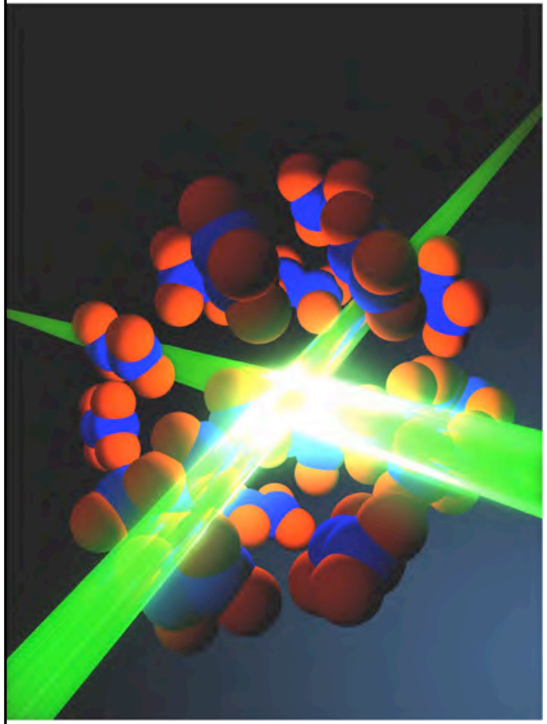
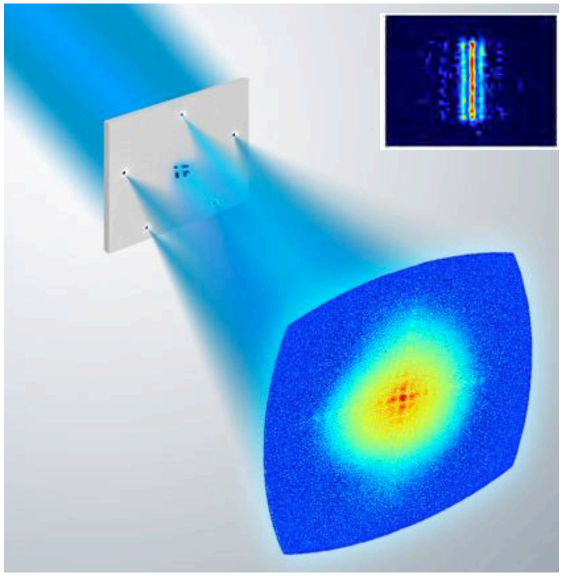
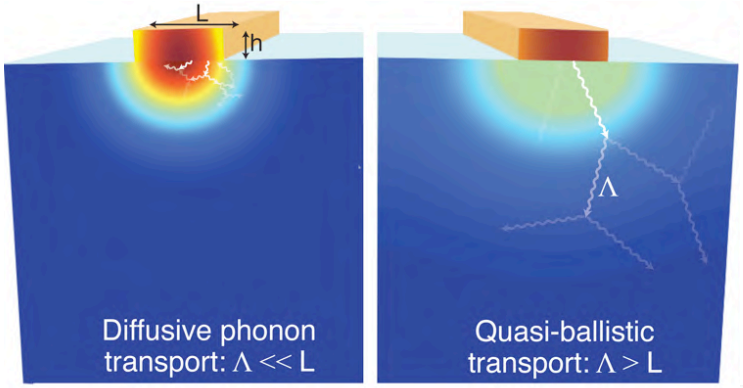
Science at the Time-scale of the Electron –Coherent Attosecond Soft and Hard X-Ray Harmonics

Henry Kapteyn and Margaret Murnane







KITP August 2010

Collaborators

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University of California, Berkeley and LBL

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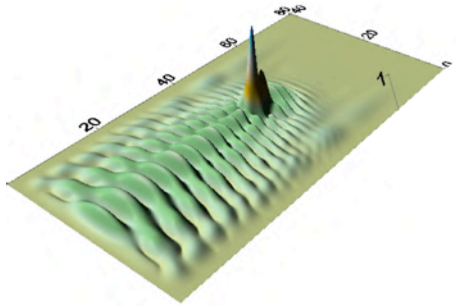
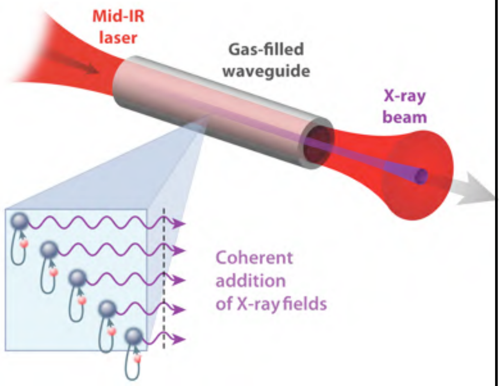
Colorado State University

Science at the timescale of the electron – so what?



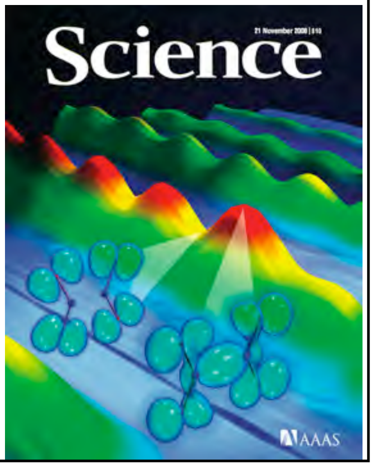
I. Bright coherent x-ray beams on a tabletop

- —Ultrafast lasers can manipulate electrons on their fundamental timescale to implement a coherent version of the x-ray tube
- —Bright coherent tabletop beams at > 0.5 keV
- —Duration of 10 attoseconds – soon zeptoseconds!



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

- —Image reactions at the level of electrons
- —Understand energy/charge transport at the nanoscale
- —Capture correlated electron dynamics (spin, molecules, materials)
- —Elemental and chemical nanoprobe of thick samples



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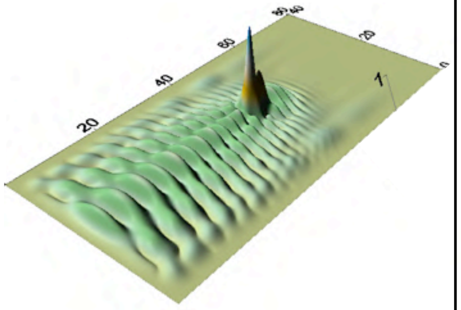

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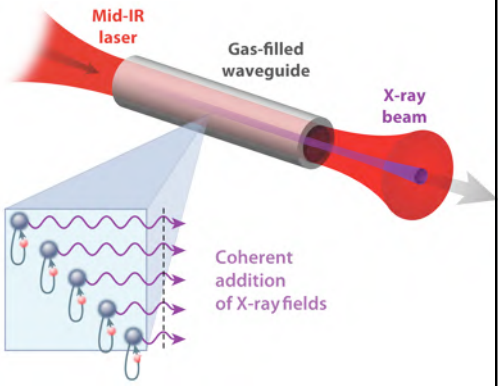

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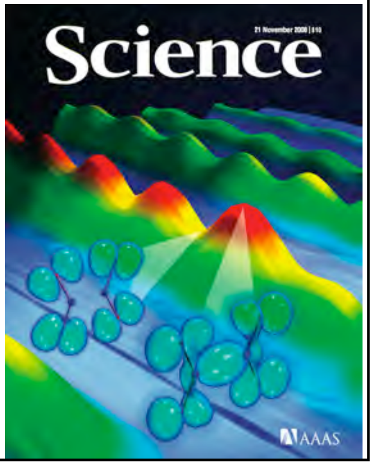
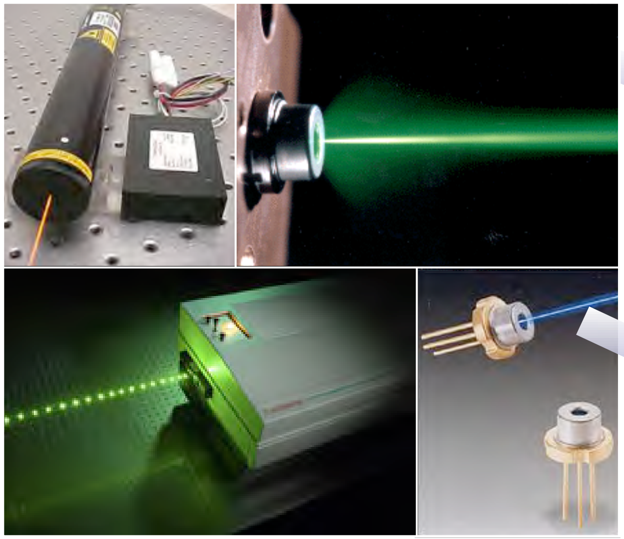
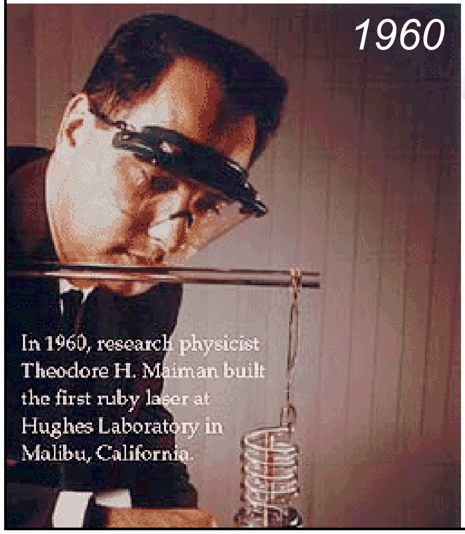
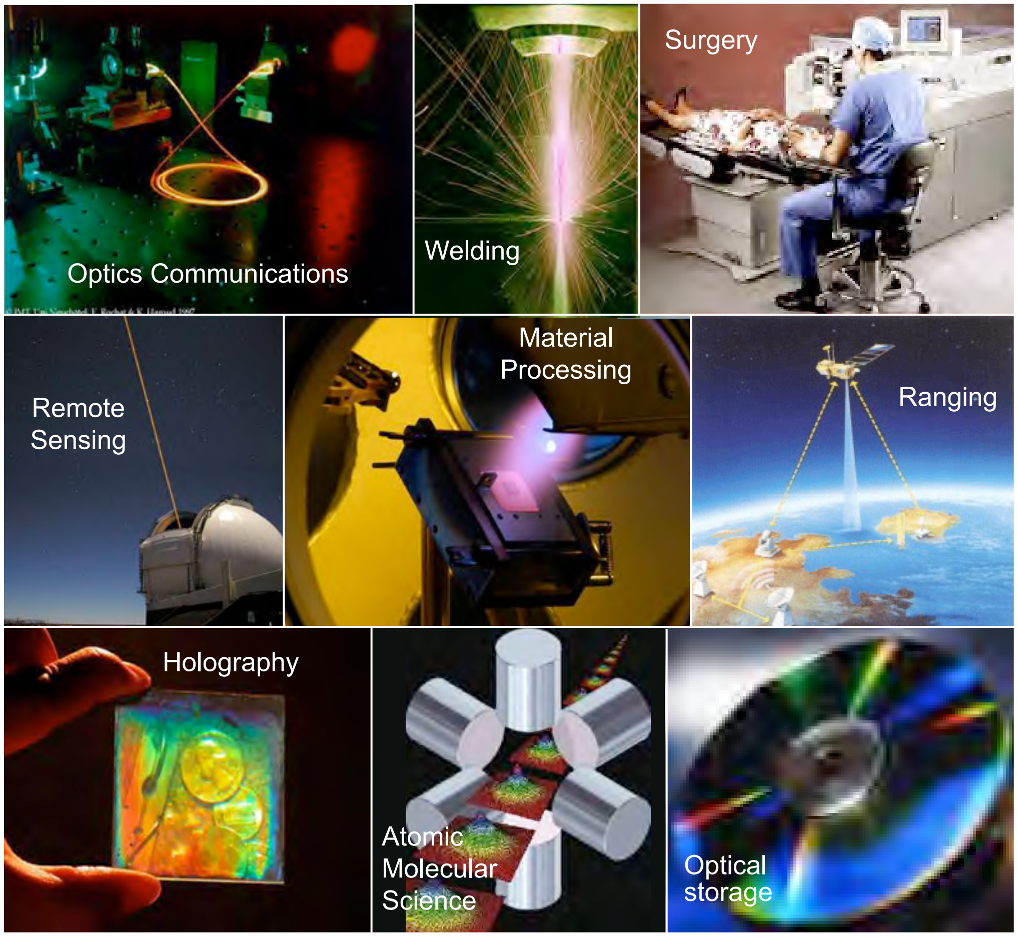


