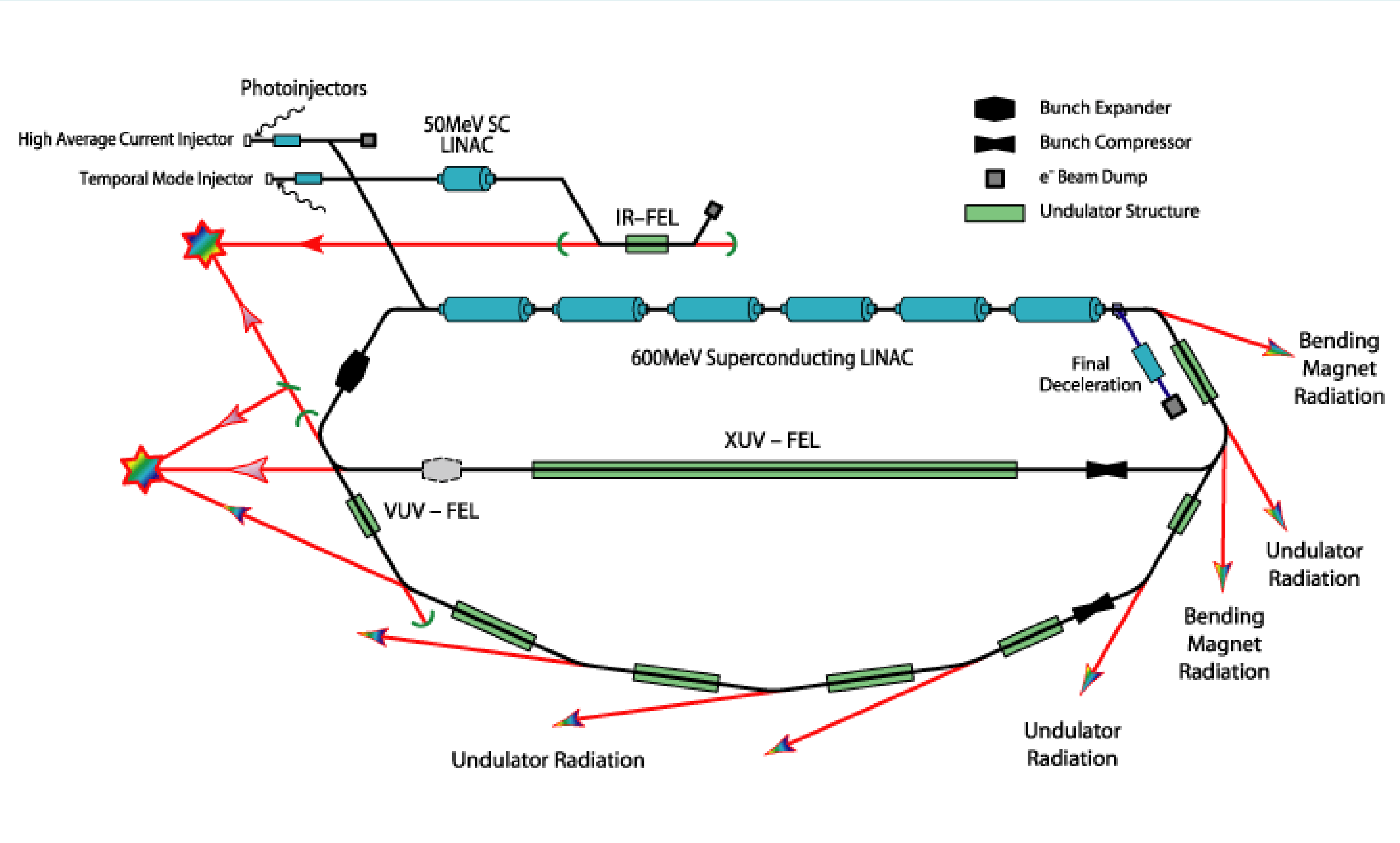
The science motivating

the UK's Fourth Generation Light Source Project

Wendy Flavell

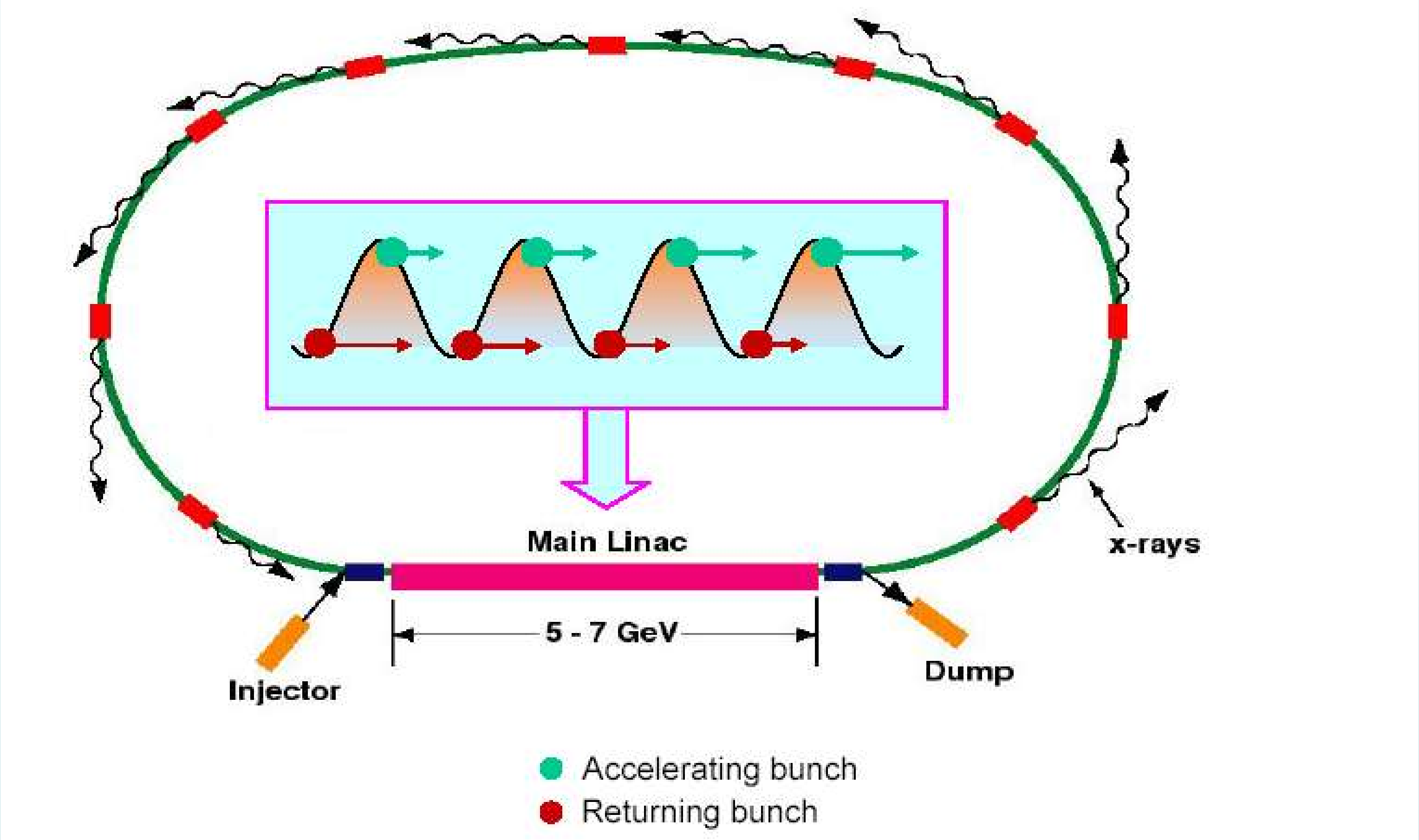
University of Manchester & CCLRC

4GLS

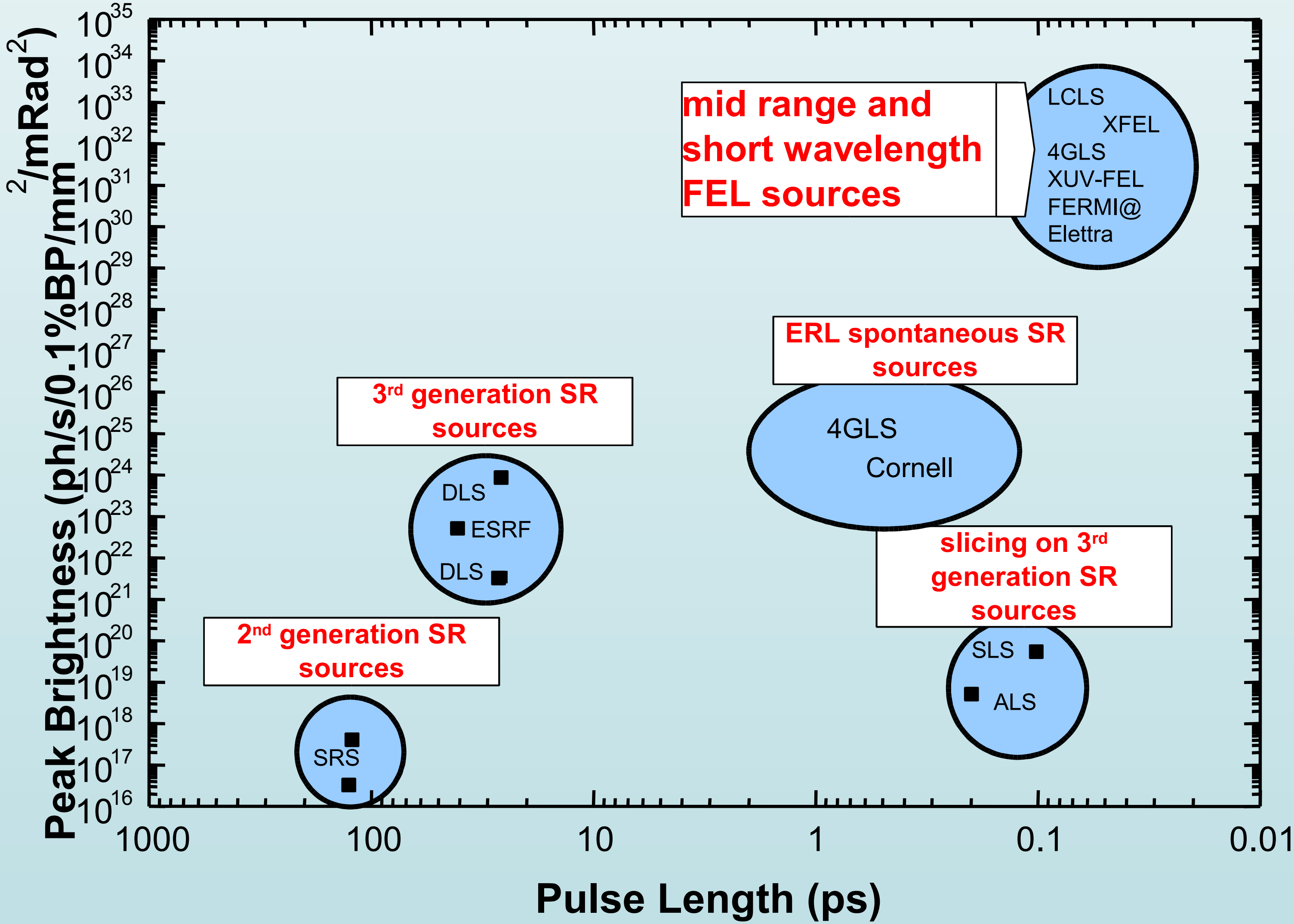


………an energy recovery linac (ERL), not a storage ring

Energy recovery in a linac

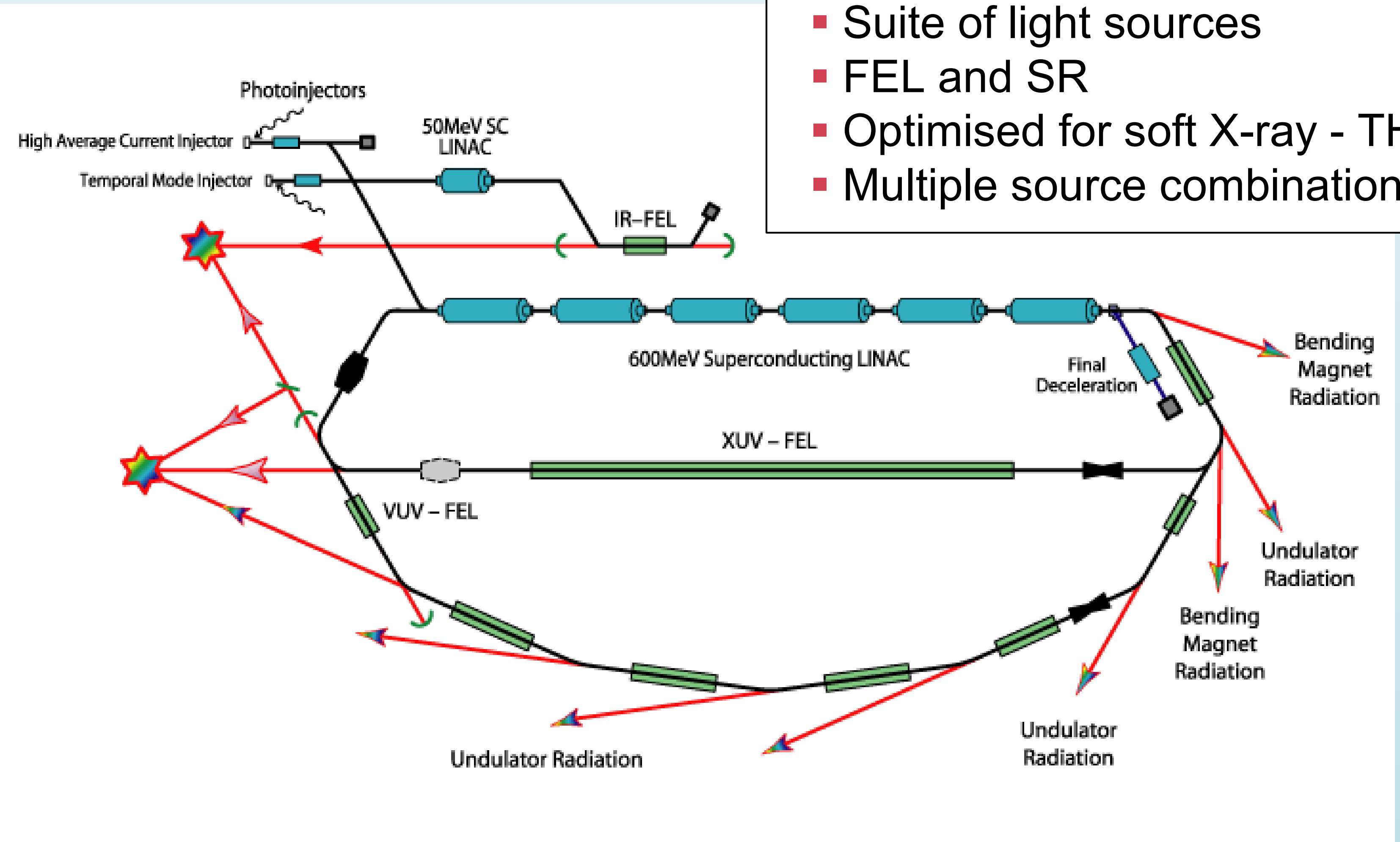


Linac-based light sources

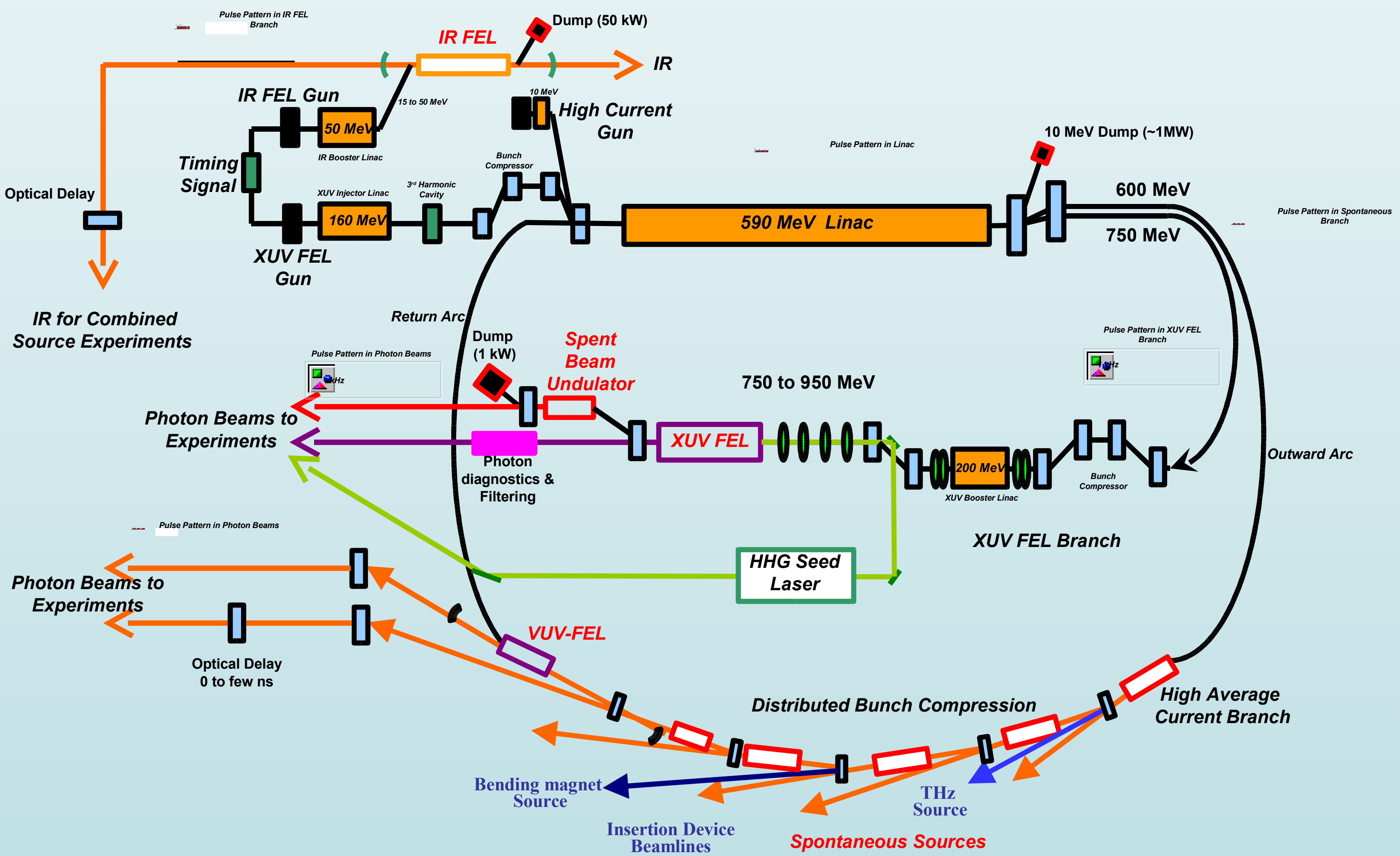


4GLS

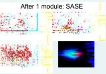
- Suite of light sources
- FEL and SR
- Optimised for soft X-ray - THz
