Trieste, 05-12-01

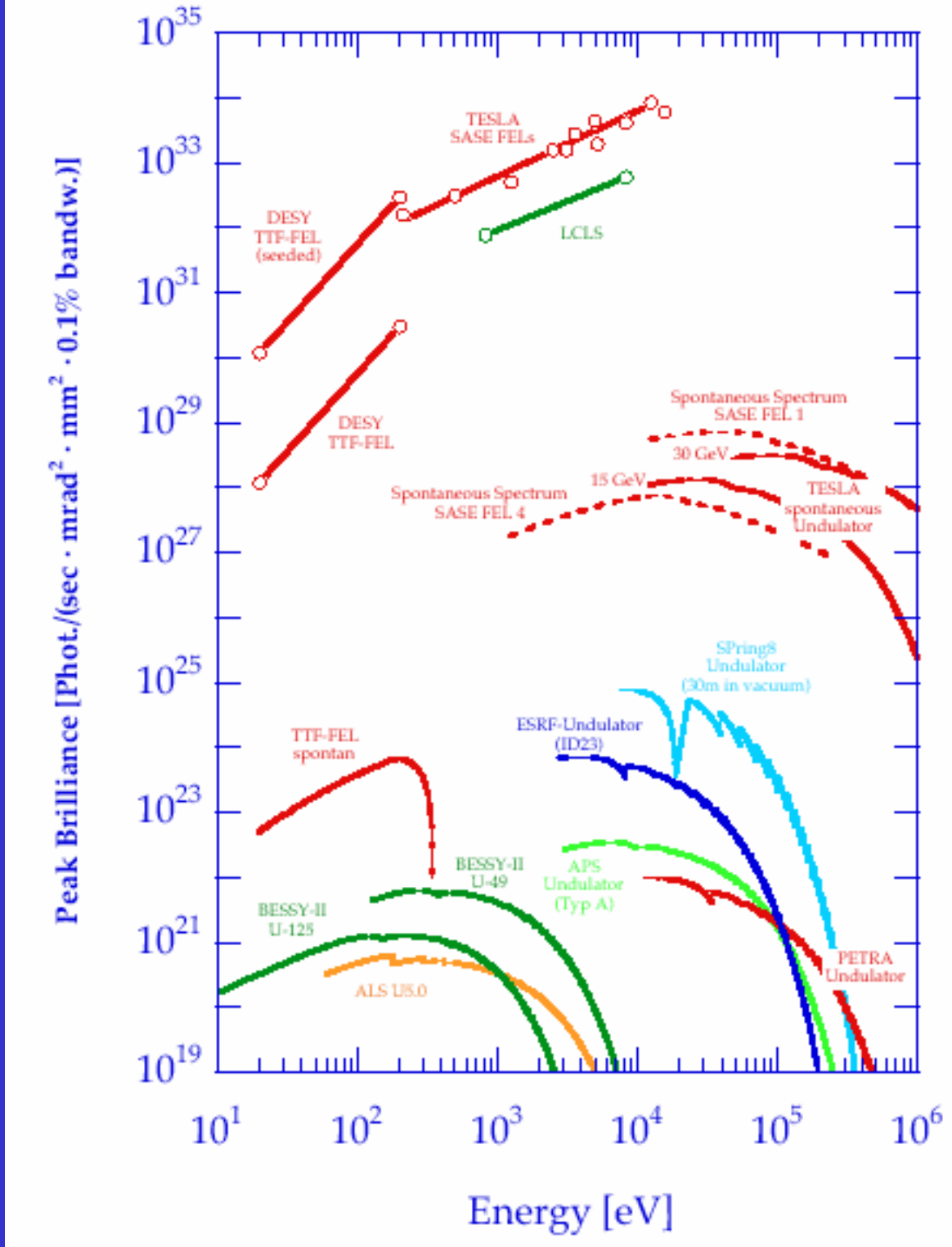
P. Perfetti

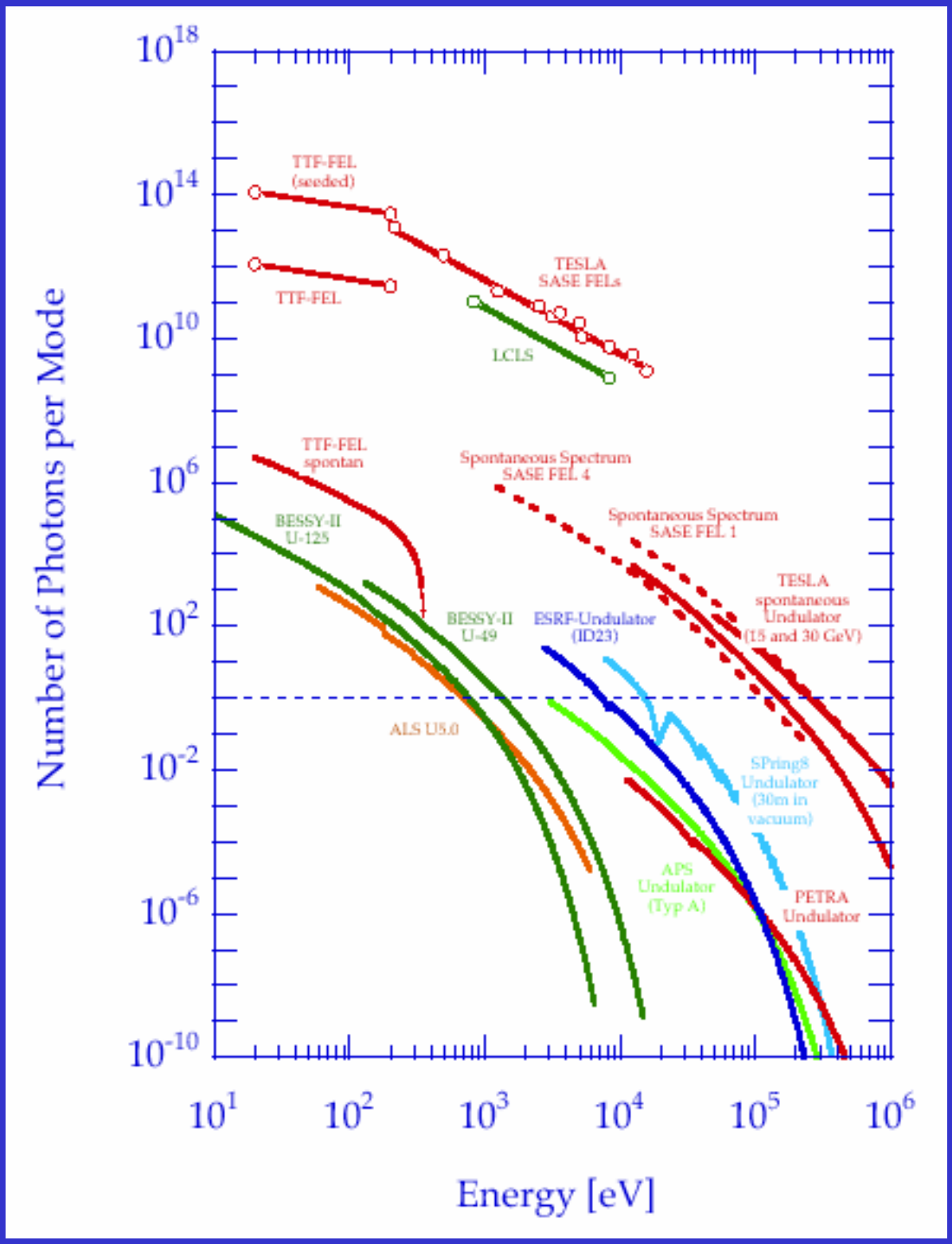
SCIENTIFIC OPPORTUNITIES WITH AN ULTRABRIGHT PULSED LASER SOURCE

OUTLINE

- FEL/SASE CHARACTERISTICS
- PERSPECTIVES IN THE SOFT X-RAY
- PERSPECTIVES IN THE HARD X-RAY

Istituto di Struttura della Materia del CNR
via del Fosso del Cavaliere 100, 001□□33 ROMA


Peak Brilliance [Phot./(sec · mrad² · mm² · 0.1% bandw.)]
10^35
10^33
10^31
10^29
10^27
10^25
10^23
10^21
10^19
10^1
10^2
10^3
10^4
10^5
10^6
Energy [eV]
TESLA
SASE FELs
LCLS
DESY
TTF-FEL
(seeded)
DESY
TTF-FEL
Spontaneous Spectrum
SASE FEL 1
30 GeV
15 GeV
Spontaneous Spectrum
SASE FEL 4
TESLA
spontaneous
Undulator
SPring8
Undulator
(30m in vacuum)
ESRF-Undulator
(ID23)
TTF-FEL
spontan
BESSY-II
U-49
APS
Undulator
(Typ A)
BESSY-II
U-125
ALS U5.0
PETRA
Undulator


Number of Photons per Mode
Energy [eV]
10^{18}
10^{14}
10^{10}
10^{6}
10^{2}
10^{-2}
10^{-6}
10^{-10}
10^{1}
10^{2}
10^{3}
10^{4}
10^{5}
10^{6}
TTF-FEL (seeded)
TTF-FEL
TESLA SASE FELs
LCLS
TTF-FEL spontan
Spontaneous Spectrum SASE FEL 4
Spontaneous Spectrum SASE FEL 1
BESSY-II U-125
BESSY-II U-49
ESRF-Undulator (ID23)
TESLA spontaneous Undulator (15 and 30 GeV)
ALS U5.0
SPring8 Undulator (30m in vacuum)
APS Undulator (Typ A)
PETRA Undulator

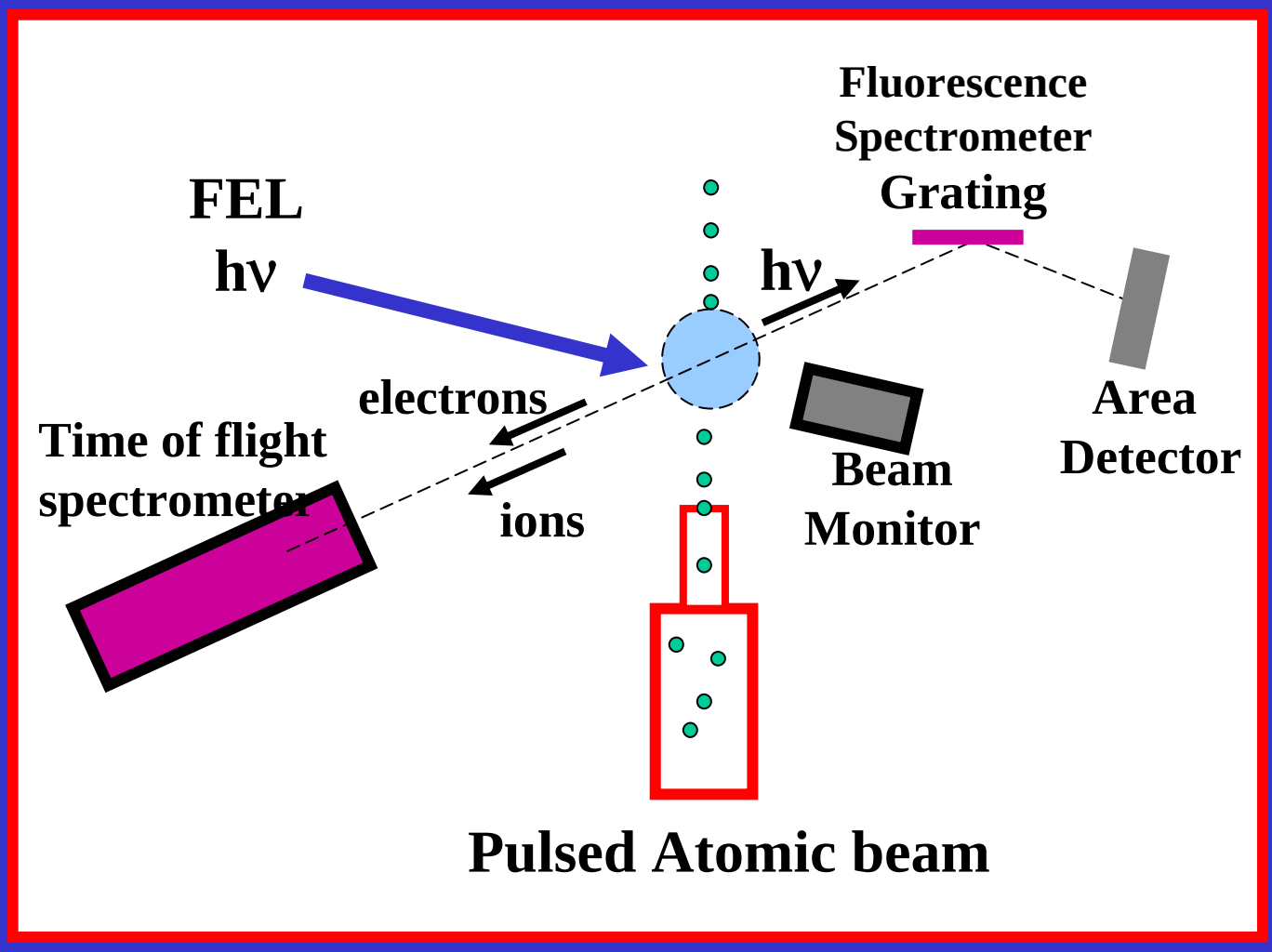
EXPERIMENTS ON IV GENERATION SR SOURCES MUST BE TAILORED ON THE MAIN CHARACTERISTICS OF THESE SOURCES

