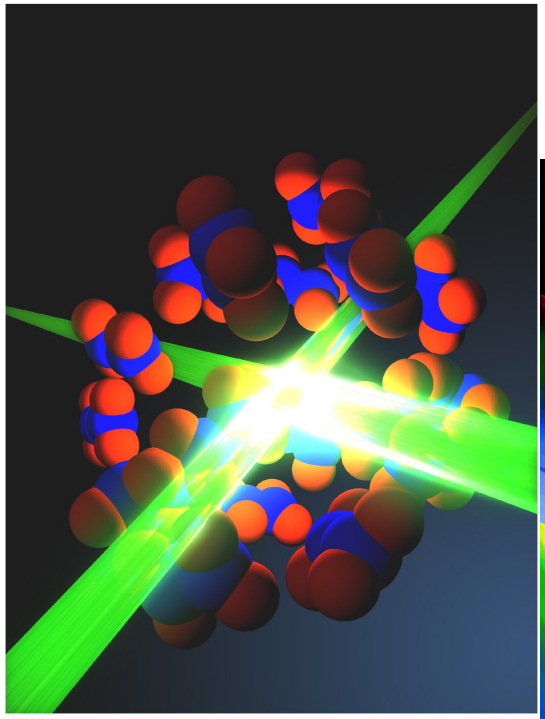
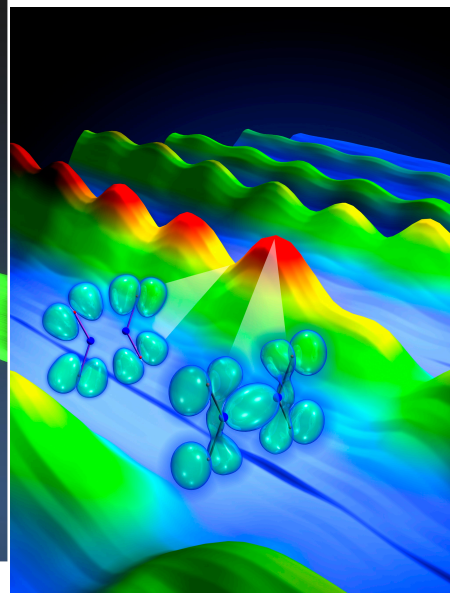
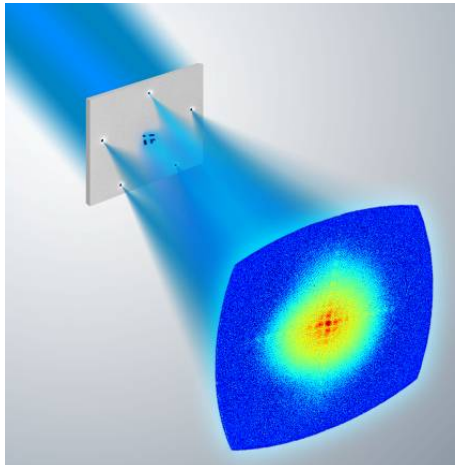
Science at the Time-scale of the Electron - Coherent X-Rays from Tabletop Ultrafast Lasers

Henry Kapteyn and Margaret Murnane







Fermilab, March 31 2010

Colorado
University of Colorado at Boulder

Collaborators

Tenio Popmintchev, Ming-Chang Chen, Paul Arpin, Ivan Christov
Wen Li, Xibin Zhou, Robynne Lock
Chan La-O-Vorakiat, Stefan Mathias, Mark Siemens
JILA, University of Colorado at Boulder

Andreas Becker, Agnieszka Becker, Ronggui Yang
University of Colorado at Boulder

Jorge Rocca, Carmen Menoni, Mario Marconi
Colorado State University

Keith Nelson
MIT

Albert Stolow, Serguei Patchkovskii
NRC Canada

Robin Santra
Argonne National Lab, Illinois

Predrag Ranitovic, Lew Cocke, Xiao-Min Tong
Kansas State University, Manhattan

Anne Sakdinawat, David Attwood, Eric Anderson
University of California, Berkeley and LBL

Tom Silva, Justin Shaw, Hans Nembach
NIST

Janwei Miao
University of California, Los Angeles

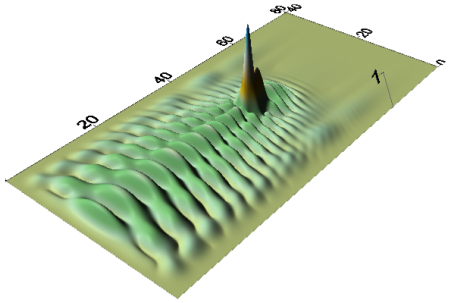
Martin Aeschlimann, Claus Schneider, Roman Adam, Patrick Grychtol
Kaiserslautern and Julich

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*

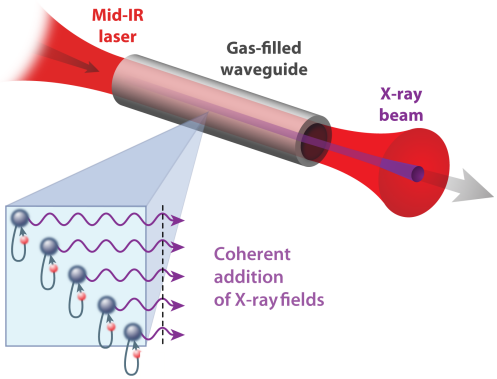
POSTDOCS *Xibin Zhou, Alon Bahabad, Predrag Ranitovic, Stefan Mathias, Tenio Popmintchev*

Science at the timescale of the electron – so what?



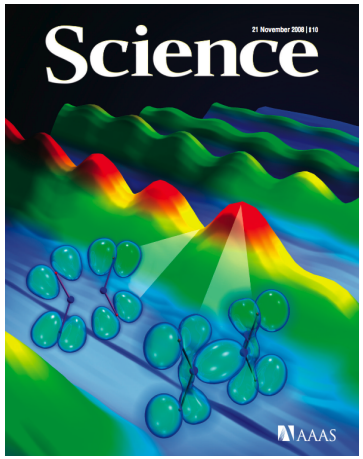
I. Bright coherent x-ray beams on a tabletop

- Ultrafast lasers can manipulate electrons on their fundamental timescale to implement a coherent version of the x-ray tube
- Fastest strobe light in existence, fastest controlled event
- 10 attosecond pulses at 0.5keV
- Zeptosecond pulses coming soon…

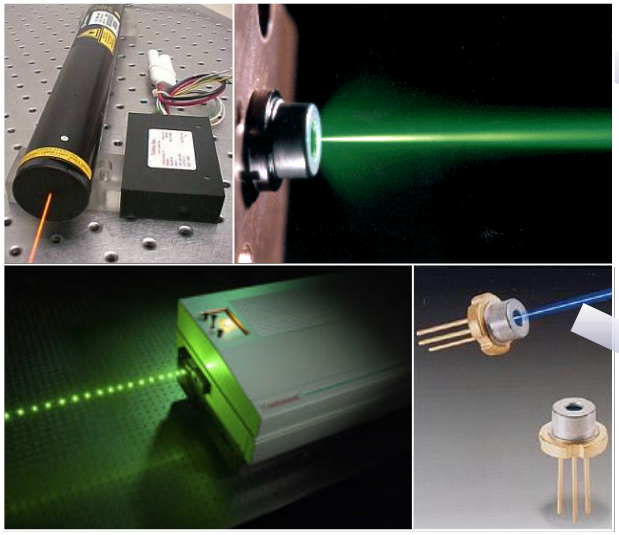


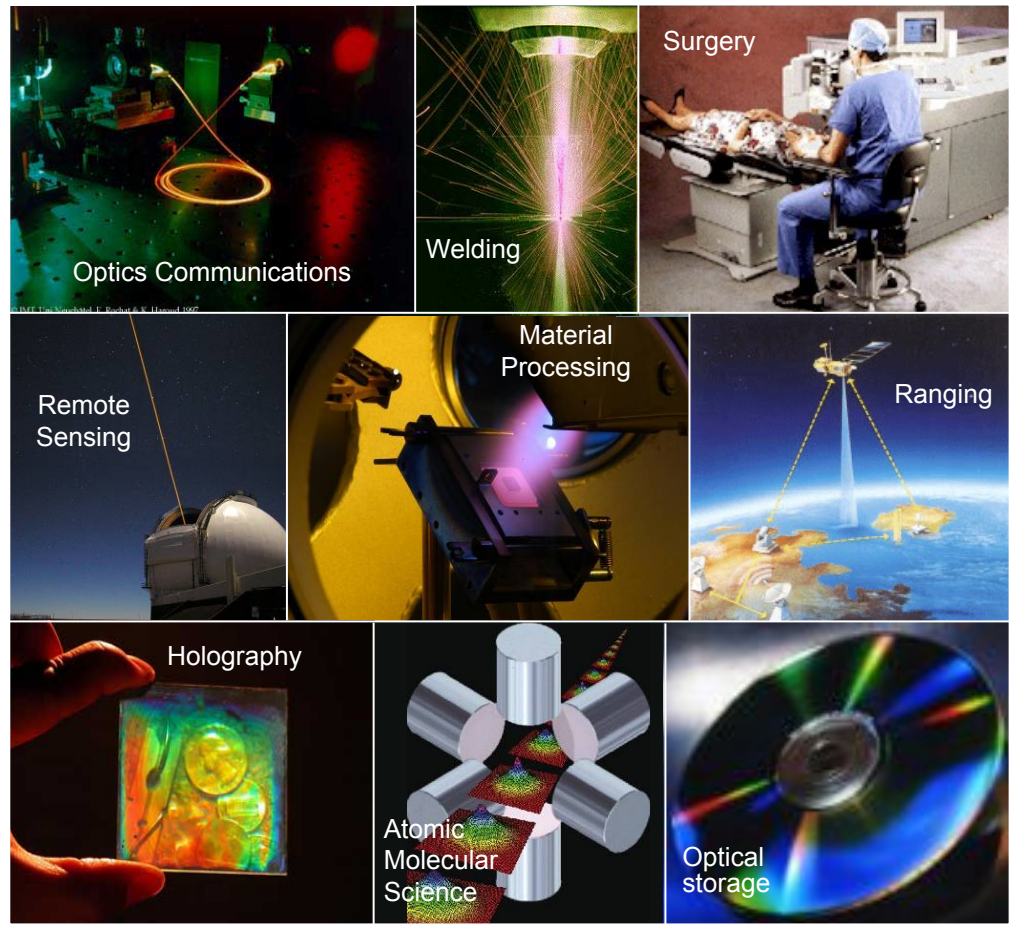
II. Ultrafast x-rays are an ideal probe of the nanoworld

- Capture correlated electron dynamics in molecules and materials
- Understand energy/charge transport at the nanoscale
- High resolution lensless imaging using coherent x-rays
- Revolutionary new technologies for science and industry

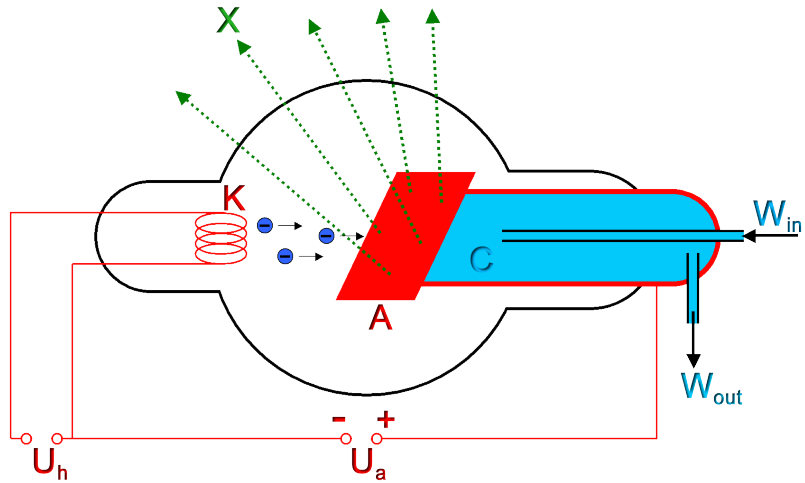


Visible laser light has greatly benefitted society

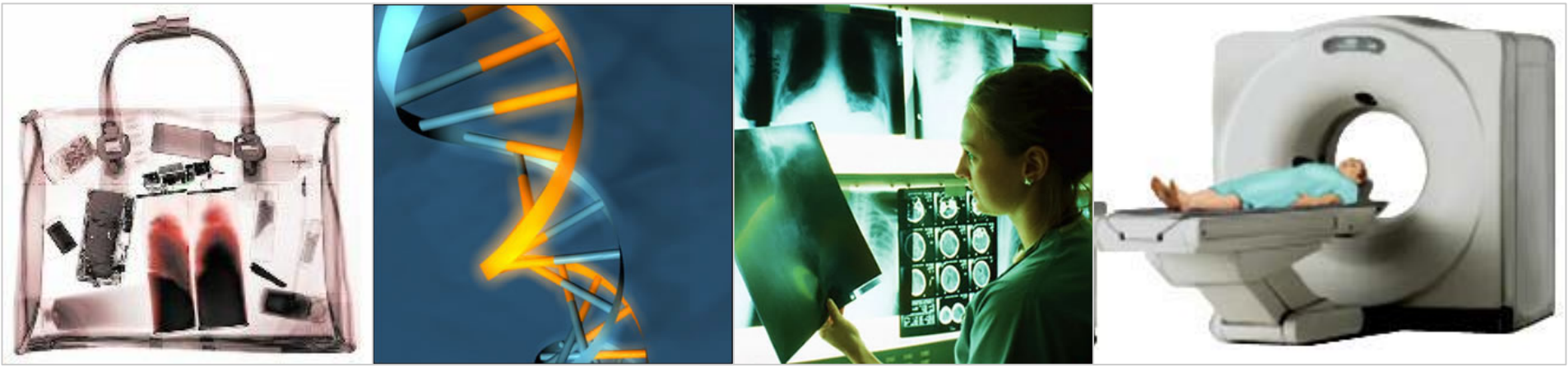




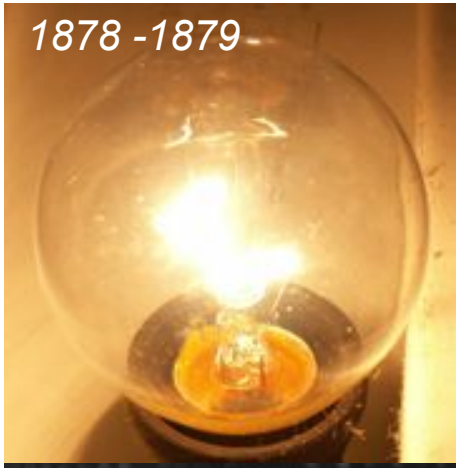
X-ray light also greatly benefits society

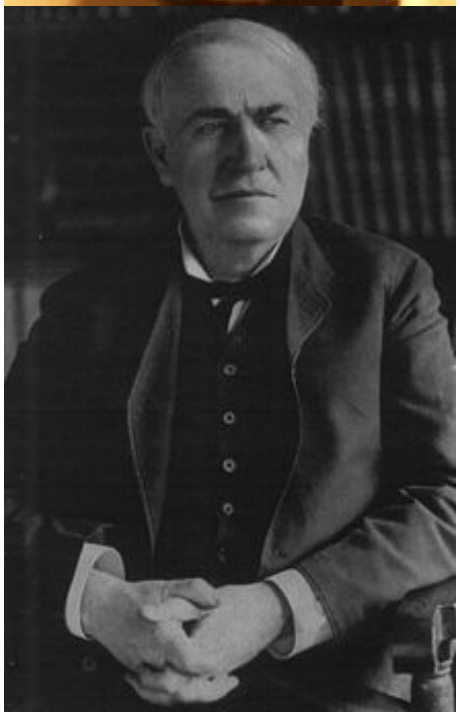


X-ray tube



Incoherent radiation



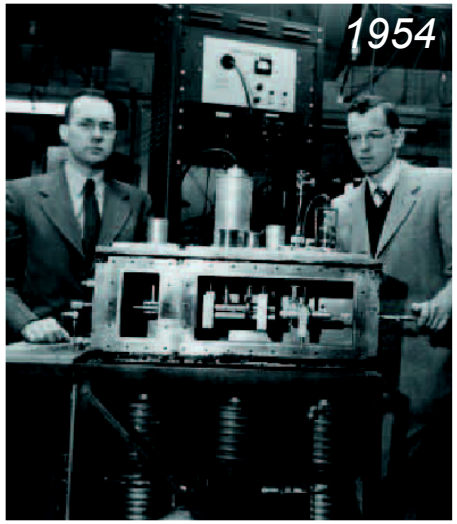


Edison

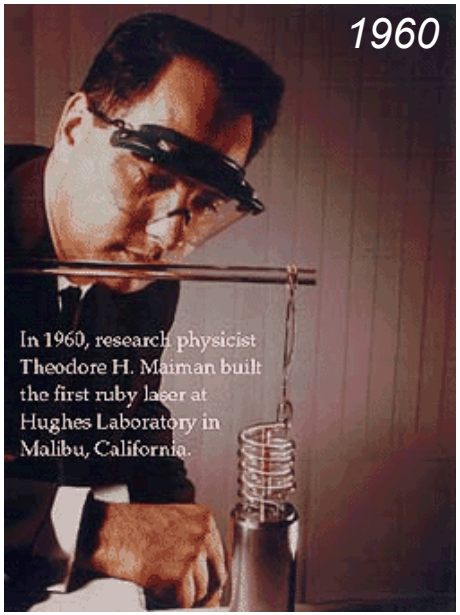
17 years later……

Wilhelm Roentgen

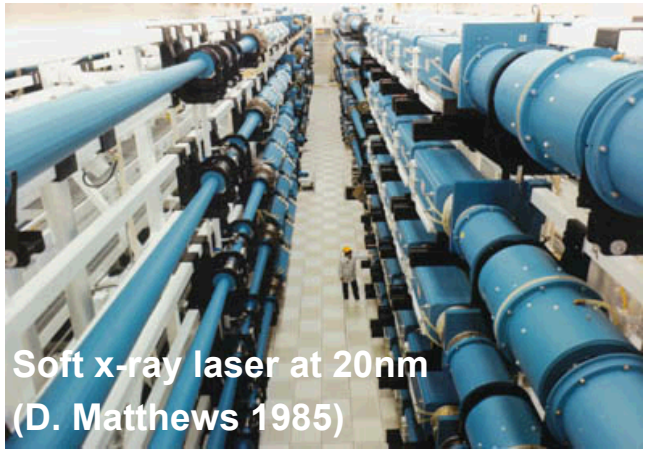
Coherent laser radiation

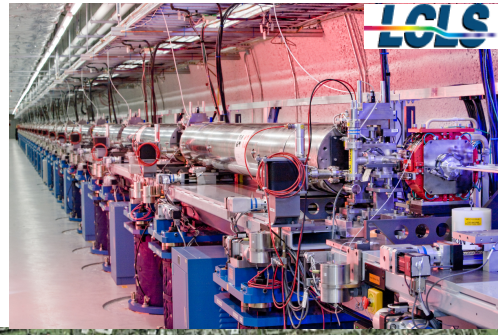


Townes and Gordon with one of their first masers (1954).

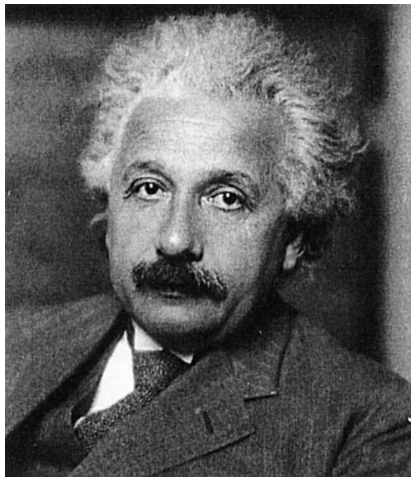


31 years later……





Why are x-ray lasers so challenging to build?



Spontaneous emission / Stimulated emission

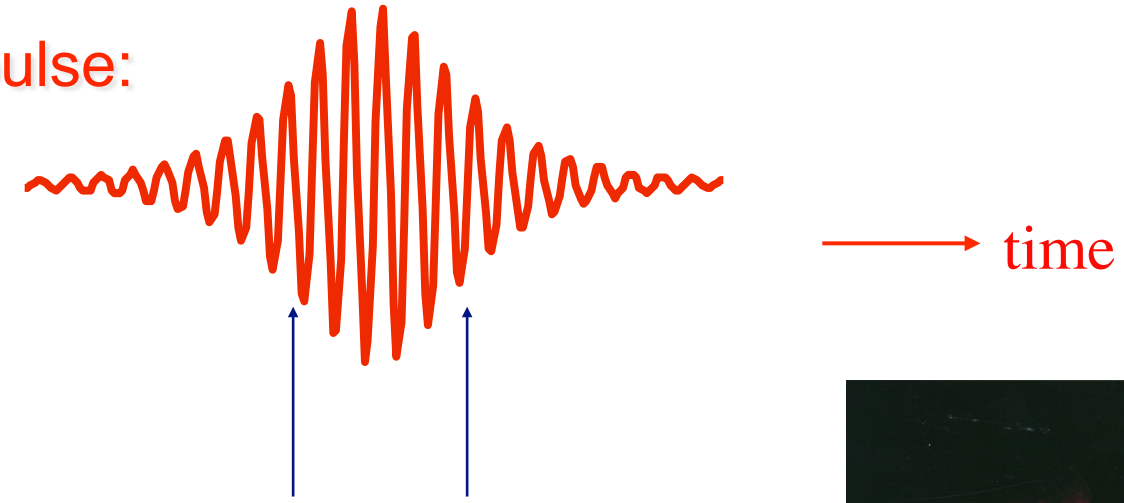
$$\frac{A_{21}}{B_{21}} = \frac{8\pi h\nu^3}{c^3} \propto \nu^3$$

$$Power \propto \left(\frac{1}{\sigma_g}\right)\left(\frac{1}{\tau}\right)(h\nu) \propto \frac{1}{\lambda^5}$$

- **1 μm -> 1 mW**
- **1 nm -> TW**
- **1 Å -> 1 PW**

Start with an ultrafast laser pulse

10 fs light pulse:

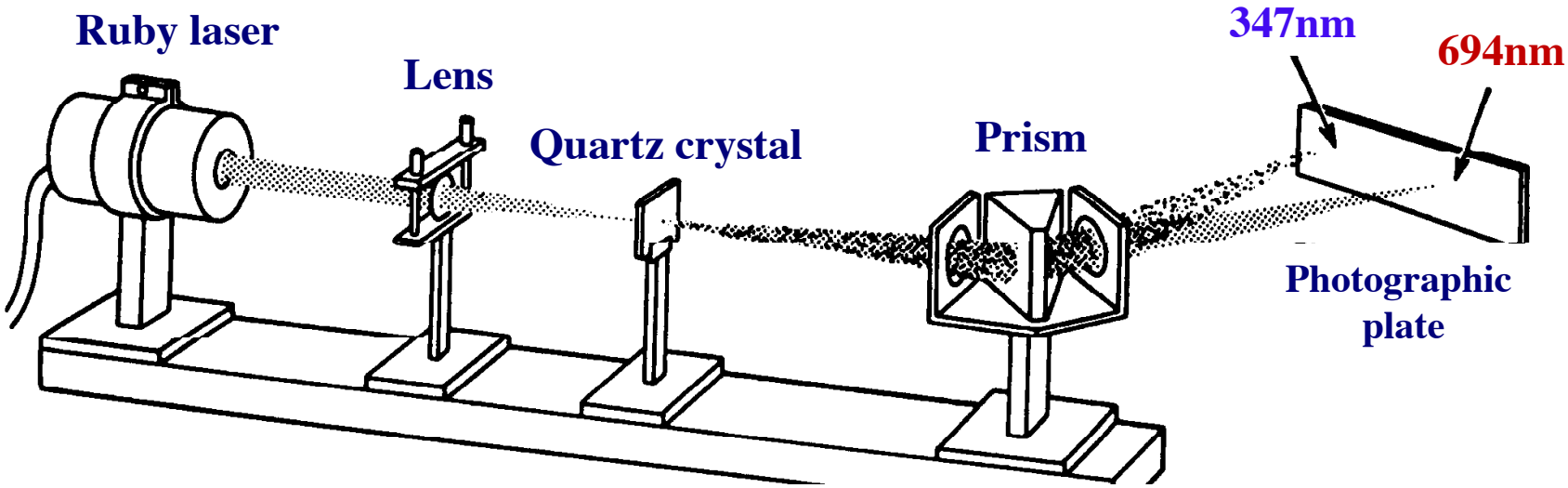


Δx=3 micrometers
= 1/50 human hair

First 10 fs Ti:sapphire laser
Optics Letters **18**, 977 (1993)

