Next Generation
Advanced Light Source
Science

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Measuring in real time

What if all you saw of a race was this bit? Would you learn anything?

This is our problem with most current SR research - reactions happen so fast we only see the products.

Next generation light sources enable us to watch chemical reactions as they happen (on fs timescales), even in nanoclusters of material.
The science need

The fundamental requirement is to understand the dynamic behaviour of matter, often in very small (nm) units, on very fast (fs) timescales.

We need not just to determine structure with high precision, but to understand how these structures work.
Energy recovery in a linac

Some world ERL light source projects:
- JLab (US)
- JAEA (Japan)
- BINP (Russia)
- ERLP (UK)
- CHESS (US)
- KEK (Japan)
- 4GLS (UK)
- BNL (US)
- APS? (US)

pulse lengths ca. 100 fs
(storage ring ca. 10’s -100 ps)
The 4GLS prototype, ERLP takes shape

March 2007
FLASH

status
operational with users
has achieved 13.8nm, 40μJ pulse

Repetition rate up to 150 Hz
Photon range 45-15 nm
$10^{13}$-$10^{14}$ ph/pulse
Pulse duration 10-50 fs

J. Schneider, DESY
Some planned X-ray FELs

LCLS (SLAC)
XFEL (DESY)
SCSS (SPring-8)
Linac-based light sources

- **Peak Brightness**: (ph/s/0.1%BP/mm²/mrad²)
- **Pulse Length**: (ps)

### Sources
- **SLS**
- **DLS**
- **SRS**
- **ESRF**
- **ERL Spontaneous SR Sources**
- **4GLS**
- **Cornell**
- **ALS**
- **SLS**
- **ALS**
- **FERMI@Elettra**
- **LCLS**
- **XFEL**
- **4GLS**
- **XUV-FEL**
- **Slicing on 3rd generation SR Sources**
A step change

The advent of world 4th generation sources will bring a step change in brightness in the VUV/XUV and X-ray regimes (and THz too!)

…..and the pulse lengths are similar to the time it takes to break a chemical bond
Dynamics and kinetics

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

TESLA technical design report, March 2001
Pump-probe:
understanding reaction pathways

- real-time catalytic reaction monitoring
- we can study dilute, rare or shortlived species and their reactions

(Anders Nilsson and colleagues, SLAC)
Pump-probe at FLASH

2-colour ionisation of Xe and He;
20 fs XUV (45.8, 89.9 eV),
5 Hz;
120 fs 800 nm Ti:sapph

Single-shot spectra

Photoemission sidebands as a pulse diagnostic

Energy and climate change

How do we combust hydrocarbons more efficiently?

Our environment

Reactions and processes in the biosphere

How do we clean up our environment?

What are the reaction pathways of the free radicals and ions contributing to atmospheric pollution?

pollutant creation & removal

What are the mechanisms of bioremediation?

environmental effects of toxins (atmospheric, inorganic, fungal) on photosynthetic systems
Cleaner catalysts by design

How do we design cleaner, more efficient catalysts?

Can we understand and optimise the synthesis of chiral pharmaceuticals?

- Studies of enantiomer-selective chemistry
- Need to understand asymmetric reaction pathways: key to improving turnover number

Asymmetric hydrogenation
Courtesy William Hems, Johnson Matthey Catalysts - Chiral Technologies
How do stars form and work?
How did life originate?

Fundamental reactions in the interstellar medium

- 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, molecular interactions on ultracold surfaces
- Interactions of biomolecules with intense CP VUV light; homochirality of life
- improving our understanding of the origins of the universe
Mode-selective control of chemical reactions

Desorption of hydrogen by resonant excitation of the Si-H vibrational stretch

Liu, Feldman, Tolk, Zhang and Cohen, Science 312 1024, 2006

Reaction diverted from thermal pathway

Potential impact on:
- the storage, transport and delivery of hydrogen for the hydrogen economy
- reactive chemistry on surfaces
Can we *control* the direction of a chemical reaction?

First observation of real-time vibrational wavepacket interference  Kiyoshi Ueda *et al.*, *PRL* 96 093002 (2006)
Can we understand electron correlation?

Exploring the behaviour of atoms and molecules in high intensity, high frequency field regimes

XUV field intensities $10^{16} - 10^{17}$ Wcm$^{-2}$

Providing data for new theory development
Multiphoton excitations of atoms, molecules, clusters...

- Coulomb explosions
- Tests of theory

First results from FLASH show Xe clusters 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), T Laarmann et al., PRL, 95, 063402 (2005))
One-shot experiments!