- **BRILLIANCE (10 orders of magnitude higher than 3rd generation SR sources)**
- **COMPLETE TRANSVERSE COHERENCE**
- **PULSE TIME STRUCTURE <100fs**

POSSIBLE APPLICATIONS IN THE SOFT X-RAY

ATOMIC AND MOLECULAR PHYSICS

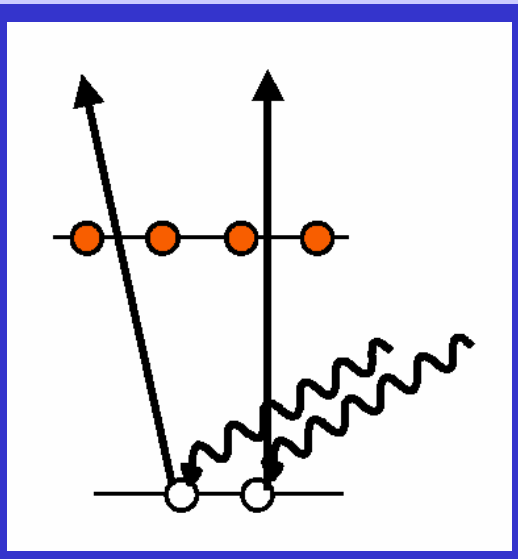
- **The interaction of a high intensity photon beam with isolated atoms and clusters is a first step in:**
- **Understanding the interaction with condensed matter**
- **To tailor new experiments**
- **To choose the right materials for the optical elements (mirrors, monochromators, slits, …….)**



set-up for experiments on isolated atoms

- The interaction of the FEL photons with the atomic beam produces ions, electrons and fluorescence photons
- The simultaneous detection of the different outgoing species will give detailed information on the Auger and fluorescence cascade resulting in highly charged final ions

MULTIPLE CORE-HOLE FORMATION



- **ELECTRON CORRELATION IS RESPONSIBLE OF MULTIPLE CORE-HOLE FORMATION**

- **FOR DEEP CORE- LEVELS THIS IS A LOW CROSS SECTION PROCESS**

- **NEED FOR THE SHORT AND INTENSE FEL PULSE RADIATION TO CREATE A SUFFICENTLY LARGE NUMBER OF PROCESSES TO BE DETECTED**

- **NEED FOR AN X-FEL**
- **NONLINEAR PHOTON-ATOM INTERACTIONS**
- **FOR INNER SHELLS: THE TWO PHOTON PHOTOIONIZATION CROSS SECTIONS ARE EXCEEDINGLY SMALL**

2-photon double ionization

$$h\nu + He \rightarrow He^{2+} + 2e$$

correlated electron-electron dynamics

[Review : Briggs and Schmidt J. Phys.B33 (2000)R1; King and Avaldi J. Phys.B33 (2000)R215]

$$nh\nu' + He \rightarrow He^{2+} + 2e$$

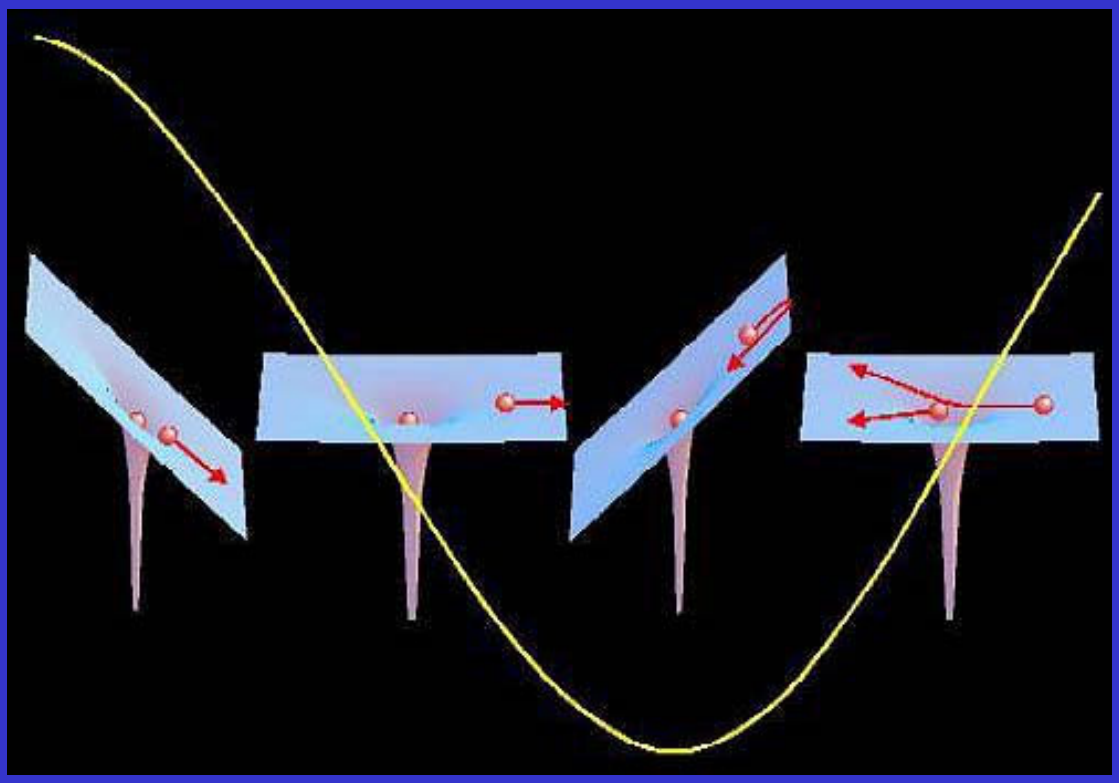
correlated emission /sequential process ?

[Frankfurt+Marburg PRL 84 (2000)443; Freiburg+Berlin PRL 84 (2000)447]

Ti-sapphire laser λ=800 nm, E=1.54 eV

Δt=220 $\rightarrow$30 fs

I= 10-100 Twatt/cm^2



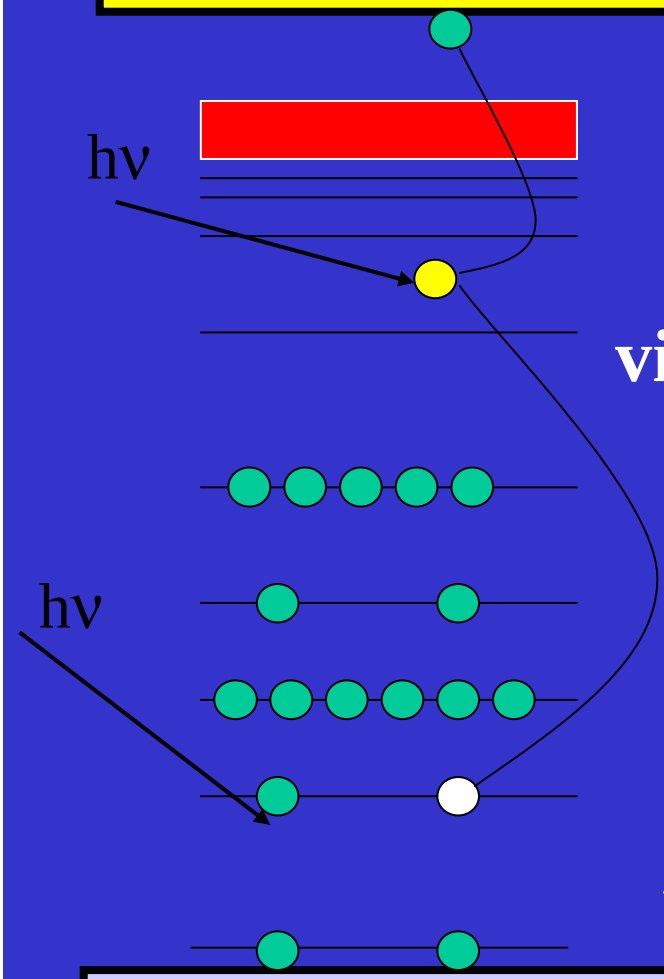
Mechanism:rescattering

X-FEL :

2-photon process

non-perturbative regime ($E_p \propto I/\omega^{2)}$

2 photon inner-shell ionization and Auger decay



virtual state $\tau \cong 10^{-14}$ s

$\Gamma \cong 100$ meV

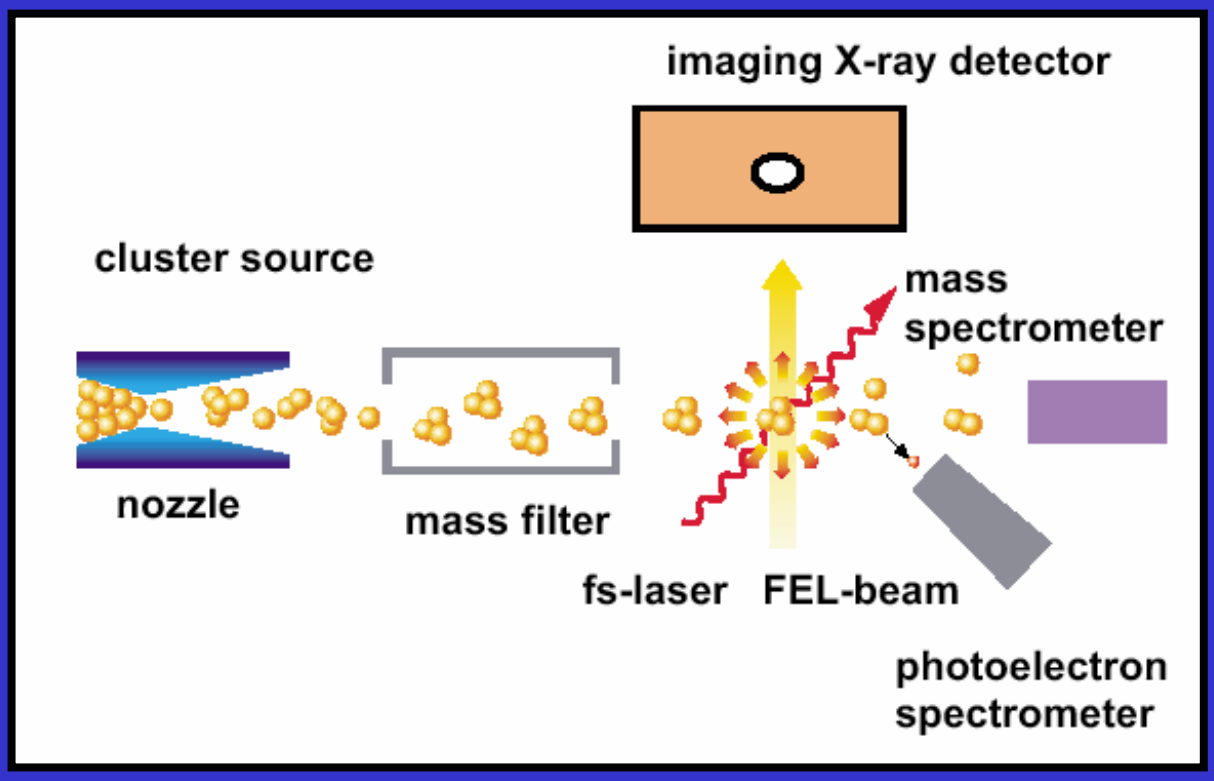
$\tau_{Auger} \cong 10^{-14}$ s

- What about the Auger decay ?
- What about the interaction between the Auger electron and the photoelectron ?