The birth of Nonlinear Optics – second harmonic generation

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



VOLUME 7, NUMBER 4 PHYSICAL REVIEW LETTERS AUGUST 15, 1961

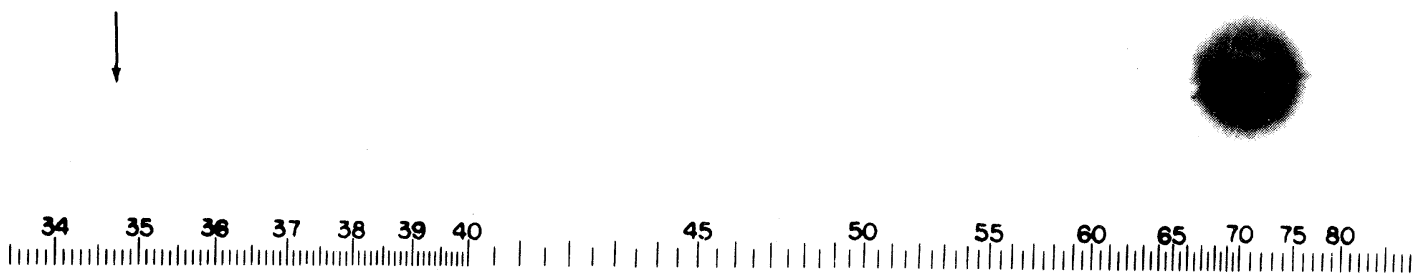


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

The birth of Nonlinear Optics – second harmonic generation

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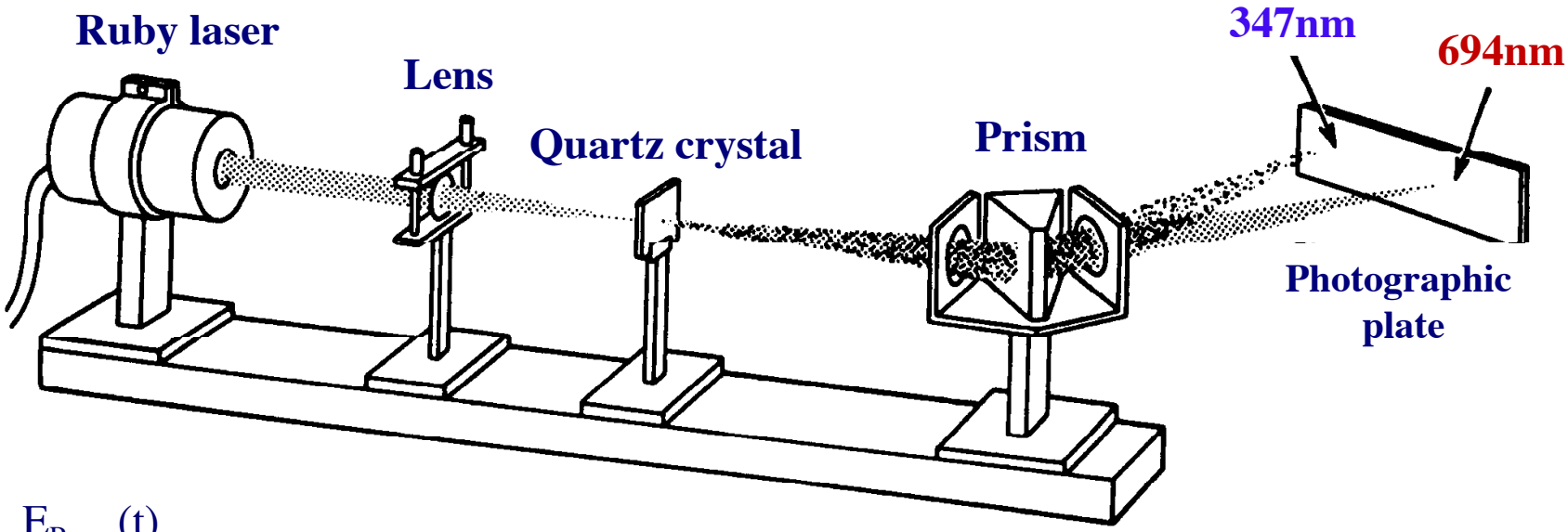
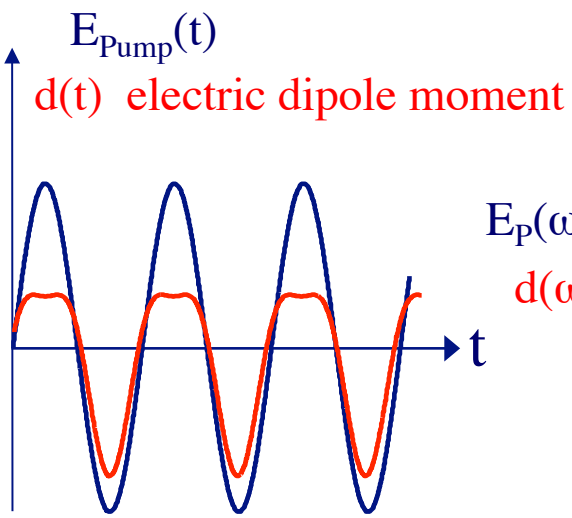
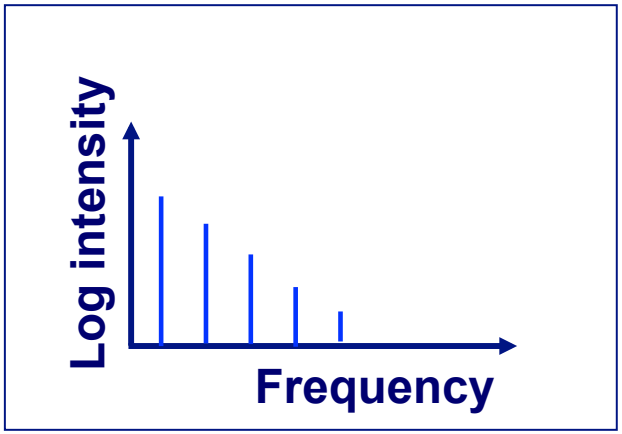




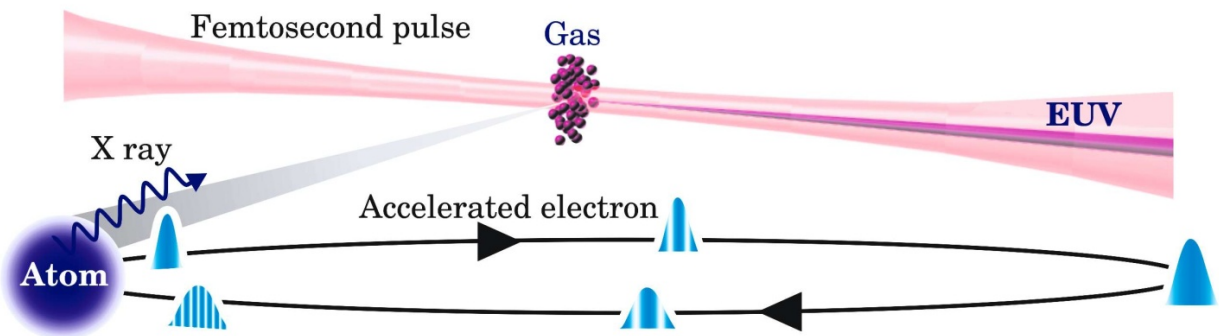
$E_P(\omega)=E_0^{i\omega t}$

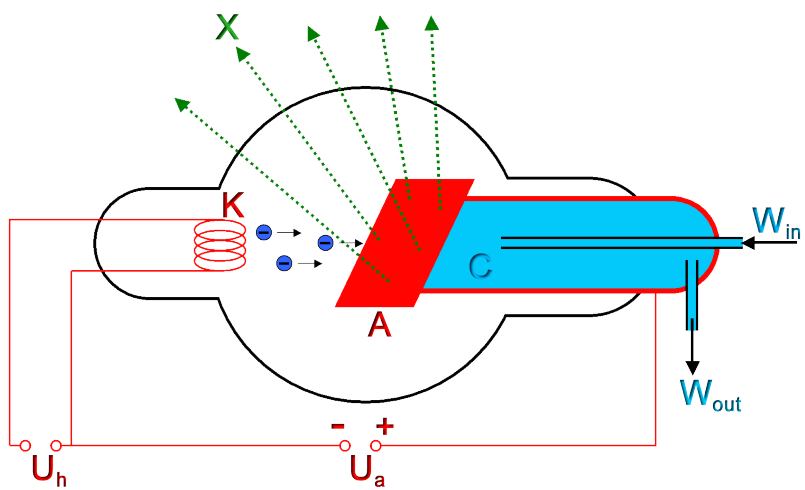
$d(\omega)=a_1 e^{i\omega t} + a_2 e^{i2\omega t} + \ldots$



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



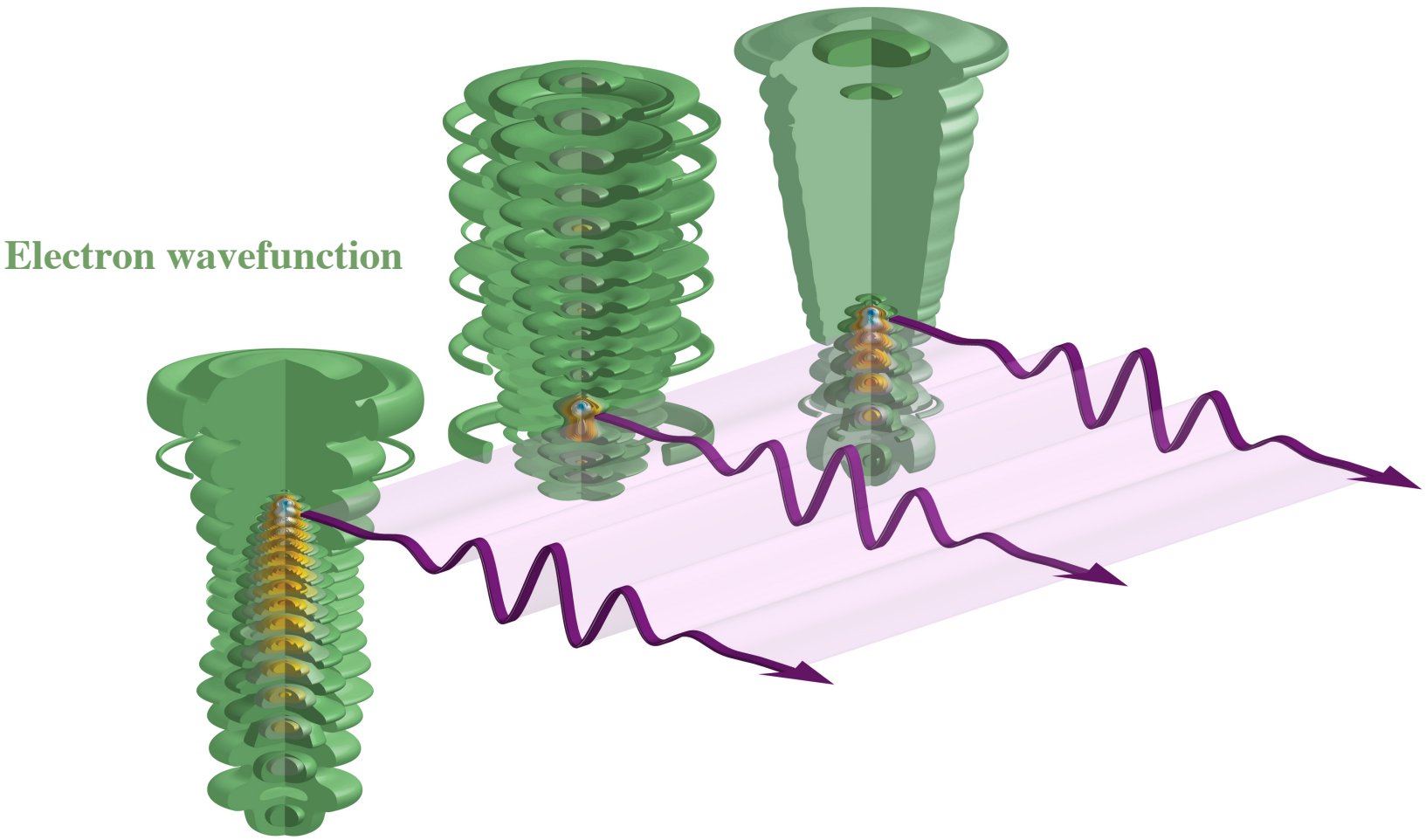


X-ray tube

*SILAP Proc. pp. 95-110 (1993); PRA **49**, 15 (1994)*

High Harmonic Generation – quantum picture

- Coherent x-rays are generated by focusing an intense laser into a gas
- Broad range of harmonics generated simultaneously from UV - keV



High Harmonic Generation - extreme nonlinear optics

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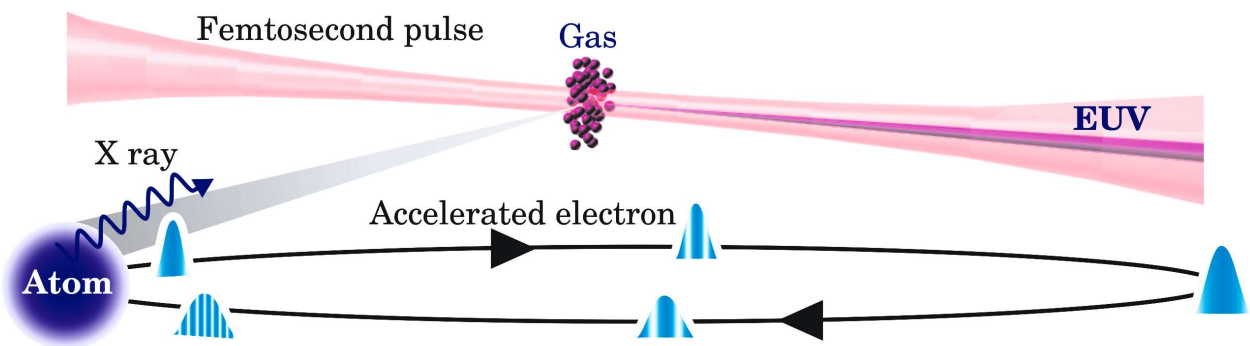
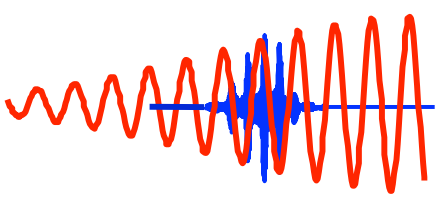
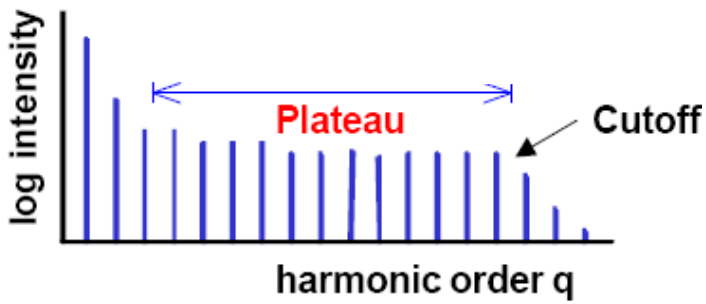




< 10 fs duration



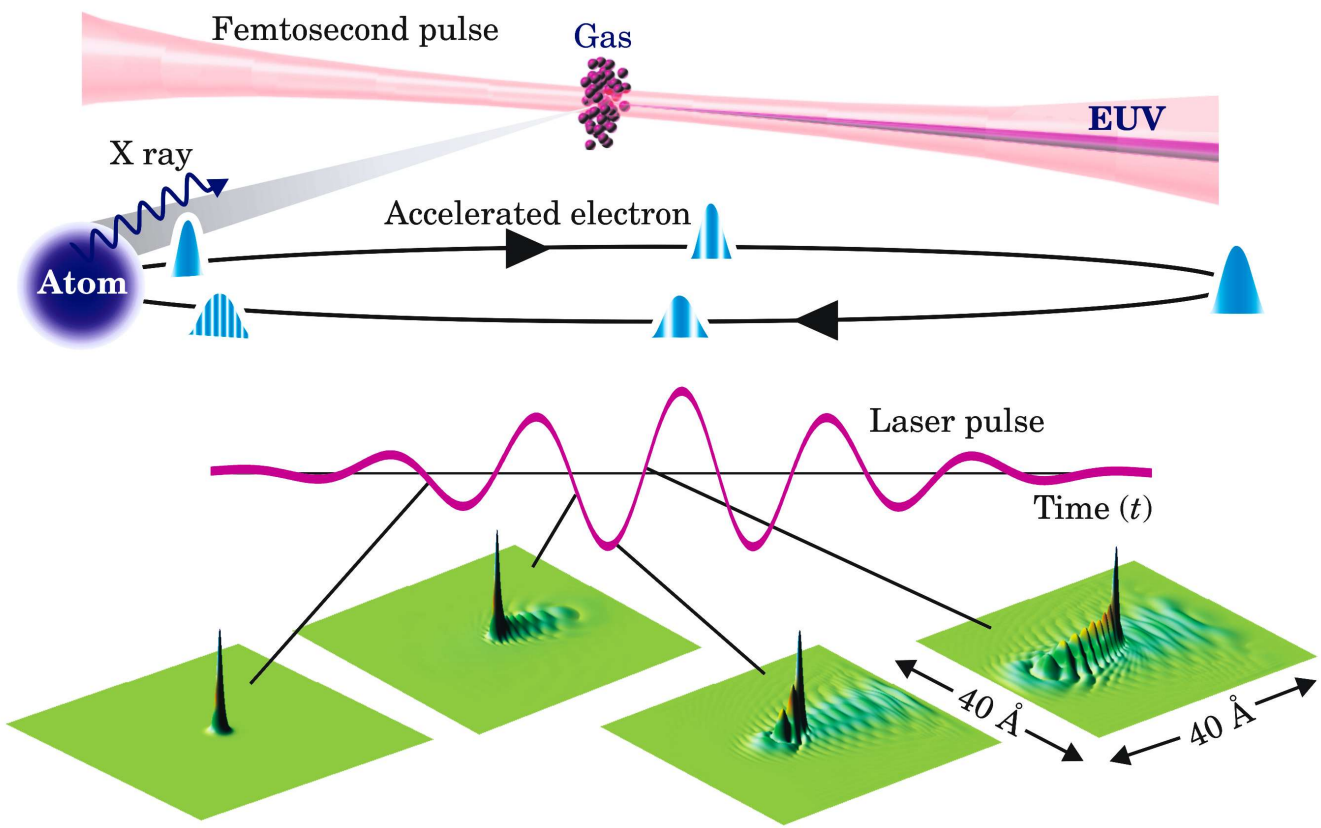
Broad frequency range UV - keV

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

Electron wriggle energy coherently converts to x-rays

High Harmonic Generation – quantum picture

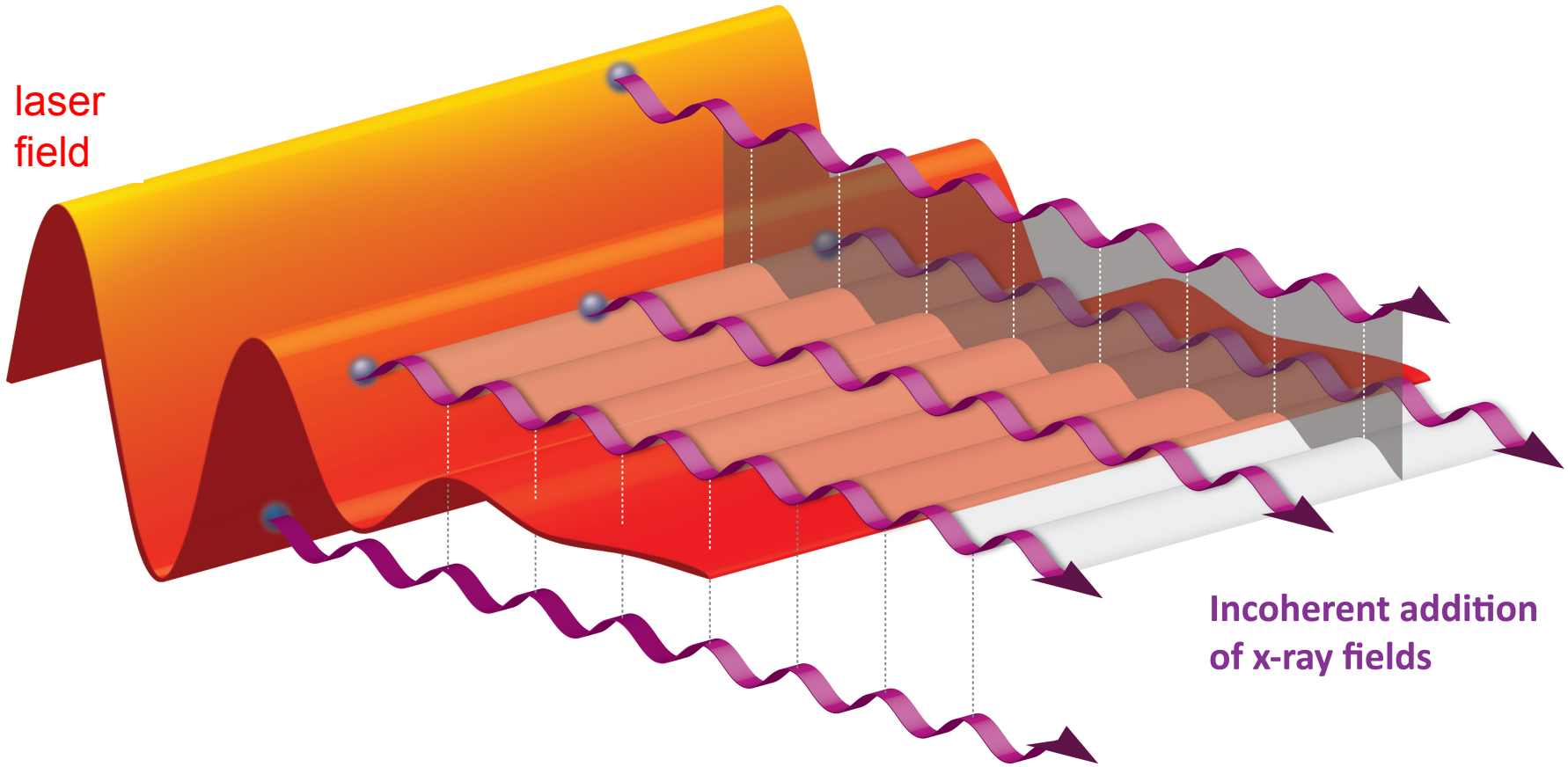
- Coherent x-rays are generated by focusing an intense laser into a gas
- Broad range of harmonics generated simultaneously from UV - keV



Electron wavefunction

Generating bright, coherent, x-ray beams

- Bright harmonics require the coherent addition of waves from many atoms
- When laser and x-ray phase velocities matched, get coherent bright output



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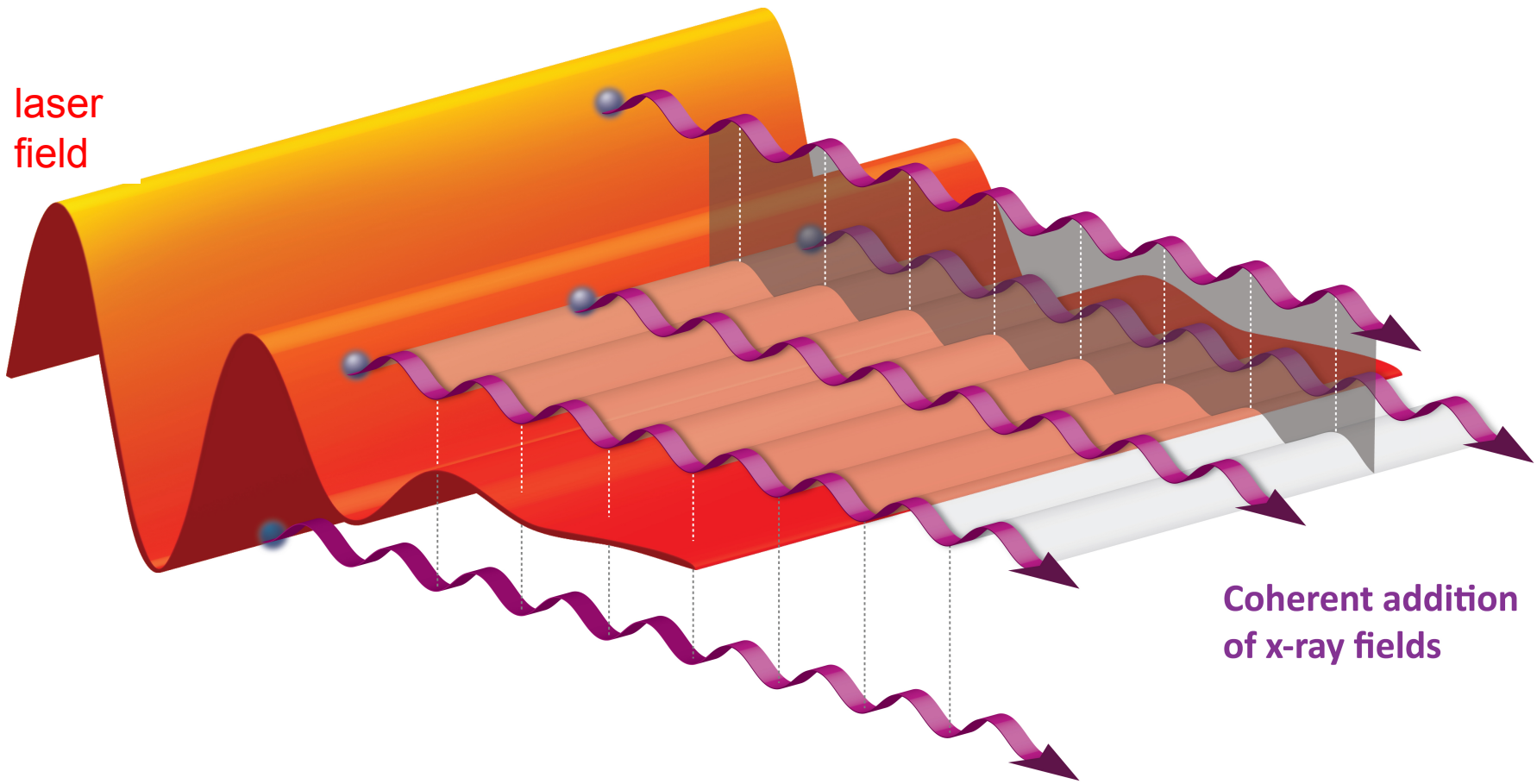
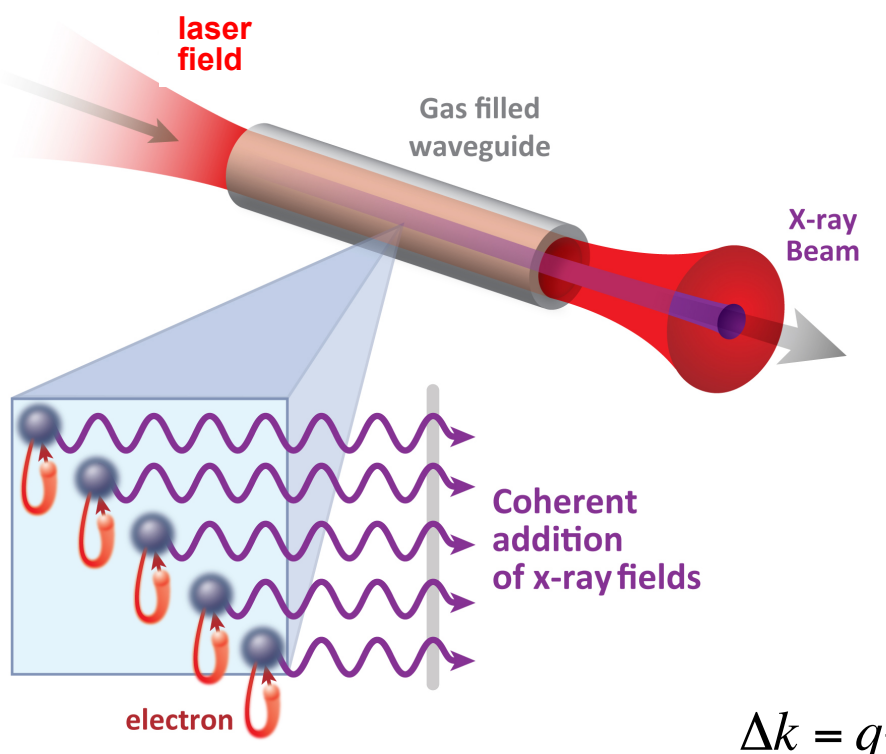
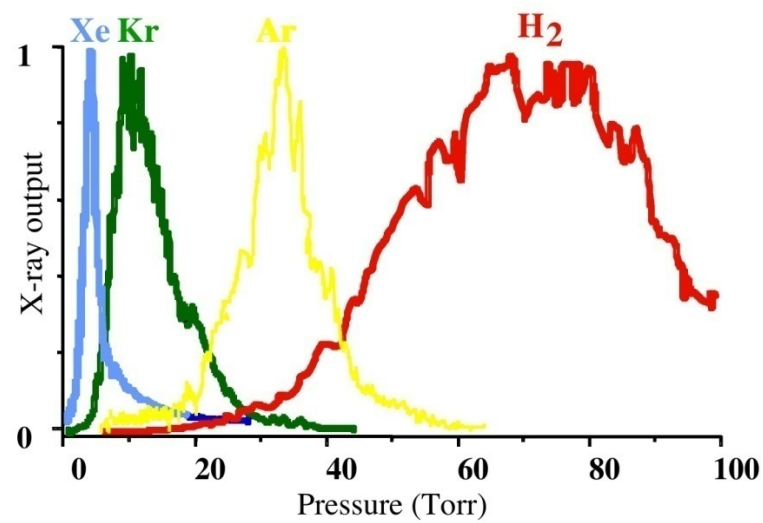


How to match the laser and x-ray phase velocities?

