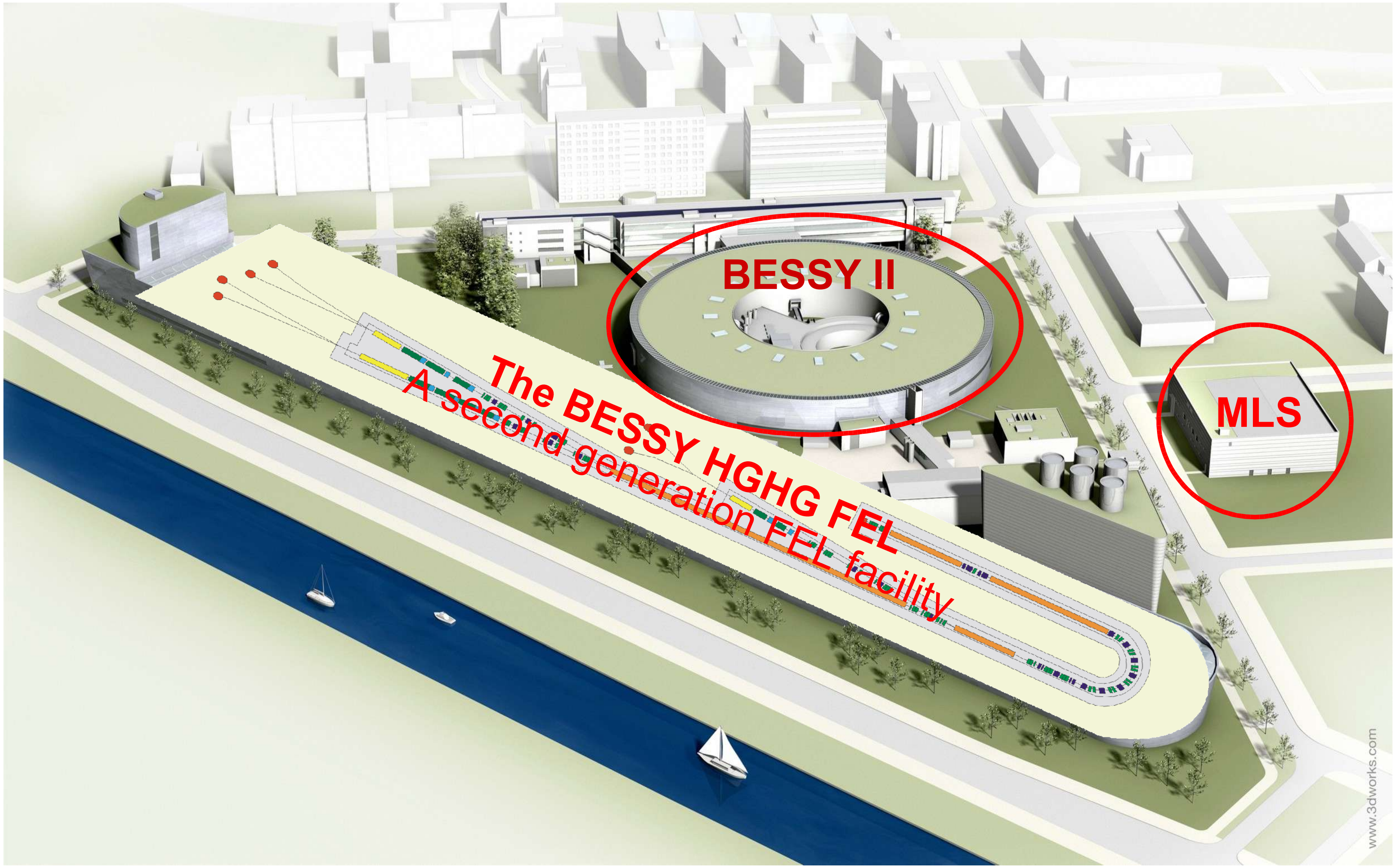
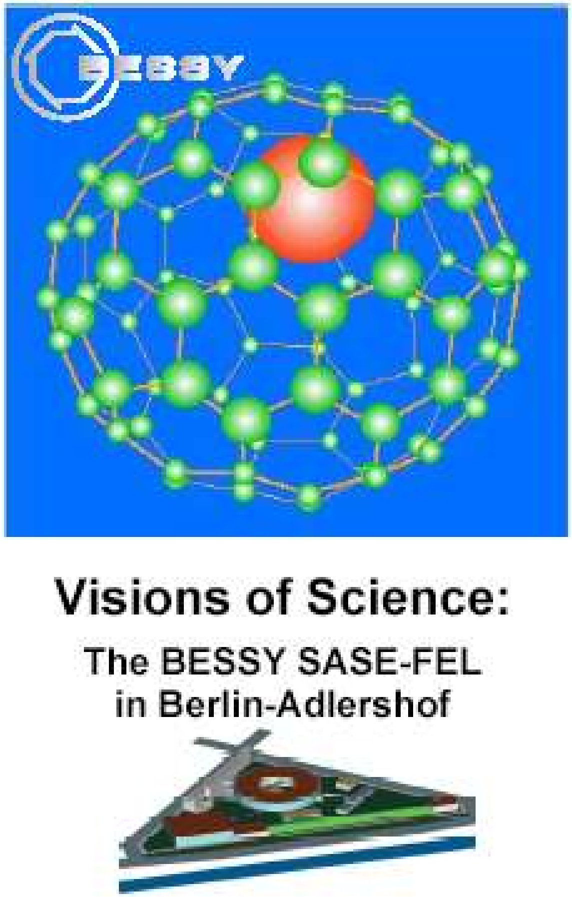
# Vision of the Future at BESSY




Berlin-Adlershof
City of
SCIENCE
TECHNOLOGY
and MEDIA
Humboldt University
BESSY
MBI

# Developing the SCIENTIFIC CASE

Two workshops with the prospective user community were organized in Blankensee (2000) and in Holzhau (2001)




**www.bessy.de/FEL**

# Welcome

BESSY FEL

## Workshop on Ultrafast Time-resolved Soft X-ray Science

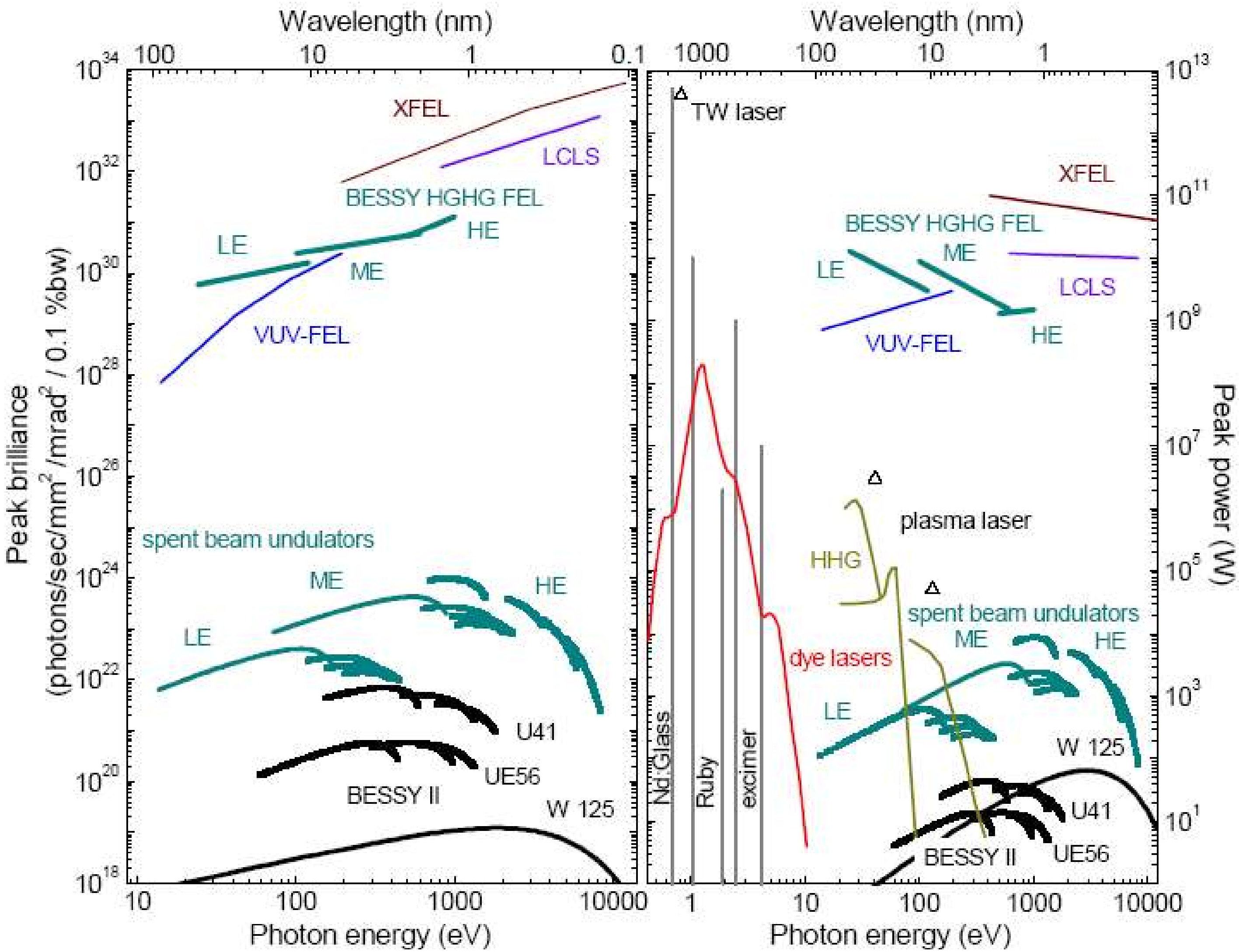
**Zeuthen, Germany, 27-29 April 2005**

SOLEIL SYNCHROTRON

BESSY

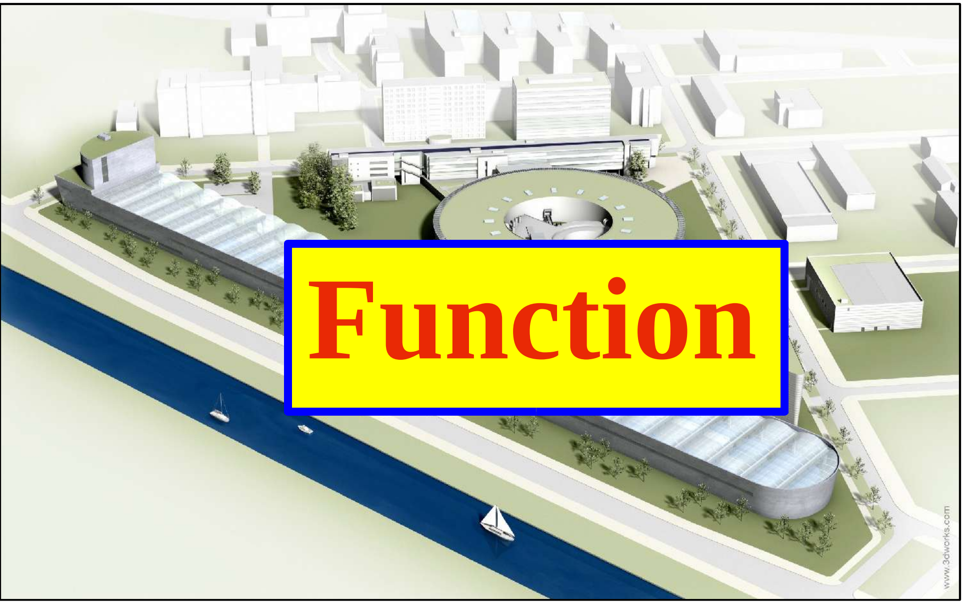
SWISS LIGHT SOURCE

# The BESSY Soft X-Ray FEL
## Peak Power and Brilliance

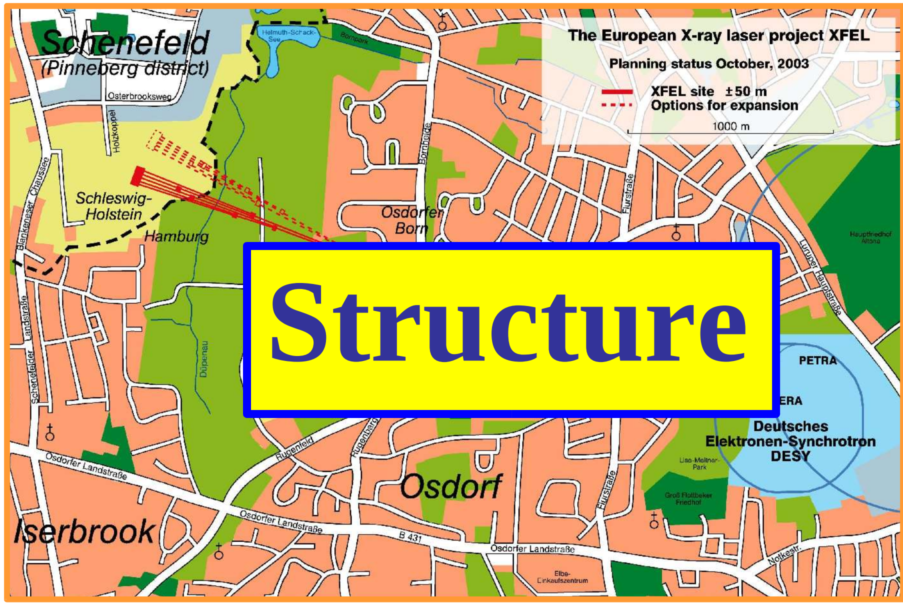



**Experiments can be more readily extrapolated by the LASER community than by the synchrotron radiation community**

## VUV and SOFT X-RAYS
## BESSY FEL




## X-RAYS
## TESLA X-FEL



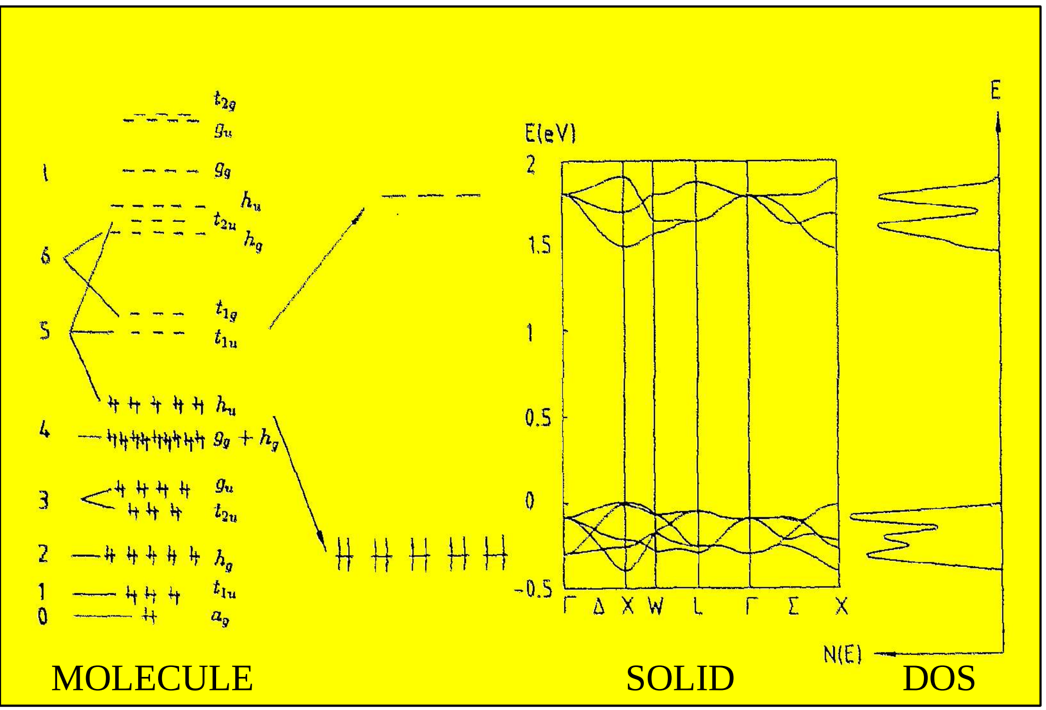

| 20 eV to 1 keV | **PHOTON ENERGY** | 500 eV to 15 keV |
|---|---|---|
| <20 fs controlled | **PULSE LENGTH** | 100 fs |
| 1 kHz (1-25 pulses) | **REPETITION RATE** | 10 Hz (7200 pulses) |
| HGHG-FEL | **SYNCHRONIZATION** | SASE-FEL |

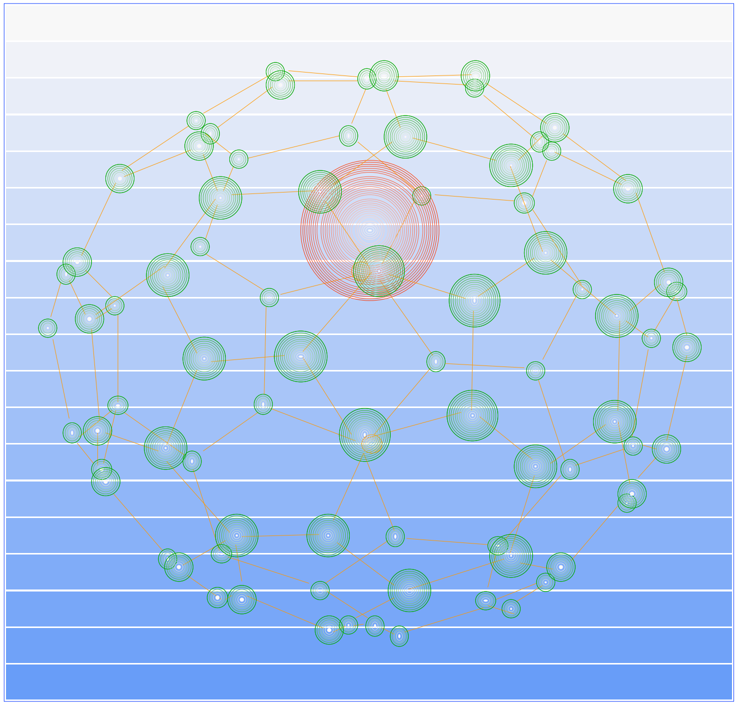
# COMPLEMENTARITY of the BESSY FEL and the TESLA X-FEL