[Rioual et al. PRA 61 (2000)044702, PRL 86 (2001) to be published]

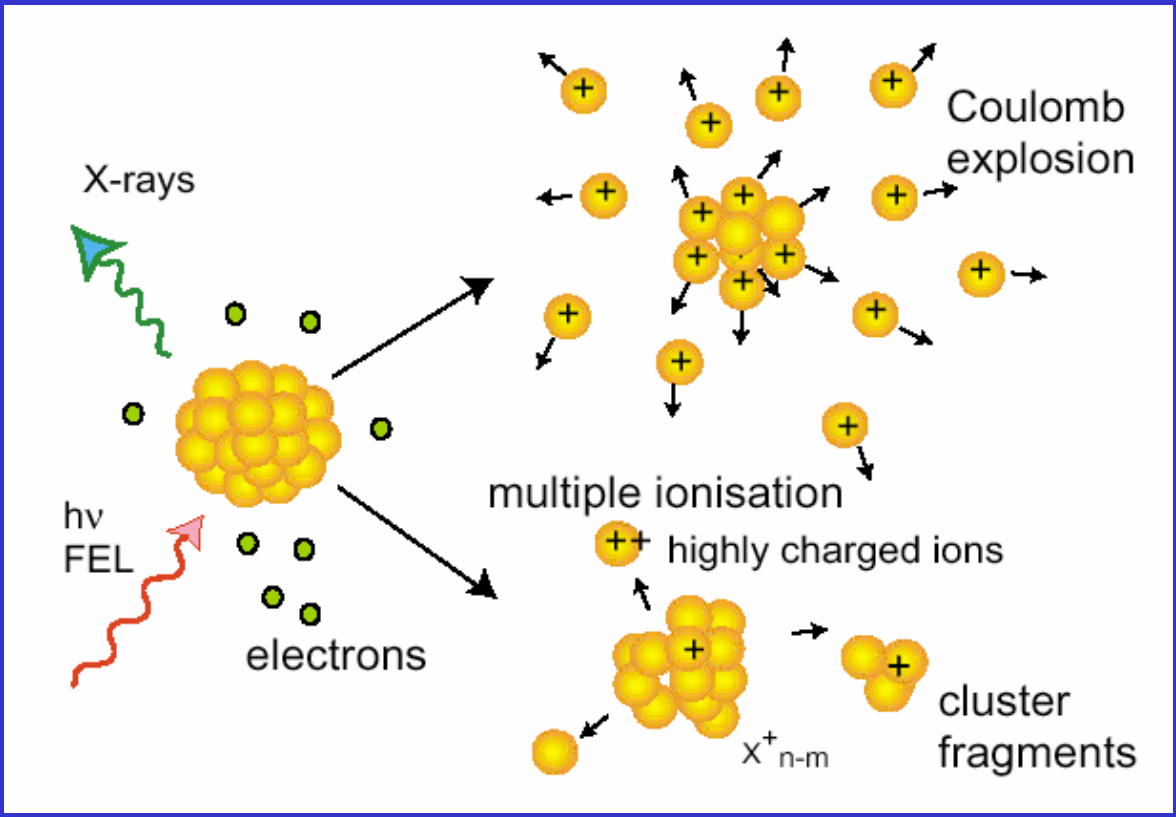
- Is it possible to observe laser assisted Auger processes ?

CLUSTER PHYSICS



- PHOTON-CLUSTER INTERACTION CAN CAUSE:
  - PHOTO-FRAGMENTATION
  - HIGHLY CHARGED IONS
  - COULOMB EXPLOSION

- DIRECT STUDIES OF FEL-CLUSTER INTERACTION
- PUMP-PROBE EXPERIMENTS USING AN EXTERNAL fs-LASER



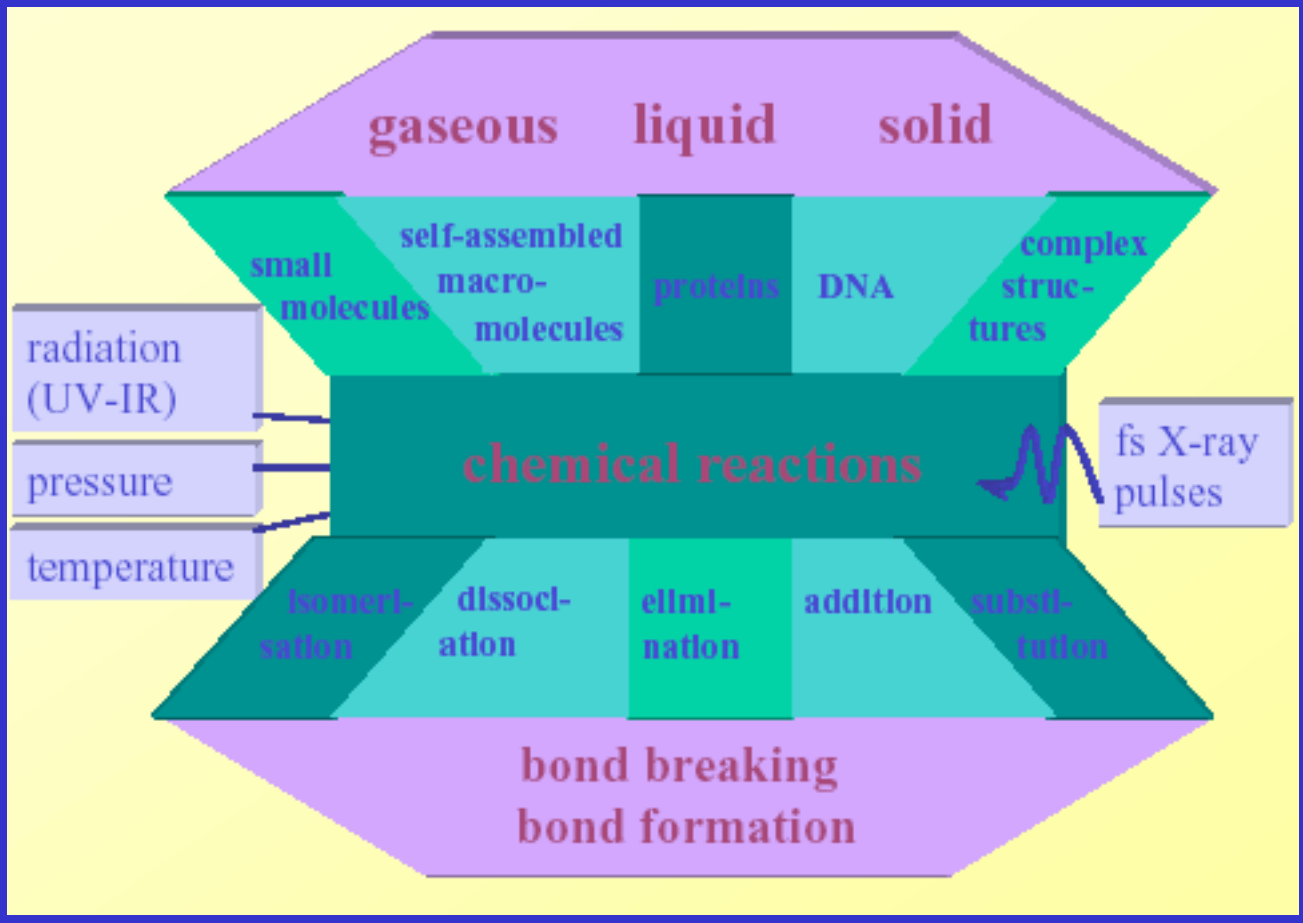
- **FOR A FOCUSED BEAM, DEPENDING ON THE ATOMIC SPECIES CROSS SECTIONS, ALL THE ATOMS IN THE CLUSTER COULD BE IONIZED CREATING A BALL OF CHARGE GIVING RISE TO A**

COULOMB EXPLOSION(C.E.)

- **C.E. ENERGIES AND CHARGE STATES ARE MEASURED BY OBSERVING THE EXPLODING FRAGMENTS**
- **THESE EFFECTS WILL GIVE INSIGHT INTO MATERIAL DAMMAGE INDUCED BY THE FEL RADIATION**

- **ELECTRONIC STRUCTURE OF OCCUPIED AND UNOCCUPIED LEVELS:**
  - **PHOTOELECTRON SPECTROSCOPY (UPS,XPS)**
  - **ABSORPTION SPECTROSCOPY (NEXAFS)**
- **GEOMETRICAL STRUCTURE:**
  - **(XANES, EXAFS)**
- **DYNAMICS:**
  - **VIBRATION**
  - **DISSOCIATION**
  - **COULOMB EXPLOSION**
  - **PUMP-PROBE EXPERIMENTS**
- **CHEMICAL, ELECTRONIC AND DYNAMIC PROPERTIES**
  - **CLUSTER ON SURFACES**
  - **CLUSTER MATTER**

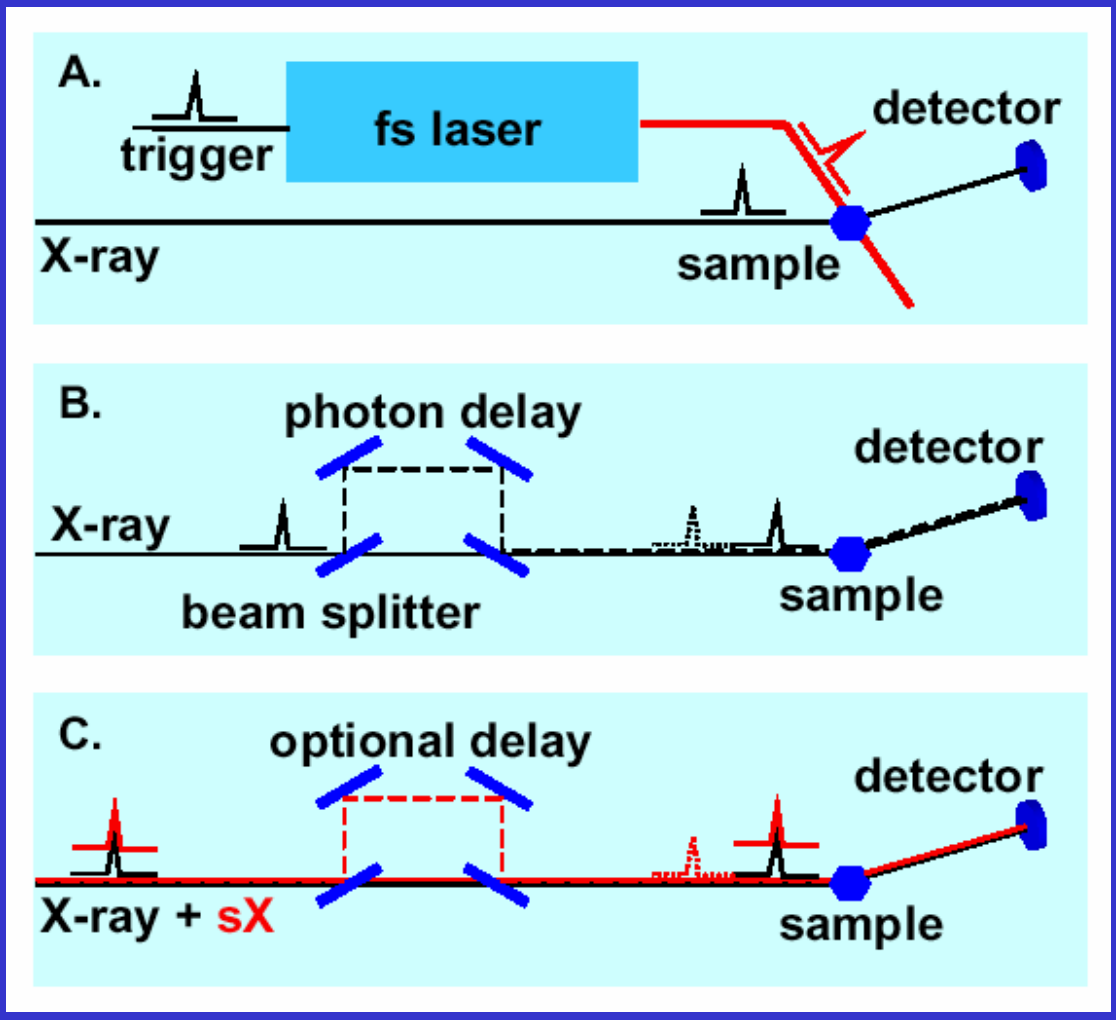
CHEMISTRY



- **THE MOST EXCITING EXPERIMENTS ARE THOSE BASED ON THE TIME STRUCTURE OF THE FEL PULSE (<100 fs) AND WE ENTER IN THE DOMAIN OF FEMTOCHEMISTRY**