- Multiple source combinations



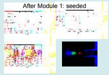
4GLS conceptual layout



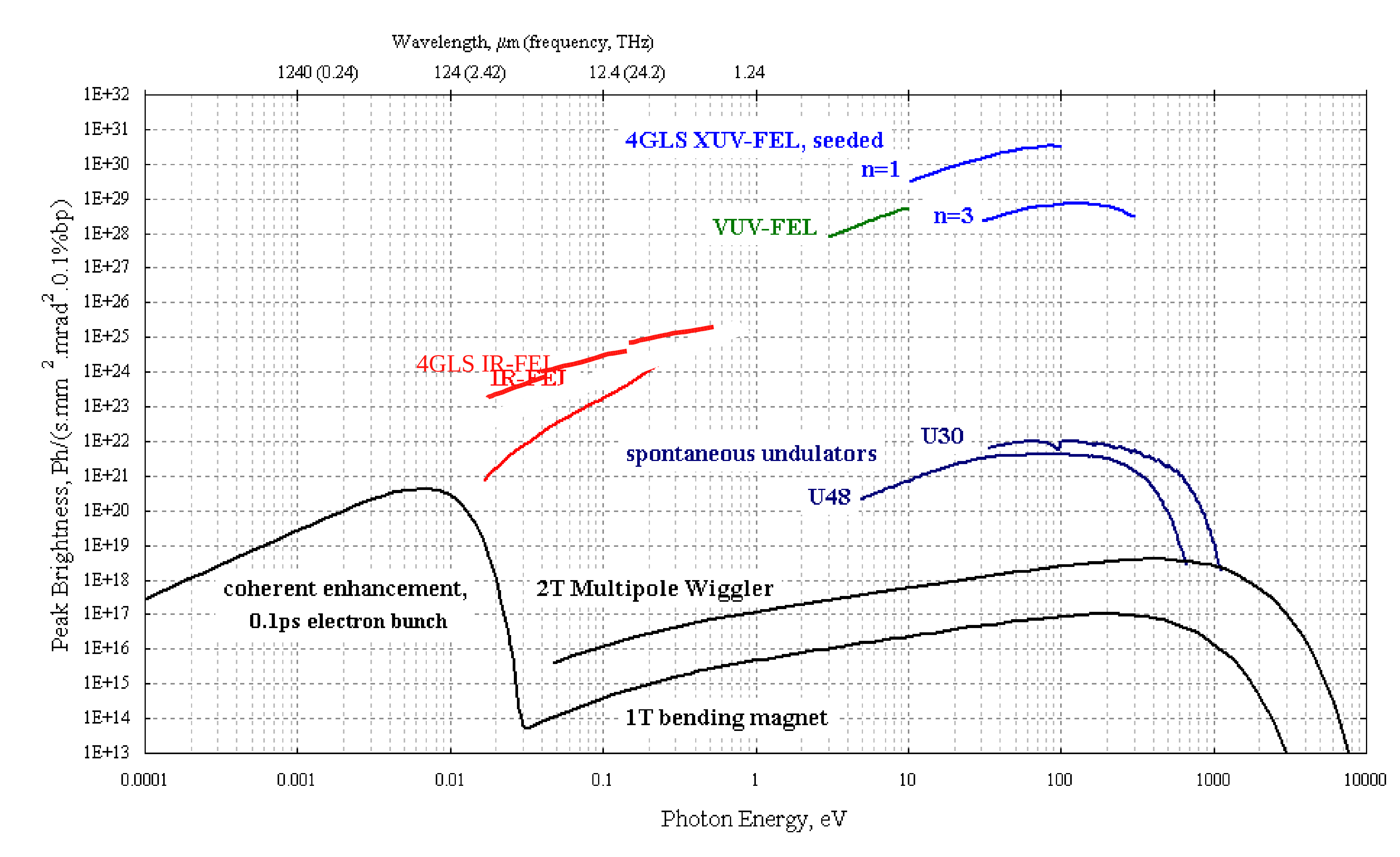
XUV FEL



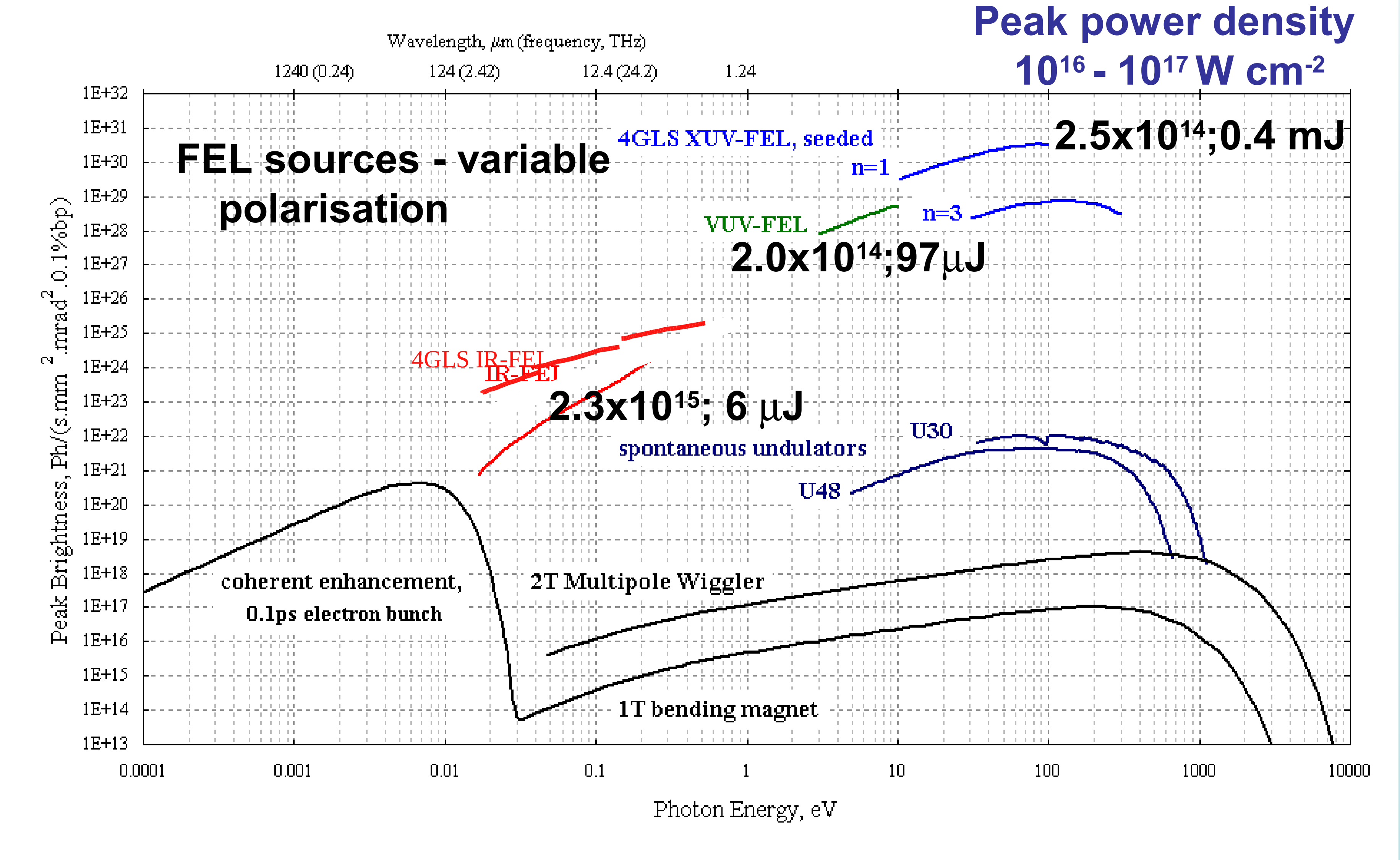
Seeding will be adopted



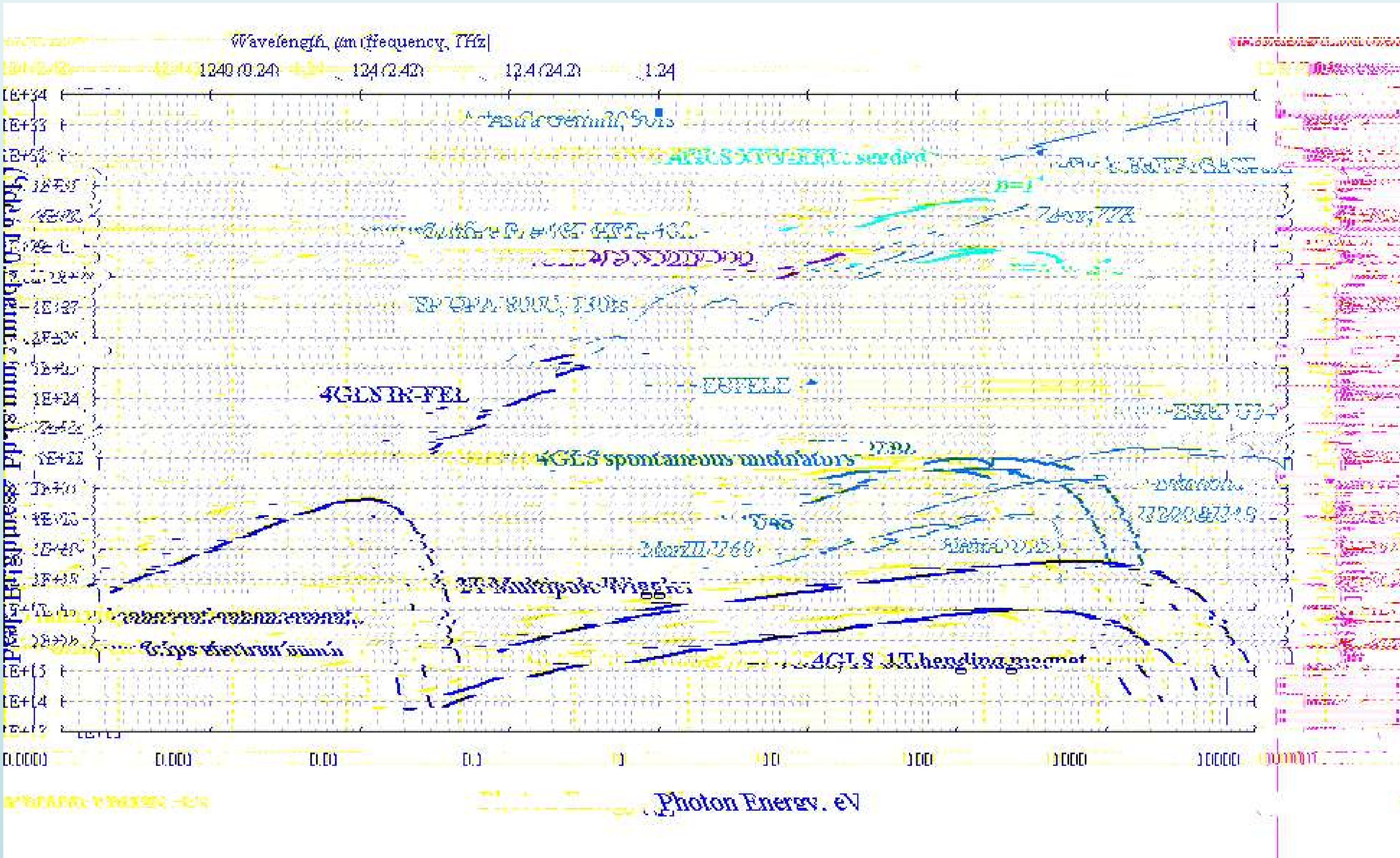
Peak Brightness



Photons per pulse and energy per pulse



4GLS: comparison with other sources



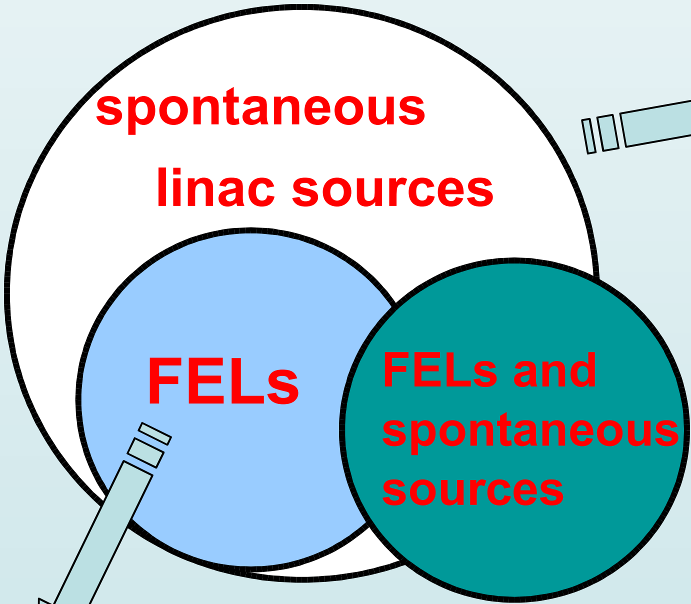
4GLS: the science need

Fundamental requirement to understand the *dynamic* behaviour of matter, often in very small (nm) units, on very fast (fs) timescales

