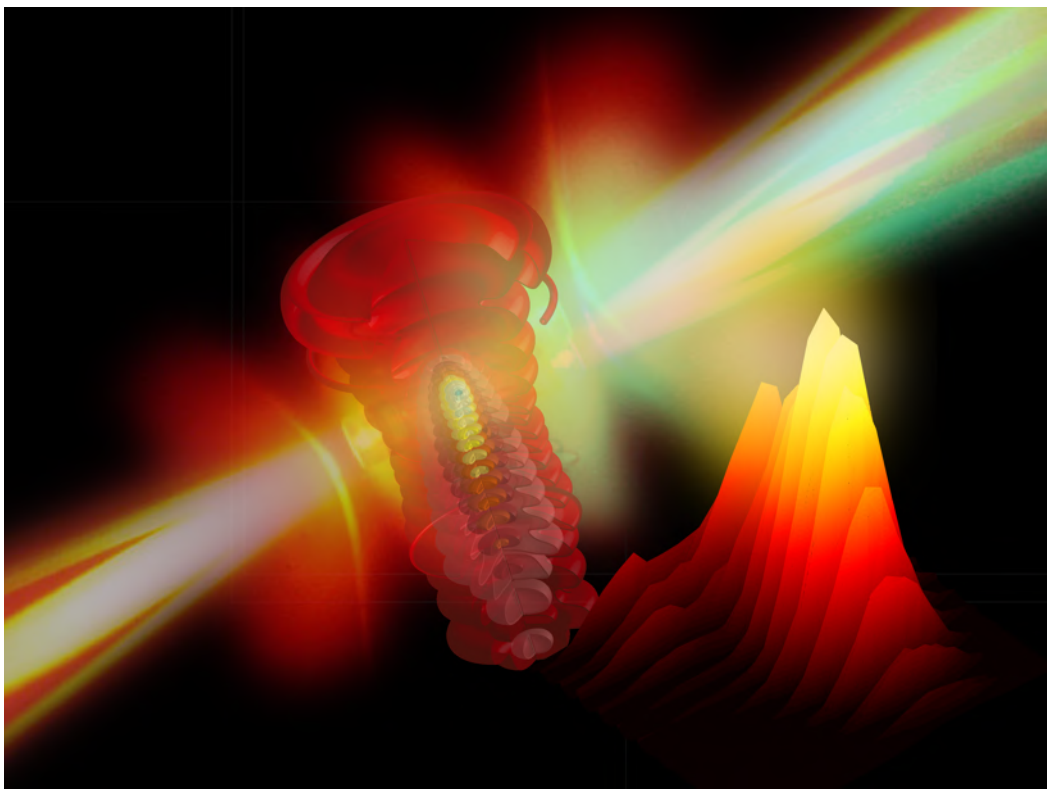
Coherent X-Rays from Ultrafast Mid-IR Lasers for Applications in Nanotechnology

Margaret Murnane and Henry Kapteyn



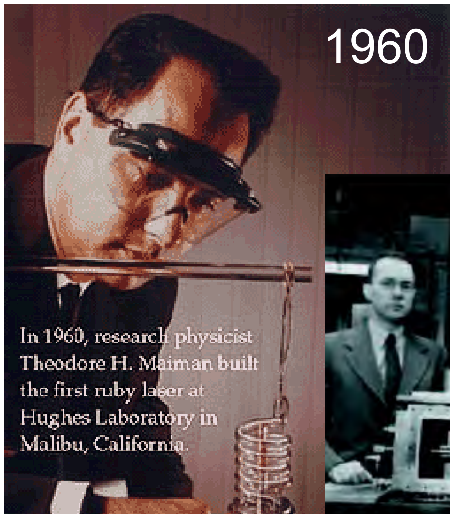
NSSEFF
NATIONAL SECURITY SCIENCE & ENGINEERING FACULTY FELLOWS

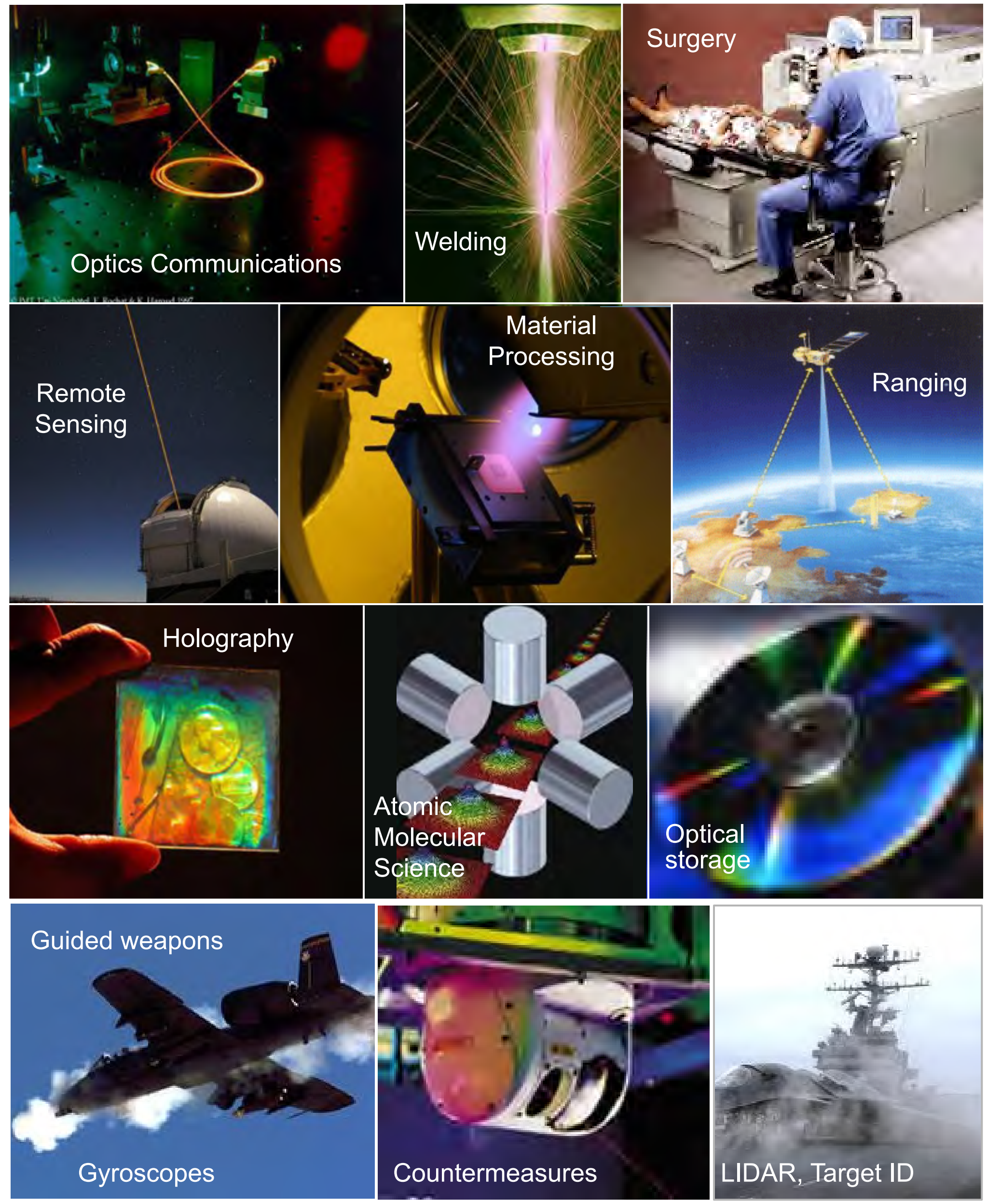
National Security Science and Engineering Faculty Fellowship Competition

7TH ANNUAL DISRUPTIVE TECHNOLOGIES CONFERENCE
Oct. 13 2010

Colorado
University of Colorado at Boulder

Lasers were a disruptive technology

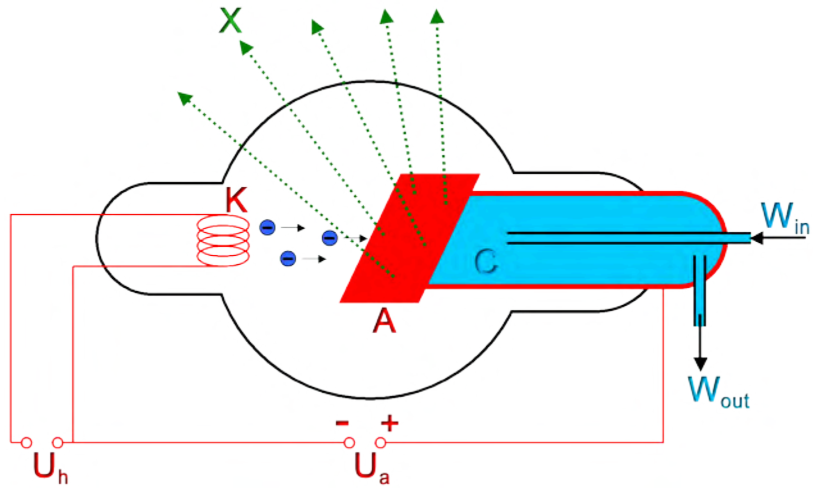




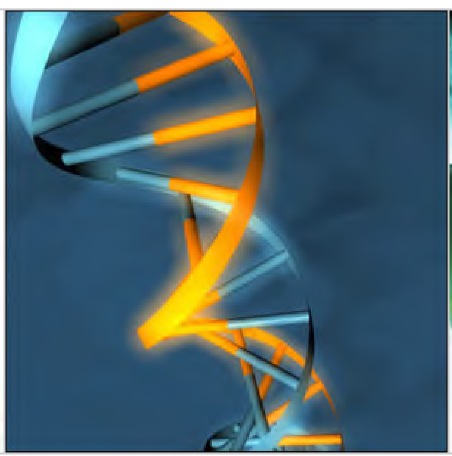
"A solution looking for a problem"
Charles Townes

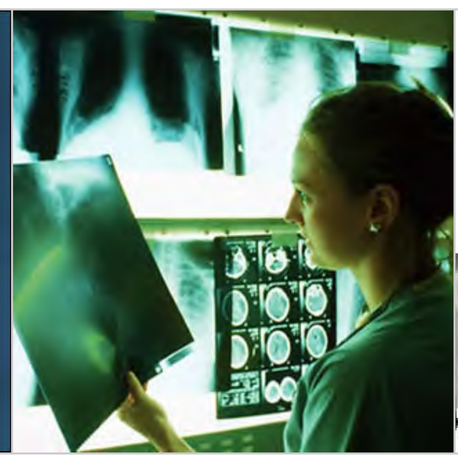
X-Ray light also greatly benefits society

Wilhelm Roentgen



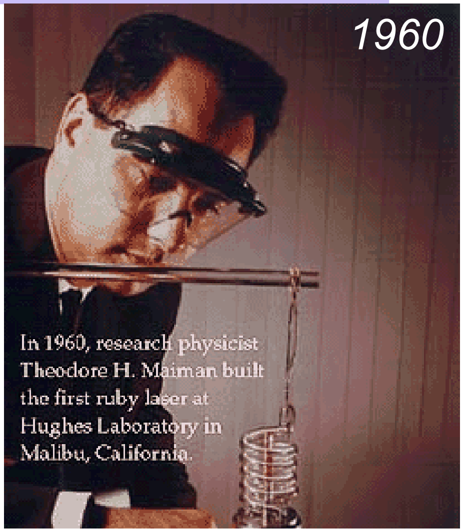
Roentgen X-ray tube

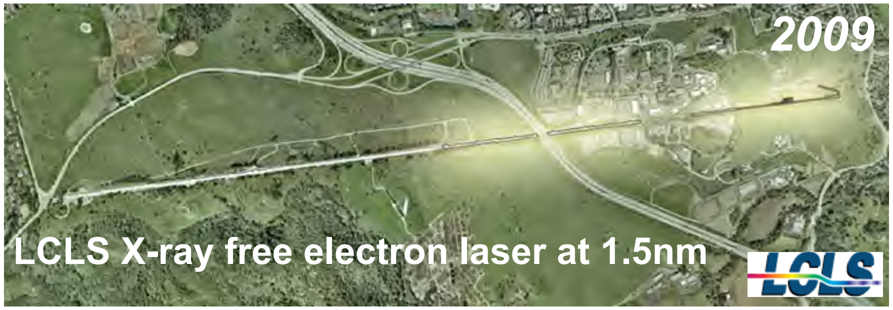




Can we build a tabletop x-ray laser?