BESSY FEL

## Electronic Structure or ‘Function’ and Structure

**Are needed to develop an understanding of all essential materials properties**

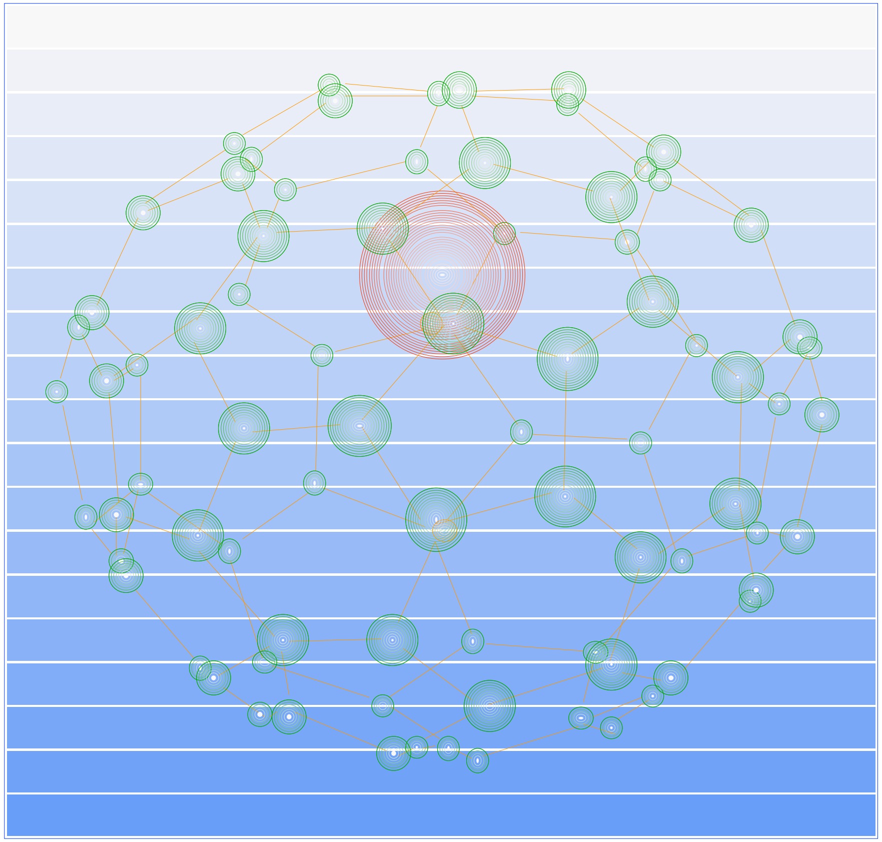


VUV and SOFT X-RAYS
BESSY FEL

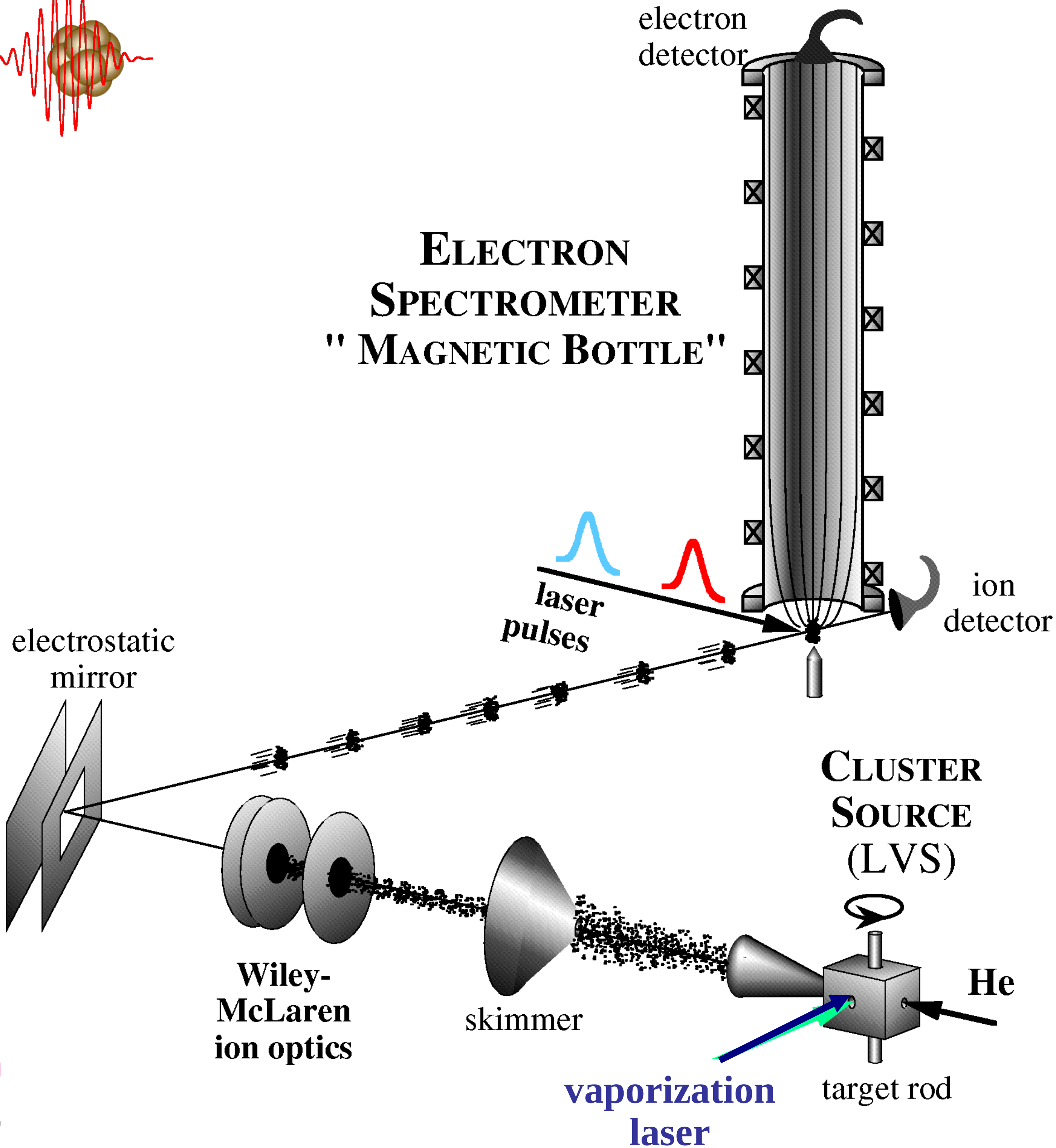
X-RAYS
TESLA X-FEL

# Clusters as New Materials



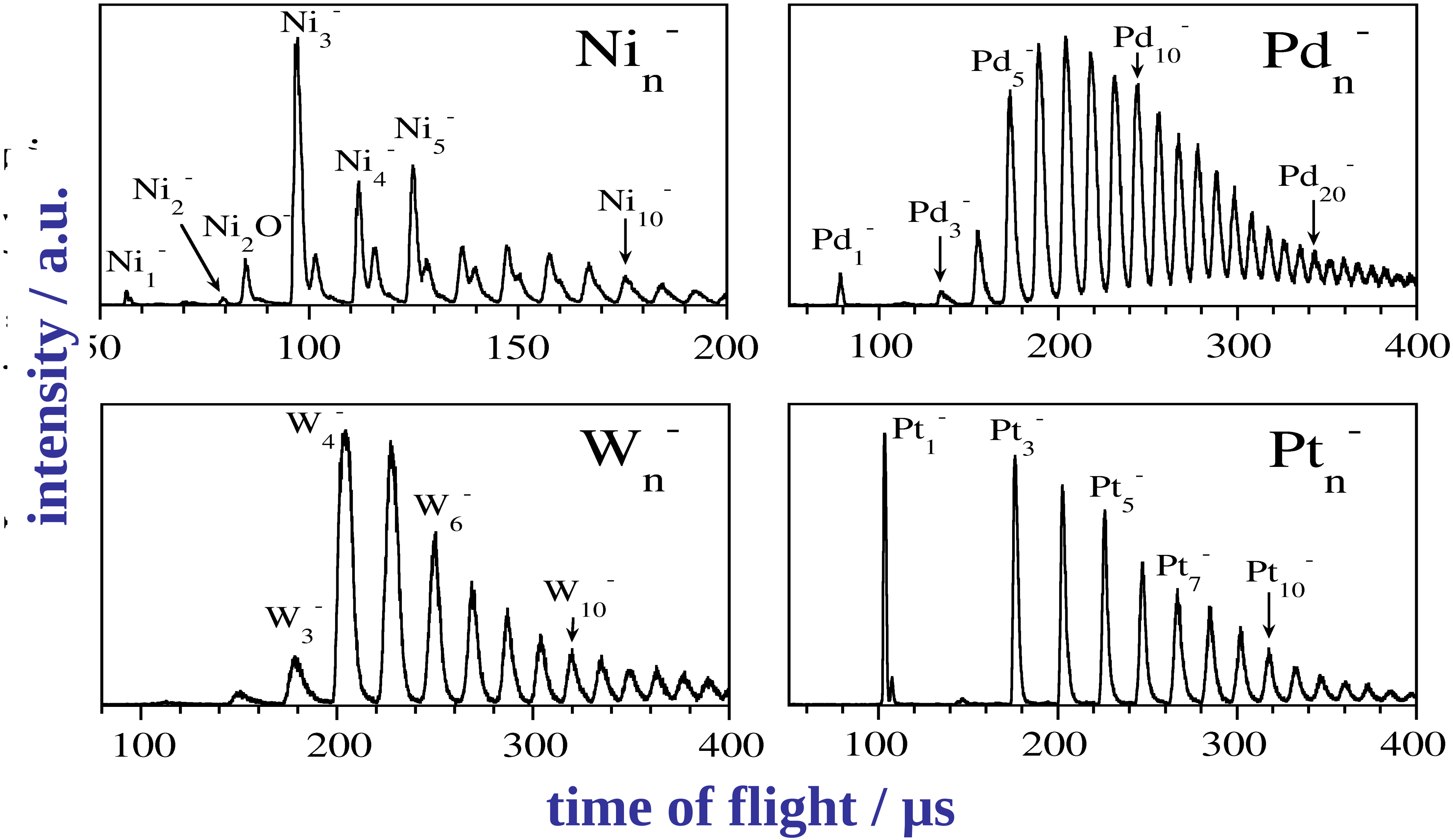
**Exploring the materials properties and dynamics of size-selected clusters**

# Photoelectron spectroscopy of mass selected clusters



# Time-of-flight mass spectra of cluster anions

BESSY FEL



# Visions of SCIENCE

BESSY FEL

## Photoelectron spectroscopy of mass selected clusters

Freie Universität Berlin

Uni Rostock

BESSY

Universität Hamburg

Uni Würzburg

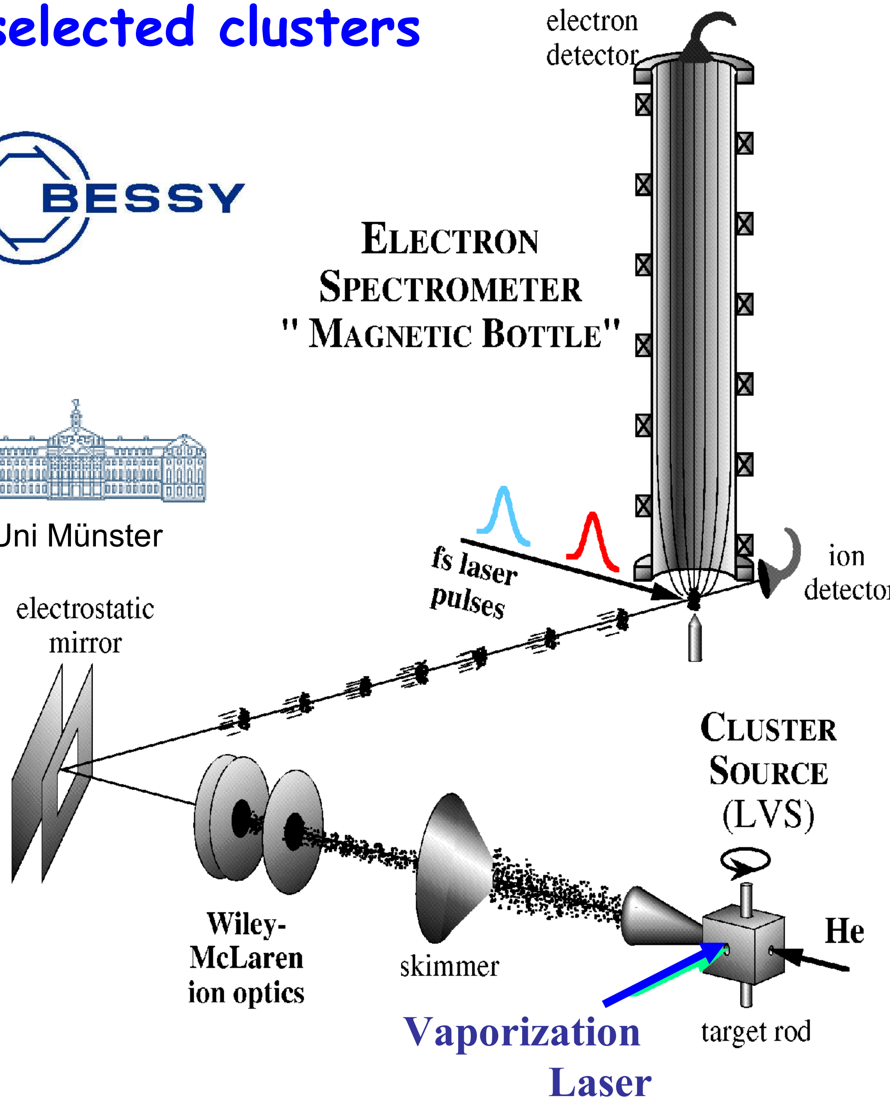
Uni Münster

JOHANN WOLFGANG GOETHE UNIVERSITÄT FRANKFURT AM MAIN

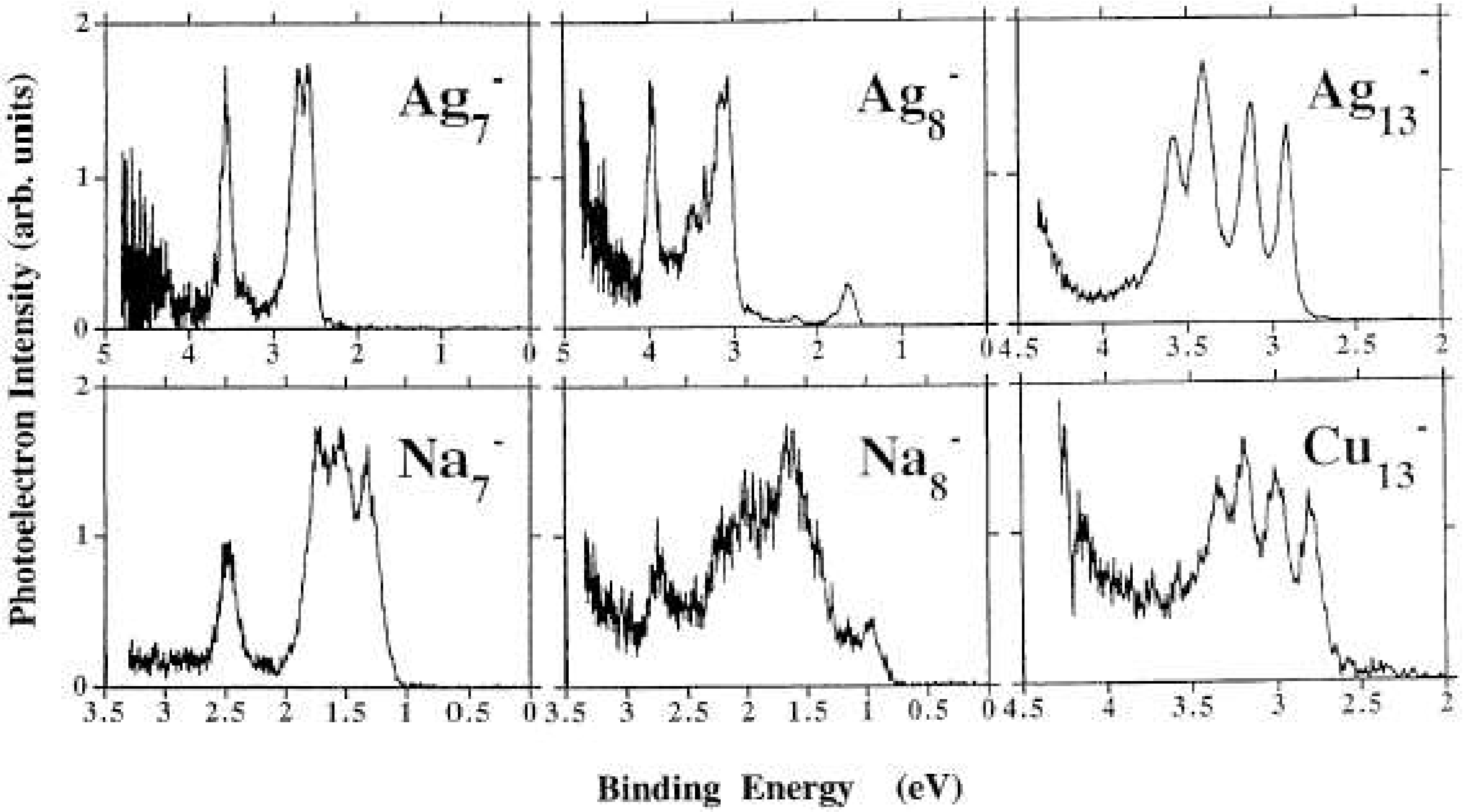
TU berlin

DESY

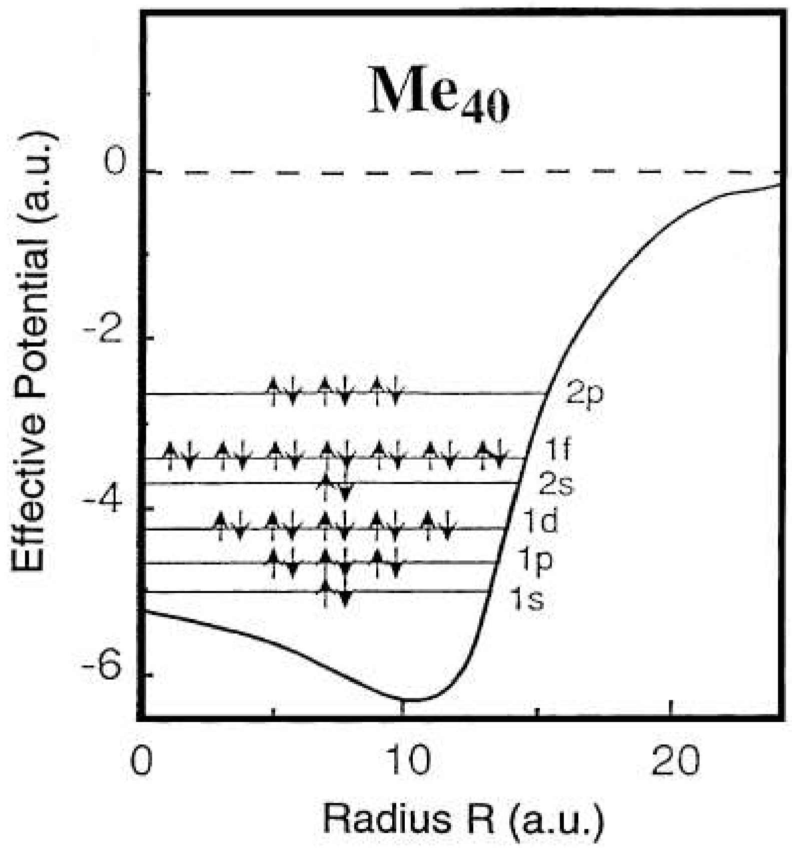
Universität Konstanz



# Searching for Electronic Shells




**Cu-, Ag-, Au-, and Alkali-Clusters clearly exhibit a shell pattern for the delocalized atomic s-electrons**



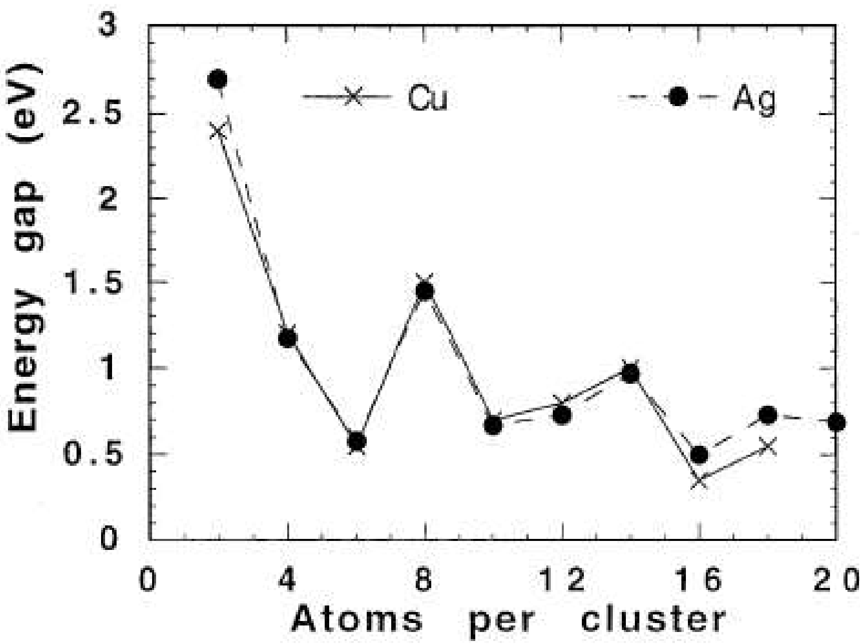

How do we **define** ,metallic' behavior ?

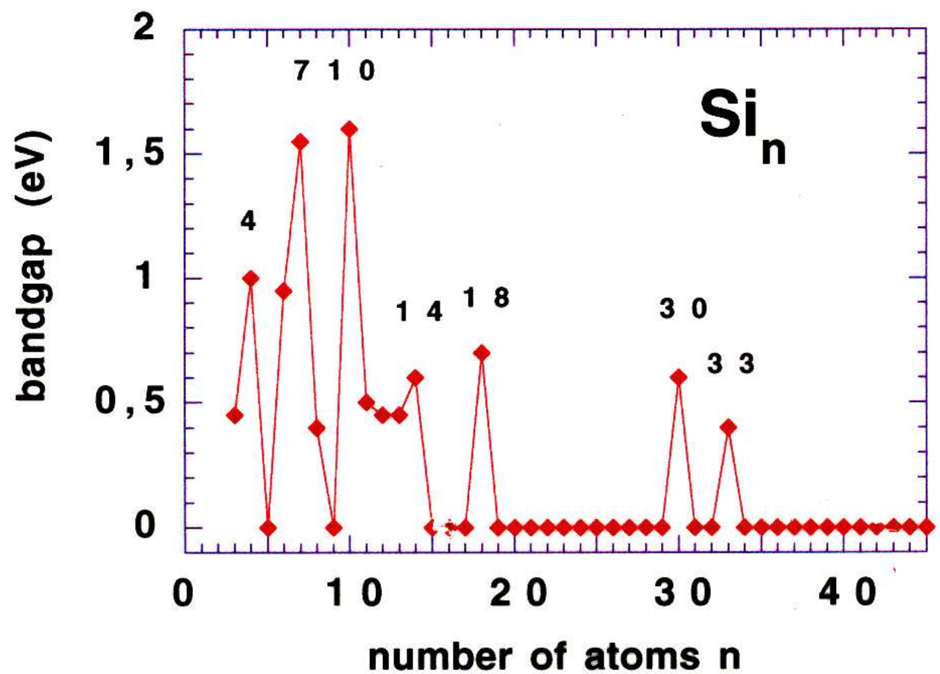
How do we **detect** ,metallic' behavior ?

**Delocalized electronic states**

**Electronic shell model**

# The 'bandgap' of small Clusters




**Electronic shells and the bandgap of Cu and Ag clusters**

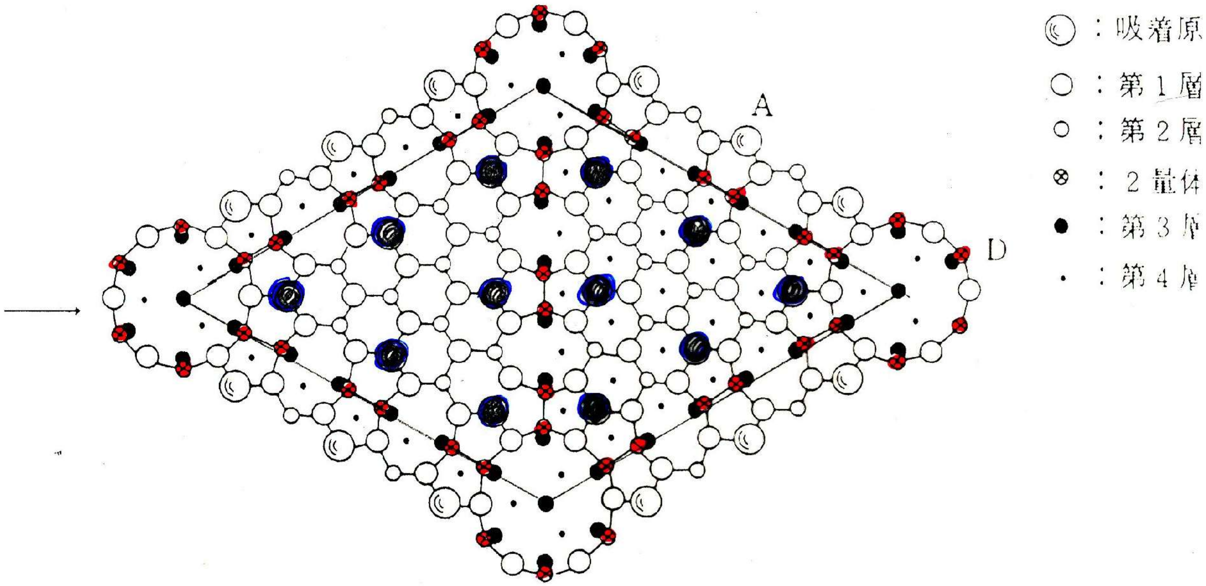
C.Y. Cha, G. Ganteför, W, Eberhardt
J. Chem. Phys. 99, 6308 (1993)
H. Handschuh, C.Y. Cha, P.S. Bechthold, G. Ganteför, W, Eberhardt
J. Chem. Phys. 102, 6406 (1995)

**The experimentally observed bandgap of small Si clusters**

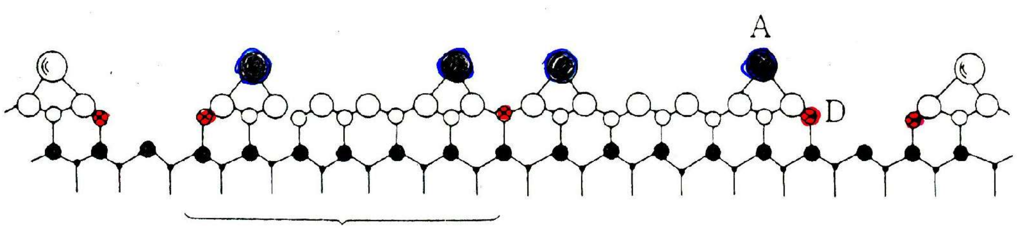
M. Maus, G. Ganteför, W. Eberhardt
Appl. Phys. A70, 535 (2000)

**With lasers only the top of the valence band is accessible**

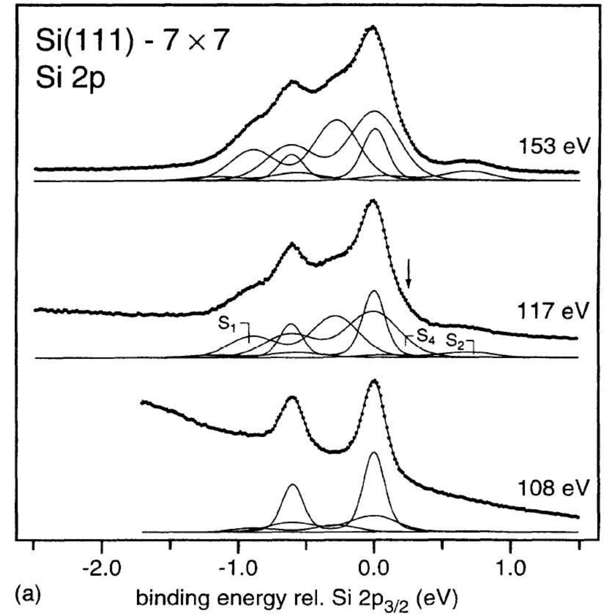
# High resolution Si 2p core level spectra of Si (111) 7x7