Need not just to determine *structure* with high precision, but to understand *how these structures work*

The need is for an ultra-high brightness *low energy* facility that allows the use of fast pulsed sources in combination

4GLS: more than the sum of the parts



- dynamics of adsorbates, films & interfaces
- nondestructive biosystem dynamics
- ultra-high spectral resolution

+

- pump-probe
  FEL+ spontaneous SR (THz>)
- dynamics in nanoparticles and microdevices, spintronics
- energy transfer in biomolecules

+

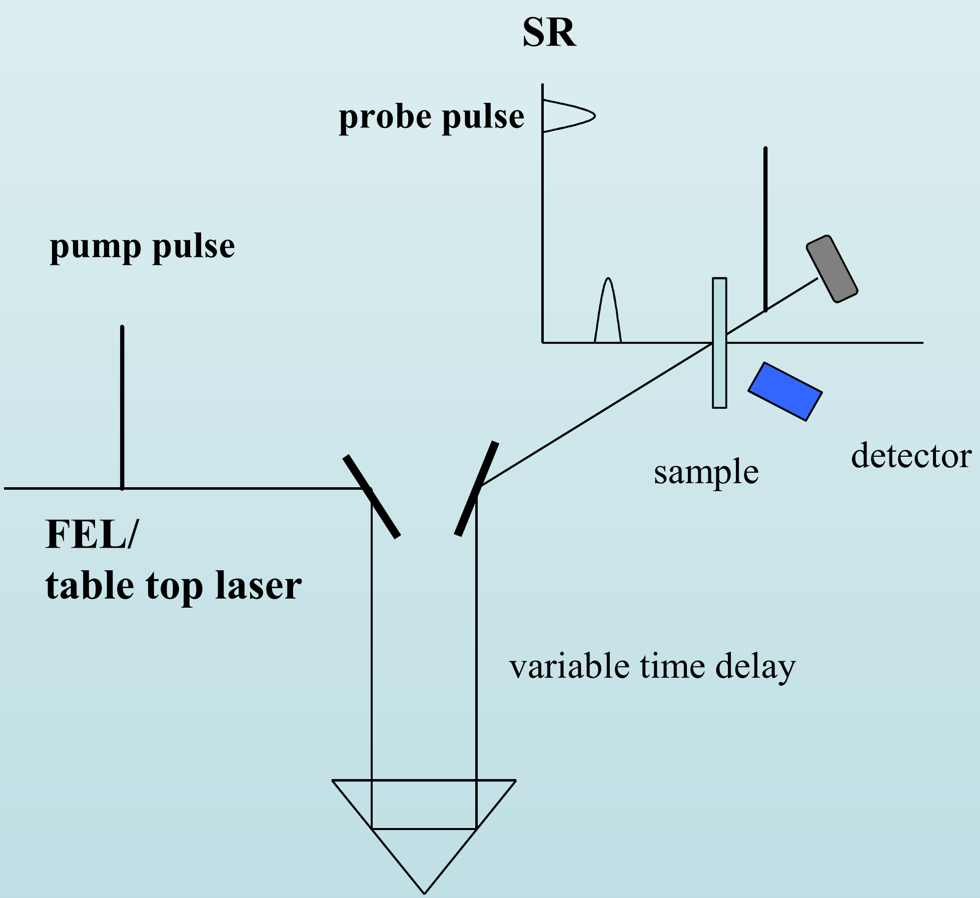
- bio and chemical fs dynamics
- materials under extreme conditions
- nonlinear processes
- plasma physics
- nano spectromicroscopy
- nano electronic structure and magnetism

4GLS: pulse combinations

Many combinations possible:

- Multiple sources
- *Naturally synchronised*
- THz-soft X-ray
- SR/FEL/tabletop lasers
- Variable pulse spacing
- Pulse quality

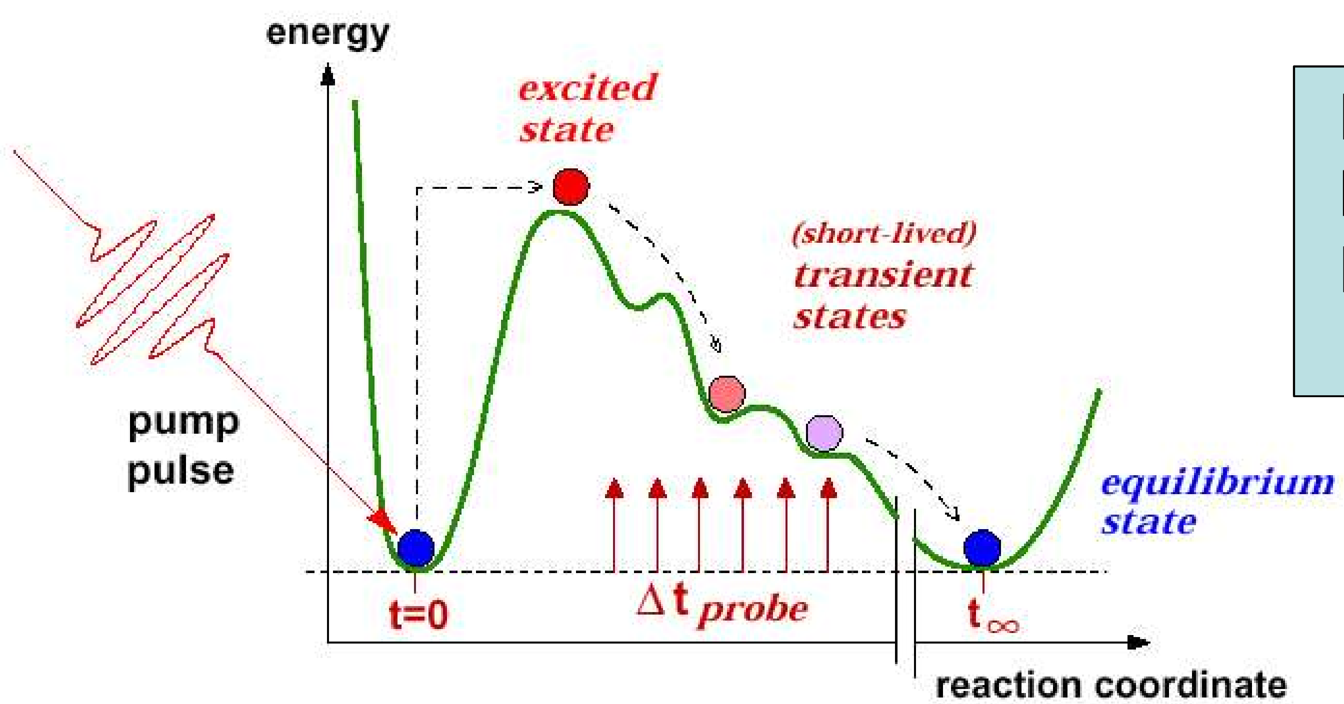
UNIQUE EXPERIMENTAL FLEXIBILITY



Dynamics and kinetics

- pulse lengths down to sub-100 fs
- real time monitoring of chemical reactions,
  bond breaking and making

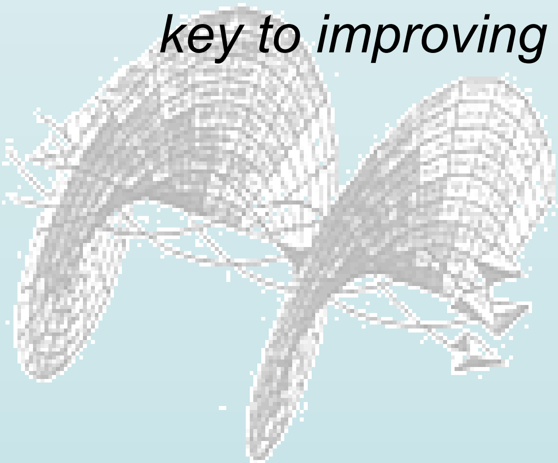
DYNAMICAL BEHAVIOUR OF REAL SYSTEMS



TESLA technical design report, March 2001

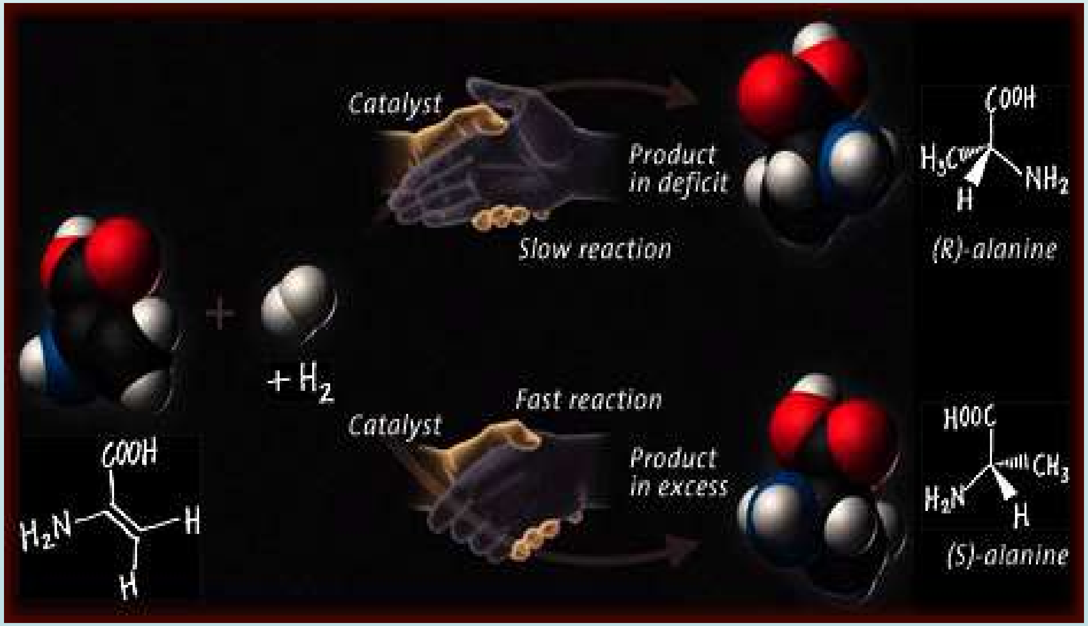
Key areas: Understanding catalytic reaction pathways

- Enzyme-catalysed processes
- Reactions contributing to atmospheric pollution
- Reactions in the interstellar medium
- Studies of enantiomer-selective chemistry:
  - *Need to understand asymmetric reaction pathways: key to improving TON*