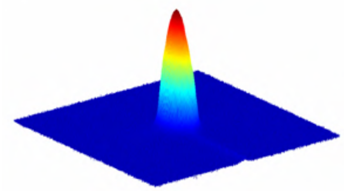
- X-ray lasers are more challenging than visible lasers
  - Power requirement scales rapidly with decreasing wavelength: TW pump for 1 nm laser!
- Alternative: new x-ray free electron lasers
  - mJ coherent x-ray pulses at 30 Hz
  - large, expensive, limited access time
- Need another approach to make coherent x-ray sources widely available

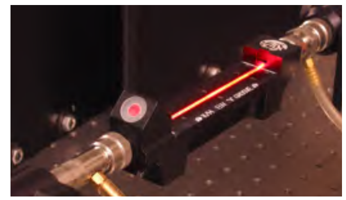




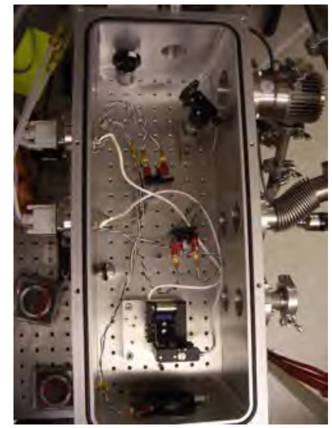
Coherent ultrafast x-ray beams on a tabletop

High average power mid-IR femtosecond laser



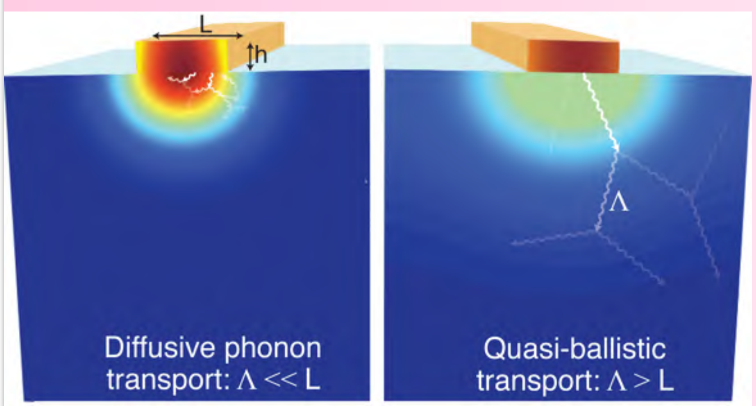


X-ray upconversion

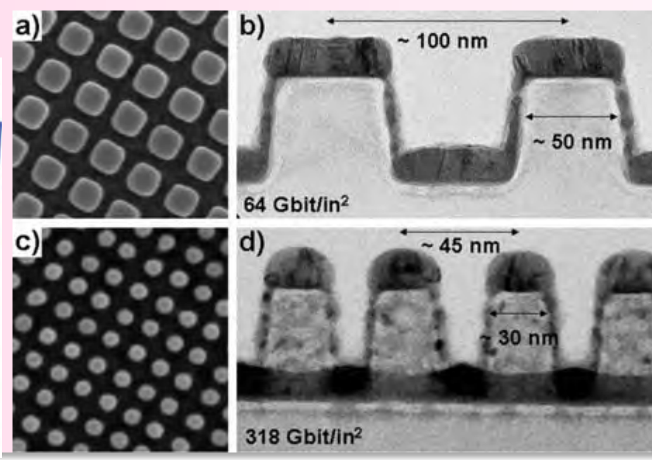


Soft x-ray microscope

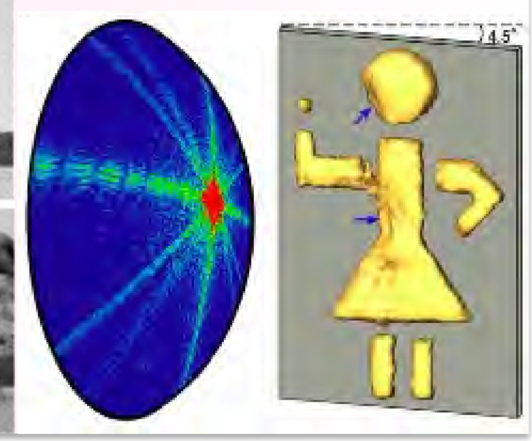
Thermal management in nanostructures



Fundamental time and space limits in magnetic materials

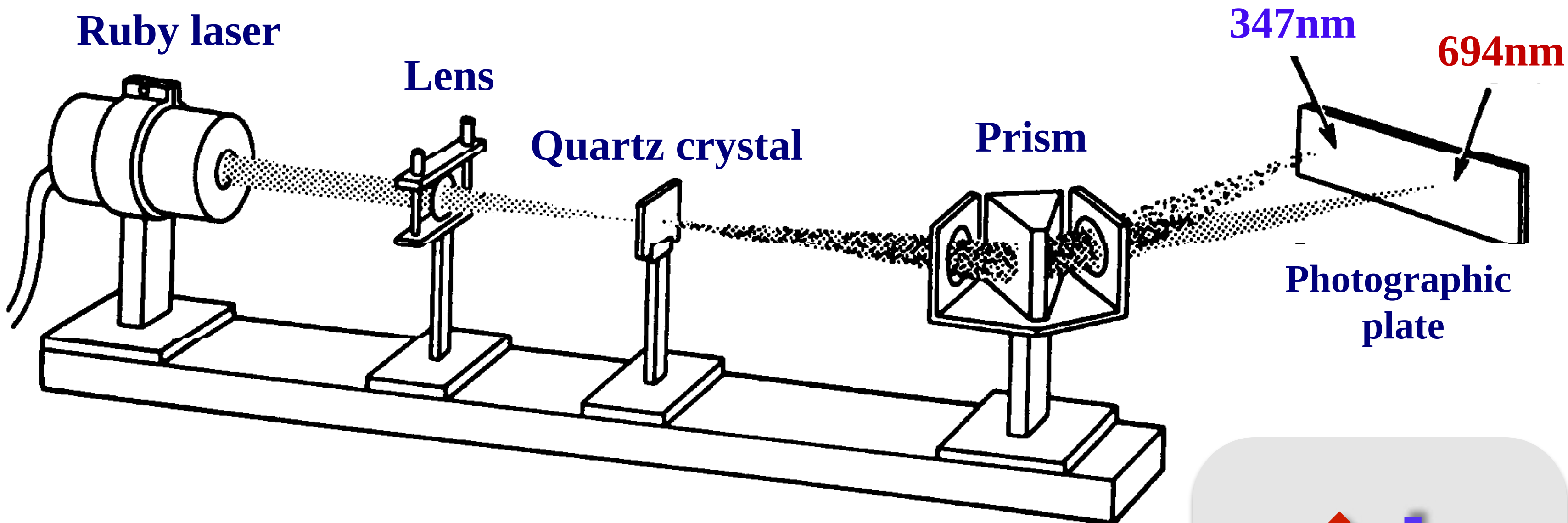


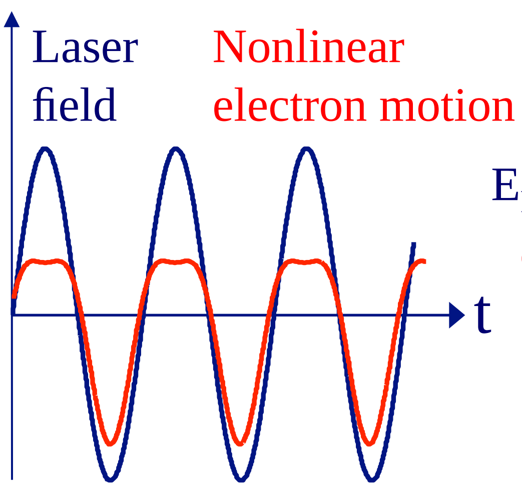
Advanced element-specific nanoimaging



The birth of Nonlinear Optics – second harmonic generation

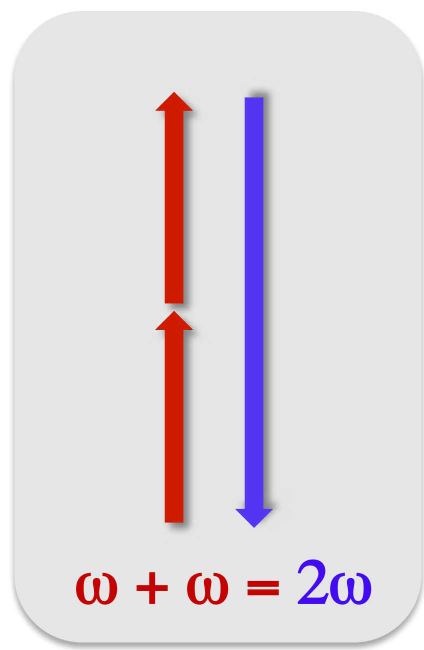
P.A. Franken et al, PRL 7, 118 (1961)





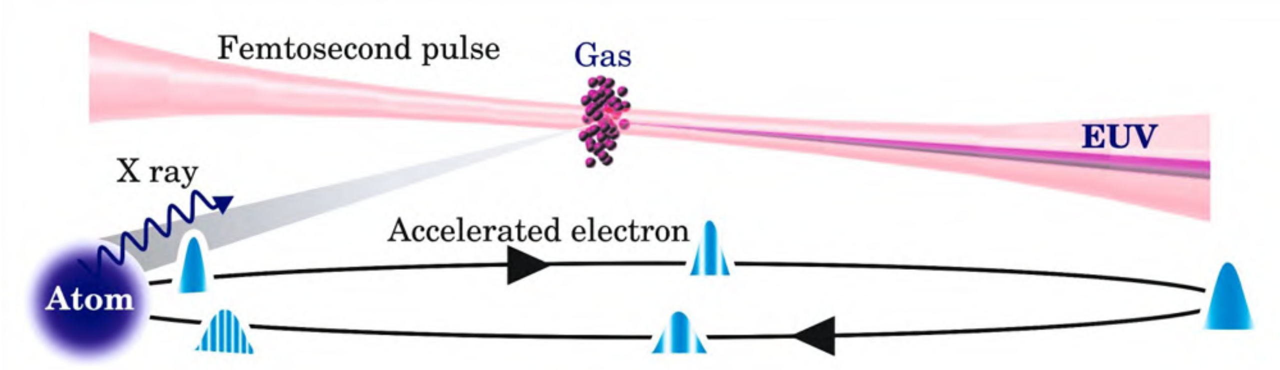
$E_P(\omega)=E_0^{i\omega t}$

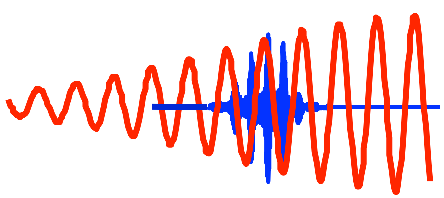
$d(\omega)=a_1 e^{i\omega t} + a_2 e^{i2\omega t} + \ldots$



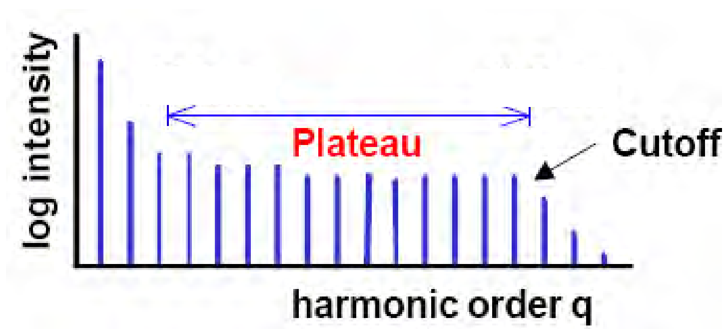
High Harmonic Generation - extreme nonlinear optics

- Coherent x-rays are generated by focusing a femtosecond laser into a gas
- Broad range of harmonics generated simultaneously from UV – keV
- Discovered in 1987, explained in 1993