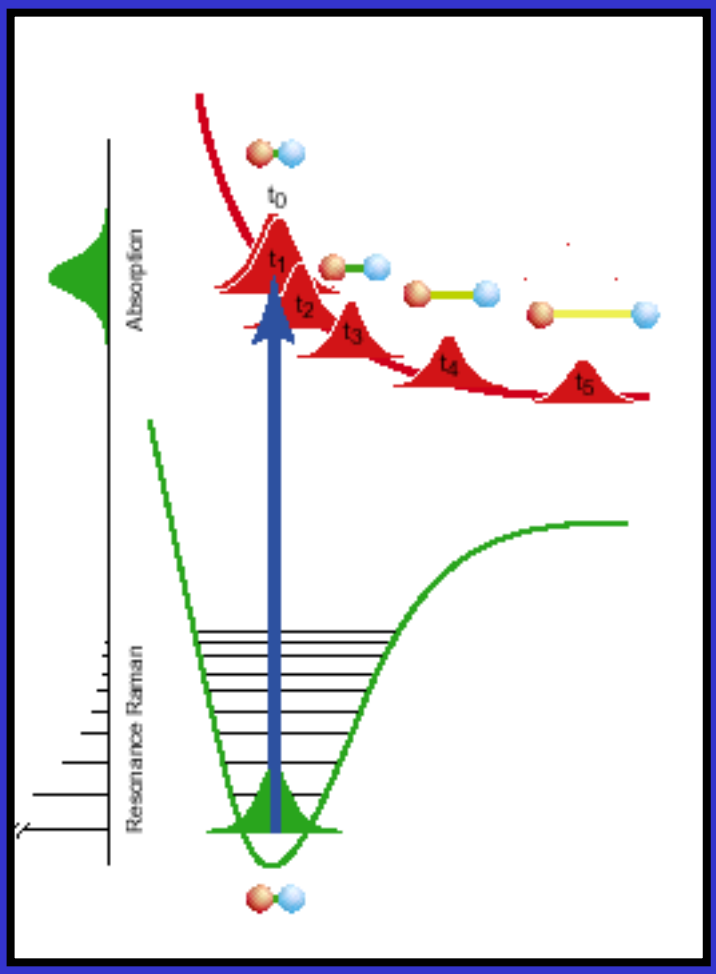
- **DYNAMICS OF CHEMICAL REACTION**
  - **MOLECULAR BOND LENGTH**
  - **ELECTRONIC STRUCTURE**
  - **MOLECULAR VIBRATION**
  - **POSITION IN SPACE**

- **A TYPICAL EXPERIMENT IN THIS FIELD IS A PUMP-PROBE EXPERIMENT**
- **PUMP: A SHORT LASER PULSE (OR THE FEL/SPONTANEOUS EMISSION) DRIVE THE EXCITATION MECHANISM**
- **THE RETARDED FEL-PULSE ALLOW THE DEFINITION OF THE TIME-EVOLUTION OF THE SYSTEM**



- **THE TIME SCALE OF A CHEMICAL REACTIONS ARE OFTEN IN THE fs SCALE**
- **E.g. THE UV PHOTODISSOCIATION OF A H_2O MOLECULE IN H+OH OCCURS IN ≅ 10 fs**
- **THE BEST SYNCHROTRON SOURCES ALLOW STUDIES IN A FEW TENS OF PICOSECONDS**
- **1999: NOBEL PRIZE IN CHEMISTRY TO AHMED ZEWAIL IN THE AREA OF ULTRAFAST SPECTROSCOPY**
- **THE SOFT-X-RAY FEL WILL EXTEND THESE STUDIES TO HIGH ENERGIES AND AND CAN USE EXPERIMENTAL METHODS TO INVESTIGATE THE THE STRUCTURAL PROPERTIES (TIME DEPENTENT XANES/ EXAFS)**

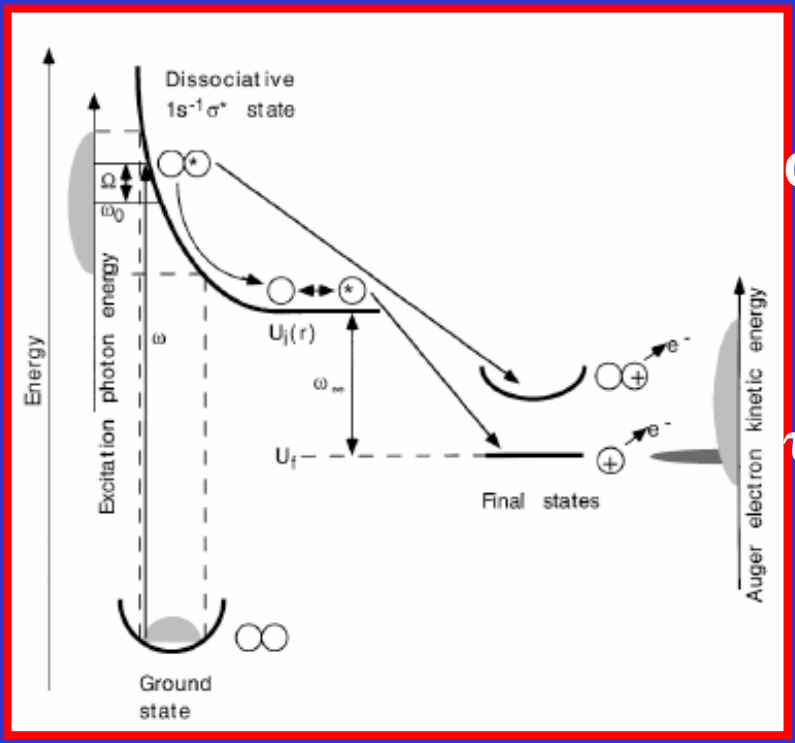
EXAMPLE OF DIATOMIC PHOTODISSOCIATION:



THE RED WAVE PACKET IS EXCITED TO A REPULSIVE STATE . THE WAVEFUNCTION OFTEN REMAIN LOCALIZED AND THE BEST WAY TO KNOW THE BOND LENGTH IS TO MEASURE IT DIRECTLY

TIME DEPENDENT XANES, EXAFS, RAMAN XAS

Doppler Splitting of In-Flight Auger Decay of Dissociating Oxygen Molecules:



$O_2 + h\nu = (O_2)^* = O^*O + OO$

$O_2^+ + e^-$ $\quad$ $O^+ + e^-$

$\tau_{Auger} = 3fs$ $\quad$ $\tau_{diss.} = 7fs$

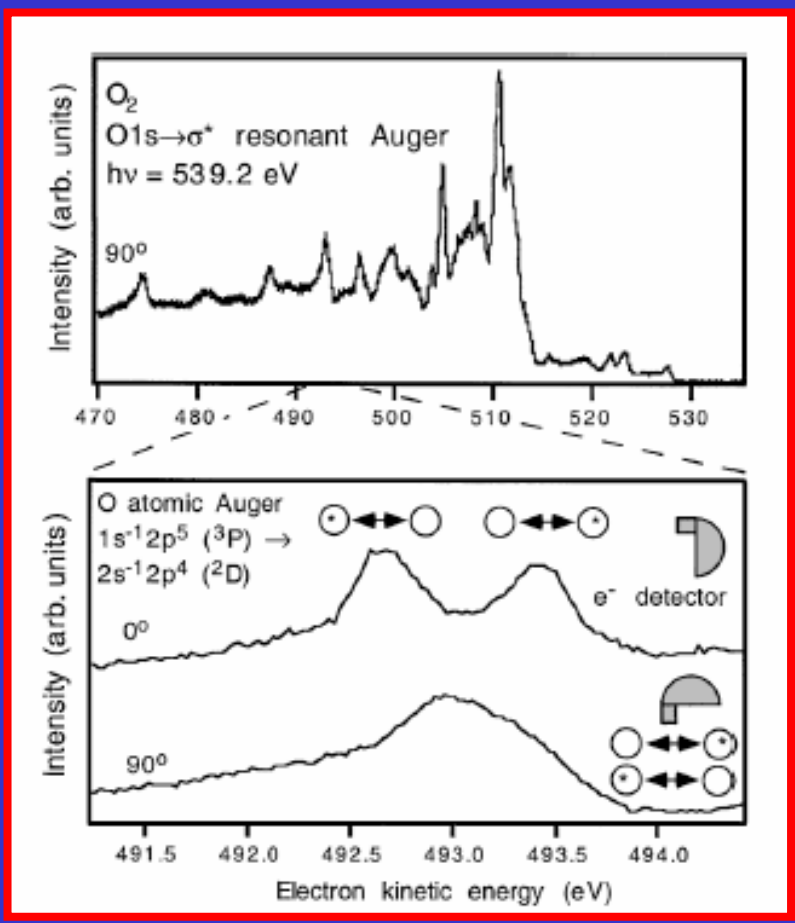


photo-absorpt.. selectivity

O ⟷ O

Intermolecular-direction parallel to polarization vector e

doppler shift is zero for

p ⊥ e

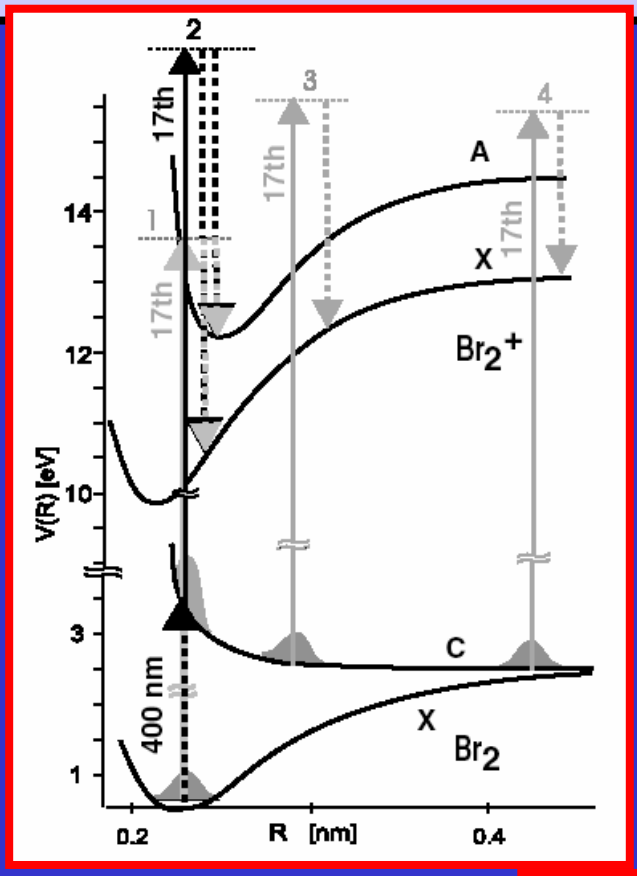
And

Opposite for

p ∥ e

O. Bjorneholm et al. Phys. Rev. Lett. 84,2826 (2000)

Ultrafast Time-Resolved Photoelectron Spectroscopy of Dissociating Br_2



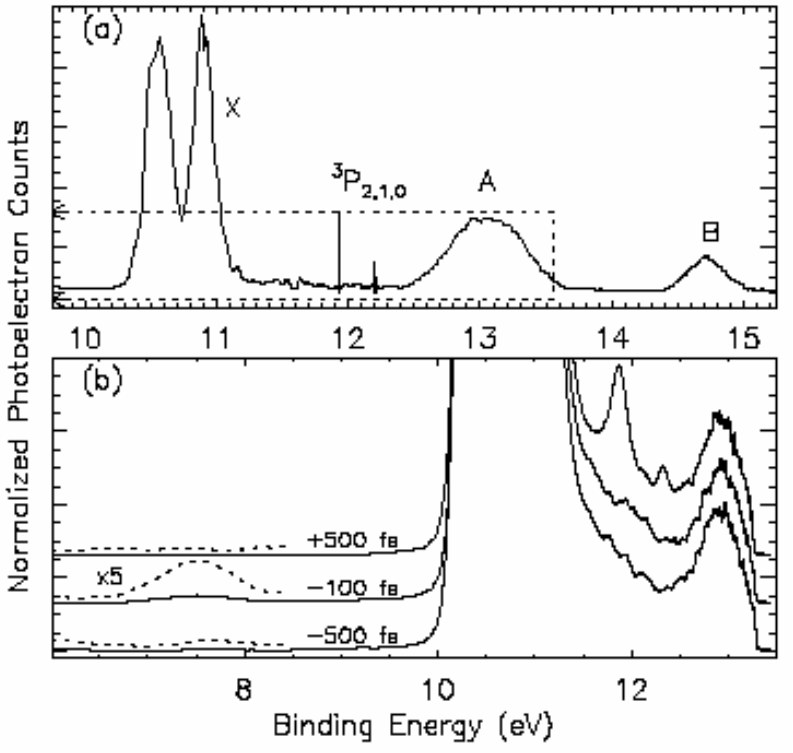
Pump: hν = 3 eV, 80 fs

Probe: table-top x-ray source
hν = 26.4 eV, 250 fs
$3x10^6$ phs/pulse

A, B, X final states with hν = 26.4 eV

with a pump-probe delay of 500 fs a clear signal from atomic species is present

40 fs is the calculated dissociation time From cross-correlation analysis



Lora Nugent-Glandorf et al. Phys.Rev.Lett.87, 103002-1 (2001)