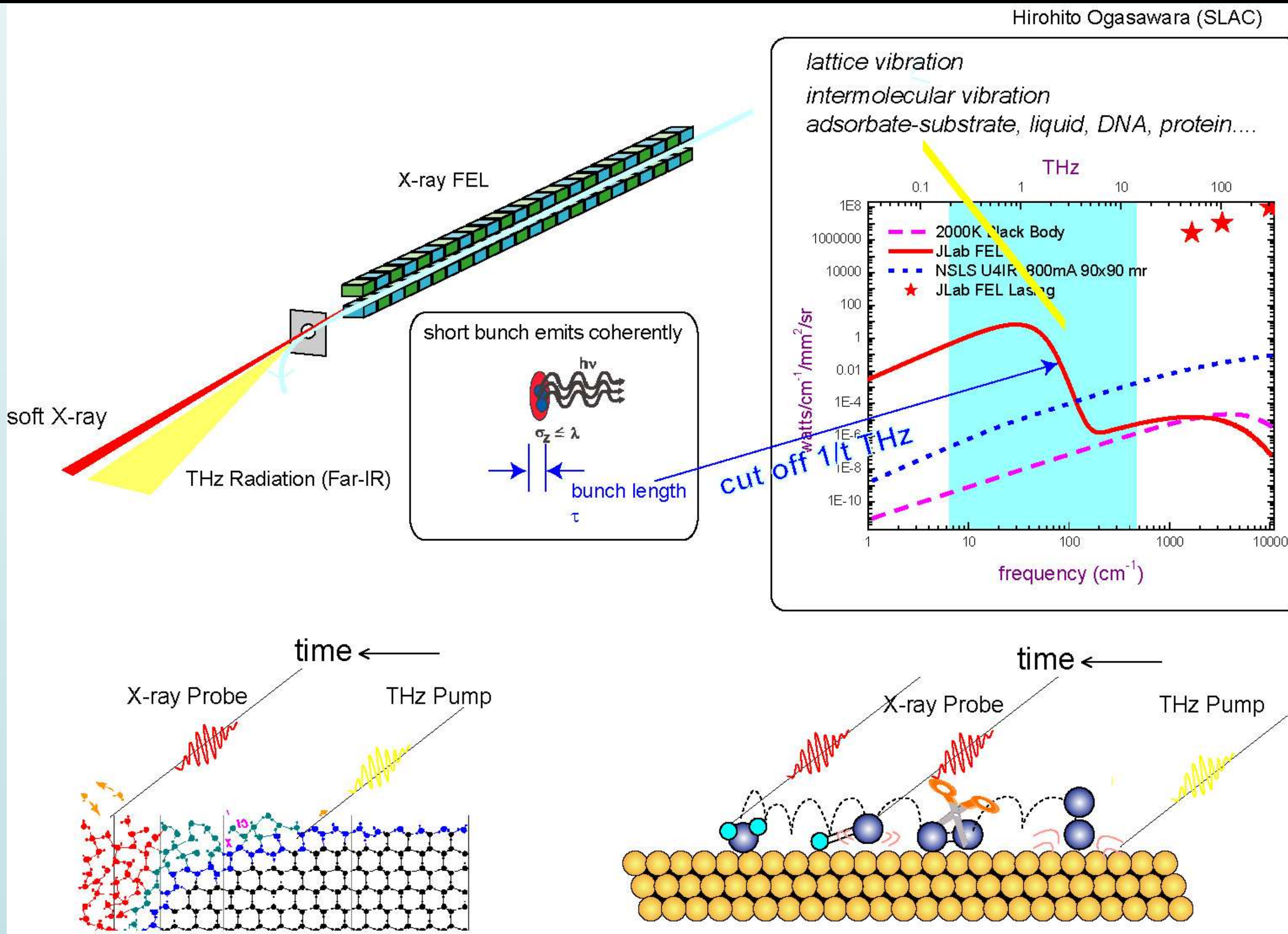
Asymmetric hydrogenation

Courtesy William Hems, Johnson Matthey Catalysts - Chiral Technologies



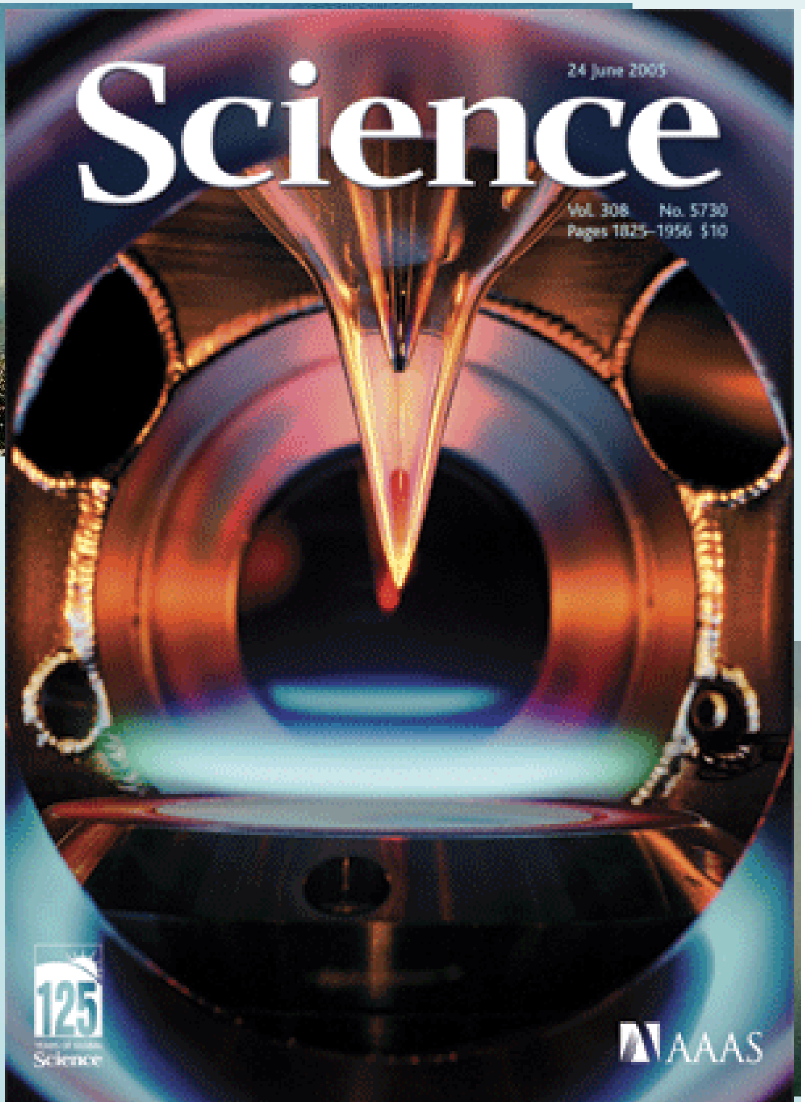
THz initiation of surface reactions

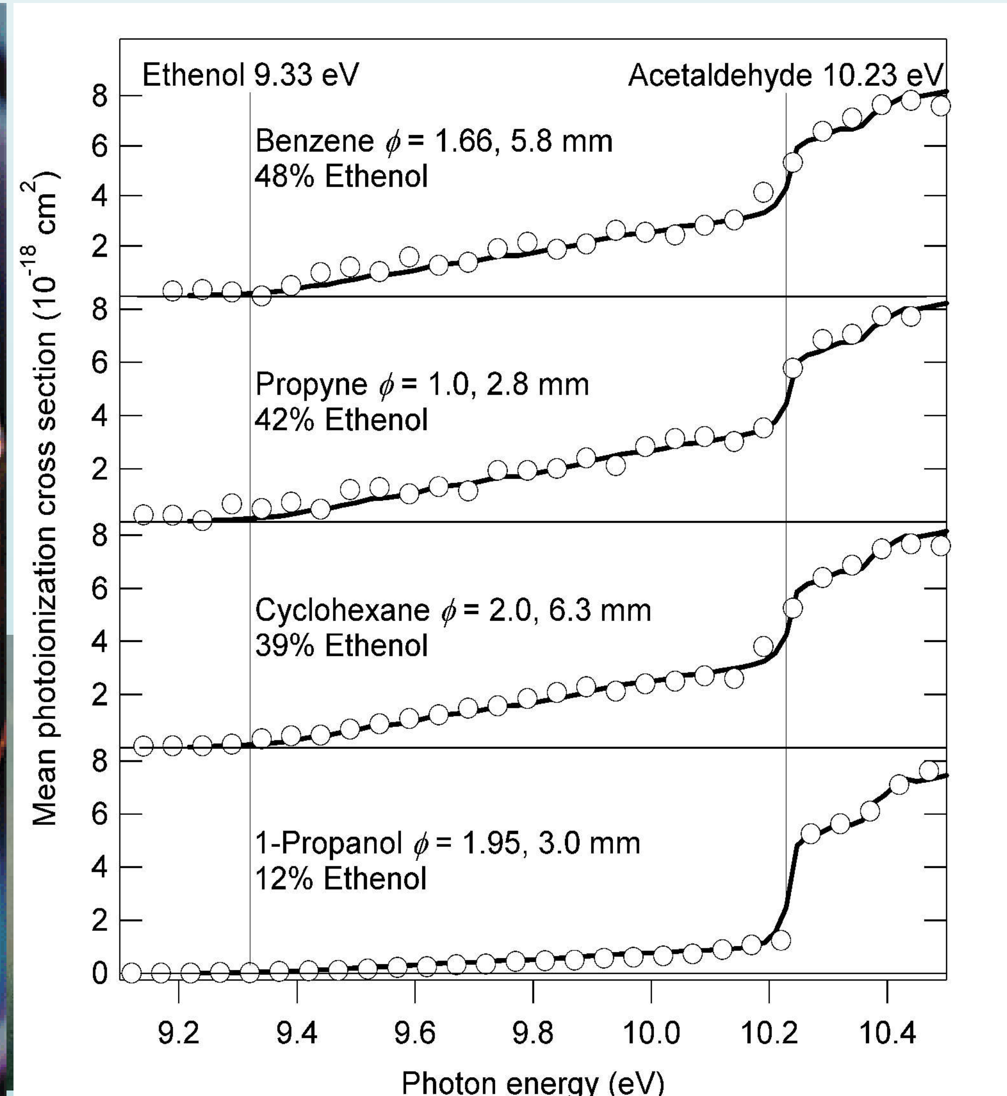
- Many reactions are driven by temperature (kT) not optical excitations
- THz pump and XPS, XES, RAIRS probe
- THz probe of H-bonding, normal modes of biomolecules.



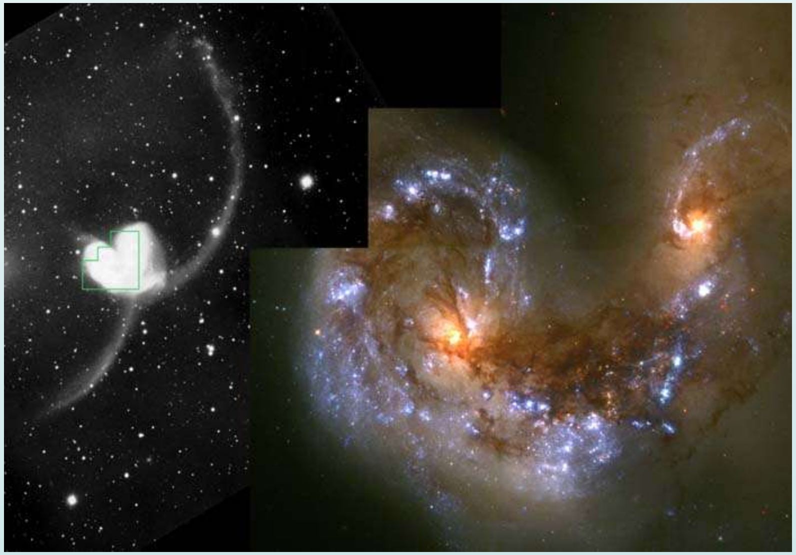
Anders Nilsson, SSRL

Reactions and processes in the biosphere



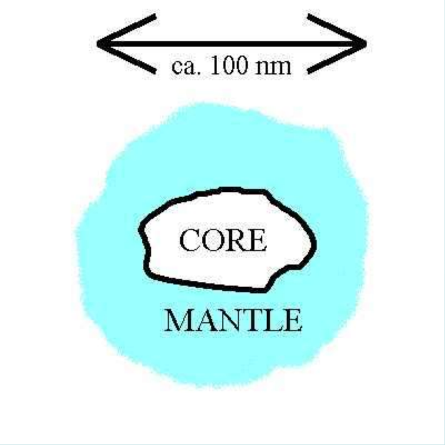


Fundamental measurements in astrophysics & astrochemistry



- studies of exotic and/or shortlived species
- key fundamental measurements on multiply charged species - remove reliance on computed parameters
- chemistry of the interstellar medium - ion-surface and gas phase interactions, formation of complex ions and molecules
- improving our understanding of the origins of the universe

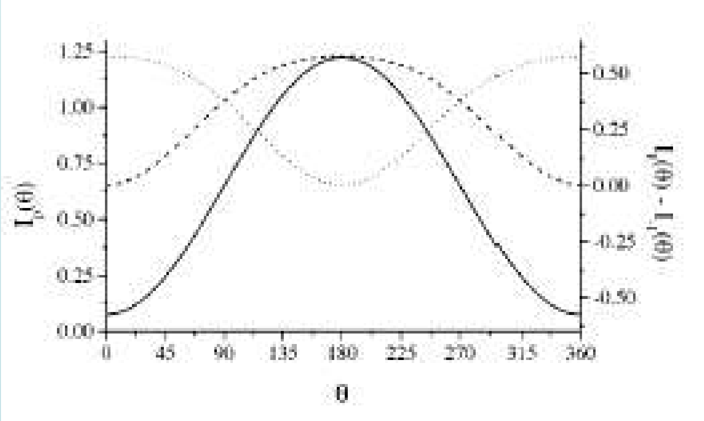
Molecular interactions on ultracold surfaces



Molecular chirality and the homochirality of life

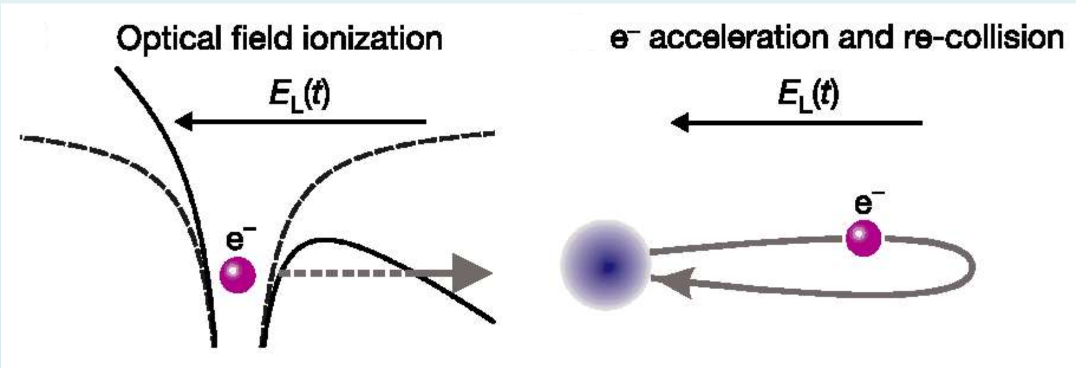
- CP light from star-forming regions is thought to have played a key role in biological evolution
- Require high quality, high intensity CP light in VUV - pulsed and CW

- CD in the angular distribution of *photoelectrons* (CDAD) from chiral molecules predicted to be *ca.* 10%!

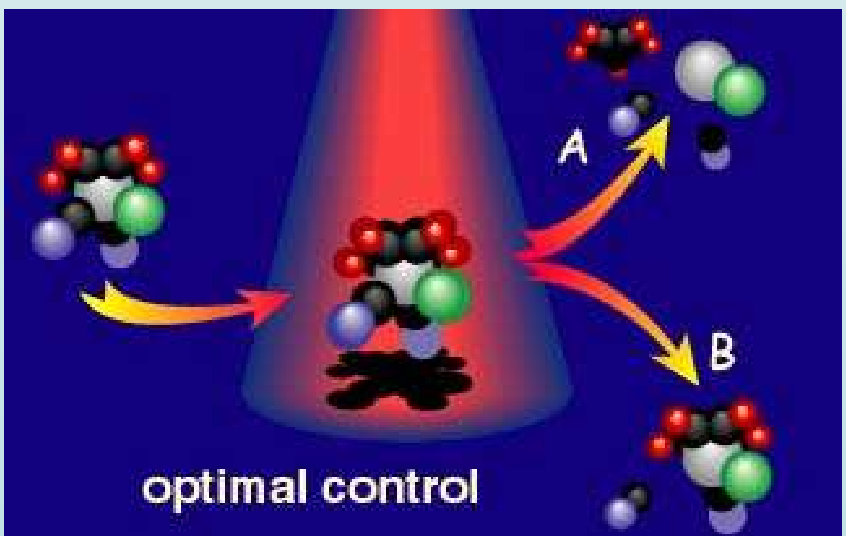


Calculated CDAD for D-glyceraldehyde
I Powis, *J Chem Phys*, **112**, 301, (2000)

Key areas: Understanding atomic and molecular dynamics - quantum chemical control

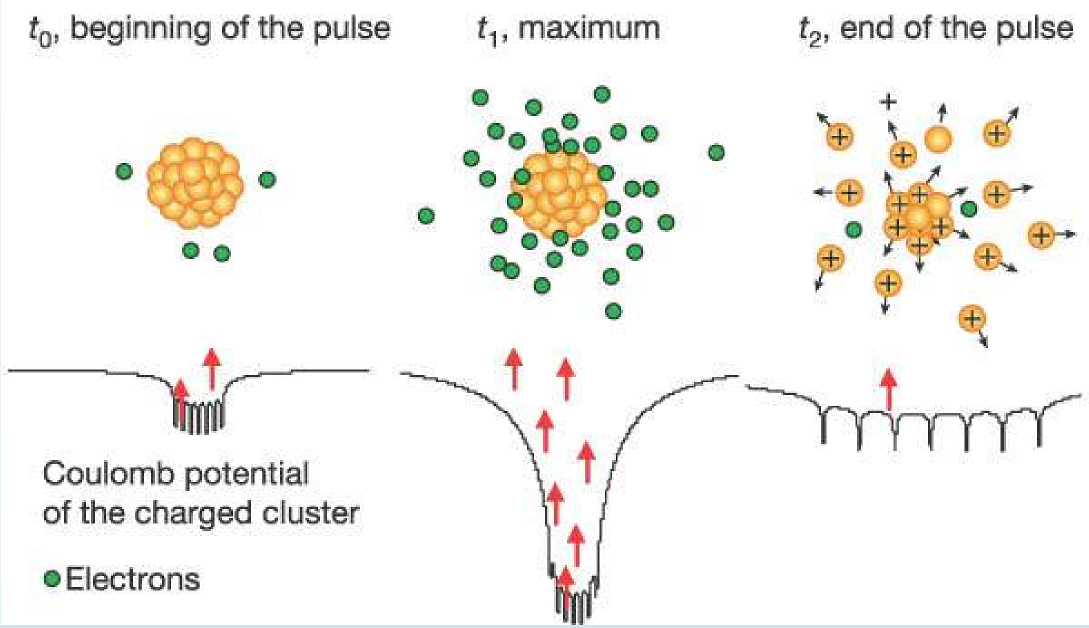


A Baltuska *et al.*, *Nature*, **421**, 611, (2003)



- Exploring the behaviour of atoms and molecules in high intensity, high field regimes
- Leading to new states of matter
- Coherent control of chemical reactions - we can control which bonds we break in a molecule, and direct the reaction.