< 10 fs duration



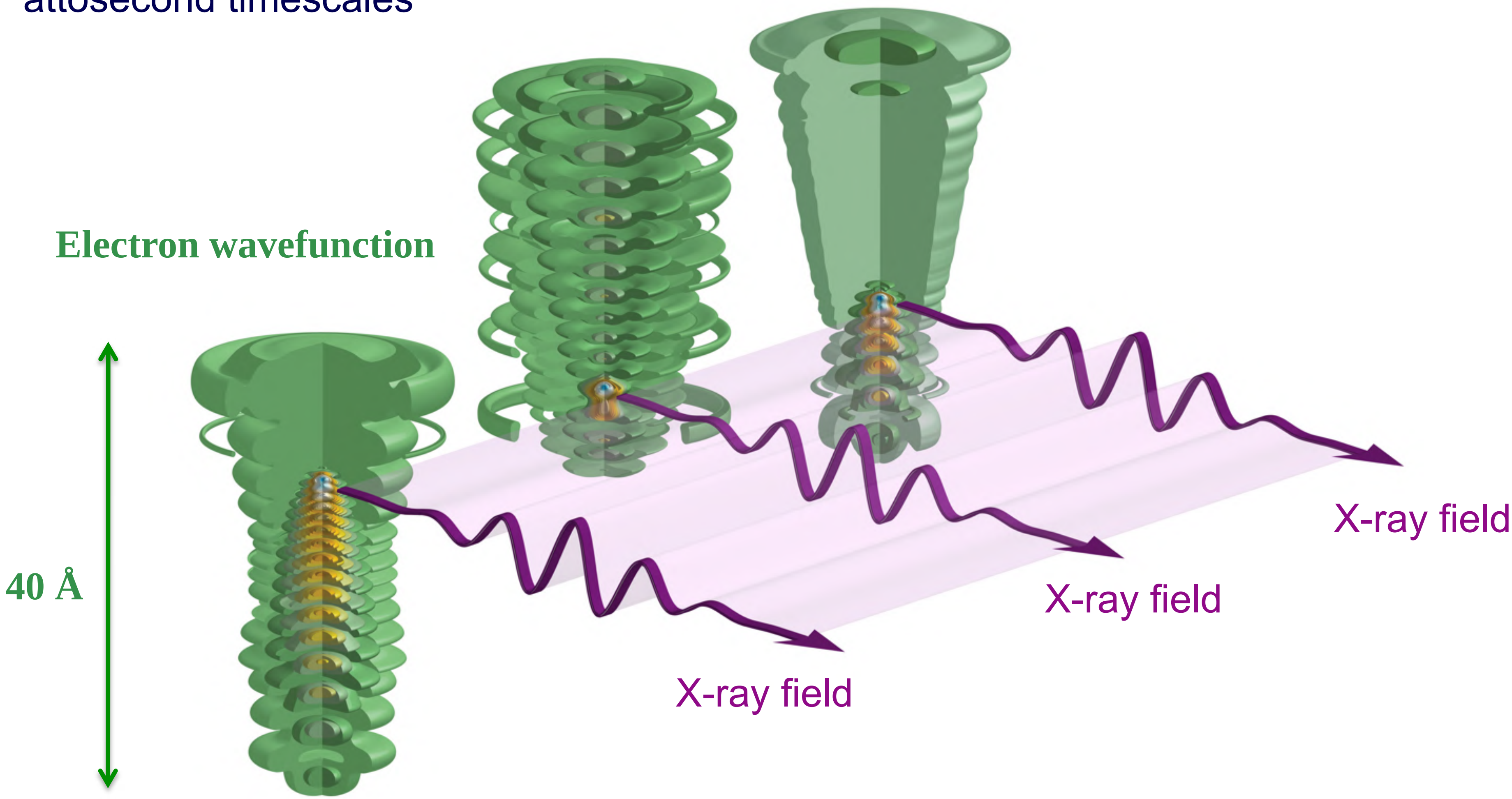
Broad frequency range UV - keV

$$h\nu_{max} \propto I_{laser}\lambda_L^{\ 2}$$

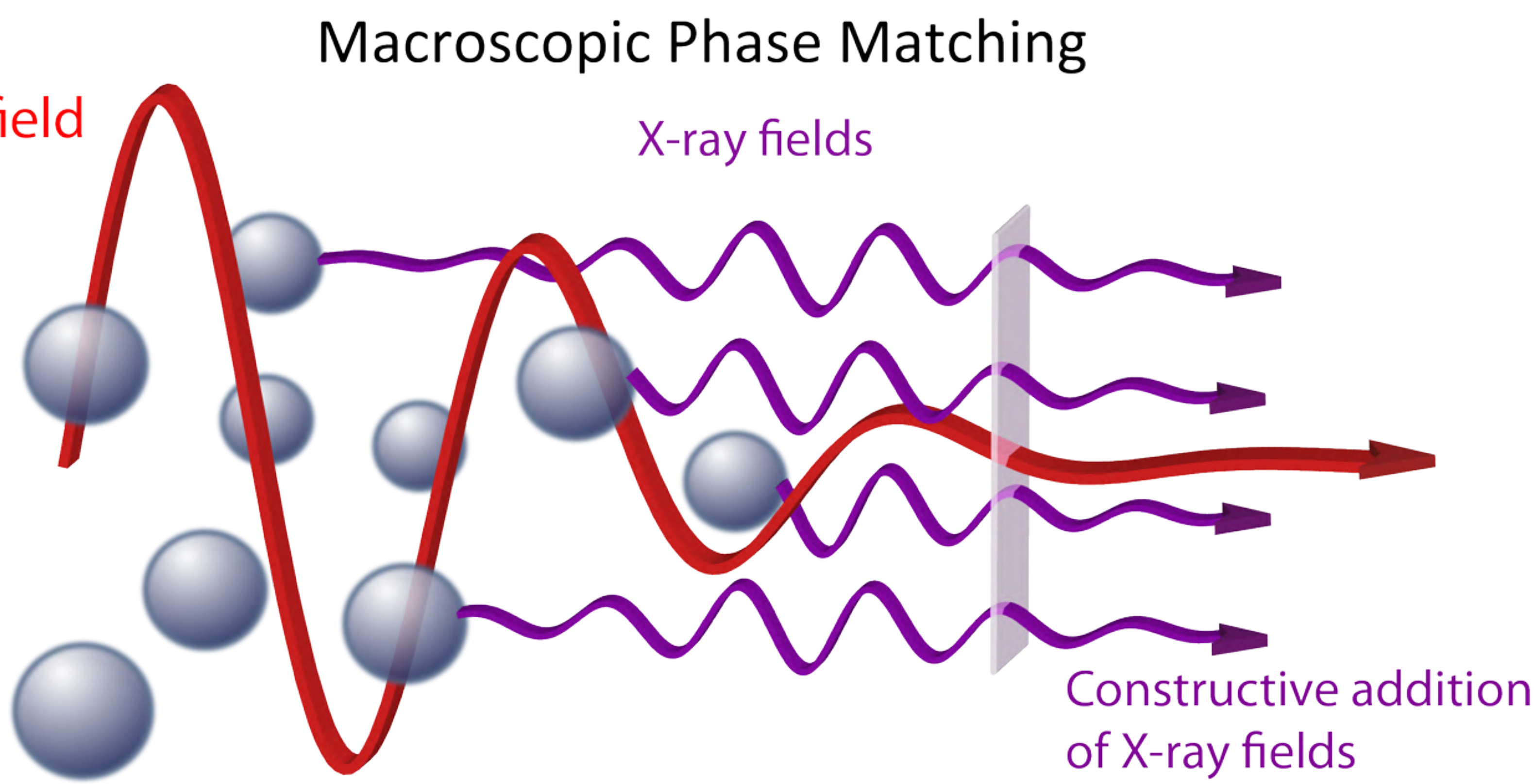
Electron wriggle energy coherently converts to x-rays

High harmonic generation – quantum picture

- Electron wavefunction is highly modulated when driven by strong laser field
- Rapidly changing dipole moment give rise to high harmonics in radiated field
- Can control x-ray emission by controlling a radiating electron on Å spatial scales and attosecond timescales

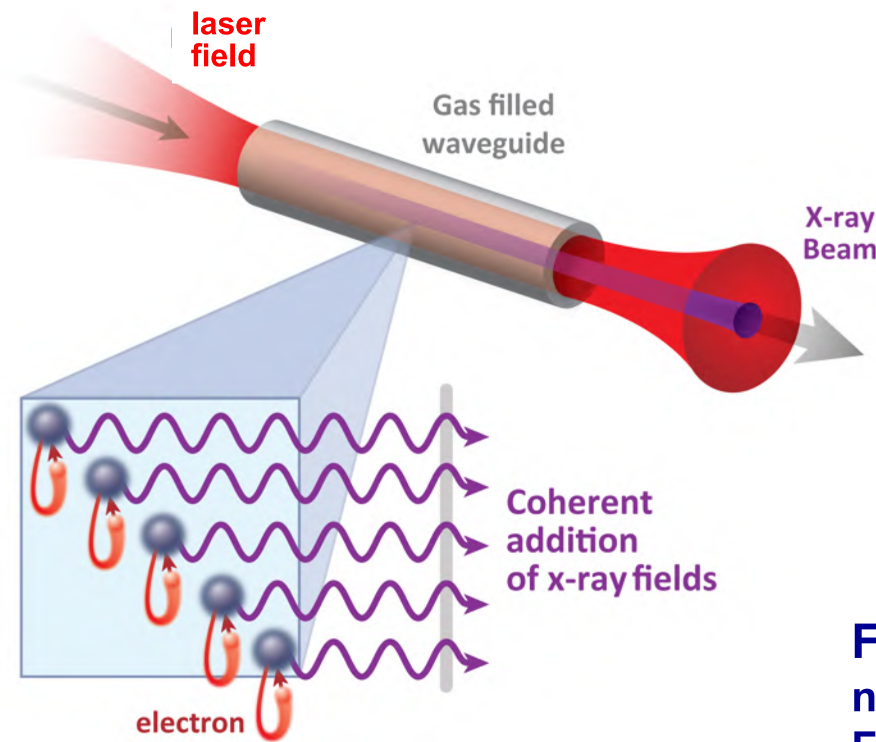


How to efficiently upconvert laser light to soft x-rays?

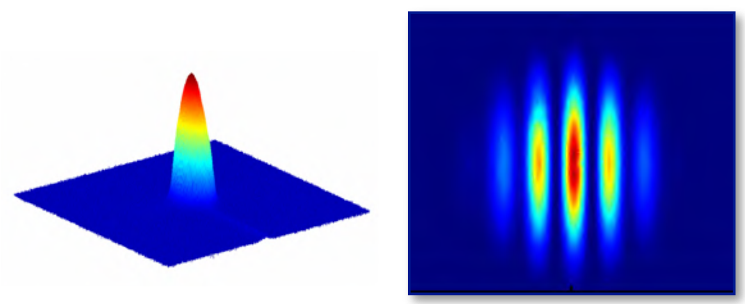


How to efficiently upconvert laser light to soft x-rays?

- Tune the gas pressure to equalize the laser and x-ray phase velocities
- Generate fully coherent, bright, soft x-ray beams
- Efficiency of ≈ 10^{-5} per harmonic in EUV region below 150 eV