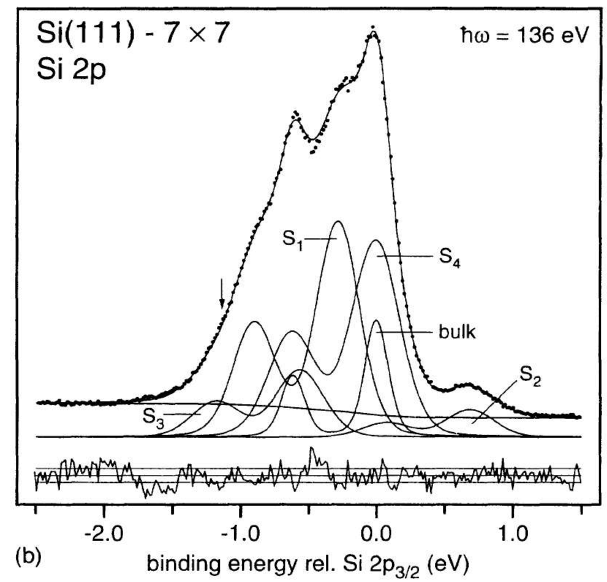



(a) 平面図



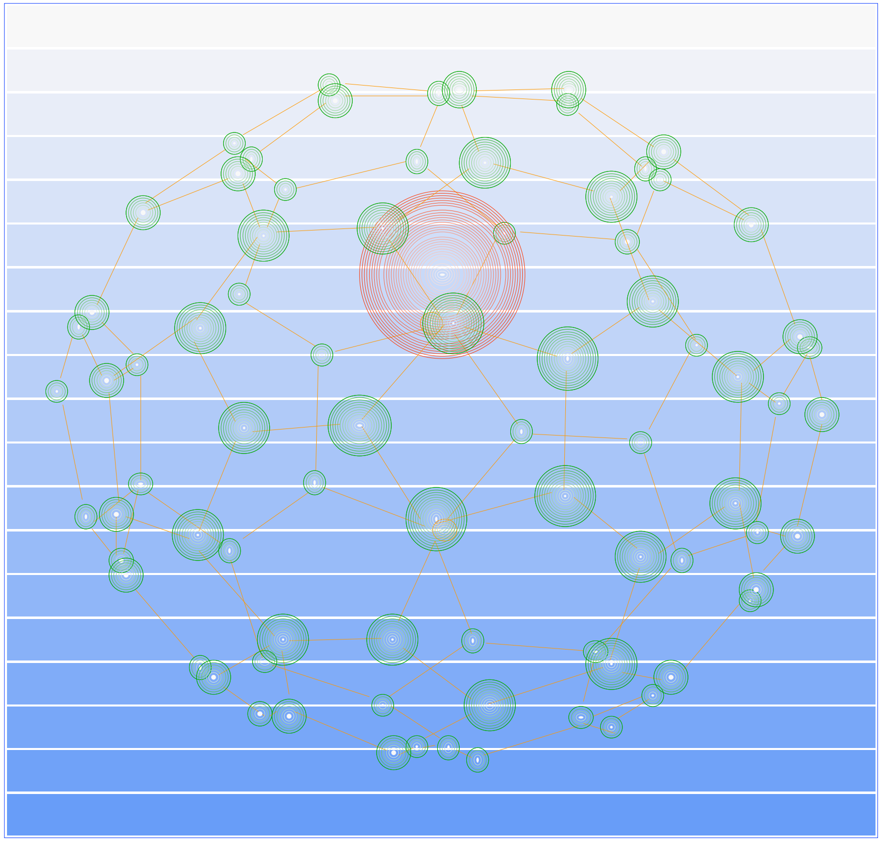

K. Takayanagi, Y. Tanishiro, S. Takahashi, M. Takahashi
Surf. Sci. **164**, 367 (1985)




J.J. Paggel, W. Theis, K. Horn
Ch. Jung, C. Hellwig, H. Petersen
Phys. Rev **B50**, 18686 (1994)

W. Eberhardt



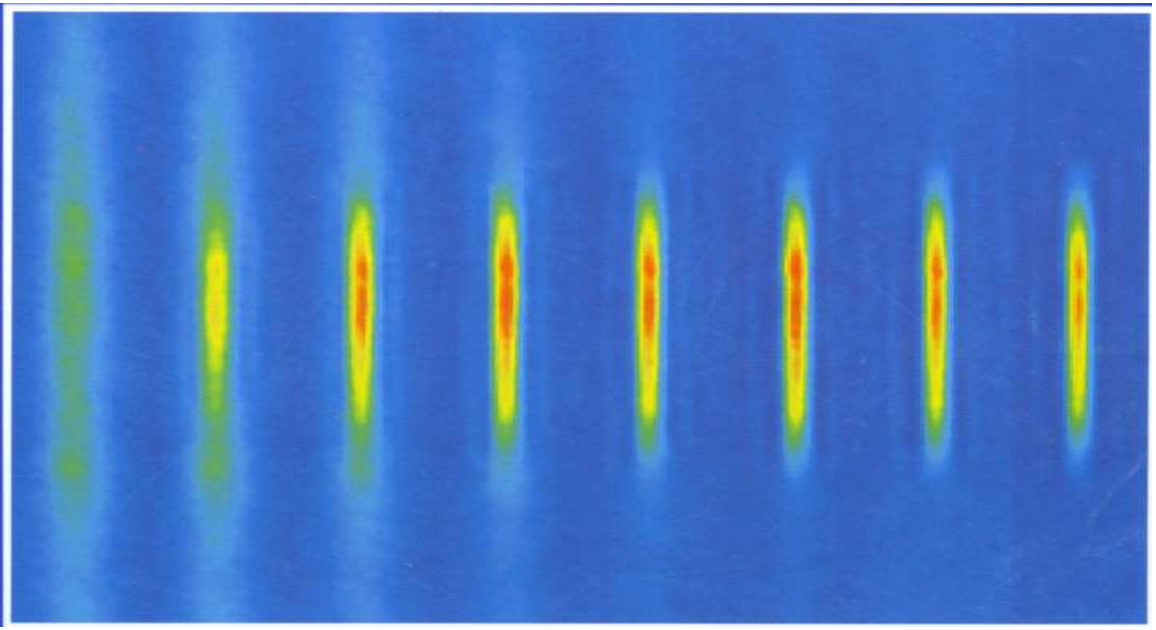
**Resonant core level photoemission spectroscopy of clusters**

**High resolution NEXAFS**

**At a Soft X-ray FEL the C, N, O 1s and the rare earth core levels are accessible**

# Visions of SCIENCE

## Atoms, Molecules and Ions
## New Fundamental Limits



**Bose condensate at different temperatures**
W. Ketterle, Phys. Bl. 53, 677 (1997)

**Spectroscopy of atoms and ions in traps**

**Quantum computing**

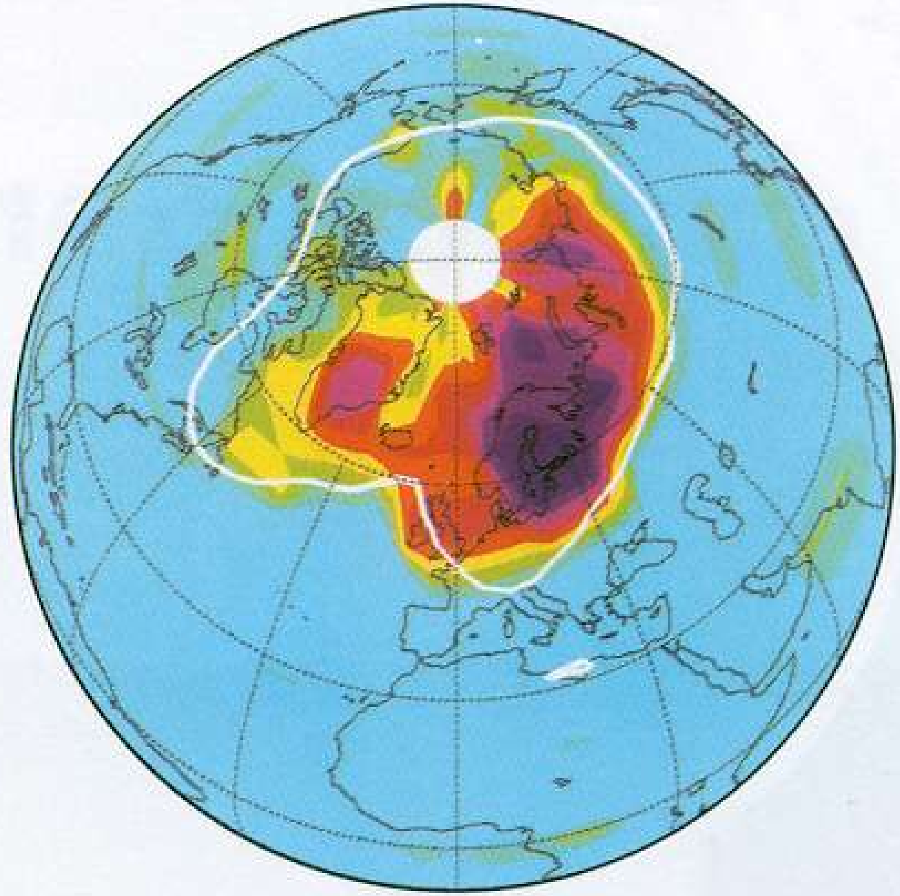
**Non-linear phenomena**

**Hollow atoms**

**Precision spectroscopy**

## Chemistry of Radicals

**Understanding the factors and processes influencing the global change in climate**

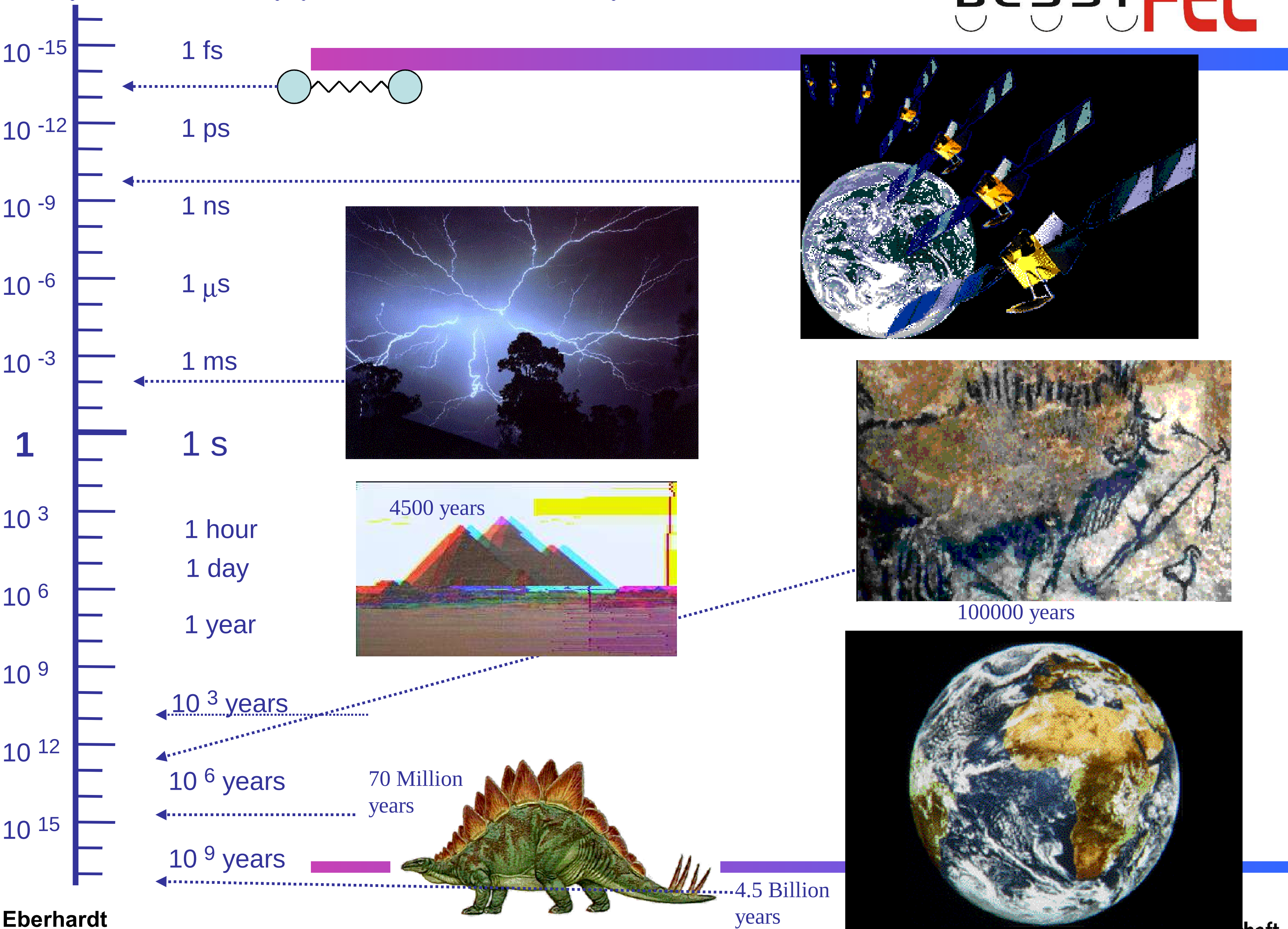


**At a Soft X-Ray FEL the C 1s, N 1s, and O 1s core levels are accessible**

**Ozone Hole in the Northern Hemisphere** (J. Waters, JPL)

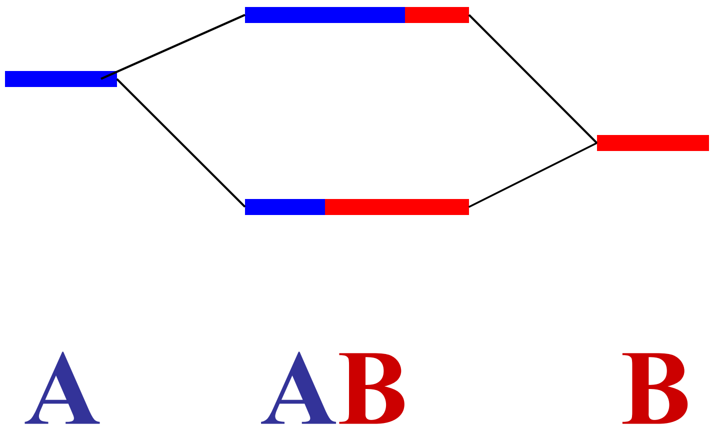
# Fs Spectroscopy: Relationship with Time

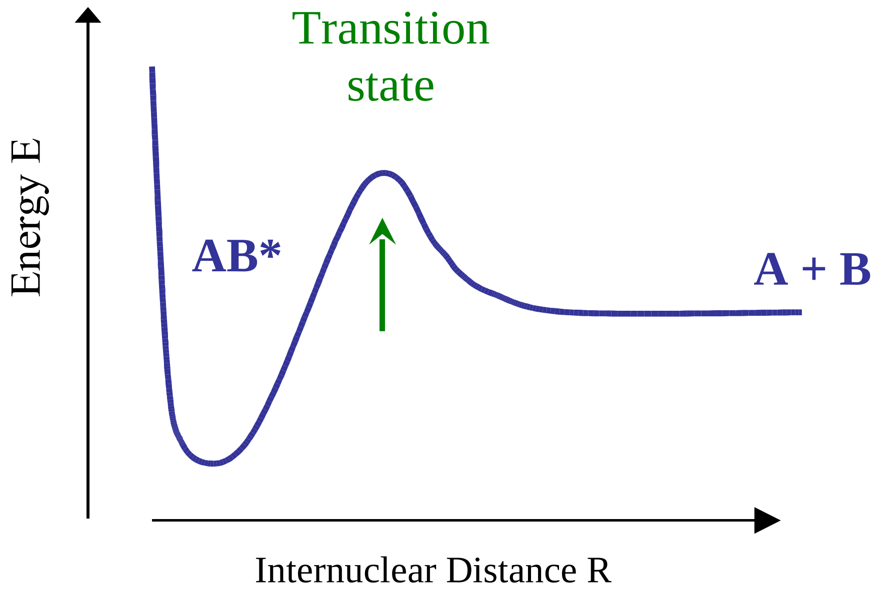




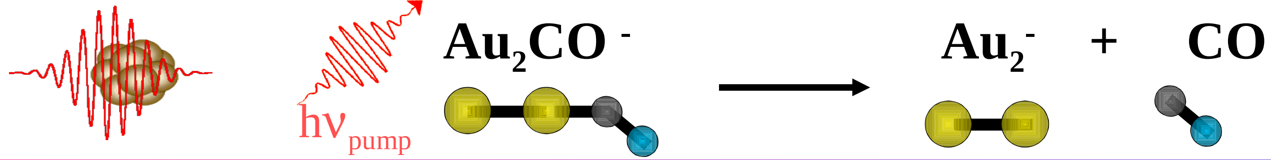
# FEMTOCHEMISTRY

## at surfaces, in molecules, and clusters

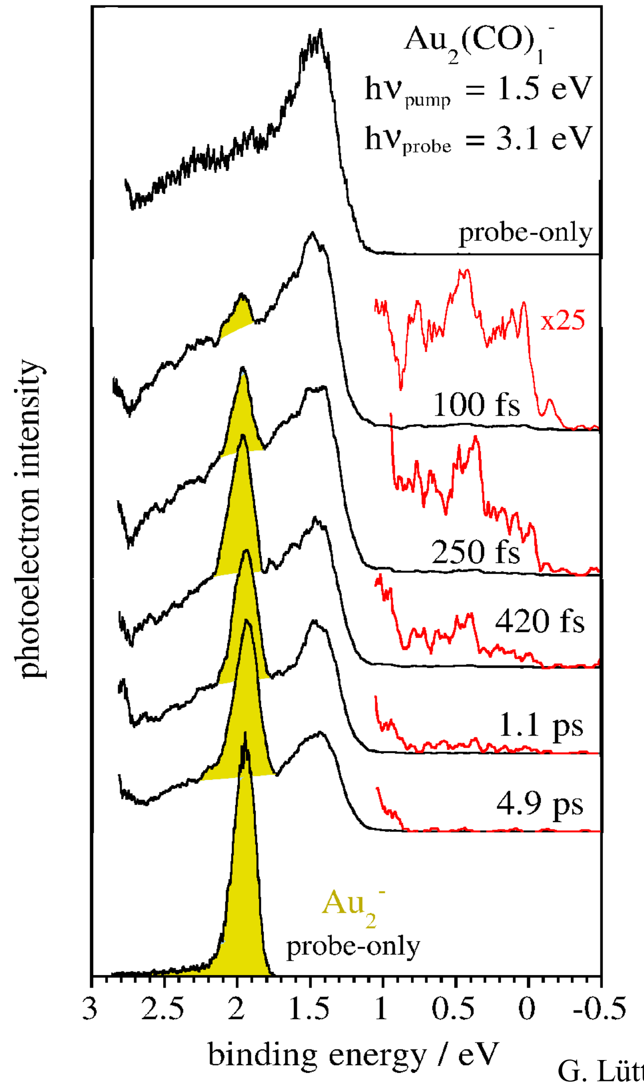



**Understanding the dynamics and formation of a chemical bond by time resolved electron spectroscopy**





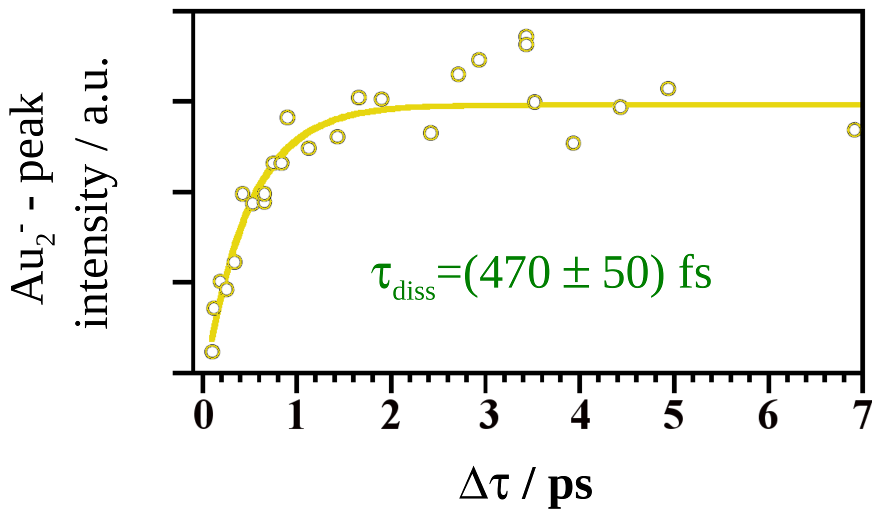



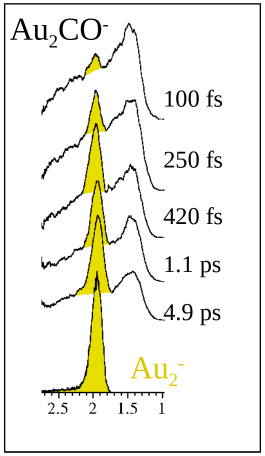

# Photon-induced dissociation

# time resolved pump probe photoemission studies

G. Lüttgens N. Pontius, M. Neeb, P.S. Bechthold, W. Eberhardt PRL **88**, 076102 (2002)

# ‘Thermal desorption’ of CO :

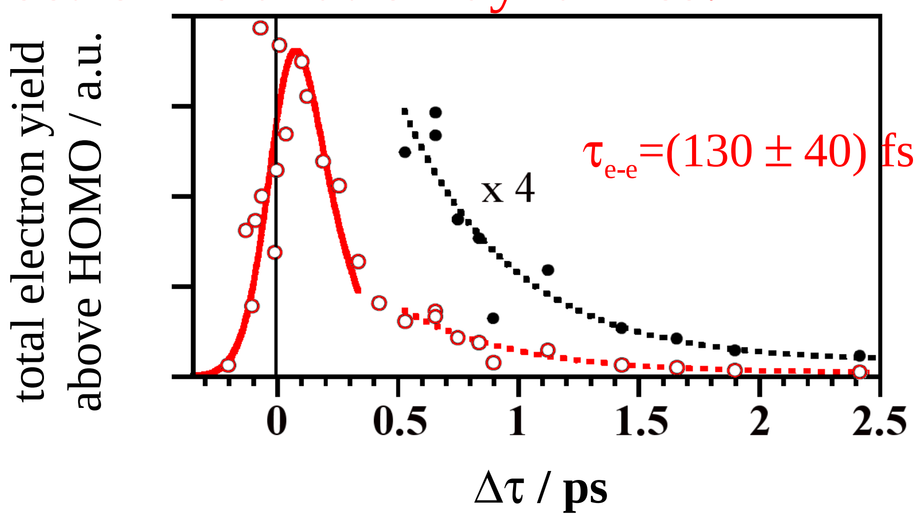


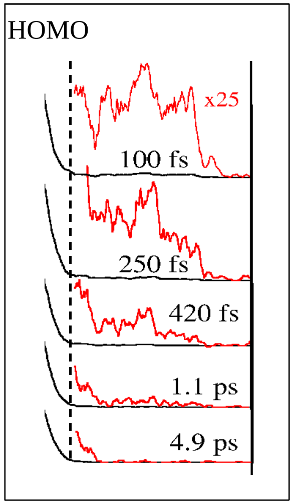




**$Au_2CO^-$**

**Desorption via phonon excitations**

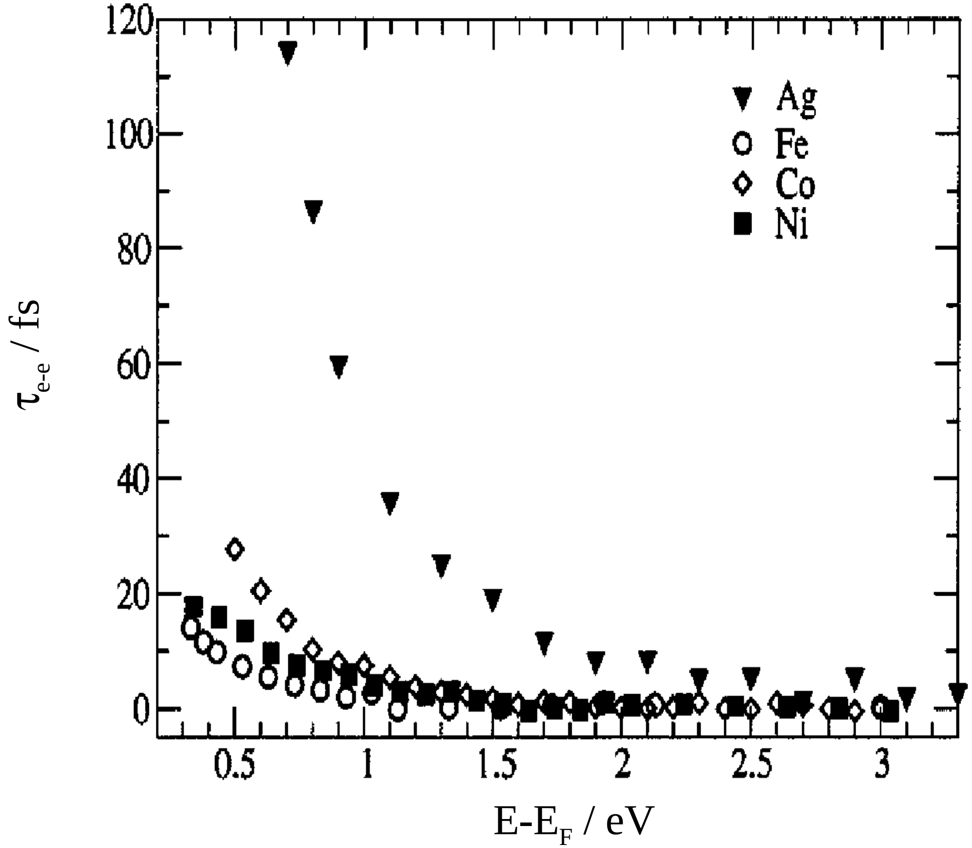
electron relaxation dynamics:

