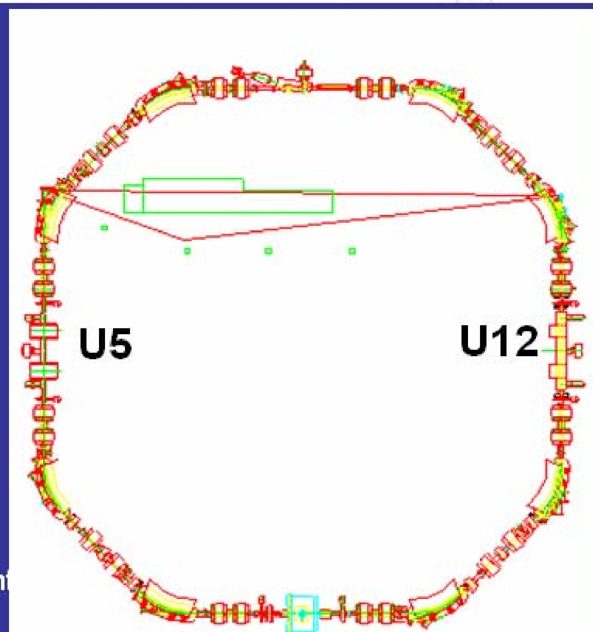



Can be tested on
VUV ring in BNL



# Completed experiments with sub-ps x-ray pulses

ALS

A. Cavalleri, M. Rini, H.H. Chong, S. Foumaux, T.E. Glover, P.A. Heimann, J.C. Kieffer, R. W. Schoenlien, "*Band-Selective measurements of electron dynamics in VO2 using femtosecond near-edge X-ray absorption*", *Phys. Rev. Lett.*, **95**, 067405 (2005)

A. Cavalleri, S. Wall, C. Simpson, E. Statz, D.W. Ward, K.A. Nelson, M. Rini, R. W. Schoenlien, "Tracking the motion of charges in a terahertz light field by femtosecond X-ray diffraction ", *Nature*, **442**, 664 (2006)

BESSY

C. Stamm, T. Kachel, N. Pontius, R. Mitzner, T. Quast, K. Holldack, S. Khan, C. Lupulescu, E.F. Aziz, M. Wietstruk, H.A. Dürr, W. Eberhardt, "*Femtosecond modification of electron localization and transfer of angular momentum in ferromagnetic Ni* ", submitted for a publication,

SLS

M. Chergui, R. Abela, submitted for a publication,