Multiphoton excitations of atoms, molecules, clusters…….



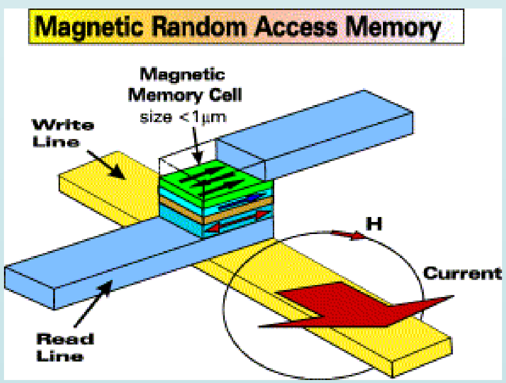
- Coulomb explosions
- Tests of theory

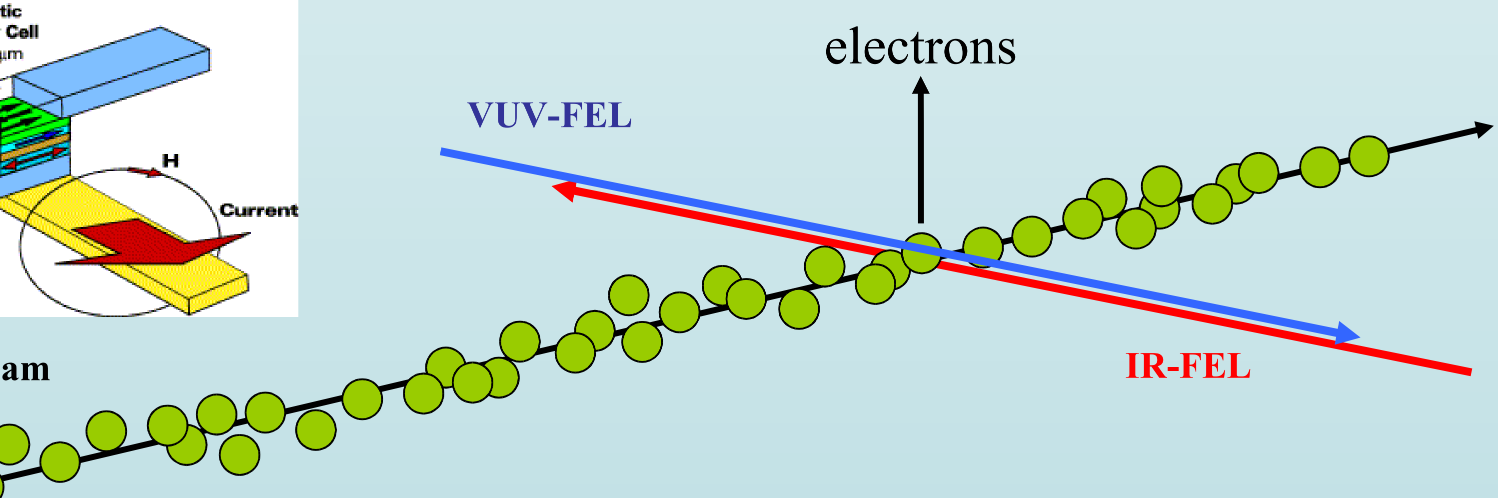
Recent first results from DESY TTF show Xe atoms undergo a Coulomb explosion in the VUV at field intensities 10x lower than predicted by existing models

(H Wabnitz et al., *Nature,* **420**, 467, (2002))

Key areas: Understanding charge and spin dynamics - new nanodevices

- The number of transistors on a Si chip doubles every few years
- Devices now so small that they are not at equilibrium when **operating**
- We need to understand how electrons and holes **move** through devices
- We need to use new approaches, e.g. **spintronics**
    - *Measurements of charge and spin dynamics in **single** nanoparticles*
    - *Next generation computers*

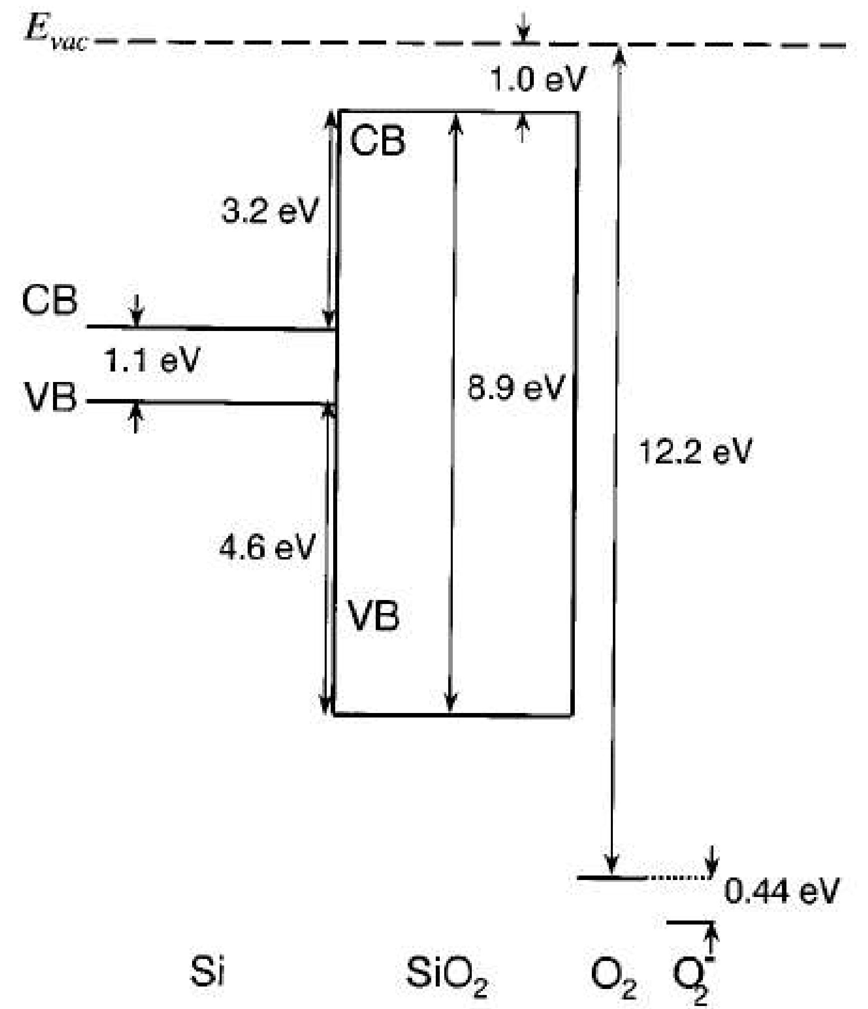




Exploitation of nanocomposites

- **Need to know:**
  - *well depth for e⁻ and h⁺*
  - *variation of band gap with size*
  - *energy level line-up*
  - *influence of defects/ adsorbates on e.g. luminescence*
  - *lifetime of excited state (exciton)*
  - *nature of charge transport between layers in microdevices*

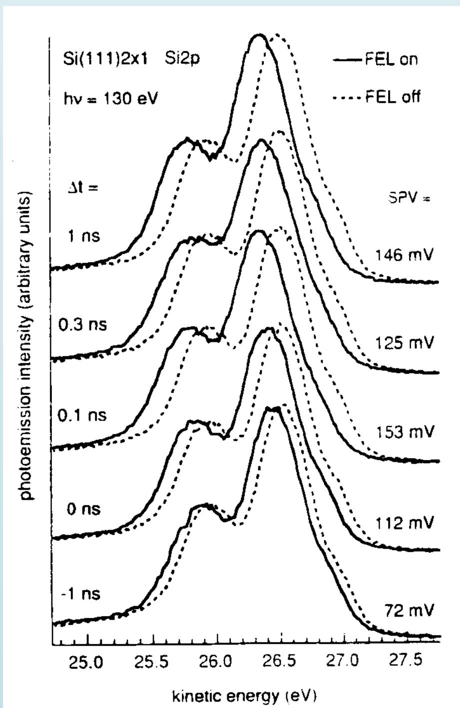
- **But for band gaps ≥UV (photovoltaics, fluorescent security tags, polymer stabilisers, sun screens,), and down to fs timescales**



J G Mihaychuk *et al., Phys Rev B*, 59, 2164, (1999)

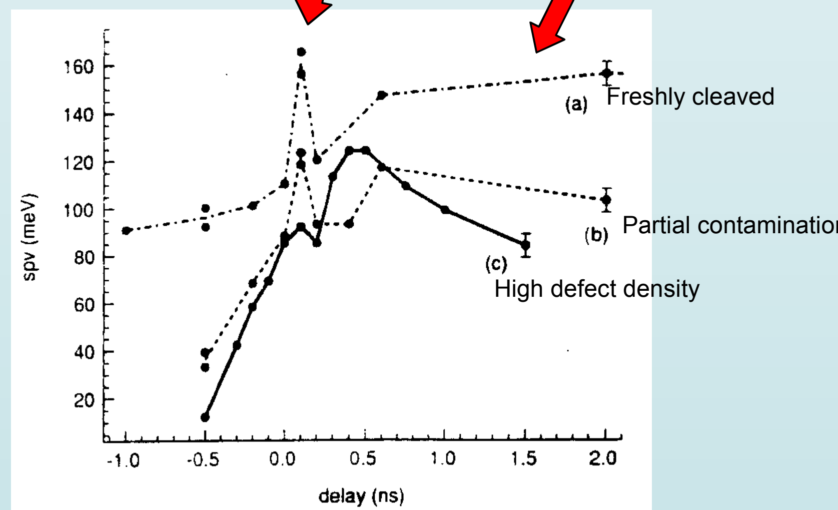
Transient charge carrier distributions measured by FEL-SR pump-probe

Sub nanosecond charge density evolution (electron/hole pair dynamics) probed with SR by monitoring the surface photovoltage effect induced by illumination of Si(111) 2x1 with FEL photons.



Oscillation due to influence of surface states

Photocarrier relaxation as per bulk



M Marsi, M E Couprie, L Nahon, D Garzella, T Hara, R Bakker, M Billardon, A Deloulbe, G Indlekofer and A Taleb-Ibrahimi , *J Electron Spectrosc Relat Phenom*, **94**, 149, (1998)

THz Pump-probe measurements of internal exciton transitions

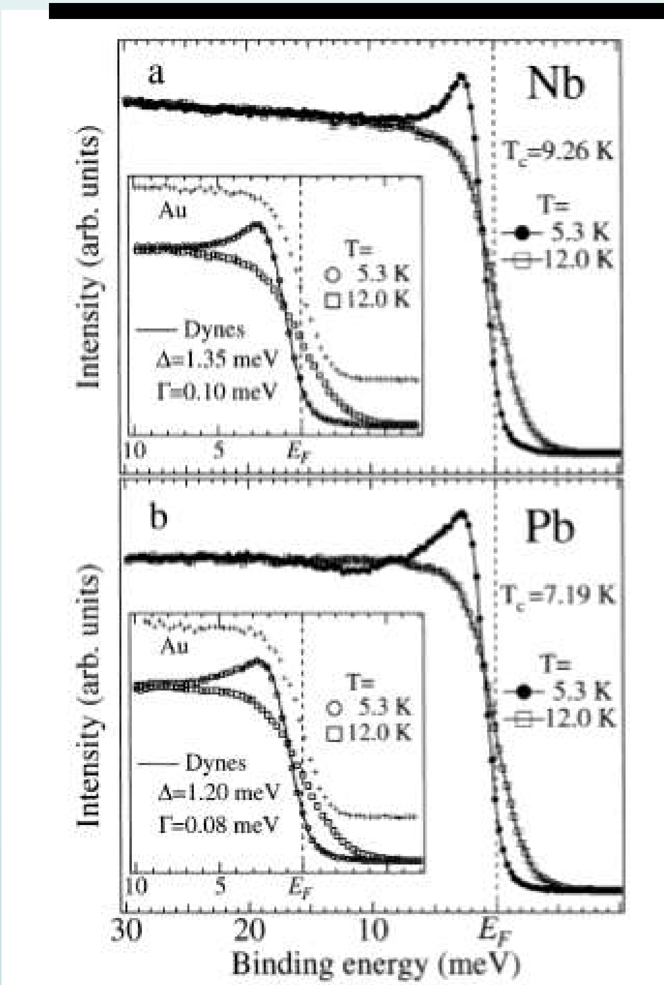
Evolution of conductivity and dielectric constant as insulating excitons form in GaAs MQWs on ps timescales

QuickTimeª and a
Photo - JPEG decompressor
are needed to see this picture.

QuickTimeª and a
Photo - JPEG decompressor
are needed to see this picture.