Coherent light has transformed society



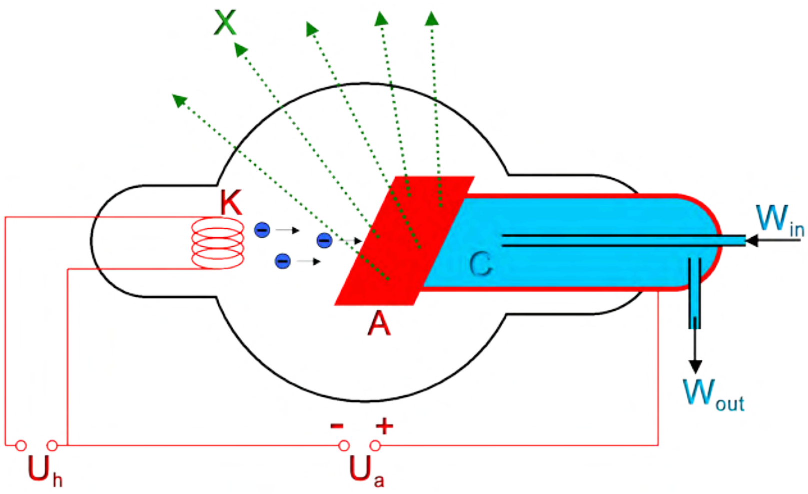

1960

In 1960, research physicist Theodore H. Maiman built the first ruby laser at Hughes Laboratory in Malibu, California.

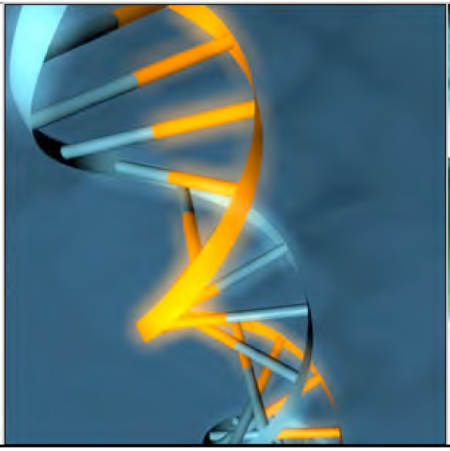


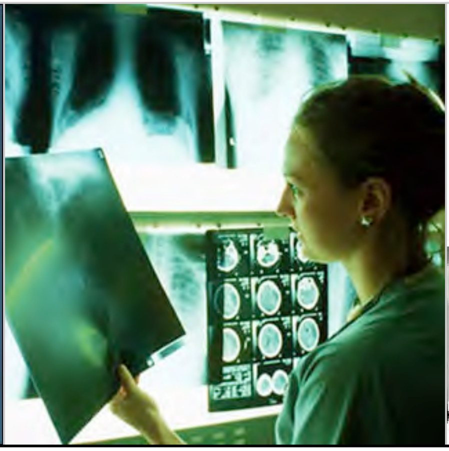
And so have x-rays…..

Wilhelm Roentgen

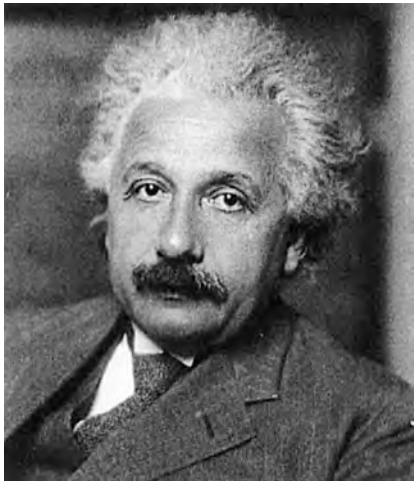


X-ray tube





So, what about coherent x-rays? X-ray lasers



Spontaneous emission / Stimulated emission

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

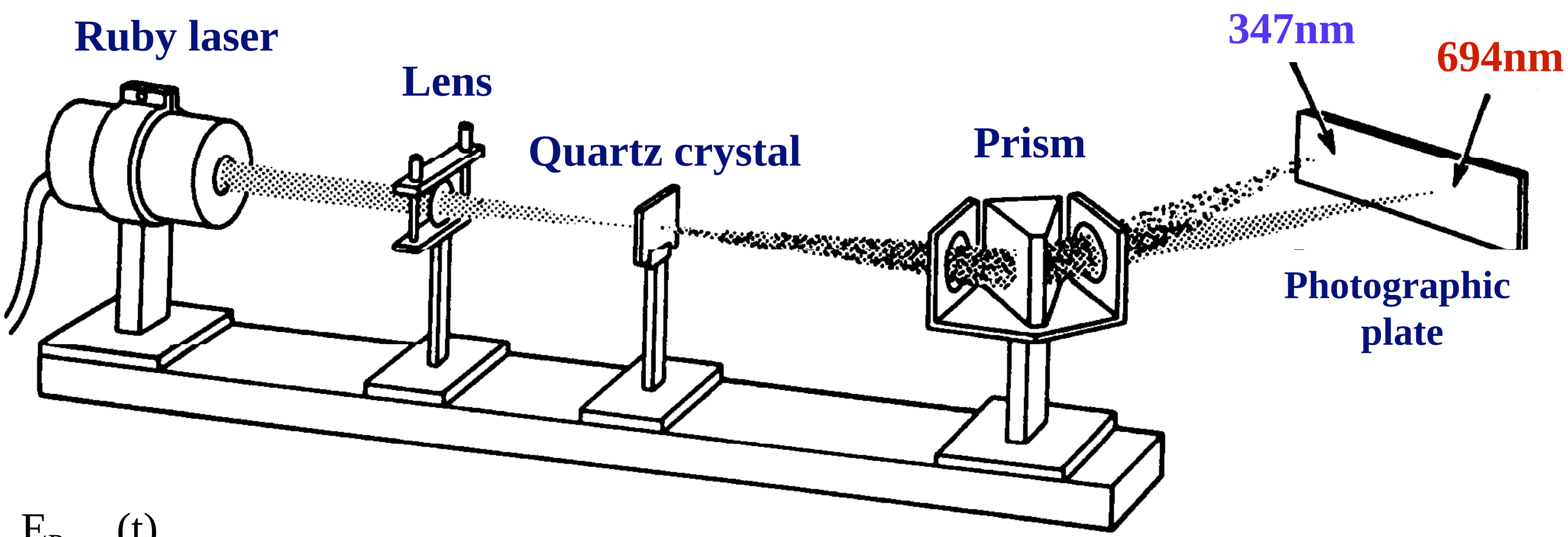
$$Power \propto \left(\frac{1}{\sigma_g}\right)\left(\frac{1}{\tau}\right)(h\nu) \propto \frac{1}{\lambda^5}$$

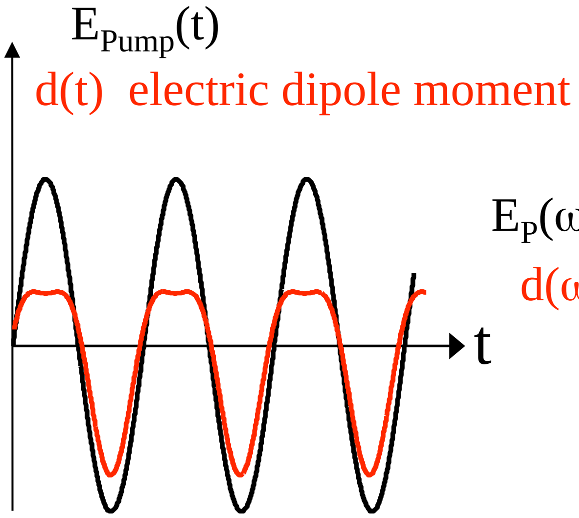
- 1 µm -> 1 mW
- 1 nm -> TW
- 1 Å -> 1 PW

LCLS X-ray free electron laser at 1.5nm

An alternative approach: Nonlinear Optics

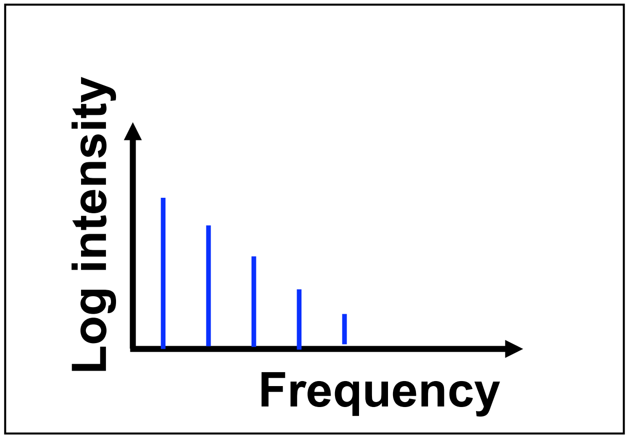
P.A. Franken et al, PRL 7, 118 (1961)





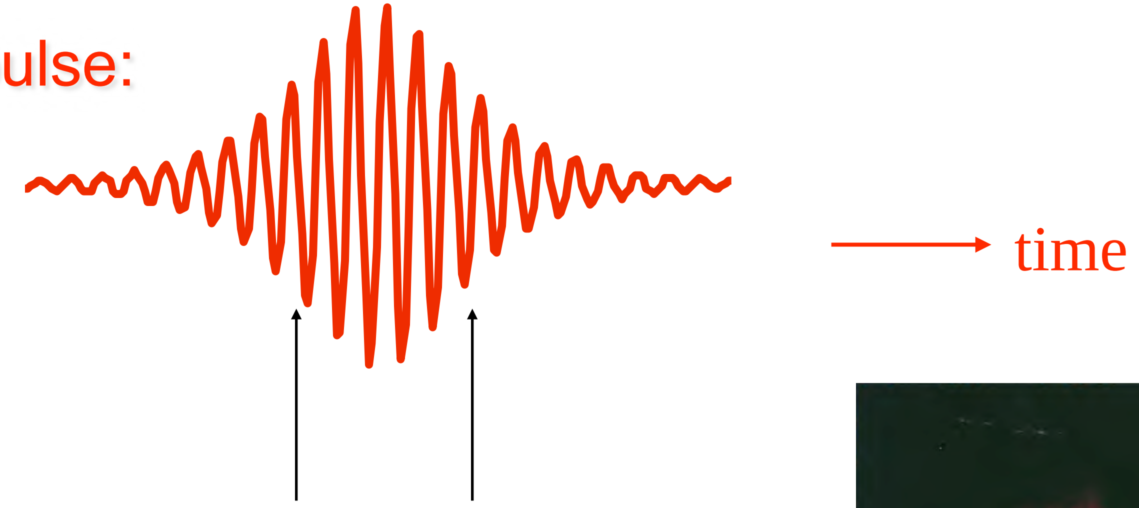
$E_P(\omega)=E_0^{i\omega t}$

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



Extreme NLO: start with an ultrafast laser pulse

10 fs light pulse:



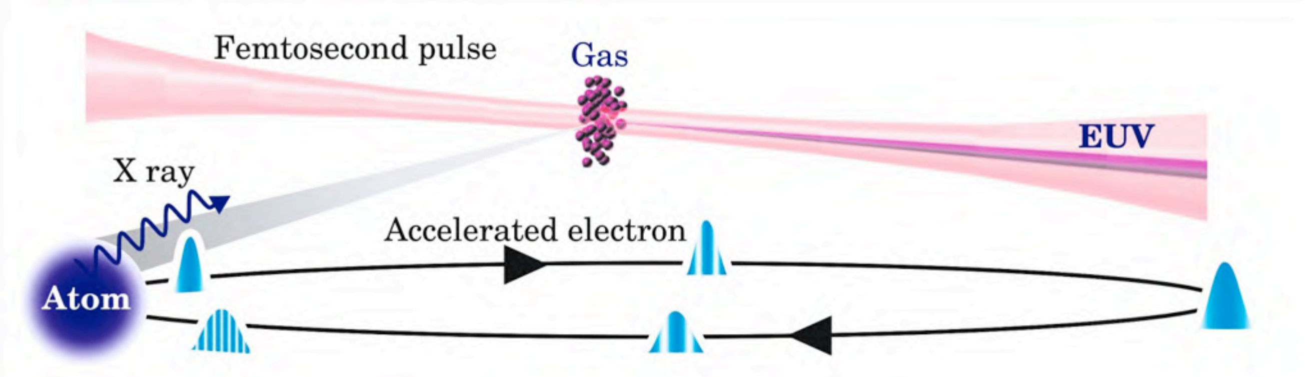
time

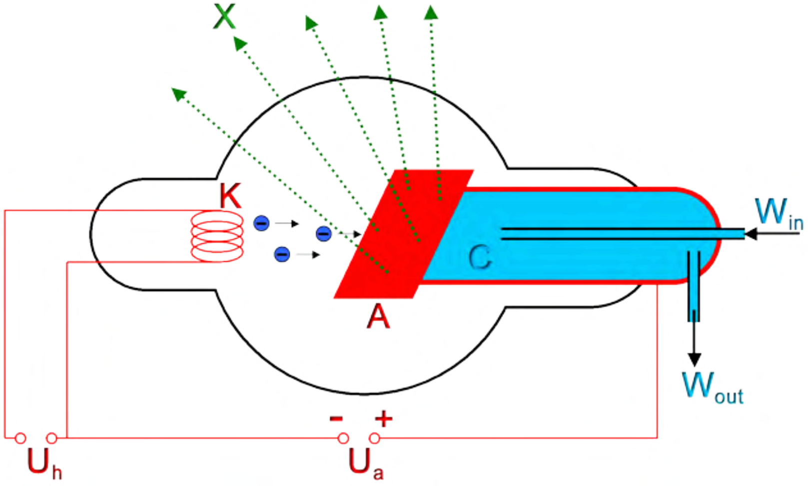
Δx=3 micrometers
= 1/50 human hair

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

High-order Harmonic Generation– NLO using unbound states

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



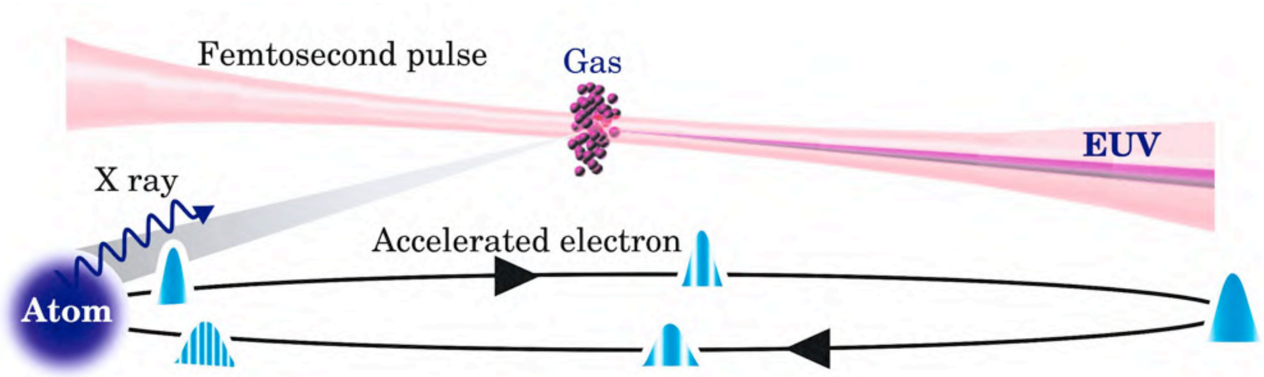


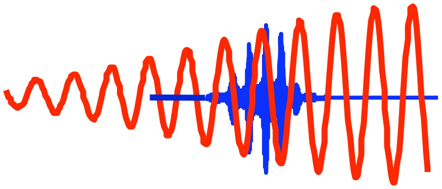
X-ray tube

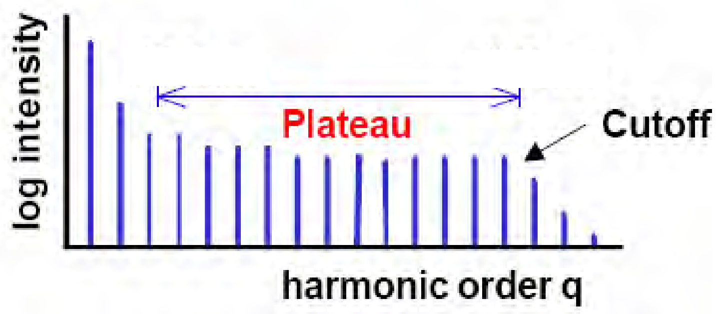
*SILAP Proc. pp. 95-110 (1993); PRA **49**, 15 (1994)*

High Harmonic Generation

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







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

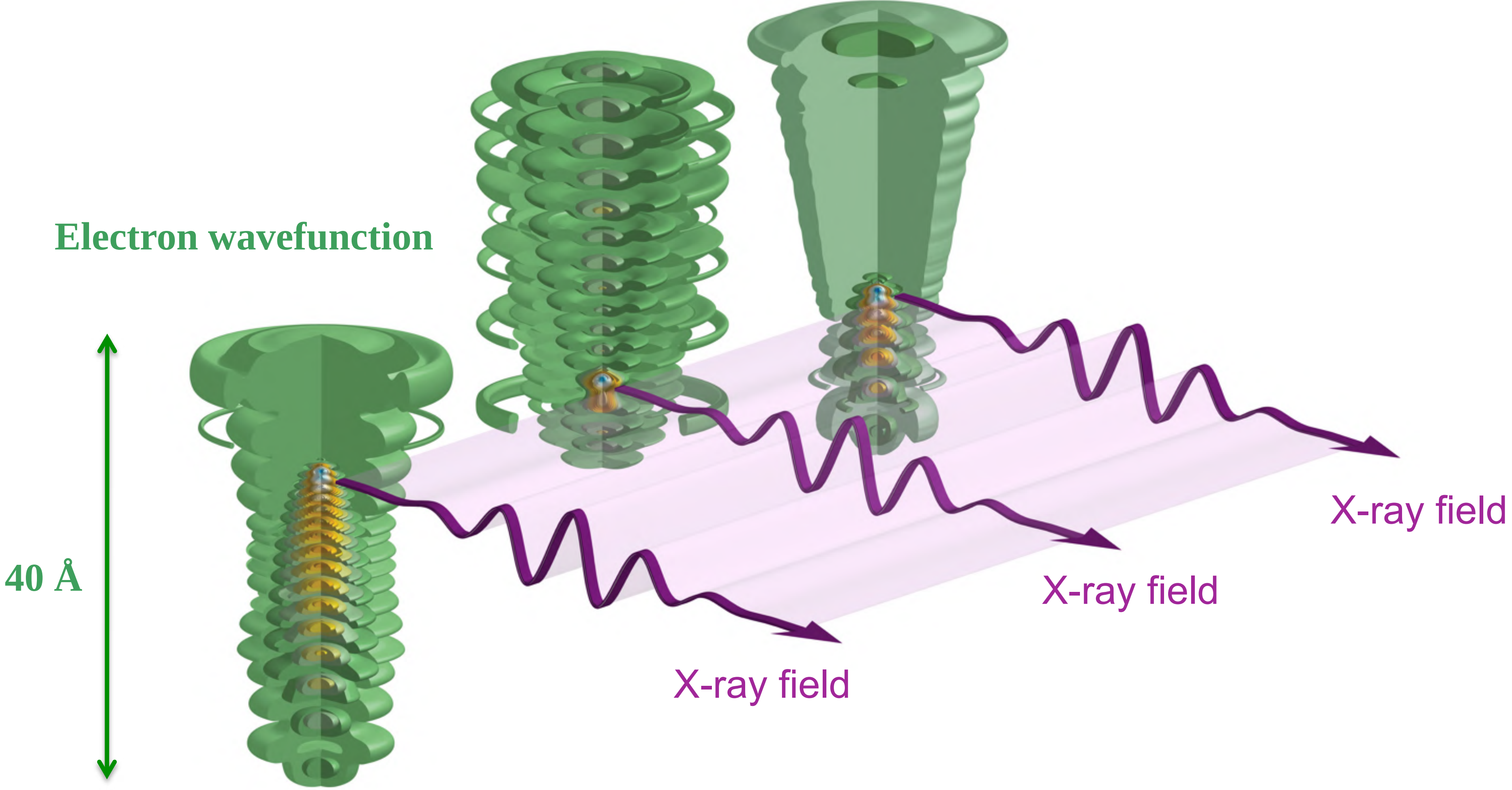
< 10 fs duration

Broad frequency range UV - keV

Electron wriggle energy coherently converts to x-rays

High Harmonic Generation – quantum picture

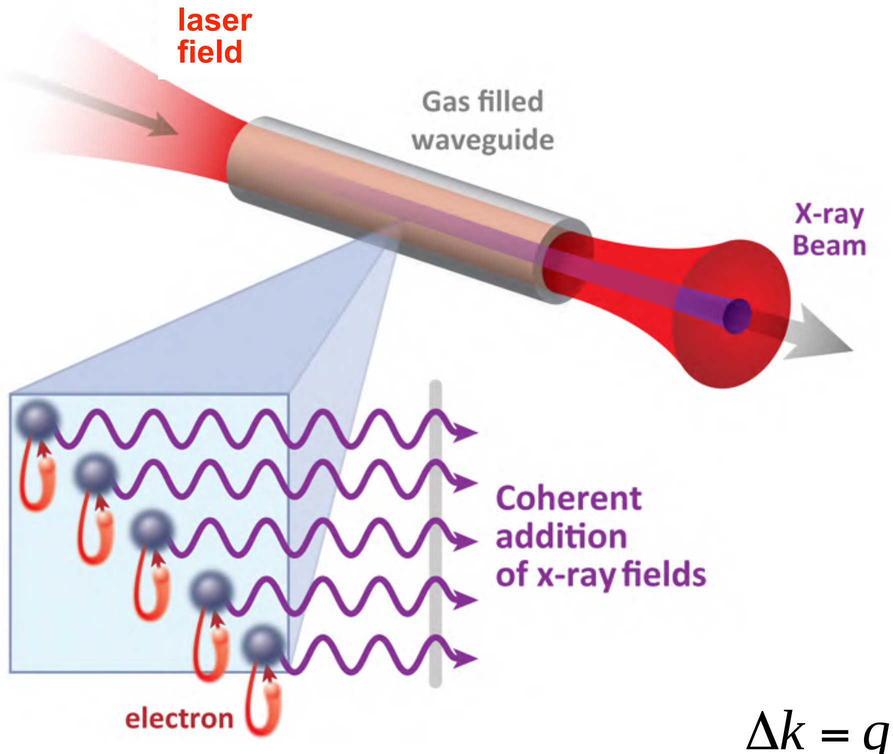
- Electron takes a few fs to leave vicinity of an atom after being ionized
- During that time, wavefunction is highly modulated due to recollisions
- Modulations give rise to high harmonics in radiated field