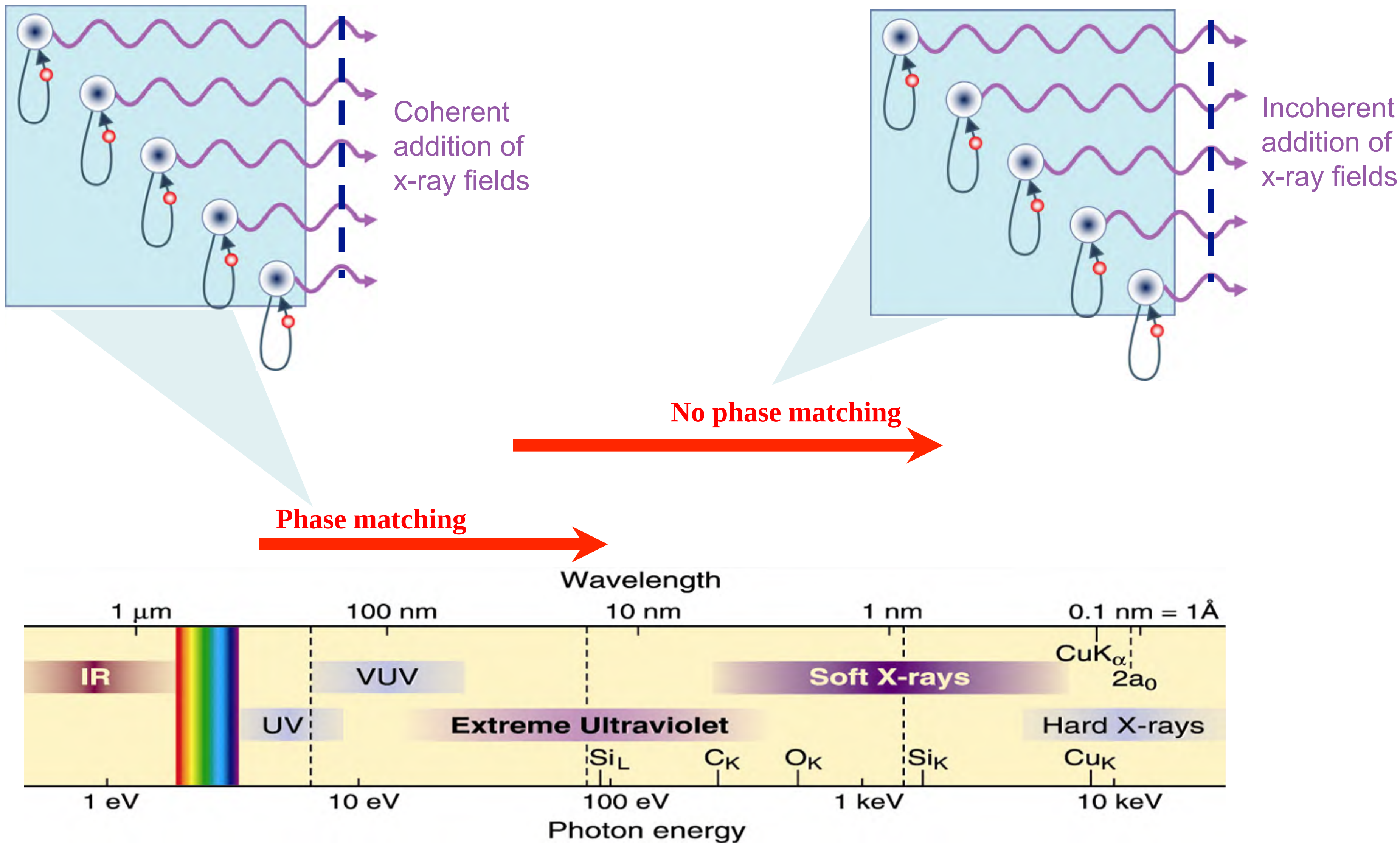
$V_{Laser}=V_{X\text{-}ray}=C$



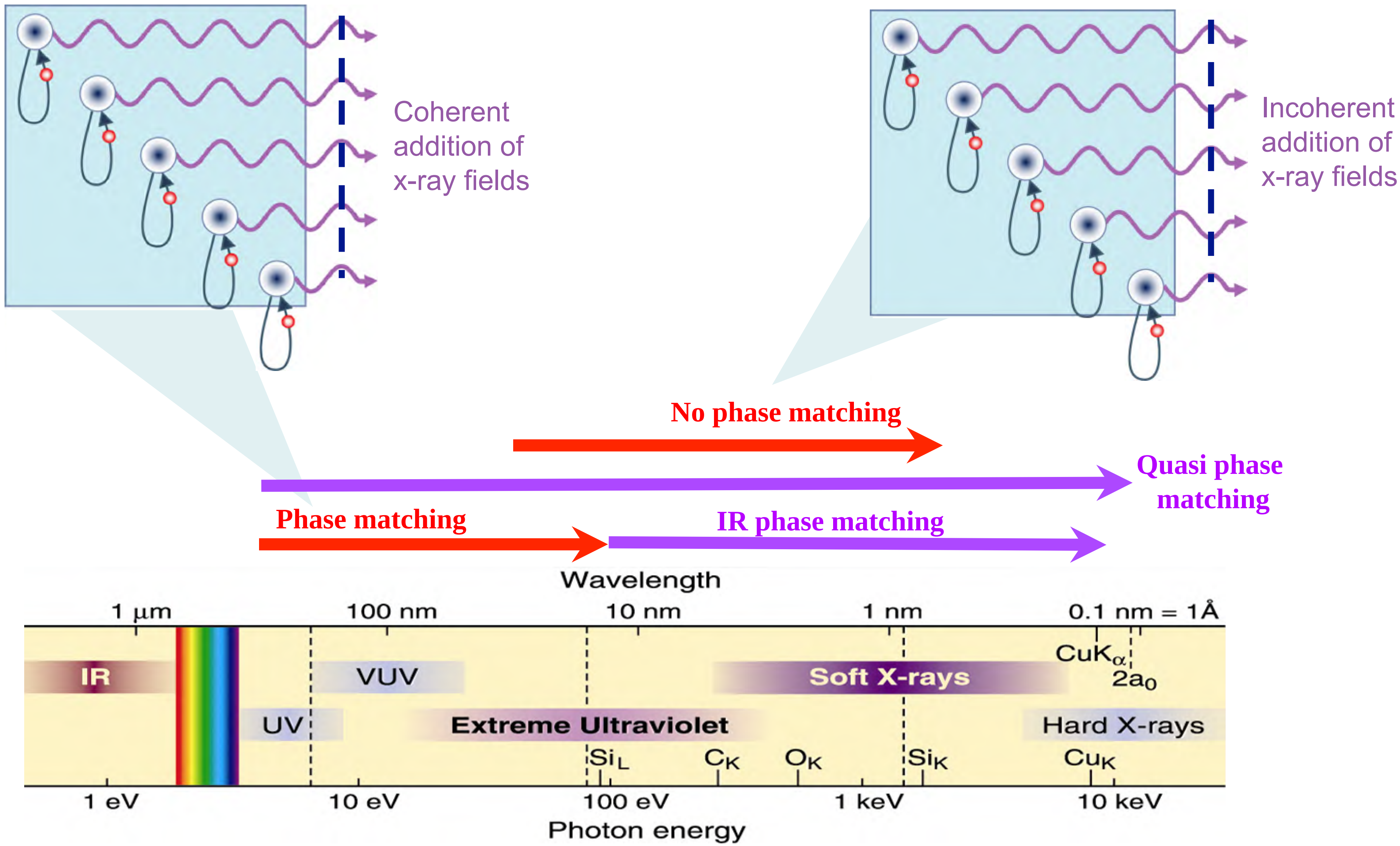
Fully coherent bright EUV and soft x-rays
nJ per harmonic, uW average powers
Femtosecond-to-attosecond duration

Science ***280**, 1412 (1998)*
Science ***297**, 376 (2002)*
Science ***317**, 775 (2007)*

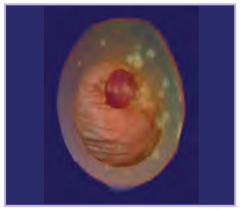
Grand challenge – how to prevent dramatic fall-off in brightness in x-ray region because x-rays interfere destructively



Disruptive technology - new phase matching schemes allow fs lasers to be upconverted to coherent hard x-rays



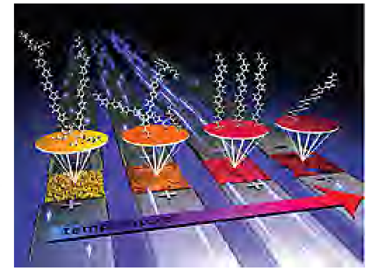
X-ray light is a unique tool for science



Bio-microscopy

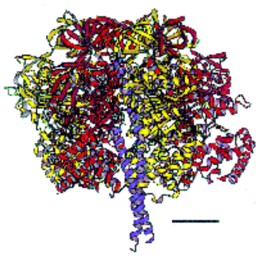
Spectro-microscopy of surfaces

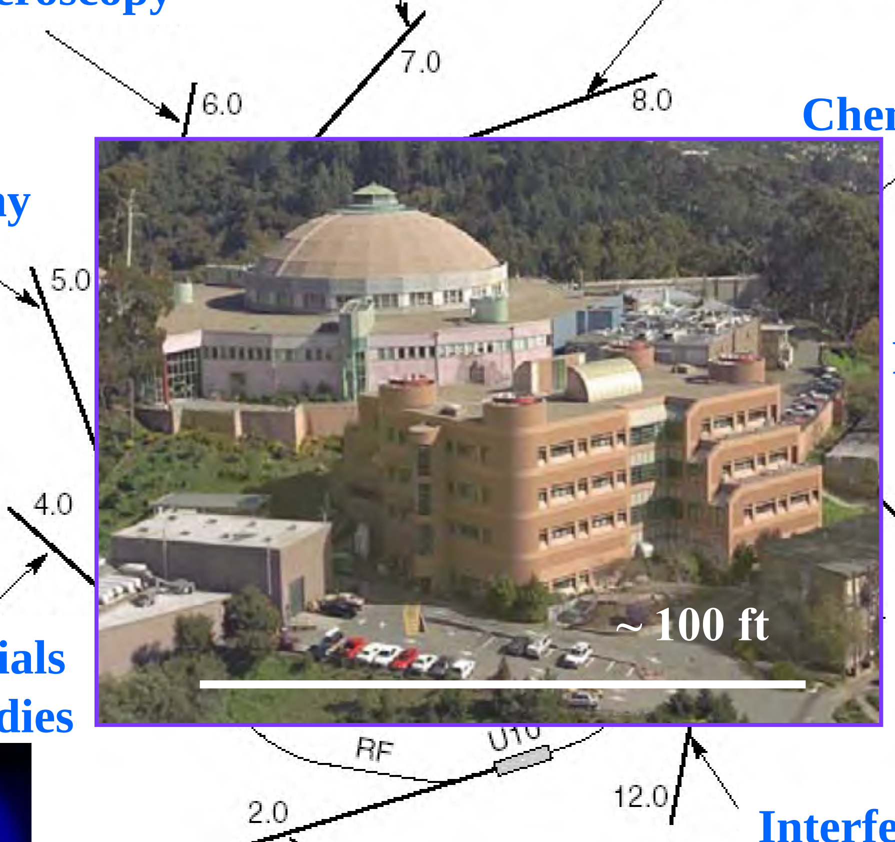
Surface science



Chemical dynamics

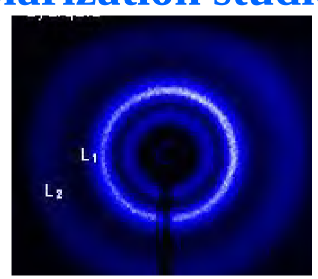
Protein crystallography



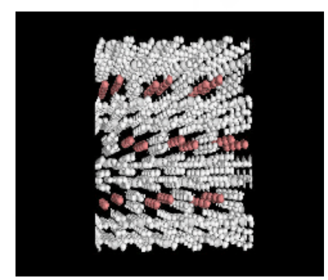


Photoemission spectroscopy

Magnetic materials
Polarization studies



Materials science and biology



Atomic and Molecular physics

Interferometry and coherent optics

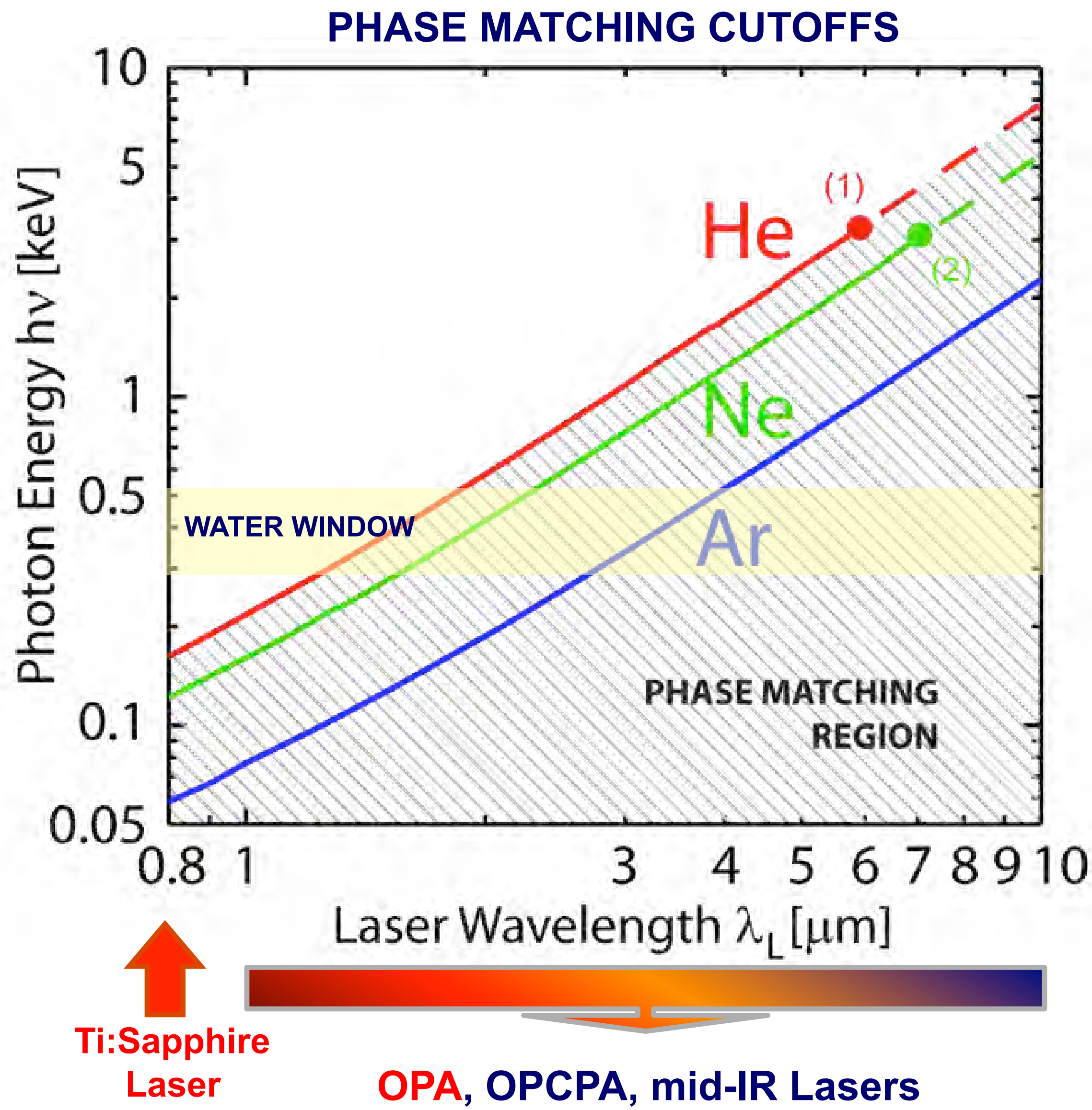
Phase matching using mid-infrared lasers