- Place gas inside a hollow fiber
- Tune the gas pressure to equalize the laser and x-ray phase velocities





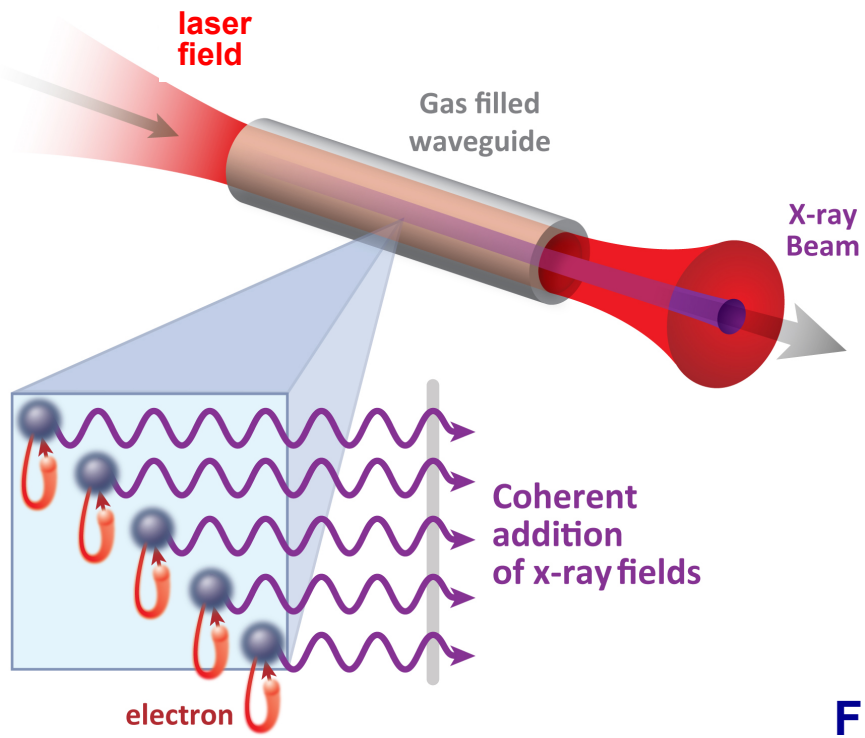
$$\Delta k = q k_{laser} - k_{HHG} = 0$$

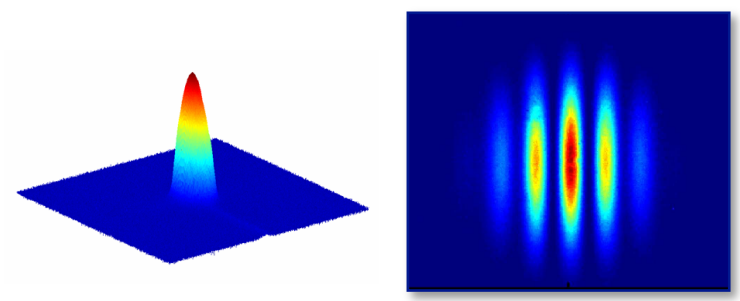
$$\Delta k = q\left\{\left(\frac{u_{11}^2 \lambda_0}{4\pi a^2}\right) - P\left((1-\eta)\frac{2\pi}{\lambda_0}\Delta\delta - \eta\left[N_{atm} r_e \lambda_0\right]\right)\right\}$$

Waveguide **Neutrals** **Plasma**

Generating bright, coherent, x-ray beams

- Tune the gas pressure to equalize the laser and x-ray phase velocities
- Generate fully coherent, bright, x-ray beams





Fully coherent bright EUV and soft x-rays

nJ per harmonic, uW average powers

Femtosecond-to-attosecond duration

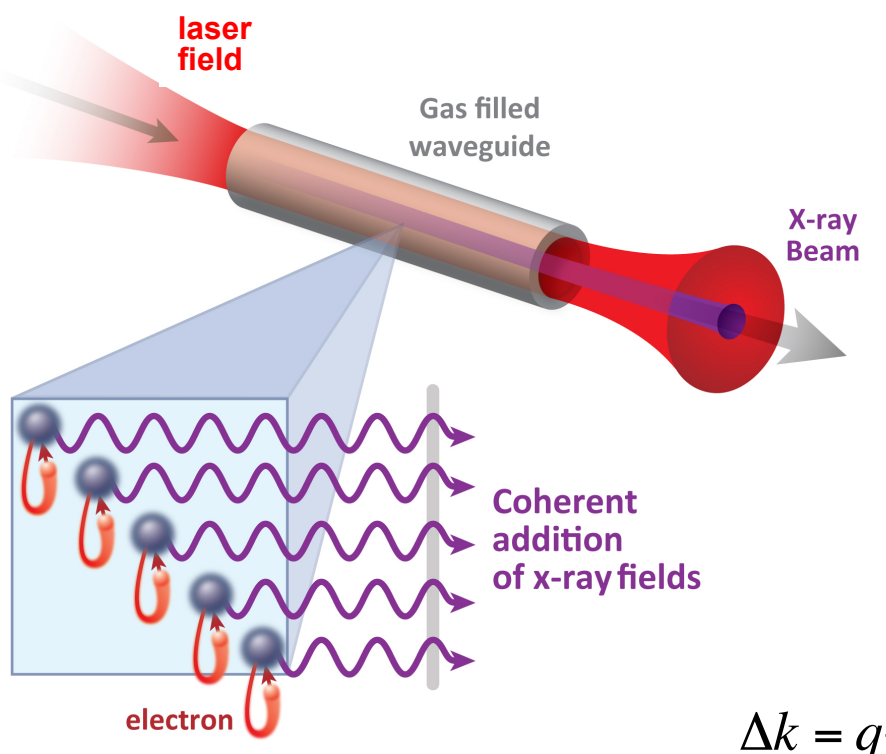
*Science **280**, 1412 (1998)*

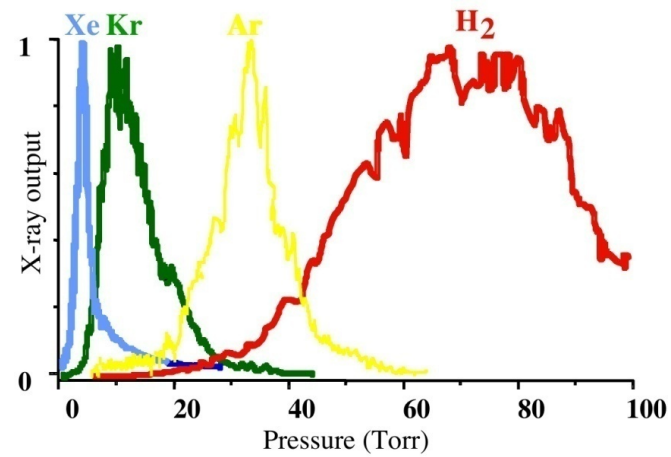
*Science **297**, 376 (2002)*

*Science **317**, 775 (2007)*

Limits of phase matching

- To generate high energy x-rays, need high electron energy - ionize gas
- Presence of plasma speeds up laser too much – no phase matching



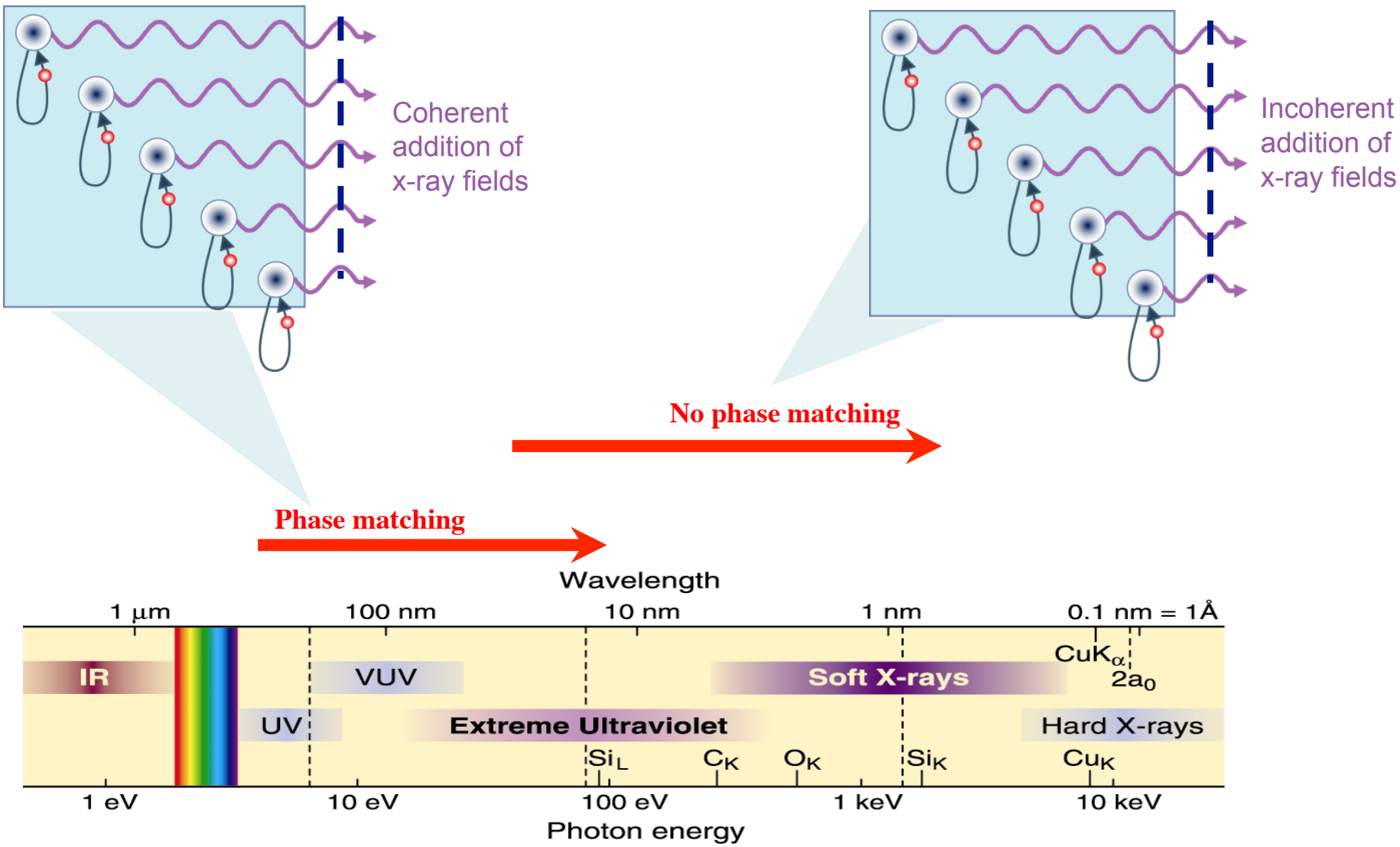


$$\Delta k = q k_{laser} - k_{HHG} = 0$$

$$\Delta k = q\left\{\left(\frac{u_{11}^2 \lambda_0}{4\pi a^2}\right) - P\left((1-\eta)\frac{2\pi}{\lambda_0}\Delta\delta - \eta\left[N_{atm} r_e \lambda_0\right]\right)\right\}$$

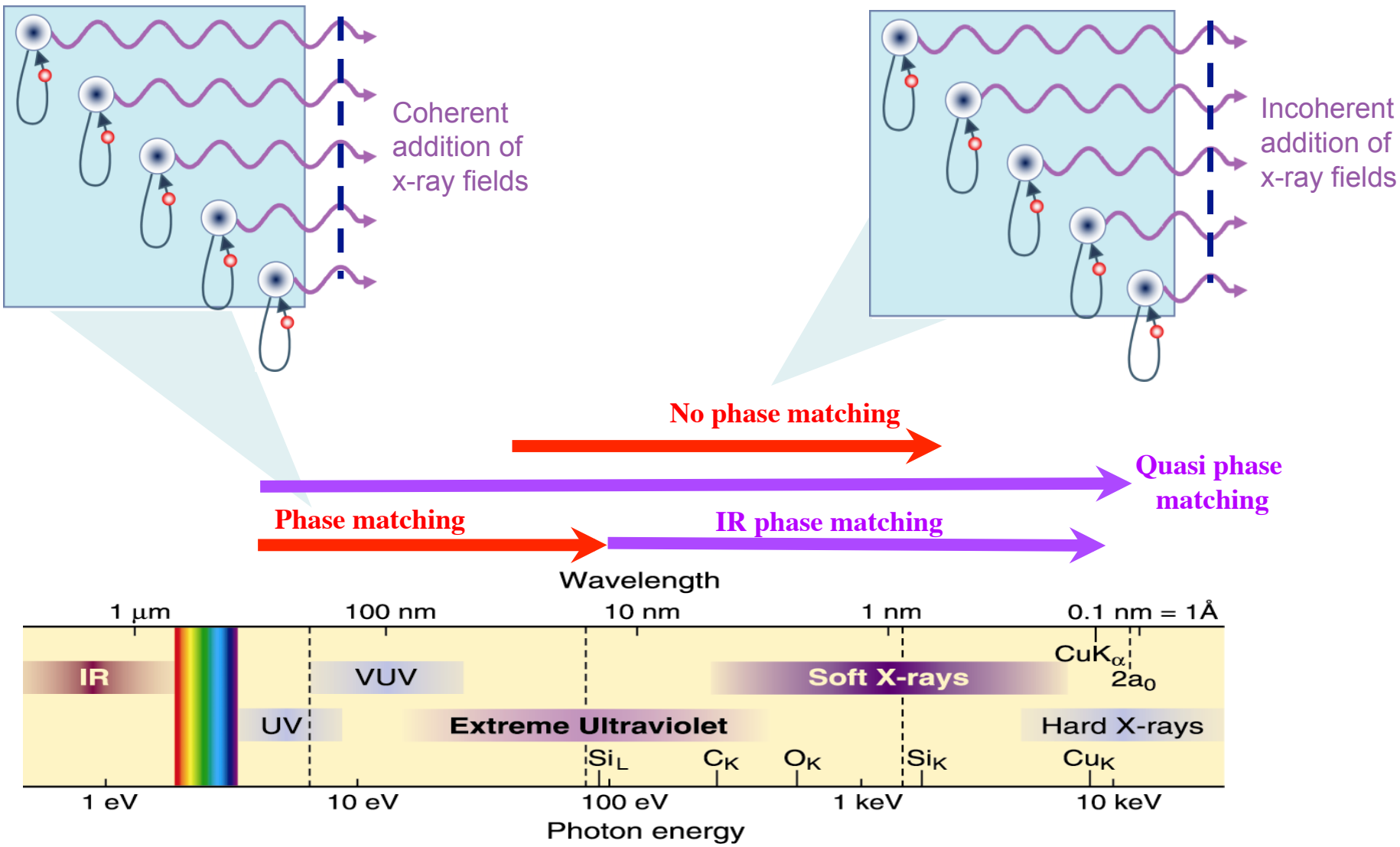
Waveguide **Neutrals** **Plasma**

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

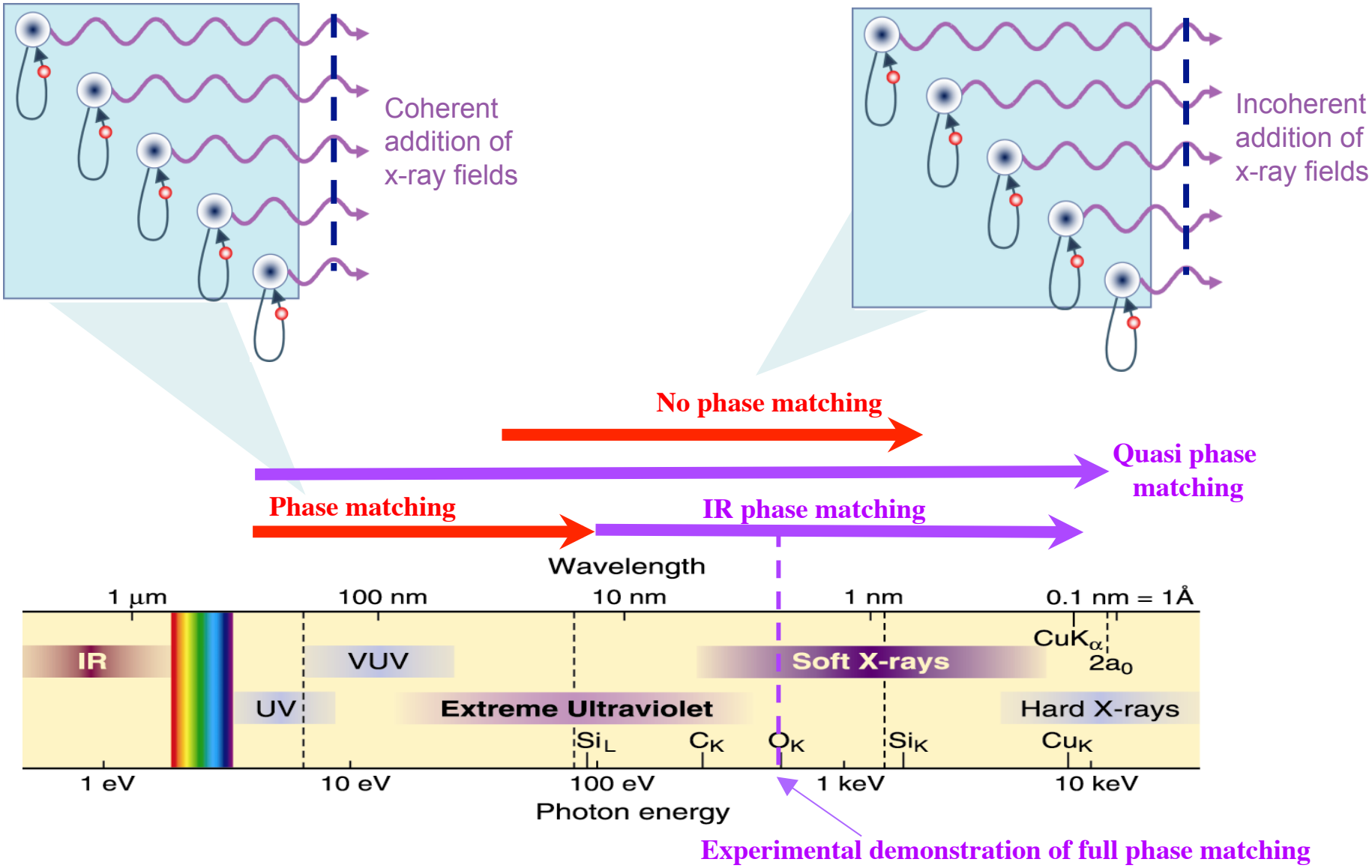


10^{11} ph/sec *10^{10} ph/sec* *10^{7} ph/sec* *100 ph/sec*

New phase matching techniques allow laser beams to be converted into laser-like beams of hard x-rays



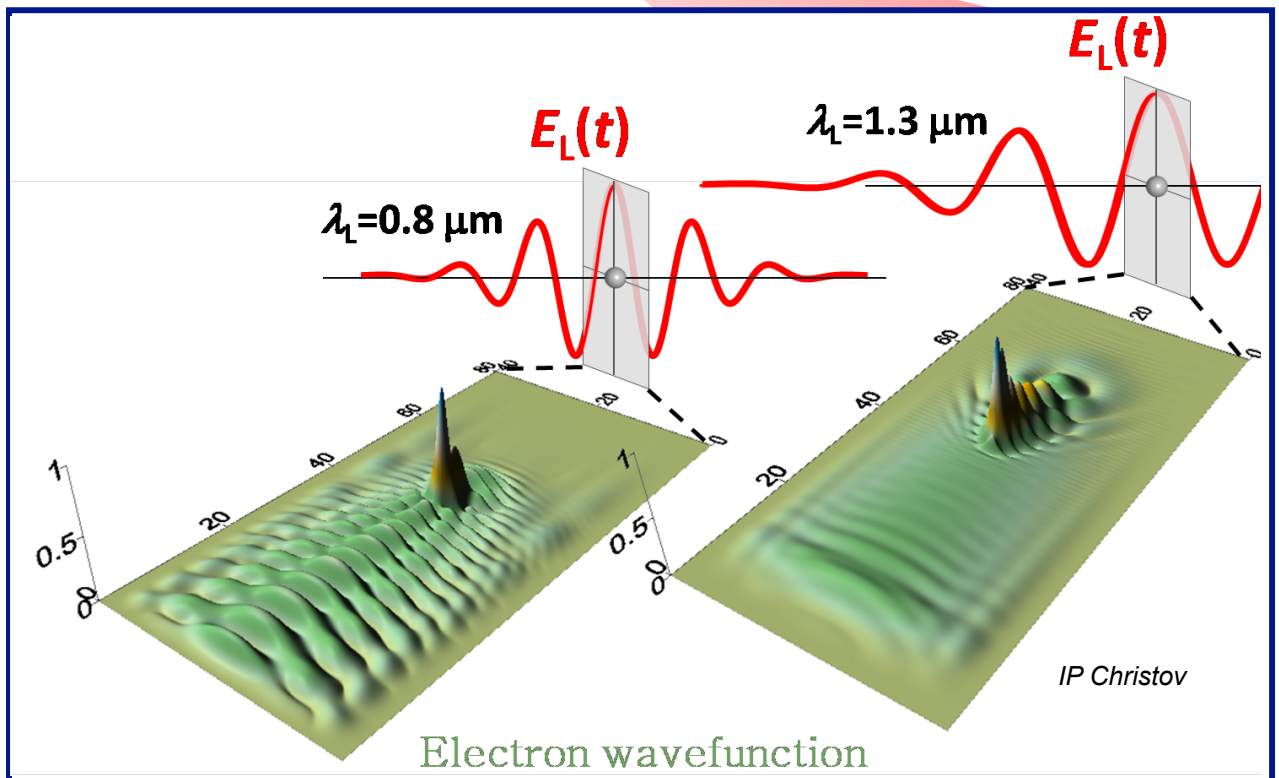
Bright soft x-ray beams demonstrated at > 0.5 keV



High harmonic generation using long wavelength lasers

Maximum photon energy: $h\nu_{cutoff} \propto I_L \lambda_L^2$

- Using long wavelength lasers allows maximum photon energy to increase
- But… quantum spread of electron wavefunction is greater for longer cycle
- Single atom yield decreases rapidly, scaling as $\lambda^{-5.5}$
- But….phase matching can overcome this limitation

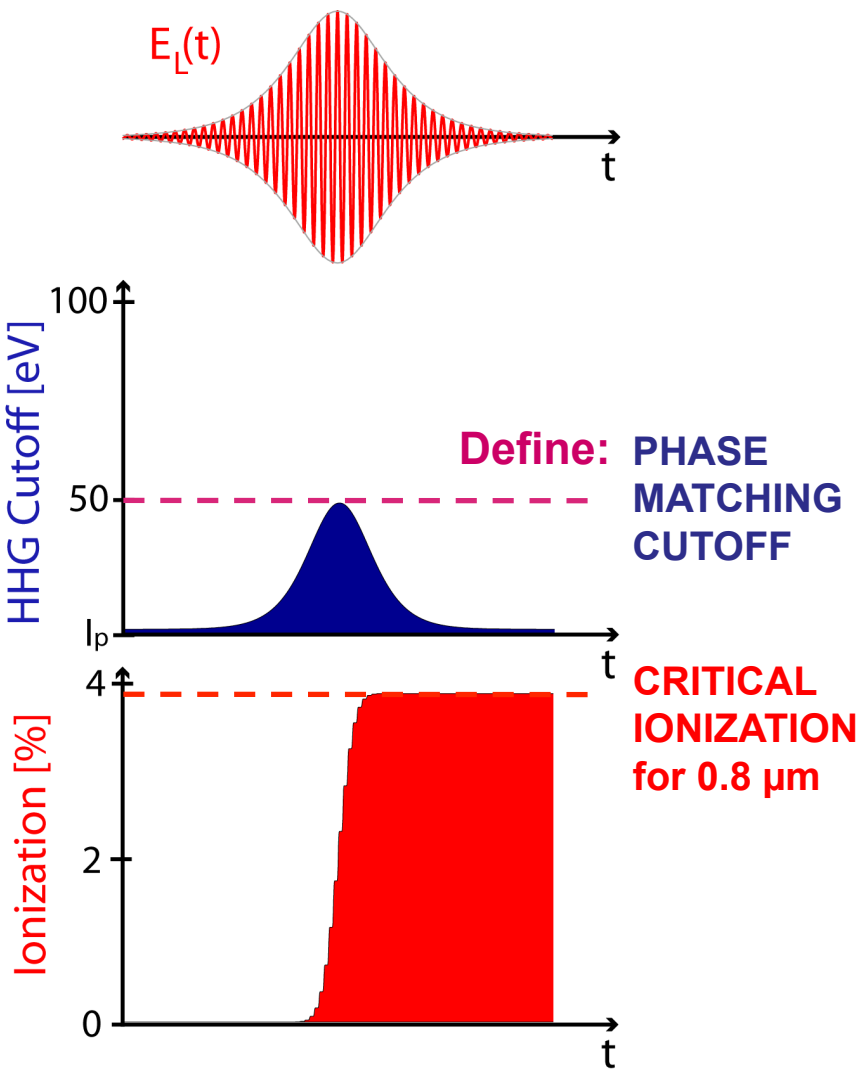


*Tate et al.,PRL **98**, 013901 (2007);*
*Schiessl et al., PRL **99** 253903 (2007);*
*Frolov et al., PRL **100**, 173001 (2008).*