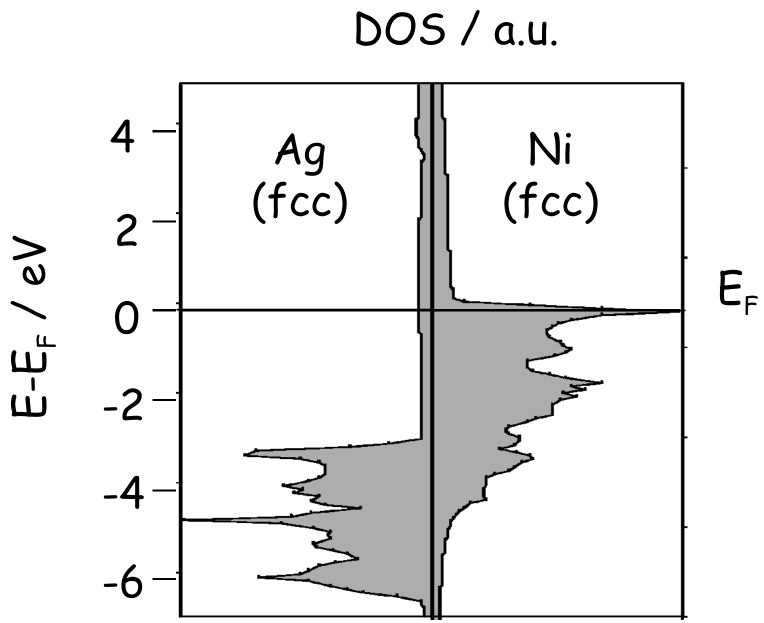


# $e^-$-$e^-$- scattering in metallic solids and at surfaces

BESSY FEL




**Synchronization is extremely important**

R. Knorren, K.H. Bennemann, R. Burgermeister, M. Aeschlimann
Phys. Rev. B 61, 9427 (2000)

# Control of the Pulse Shape <==> Coherent control (of the process)

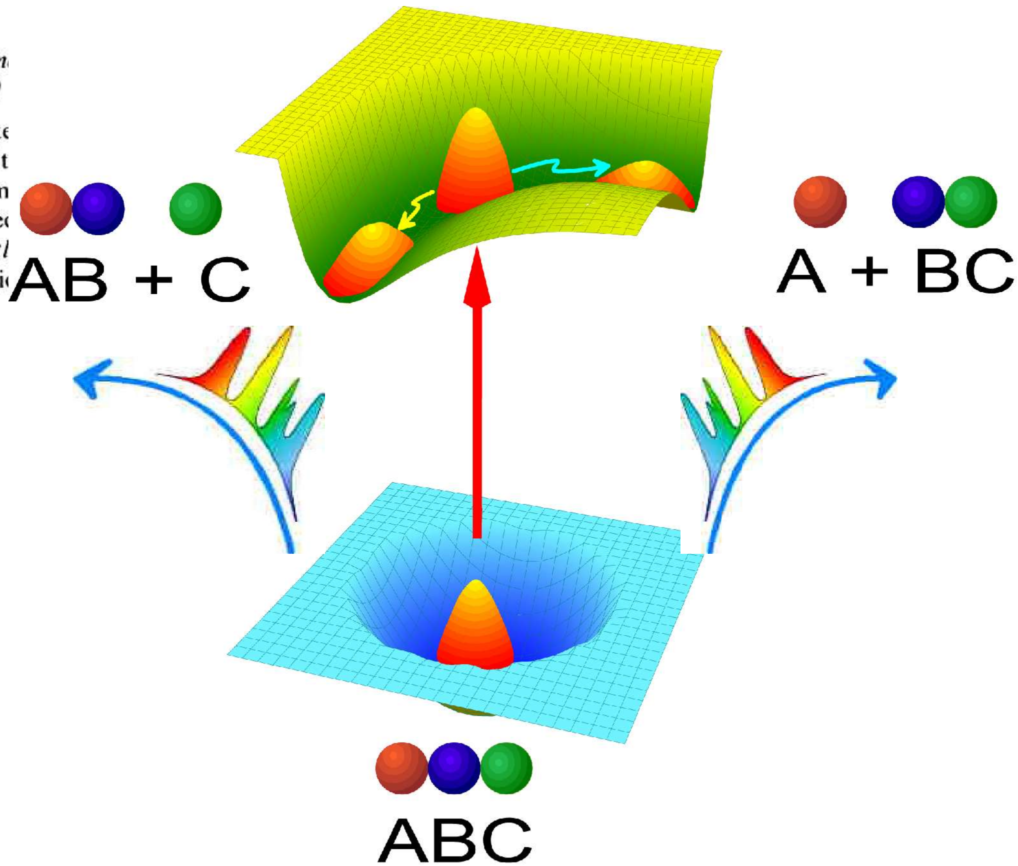
VOLUME 68, NUMBER 10 PHYSICAL REVIEW LETTERS 9 MARCH 1992

## Teaching Lasers to Control Molecules

Richard S. Judson[a]
*Center for Computational Engineering, Sandia National Laboratories, Livermore, California 94551-0969*

Herschel Rabitz
*Department of Chemistry, Princeton University, Prin*
(Received 26 August 1991)

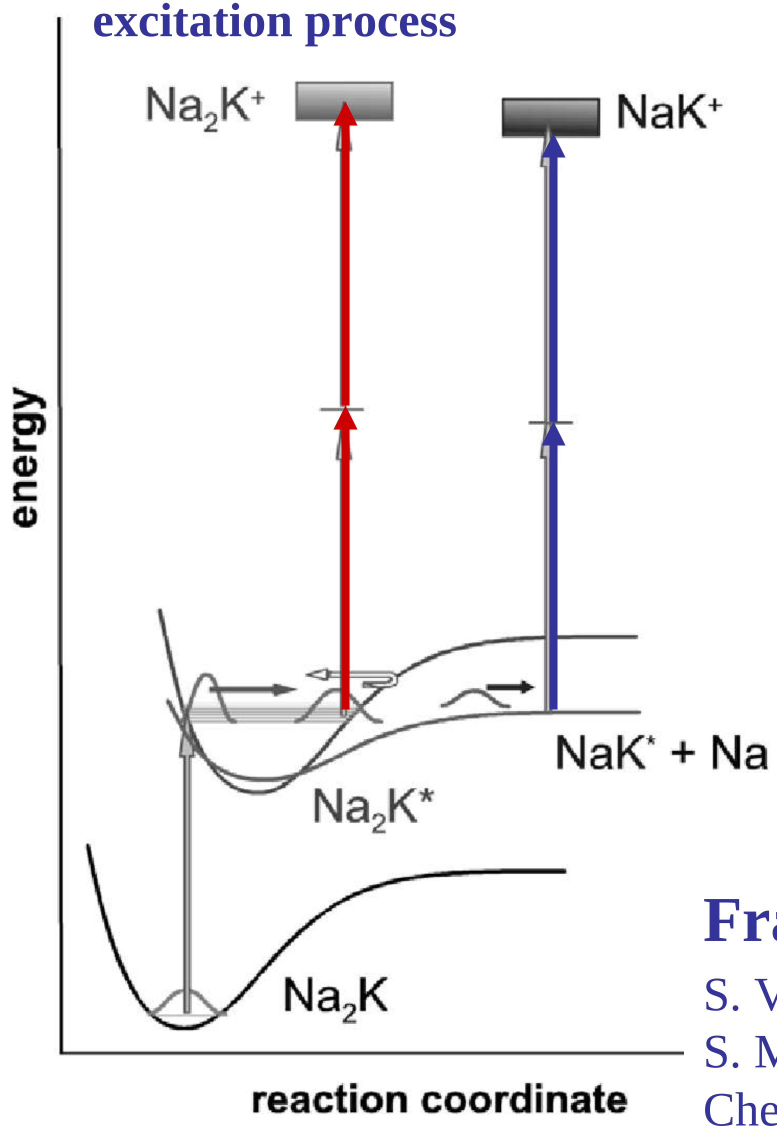
We simulate a method to teach a laser pulse sequences to excite
learning procedure to direct the production of pulses based on "fit
ratory measurement device. Over a series of pulses the algorithm
perimental apparatus, which consists of a laser, a sample of mole
as an analog computer that solves Schrödinger's equation *exactl*
paratus that learns to excite specified rotational states in a diatomi

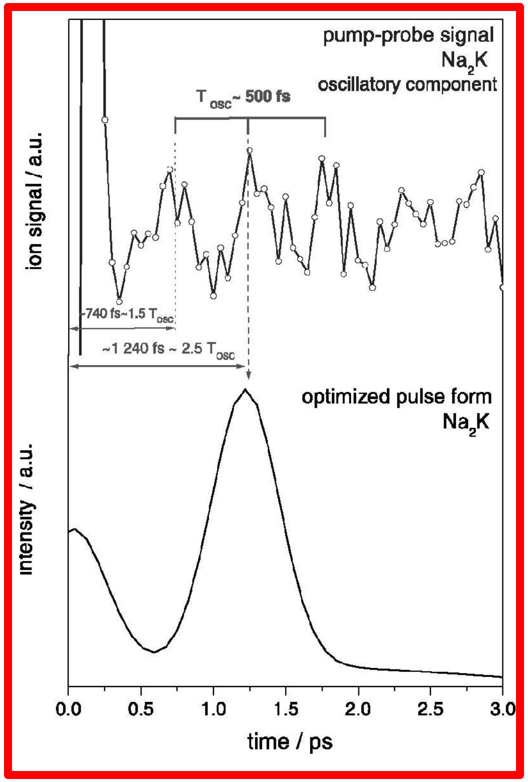


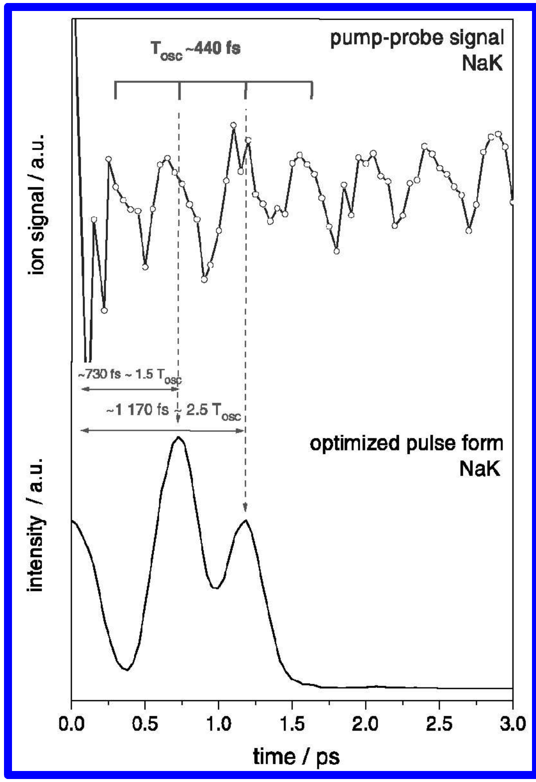

**The HGHG FEL delivers (double) pulses with controlled shape**

# Control of the Pulse Shape <==> Coherent control (of the process)

## Schematics of the 3 photon excitation process



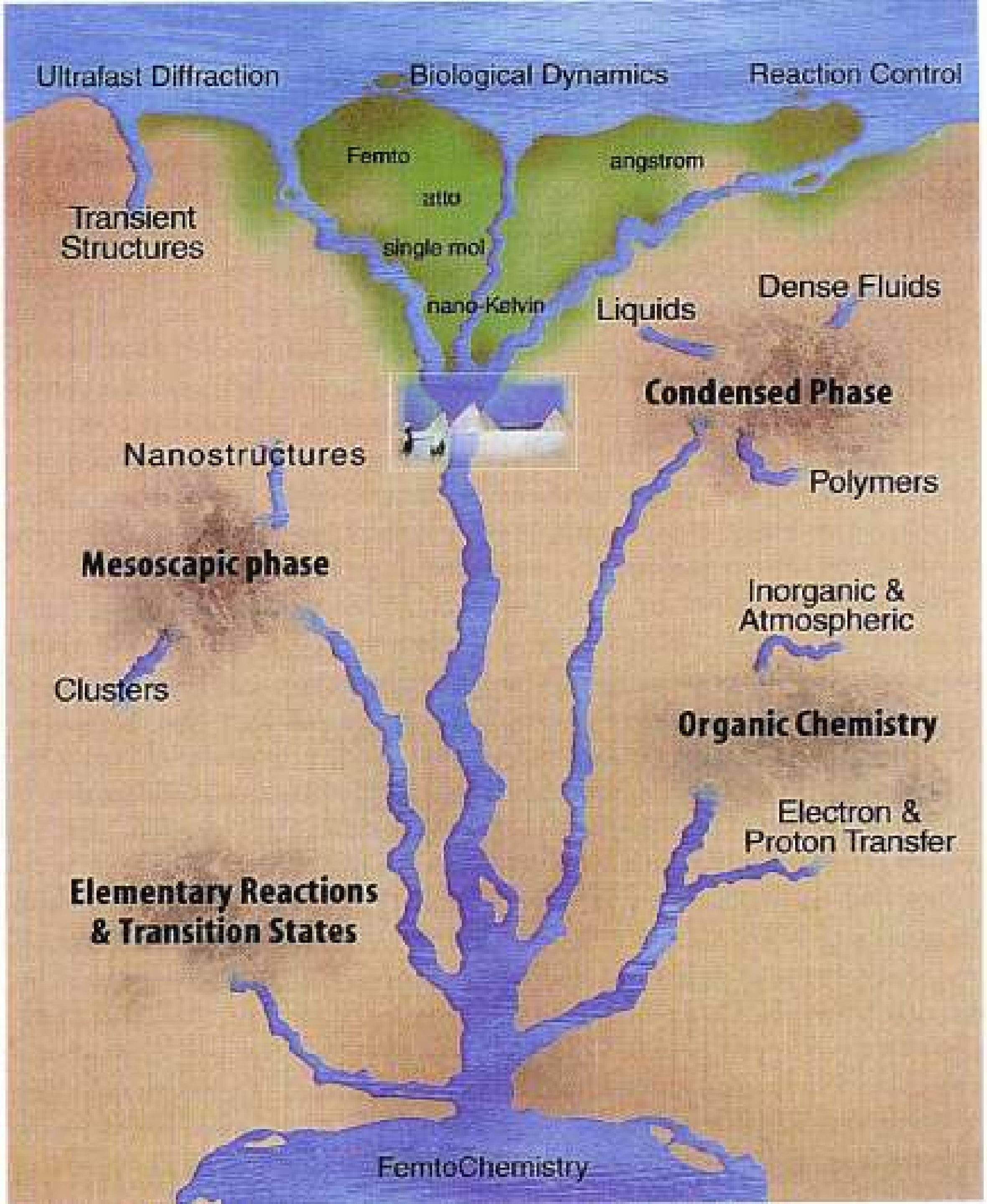




## Fragmentation of $Na_2K$

S. Vajda, A. Bartelt, E.C. Kaposta, T. Leisner, C. Lupulescu
S. Minemoto, P. Rosendo-Francisco, L. Wöste
Chem. Phys. **267**, 231 (2001)

BESSY FEL




# Femto-Science

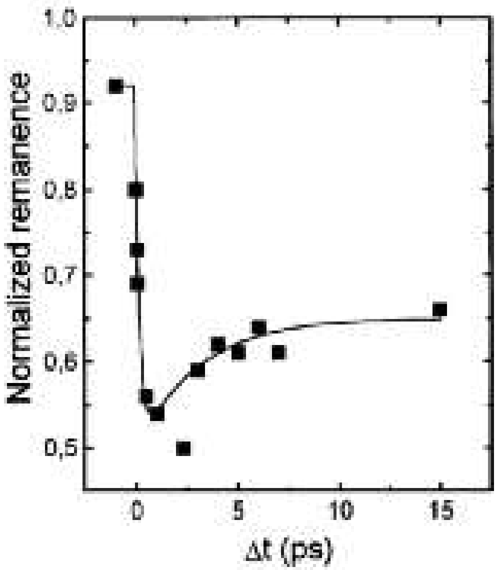
Ahmed H. Zewail
Nobel Prize 1999

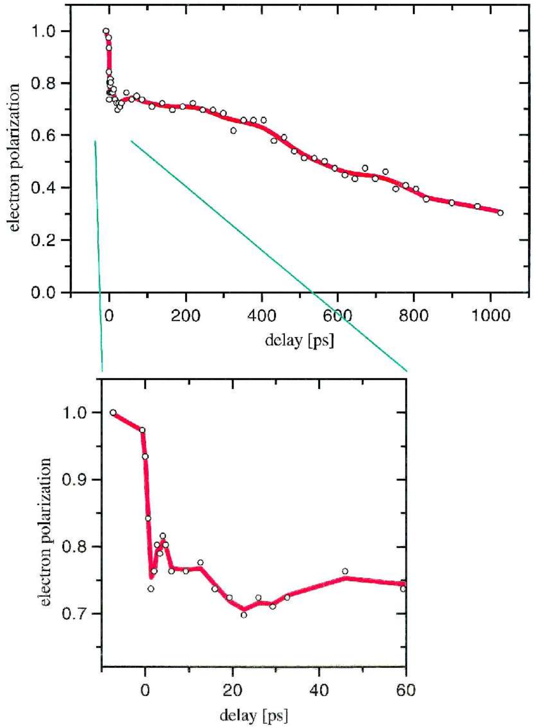
Leibniz Gemeinschaft

## Dynamic optical measurements (Kerr, SHG)

**E. Beaurepaire, J.C. Merle, A. Daunois J.Y. Bigot, PRL 76, 4520 (1996)**

**J. Hohlfeld, E. Matthias, R. Knorren, K.H. Bennemann, PRL 79, 5149 (1997)**



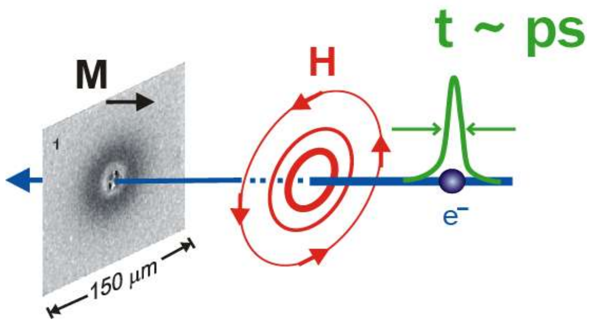




## Spin polarized photoemission with fs laser pulses

**A. Scholl, L. Baumgarten, R. Jacquemin W. Eberhardt, PRL 79, 5146 (1997)**

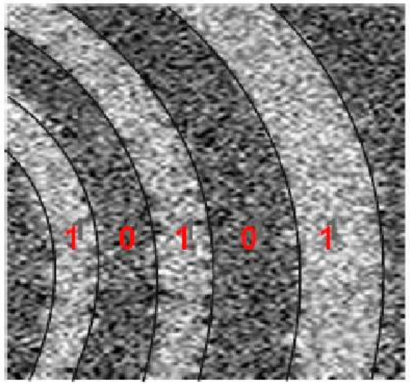
# Excitation by magnetic field pulse

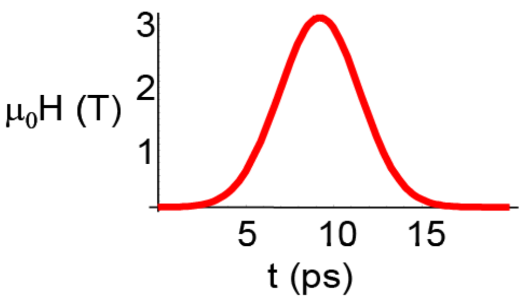
relativistic electrons shot through magnetic samples




linear accelerator
(SLAC, Stanford)