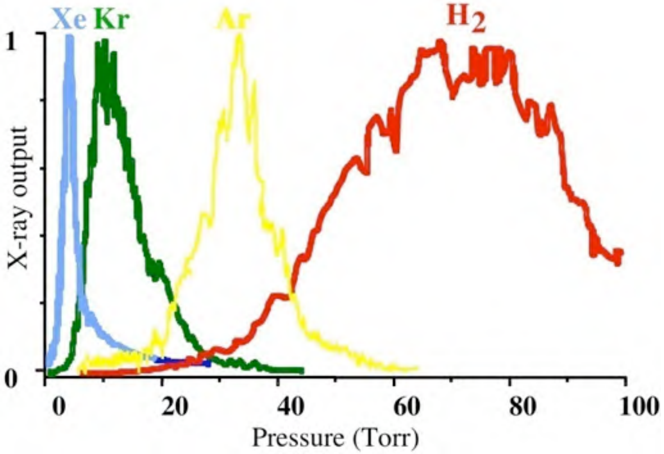
Simulations by Ivan Christov

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



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



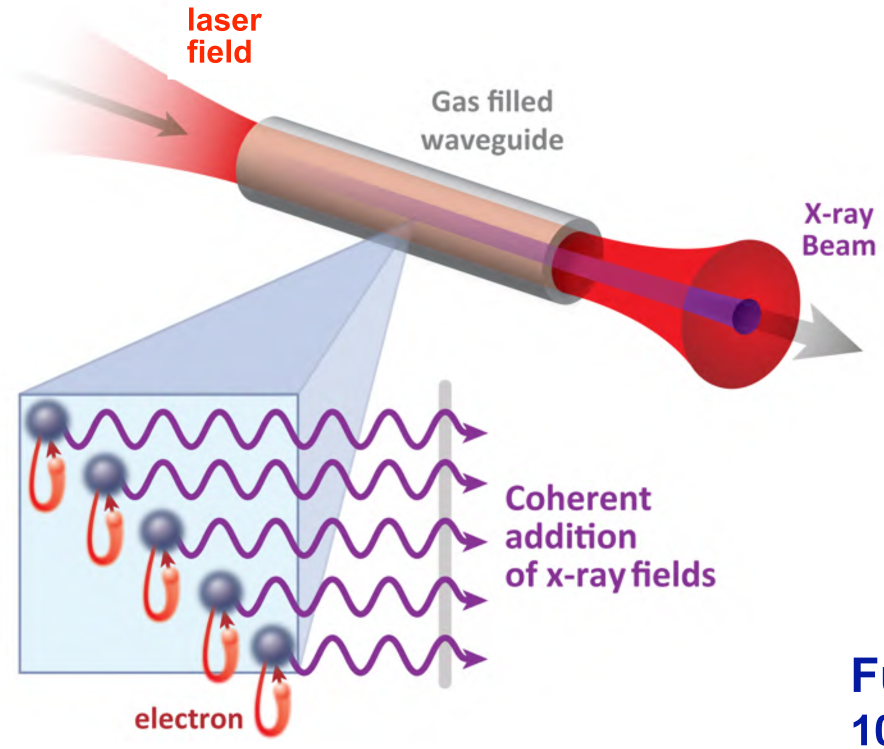
$$\Delta k = qk_{laser} - k_{HHG} = 0$$

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

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, harmonics in EUV region < 150 eV



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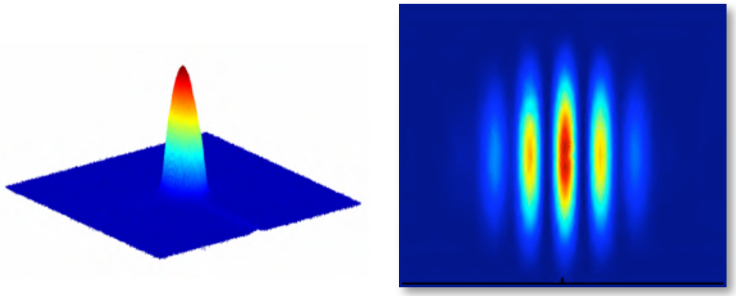


Fully coherent bright EUV and soft x-rays

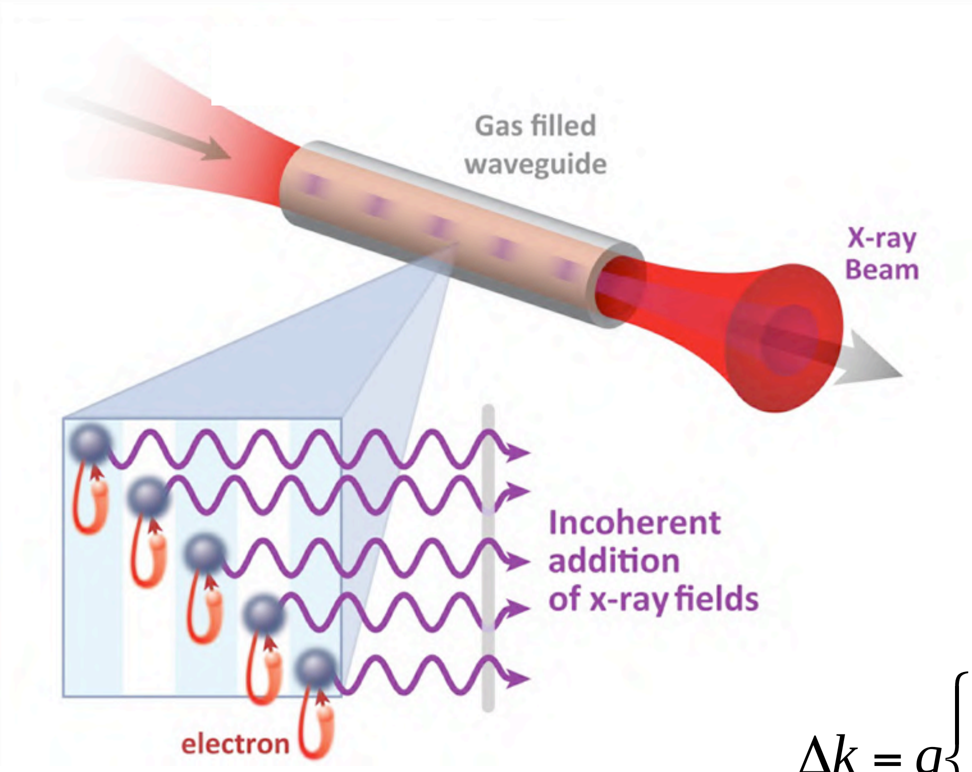
10^{-5} or nJ per harmonic, uW average powers

Femtosecond-to-attosecond duration

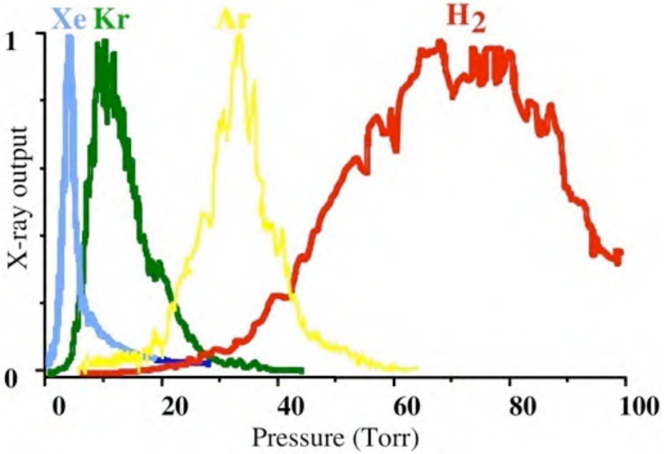
Phys. Rev. Lett. ***78****, 1251 (1997)*
Science ***280****, 1412 (1998)*
Phys. Rev. Lett. ***83****, 2187 (1999)*
Science ***297****, 376 (2002)*

Limits of phase matching

- To generate high energy x-rays, need high electron energy - ionize gas
- Presence of plasma speeds up laser –no phase matching above 150 eV



$V_{laser} = V_{X\text{-}ray} \neq C$



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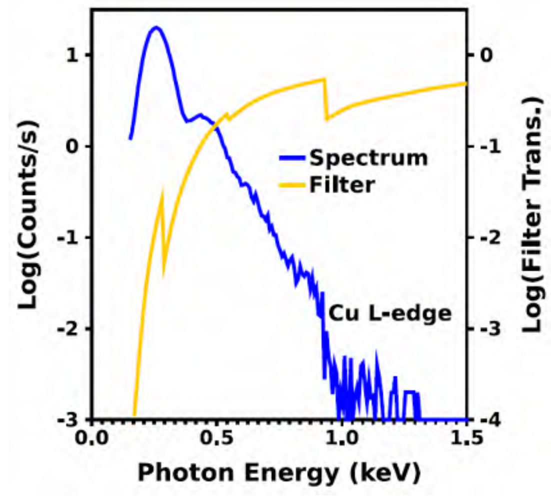
Waveguide **Neutrals** **Plasma**

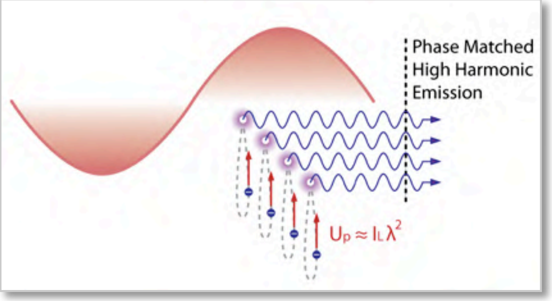
Generating very high energy harmonics

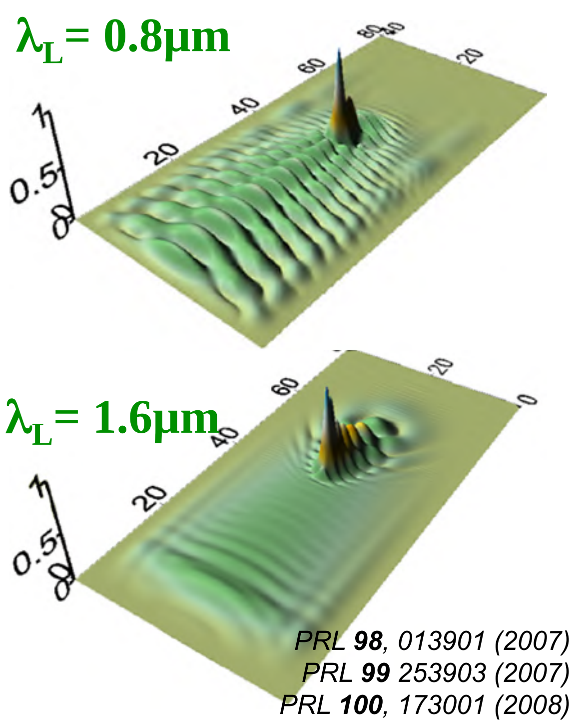
Single atom cutoff photon energy: $h\nu_{cutoff} = I_p + 3.2\, I_L \lambda_L^2$