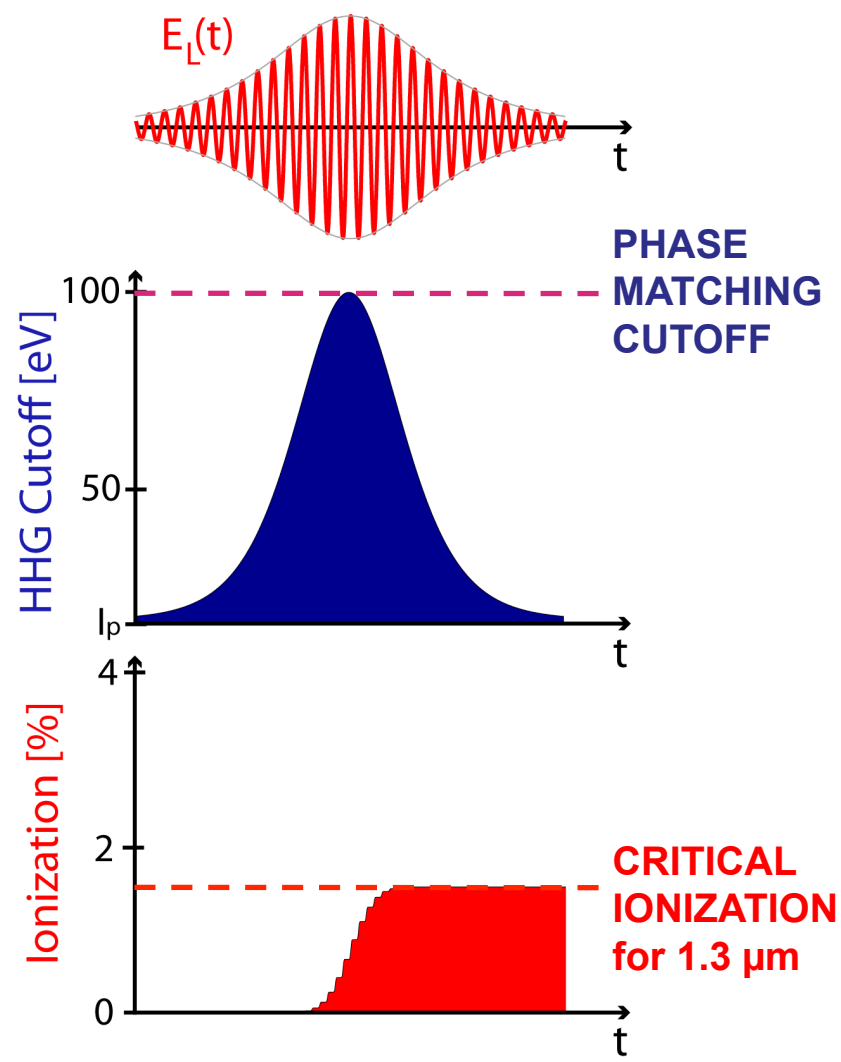
What about *phase matching* using longer laser wavelengths?

$$h\nu_{cutoff} = I_p + 3.2\, I_L \lambda_L^2$$

Ar
0.8 μm laser, 8 cycles FWHM



Ar
1.3 μm laser, 8 cycles FWHM



Predictions for phase matching using IR lasers

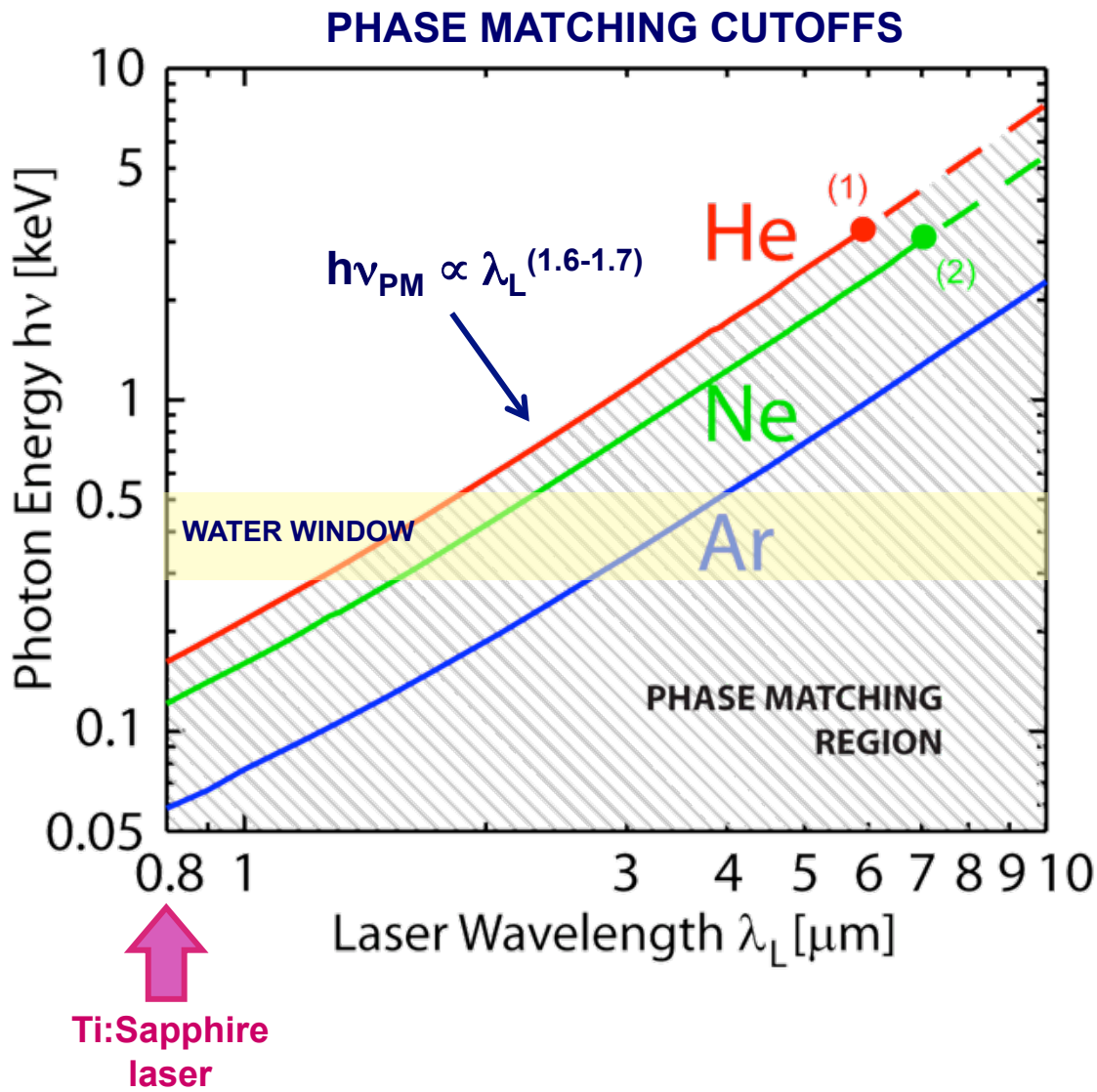
$$h\nu_{max} \propto I_L \lambda_L^2$$

IR lasers need lower intensity for a given harmonic energy

Lower laser intensity = > lower ionization, better PM

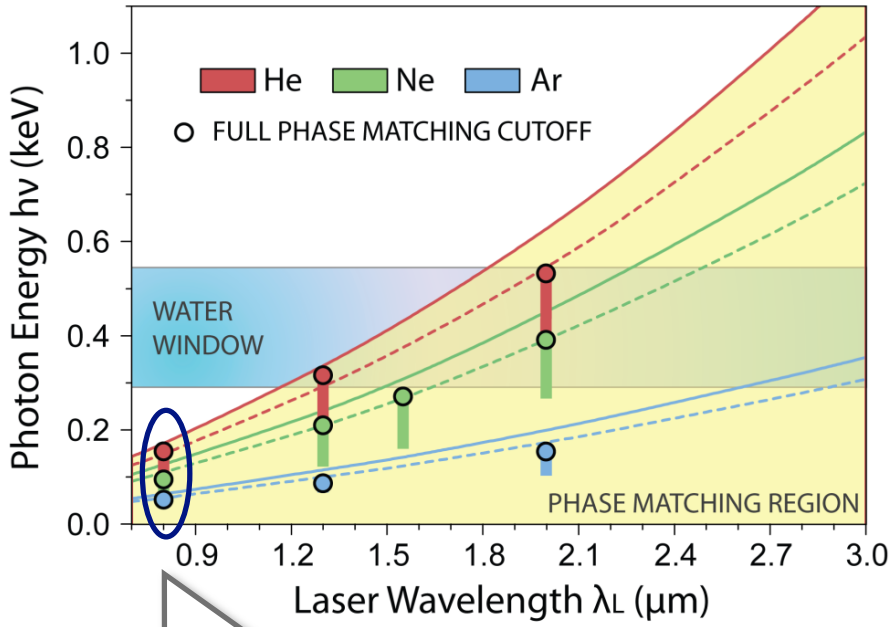
Single atom response also lower for mid-IR drivers (λ^{-5})

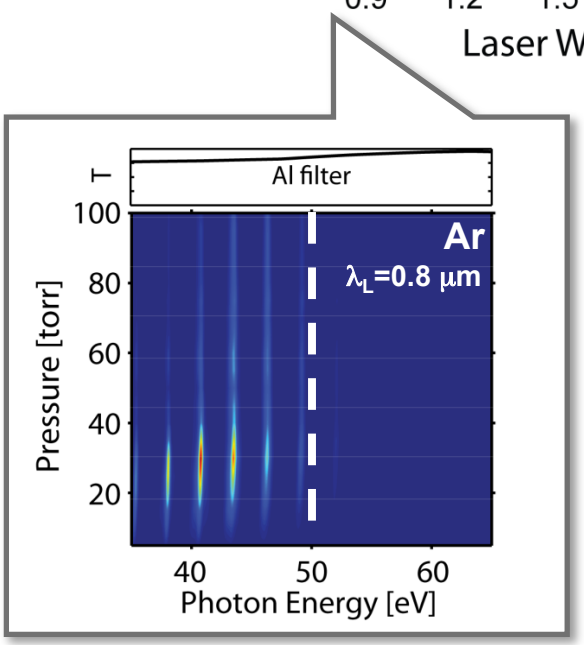
BUT phase matching pressure and gas transparency increase!



T. Popmintchev et al.
CLEO Postdeadline CPDA9 (2008)
*Optics Letters **33**, 2128 (2008)*
*PNAS, **106**, 10516 (2009)*
Nature Photonics, tbp (2010)

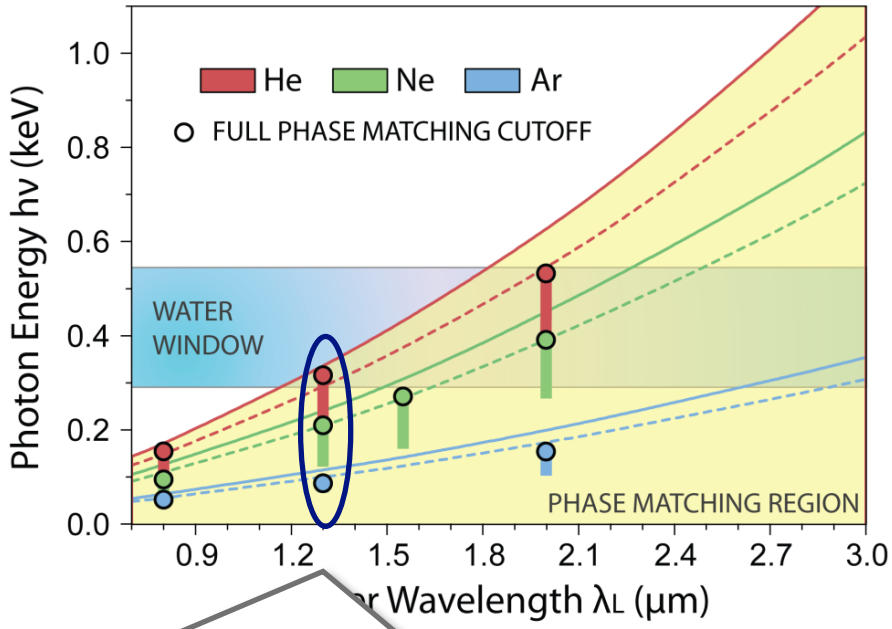
Full phase matching at >0.13keV using λ_L= 0.8μm



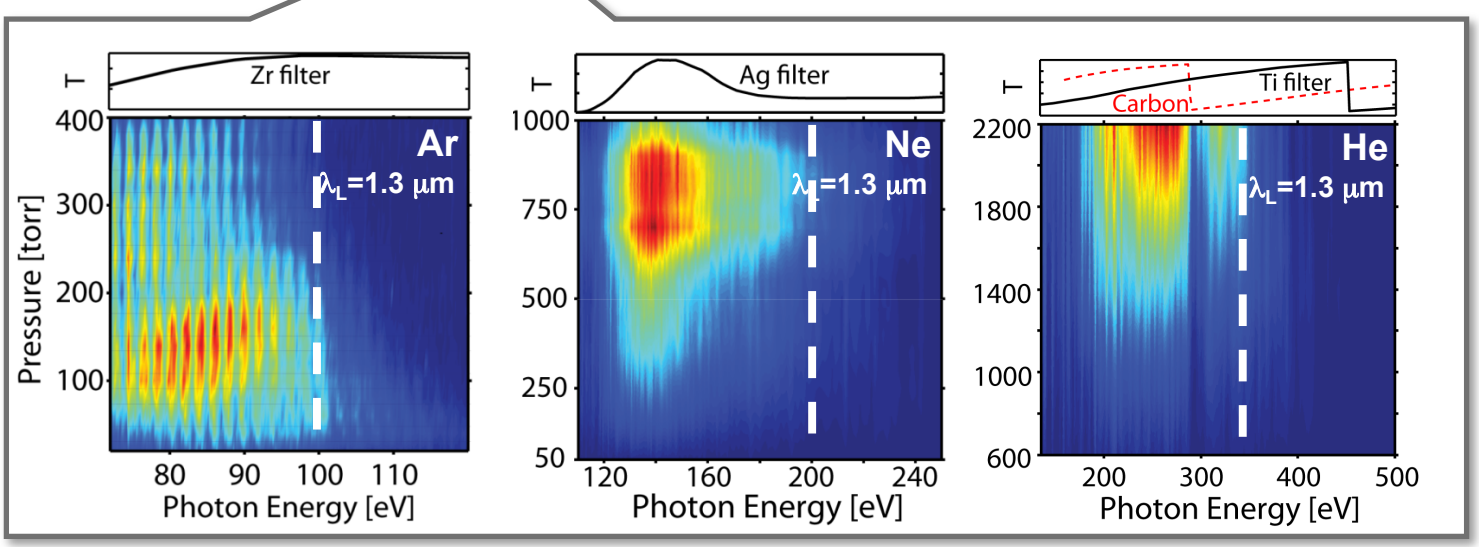


Popmintchev et al.,
CLEO Postdeadline CPDA9 (2008);
Opt. Lett. ***33****, 2128 (2008);*
PNAS ***106****, 10516 (2009);*
Nature Photonics to be publ. (2010)

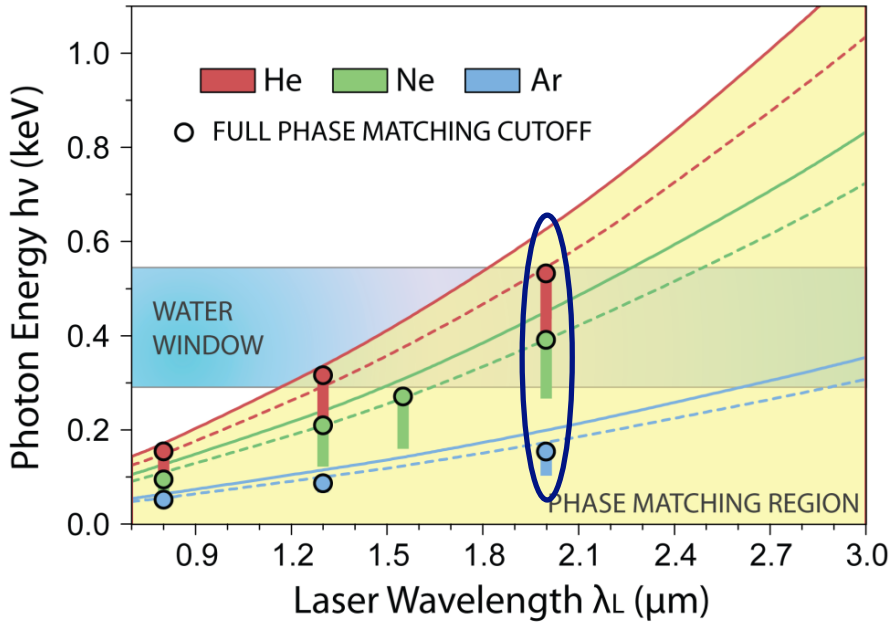
Full phase matching at >0.3keV using λ_L = 1.3μm

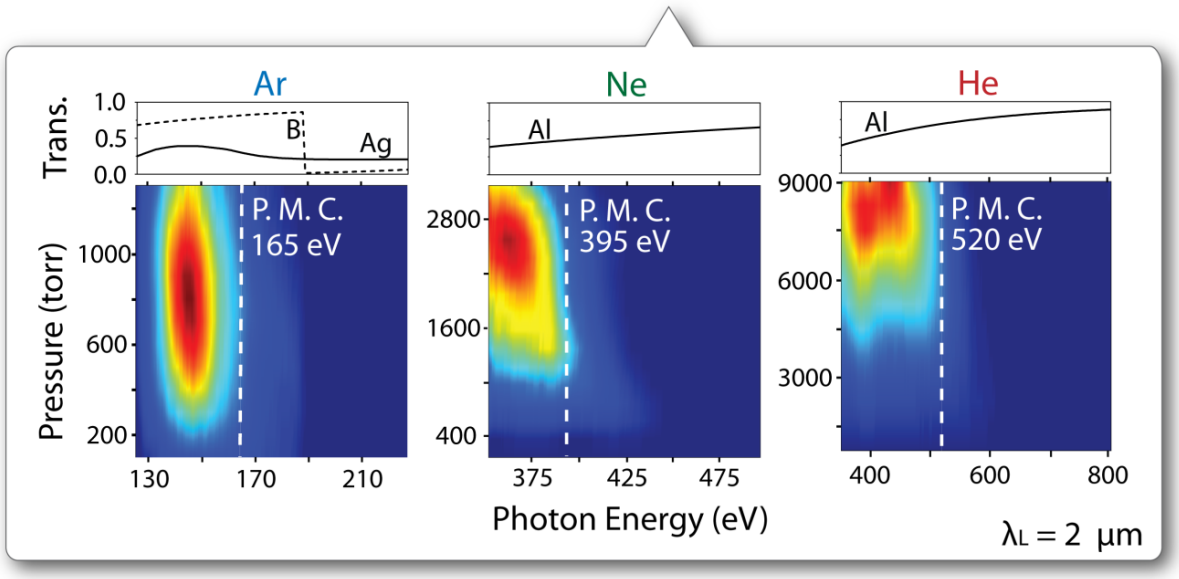


Popmintchev et al.,
CLEO Postdeadline CPDA9 (2008);
Opt. Lett. ***33****, 2128 (2008);*
PNAS ***106****, 10516 (2009);*
Nature Photonics to be publ. (2010)

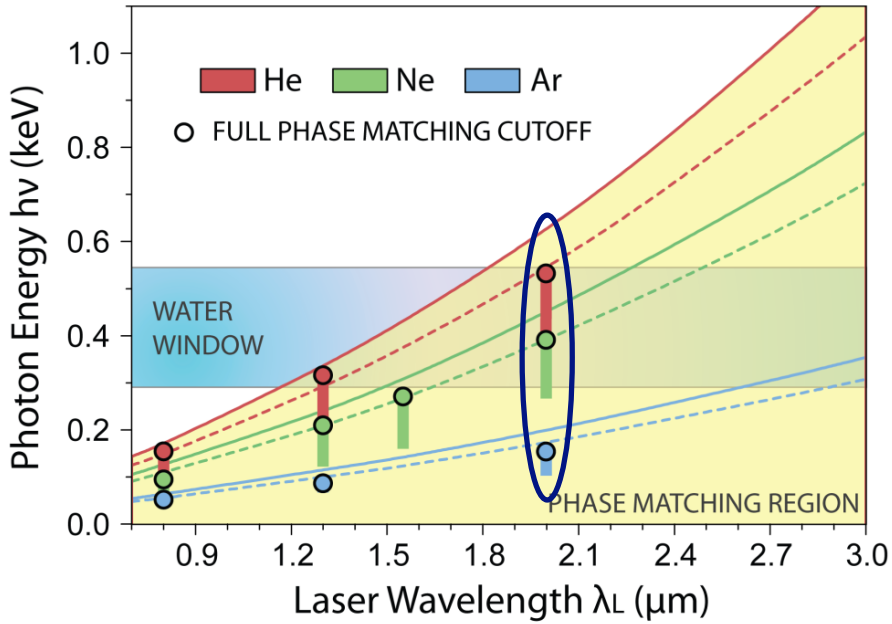


Full phase matching at >0.5keV using λ_L = 2μm

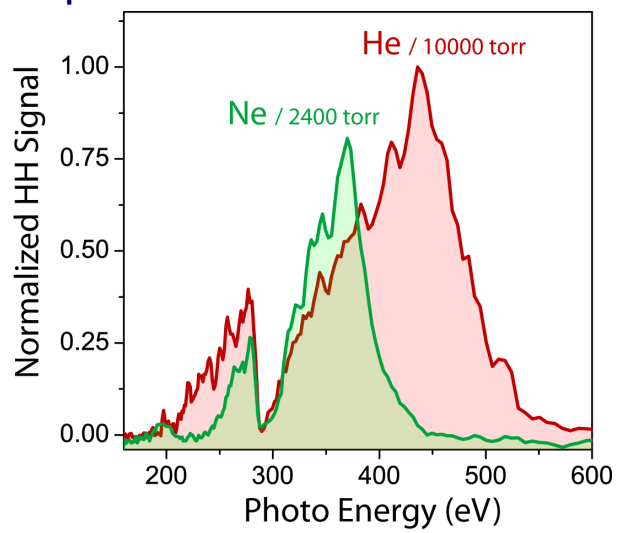


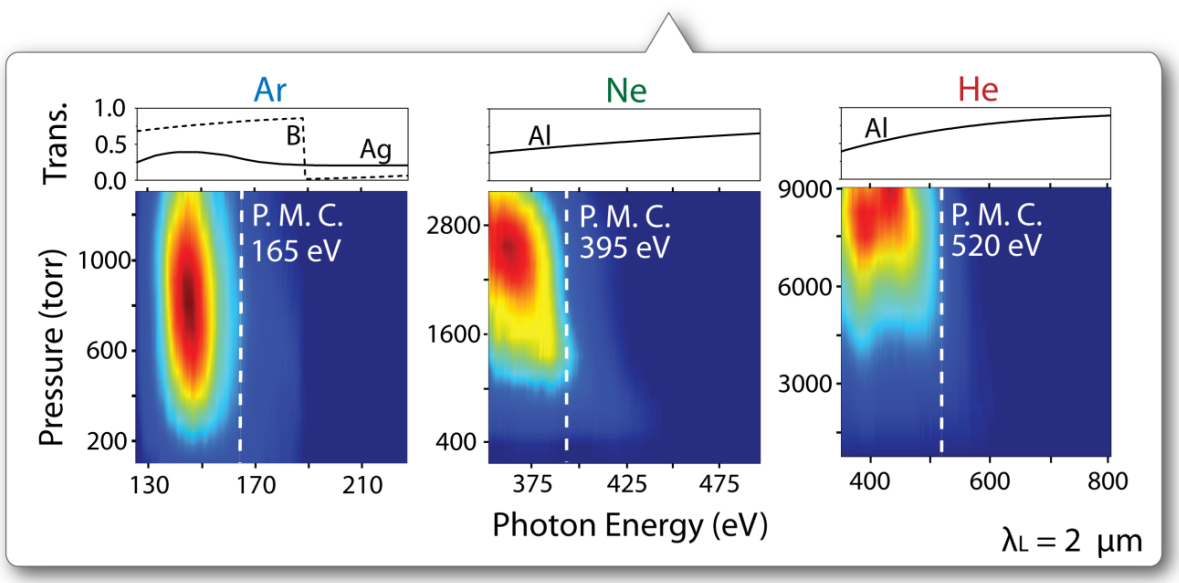


Bright spatially coherent laser-like output

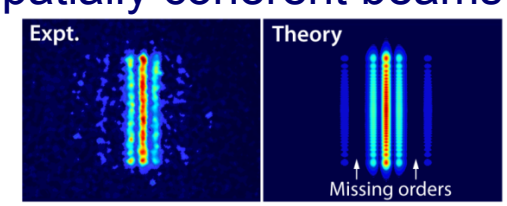


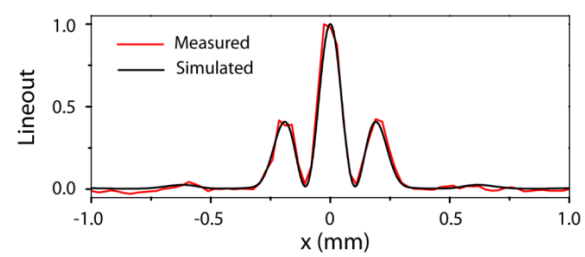
HHG flux high: 10^6 photons/shot in $\lambda/\Delta\lambda$ of 100