*Kaindl et al., Nature, **423**,734 (2003)*

Ultra-high resolution photoemission

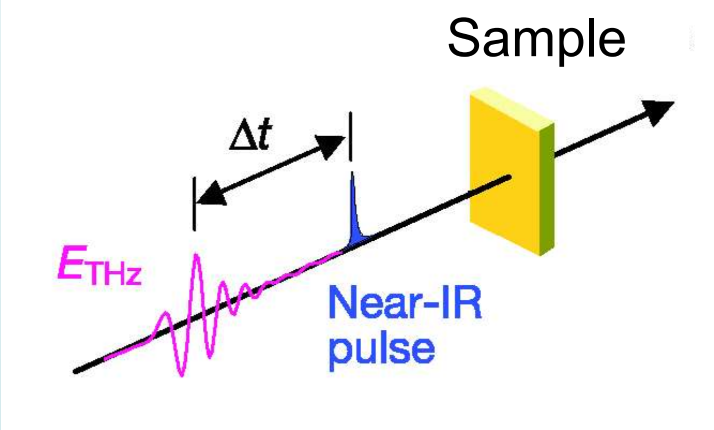


T Kiss, T Yokoya, A Chainani and S Shin, *J Electron Spectrosc Relat Phenom,* **114-116**, 635, (2001)

- Measurement of superconducting gap, anisotropy, phase diagram in HTc *and* conventional superconductors
- Mechanisms of superconductivity
- Related phenomena in correlated solids - m-nm trans., GMR, spin transitions etc.
- Chemical searchlight through resonant photoemission

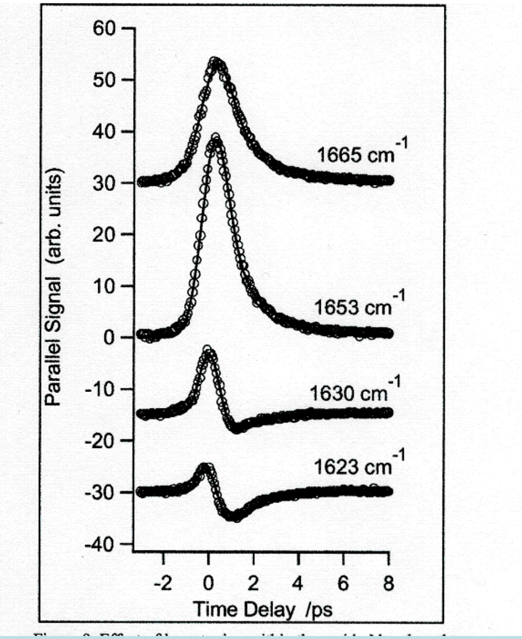
Key areas: Function of biomolecules in living systems

- Spectroscopic probes of biomolecule secondary and tertiary structure in solution, at interfaces, in membranes
- Time-resolved ‘single-shot’ measurements

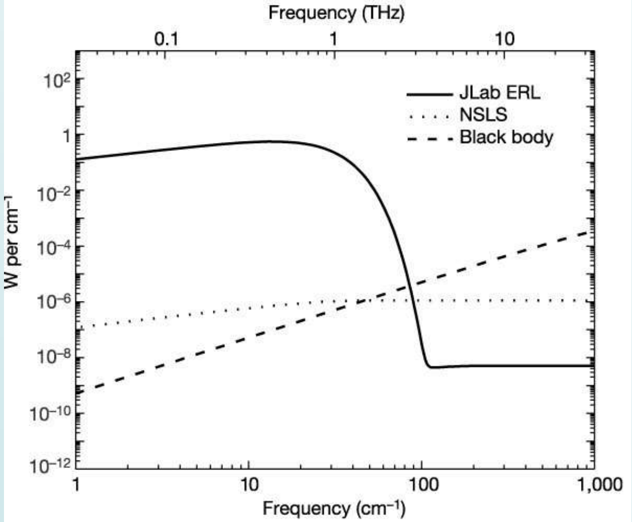


The effect of laser tuning within the amide I band on the pump-probe signal, indicative of anharmonicity in the vibration

K A Peterson *et al.*, *Proc SPIE,* **3153,**147, (1997)



Intramolecular vibrational relaxation of surface complexes



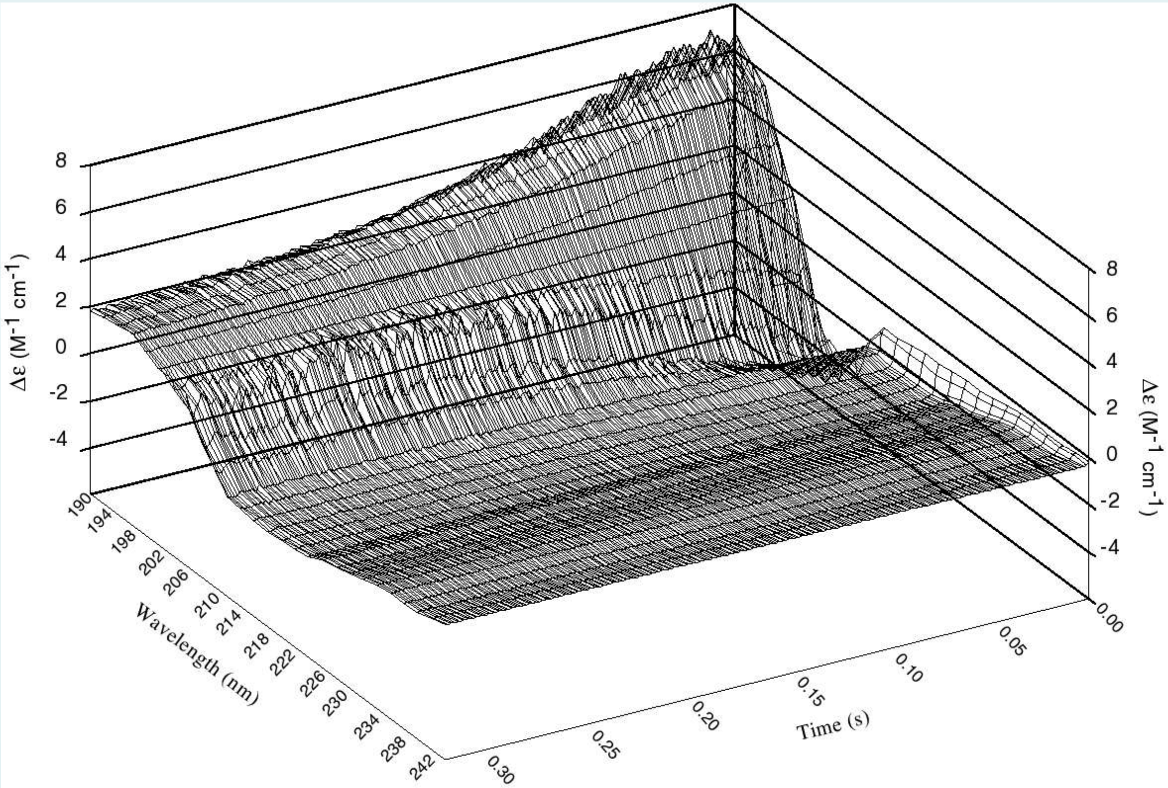
*The **enormous** power gains achievable in the far - IR using an ERL source*

G L Carr *et al, Nature*, **420**, 153, (2002)

- surface dynamics of large adsorbed molecules
- essential to improving understanding of adsorption, reaction and desorption at any catalyst surface
- *low frequency* surface-adsorbate modes
- Spatial and temporal overlap of FEL and BM beams in RAIRS geometry

UV CD

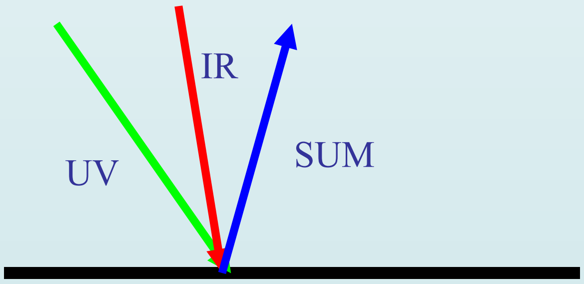
- UV -CD: chirality of protein secondary and tertiary structure
- UK has international profile (through work at SRS)



Stopped-flow SRCD, β-lactoglobulin folding, G Jones

- work possible on few ng quantities of proteins (10mg currently)
- Time-resolved work into ps regime - one-shot expts. close?

Double resonance Sum Frequency Spectroscopy and imaging: New Horizons



IR $\lambda > 10\ \mu m$
UV, tune to optimise non-resonant interaction

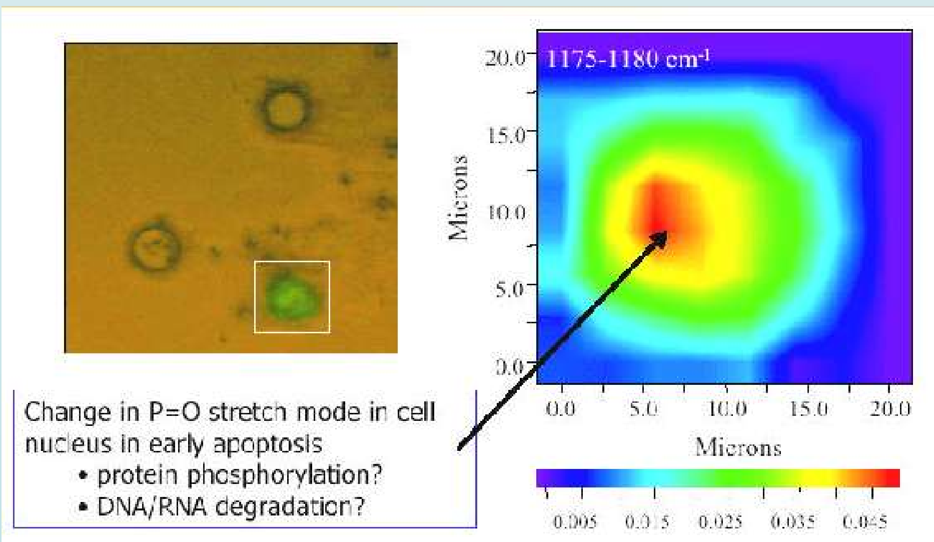
- Conformation of large admolecules
- Adsorbate dynamics
- Membrane rafts & proteins, lipid bilayers, oxide catalysts……….
- Exploits tuneability of BOTH FELS
- Enormously widens range of surfaces and vibrations

- Imaging applications using near-field SFG signal (SNOM probe): below diffraction limit

Key areas:nanoscale dynamic imaging - new approaches to early diagnosis

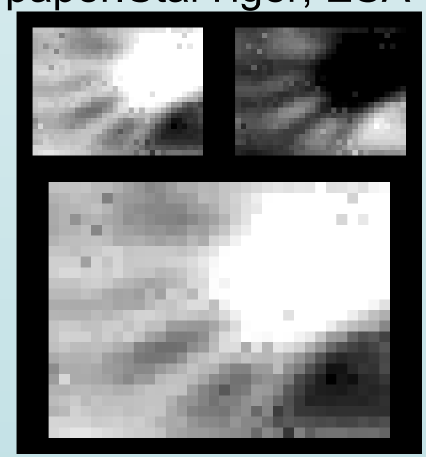
- intercellular signalling, receptor systems on membrane rafts
- functional imaging in live cells, effects of biomolecules/pollutants, *in vivo* study of radiation damage
- material-biological matrix interface, surface nanostructuring
  - *New techniques to improve early diagnosis of disease*

Cell changes during apoptosis (P Dumas, SR IR, LURE)



- Overcome diffraction limit using near-field imaging/IR FEL: 30-50 nm resolution

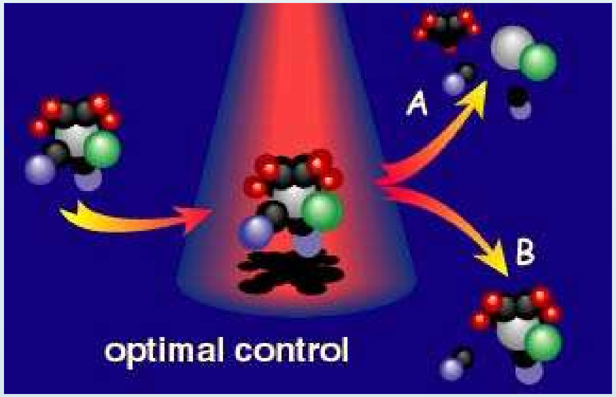
THz image of a hand taken through 15mm of paper.StarTiger, ESA



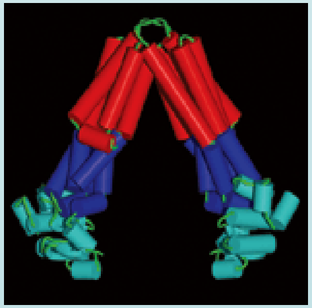
- ERLs are the world’s most intense THz sources (10’s W output)

Summary of key impact areas

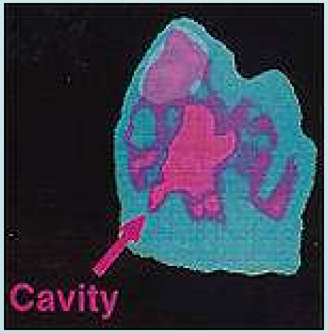
- Understanding catalytic reaction pathways
- Understanding atomic and molecular dynamics - quantum chemical control