Nature ***433****, 596 (2005)*

Ionization high - no phase matching





$\lambda_L = 0.8\mu m$

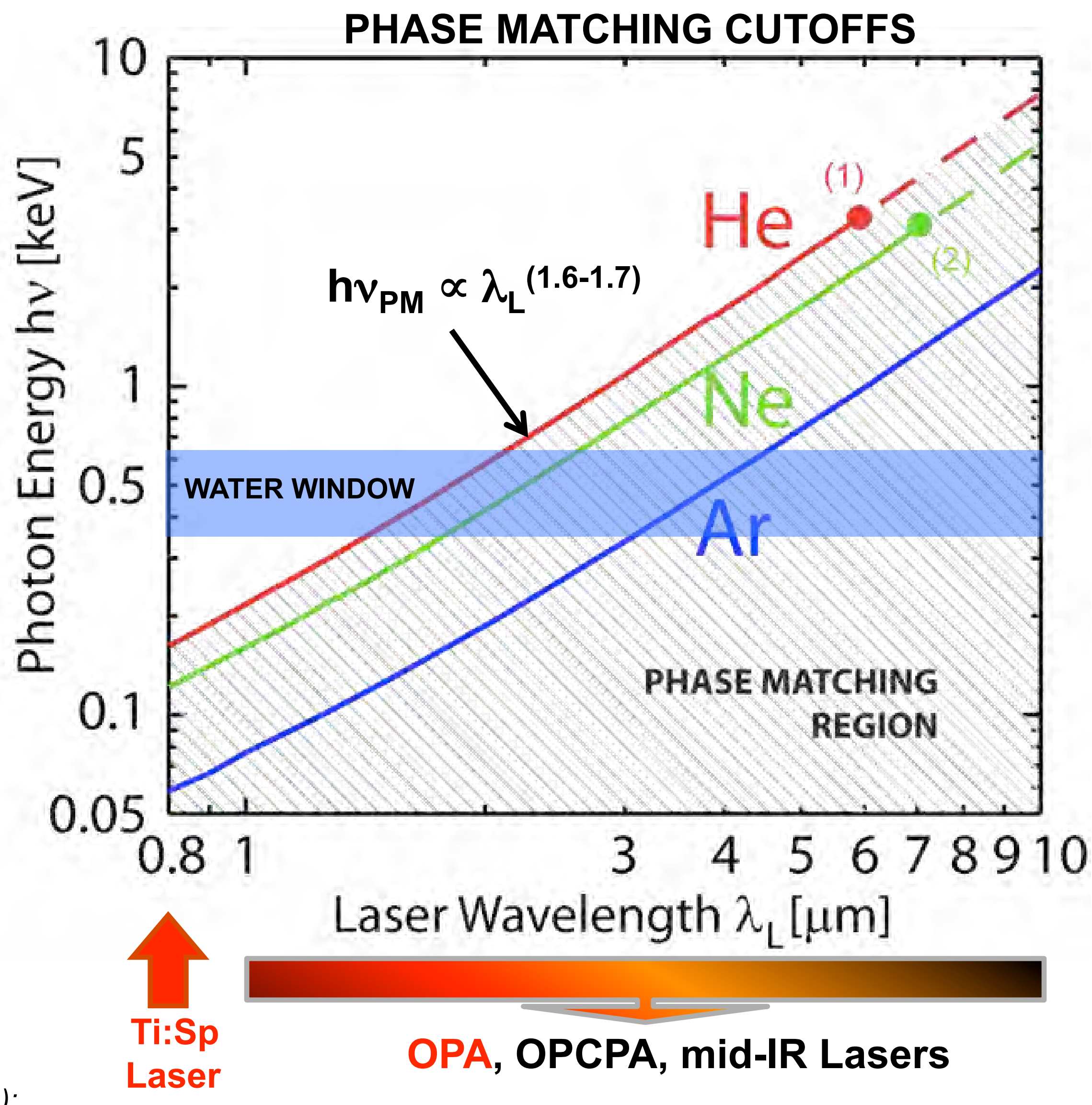
$\lambda_L = 1.6\mu m$

PRL ***98****, 013901 (2007)*
PRL ***99*** *253903 (2007)*
PRL ***100****, 173001 (2008)*

Single atom yield $\propto \lambda_L^{-5.5}$

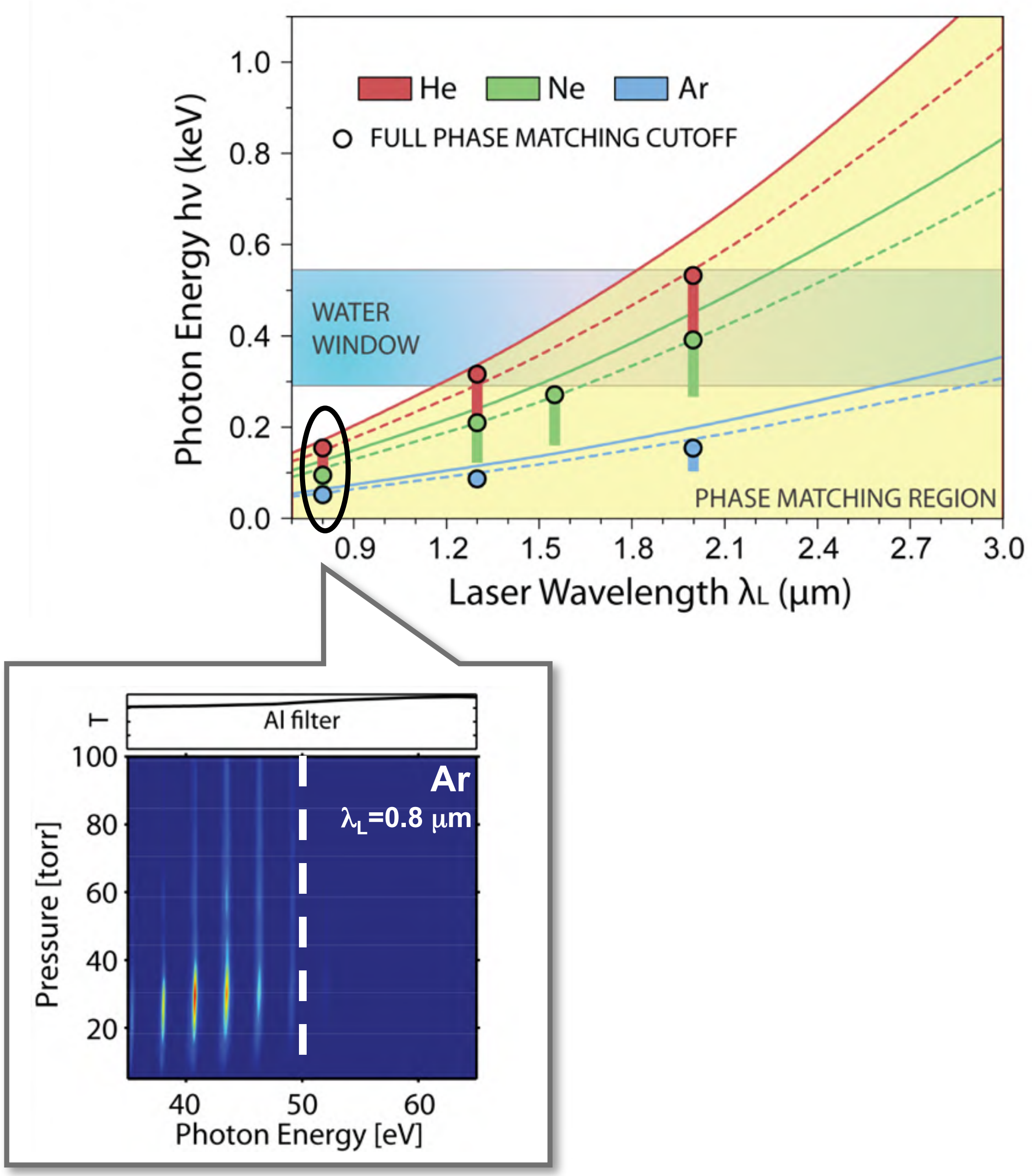
Phase matching using mid-infrared lasers

- IR lasers need lower intensity for a given harmonic energy
- Lower laser intensity = > lower ionization, better phase matching
- Predict $\mathbf{h\nu_{PM}} \propto \lambda_L^{(1.6\text{-}1.7)}$
- Single atom response also lower for mid-IR drivers ($\lambda^{-5.5}$)
- **BUT** phase matching pressure and gas transparency increase and compensate for low yield!