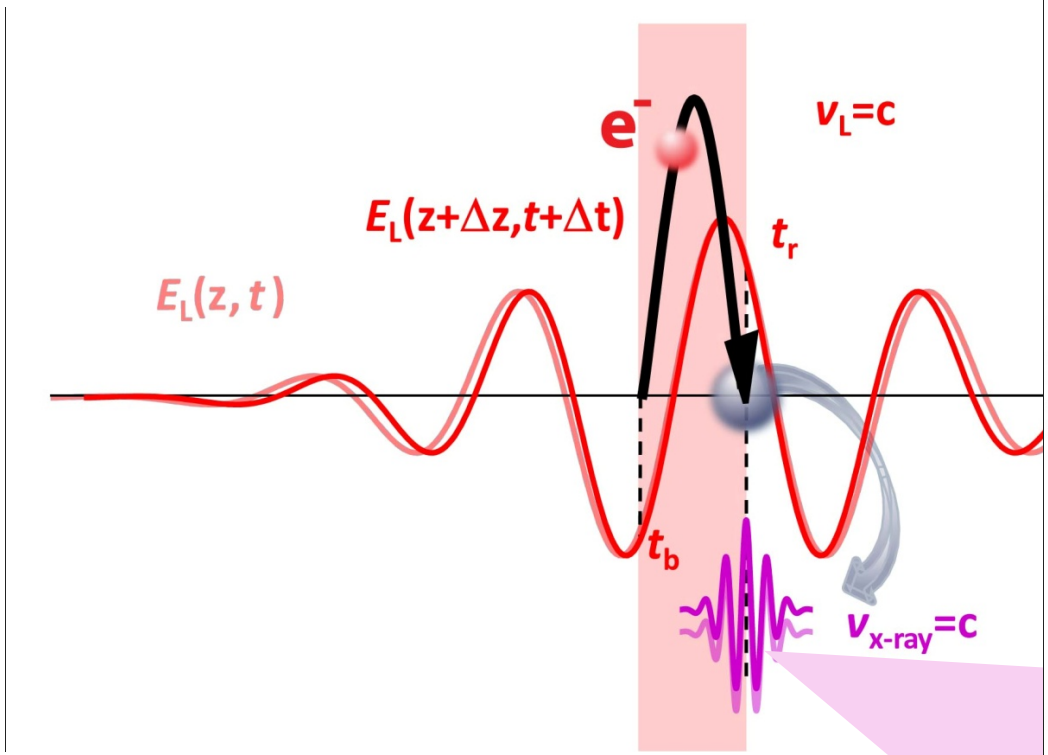


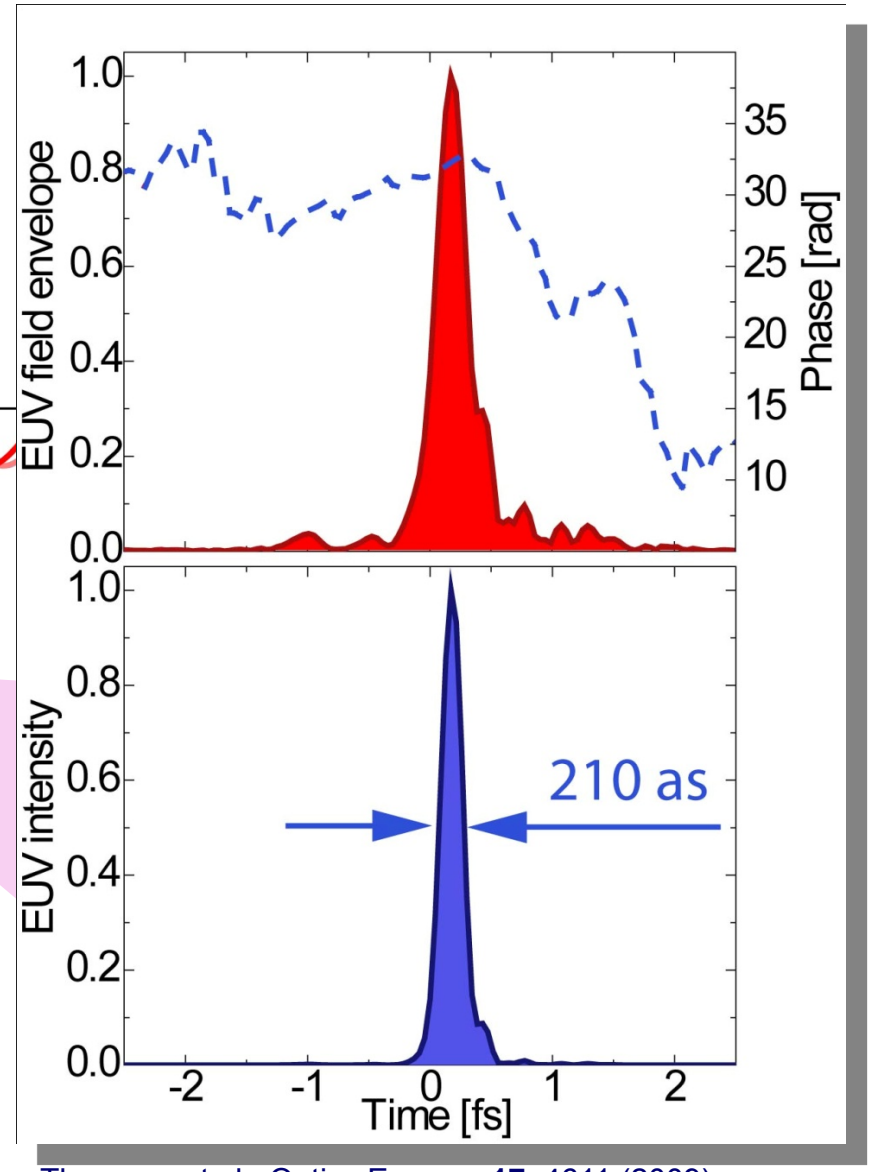
Spatially coherent beams





200 attosecond pulse generation at 50eV

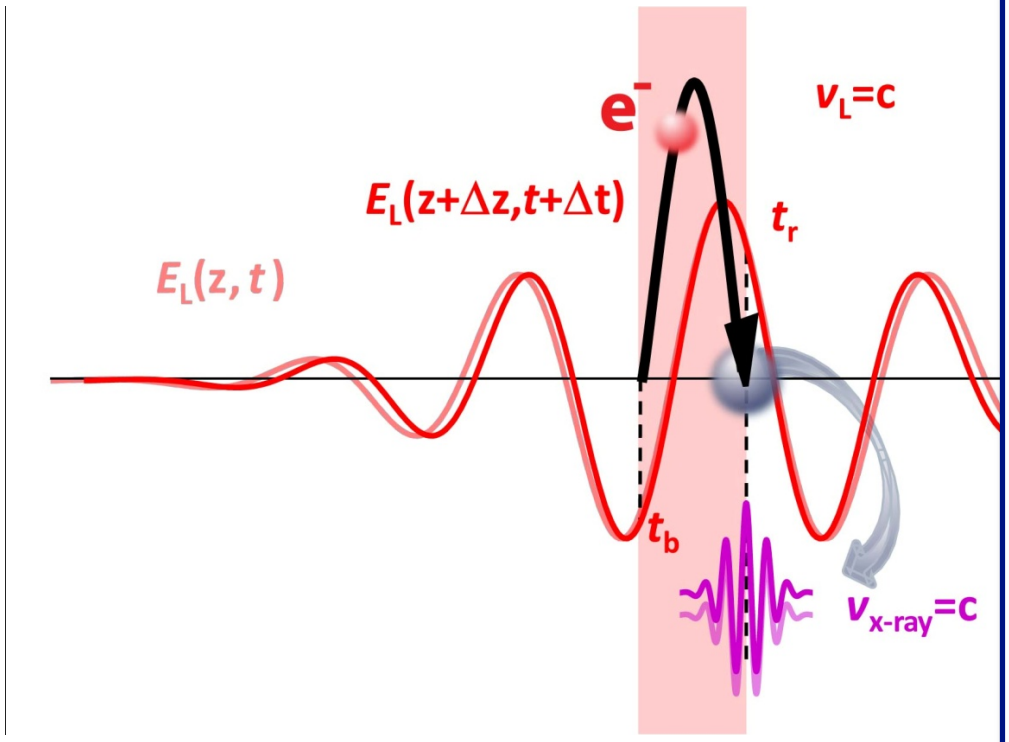


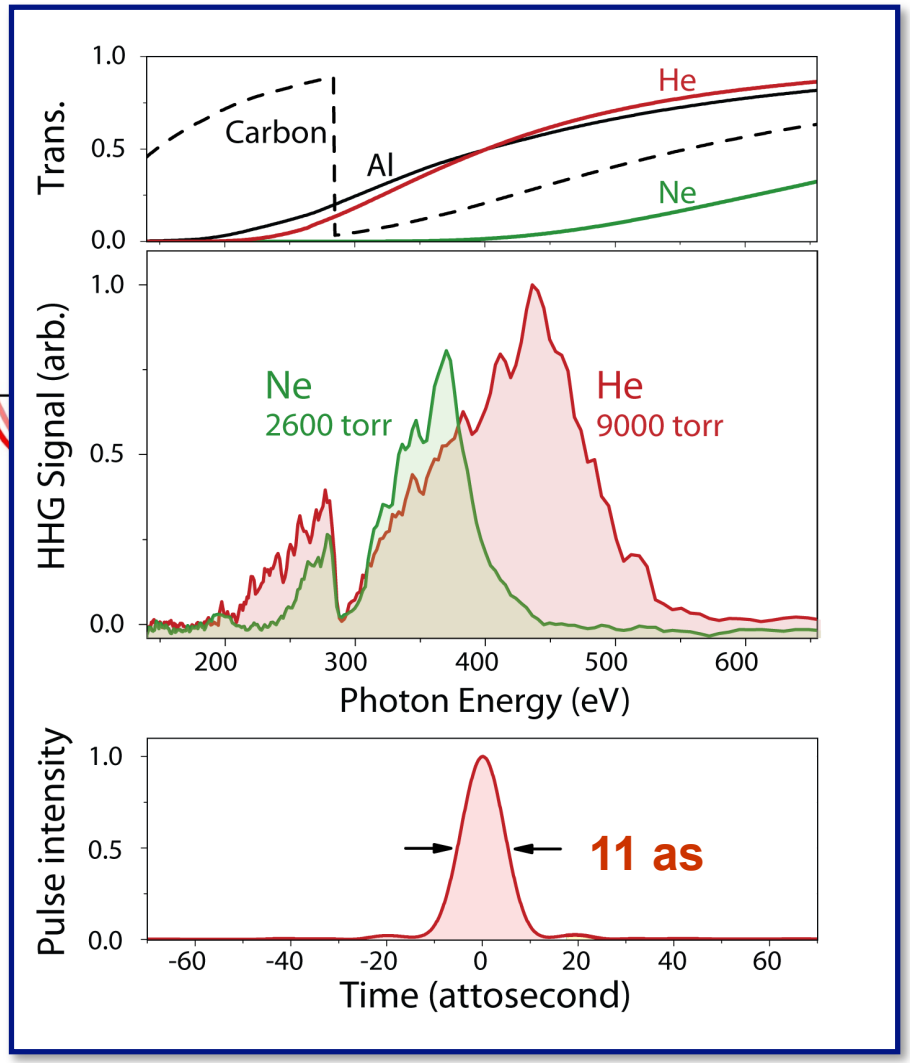


- Phase matching occurs over 1 laser cycle
- Using 15 fs driving laser at 0.8 μm, 200 attosecond pulses can easily be generated

Thomann et al., Optics Express **17**, 4611 (2009)

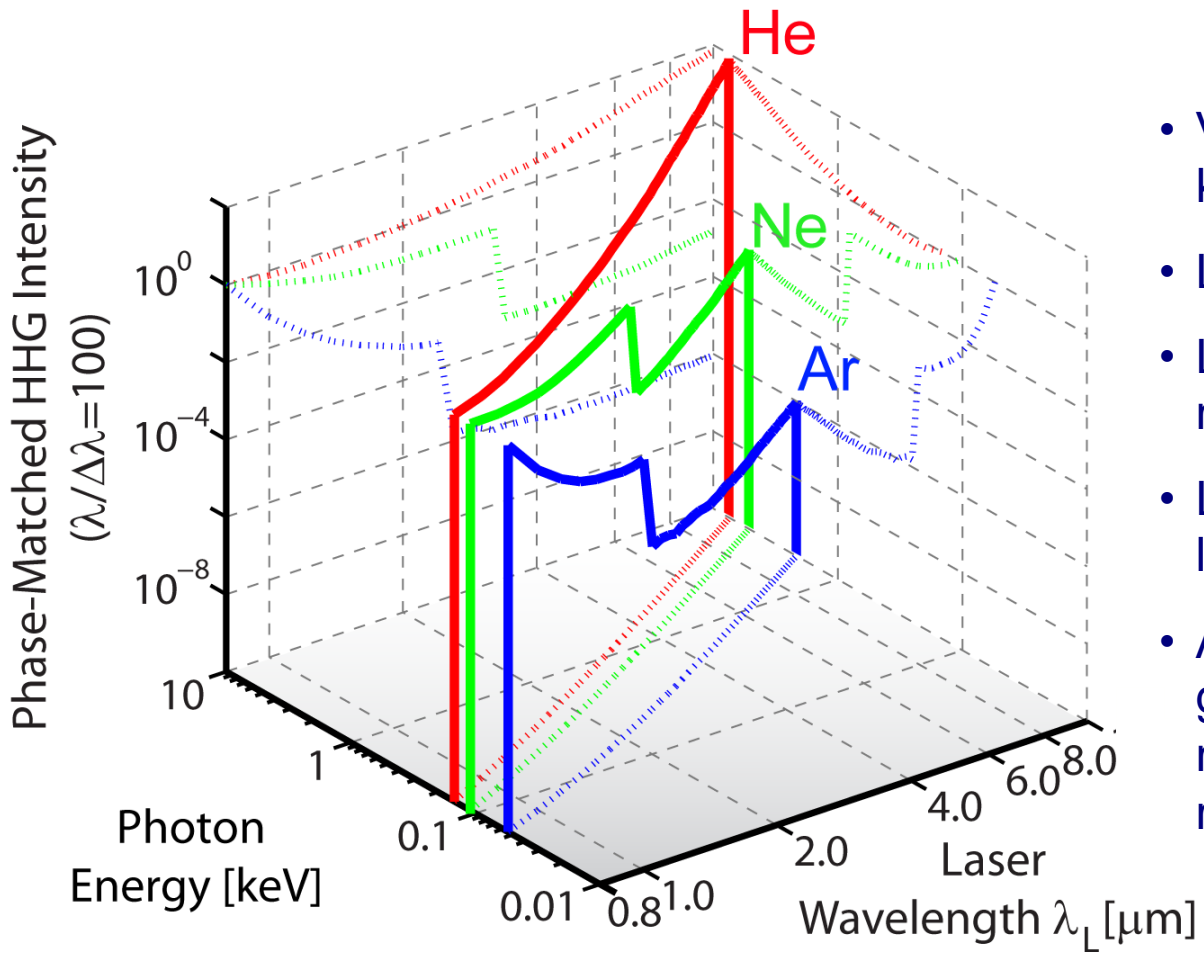
10 attosecond pulse generation at 400eV





- Phase matching occurs over 1 laser cycle
- Using 35 fs driving laser at 2 μm, 10 attosecond pulses can easily be generated
- Predict zeptosecond pulses at keV

Scaling of phase matched HHG flux to keV and beyond

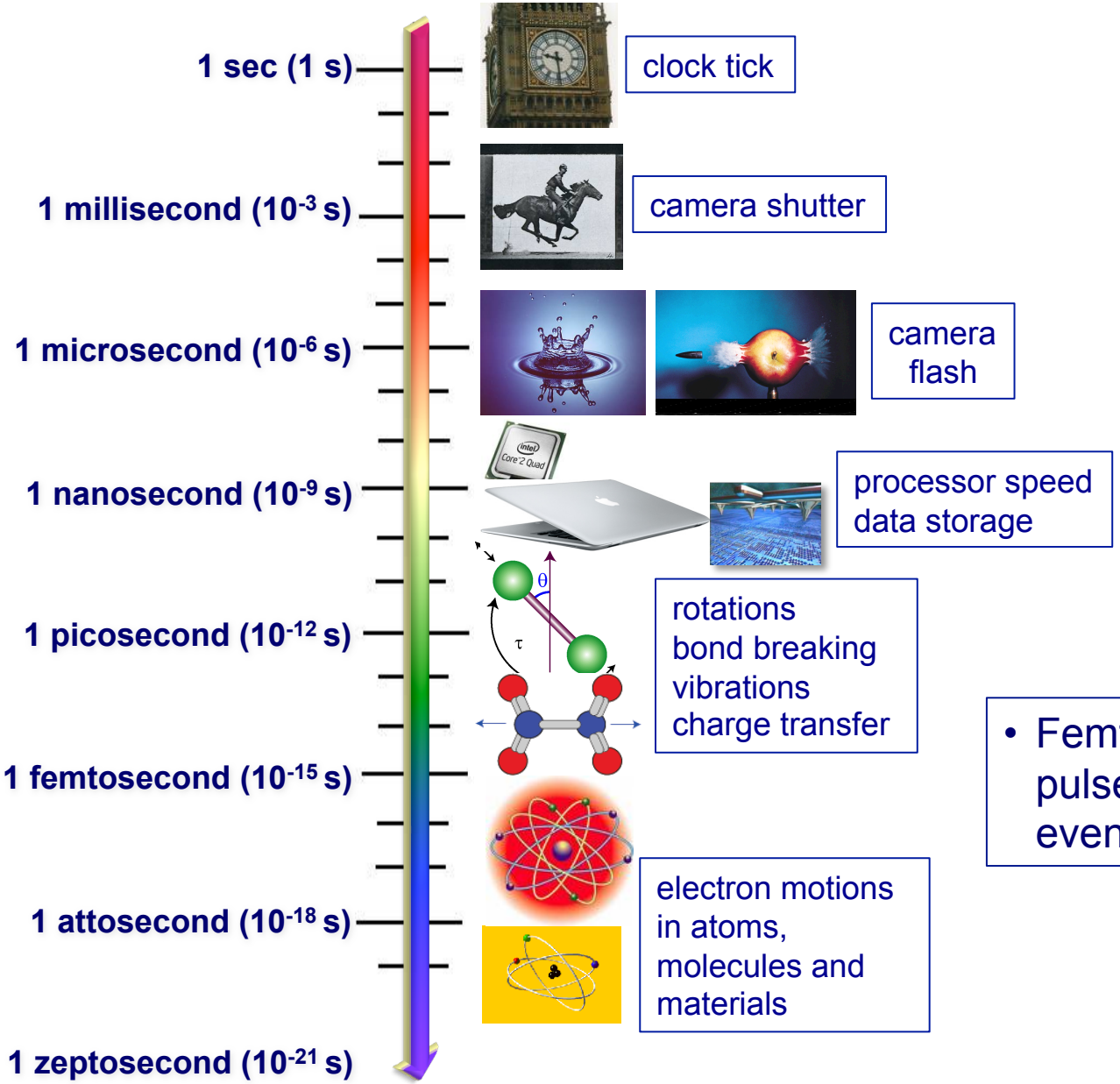


- Very favorable scaling to multi-keV region!!
- Low gas absorption of HHG
- Large pressure-length products mitigate the low χ_{eff}
- Low nonlinear distortion of laser pulse due to ionization
- At laser wavelengths > 3μm, group velocity mismatch and magnetic field effects may reduce the HHG flux

$$dI_q \propto \frac{\omega_q^{\,2}\rho^2\left|s_q\right|^2}{\alpha_q^{\,2}+\Delta k^2}\left(1+e^{-\frac{L}{L_{abs}}}-2e^{-\frac{L}{2L_{abs}}}\cos\Delta kL\right)$$

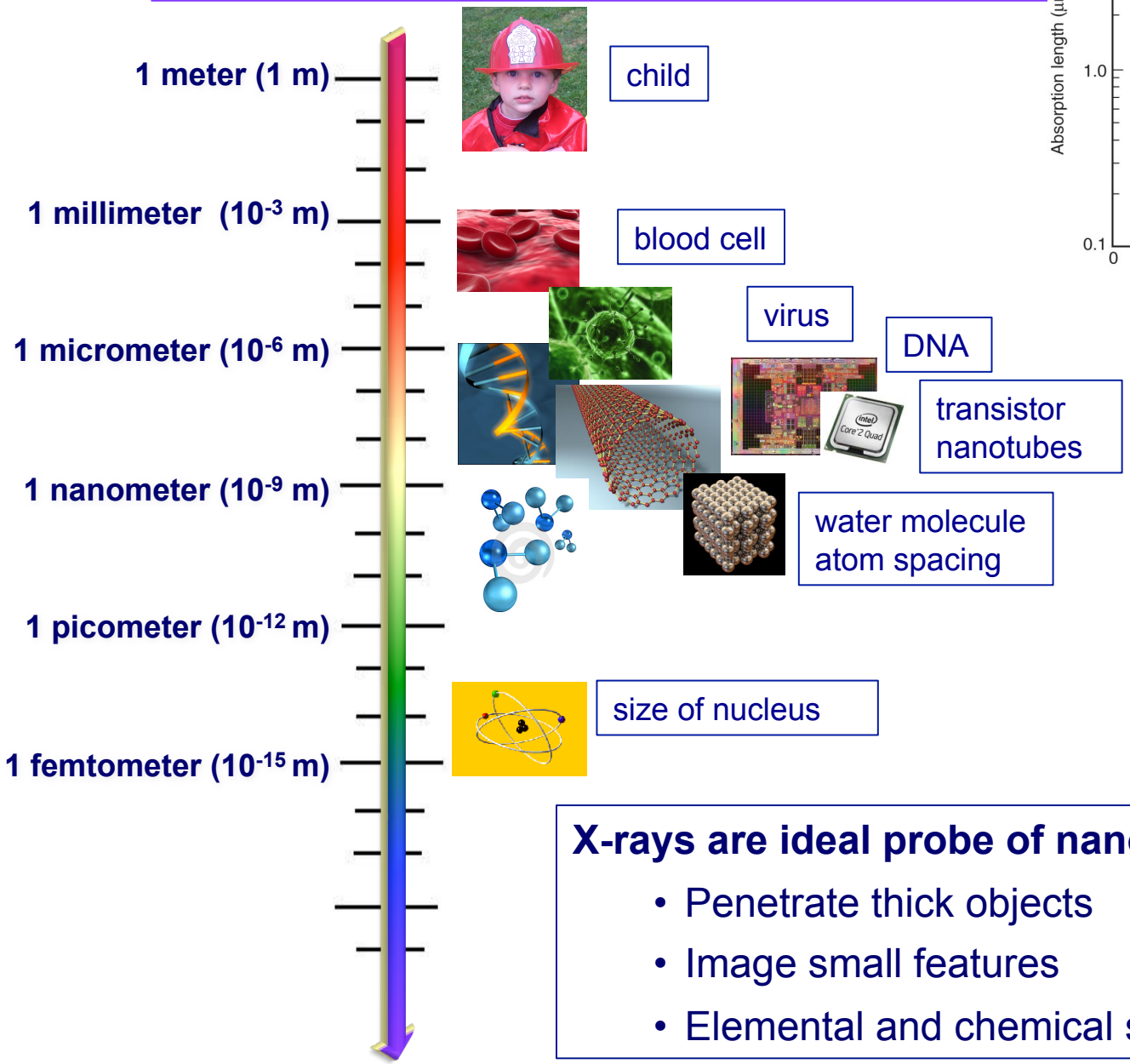
*PNAS **106**, 10516 (2009)*
Nature Photonics to be pub (2009)

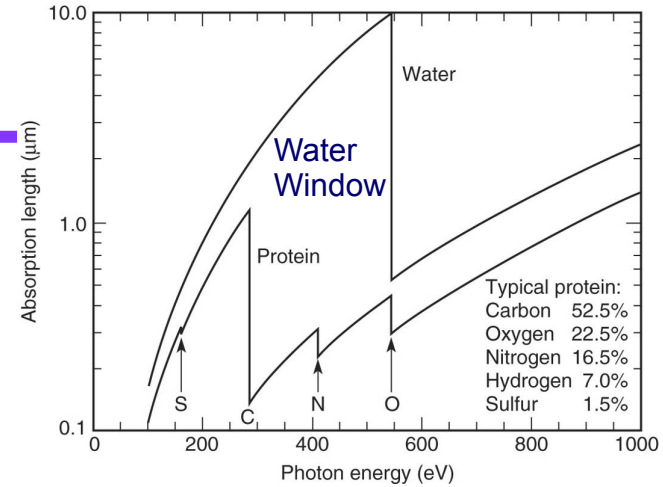
Characteristic time scales of the nanoworld

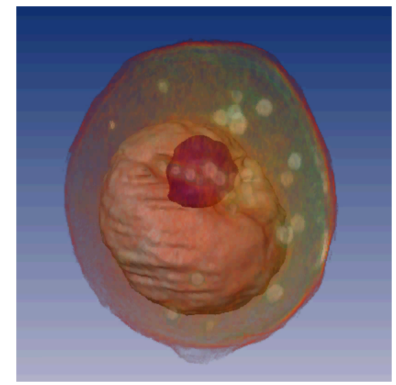


- Femtosecond to attosecond pulses can capture motion - even at the level of electrons

Characteristic length scales





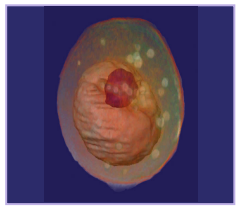


Carolyn Larabell, LBL

X-rays are ideal probe of nanoworld:

- Penetrate thick objects
- Image small features
- Elemental and chemical specificity

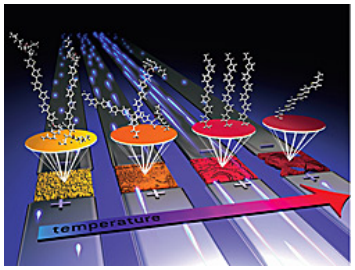
X-ray light is a unique tool for science