HETEROGENEOUS CATALYSIS

- **PHOTOELECTRON SPECTROSCOPY IS ONE OF THE MOST FREQUENTLY USED TECHNIQUES TO STUDY H.C.**

- **IT PROVIDES INFORMATION ABOUT THE CHEMICAL PROPERTIES OF THE NEAR-SURFACE REGION**

- **IDENTIFICATION OF THE DIFFERENT REACTION PRODUCTS AND THEIR TIME EVOLUTION**

- **LIMITATION OF PRESENT S.R. SOURCES: LONG DATA ACQUISITION TIME DUE TO LOW PHOTON FLUX**

- **FEL RADIATION WILL PERMIT XPS DATA ACQUISITION WITH UNPRECEDENTED SPEED**
- **(POSSIBILITY FOR TIME-RESOLVED AND LATERALLY RESOLVED XPS MAPPING)**

MICROSCOPY AND SPECTROMICROSCOPY

- **CONTRAST IN THE SOFT-X-RAY ARE MAINLY DETERMINED BY:**
  - **PHOTOELECTRIC ABSORPTION**
  - **PHASE SHIFT**

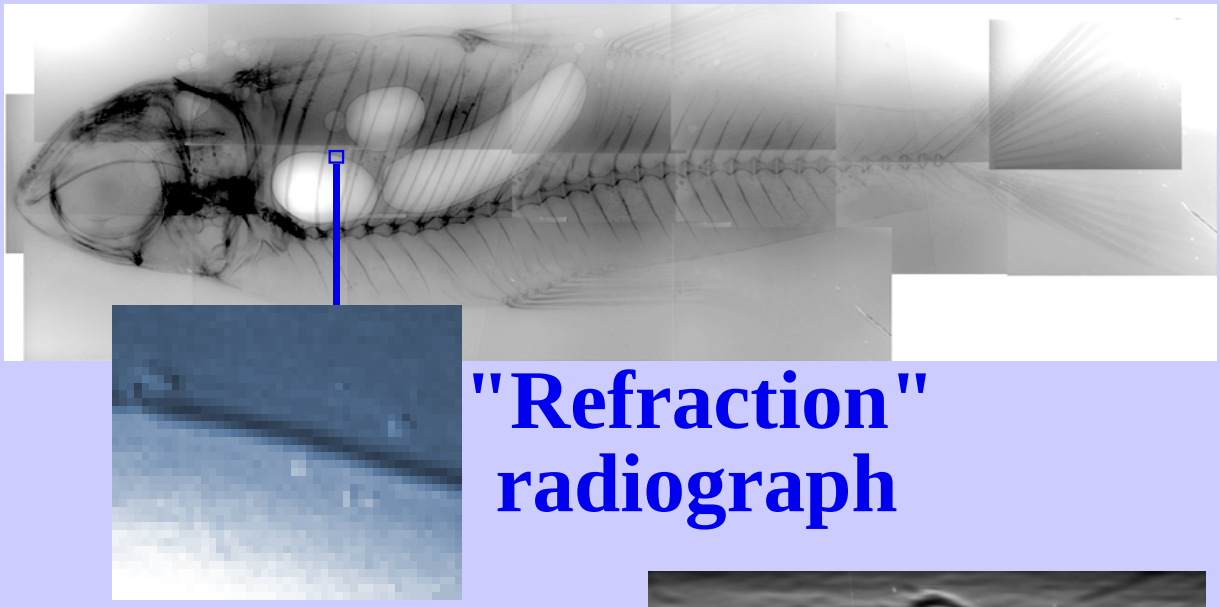
- **THE BEST SPATIAL RESOLUTION OBTAINED UP TO NOW IS ≅ 30 nm (ZONE PLATE, PEEM)**

- **CONTRIBUTION TO THIS LIMIT COMES FROM ABERRATIONS OF THE OPTICAL ELEMENTS SCALING WITH THEIR DIMENSIONS**

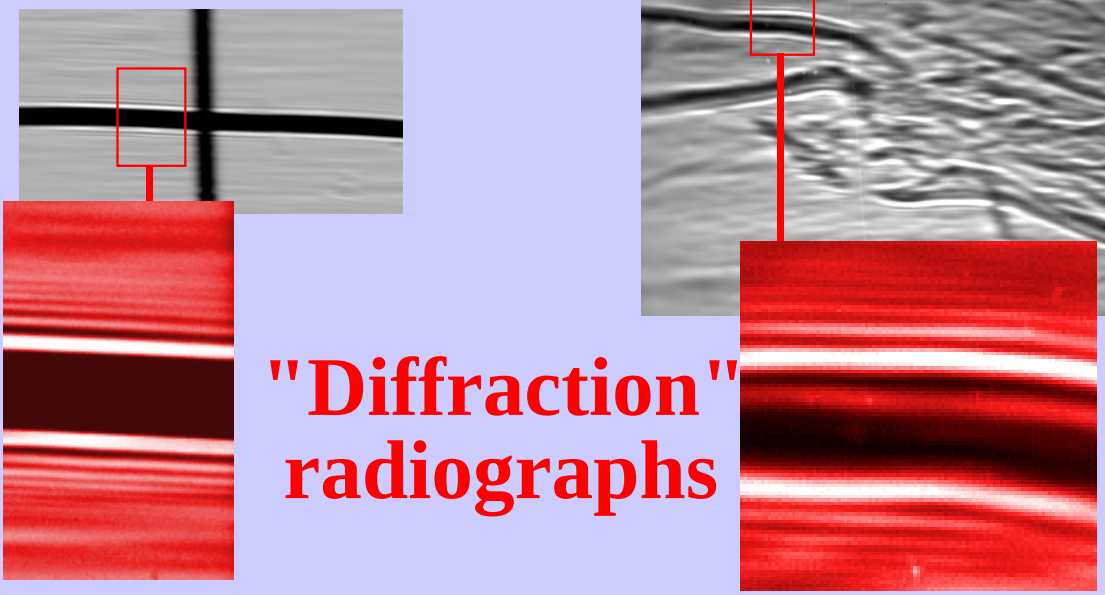
- **THE HIGH BRILLIANCE OF THE FEL ALLOWS THE REDUCTION OF THE DIMENSIONS AND A BETTER LATERAL RESOLUTION**

- **OF EXTREME INTEREST FOR BIOLOGY IS THE WATER WINDOW (280-530 eV)**
- **IN A RECENT REVIEW (J. KIRZ ET AL. 1995) MANY APPLICATIONS ARE SUMMARIZED:**
- **CHROMOSOMES**
- **MALARIA INFECTED ERYTROCYTES**
- **CALCIFIED TISSUES**
- **MUSCLES**
- **LIPID MEMBRANES**
- **POLYMERS**
- **ETC.**

Coherence-Enhanced Radiography
Refraction vs Diffraction Mechanisms



"Refraction" radiograph

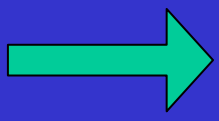


"Diffraction" radiographs

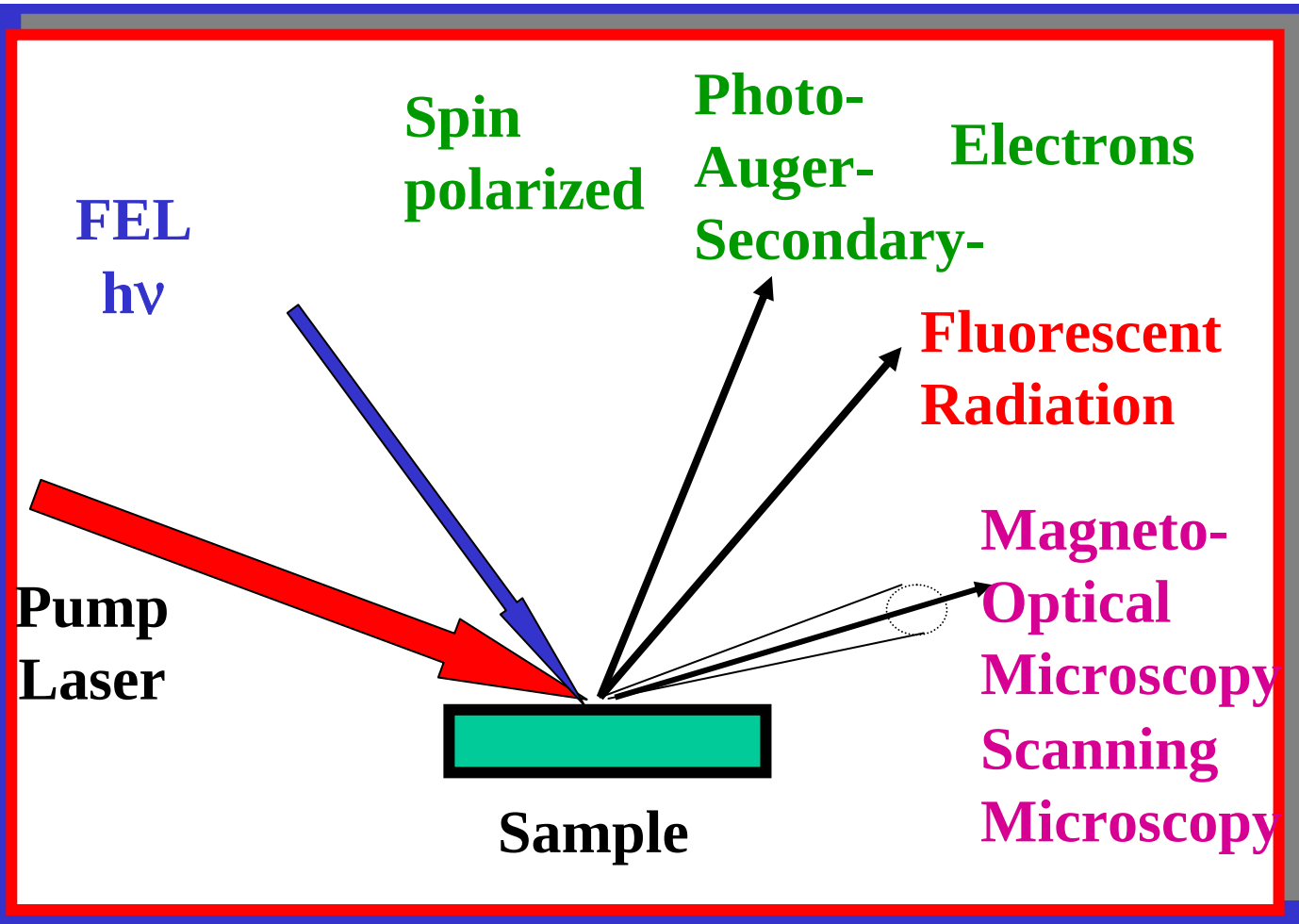
(Y. Hwu et al., data taken at the Pohang source)

SOLID STATE
HIGH RESOLUTION PHOTOELECTRON SPECTROSCOPY

- THE ACTUAL LIMIT IN ENERGY RESOLUTION OF PRESENT SPECTROMETERS IS $\Delta E \cong 10$ meV

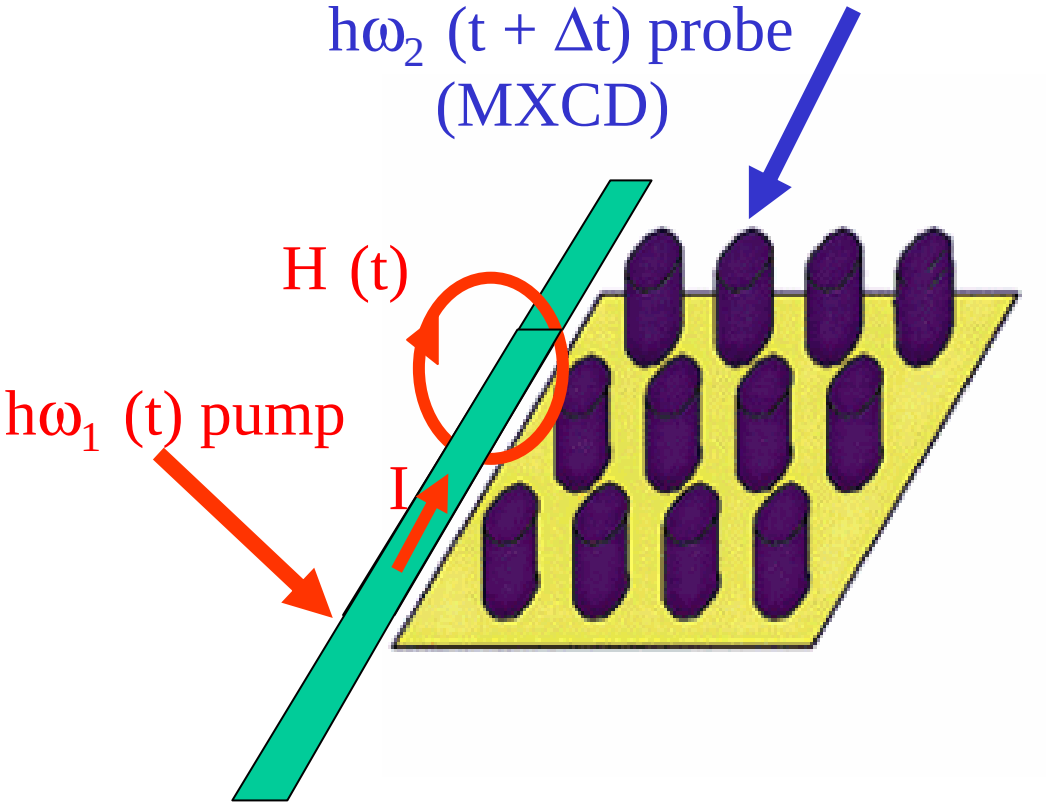
- INCREASING RESOLUTION  DECREASING PHOTON FLUX AND COUNTS RATE

- THE HIGH FEL PHOTON FLUX ALLOWS TO REDUCE ΔE DOWN TO $\cong 1$ meV
    - SPECTROSCOPY AT AND CLOSE TO THE FERMI SURFACE, TO UNDERSTAND TRANSPORT, MAGNETIC PHENOMENA, SUPERCONDUCTIVITY, HIGHLY CORRELATED MATERIALS
    - DILUITE CONCENTRATIONS OF ATOMS IN SOLIDS, SURFACES, INTERFACES