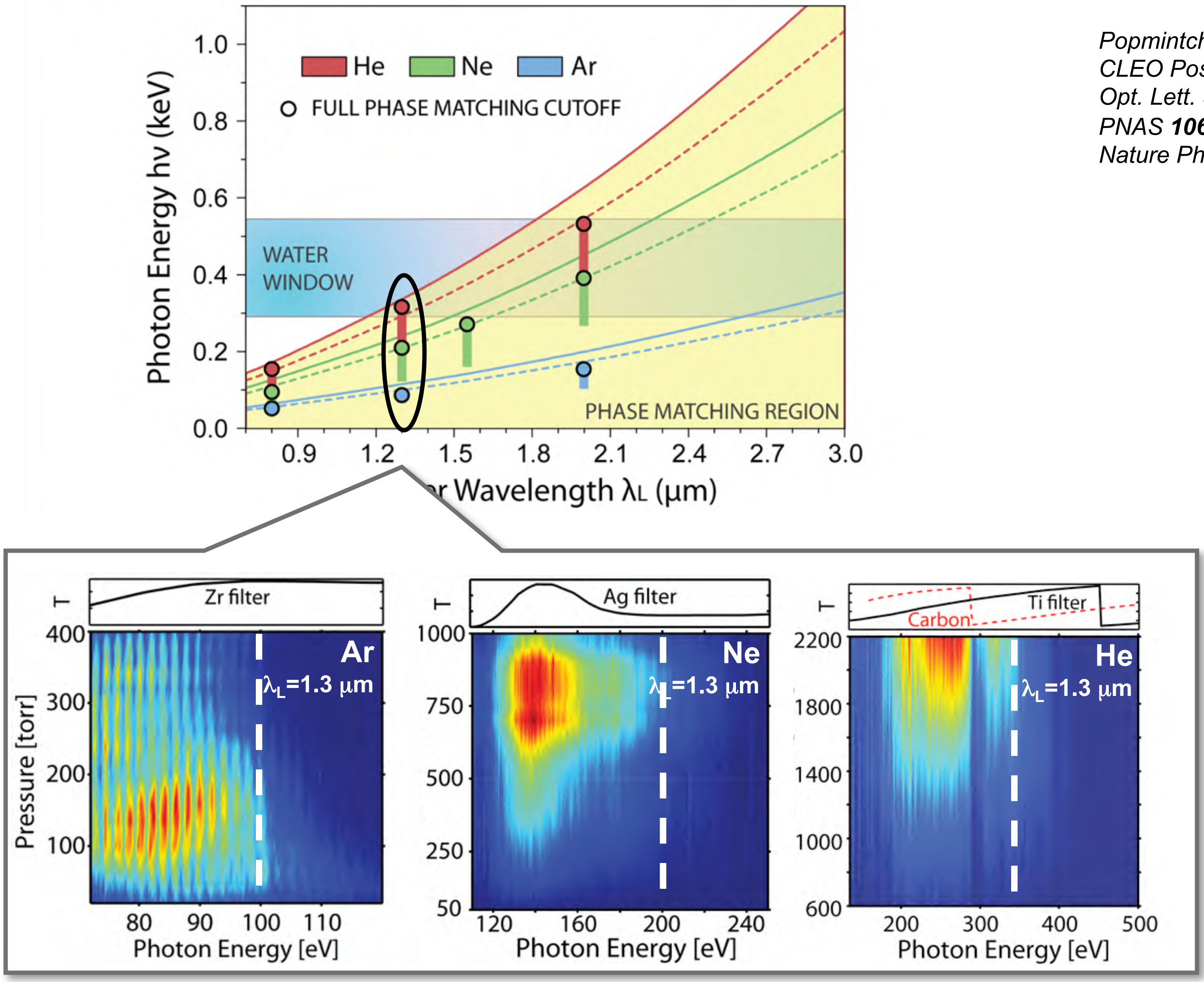
T. Popmintchev et al. CLEO Postdeadline CPDA9 (2008)
*Optics Letters **33**, 2128 (2008); PNAS, **106**, 10516 (2009);*
Nature Photonics, tbp (2010)

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



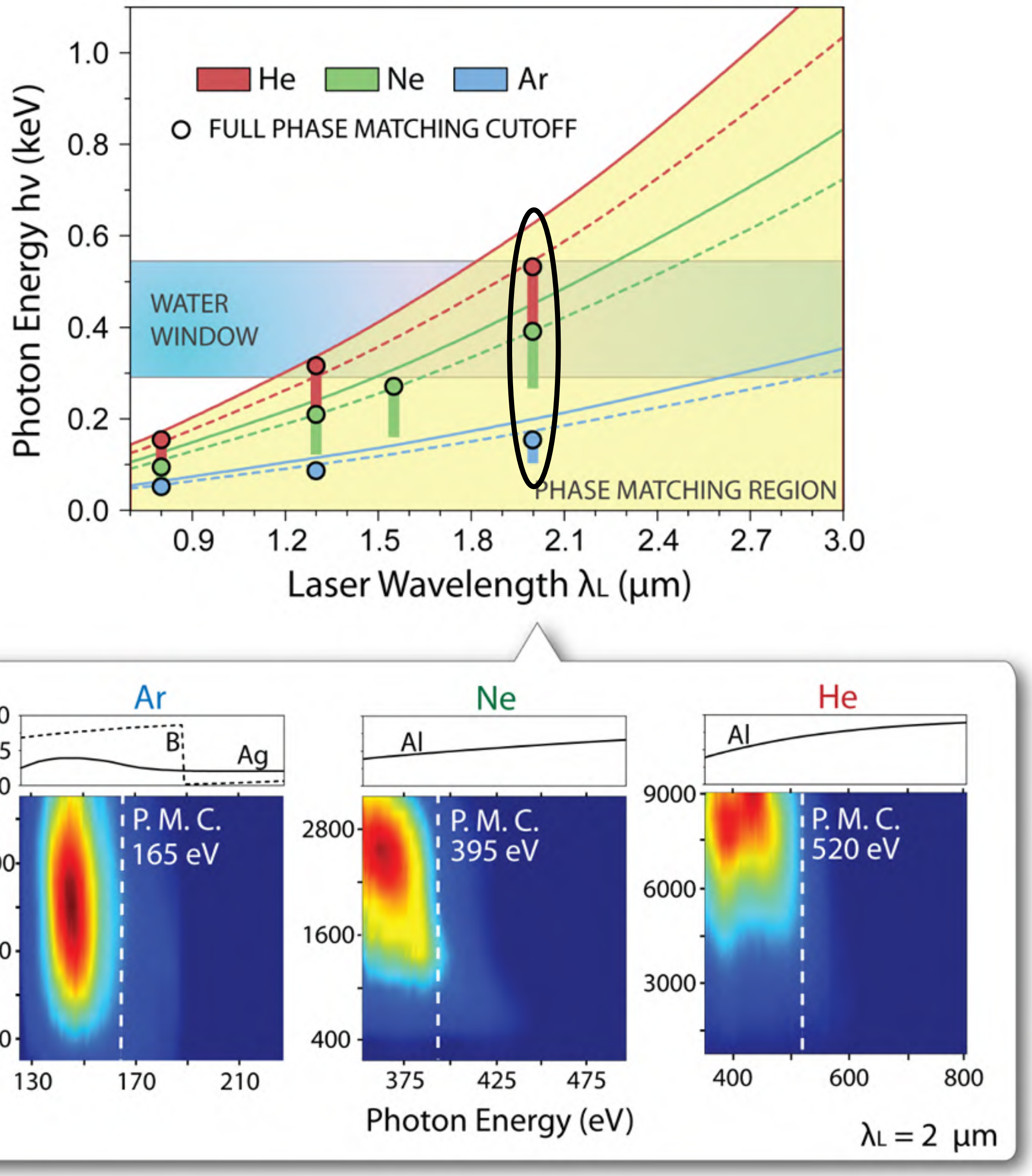
Popmintchev et al.,
CLEO Postdeadline CPDA9 (2008);
Opt. Lett. ***33****, 2128 (2008);*
PNAS ***106****, 10516 (2009);*
Nature Photonics to be publ. (2010)

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

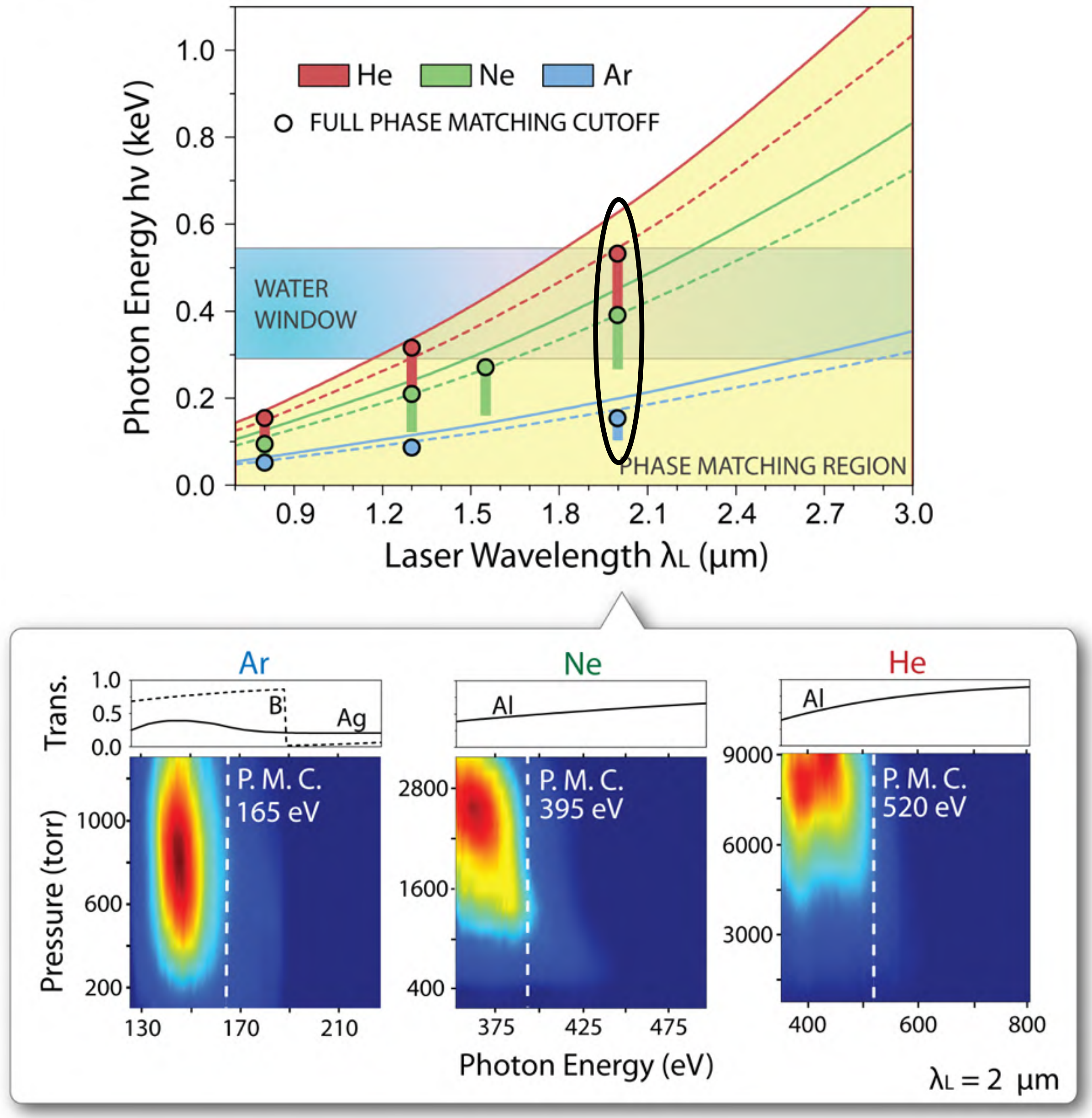


Popmintchev et al.,
CLEO Postdeadline CPDA9 (2008);
Opt. Lett. ***33****, 2128 (2008);*
PNAS ***106****, 10516 (2009);*
Nature Photonics to be publ. (2010)

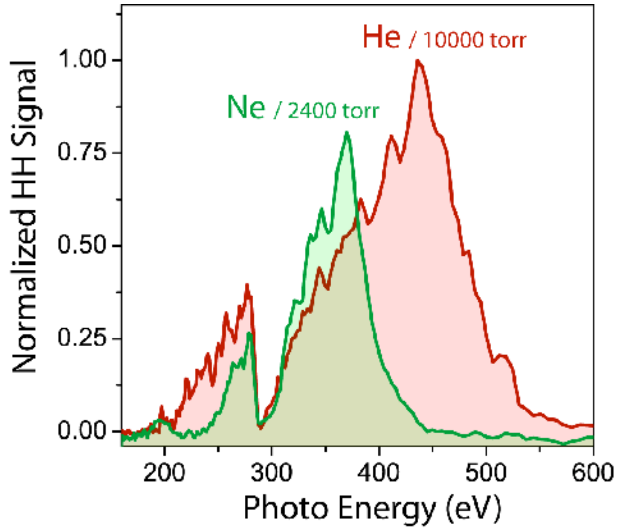
Full phase matching at >0.5 keV using λ_L= 2 μm