Bio-microscopy

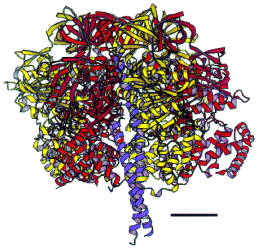
Spectro-microscopy of surfaces

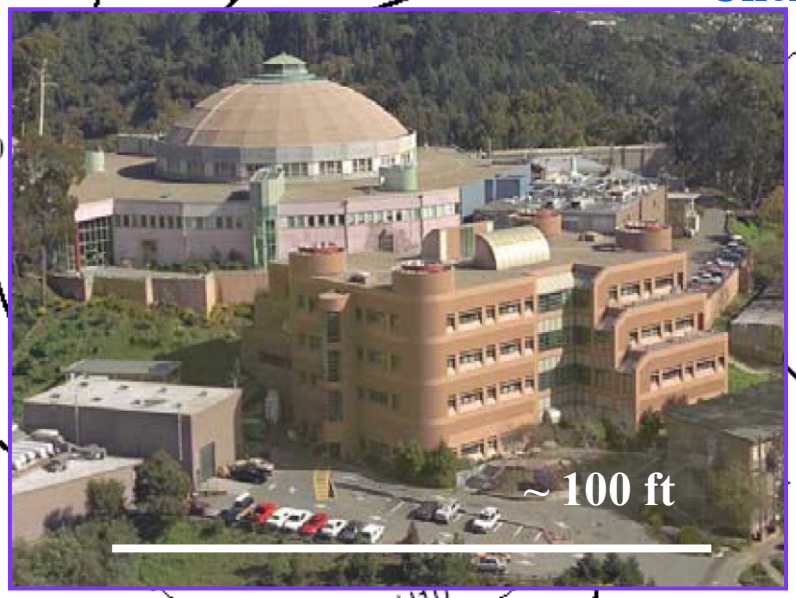
Surface science



Chemical dynamics

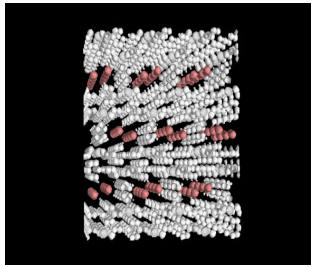
Protein crystallography



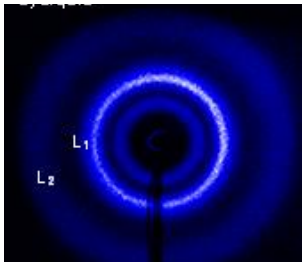


Photoemission spectroscopy

Materials science and biology



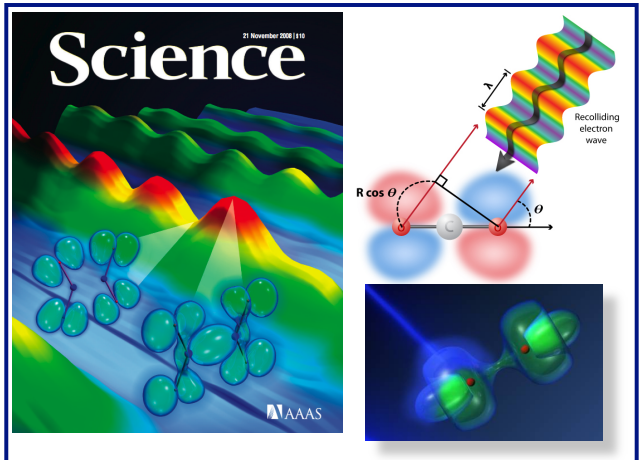
Magnetic materials
Polarization studies



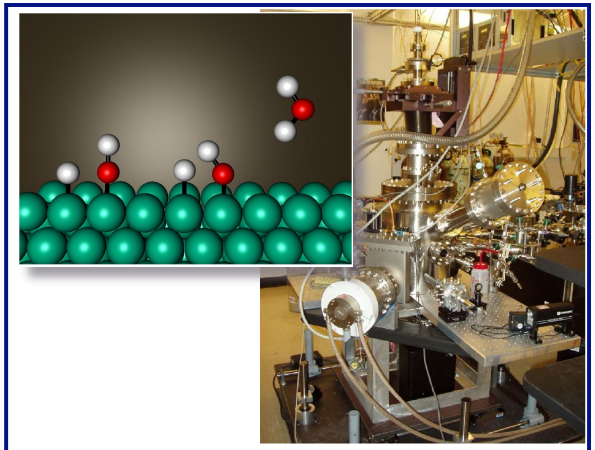
Interferometry and coherent optics

Atomic and Molecular physics

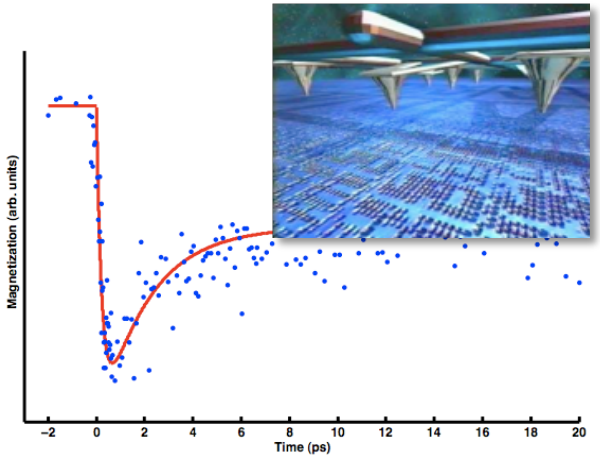
Applications of coherent ultrafast x-rays span broad range of science



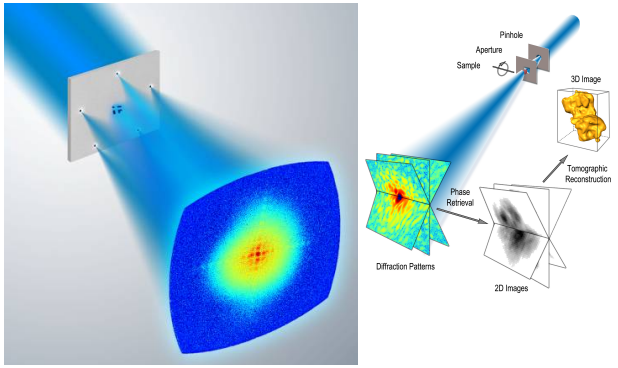
Molecular imaging: image changing electronic orbital and molecular structure



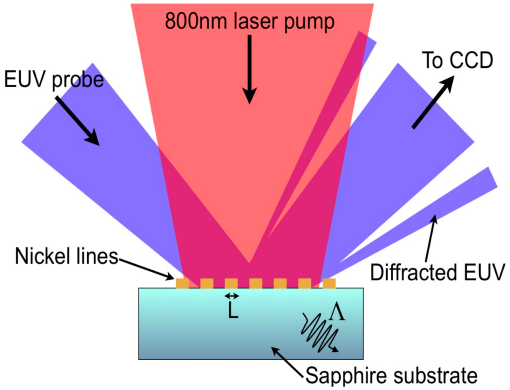
Surface science: probe electronic dynamics on catalysts, photovoltaics



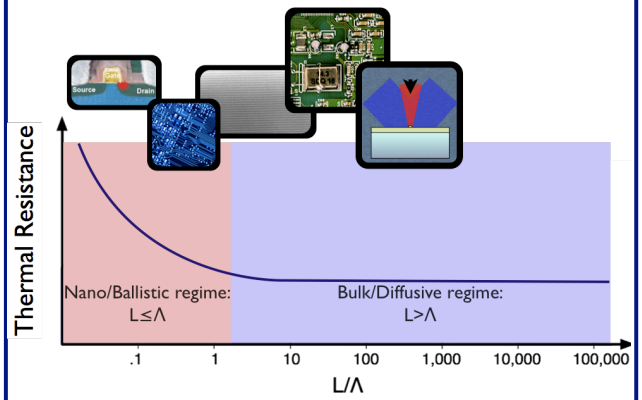
Magnetics: Probe nanodomains, magnetic dynamics



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

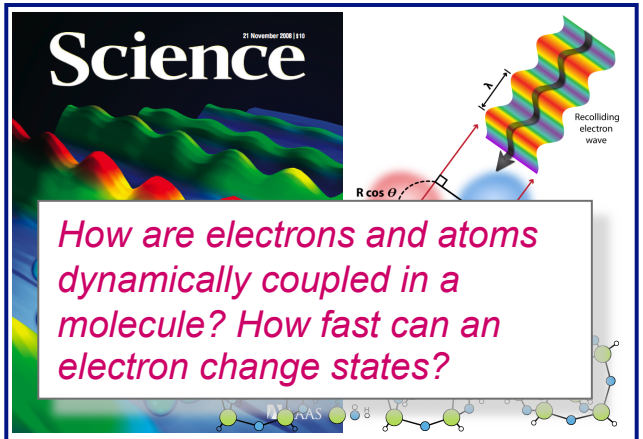


High frequency acoustic metrology: Characterize thin films, interfaces, adhesion



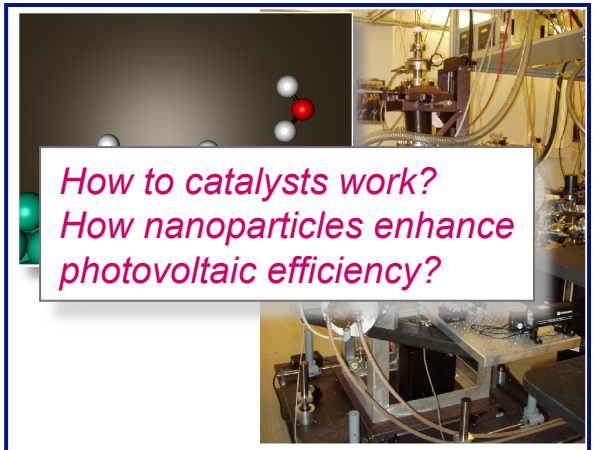
Nanothermal transport: probe heat flow in nanostructures

HHG addresses important technological or scientific questions



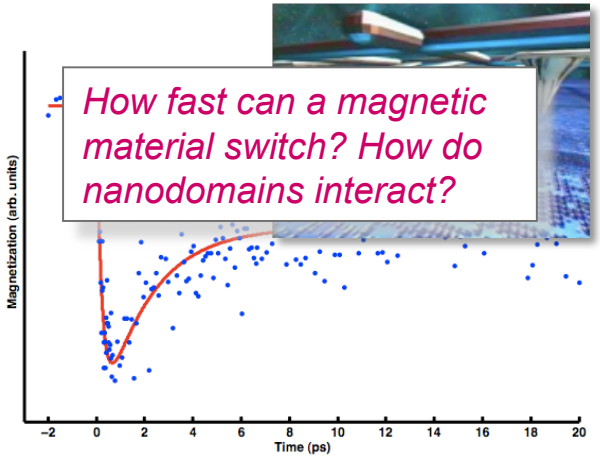
How are electrons and atoms dynamically coupled in a molecule? How fast can an electron change states?

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

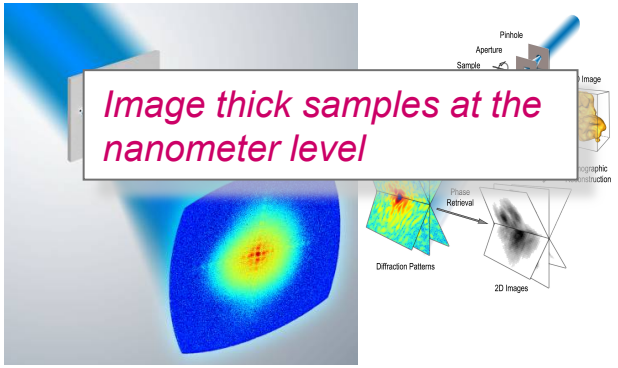
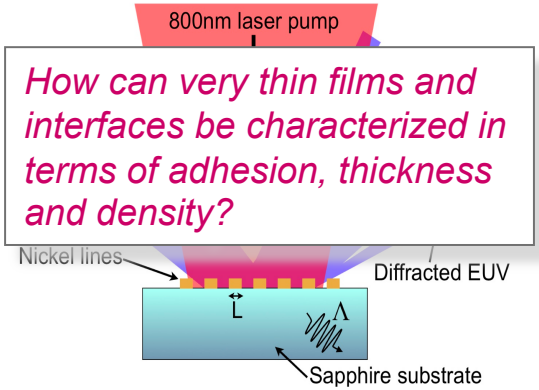


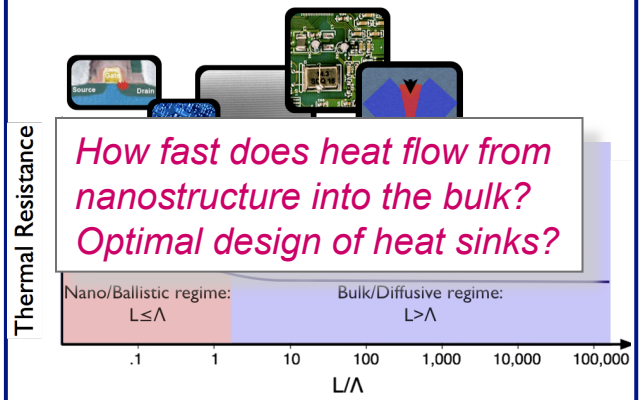
Image thick samples at the nanometer level

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?

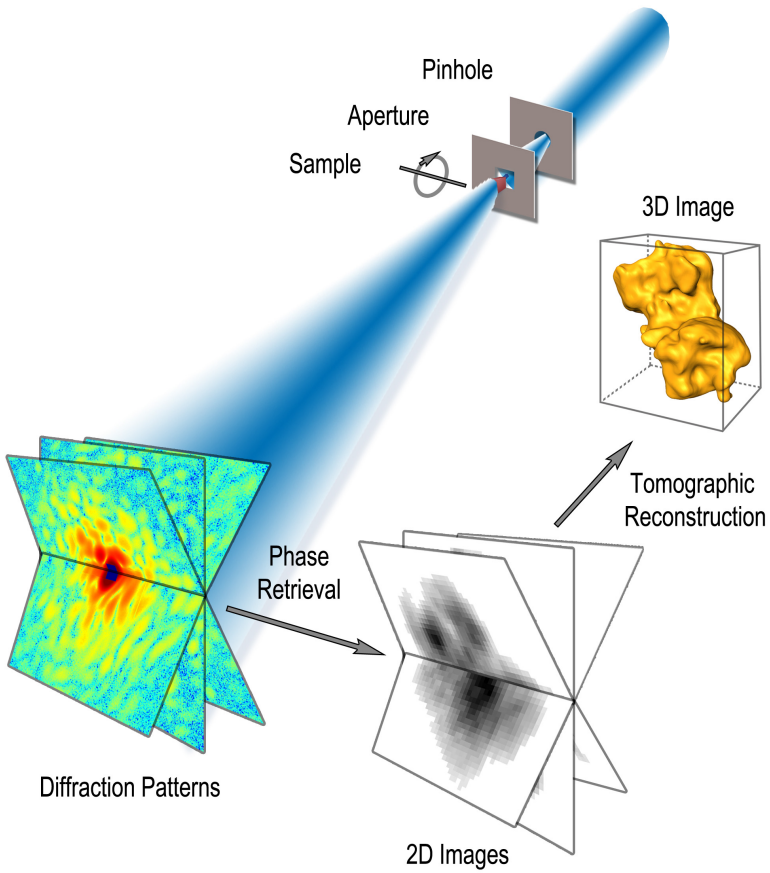
High frequency acoustic metrology: Characterize thin films, interfaces, adhesion



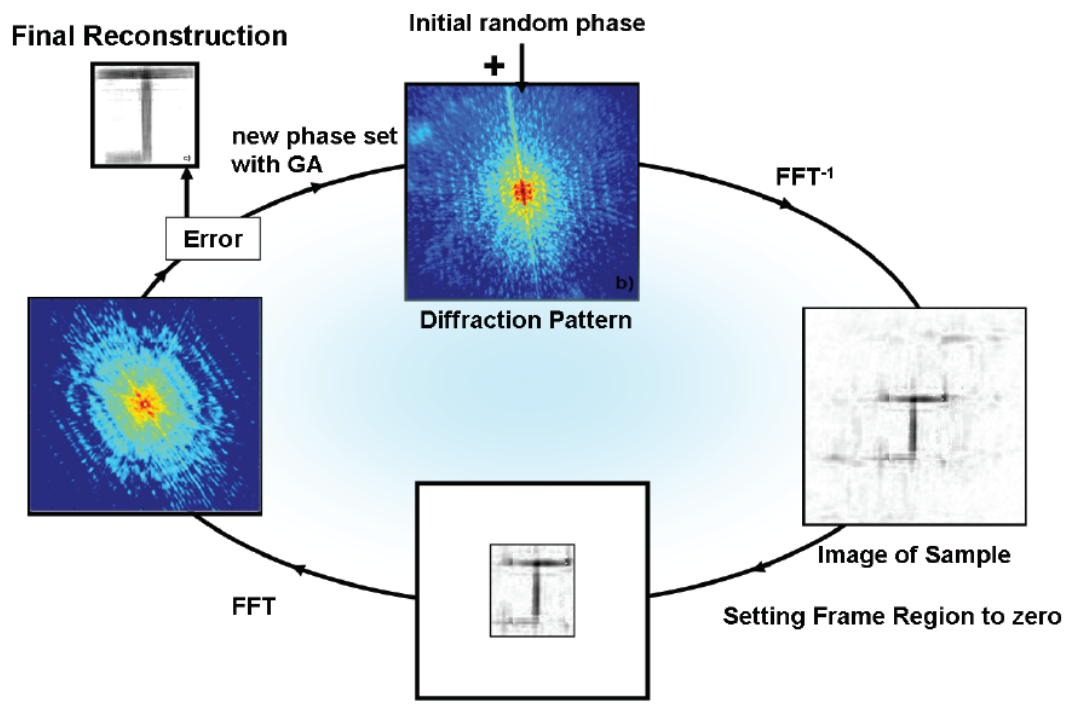
How fast does heat flow from nanostructure into the bulk? Optimal design of heat sinks?

Nanothermal transport: probe heat flow in nanostructures

Lensless microscopy using coherent x-ray beams

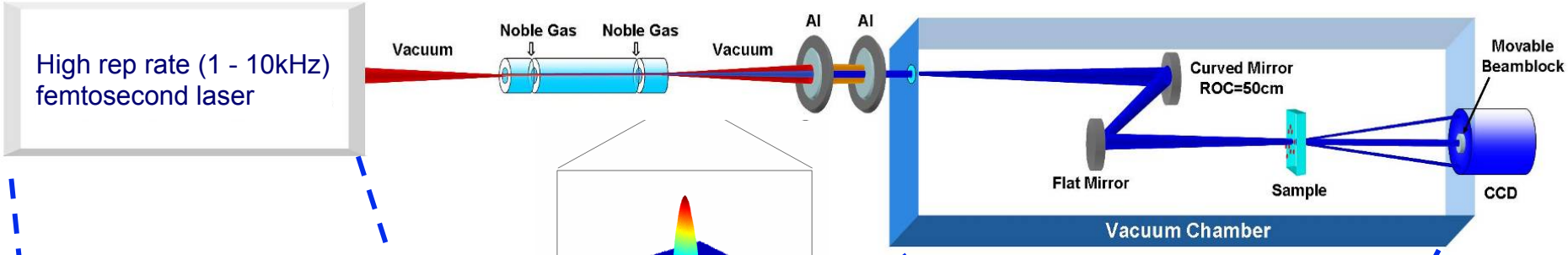


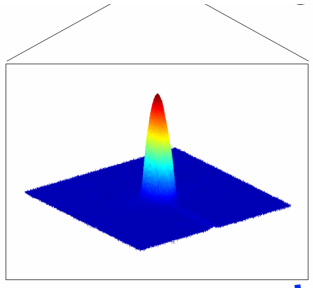
- No aberrations - diffraction-limit in theory
- Image thick samples
- Inherent contrast of x-rays
- Robust geometry, insensitive to vibrations
- Requires a coherent beam of light and an isolated sample



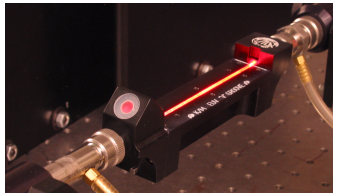
Sayre, Acta Cryst 5, 843 (1952)
Miao et al., Nature 400, 342 (1999)

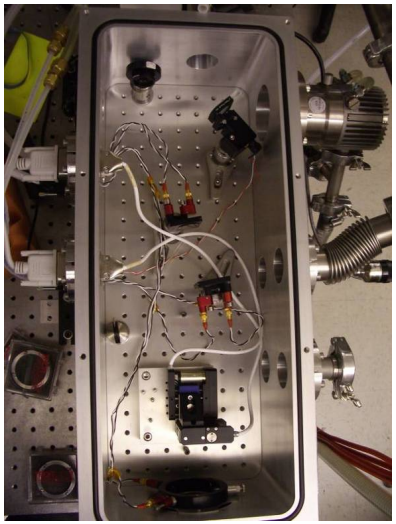
Lensless imaging using tabletop soft x-ray sources





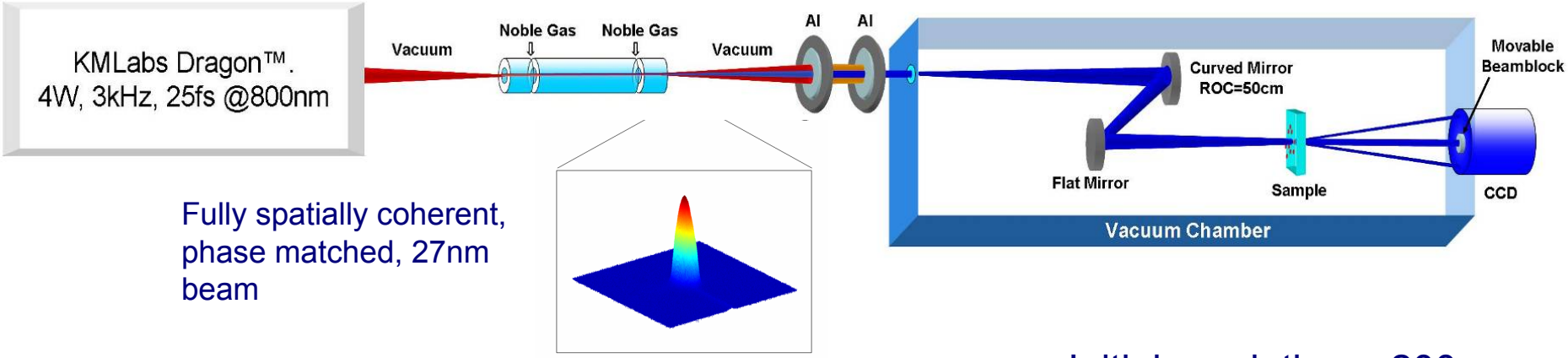
X-ray beam





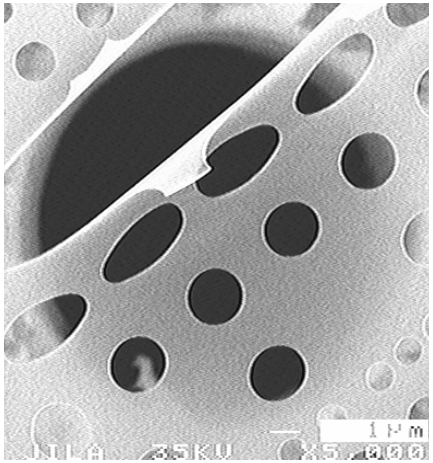
High average power fs laser system + High harmonic converter + Coherent diffractive microscope

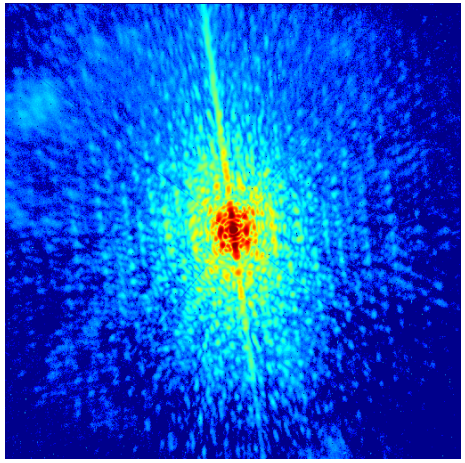
Lensless coherent imaging - first results 200nm

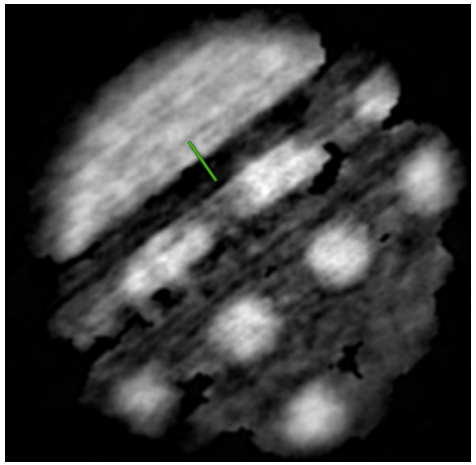


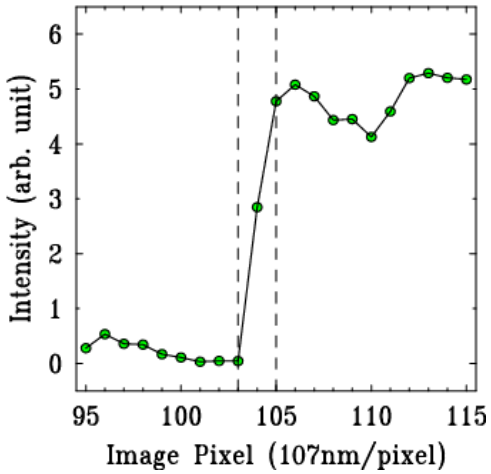
Fully spatially coherent, phase matched, 27nm beam

Initial resolution ≈ 200nm







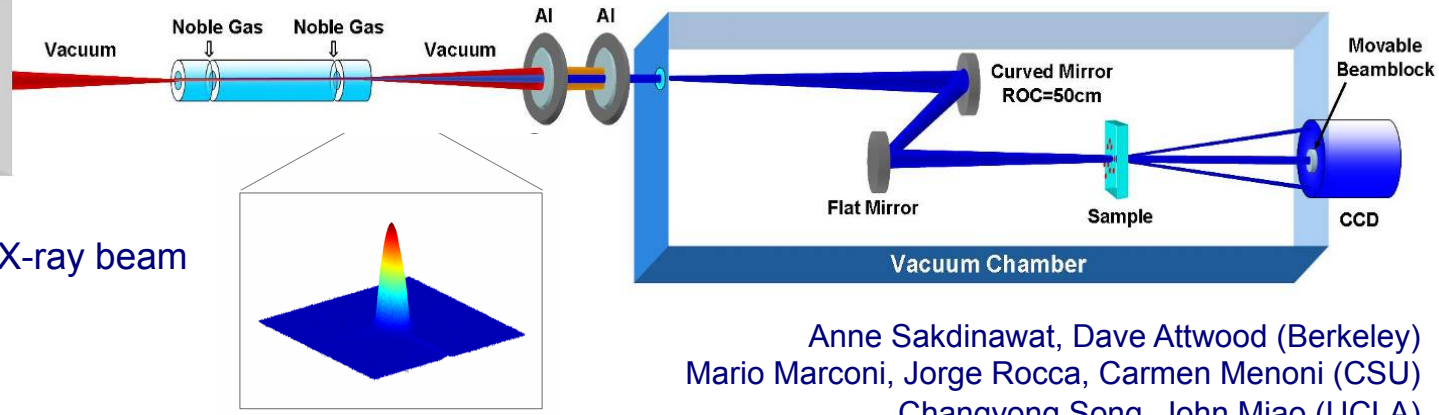


sample (SEM image) diffraction pattern sample (reconstruction)