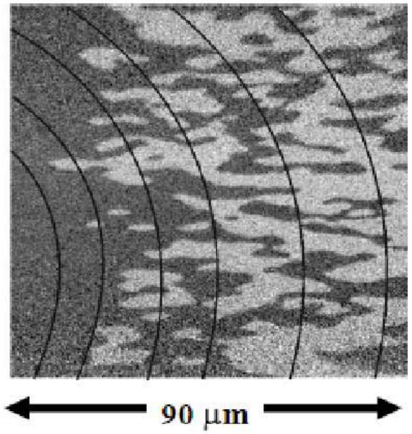
field pulse
duration:
3 ps



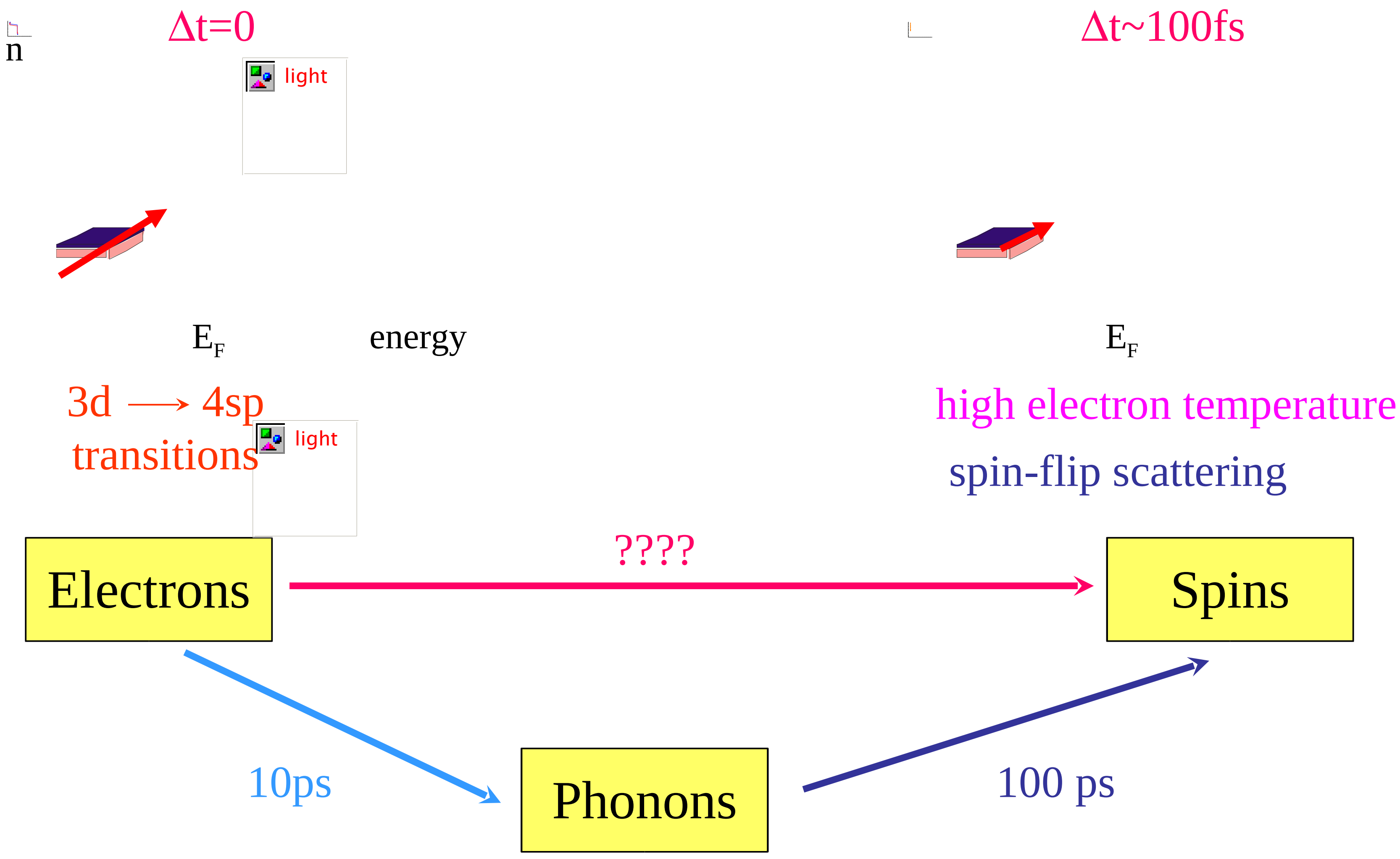

moving electrons
generate ultrashort
magnetic field

100 fs

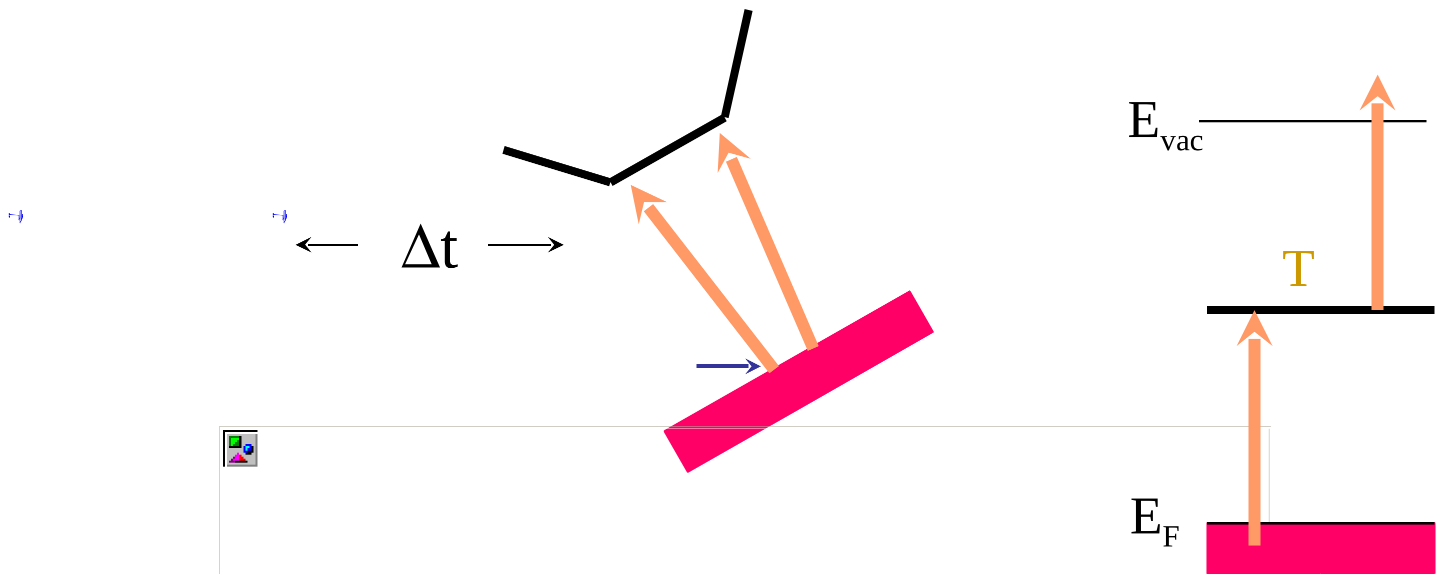



I.Tudosa, C. Stamm, A.B. Kashuba, F. King, H.C. Siegmann, J. Stöhr, G. Ju, B. Lu, H.D. Weller
NATURE 428, 831 (2004)
C. Stamm, I. Tudosa, H.C. Siegmann, J. Stöhr, A.Yu. Dobin, G. Woltersdorf, B. Heinrich, A. Vaterlaus
Phys. Rev. Lett. 94, 197603 (2005)

# Energy relaxation mechanisms



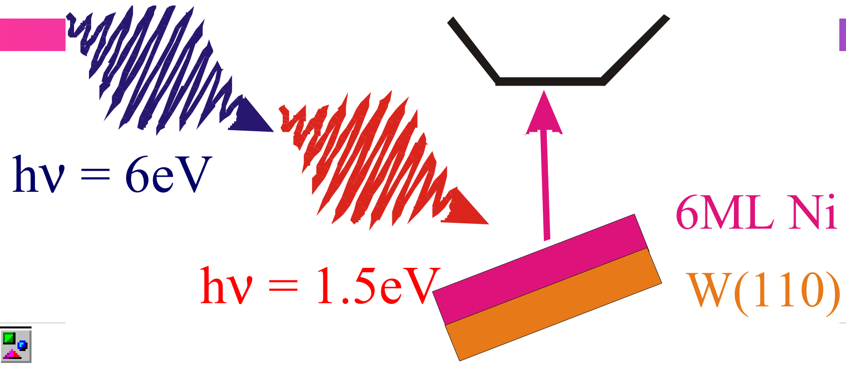
# Time-resolved two-photon photoemission

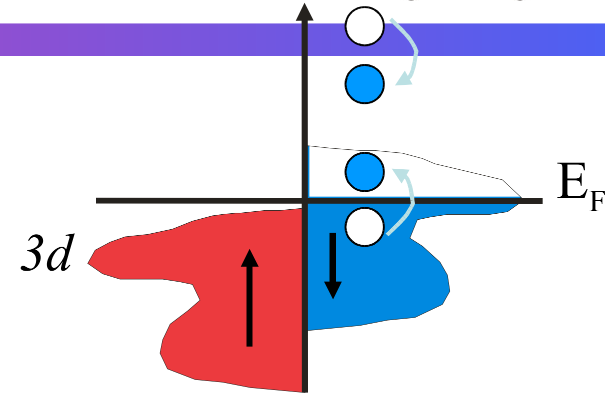



**FWHM → T**

H.S. Rhie, H.A. Dürr, W. Eberhardt, **PRL 90**, 247201 (2003)

# Fs electron dynamics in Ni/W(110) films



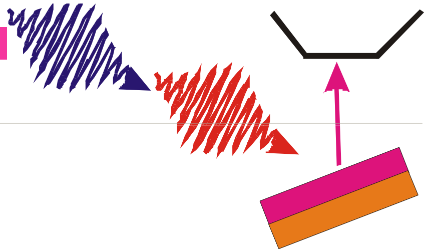

time-resolved photoemission

number of electrons above $E_F$



H.S. Rhie, H.A. Dürr, W. Eberhardt, **PRL 90**, 247201 (2003)

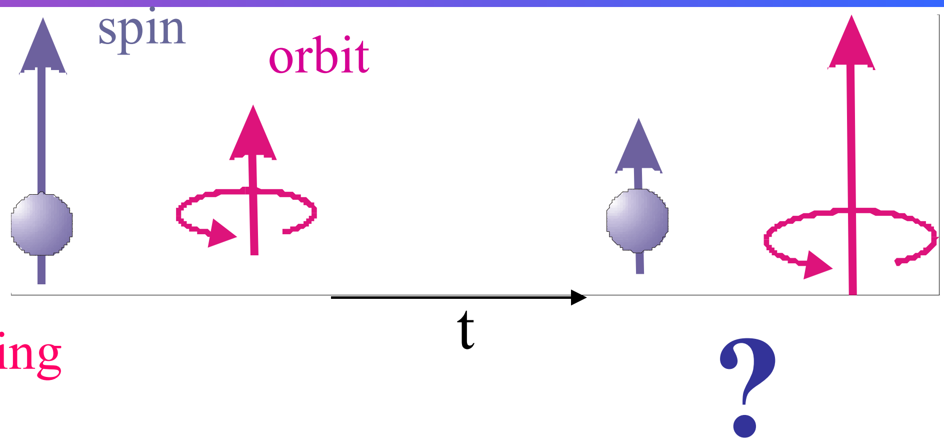
# Ultrafast spin dynamics in Ni





H.S. Rhie, H.A. Dürr, W. Eberhardt, **PRL 90**, 247201 (2003)

# Ultrafast energy relaxation in Ni




Spin excitations

spin-orbit coupling
~ 300 fs

hν

light%20horizo

Electrons

electron-phonon coupling
~ 2 ps

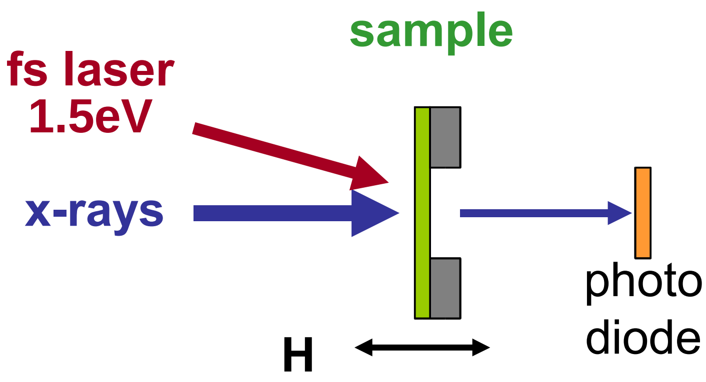
Phonons



H.S. Rhie, H.A. Dürr, W. Eberhardt, **PRL 90**, 247201 (2003)

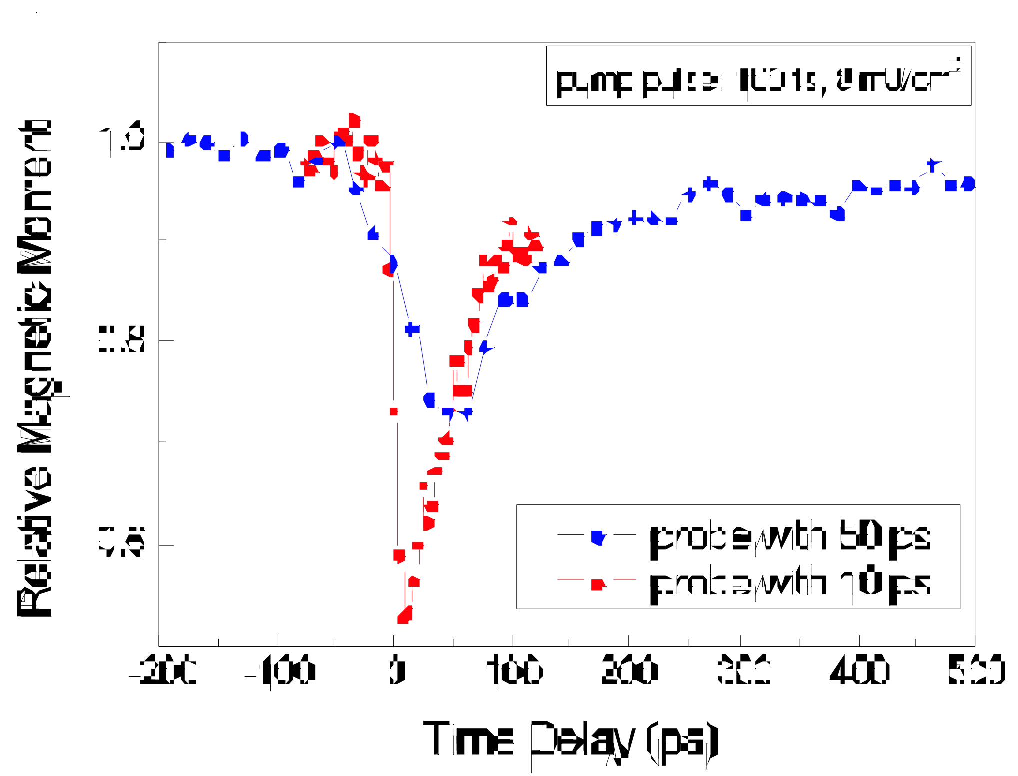
# laser pump - x-ray probe

## x-ray transmission setup




correlate 1.5 eV (fs laser) with 700-900eV (x-ray)

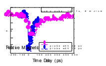
## dynamic magnetic signal



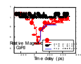
# Pump-Probe Spectra

BESSY FEL

single-bunch vs.
low-alpha mode



re-establishing the
magnetic moment

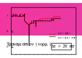


**Dynamic response**
limited by x-ray pulse
length and jitter

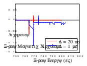
**heat transfer**

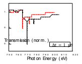
| | |
|---|---|
| perpendicular | 100 nm in 100 ps |
| lateral | 1 mm in 1 µs |

# Time resolved (Dynamic) XMCD-Spectra



Dichroism





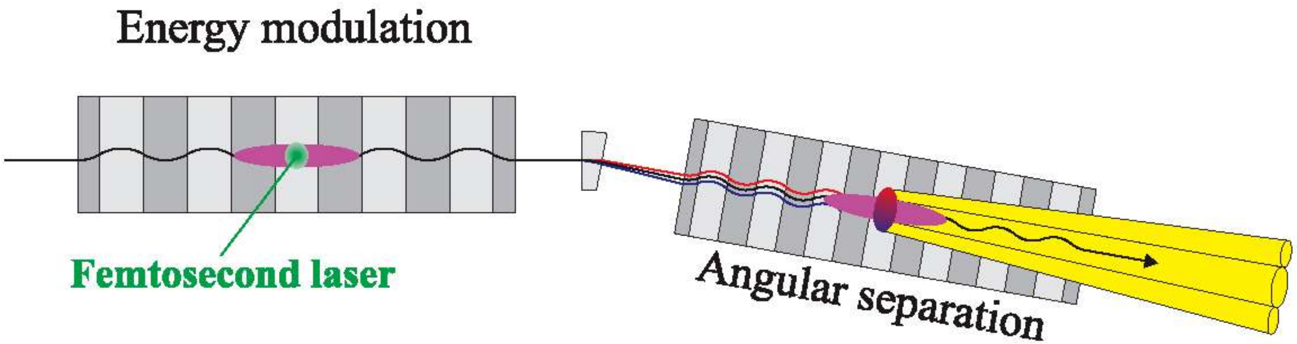
sum rule analysis:

S, L decrease by 35%

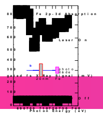
# Fs Slicing

## Fs slicing at BESSY II

- ALS pioneered fs slicing with signal:background ~ 1:1
- BESSY pioneered angular separation scheme with signal:background >10:1
- SLS (2006) and SOLEIL (~2008) will implement angular separation




**Planning/commissioning:** S. Khan, H. Dürr, C. Stamm, C. Lupulescu, T. Kachel, T. Quast, K. Holldack, A. Erko, A. Firsov
**External user operation (start 2006)**



# Fs Slicing

## HGHG-FEL Test Bench

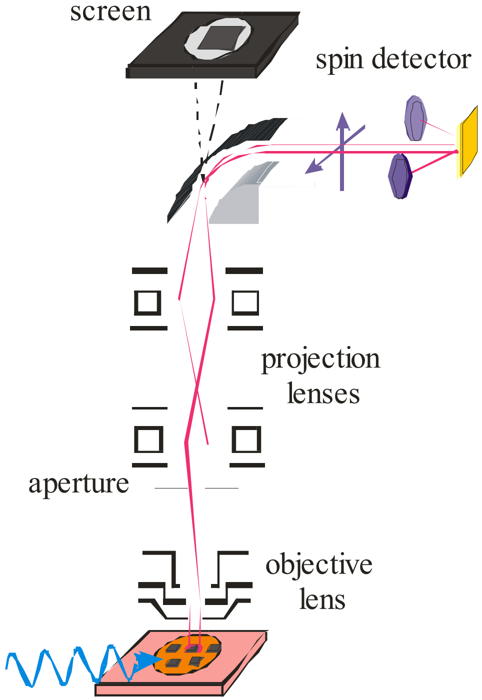
Laser electron interaction
Synchronization
Energy transfer
THz-Diagnostics

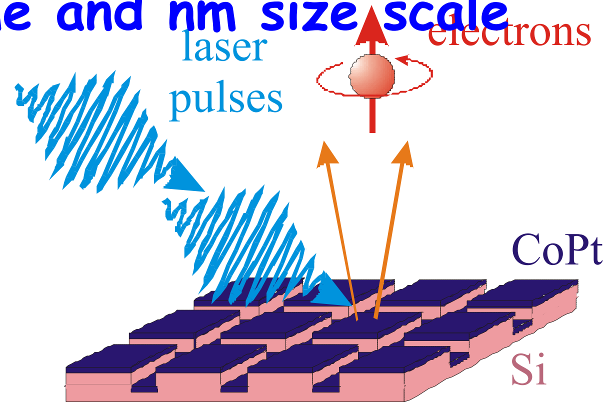
Pump-probe experiments
Synchronization
Beam transfer
X-ray pulse characterization