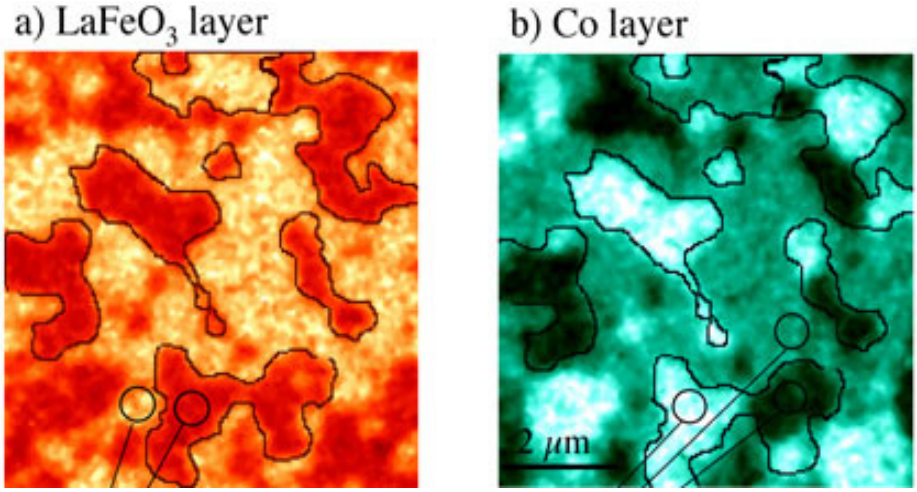


- Spin-Resolved photoelectron Spectroscopy on Magnetic surfaces
  - Pump: laser beam for heating the sample for T>Tc the spin alignment is destroyed
  - on what time-scale the rise in lattice temperature (phonon population) destroy the spin alignment
  - Dynamics: spin-lattice relaxation (nsec fsec ?)
  - Important for magneto-optical recording
  - Probe: FEL radiation → spin-polarized photo-, Auger-electrons

Probe Charge and Spin Dynamics (fs < Δt < ps)

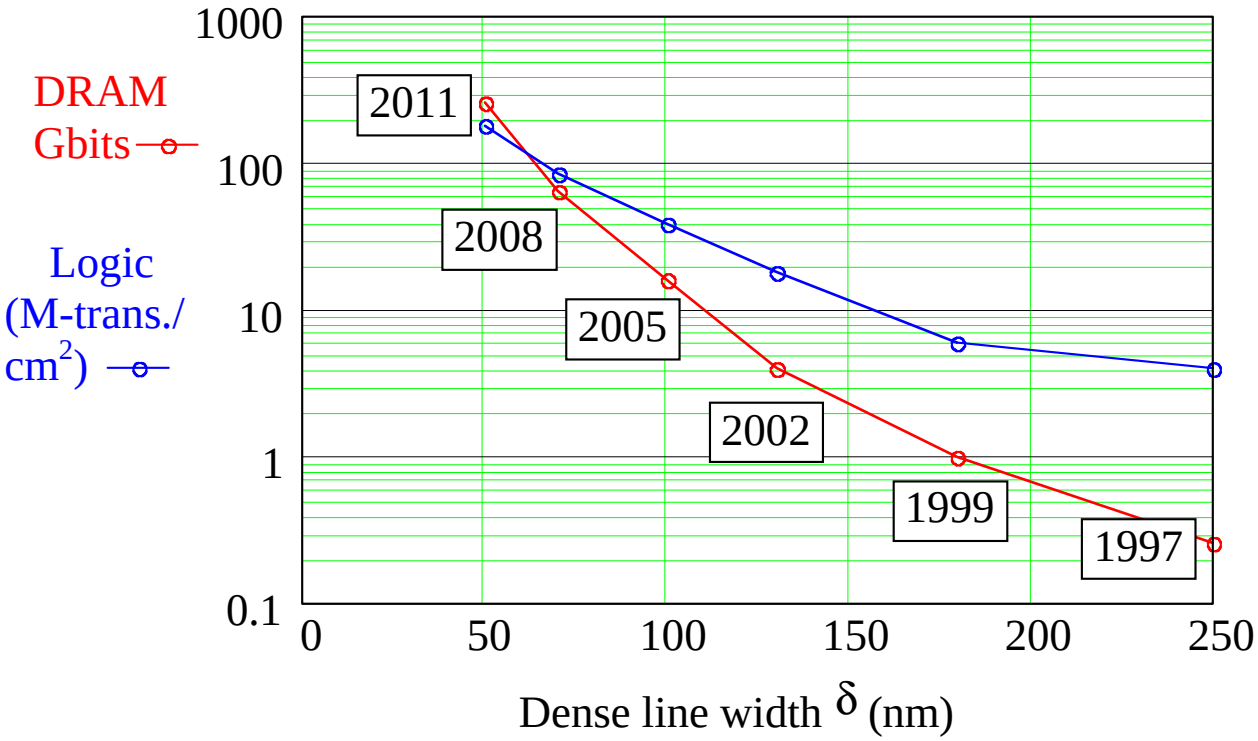


Magnetic Microscopy (XMCD)
F. Nolting et al., Nature 405, 767 (2000)



NANOLITOGRAPHY

DRAM (Dynamic Random Access Memory) in bits/chip and Logic (transistors/cm^2) according with the National Techn. Roadmap of the Semiconductor Industry Assoc. April 1999.

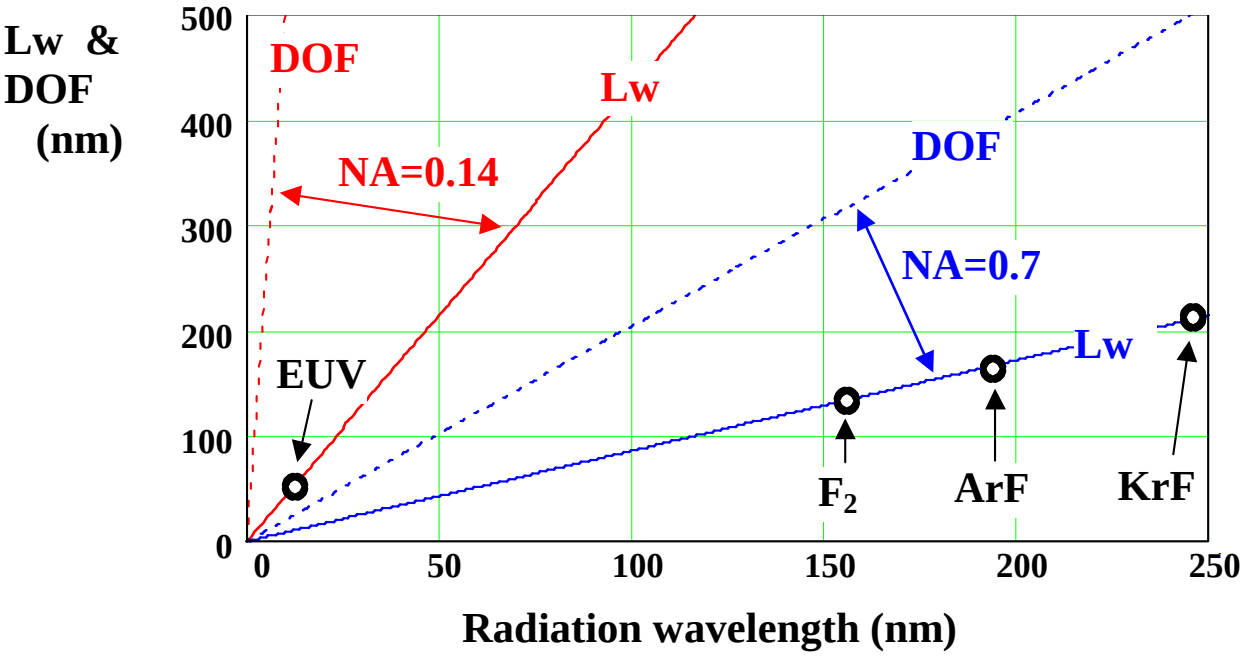


Candidate technologies for sub-100 nm lithography:

- **Optical-L: by using the F_2 laser at λ=157 nm**
- **X-ray-L: proximity lithography @ hν~ 1.2 keV**
- **EUV-L: projection lithography @ hν~ 90 eV**
- **EPL: Electron Projection Lithography @ e^--beam**
- **IONS-L: projection lithography @ ions-beam**

Dense line width "Lw" (solid) and Depth Of Focus "DOF" (dashed) vs radiation wavelength "λ".

Lw = k1·λ/NA **k1~ 0.6** **NA=numerical aperture**
DOF= k2· λ/NA^2 **k2~ 0.5**



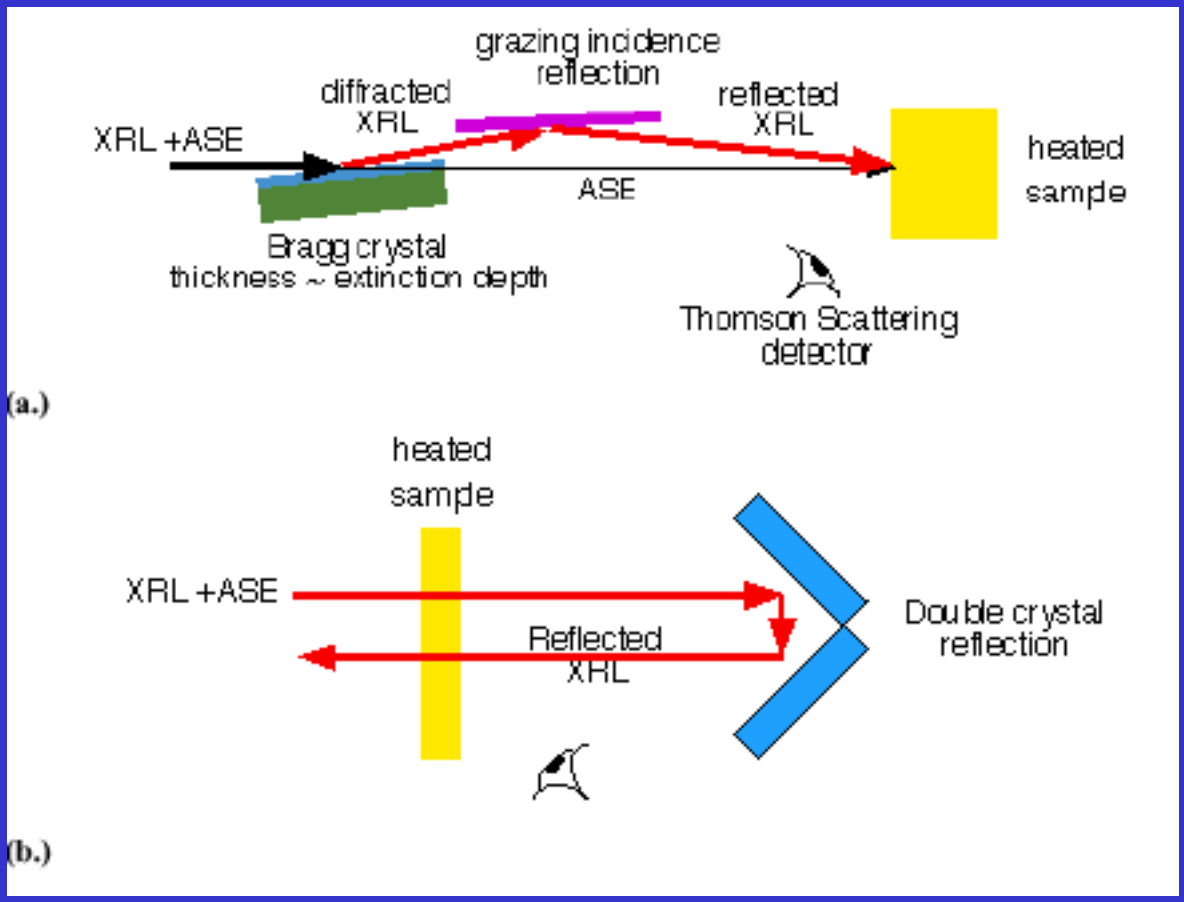
Power requirements (T. Silvfast, J. Q. Elect, Vol 35, 1999):