- Understanding charge and spin dynamics - new nanodevices

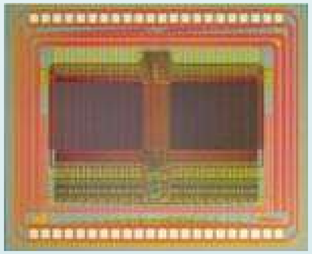


- Understanding the function of biomolecules in living systems



- Nanoscale dynamic imaging - new approaches to early diagnosis

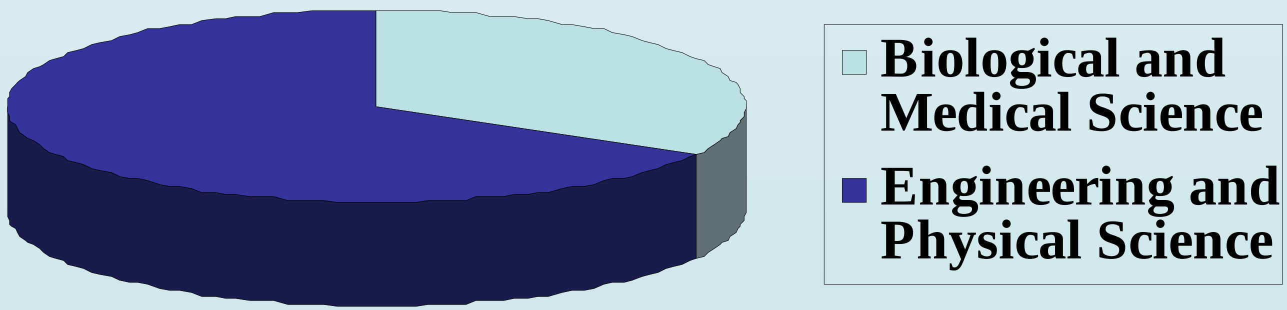
The user community



EPSRC:
nanotechnology, dynamics in microdevices

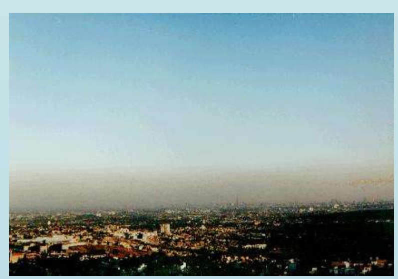
MRC:
medical imaging

PPARC:
astrophysics



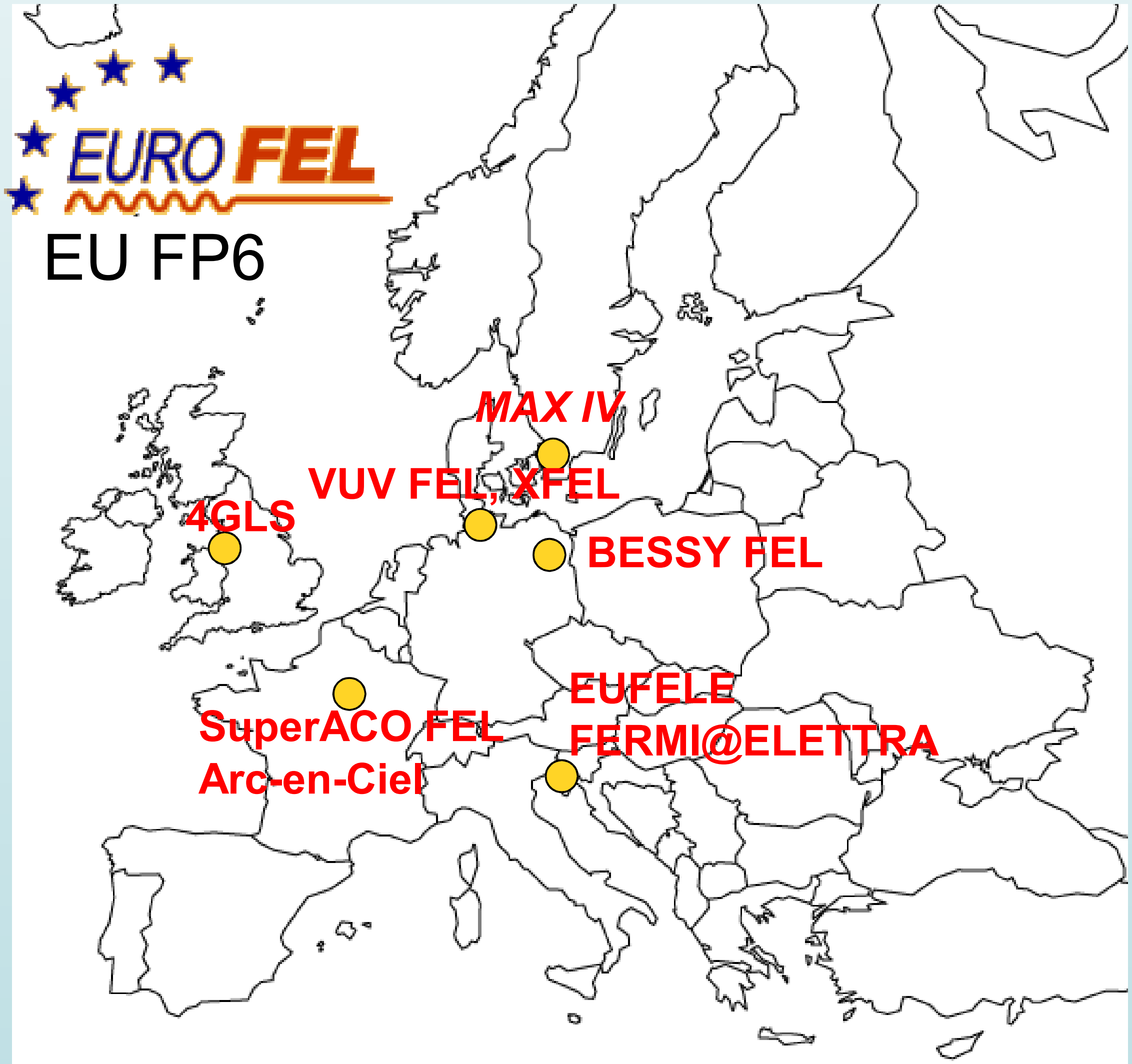
Estimated user base *ca.* 1000,
> 300 already actively engaged

NERC:
atmospheric reactions, bioremediation



BBSRC
biomolecular dynamics

The international picture



International development programmes:

- JLab
- Cornell
- Stanford
- Rossendorf
- DESY
- Fermi@ELETTRA
- EUROFEL

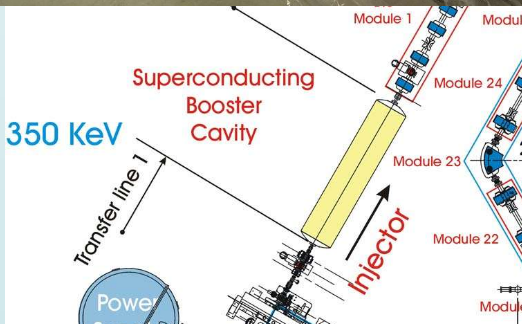
elettra

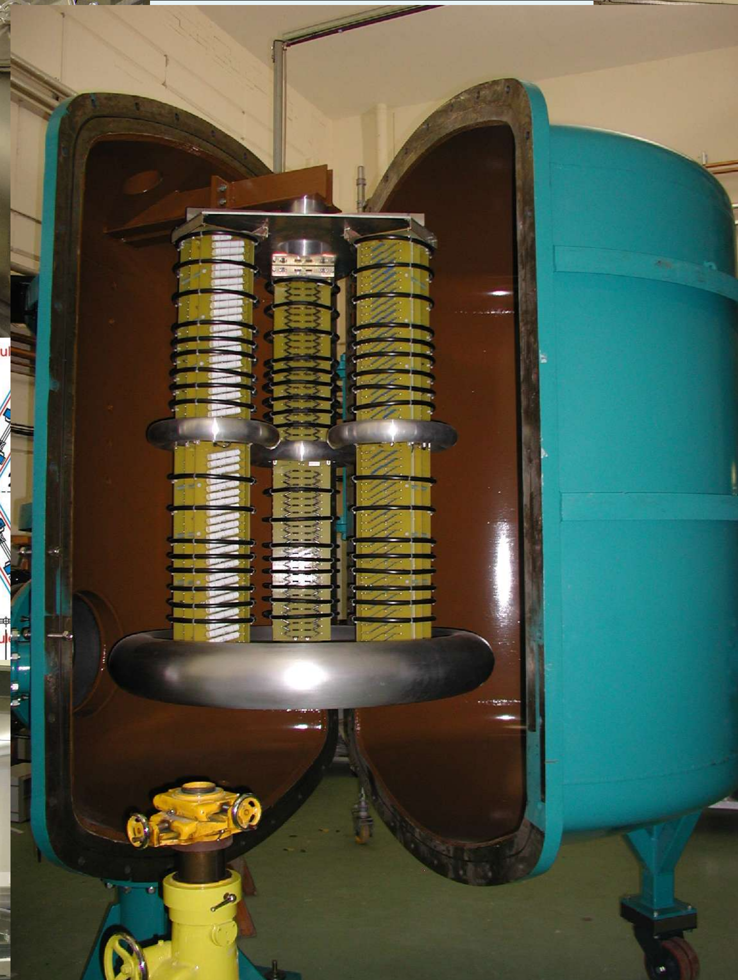
Exploring the Nature of Matter
Jefferson Lab

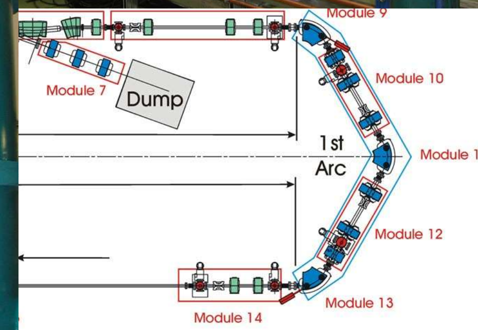
4GLS: timescales

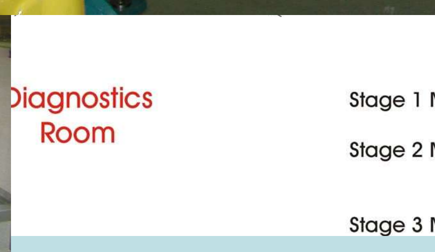
✓ **April 02**	Scientific case approved (Gateway 0)
✓ **November 02**	Business case approved (Gateway 1)
✓ **April 03 & 04**	£13.9 M funding for prototype accelerator (ERLP) and R&D (OST £8 M, CCLRC £5.9 M)
✓ **Feb 05**	EUROFEL R&D work funded (Euro 9M)
✓ **March 05**	Funding for 4GLS Technical Design (CCLRC £1.6 M)
✓ **Dec 05**	£3 M NWSF funding for ERLP science
▪ **Start 07**	Prototype operational, report on 4GLS phase I
▪ **2007 ??**	Approval for 4GLS procure and start build
▪ **2011 ??**	Facility available to researchers