Phys. Rev. Lett. 99, 098103 (2007); Nature, News & Views 449, 553 (2007)

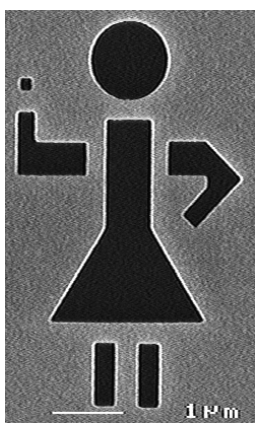
Lensless coherent imaging – 90 nm resolution

High harmonic source (30nm)

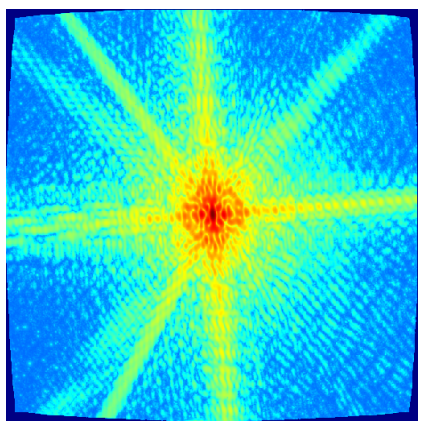


X-ray beam

Anne Sakdinawat, Dave Attwood (Berkeley)
Mario Marconi, Jorge Rocca, Carmen Menoni (CSU)
Changyong Song, John Miao (UCLA)
Richard Sandberg, Daisy Raymondson, MM, HK (JILA)

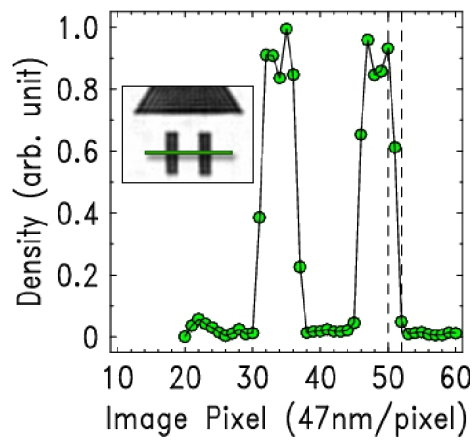


1 μm
sample (SEM image)



HHG diffraction pattern

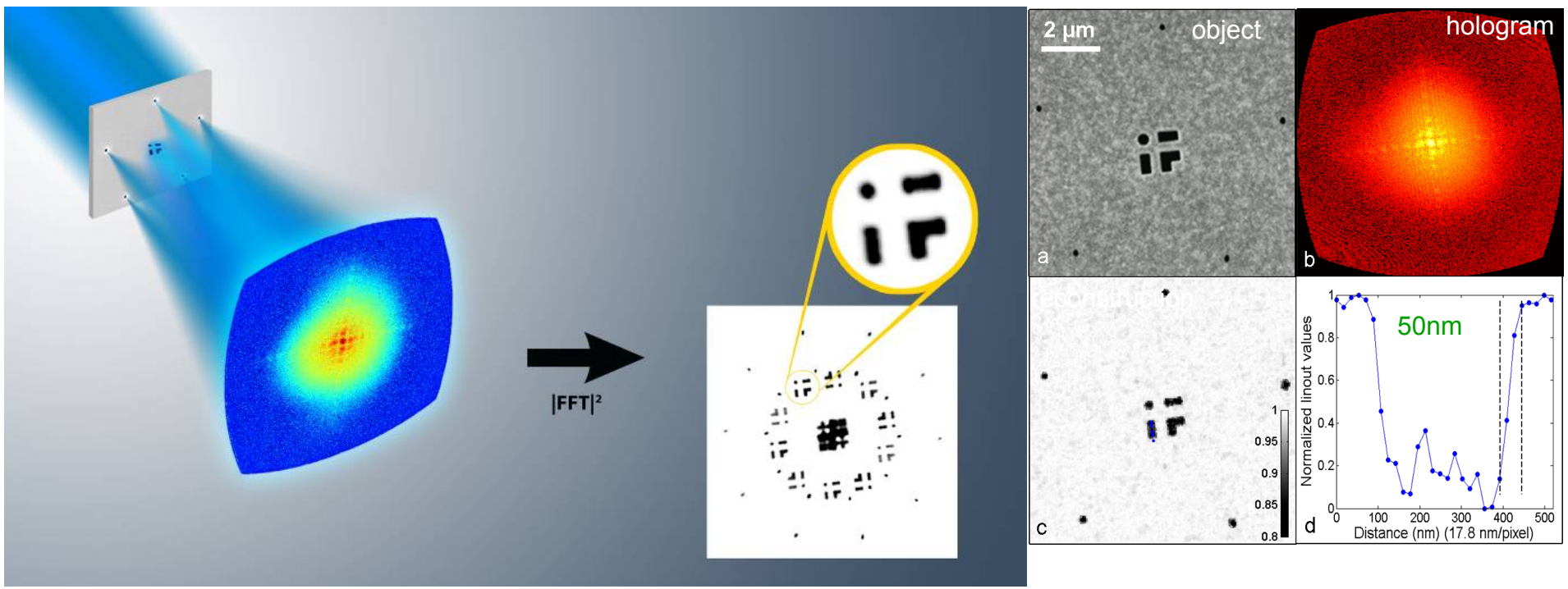
reconstruction



PNAS 105, 24 (2008); Nature Photonics 2, 64 (2008)

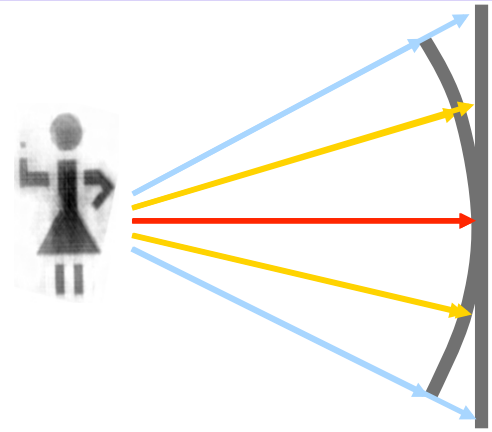
Lensless coherent imaging – current results 50nm

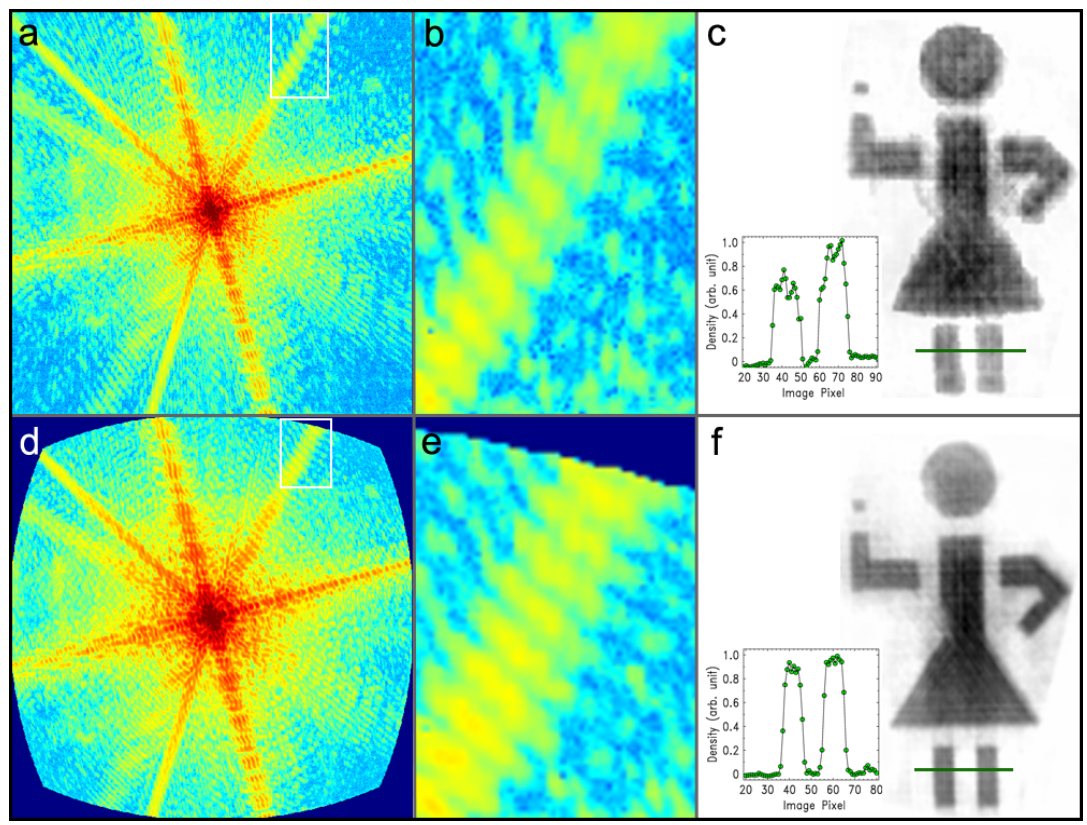
- Combine lensless imaging with holography for faster, high resolution imaging
- Resolution of 50 nm represents 1.6 λ
- Future: sub-10 nm imaging of thick samples with element specificity
- Applications in bioimaging, magnetics, nano, thermal, lithography…….



Curvature correction at high NA imaging

- High NA means high angle diffraction - which violates paraxial approximation
- Resample diffraction pattern from flat CCD to spherical field for curvature correction
- Demonstrates lensless imaging can be used to obtain near λ resolution !

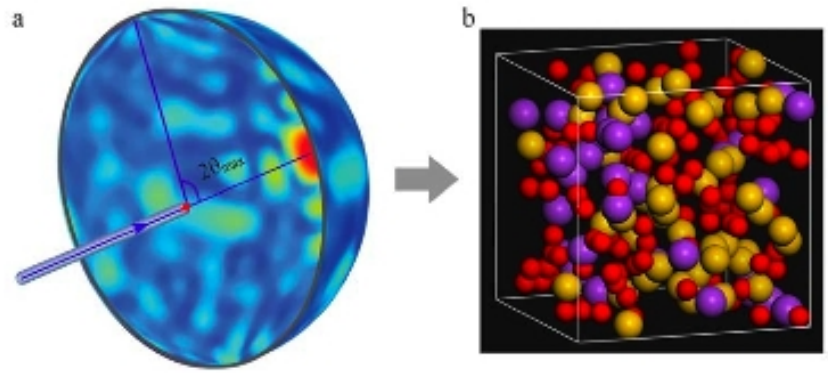




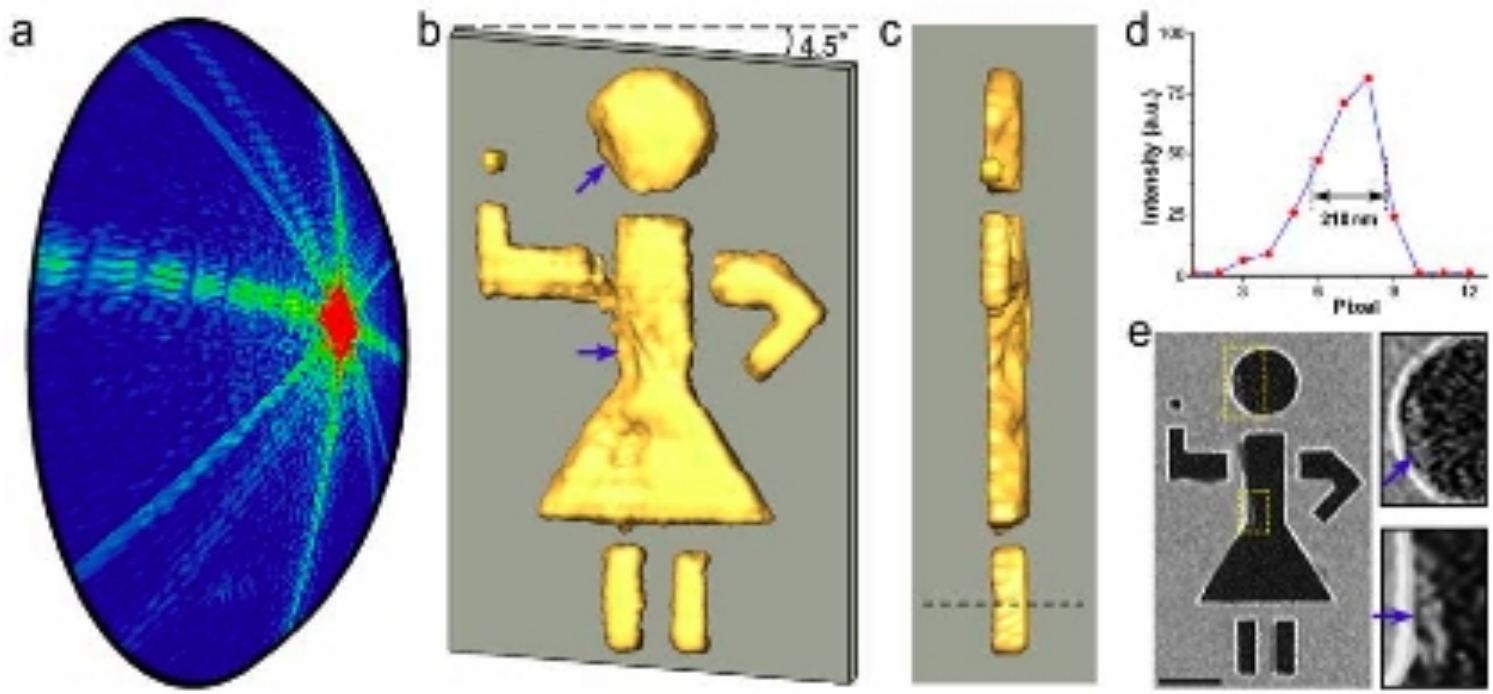
PNAS 105, 24 (2008)

Ankylography - 3D information from 2D pattern

When diffraction is sampled at very high angles, 3D information can be extracted from a 2D pattern!

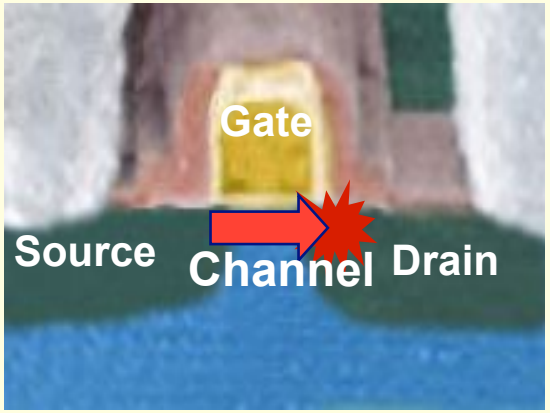


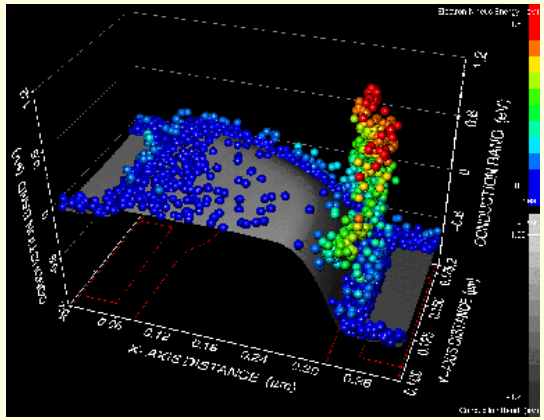
John Miao et al.
*Nature **463**, 214 (2010)*



Energy transport at the nanoscale

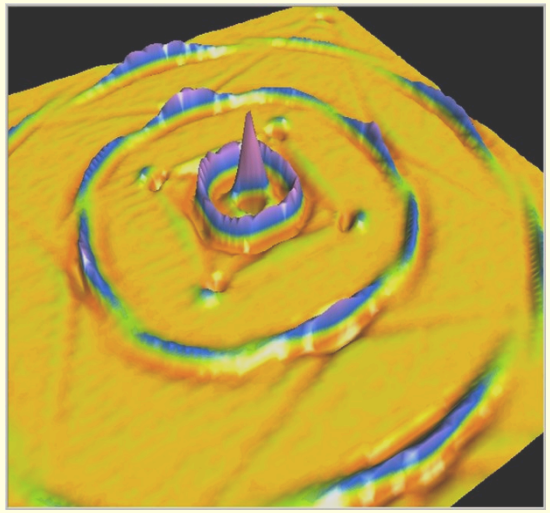
Thermal

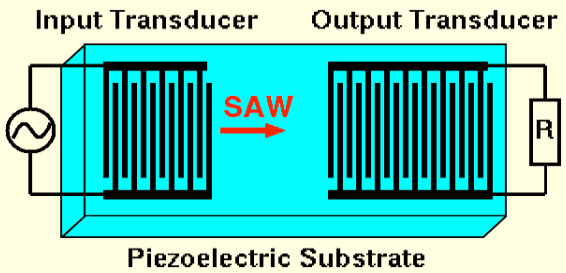




MOSFET (IBM)

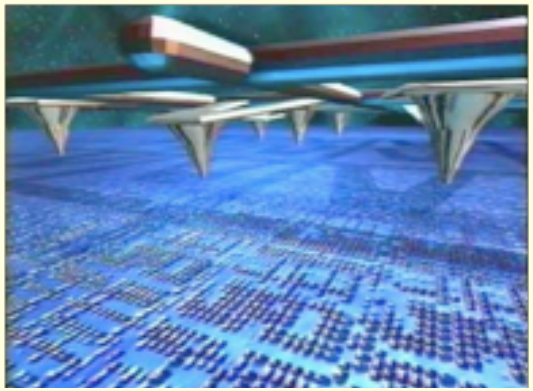
Acoustic



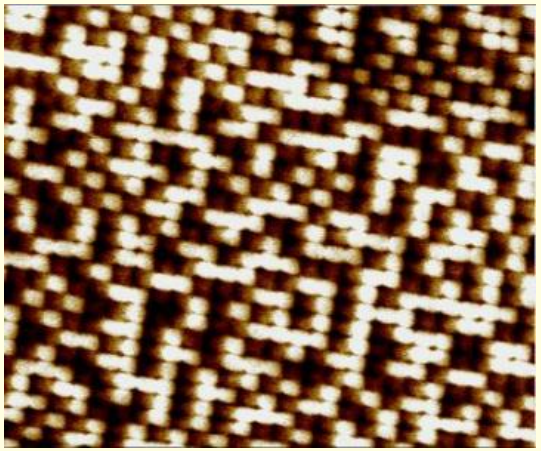


Surface Acoustic Waves (O. Wright)

Magnetic



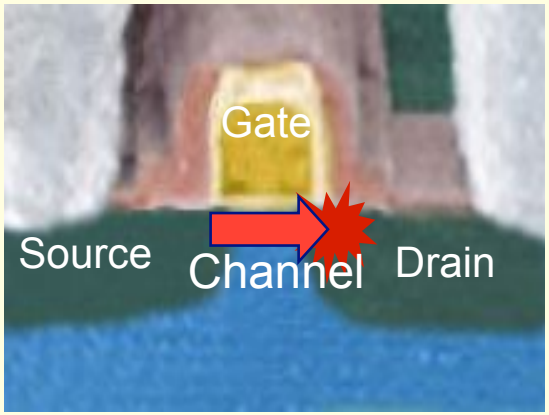
Data storage (IBM)



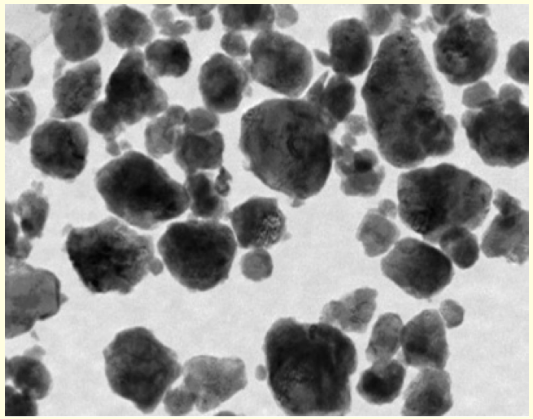
Magnetic nanodots NIST

Motivation

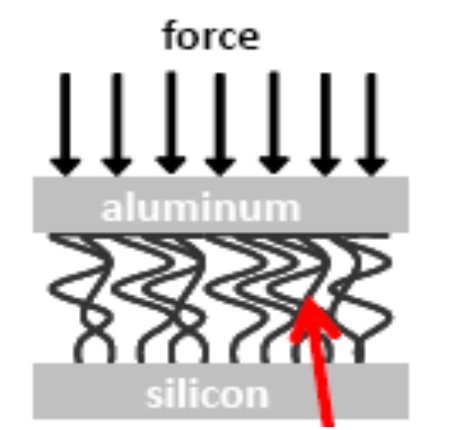
IC design



TE material



Thermal interface



Understanding heat flow away from a nanoscale hotspot is important in thermal management and novel material design

Requirements for tool:

- Non-contact
- Non-destructive
- ~1ps temporal resolution
- ~50nm spatial resolution

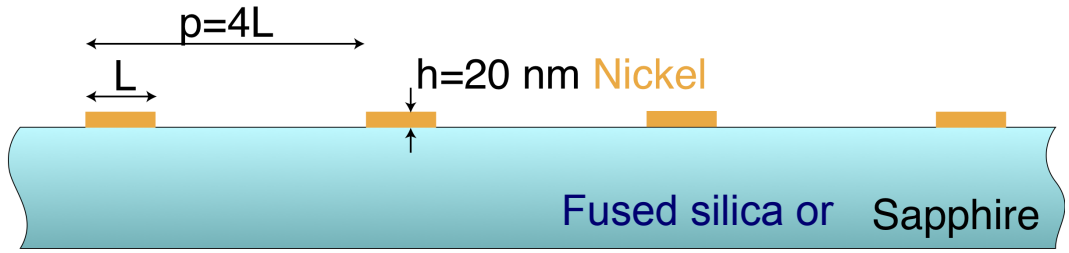
Coherent ultrafast x-ray beams

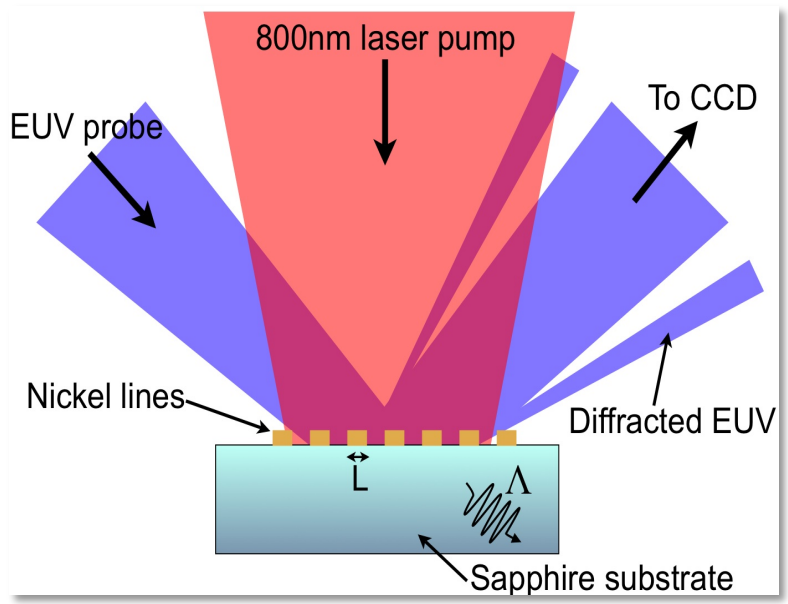
Nanoscale heat flow from hot nanowire into heat sink

- Heat is carried by phonons
- In the macroscopic world, Fourier Law applies

$$q = -k\nabla T$$

- What happens when a nanostructure is smaller than the phonon mean free path?
- Existing theories of nanoscale heat dissipation disagree