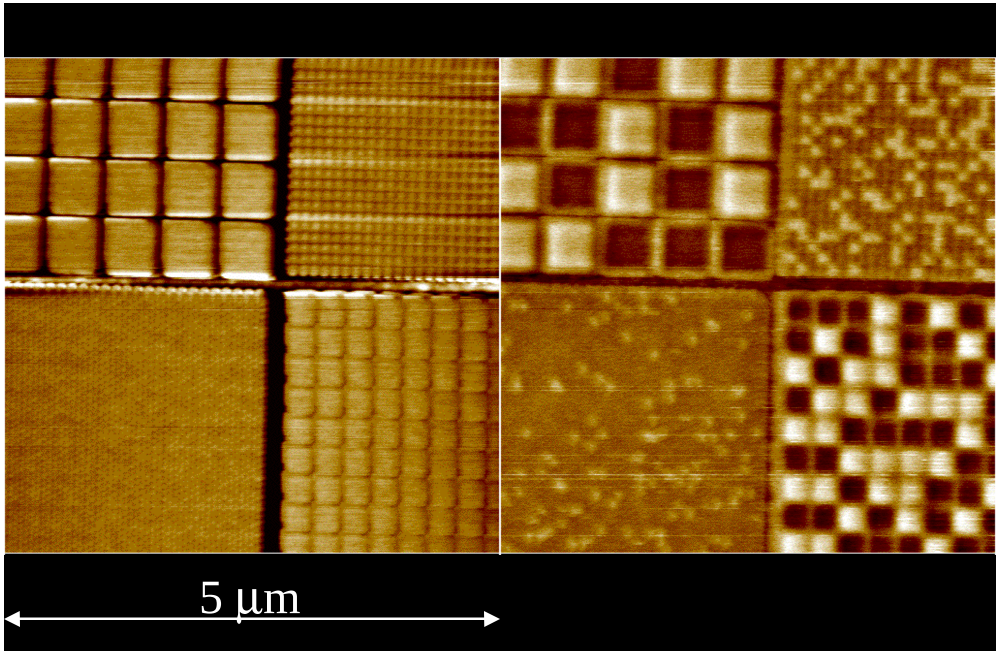
# Visions of SCIENCE

BESSY FEL

## Magnetization dynamics on the fs time and nm size scale



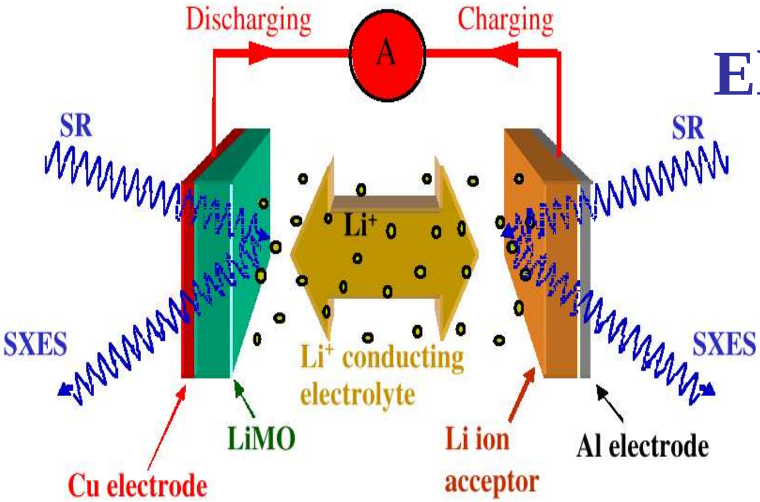




Hitachi Global Storage Technologies

S-PEEM
Spin Polarized Photo-Electron Emission Microscopy

## Improvements in Technology

# In-situ process monitoring using all photon related spectroscopies

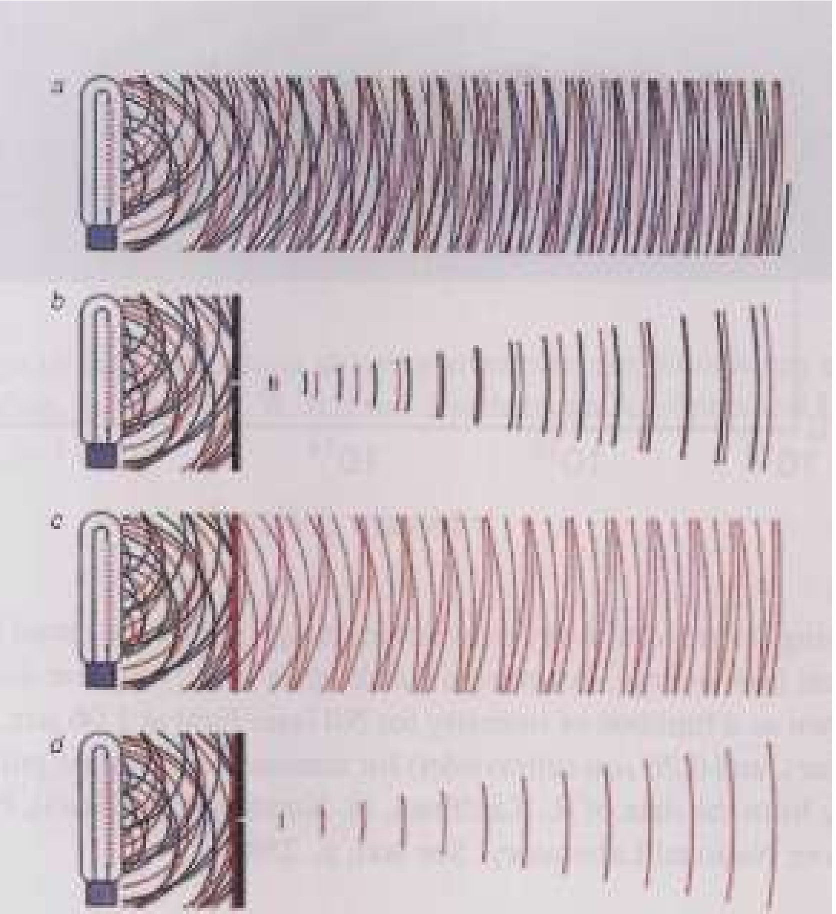



Electrochemistry
Corrosion
Lubrication
Catalysis

Spectroscopy of battery electrodes under operational conditions

Pattern formation during a chemical reaction
G. Ertl FHI Berliin

# Coherent light scattering --- Holography

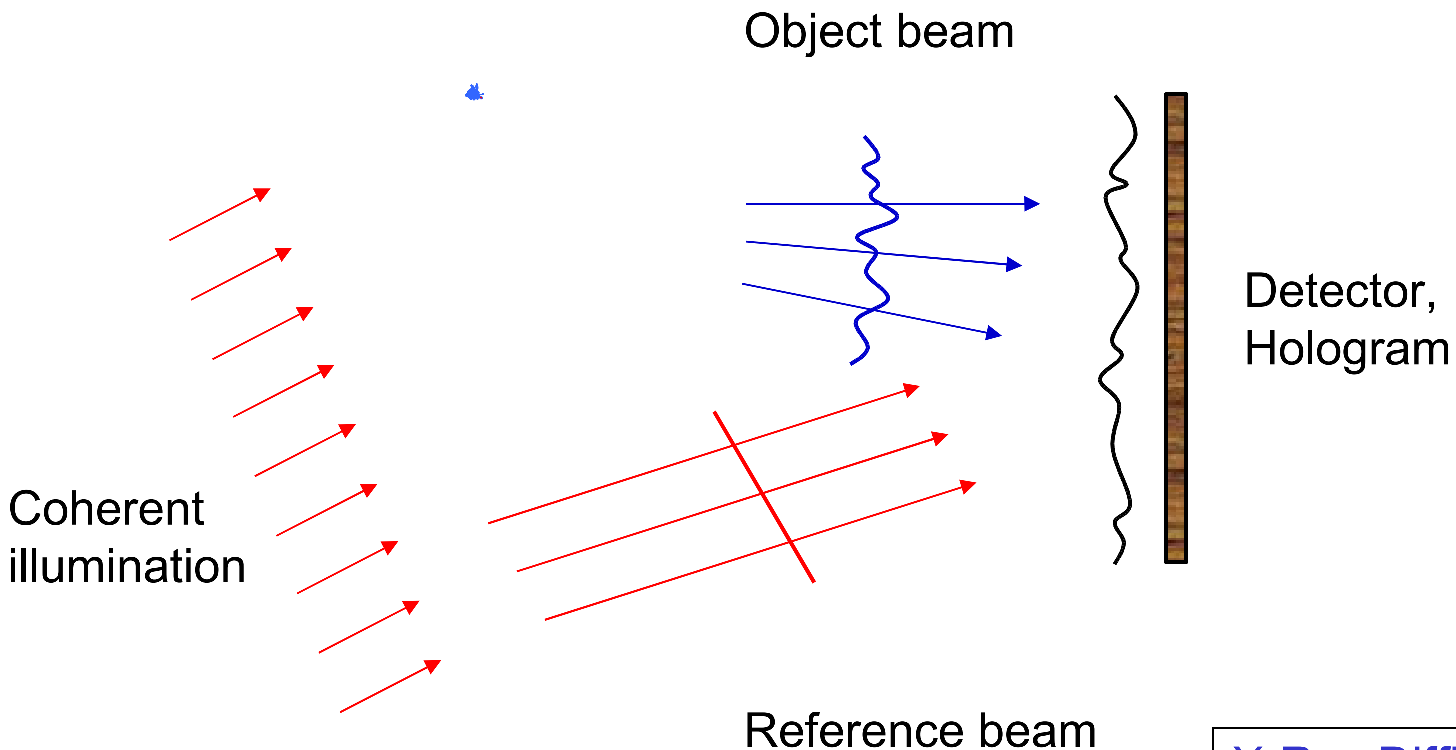


A. Schawlow, Sci. Am. **219**, 120 (Sept 1968)

The FEL
delivers not only
transverse but also
longitudinal
coherence

spatial & spectral filtering

# Holography Principle

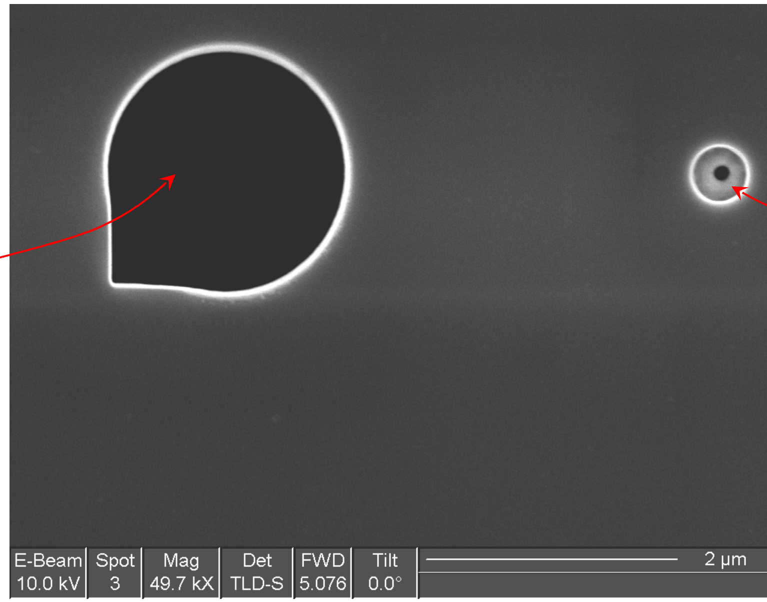



X-Ray Difficulties:
- low coherent flux
- beam splitter

## Microstructured Mask

Focussed Ion Beam:
W.F. Schlotter

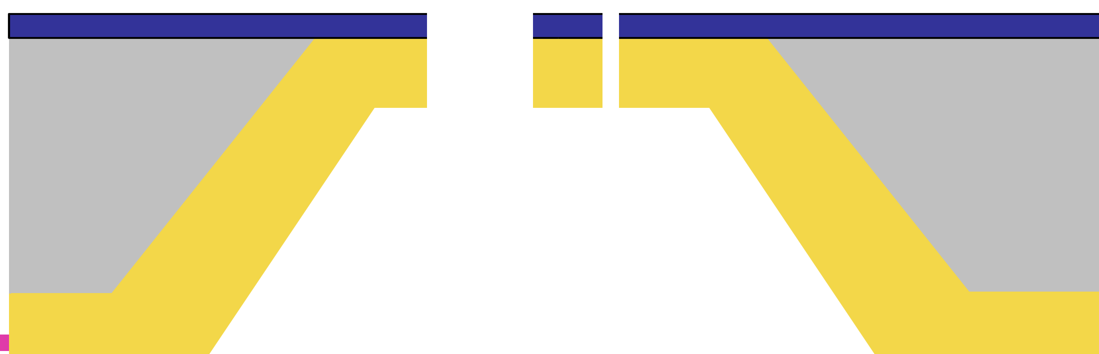
top




Object area
⌀ 2 µm

Reference hole
⌀ 100 nm

side



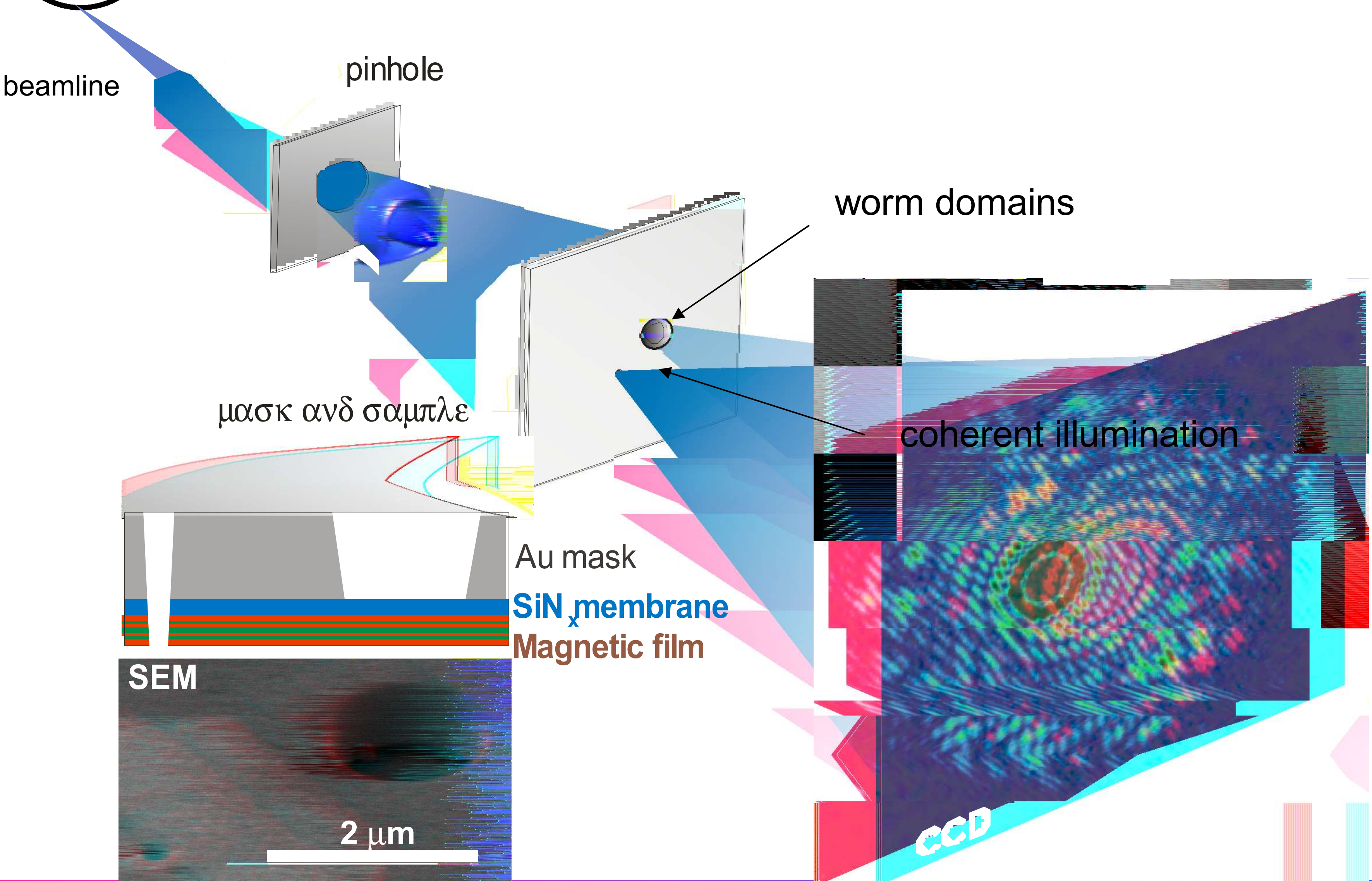
← $Si_3N_4$ (100nm) Membrane

← Si Substrate

← Au (600nm)

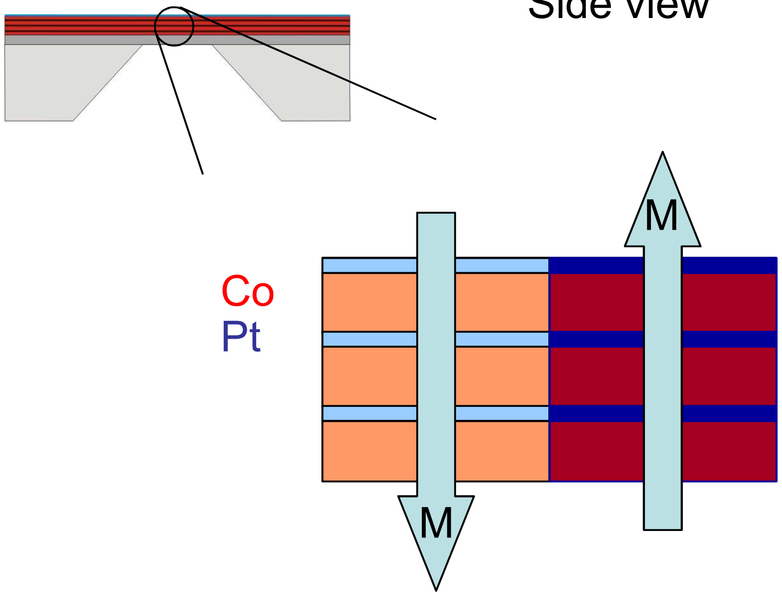
# Fourier Transform Holografy



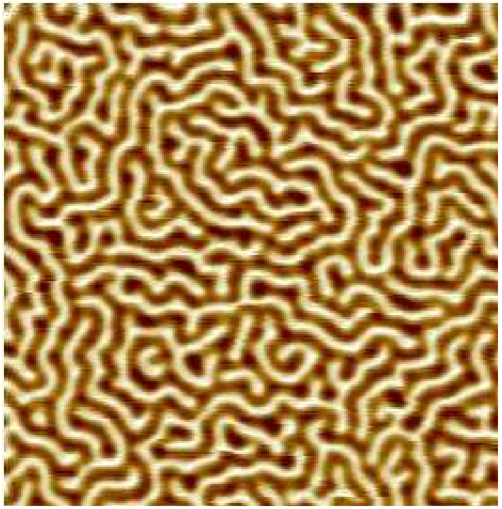


# Novel Magnetic Recording Media

Side view




MFM, top view



5 µm x 5 µm

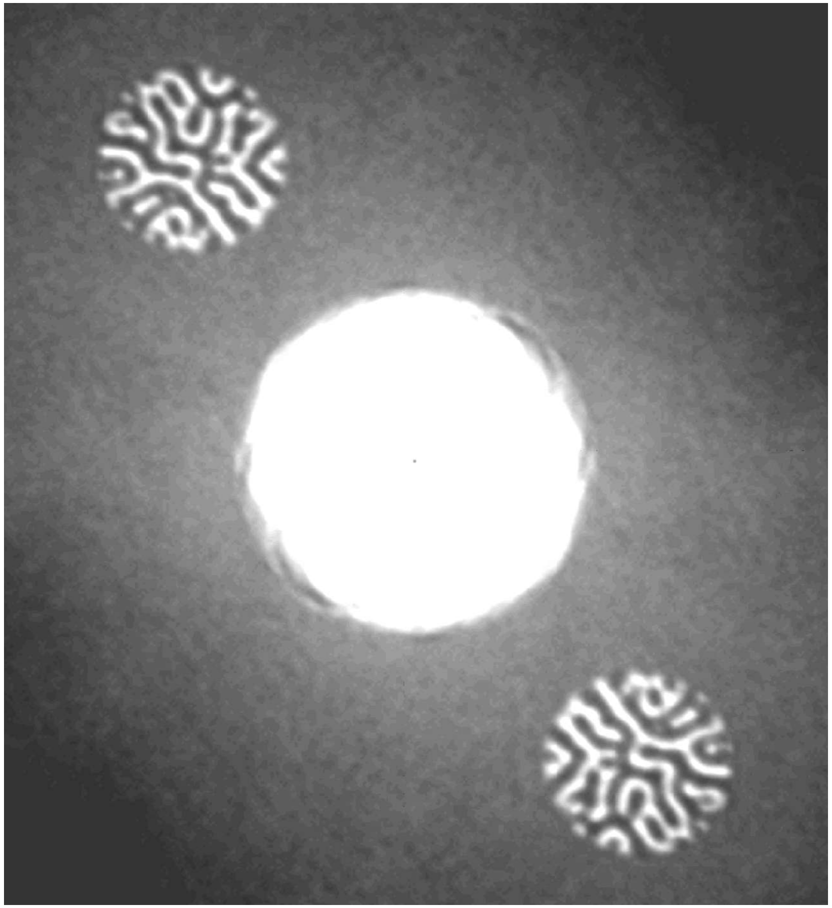
**Sample: O. Hellwig**

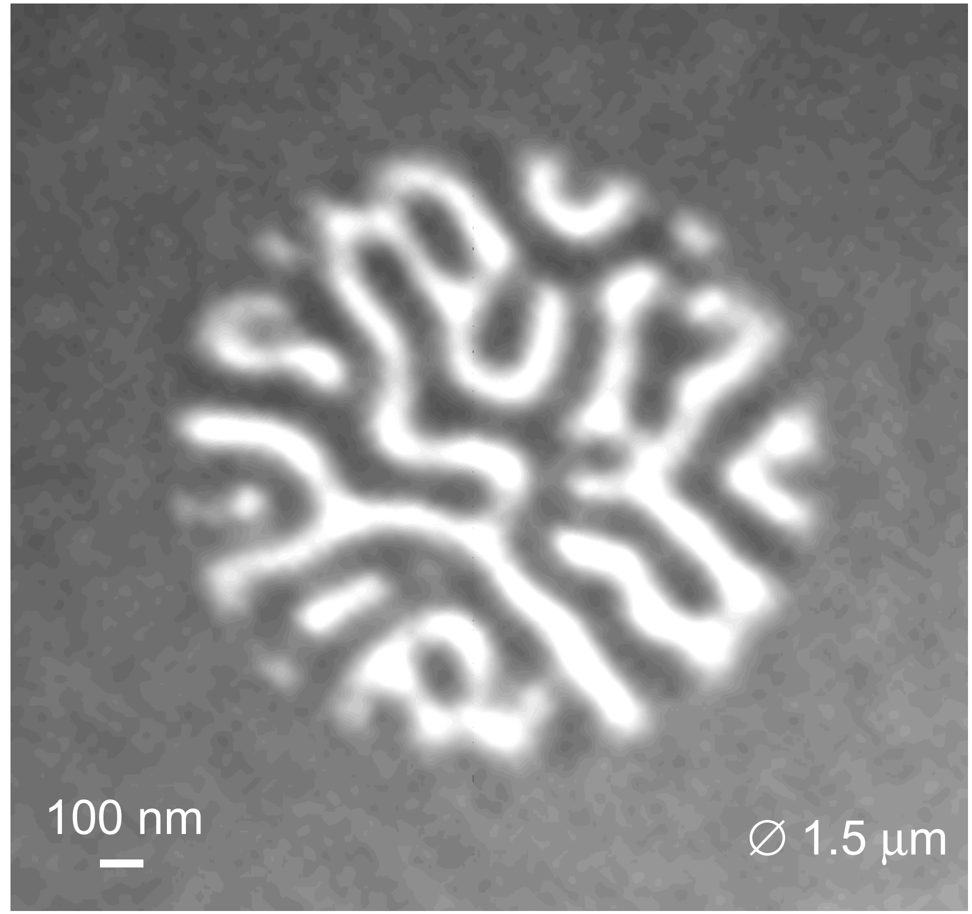
$SiN_x$ / Pt (24 nm) /
[Co (1.2 nm) / Pt (0.7 nm)]$_{50}$ /
Pt (1.5 nm)

perpendicular anisotropy

Contrast mechanism:
Circular magnetic dichroism

# Magnetic Image

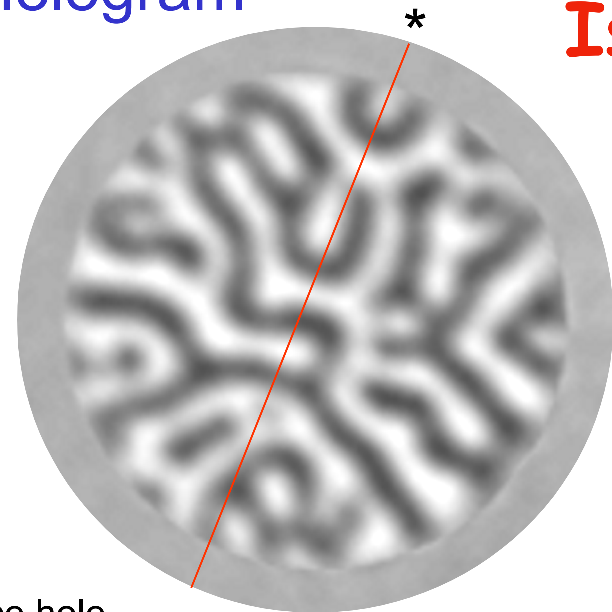




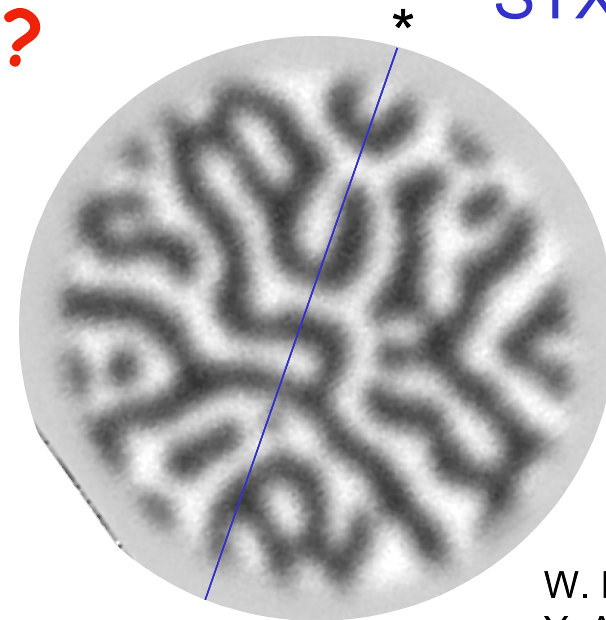
# Is it real?