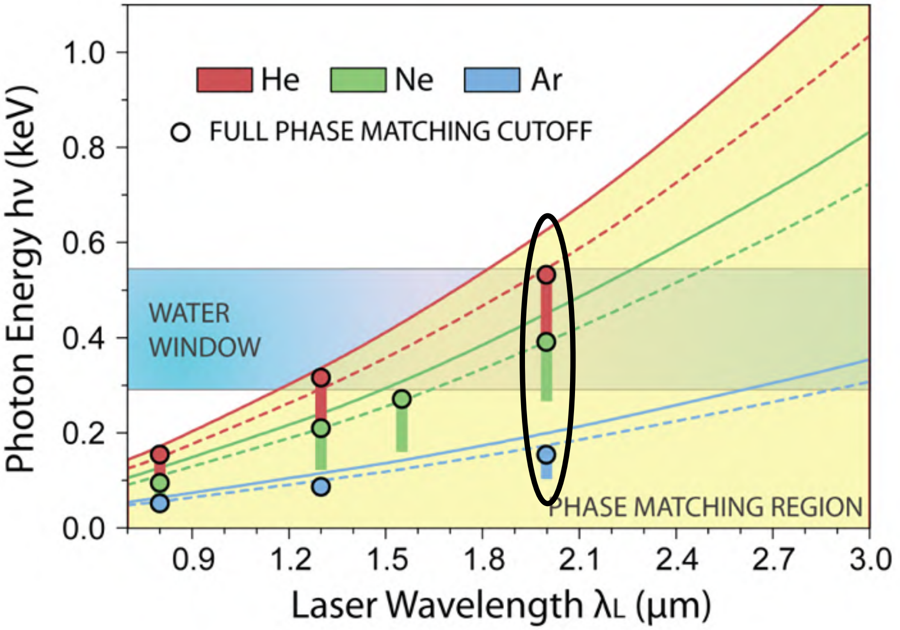
Broad soft x-ray supercontinuum spans > 300 eV

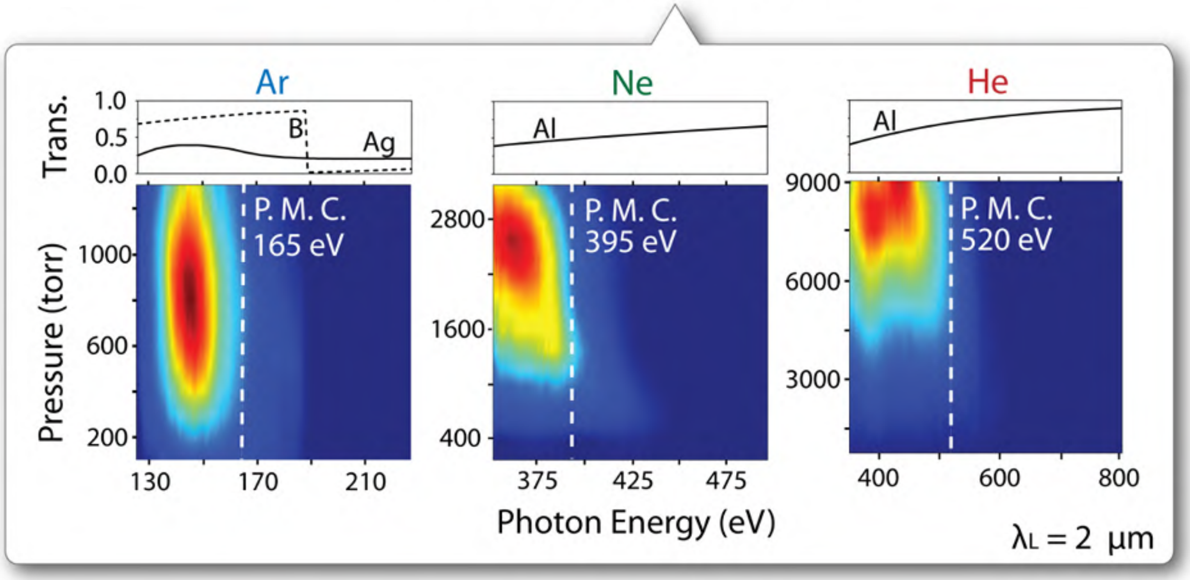


HHG flux high: 10^6 photons/s in $\lambda/\Delta\lambda$ of 100



Bright spatially coherent laser-like output





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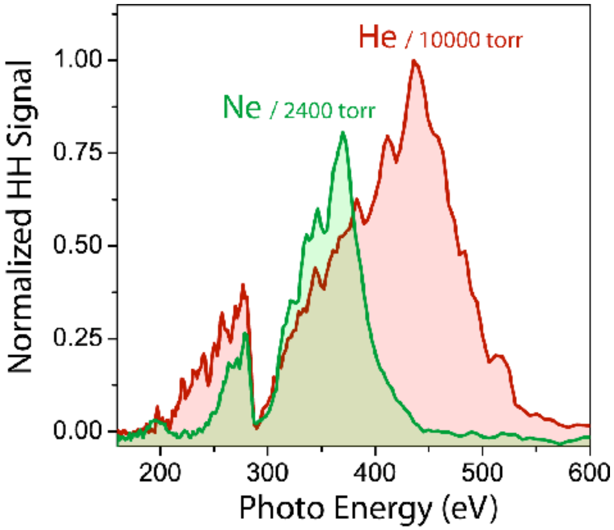
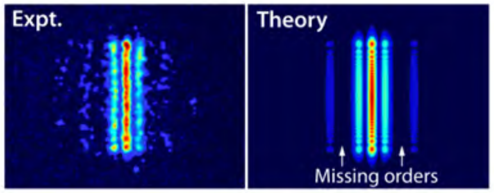
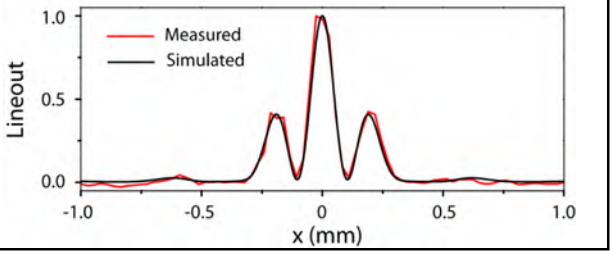
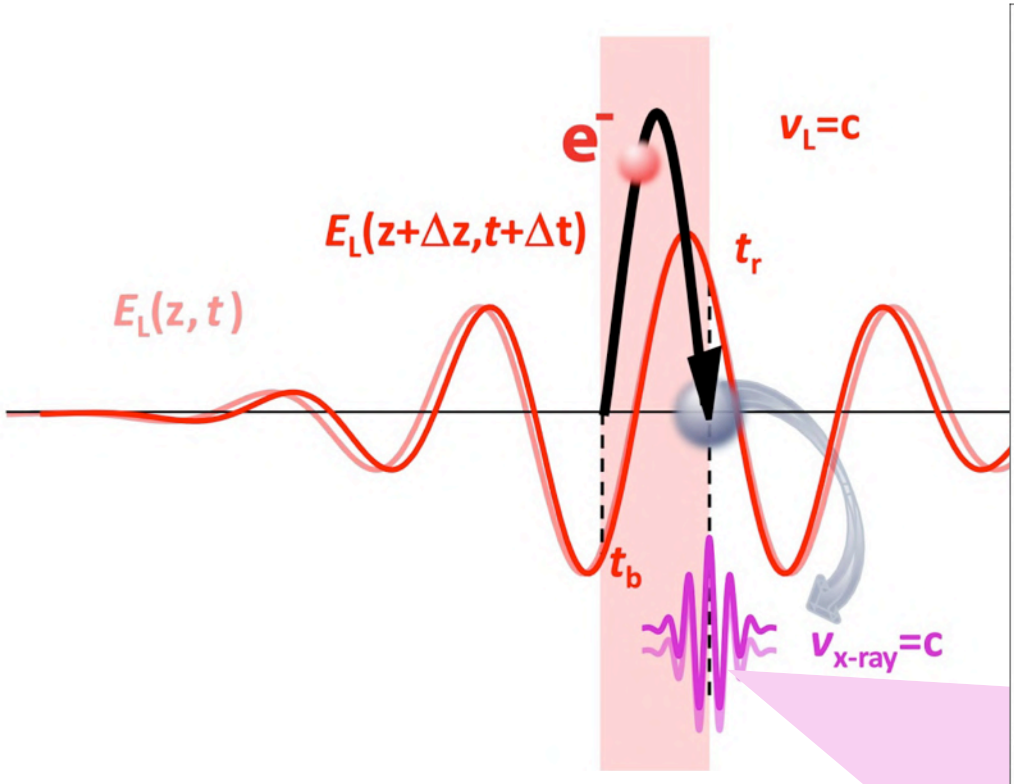