Mid-IR lasers need lower intensity for a given harmonic energy: $h\nu_{max} \propto I_{laser}\lambda_L^2$

Lower laser intensity = > lower ionization, better phase matching

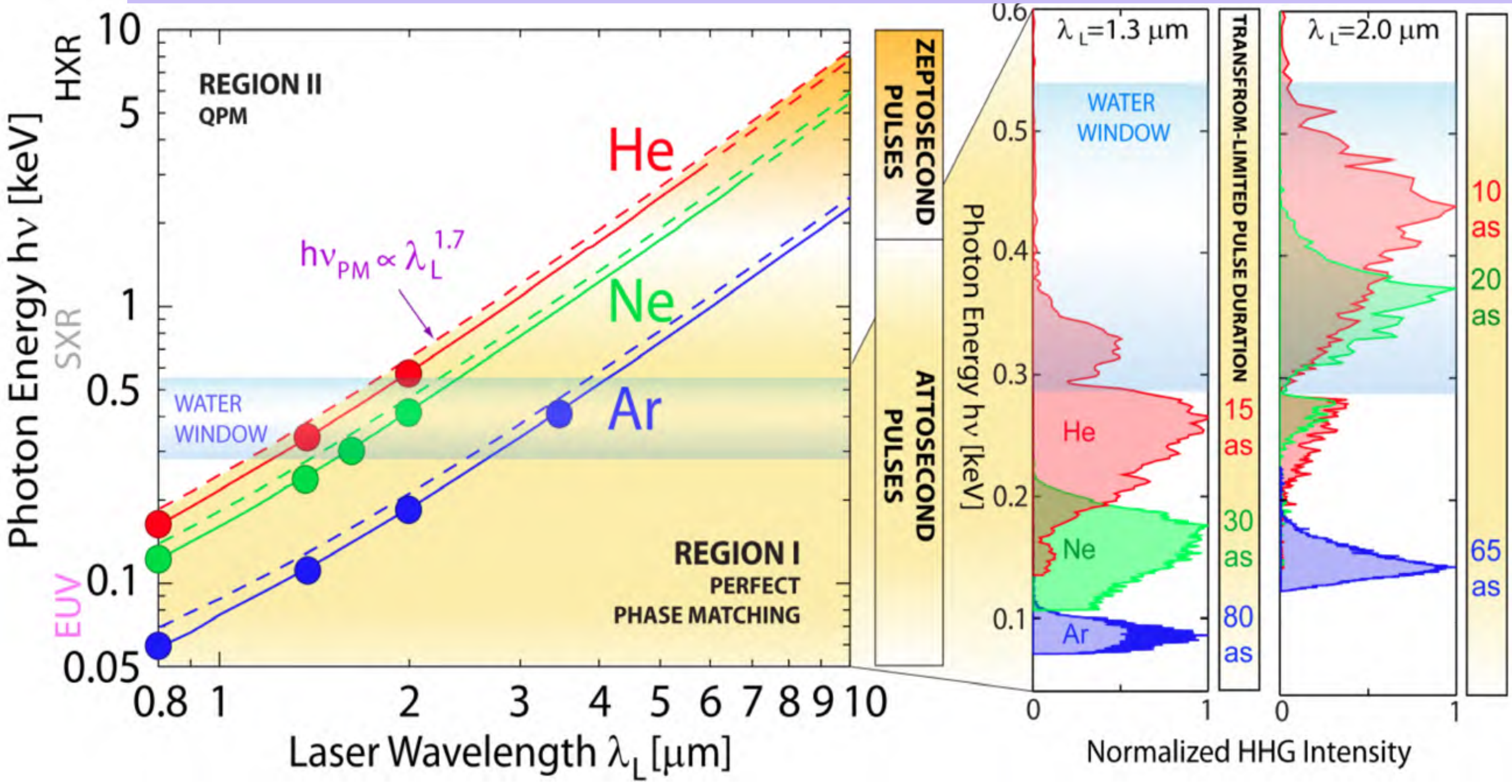
Single atom yield is lower for mid-IR drivers (λ^{-7})

BUT phase matching pressure and gas transparency increase!

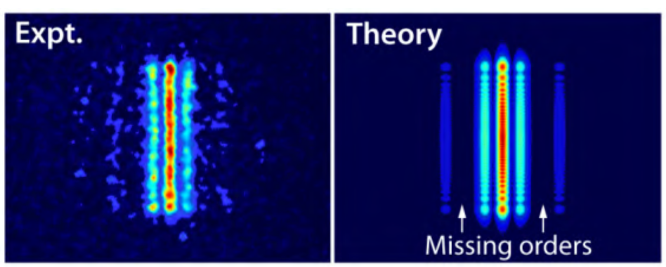


*T. Popmintchev et al. Optics Letters **33**, 2128 (2008); PNAS, **106**, 10516 (2009); PRL, tbp (2010); Nature Photonics, tbp (2010)*

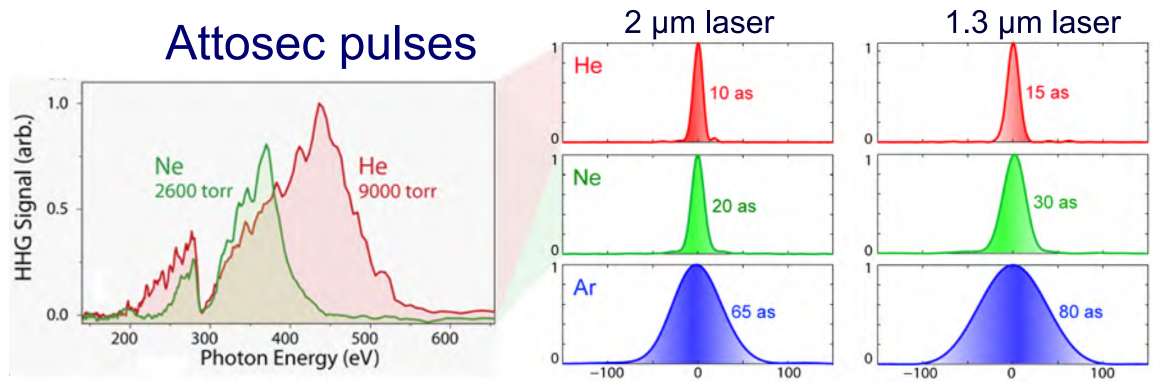
Broad x-ray supercontinuum in laser-like beam



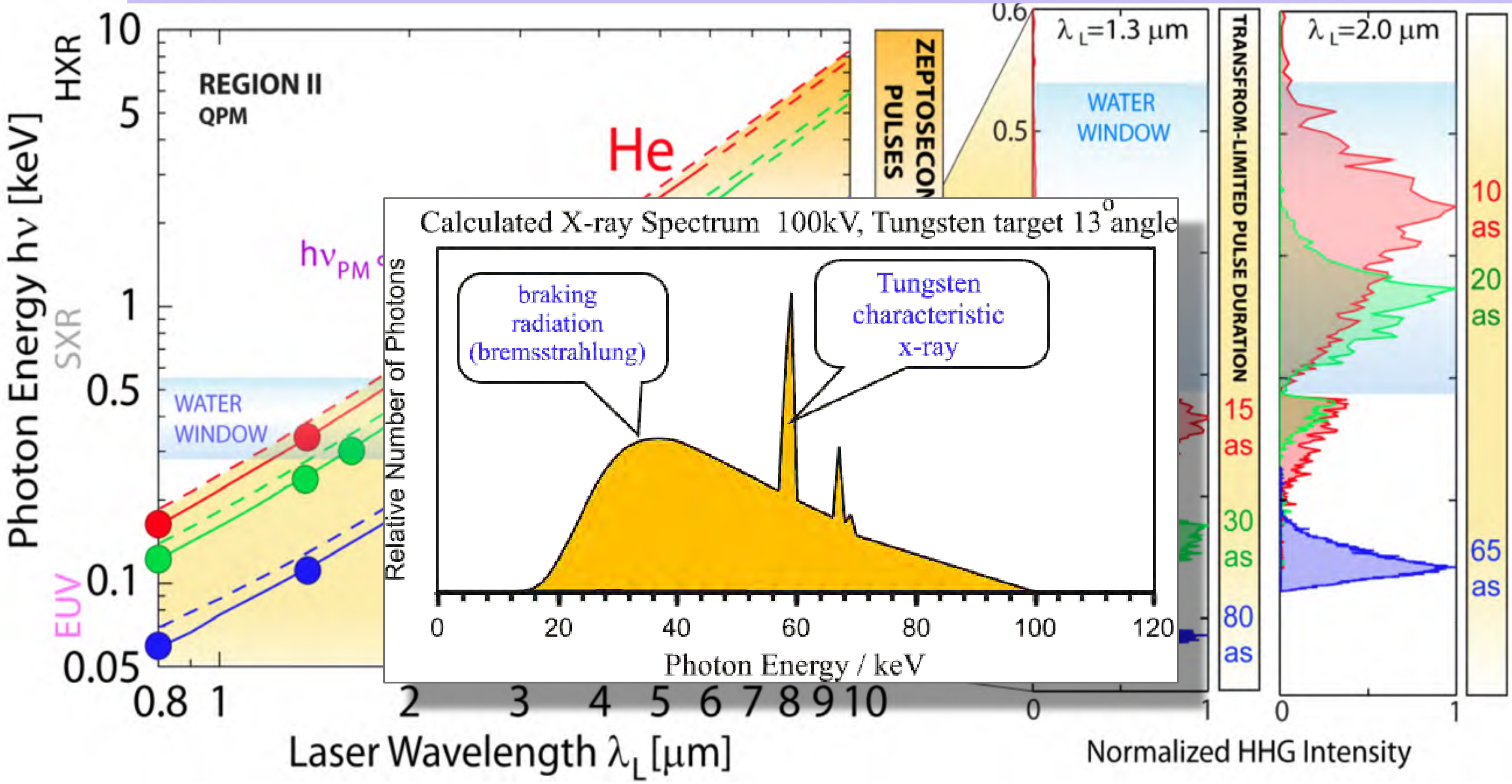
Spatially coherent beams at 3nm



Attosec pulses



Coherent version of x-ray tube



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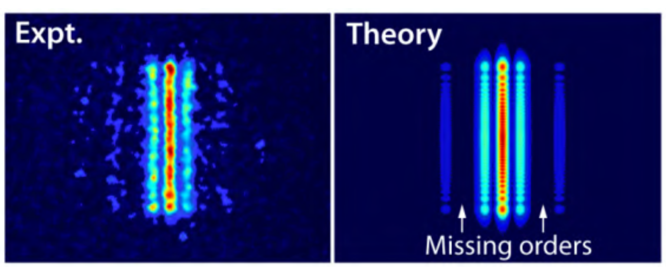

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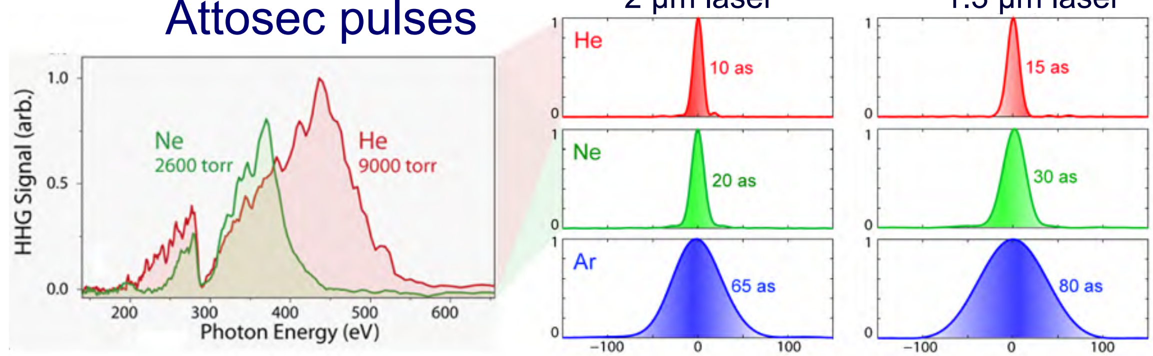
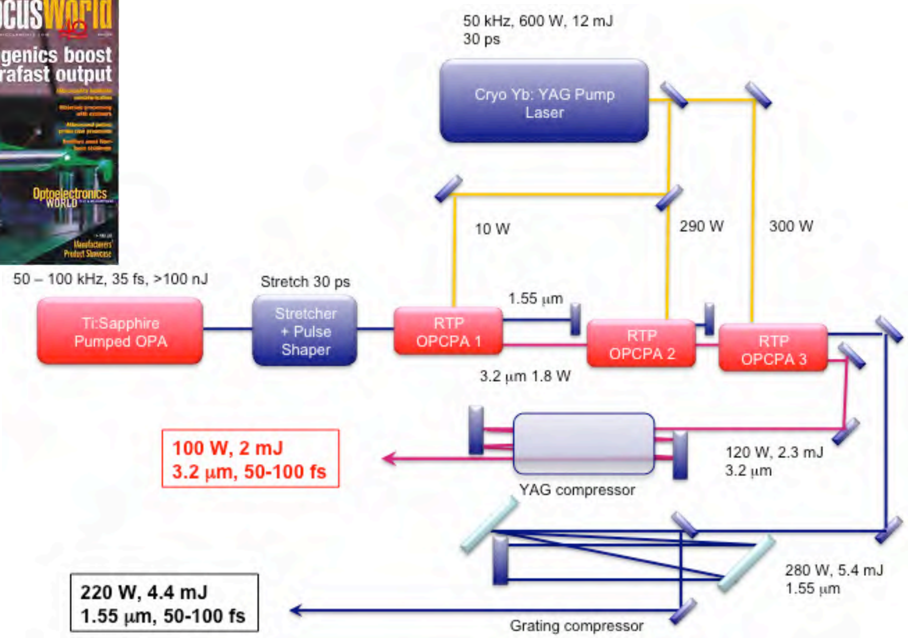