On the 300 mm wafer: 120 mW (for 60 Wafer*layer/hour)
From mercury lamp (λ=248 nm): 1 kW
From excimer lasers (ArF, KrF): few Watts
From F_2 laser (λ=157 nm): few Watts
From EUV source: 3 W (on the condenser mirror in 2.5% refl. band-width @ 13 nm)

HARD X-RAY

- **X-FEL CAN BE USED TO PRODUCE AND/OR TO STUDY WARM DENSE PLASMA**
- **IMPORTANT STUDIES FOR FUSION PROCESSES**
- **WARM DENSE MATTER REFERS TO THAT PART OF THE DENSITY-TEMPERATURE PHASE DISGRAM WHERE STANDARD THEORIES OF CONDENSED MATTER PHYSICS AND PLASMA STATISTICS PHYSICS ARE NOT VALID. THE WARM DENSE MATTER IS A STATE INTERMEDIATE BETWEEN THE SOLID STATE AND THE PLASMA; A POSSIBLE STATE OF: PLANETS INTERIOR, COLD DENSE STARS, PLASMA DIRECTLY CREATED FROM CONDENSED MATTER (LASER ABLATION, INERTIAL FUSION PROCESSES)**

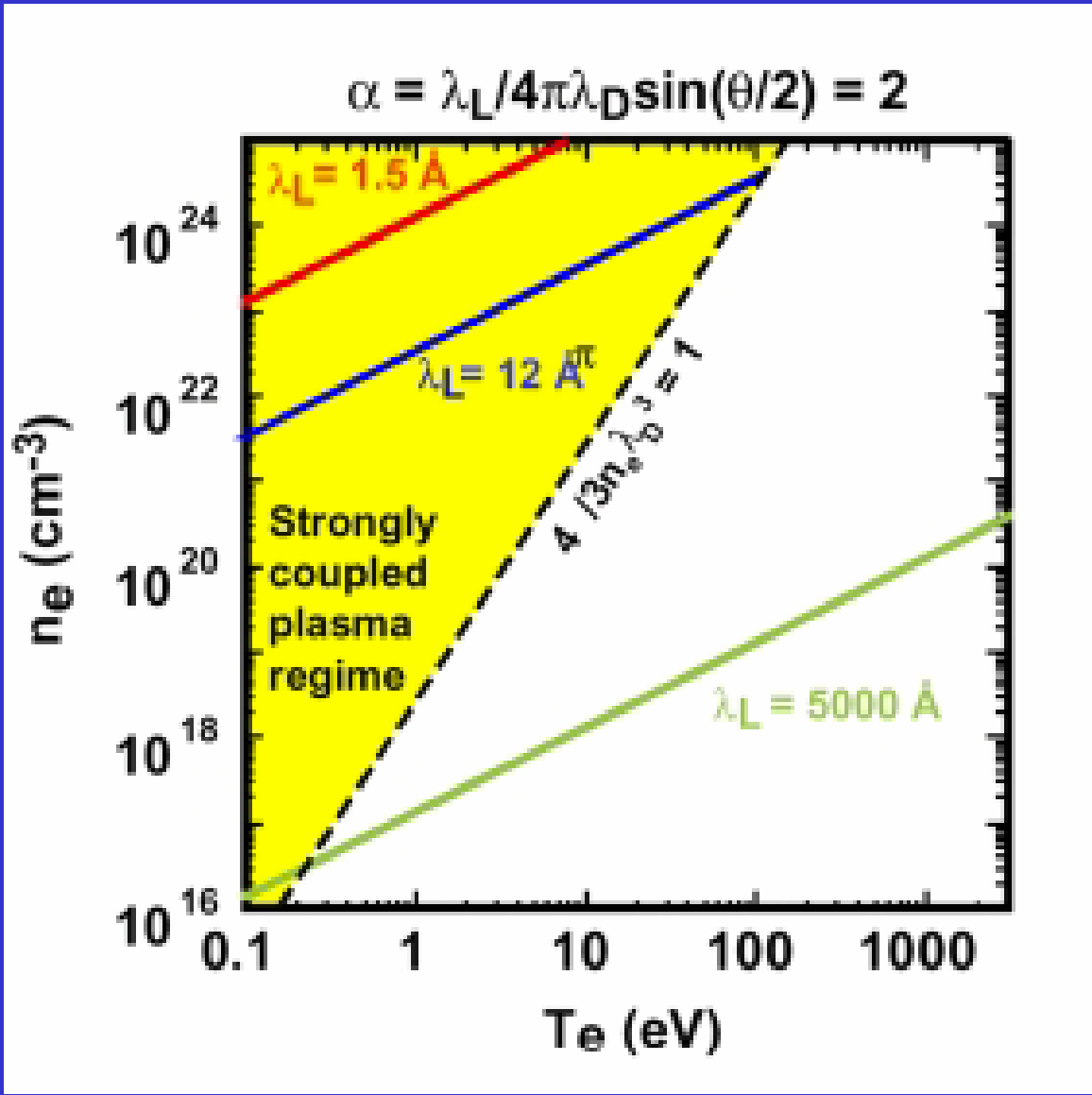
- **PUMP-PROBE EXPERIMENTS**
- **PUMP: SPONTANEOUS EMISSION**
- **PROBE: X-FEL EMISSION**
- **THOMSON SCATTERING GIVE INFORMATION ON THE IONIZATION STATE, DENSITY, TEMPERATURE AND MICROSCOPIC PROPERTIES OF PLASMA**



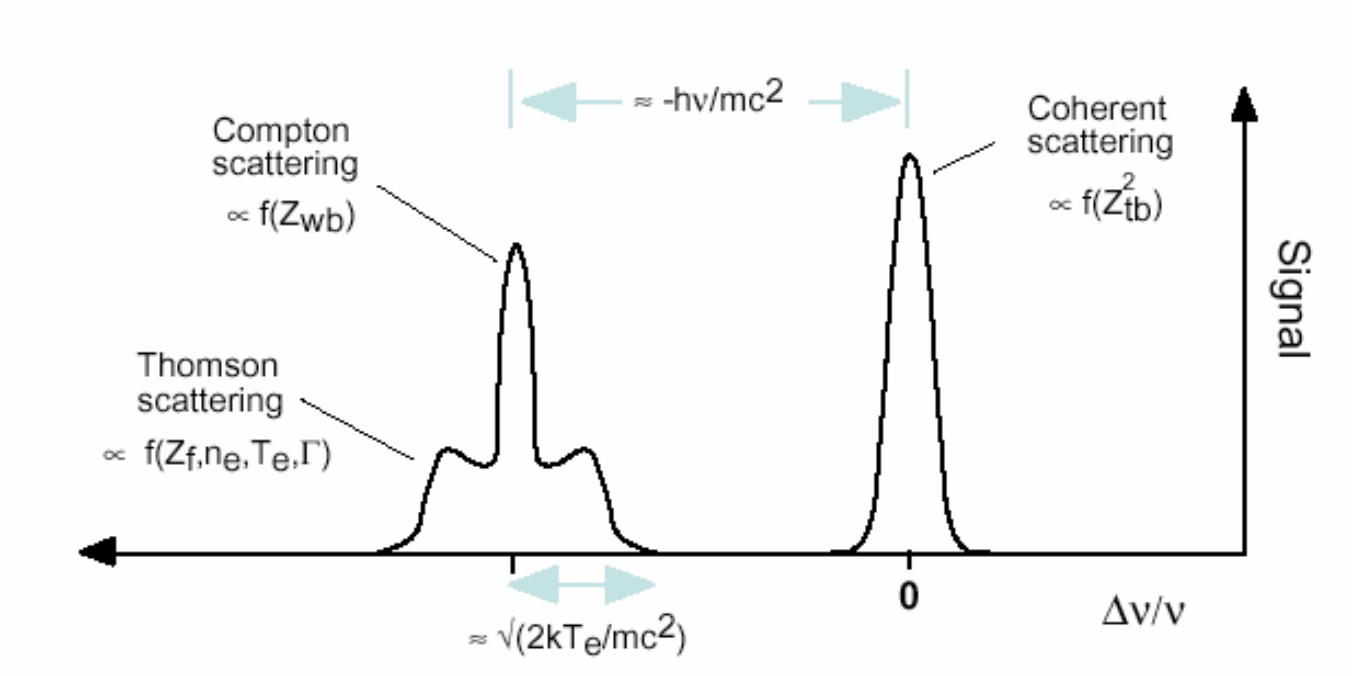
WITH AN X-FEL OF 1.5 Å WAVELENGTH

IT IS POSSIBLE TO ANALYSE PLASMA

WITH DENSITY > 10^{23} cm^{-3}



THOMSON SCATTERING



- **Coherent unshifted peak: intensity proportional to tightly bounded electrons (z_{tb}^2/atom)**
- **Incoherent Compton peak: intensity proportional to weakly bounded electrons (z_{wb}/atom)**
- **Thomson scattering: spectral integrated intensity is proportional to free electrons (z_f / atom)**

BIOLOGY: STRUCTURAL STUDIES

- **STRUCTURES AVILABLE (END 2000)**
  X-RAY 10143
  NMR 2128
- **PROTEIN CODED IN THE HUMAN GENOME 100.000-140.000**
- **THE BOTTLENECK IS THE NEED FOR CRYSTALS**
- **MANY BIOLOGICAL COMPLEXES CANNOT BE CRYSTALLISED**
- **AN X-FEL SOURCE CAN CHANGE THE SCENARIO COMPLETELY: THE GOAL IS TO BE ABLE TO PERFORM DIFFRACTION STUDIES ON SINGLE MACROMOLECULES, VIRUS, NANOCRYSTALS, …..**