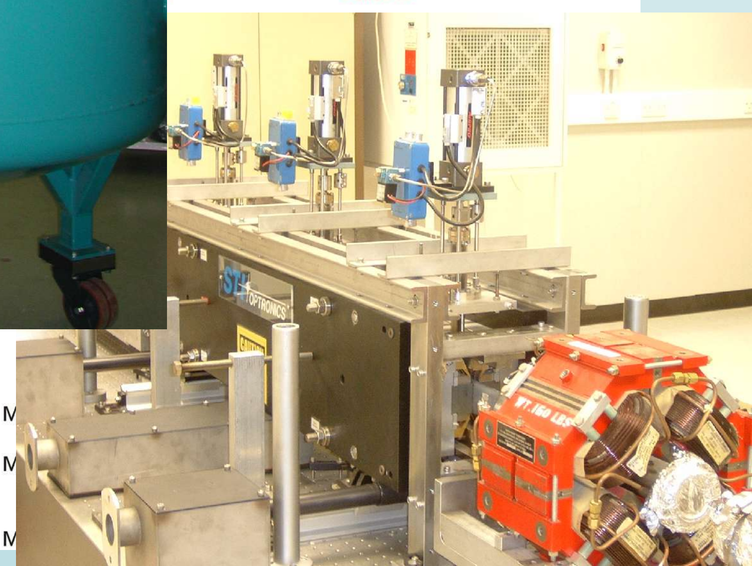
ERL Prototyp



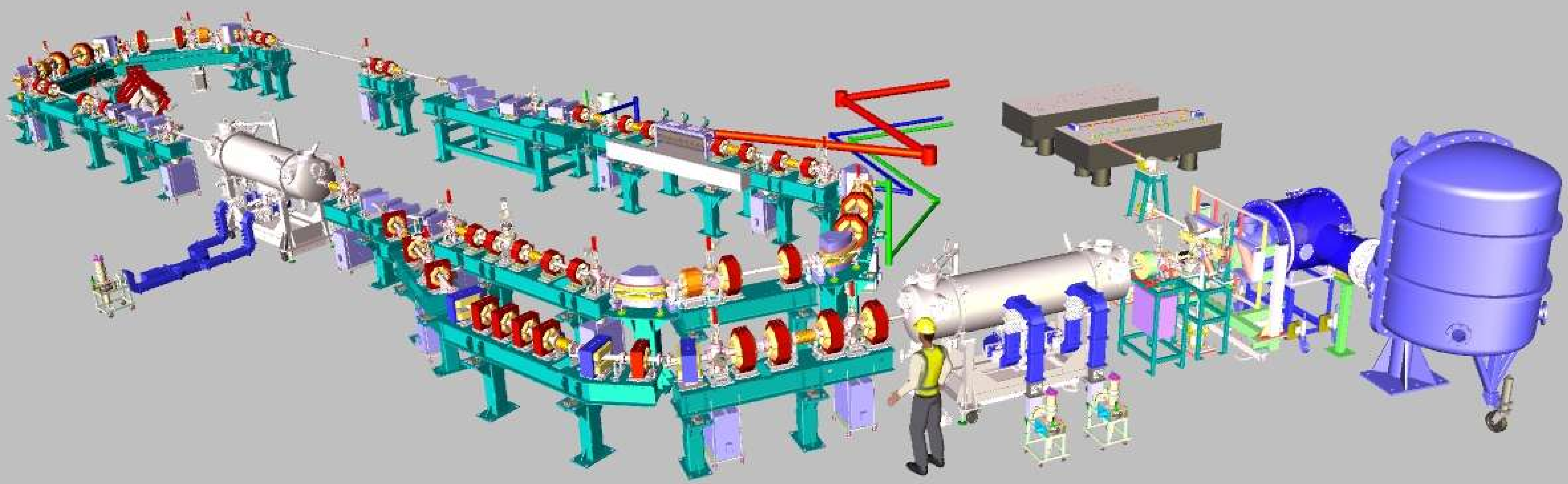




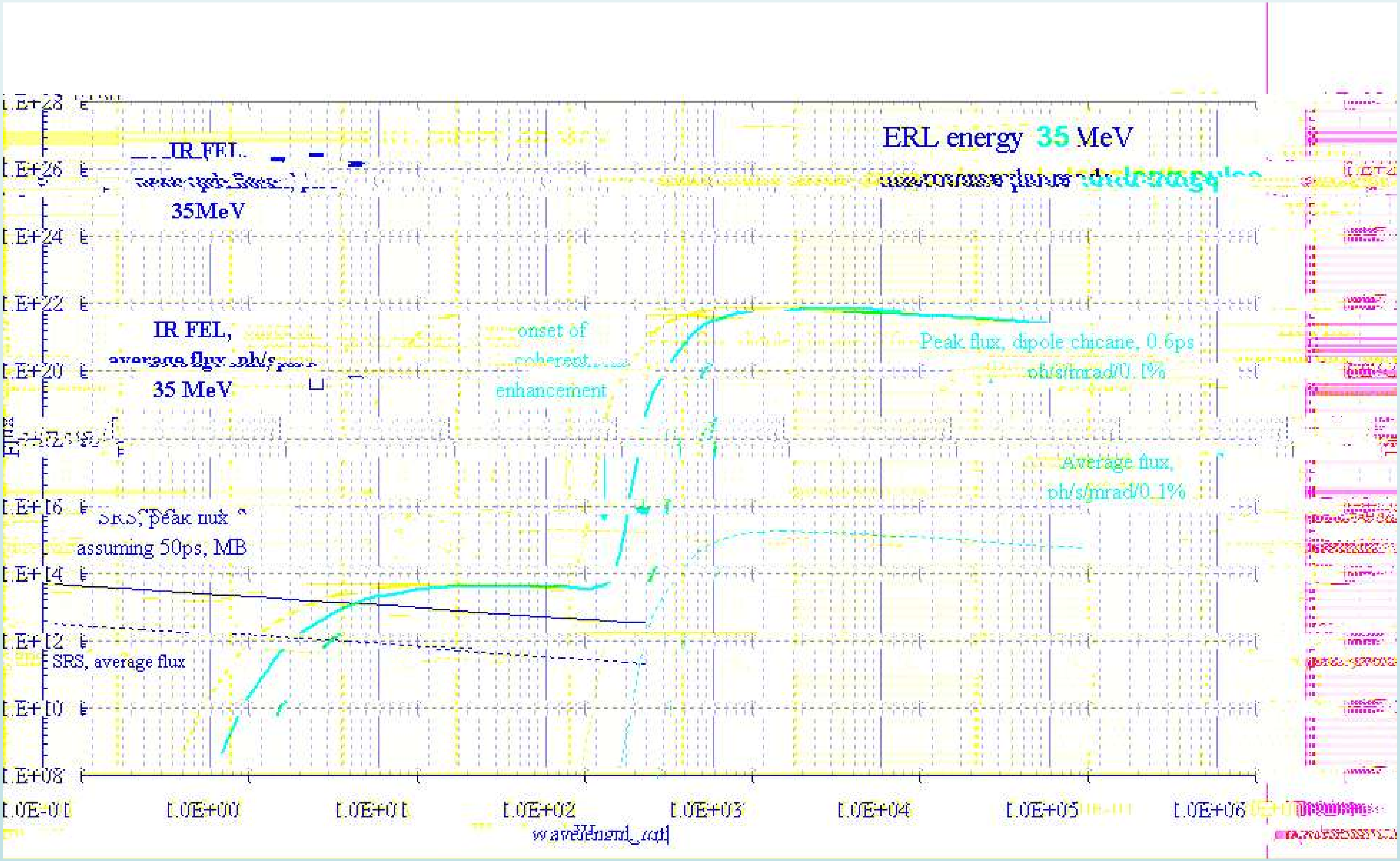




ERLP Nov 05

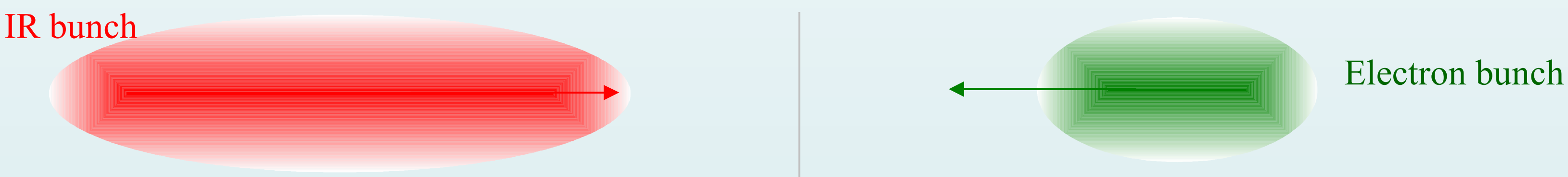


ERLP output

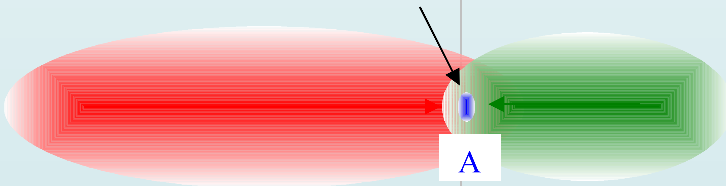


ERL energy 35 MeV
IR FEL,
35MeV
IR FEL,
35 MeV
onset of
coherent
enhancement
Peak flux, dipole chicane, 0.6ps
ph/s/mrad/0.1%
Average flux,
ph/s/mrad/0.1%
assuming 50ps, MB
SRS, average flux
1.E+28
1.E+26
1.E+24
1.E+22
1.E+20
1.E+16
1.E+14
1.E+12
1.E+10
1.E+08
1.0E-01
1.0E+00
1.0E+01
1.0E+02
1.0E+03
1.0E+04
1.0E+05
1.0E+06

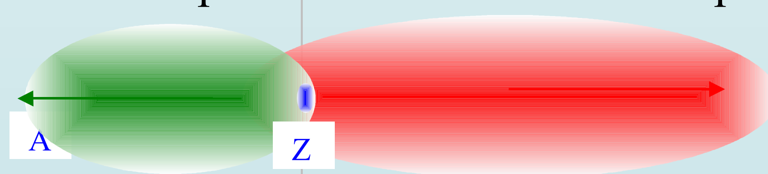
IR-electron bunch collision



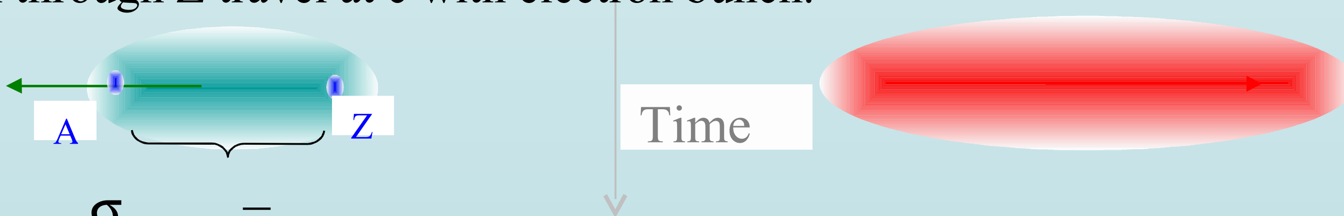
First x-rays of collision produced when bunches 1st meet.



Last x-rays of collision produced when bunches separate.



X-rays A through Z travel at c with electron bunch.



$\sigma_{\text{x-rays}} = \sigma_{\text{electrons}}$

$$h\nu_X = h\nu_L 2\gamma^2 \frac{(1-\cos\phi)}{(1+\gamma^2\theta^2)}$$

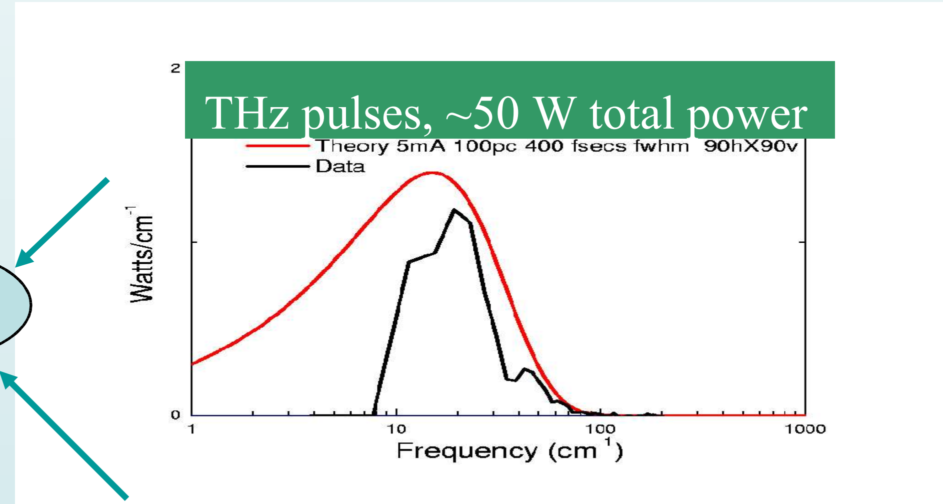
Courtesy George Neil, JLab

Simultaneous production of 3 μm, THz, and 10 keV X-ray femtosecond pulses

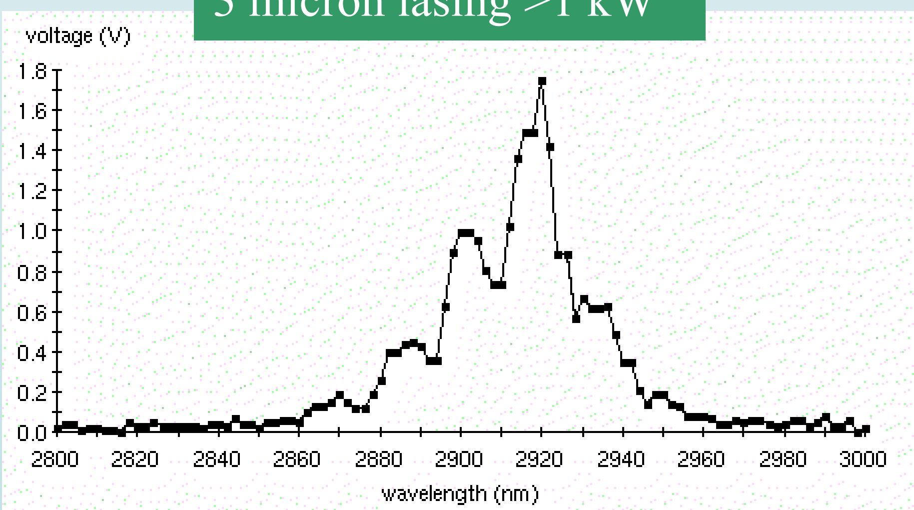
- **800 fsec pulses at 37.4 MHz**
- **Synchronized to <<psec levels (same beam!)**
- **All three wavelengths at world class fluxes**

Sample

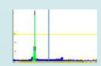
THz pulses, ~50 W total power



3 micron lasing >1 kW



10 keV X-ray > 10^5 ph/sec/0.1% BW

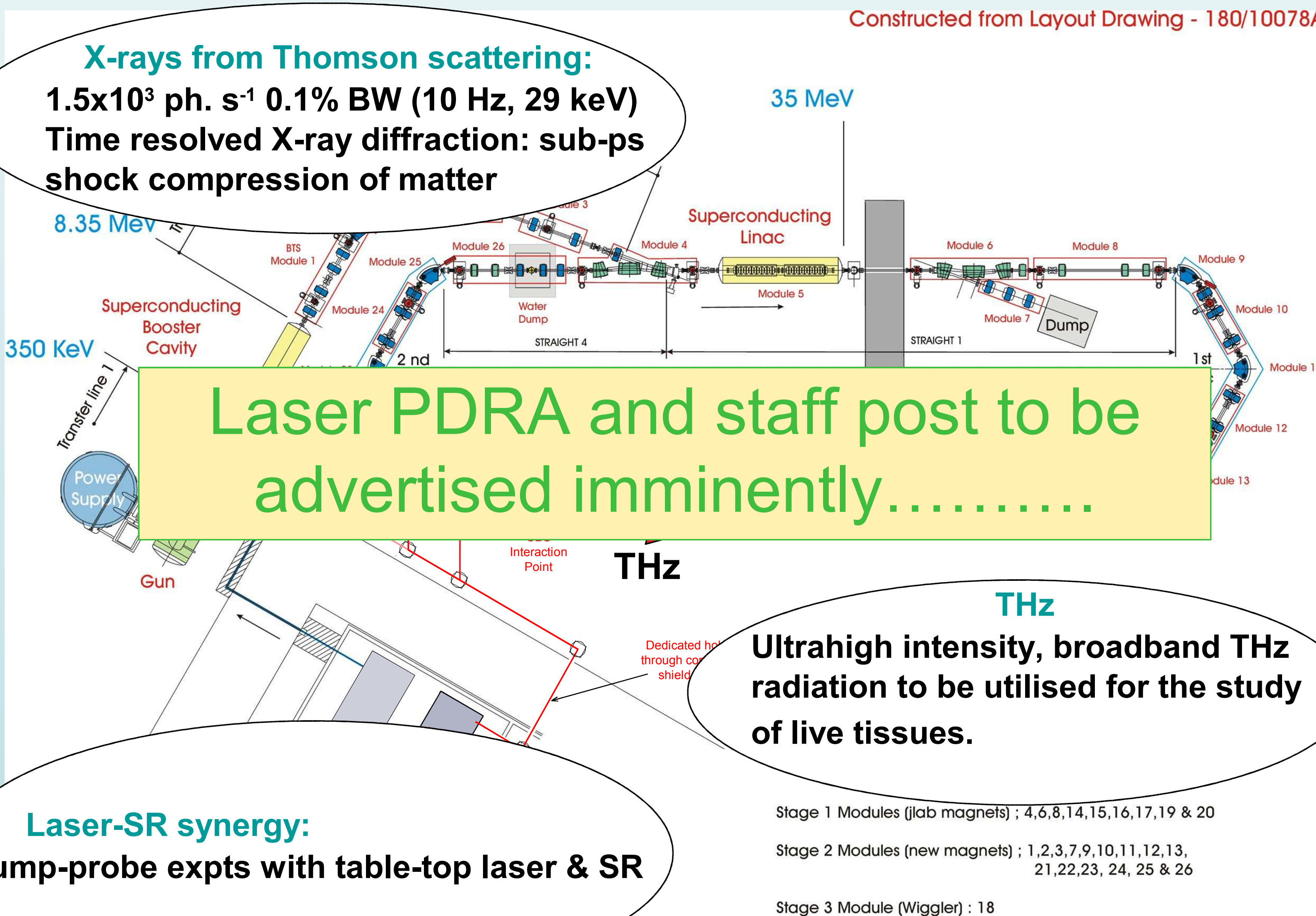


0 16 32

X-ray energy (keV)

Courtesy George Neil, JLab

NW Science Fund Award £3 M, 3 years from Dec 05



X-rays from Thomson scattering:

1.5x10³ ph. s⁻¹ 0.1% BW (10 Hz, 29 keV)
Time resolved X-ray diffraction: sub-ps shock compression of matter

Laser PDRA and staff post to be advertised imminently……….

THz

Ultrahigh intensity, broadband THz radiation to be utilised for the study of live tissues.

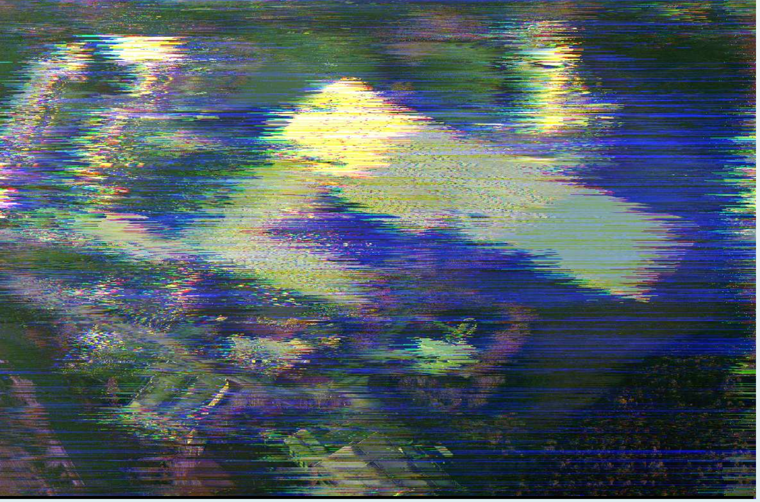
Laser-SR synergy:

Pump-probe expts with table-top laser & SR

In summary...

4GLS combines superconducting ERL, SR and FEL technology in a fully integrated multi-source facility

- unique capability in pump-probe experiments
- THz - soft X-ray
- science programme complementary to XFEL and LCLS
- multiuser facility
- cost-effective delivery (*ca. £180 M)*
- substantial user benefits of ERL e.g.
  - infinite beam lifetime and short pulse SR sources
  - flexible control of pulse structure
  - ERL symmetrical beam and small emittance
- easy upgrade paths and additions e.g.
  - recirculate beam for hard X-rays
  - Thomson scattering
- ERLP is Europe’s most intense broadband THz source



4GLS

LIGHT YEARS AHEAD

Acknowledgements

- The 4GLS Team
- The 4GLS International Advisory Committee
- The 4GLS Steering Committee
- All contributors to the Science and Business Cases
- Funding: OST/DTI, CCLRC, NWDA and EU

Further information

http://www.4gls.ac.uk

[redacted]

[redacted]