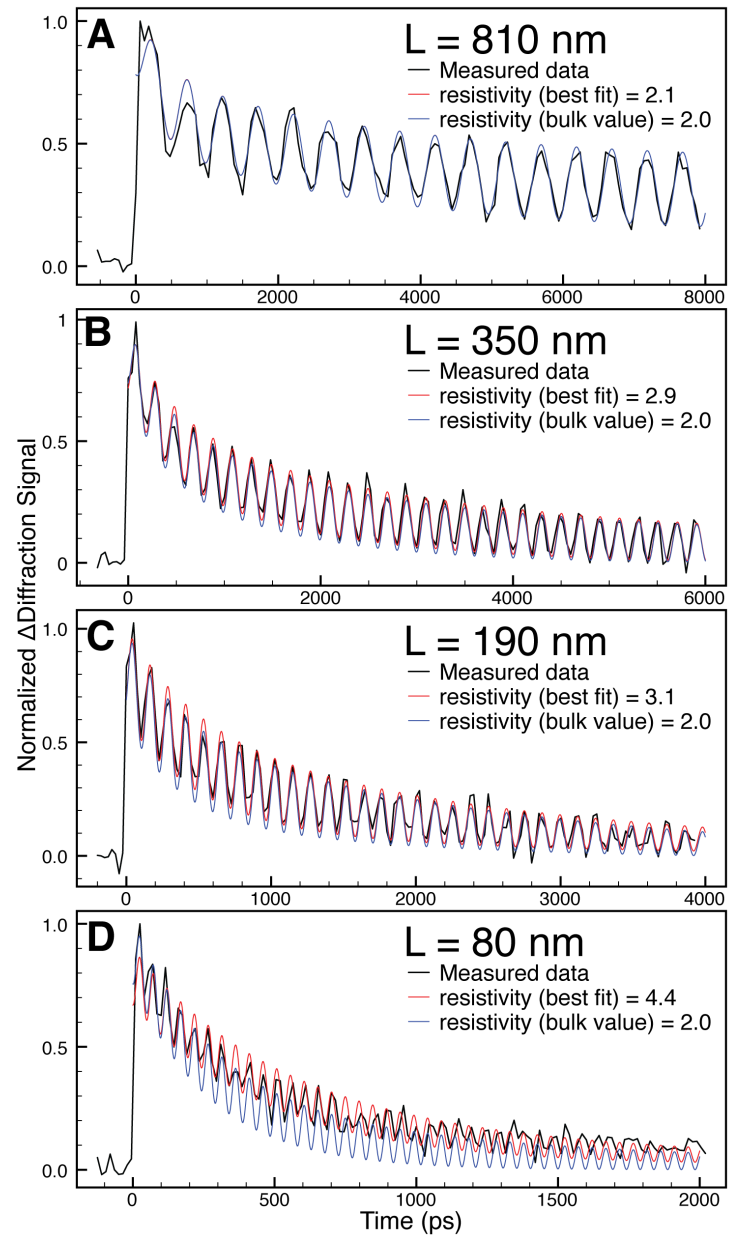


Observe deviation from Fourier Law for small linewidths

- Heat is carried by phonons
- In the macroscopic world, Fourier Law applies

$$q = -k\nabla T$$

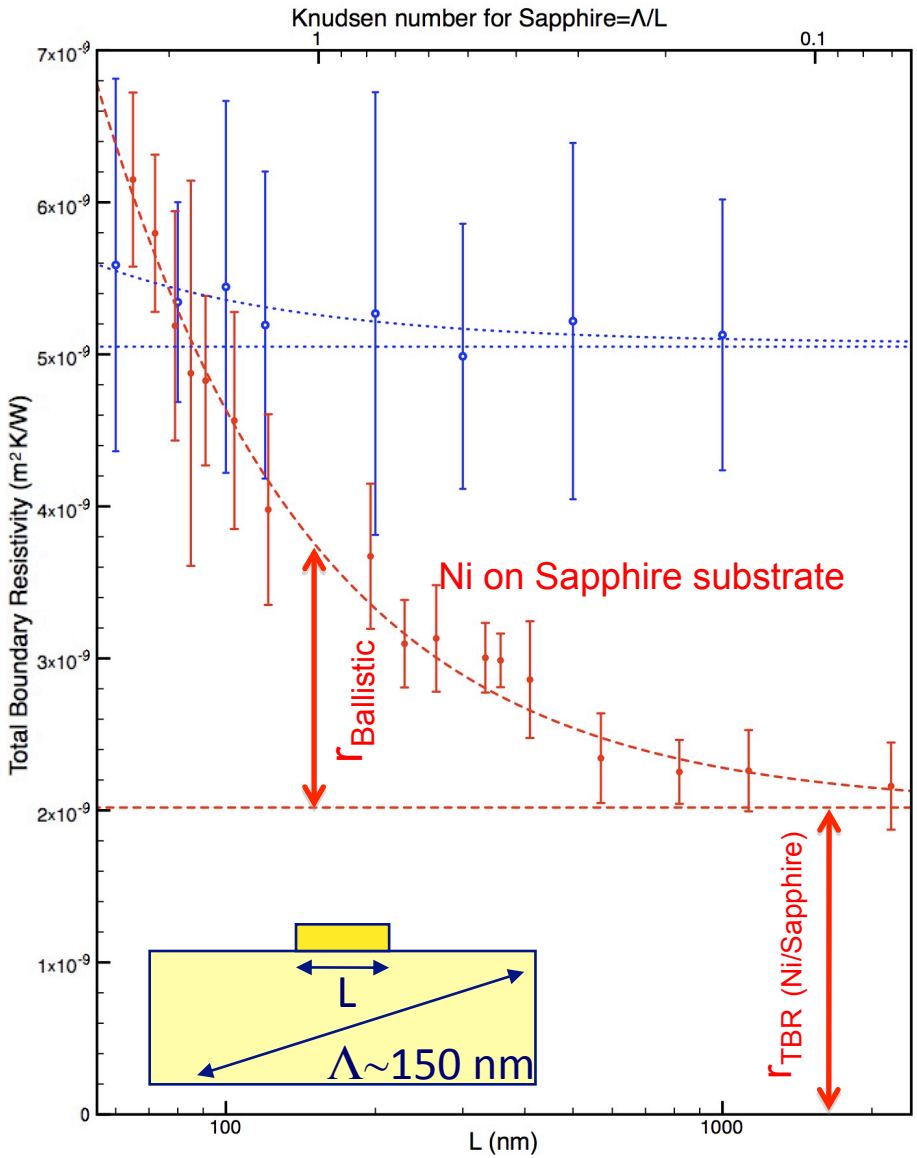
- What happens when a nanostructure is smaller than the phonon mean free path?
- Existing theories of nanoscale heat dissipation disagree



Nanoscale heat flow from hot nanowire into heat sink

- **Fused silica substrate:**
  - Λ ~ 2 nm
  - Measured interfacial resistivity is L-independent
  - Diffusive transport to even the smallest linewidths

- **Sapphire substrate:**
  - Λ ~ 100 - 150 nm
  - Observed quasi-ballistic boundary resistance ~Λ/L
  - r_{BD} 3x higher than bulk at 65nm

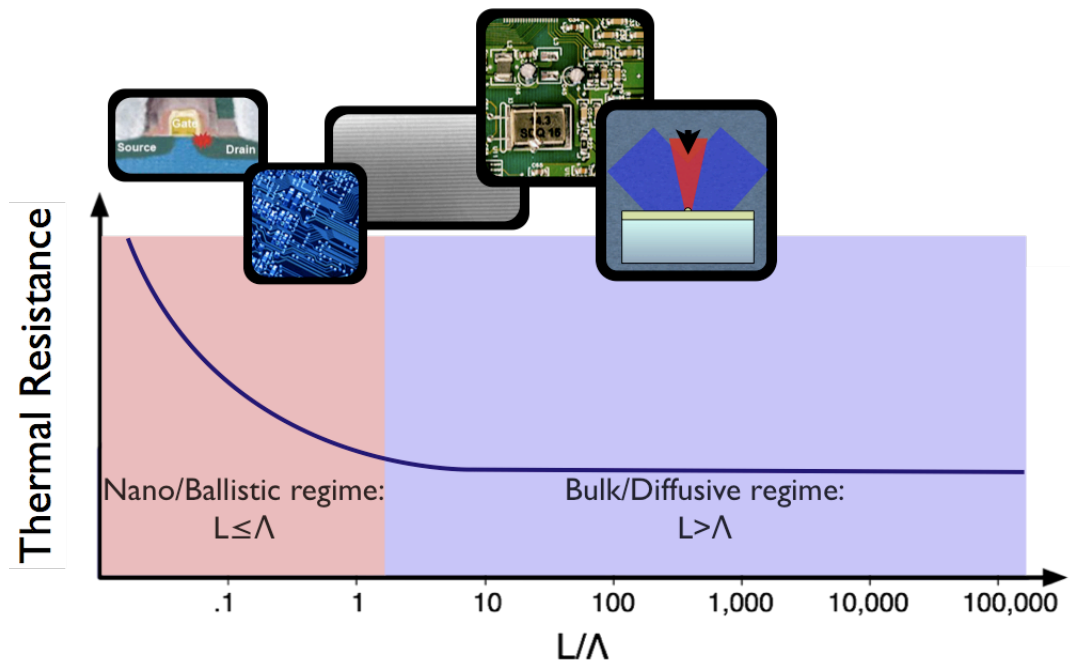


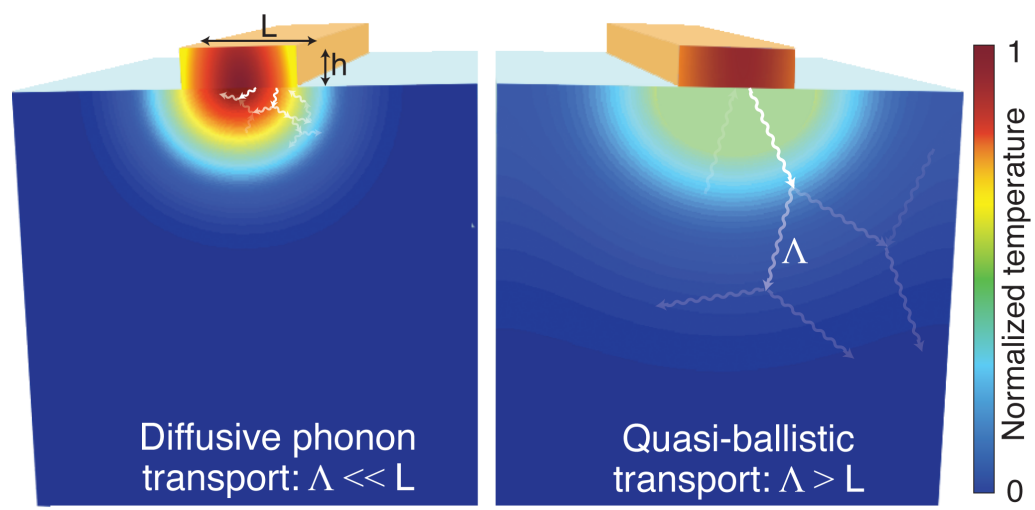
Nanoscale heat flow from hot nanowire into heat sink

- Heat is carried by phonons
- In the macroscopic world, Fourier Law applies

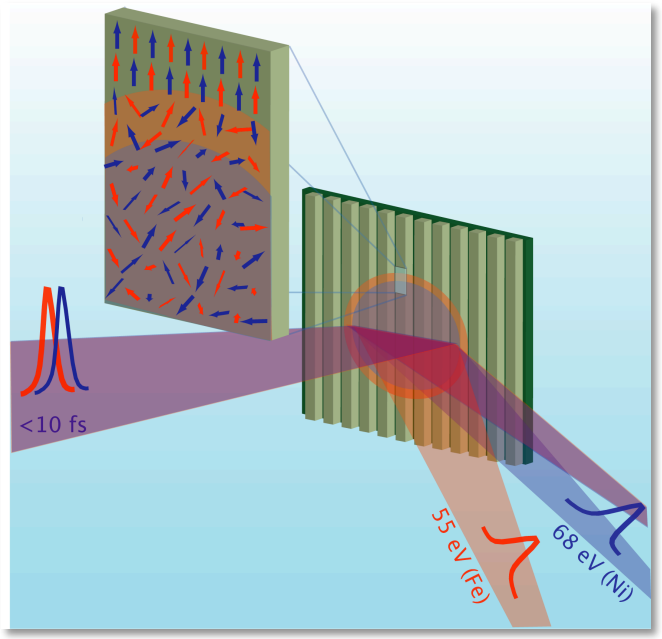
$$q = -k\nabla T$$

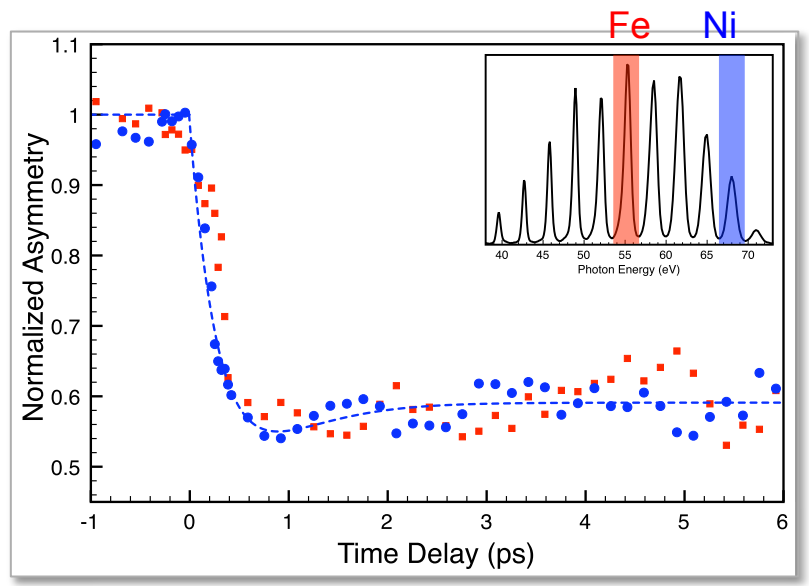
- What happens when a nanostructure is smaller than the phonon mean free path?
- Existing theories of nanoscale heat dissipation disagree
- Fourier law over-estimates the heat flow - need to think of interface ≈ phonon mean free path
- Ronggui Yang, Keith Nelson, Erik Anderson (Nature Materials **9**, 26 (2010))





Ultrafast, Element-Specific, Demagnetization Dynamics

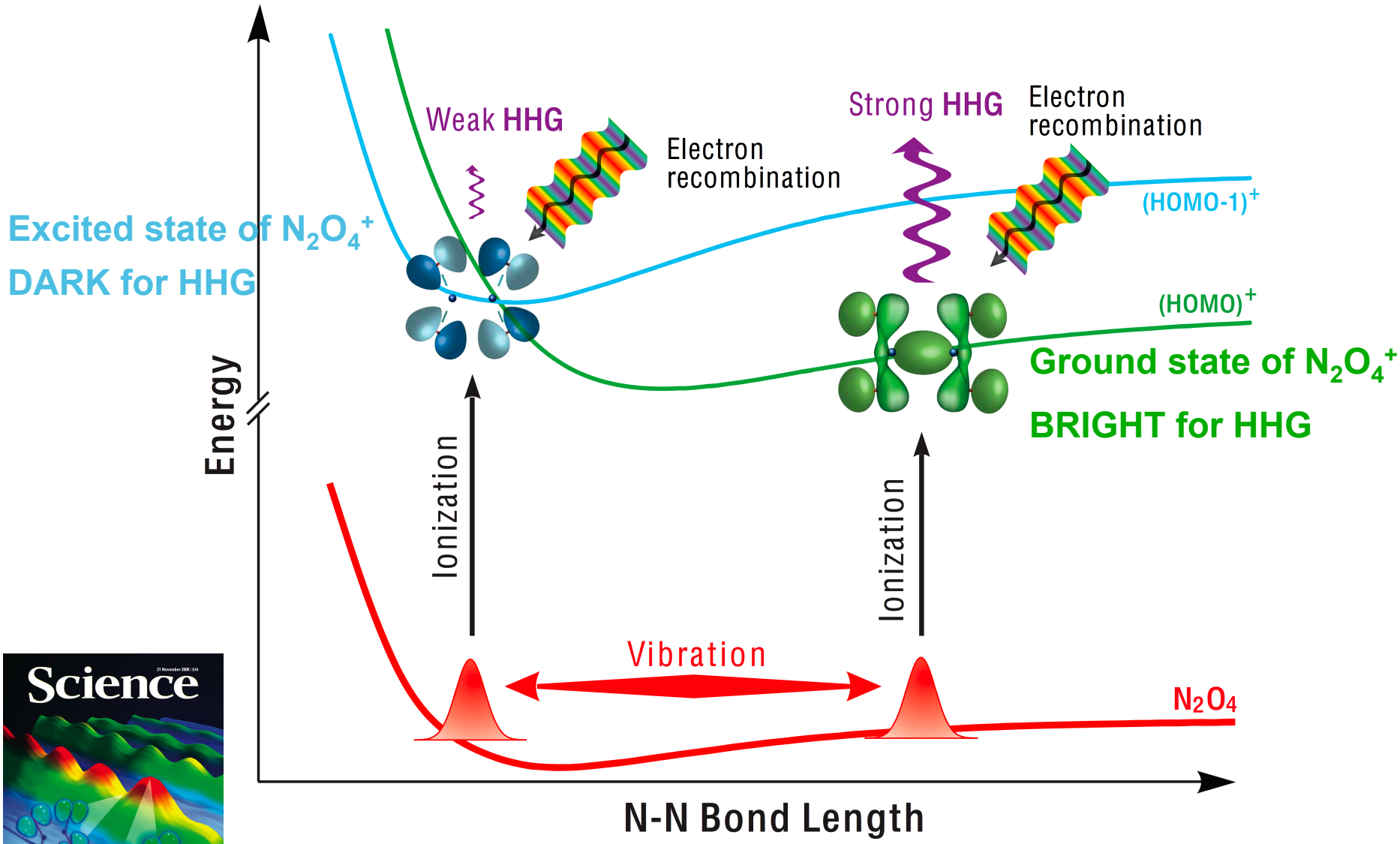


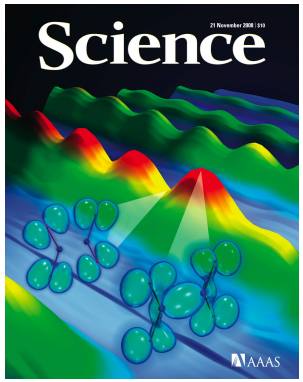


Phys. Rev. Lett. ***103****, 257402 (2009)*

- Use femtosecond laser to demagnetize a complex magnetic material
- Probe magnetization decay using ultrafast x-rays
- First experiment to –
  - Measure elementally-specific magnetization dynamics in a compound sample (Fe/Ni)
  - Achieve ultrafast time resolution in x-ray region (55 fs)
  - Magnetization decays within ≈ 400 fs for both Fe and Ni
- Important applications in understanding limiting speed of magnetic switching, coherent magnetics, spintronics

Capturing electron dynamics in molecules

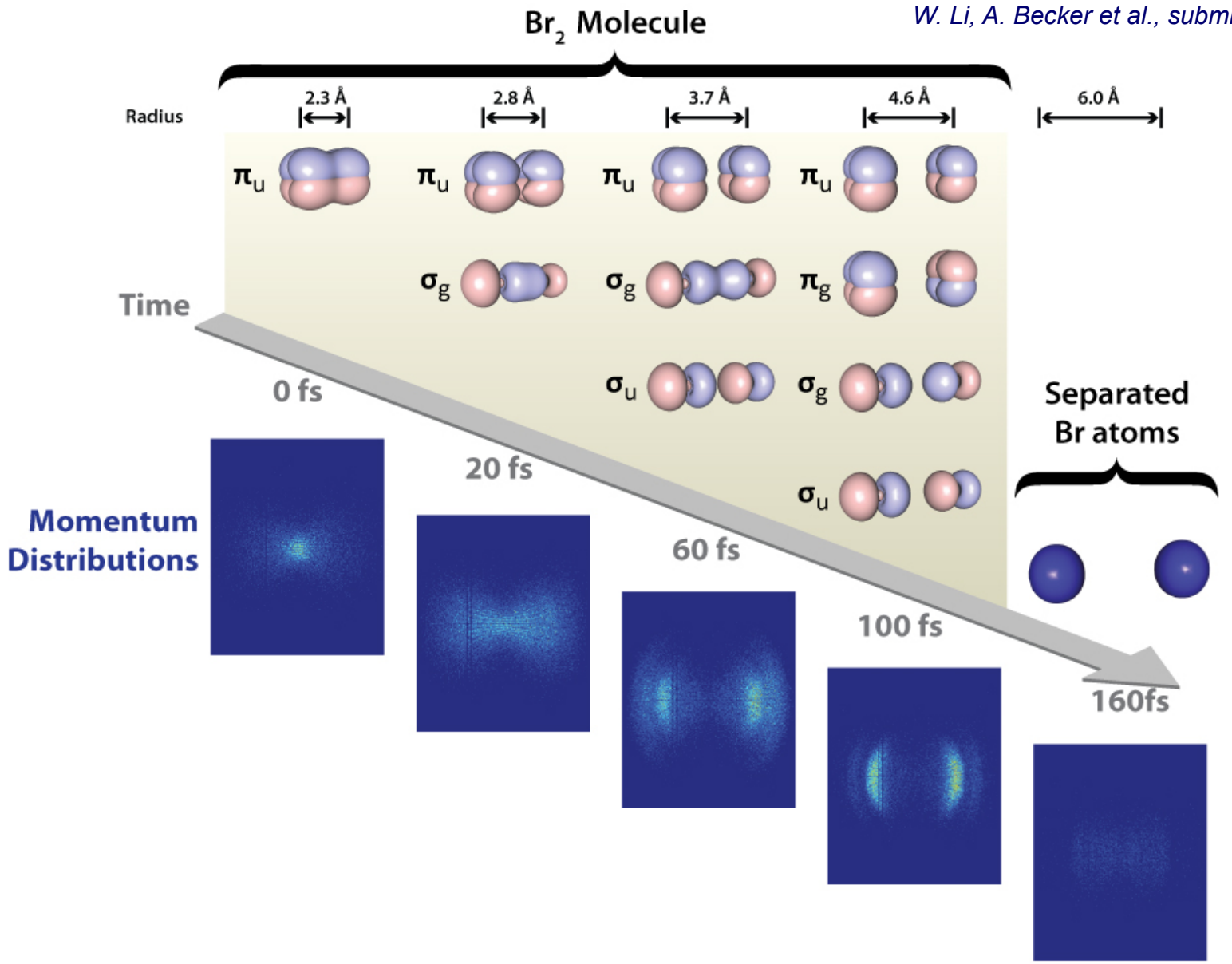




*Wen Li et al., Science **322**, 1207 (2008)*

Following electron orbital dynamics as a bond breaks

W. Li, A. Becker et al., submitted (2009)



Following electron orbital dynamics as a bond breaks

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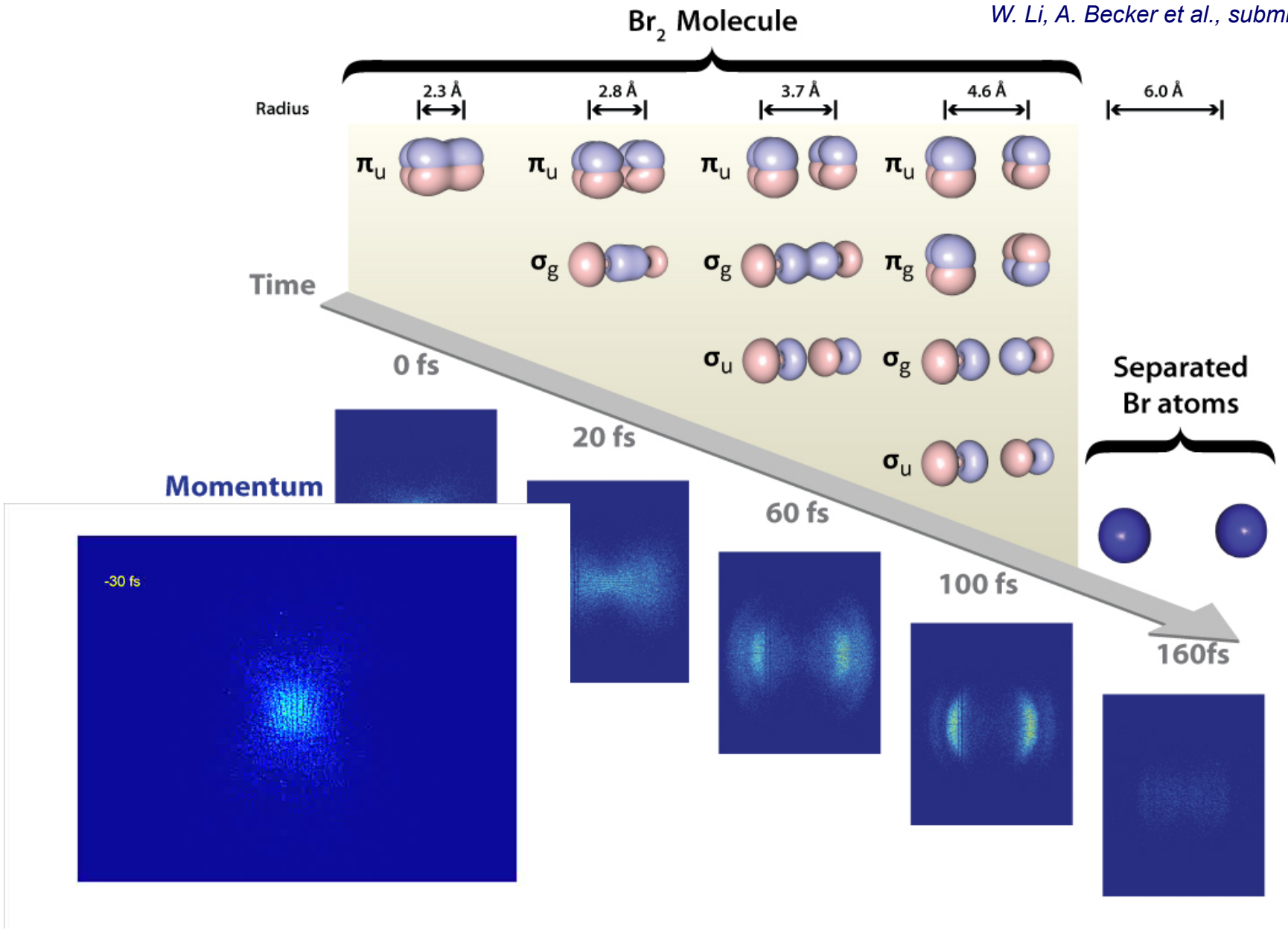
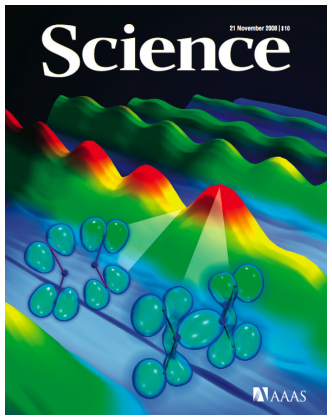
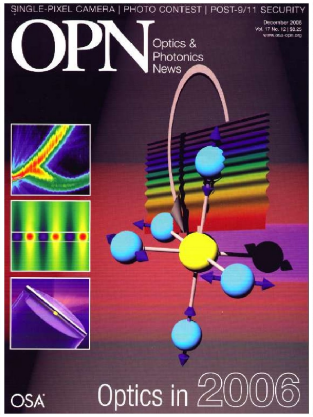
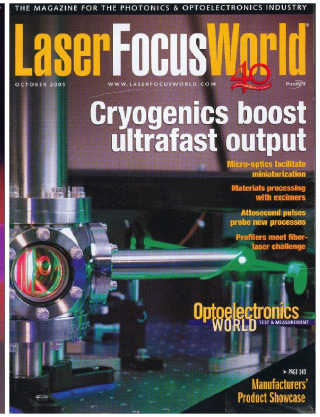
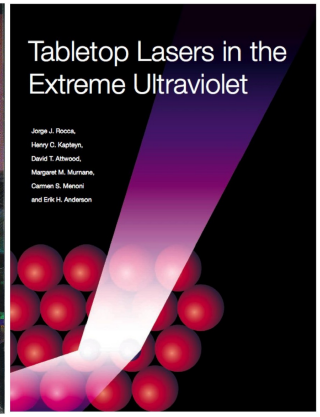


Future

- Take attosecond electron rescattering physics, discovered just over 20 years ago, to generate coherent x-rays and electrons
- Now have soft x-ray laser beams that contain many “colors” of x-rays all at once, with excellent prospects for hard x-ray laser beams on a tabletop
- 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 NSF, DOE, DOD