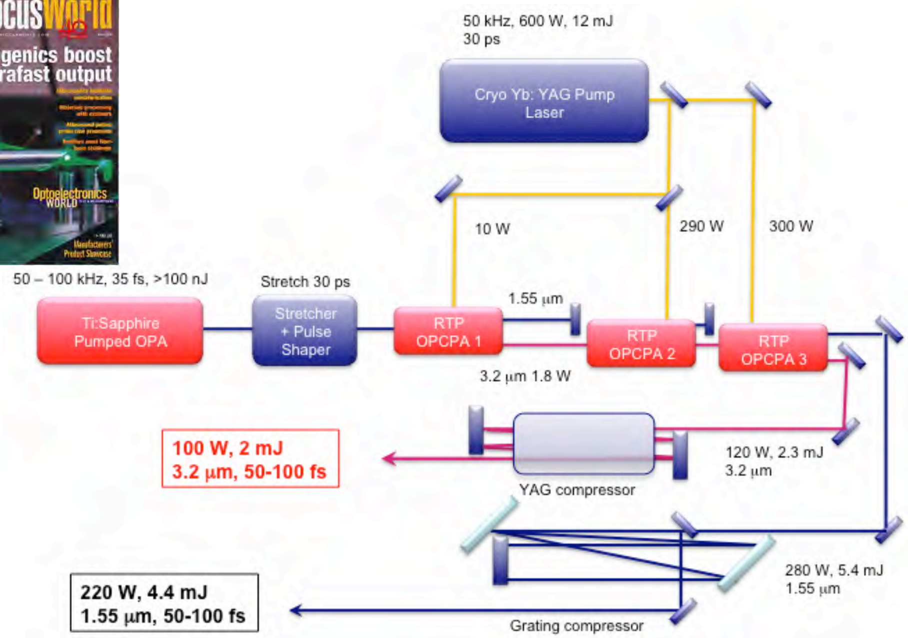
Mid-IR fs lasers with sub-kW average powers

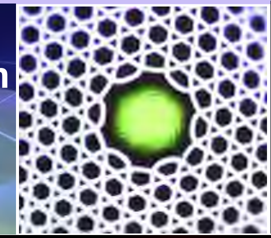


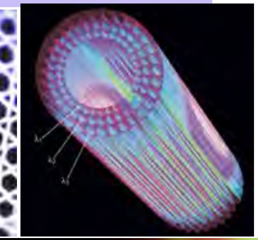
- OPCPA
- Mid-IR laser materials
- Cryogenic cooling enables high av. Power
- Tabletop footprint

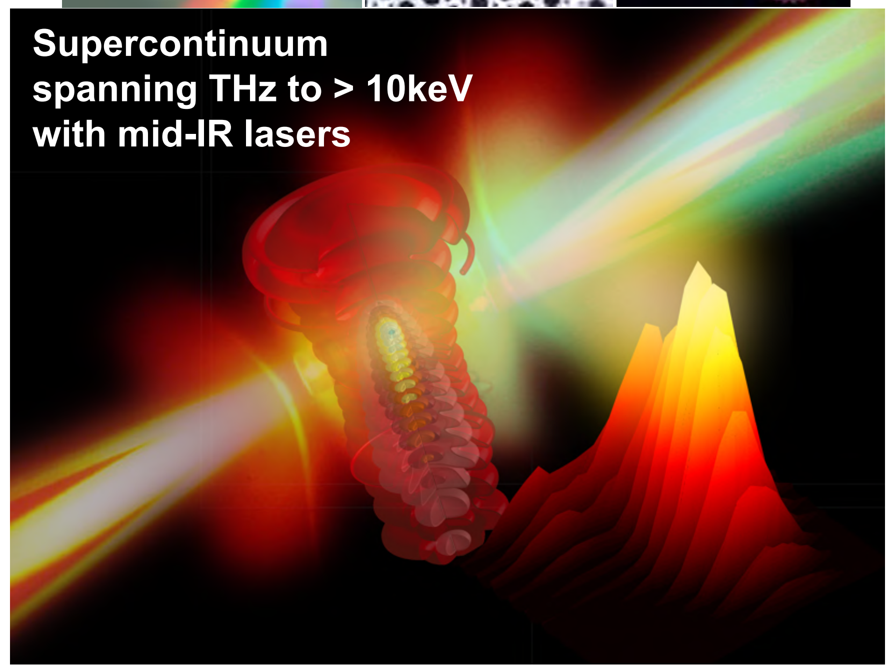
- Efficiency of femtosec lasers in mid-IR approaching that of continuous lasers
- 50kHz, 3mJ, mid-IR lasers will generate 50 μW in $\lambda/\Delta\lambda \approx 100$ up to 1 keV
- Many applications in supercontinuum generation, remote chemical sensing, countermeasures, filamentation, micromachining and imaging

Mid-IR fs lasers with sub-kW average powers









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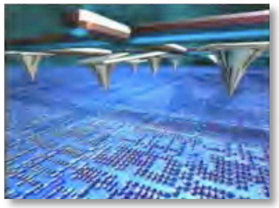
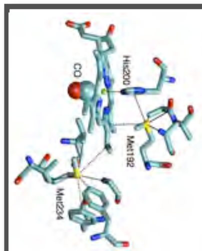
Applications of ultrafast coherent x-rays



NANOSCIENCE (50nm)
- **Ultrafast, elemental, magnetic switching speeds (thin samples)**
- **Nanoscale heat flow**



EXAFS/PEEM/ESCA

Image reactions at the level of atoms and electrons

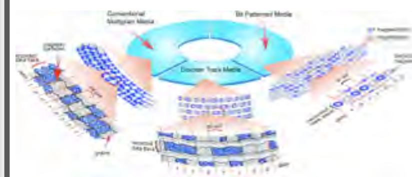
Understand and control function

METROLOGY FOR EUV LITHOGRAPHY

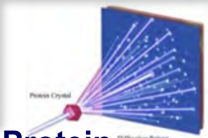
BIO-IMAGING

Cellular and materials tomography (10nm, elemental)

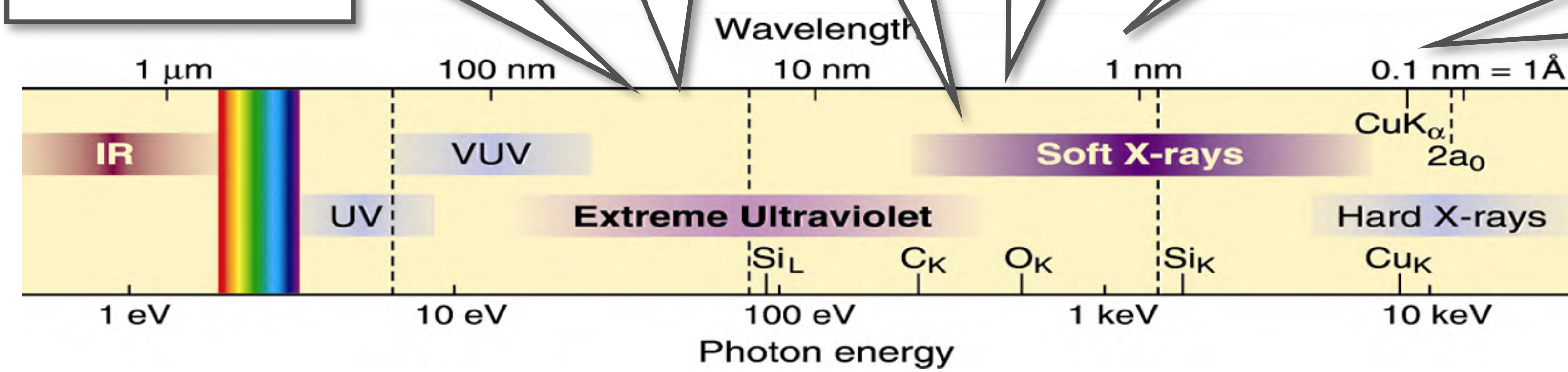
NANOSCIENCE (< 10nm)



Dynamic imaging of advanced nano-structures (magnetic, thermal etc.)

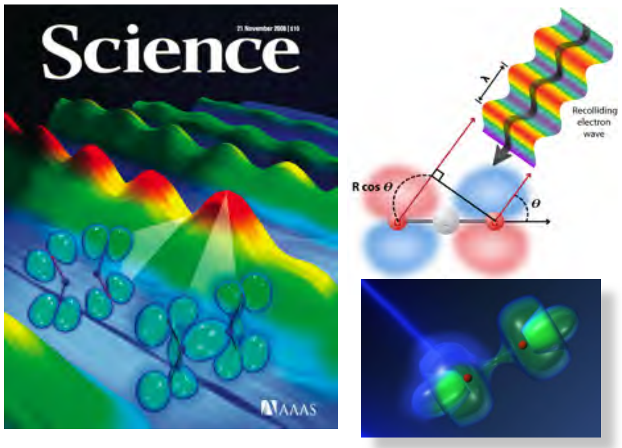


Protein crystallography

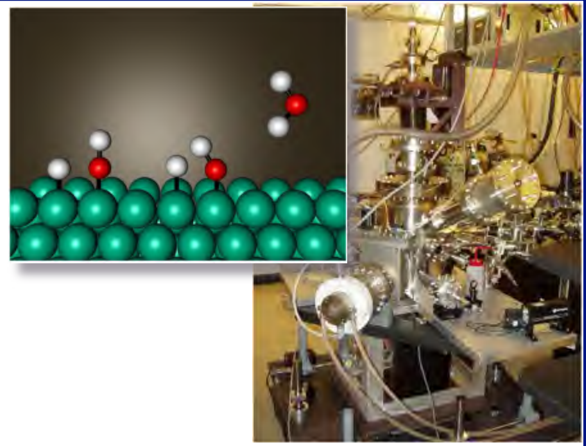


Coherent soft x-rays are ideal probes of nanoworld:
- Penetrate thick objects and image small features
- Elemental and chemical specificity if HHG can extend to x-ray absorption edges
- Tabletop applications to date limited to ≈ 100 eV

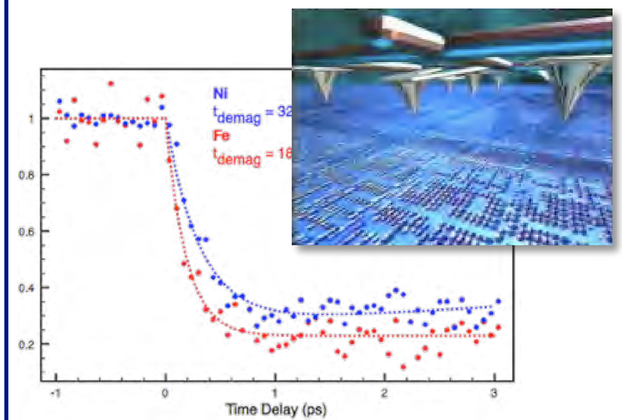
Ultrafast x-rays address important technological and scientific questions



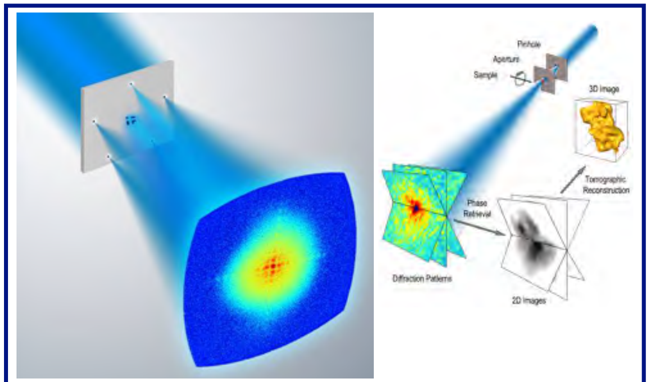
Molecular imaging: image changing electronic orbital and molecular structure