# Is it good?

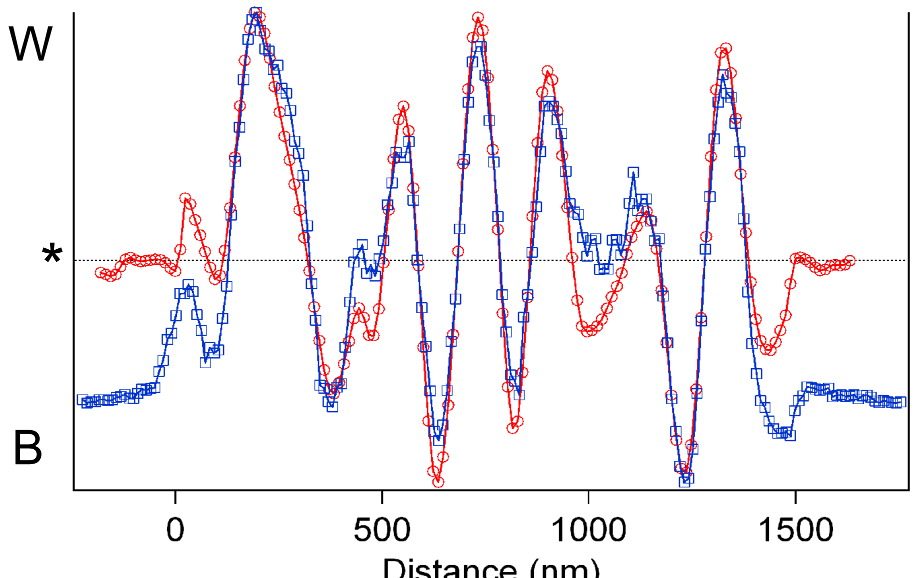
FT Hologram




STXM




Reference hole
∅ 100 nm

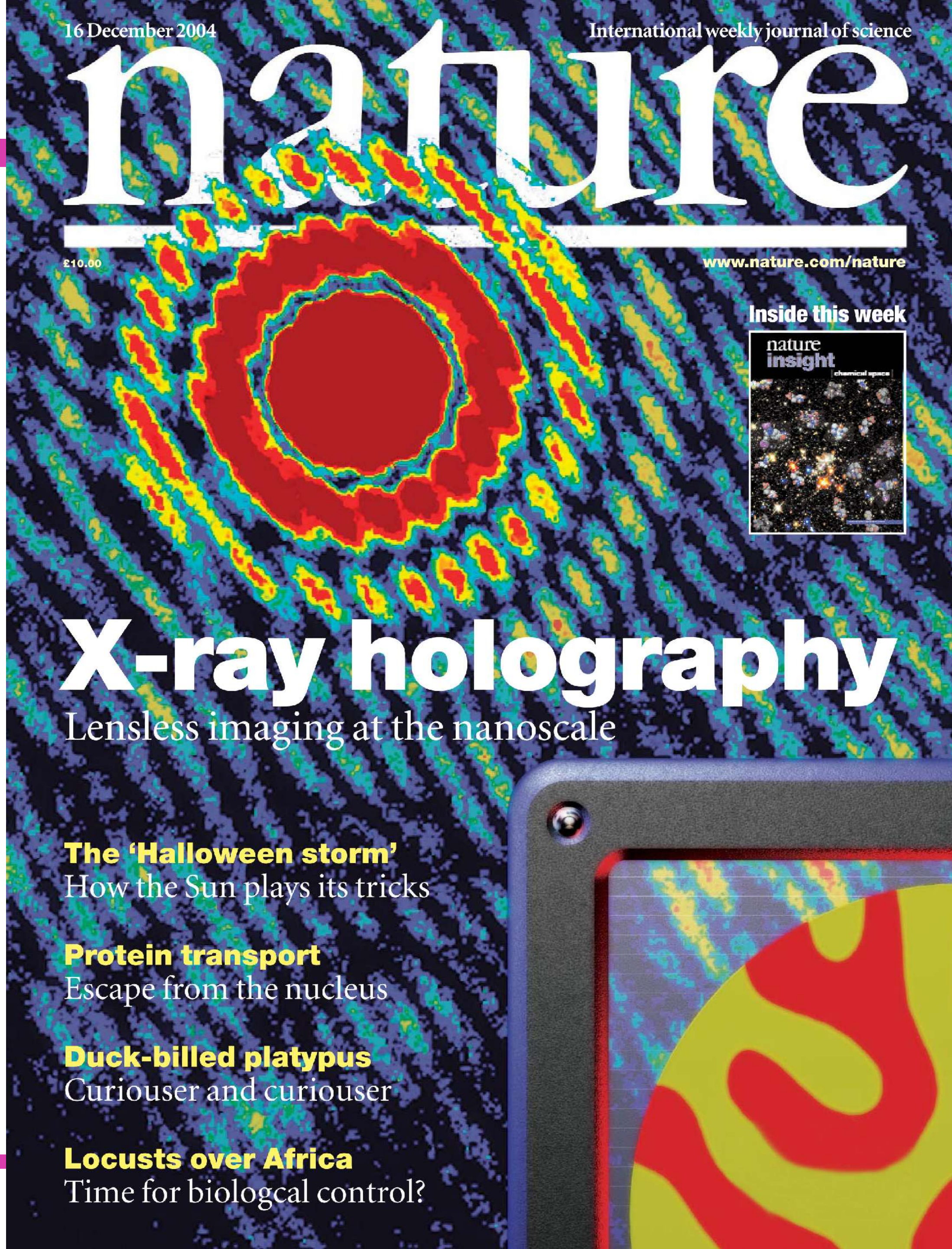



W. F. Schlotter
Y. Acremann

Resolution
30 - 40
nm

S. Eisebitt
J. Lüning
W.F. Schlotter
M. Lörgen
O. Hellwig
W. Eberhardt
J. Stöhr

NATURE **432**, 885 (2004)


16 December 2004
International weekly journal of science
nature
£10.00
www.nature.com/nature
Inside this week
nature insight
X-ray holography
Lensless imaging at the nanoscale
**The 'Halloween storm'**
How the Sun plays its tricks
**Protein transport**
Escape from the nucleus
**Duck-billed platypus**
Curiouser and curiouser
**Locusts over Africa**
Time for biologcal control?

BESSY

LELAND STANFORD JUNIOR UNIVERSITY
ORGANIZED 1891

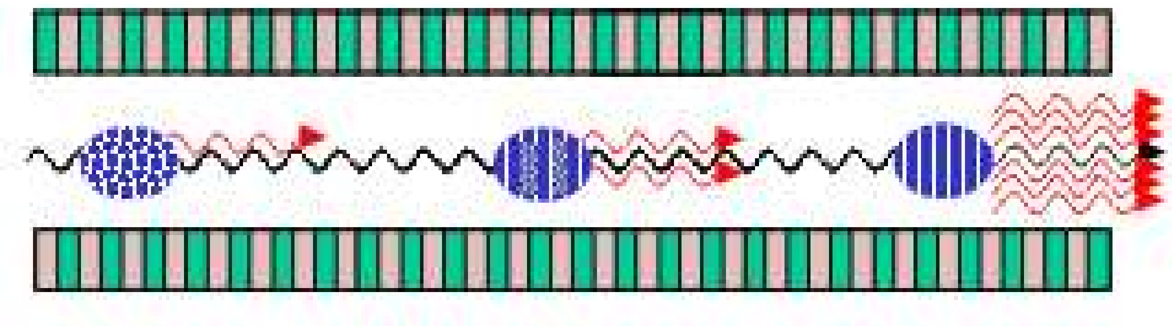
SSRL

IBM
Almaden Research Center

Leibniz Gemeinschaft

# Vision: Femtosecond Snapshots

Free Electron Laser



- coherent flux sufficient to image with a single pulse
- pulse duration 20 fs

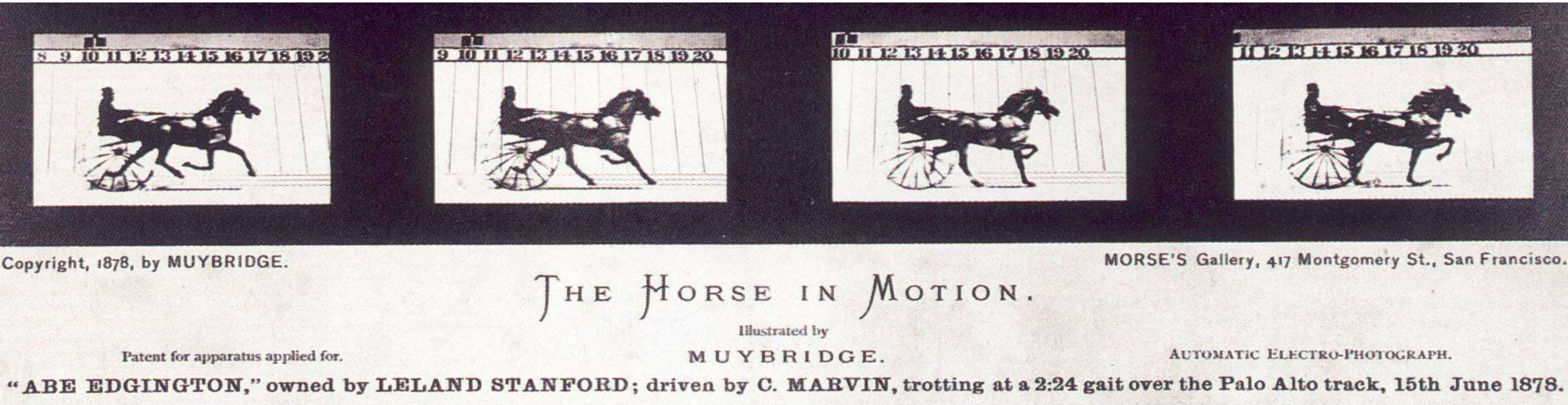
**Life Sciences**

**Magnetism**

**Materials Sciences**

**Process Monitoring**

- holography & oversampling phasing benefit from increased coherence



# Future: Patterned Magnetic Media

LETTERS

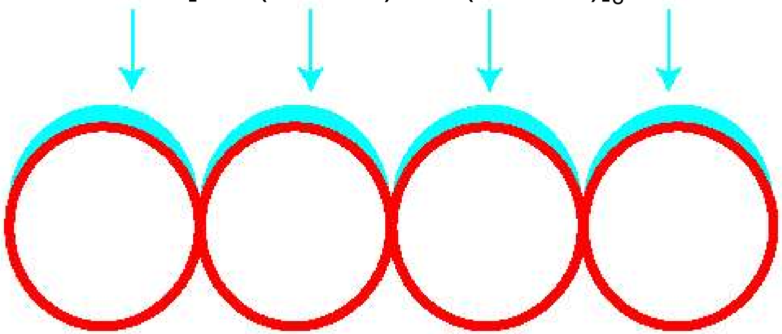
## Magnetic multilayers on nanospheres

MANFRED ALBRECHT[1]*, GUOHAN HU[2], ILDICO L. GUHR[1], TILL C. ULBRICH[1], JOHANNES BONEBERG[1], PAUL LEIDERER[1] AND GÜNTER SCHATZ[1]

Nature Materials **4**, 203 (2005)

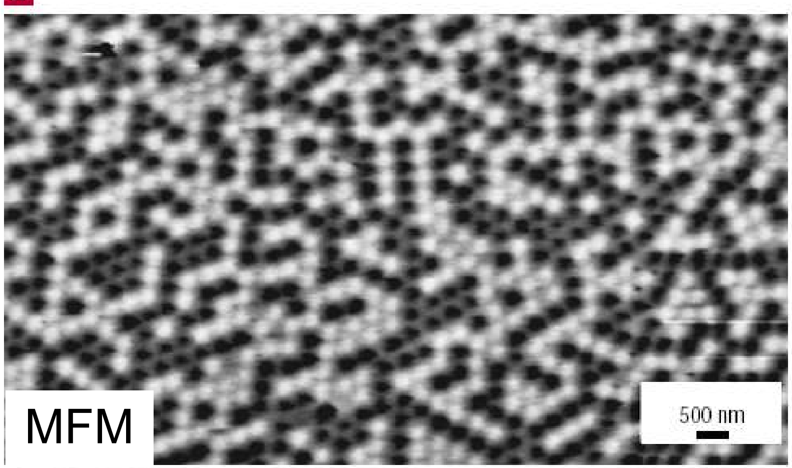
a

[Co (0.3 nm) Pd (0.8 nm)]$_8$

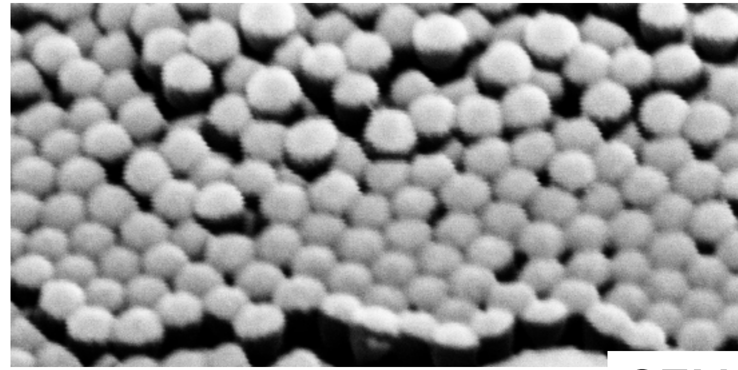


∅ 330 nm

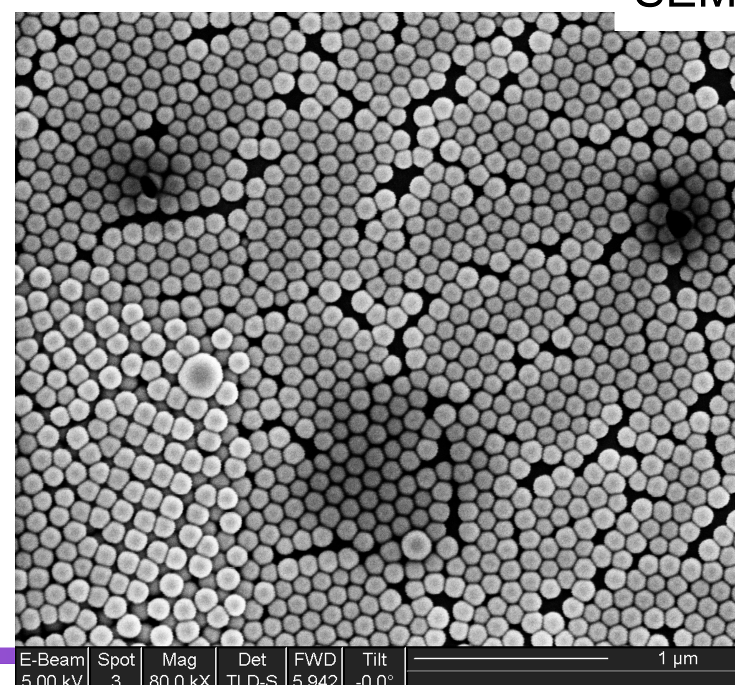
b




∅ 110 nm

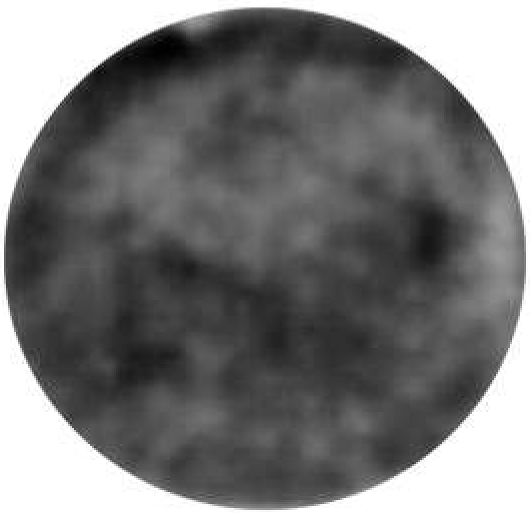


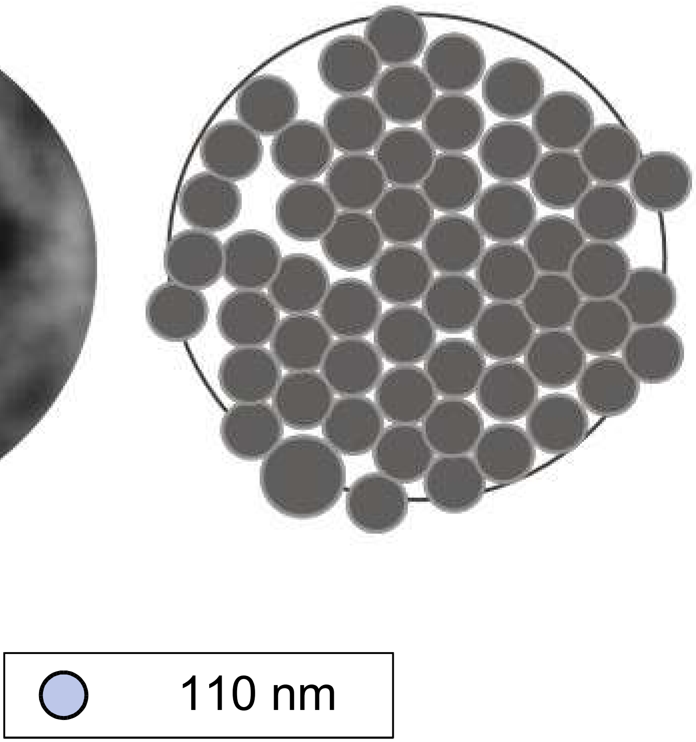
SEM

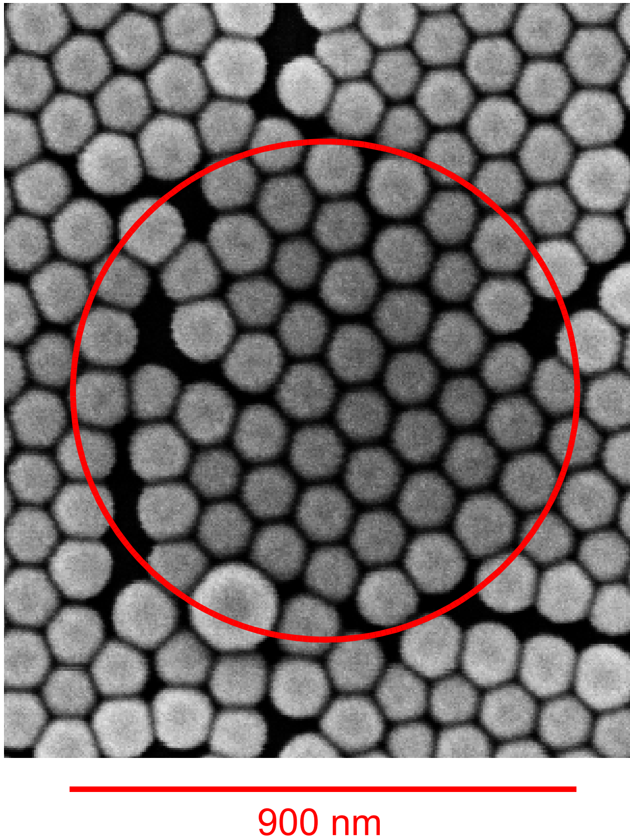


# Flipping the bits in an applied field







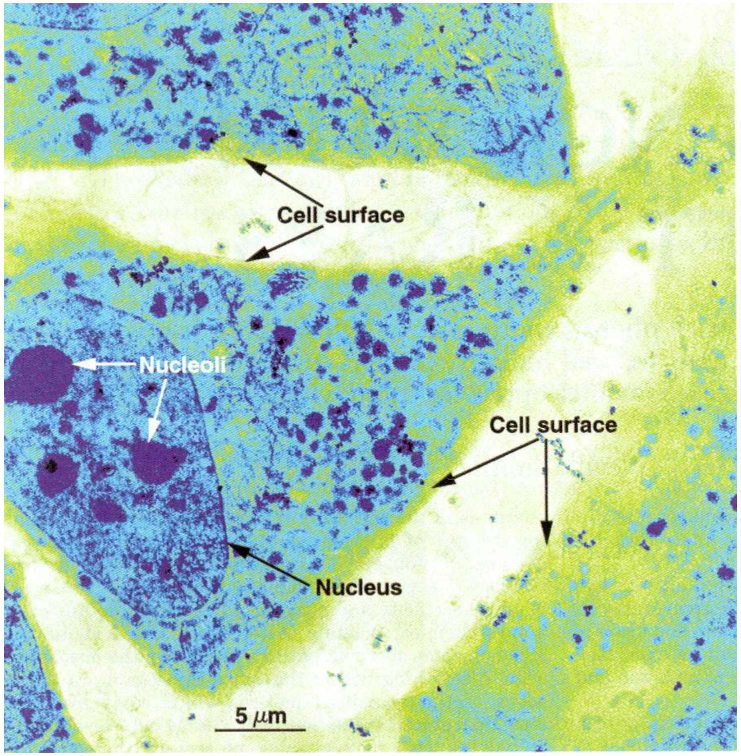




O. Hellwig, S. Eisebitt, W.F. Schlotter, J. Lüning (unpublished)

# Visions of SCIENCE

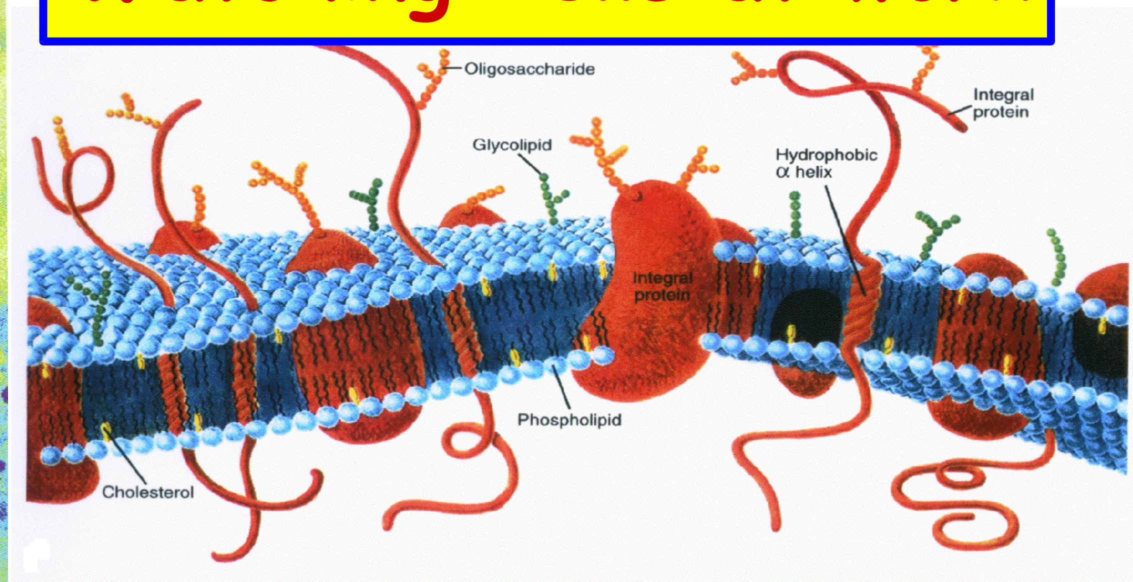
BESSY FEL

## Dynamics in Biological Systems (in Water)




Transmission micrograph of mouse 3T3 fibroblasts, taken in the ‚water window‘
(C. Larabell ALS Berkeley)

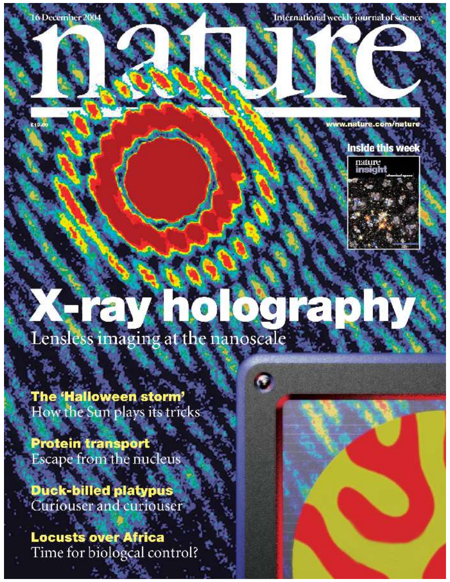
**Watching Cells at work**

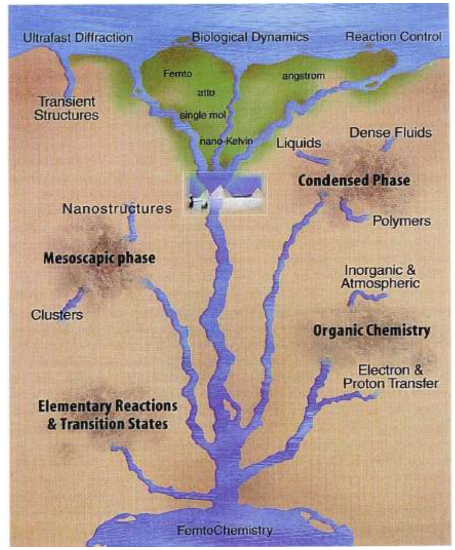


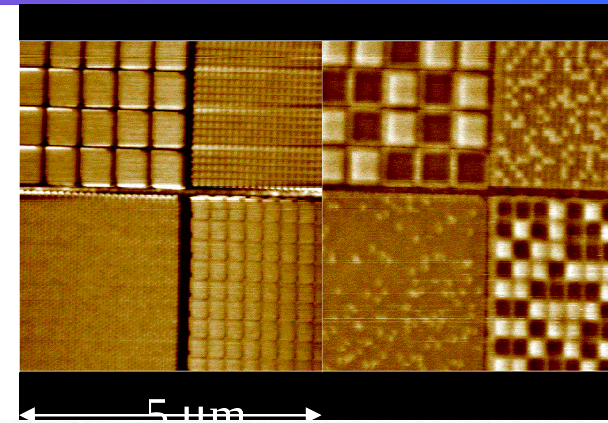

**Coherent imaging and time resolved (single pulse), chemical state sensitive microscopy of cells and membranes**

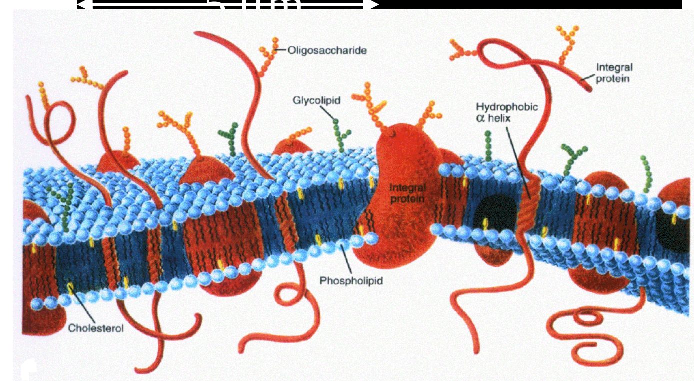
**Functional Cycles (photosynthesis, enzymatic reactions)**
**Functional Systems (ion channels, molecular motors, pumps)**