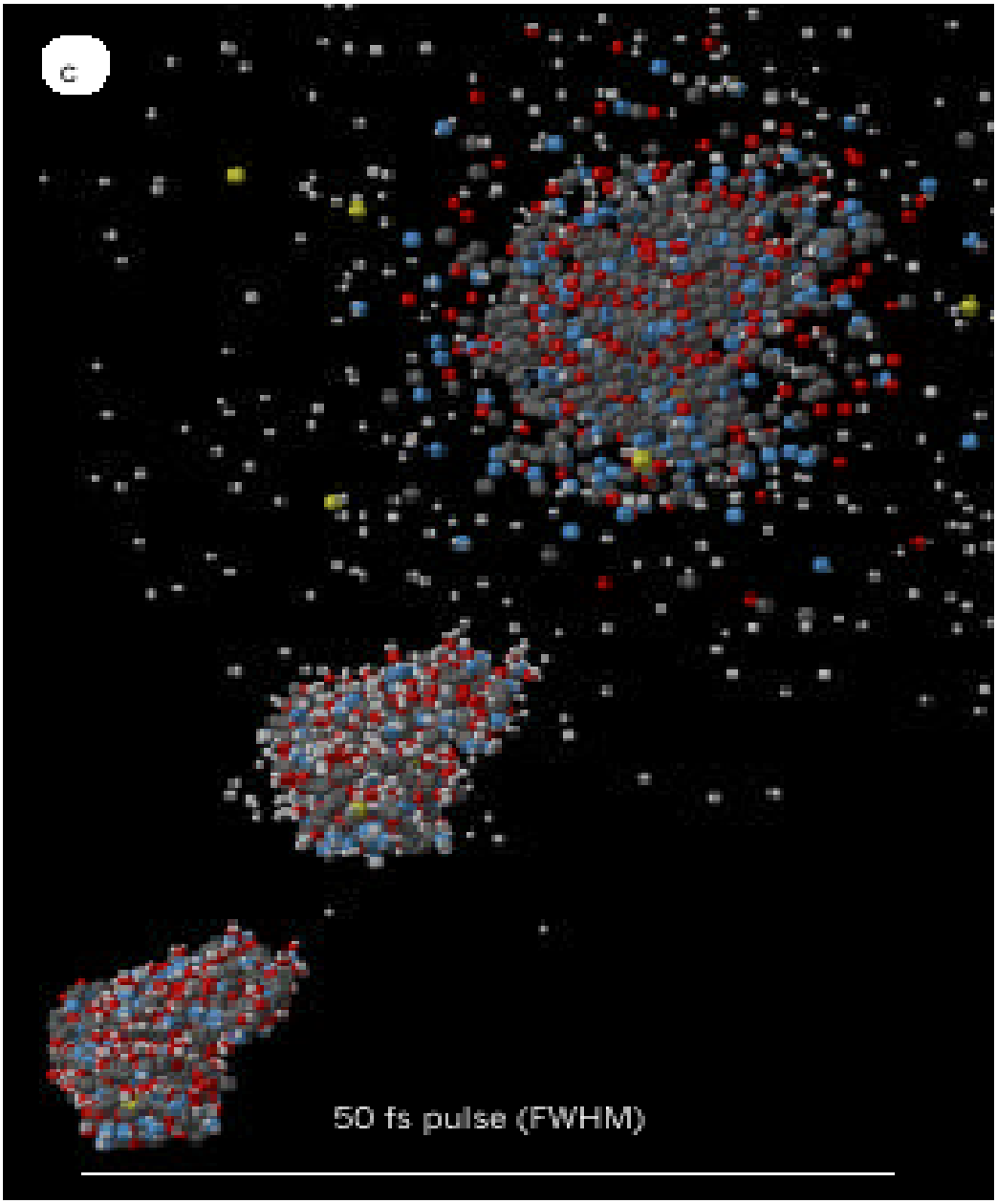
- **SINGLE-SHOT EXPERIMENTS**

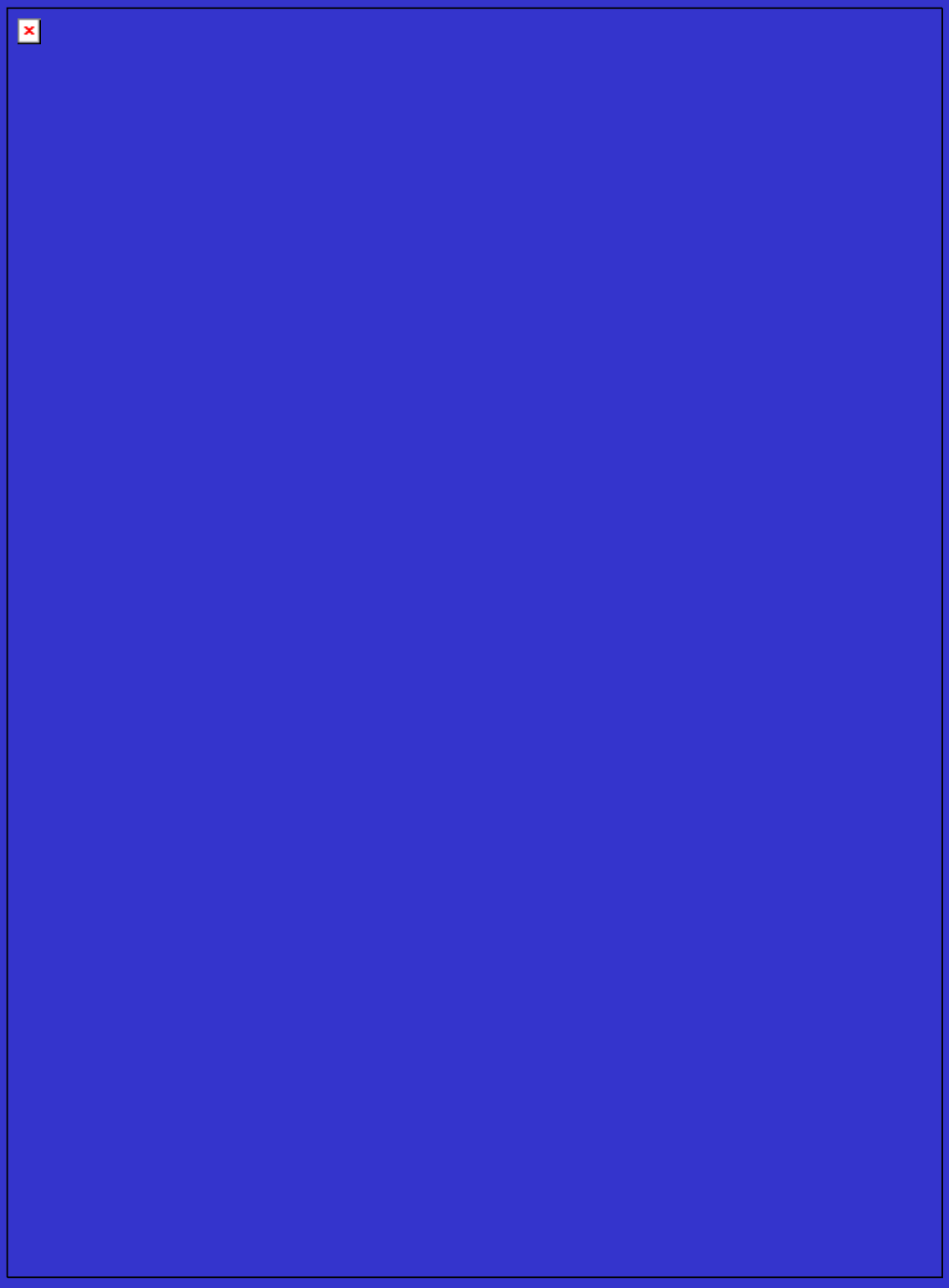
- **X-RAY HOLOGRAPHY**

- **X-RAY TOMOGRAPHY**

- **THE BOTTLENECK FOR MANY OF THIS KIND OF EXPERIMENTS IS RADIATION DAMAGE**
- **THE X-FEL ADVANTAGE IS THE SHORT PULSE LENGTH <200 fs**
- **MODELLIZATION OF RADIATION DAMAGE ON THE BASIS OF THE : PHOTOELECTRIC EFFECT (95%) AND OF ANELASTIC (COMPTON/RAMAN) EFFECTS SHOWS THAT IN THE MOLECULE A LARGE NUMBER OF POSITIVE CHARGES ARE FORMED GIVING RISE TO A COULOMB EXPLOSION**
- **NEED FOR SINGLE-SHOT EXPERIMENTS**
- **THE DEGREE OF CONVERSION OF POTENTIAL ENERGY INTO KINETIC ENERGY DURING THE X-RAY EXPOSURE IS INERTIA LIMITED AND STRONGLY DEPENDS ON THE PULSE DURATION**
- **THE MODEL SHOWS THAT A SERIOUS DAMAGE OCCURS AFTER 100 fs**

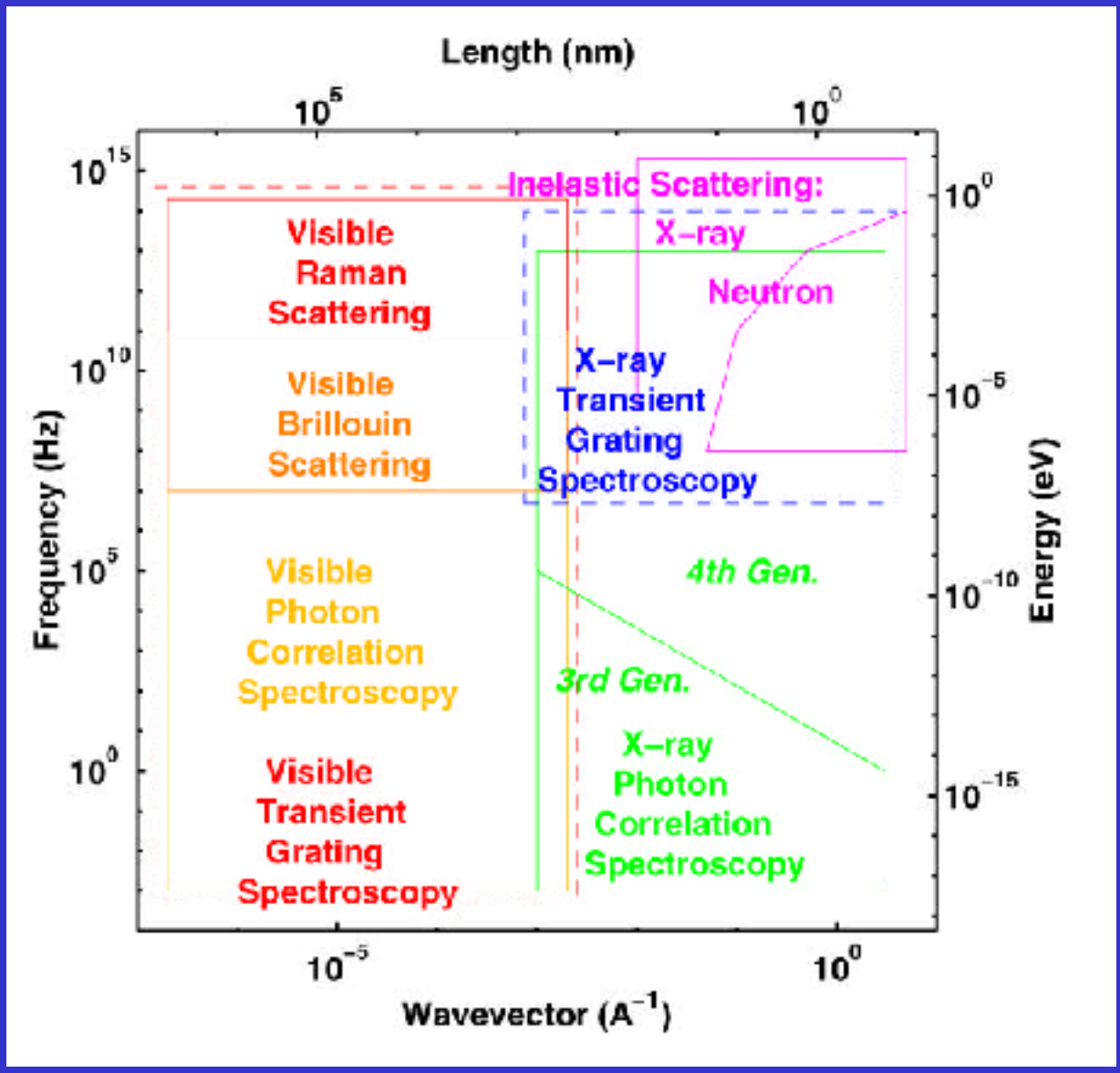
R.Neutze, R.Wouts, D. van der Spoul,,
E. Weckert, J. Hajdu; Nature 406, 752, (2000)





DINAMICAL PROCESSES IN CONDENSED MATTER

- **THE EXPERIMENTAL CHALLENGE IS TO MEASURE THE DYNAMIC STRUCTURE FACTOR S(Q, ω), OR THE CORRESPONDING RESPONSE FUNCTION S(Q, t) IN THE APPROPRIATE REGION OF THE (ω - Q) SPACE**
- **THE X-FEL ALLOWS TO COVER REGIONS OF THE (ω - Q) SPACE INACCESSIBLE BY OTHER RADIATION SOURCES**
- **POSSIBLE EXPERIMENTS:**
  - **POLYMERS**
  - **CHARGE DENSITY WAVES**
  - **QUASICRYSTALS**
  - **SURFACES**
  - **DEFECTS IN CRYSTALS**
  - **FERROELECTRICS**
  - **MAGNETIC MATERIALS**
  - **…….**


Length (nm)
10^{5}
10^{0}
10^{15}
10^{10}
10^{5}
10^{0}
Frequency (Hz)
10^{0}
10^{-5}
10^{-10}
10^{-15}
Energy (eV)
Inelastic Scattering:
X-ray
Neutron
Visible
Raman
Scattering
Visible
Brillouin
Scattering
X-ray
Transient
Grating
Spectroscopy
Visible
Photon
Correlation
Spectroscopy
4th Gen.
3rd Gen.
Visible
Transient
Grating
Spectroscopy
X-ray
Photon
Correlation
Spectroscopy
10^{-5}
10^{0}
Wavevector (A^{-1})

CONCLUSIONS

THE EXTRAORDINARY CHARACTERISTICS OF THE X-FEL

- **BRILLIANCE**
- **COHERENCE**
- **PULSE TIME STRUCTURE < 100fs**

MAKE THE NEW SOURCES COMPLEMENTARY RATHER THAN COMPETITIVE TO 3RD GENERATION (SR) FACILITIES