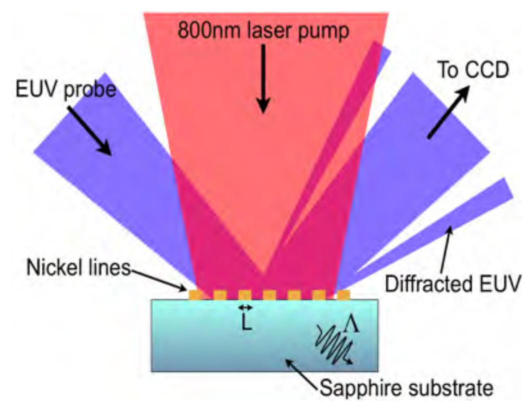
Surface science: probe electronic dynamics on catalysts, photovoltaics



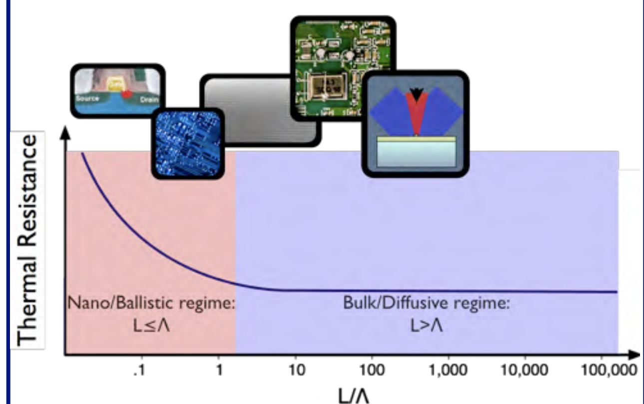
Magnetics: Probe nanodomains, magnetic dynamics



Nanoimaging: High resolution 3D imaging of thick samples using coherent lensless imaging

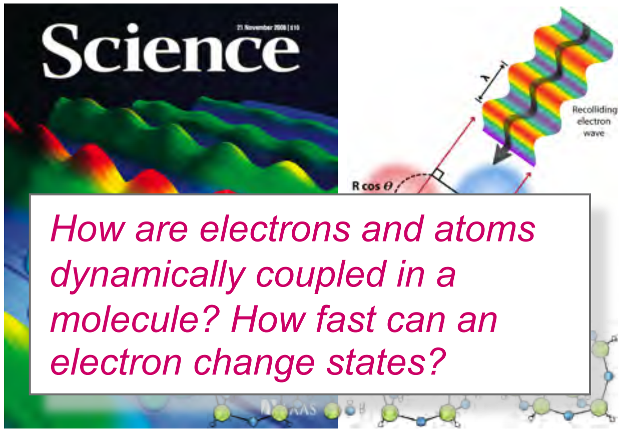


High frequency acoustic metrology: Characterize thin films, interfaces, adhesion



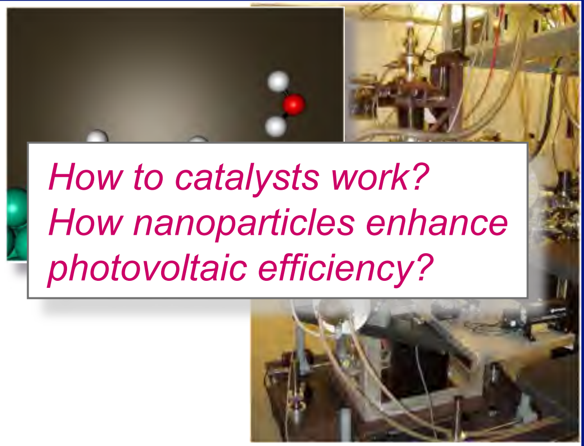
Nanothermal transport: probe heat flow in nanostructures

Ultrafast x-rays address important technological and scientific questions



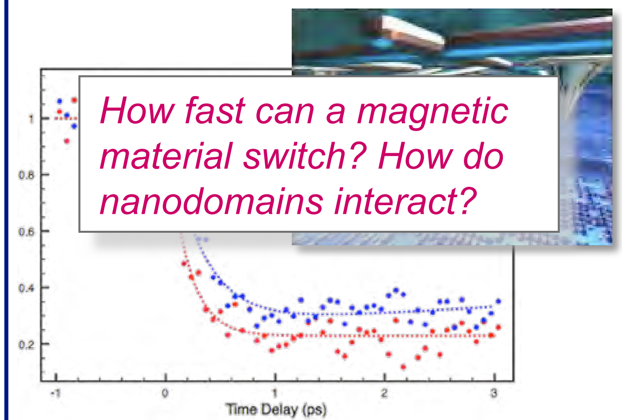
How are electrons and atoms dynamically coupled in a molecule? How fast can an electron change states?

Molecular dynamics and imaging: probe coupled electronic orbital and molecular dynamics



How to catalysts work? How nanoparticles enhance photovoltaic efficiency?

Surface science: probe electronic dynamics on catalysts, photovoltaics



How fast can a magnetic material switch? How do nanodomains interact?

Magnetics: Probe nanodomains, magnetic dynamics

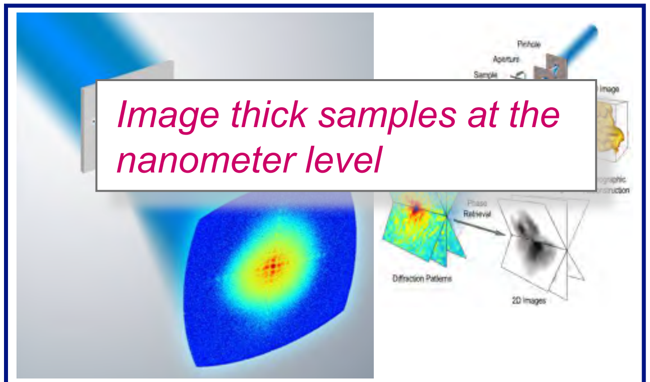
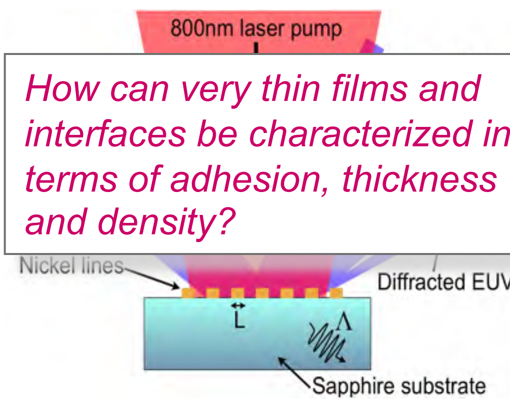


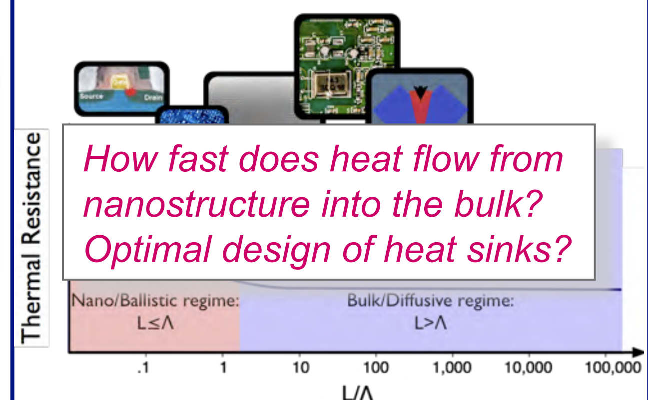
Image thick samples at the nanometer level

Nanoimaging: High resolution 3D imaging of thick samples using coherent lensless imaging



How can very thin films and interfaces be characterized in terms of adhesion, thickness and density?

High frequency acoustic metrology: Characterize thin films, interfaces, adhesion

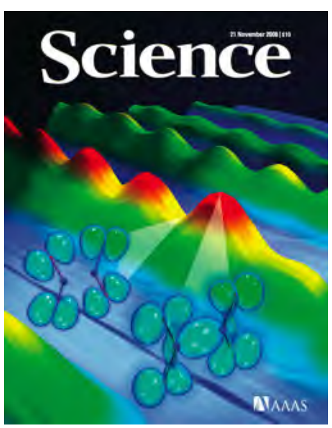


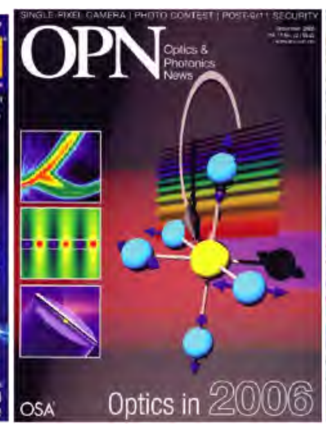
How fast does heat flow from nanostructure into the bulk? Optimal design of heat sinks?

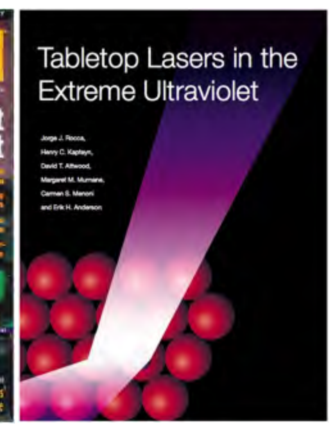
Nanothermal transport: probe heat flow in nanostructures

Future

- Take attosecond electron rescattering physics, discovered just over 20 years ago, to generate coherent x-rays
- Using mid-IR lasers, can generate coherent x-ray waveforms, with excellent prospects for hard x-ray laser beams on a tabletop……limit??
- Use x-ray lasers to visualize, interact with, and control the nanoworld, to manipulate electrons, atoms and molecules in quantum systems
- Table-top microscopes, nanoprobes and x-ray imaging with unprecedented spatial and temporal resolution
- Thanks to NSSEFF!!







Students and postdocs

STUDENTS *Paul Arpin, Tory Carr, Ming-Chang Chen, Michael Gerrity, Betsy Hall, Craig Hogle, Kathy Hoogeboom, Robynne Lock, Chan La-O-Vorakiat, Qing Li, Matt Seaberg, Dimitar Popmintchev*

POSTDOCS *Xibin Zhou, Alon Bahabad, Predrag Ranitovic, Stefan Mathias, Tenio Popmintchev*

Example recent publications since 2008

1. T. Tenio Popmintchev, M.C. Chen, O. Cohen, M.E. Grisham, J.J. Rocca, M.M. Murnane, H.C. Kapteyn, “Phase-Matching of High Harmonics Driven by Mid-Infrared Light,” Optics Letters **33**, 2128 (2008).

2. W. Li, X. Zhou, R. Lock, S. Patchkovskii, A. Stolow, H.C. Kapteyn, M.M. Murnane, “Time-resolved Probing of Dynamics in Polyatomic Molecules using High Harmonic Generation”, Science **322**, 1207 (2008).

3. E. Gagnon, V. Sharma, W. Li, A.S. Sandhu, R. Santra, P. Ranitovic, C.L. Cocke, M.M. Murnane, H.C. Kapteyn, “Observing the birth of electronic Feshbach resonances and delayed autoionization in soft x-ray induced molecular dissociation,” Science **322**, 1081 (2008).

4. R.L. Sandberg, D.A. Raymondson, C. La-o-vorakiat, A. Paul, K. Raines, J. Miao, M.M. Murnane, H.C. Kapteyn, B.F. Schlotter, “Closing the Gap to the Diffraction Limit: Tabletop Soft X-Ray Fourier Transform Holography with 50 nm Resolution,” Optics Letters **34**, 1618 (2009).

5. M.E. Siemens, Q. Li, M.M. Murnane, H.C. Kapteyn, R. Yang, E. Anderson, K. Nelson, “High-Frequency Surface Acoustic Wave Propagation in Nanostructures Characterized by Coherent Extreme Ultraviolet Beams”, Applied Physics Letters **94**, 093103 (2009).

6. T. Popmintchev, M.C. Chen, Alon Bahabad, M. Gerrity, P. Sidorenko, O. Cohen, I.P. Christov, M.M. Murnane, H.C. Kapteyn, “Phase matched upconversion of coherent ultrafast laser light into the soft and hard x-ray regions of the spectrum”, PNAS **106** (26), 10516 (2009).

7. Margaret M. Murnane and John Miao, “Ultrafast X-Ray Photography”, Nature **460**, 1088 (2009).

8. C. La-O-Vorakiat, M. Siemens, M.M. Murnane, H.C. Kapteyn, S. Mathias, M. Aeschlimann, et al. , “Ultrafast Soft X-Ray Magneto-Optics at the M-edge Using a Tabletop High-Harmonic Source”, Physical Review Letters **103**, 257402 (2009).