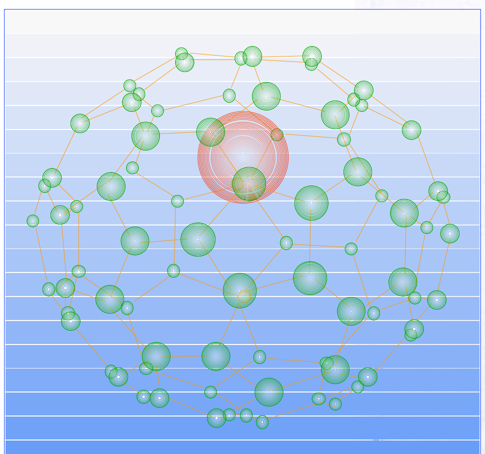
# Summary

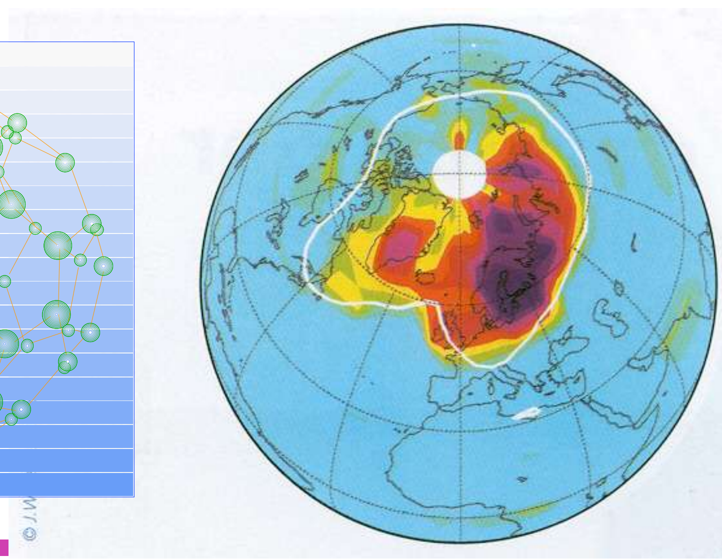

16 December 2004
nature
International weekly journal of science
www.nature.com/nature
Inside this week
X-ray holography
Lensless imaging at the nanoscale
The 'Halloween storm'
How the Sun plays its tricks
Protein transport
Escape from the nucleus
Duck-billed platypus
Curiouser and curiouser
Locusts over Africa
Time for biologcal control?

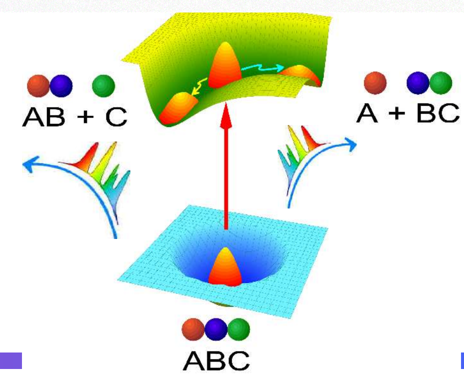

Ultrafast Diffraction
Biological Dynamics
Reaction Control
Femto
atto
angstrom
single mol
nano-Kelvin
Transient Structures
Liquids
Dense Fluids
Condensed Phase
Nanostructures
Polymers
Mesoscapic phase
Inorganic & Atmospheric
Clusters
Organic Chemistry
Electron & Proton Transfer
Elementary Reactions & Transition States
FemtoChemistry




Oligosaccharide
Integral protein
Glycolipid
Hydrophobic α helix
Integral protein
Phospholipid
Cholesterol

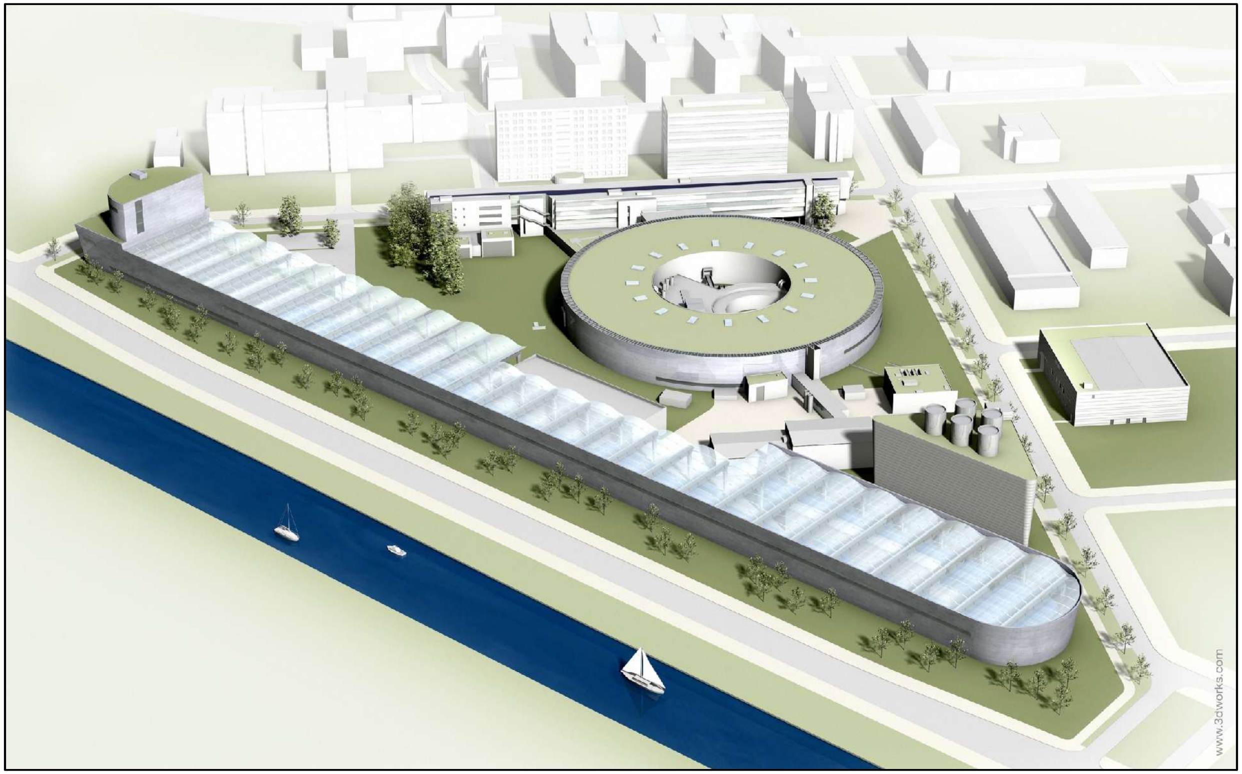





AB + C
A + BC
ABC

# The BESSY Soft X-Ray FEL

BESSY FEL



DESY

TSB TECHNOLOGIESTIFTUNG INNOVATIONSZENTRUM BERLIN

CORNELL

FZR

BROOKHAVEN NATIONAL LABORATORY

Technische Universität Dresden

MAX-PLANCK-GESELLSCHAFT

MPG Bauabteilung

Leibniz Gemeinschaft

# The BESSY Soft X-Ray FEL

BESSY FEL

## Design Goals:

**Unprecedented Spectral, Spatial, and Temporal Resolution for Soft X-Ray Science**

Photon energy range : 24 eV up to 1 keV (51 nm > $\lambda$ > 1,2 nm)

Peak power

Pulse length ... fs)

Synchroniz...

**Cascaded HGHG design**

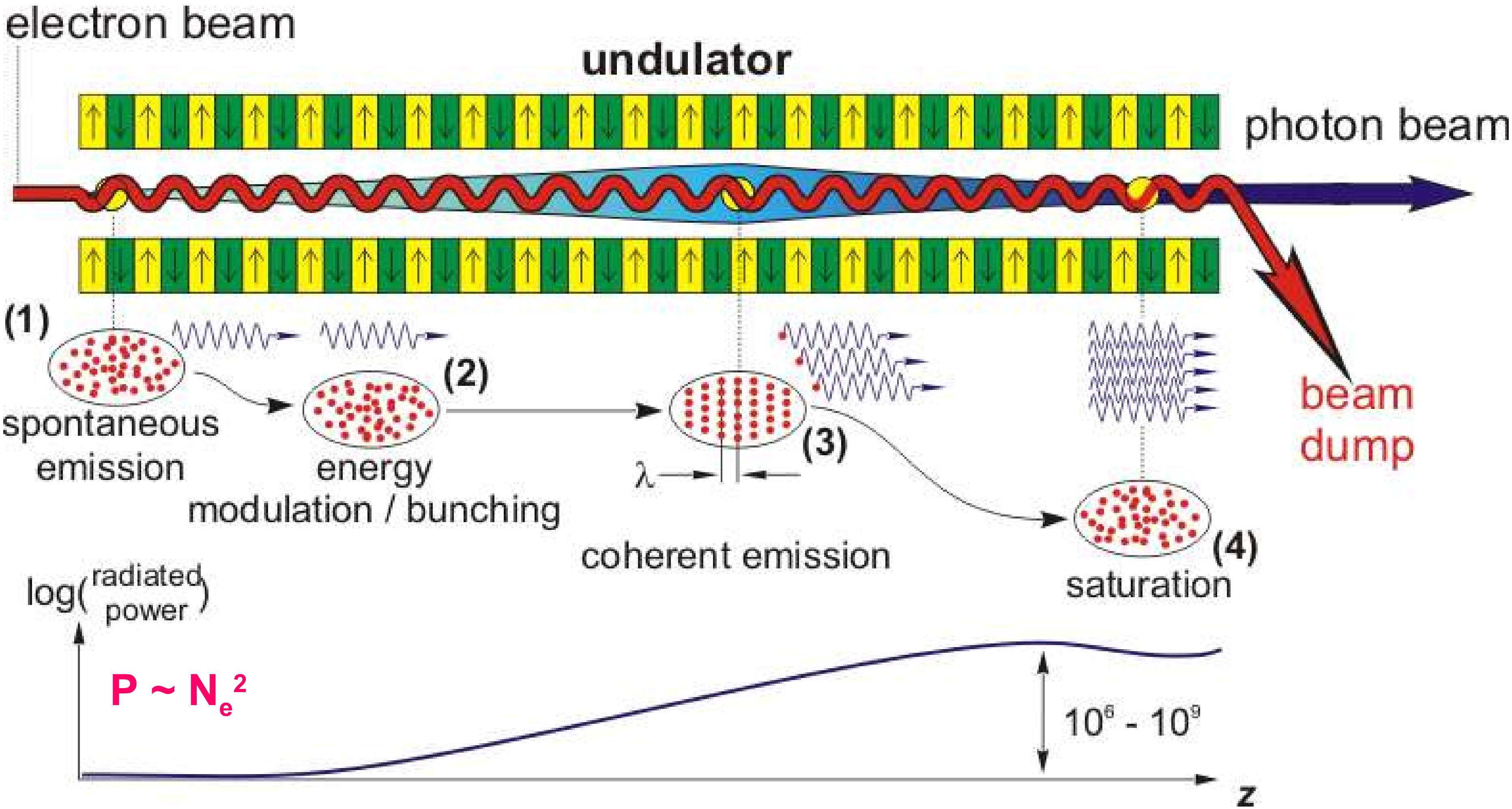
**offering control and reproducibility**

**from pulse to pulse**

Pulse sequence: 1 kHz each FEL , SC injector 25 kHz

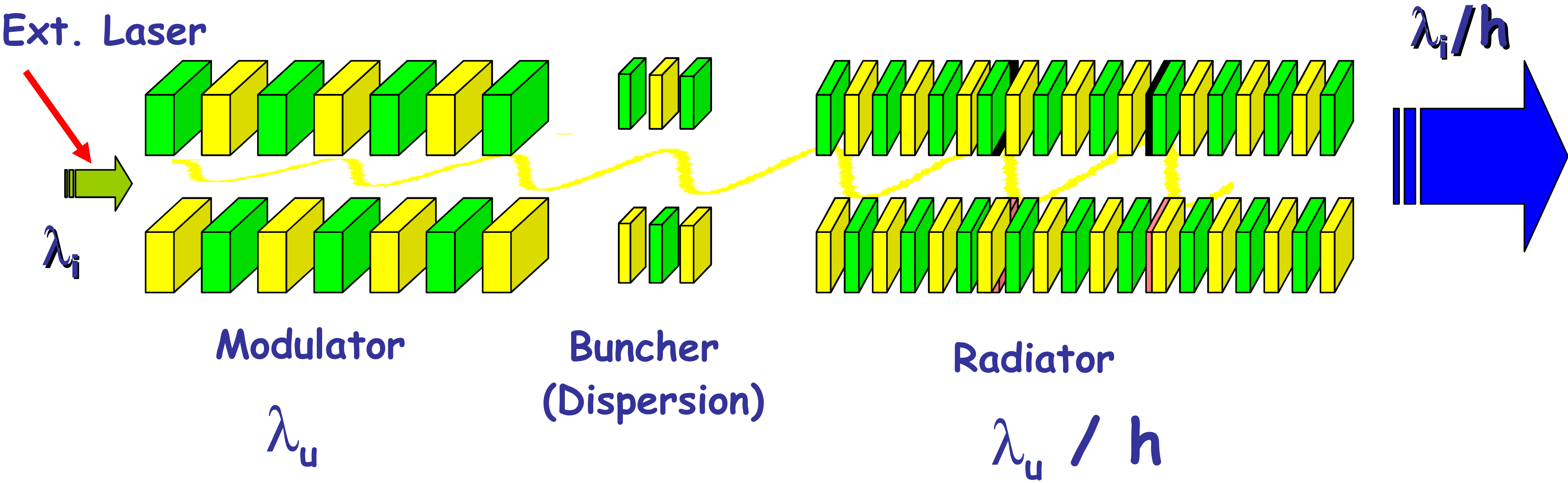
Photon polarisation: variable, selectable

# SELF AMPLIFIED SPONTANEOUS EMISSION „SASE“




Kondratenko, Saldin 1980

# High Gain Harmonic Generation (HGHG)




- Longitudinal coherence
- Ultra-fast pulses < 20 fs
- Reproducible pulse shape
- Non „chaotic" light
- Intrinsic synchronization
- Attosecond HHG option

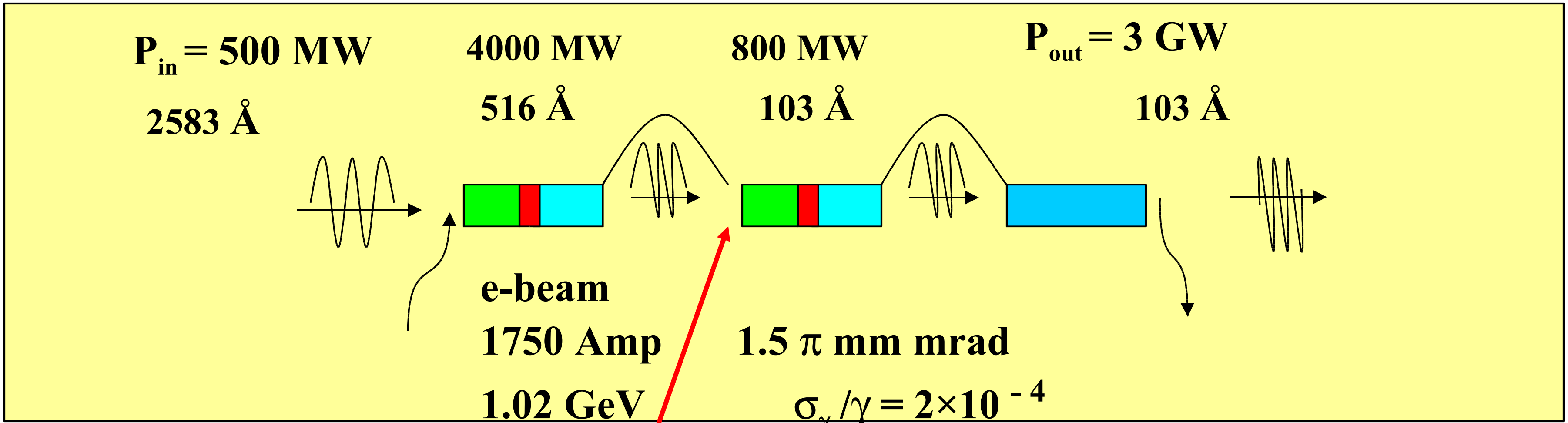
$$\lambda_w = \frac{\lambda_u}{2\gamma^2}\left(1 + \frac{K^2}{2} + \gamma^2\Theta_0^2\right)$$

**L.H. Yu, et. al. BNL**

**FEL Prize 2003**

# The BESSY Low-Energy HGHG FEL

BESSY FEL




‚attosecond' HHG Option (51.6 nm)

| | | | |
|---|---|---|---|
| | [illegible] | [illegible] | [illegible] |
| [illegible] | [illegible] | [illegible] | [illegible] |
| [illegible] | [illegible] | [illegible] | [illegible] |
| | | | |
| [illegible] | [illegible] | [illegible] | [illegible] |

# HGHG vs. SASE at $\lambda$ = 10.3 nm

BESSY FEL

**Controlled, synchronized, reproducible pulse shape**

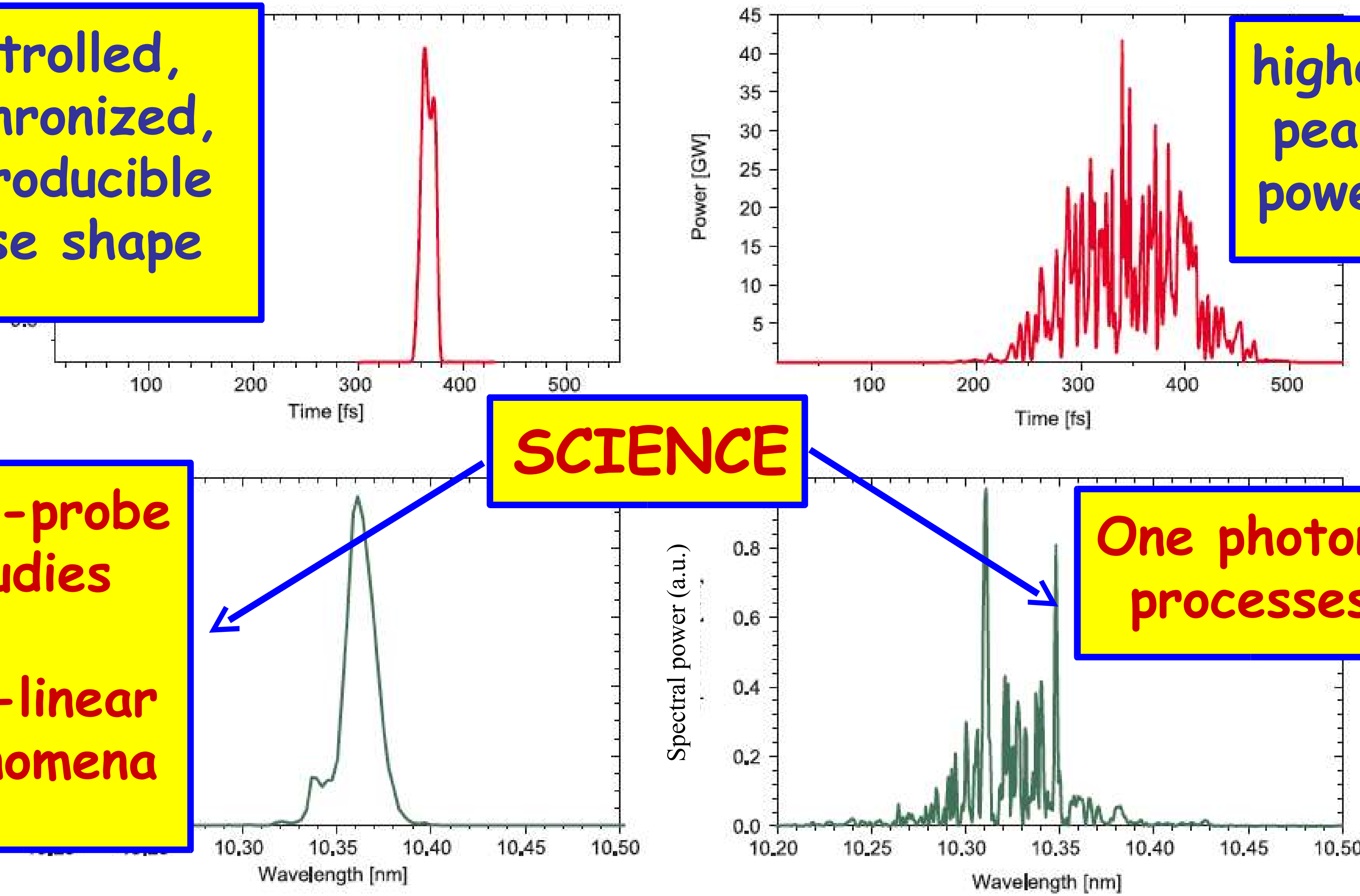
**higher peak power**

**SCIENCE**

**Pump-probe studies**

**Non-linear phenomena**

**One photon processes**



# SASE vs. HGHG Comparison

- SASE offers higher peak brilliance by about one order of magnitude
- HGHG is limited to energies < 1keV

but

- HGHG offers a controlled and reproducible temporal lineshape
- HGHG is intrinsically synchronized on the fs timescale

- **SASE is well suited for one photon processes**
- **HGHG is needed for pump-probe and multi-(n)-photon processes**

**$S \sim I^n$**

# Coworkers

BESSY FEL

| Clusters | Magnetism | Holografy |
| --- | --- | --- |
| M. Neeb | H. Dürr | S. Eisebitt |
| J. Stanzel | C. Stamm | M. Lörgen |
| F. Burmeister | C. Lupulescou | O. Hellwig |
| N. Pontius | K. Holldack | |
| G. Lüttgens | T. Kachel | J. Lüning } SSRL |
| | S. Khan | W.F. Schlotter } SSRL |
| P.S. Bechthold(IFF) | R. Mitzner | J. Stöhr } SSRL |
| | T. Quast | |