Simulated coulomb explosion of a T4 lysozyme molecule caused by a $3 \times 10^{12}$ photon per (0.1mm)$^2$ pulse of X-rays

A tool for plasma physics

Generation and study of extreme states of matter
- Temperatures up to $10^7$ K
- Pressures up to Gbar
- Improved models of stellar formation
- Generation of dense plasmas
- Studies of warm dense matter

http://www.t4.lanl.gov/CECAM/

Density-temperature diagram for astrophysical objects
TESLA technical design report, March 2001
Time-resolved diffraction

- structural changes, phase transitions, irreversible changes, cluster vibrations
- via pump-probe approach
- e.g. surface melting transitions
- study the disorder emerging within 100 fs timescale
Single molecule diffraction

Calculated scattering patterns of a single large biomolecule, Rubisco, with 15% damage-induced errors

XFEL technical design report 2nd draft 2006
Diffractive imaging at FLASH

FLASH 32 nm, 25 fs, $4 \times 10^{14}$ Wcm$^{-2}$, single shots, 3 μm structure on 20 nm-thick SiN film

Can we measure the mechanisms of energy, electron, proton and chemical transport at the cell membrane *in real time*?

How does energy transfer around a biomolecule and between a biomolecule and a substrate?
How do enzymes achieve high catalytic rates?

H-transfers can happen $10^{15}$ times faster than available theory predicts!

Small-scale promoting vibrations/motions may promote H- and electron transfer by quantum tunnelling mechanisms.


N Scrutton, M Sutcliffe, P Gardner, G Williams
EPSRC Life Science Interface funding
Human health

How do we diagnose disease (such as skin cancer) earlier and improve treatment?

How are cells damaged and repaired?

How do cells signal in the extracellular matrix? What is the action of a drug?

How are wounds healed and bones repaired?

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

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

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

THz diagnosis of basal cell carcinoma, Teraview
Breaking the diffraction limit; imaging at subcellular resolution with an IR FEL

Distribution of functional groups in a single cell
IR SNOM: resolution = $\lambda/30$
AFMIR: resolution = $\lambda/100$

$\lambda = 6.1 \, \mu m$ amide C=O stretch band
$\lambda = 6.45 \, \mu m$ reflection of sulphur, key component of amino acids
$\lambda = 6.95 \, \mu m$ sulfide cell growth medium stretch band
$\lambda = 7.6 \, \mu m$ $-\text{CH}_3$ stretch band
$\lambda = 8.05 \, \mu m$ phosphorus stretch band, component of DNA and RNA

A Dazzi et al., Optics Letters, 30, 2388 (2005)
A Cricenti, Biophysical J., 85, 2705 (2003), Vanderbilt FEL
Understanding carrier dynamics - developing new nanodevices

What happens after CMOS? 
by 2010, there will be 21 atoms in the gate legs of a transistor

How do carriers move in devices? 
we can no longer easily predict where they are

How do we make more efficient optoelectronic nanomaterials, photovoltaics, high k dielectrics?
How do we combine semiconductor technology and magnetism?
How does electron spin transport across a boundary?
Can we manipulate spin on fast timescales?

Pools of spin polarised electrons in GaAs probed using 100 fs pulses of 1.5 eV CP light
(D D Awschalom et al., Scientific American, 286, 53, (2002))
SR&FEL: more than the sum of the parts

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

- spontaneous linac sources
- FELs
- FELs and spontaneous sources

- 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
Summary

- 4th generation sources open up completely new science vistas
  - with huge potential for dynamics and imaging of nanoscale objects

- complementary to 3rd generation SR sources
  - primarily giving dynamic information, much higher brightness and shorter pulse length

- complementary to tabletop laser sources
  - superb coverage in THz, VUV, planned extensions to XUV and X-ray

- they bring together the SR and laser communities
  - resulting in a ferment of scientific excitement!

- we need talented accelerator scientists to deliver them
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4GLS-New Science with Next Generation Light

An information and interaction meeting for potential users

Daresbury Laboratory
Friday 6th July 2007

Invited speakers:
Marc Vraikling (AMOLF, Amsterdam, Netherlands),
Christian Bressler (EPFL, Lausanne, Switzerland)
Jean-Michel Ortega (CLIO, Université Paris-Sud, France),
Kevin Kubarych (University of Michigan, USA)
Richard Callow (Royal Institution of Great Britain, and University College London, UK),
Jon Marangos (Imperial College London, UK)

- The purpose of the meeting is to inform, and consult with, potential users on the evolving science programme and design of 4GLS, as work on the technical design of 4GLS progresses.
- A number of international experts will give presentations describing the key science that will be achieved.
- Discussion sessions will ensure that the evolving aspirations of the user community continue to be met as the detailed design parameters are confirmed.
- There will be an opportunity to visit the 4GLS prototype, ERLP.

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