9. Mark Siemens, Qing Li, Ronggui Yang, Keith Nelson, Erik Anderson, Margaret Murnane and Henry Kapteyn, “Measurement of quasi-ballistic heat transport across nanoscale interfaces using ultrafast coherent soft x-ray beams”, Nature Materials **9**, 26 (2010).

10. K. S. Raines, S. Salha, R. L. Sandberg, H. D. Jiang, J. A. Rodriguez, B. P. Fahimian, H. C. Kapteyn, J. C. Du, and J. W. Miao, "Three-dimensional structure determination from a single view," Nature **463**, 214 (2010).

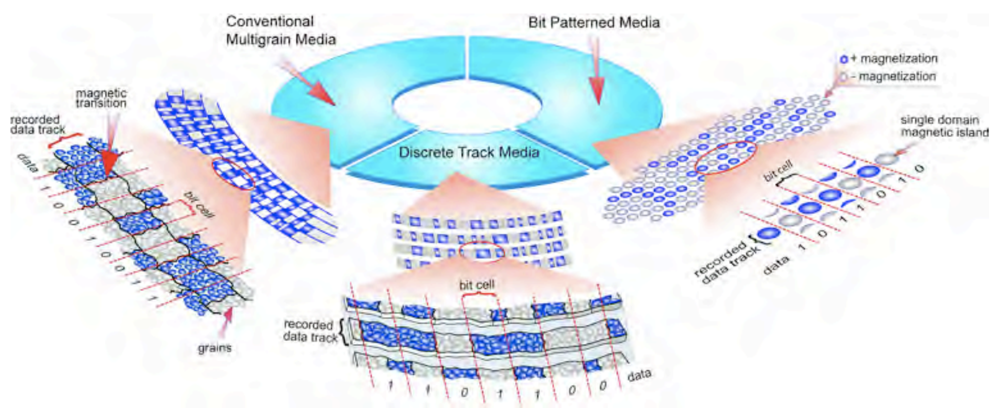
11. Alon Bahabad, Margaret. M. Murnane and Henry C. Kapteyn, “Quasi Phase Matching of Momentum and Energy in Nonlinear Optical Processes”, Nature Photonics **4**, 570 (2010).

12. M.C. Chen, P. Arpin, T. Popmintchev, M. Gerrity, B. Zhang, M. Seaberg, M.M. Murnane and H.C. Kapteyn, “Bright, Coherent, Ultrafast Soft X-Ray Harmonics Spanning the Water Window from a Tabletop Source”, to be published in Physical Review Letters (2010).

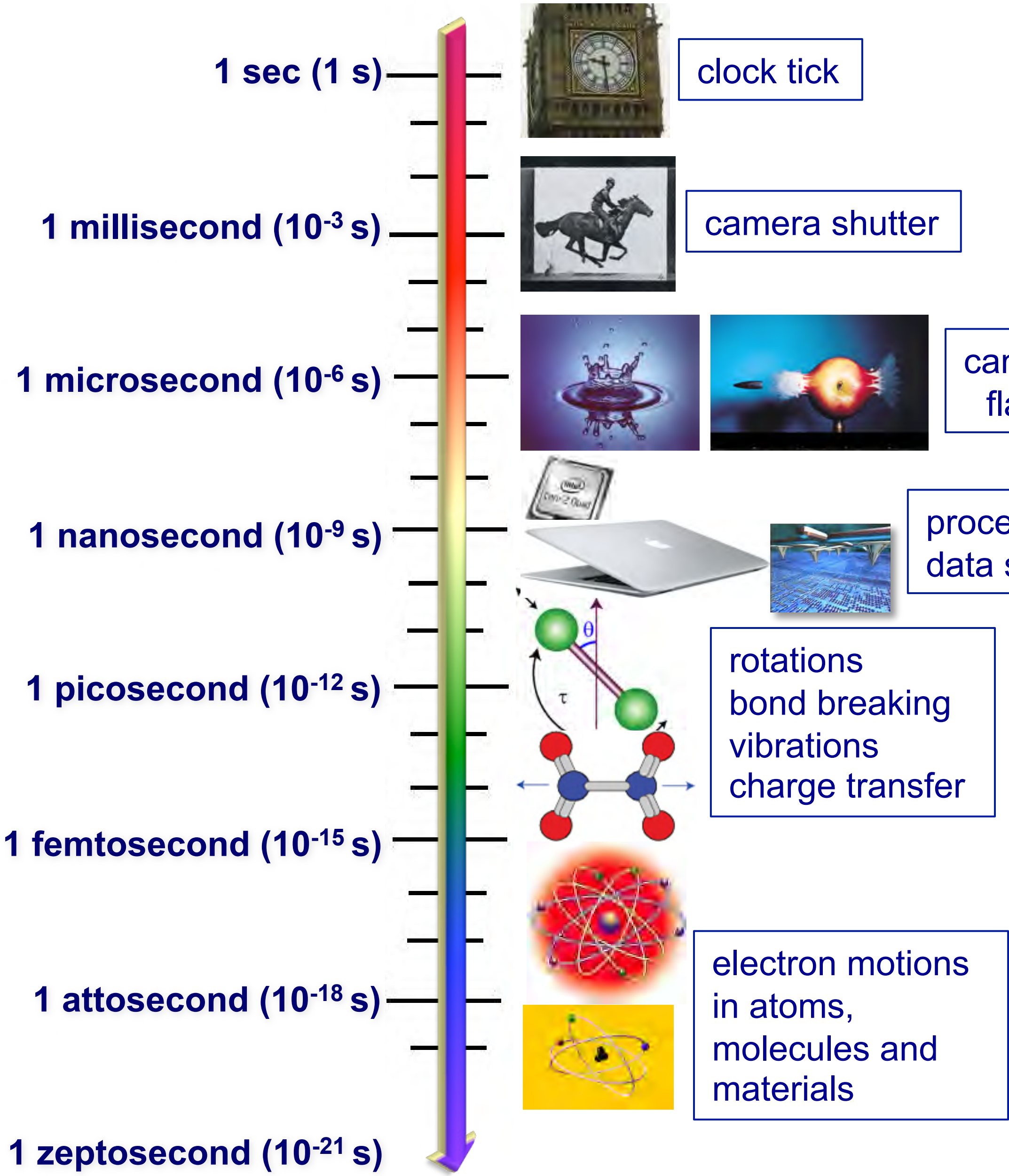
13. T. Popmintchev, M.M. Murnane and H.C. Kapteyn, “Photonics at the Time-scale of the Electron – Bright Coherent X-Rays from Tabletop Ultrafast Lasers”, to be published in Nature Photonics (2010).

Questions we are addressing in nanotechnology

- How to nanodomains interact?
- How fast can the magnetic state switch?
- How does light directly couple to spins?
- What is the optimal designs for nano-bit patterned media?
- Optimal thermal design at the nanoscale
- Element- and chemical-specific image at the nanoscale
- Thin film metrology
- Many more...

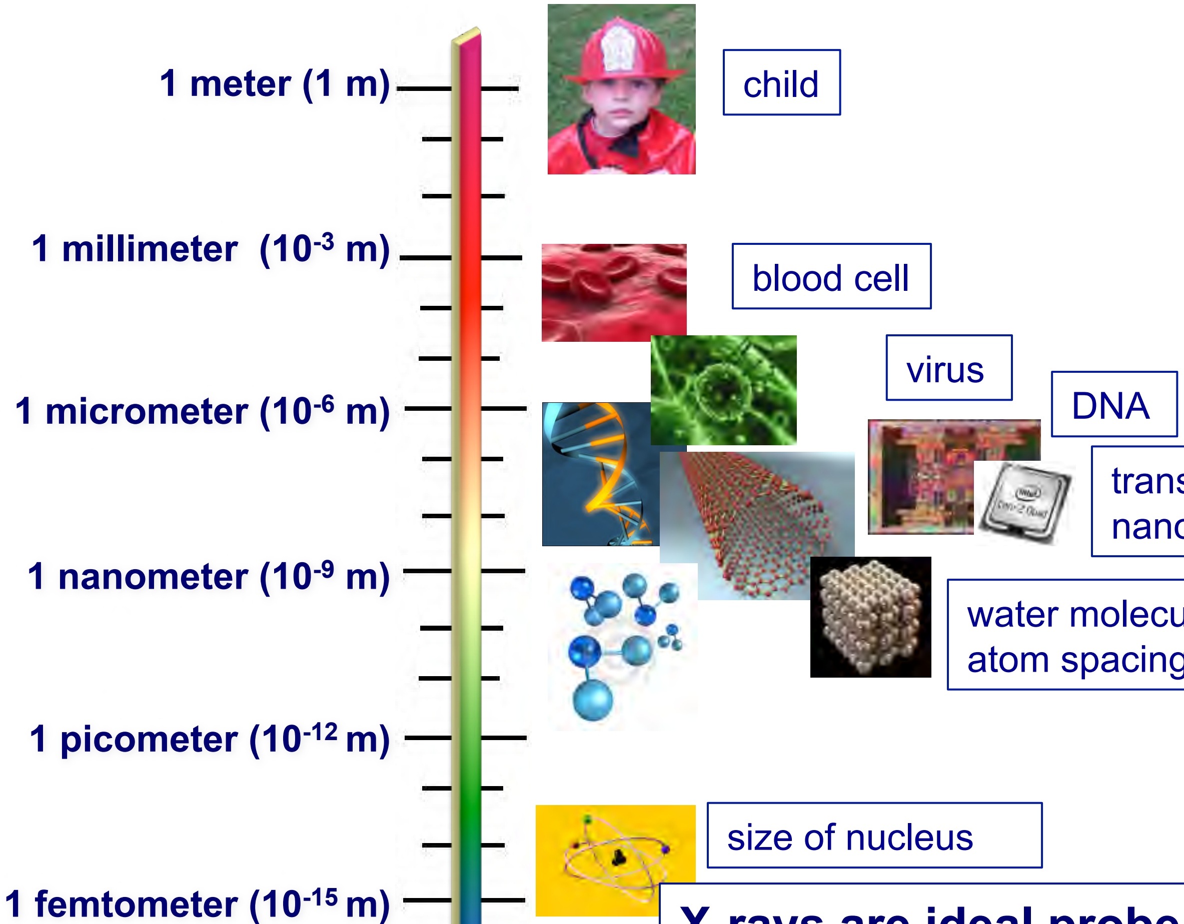


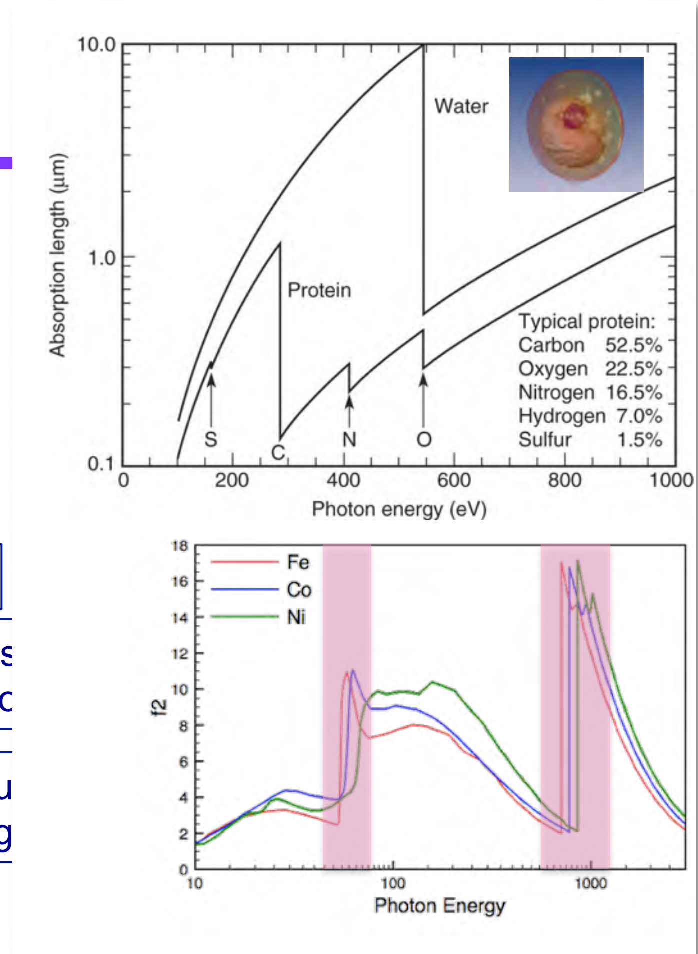
Characteristic time scales of the nanoworld



- Femtosecond to attosecond pulses can capture and manipulate all motion in the natural world, even at the level of electrons

Characteristic length scales





X-rays are ideal probe of nanoworld:

- Penetrate thick objects
- Image small features
- Elemental and chemical specificity
- Surface sensitivity from photoemission