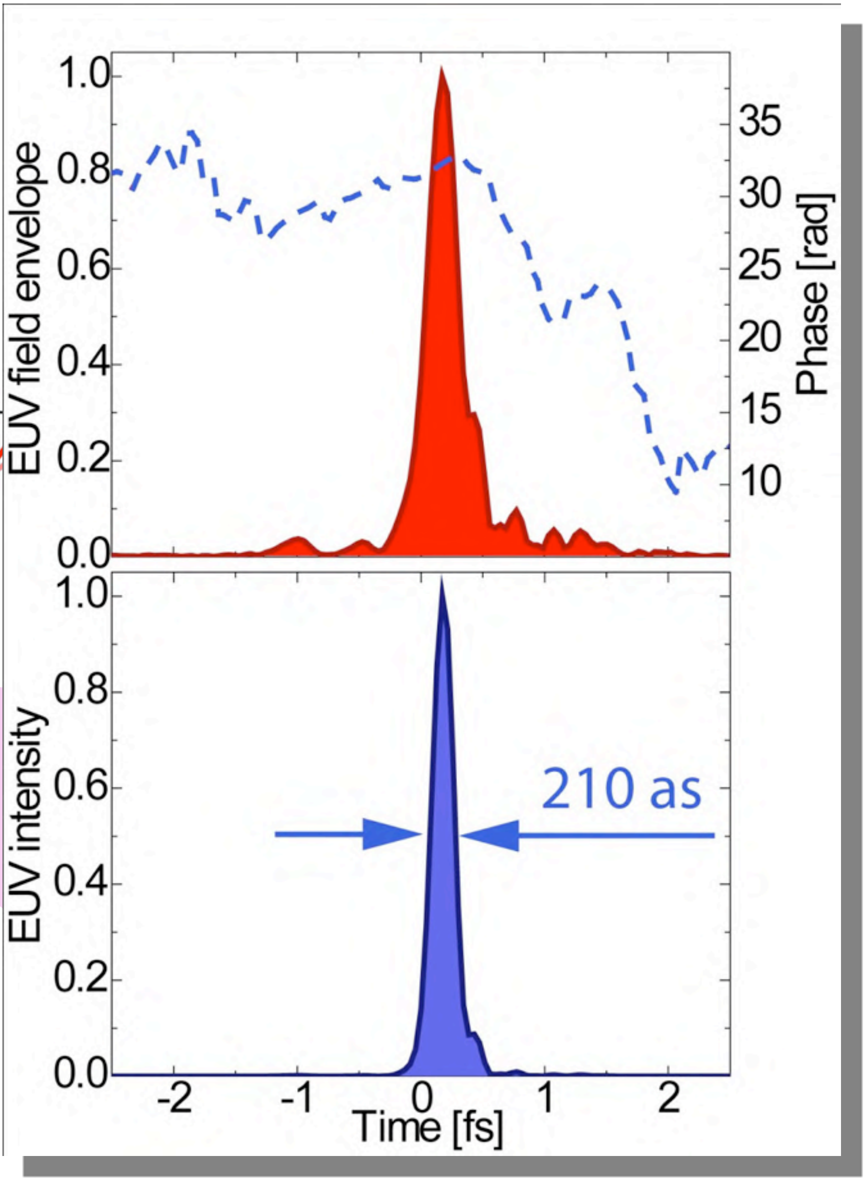
Spatially coherent beams





200 attosecond pulse generation at 50eV

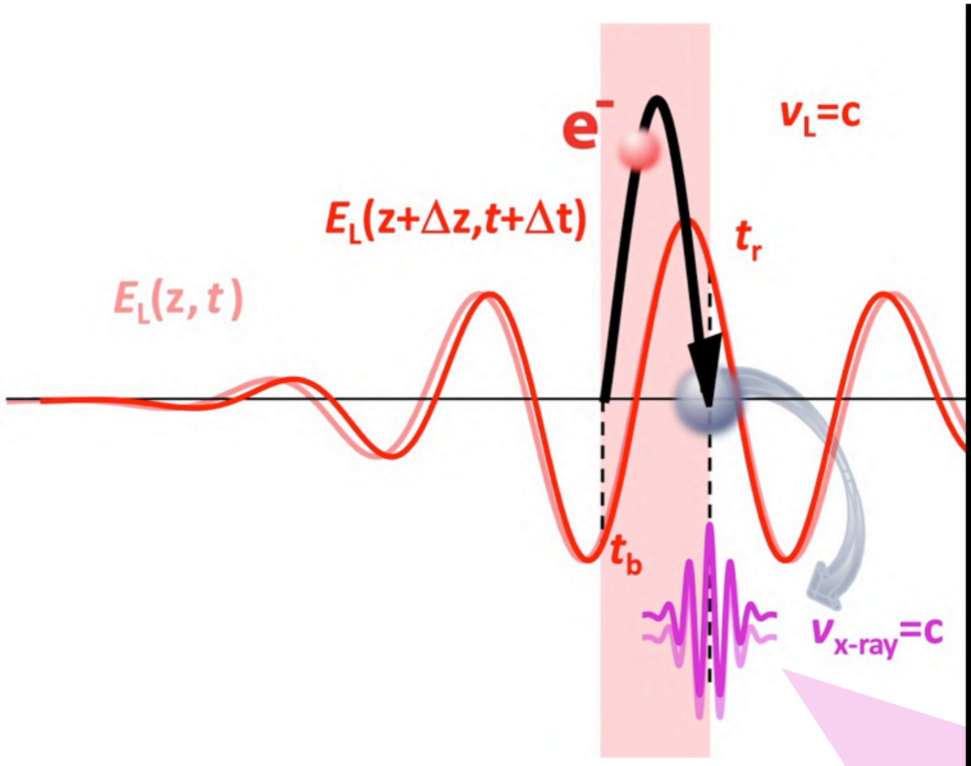


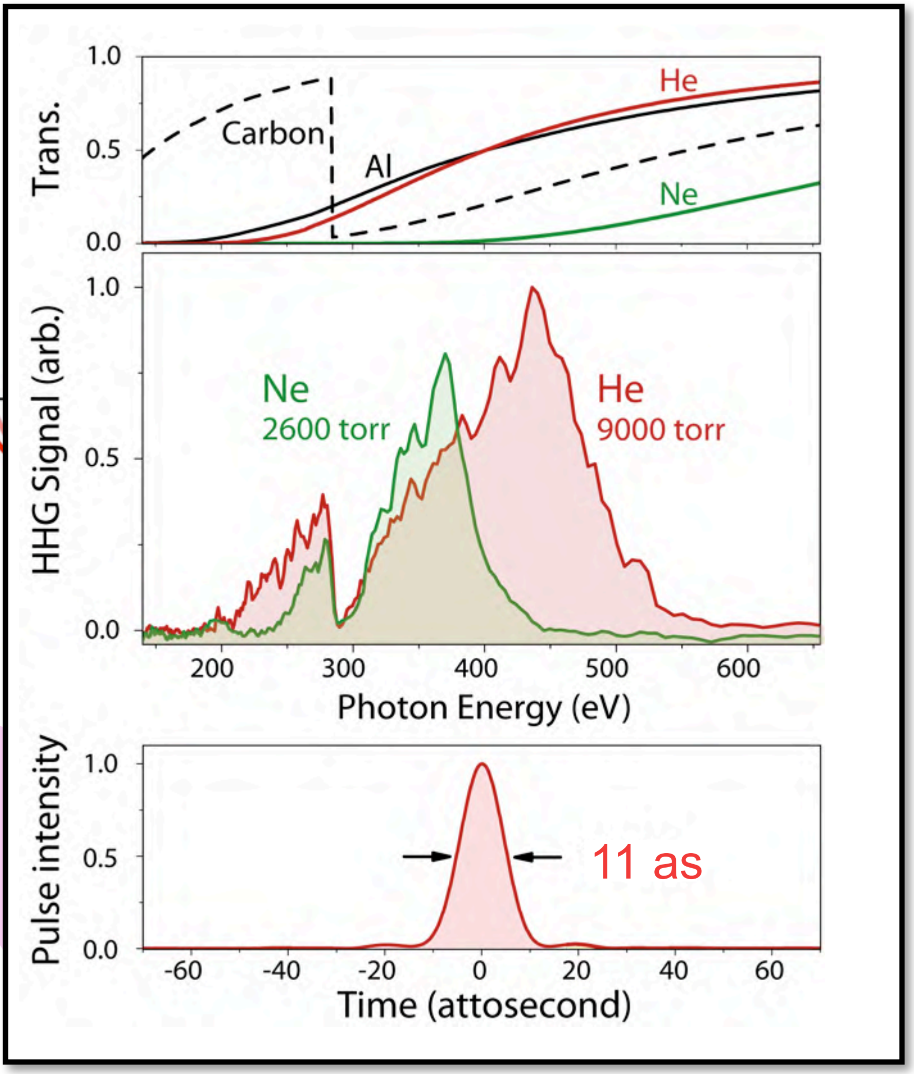


Thomann et al., Optics Express **17**, 4611 (2009)

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

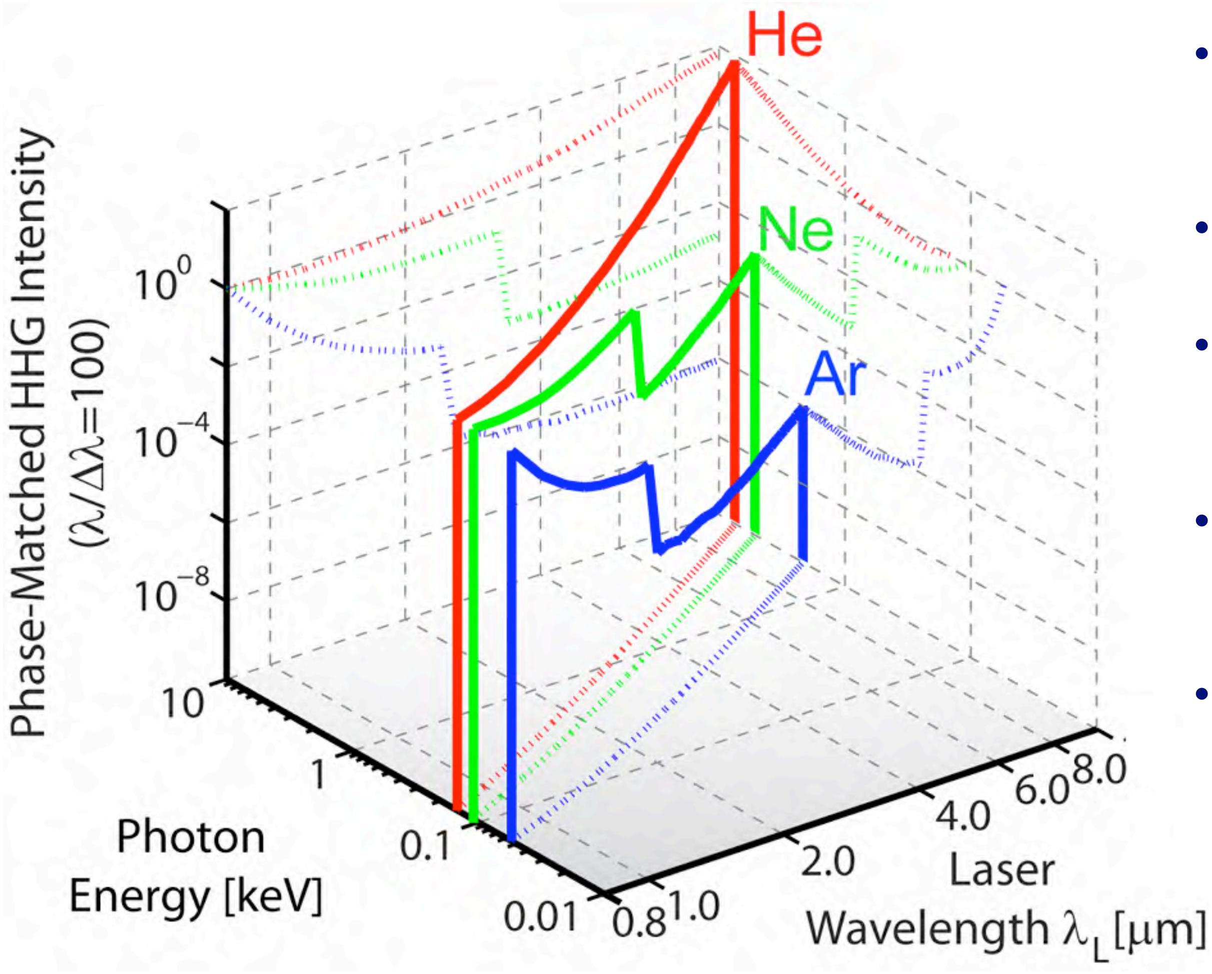
10 attosecond pulse generation at 400eV





- 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

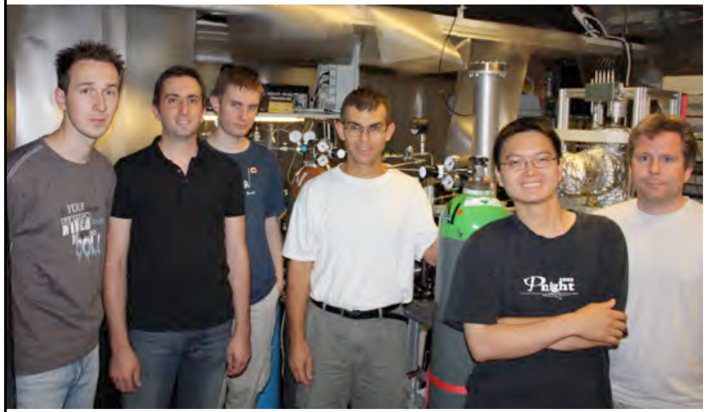


- Very favorable scaling to multi-keV region!!
- Low gas absorption of HHG
- Large pressure-length products mitigate the low χ_{eff}
- Low nonlinear distortion of laser pulse due to ionization
- At laser wavelengths > 3 μm, group velocity mismatch and magnetic field effects may reduce the HHG flux

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

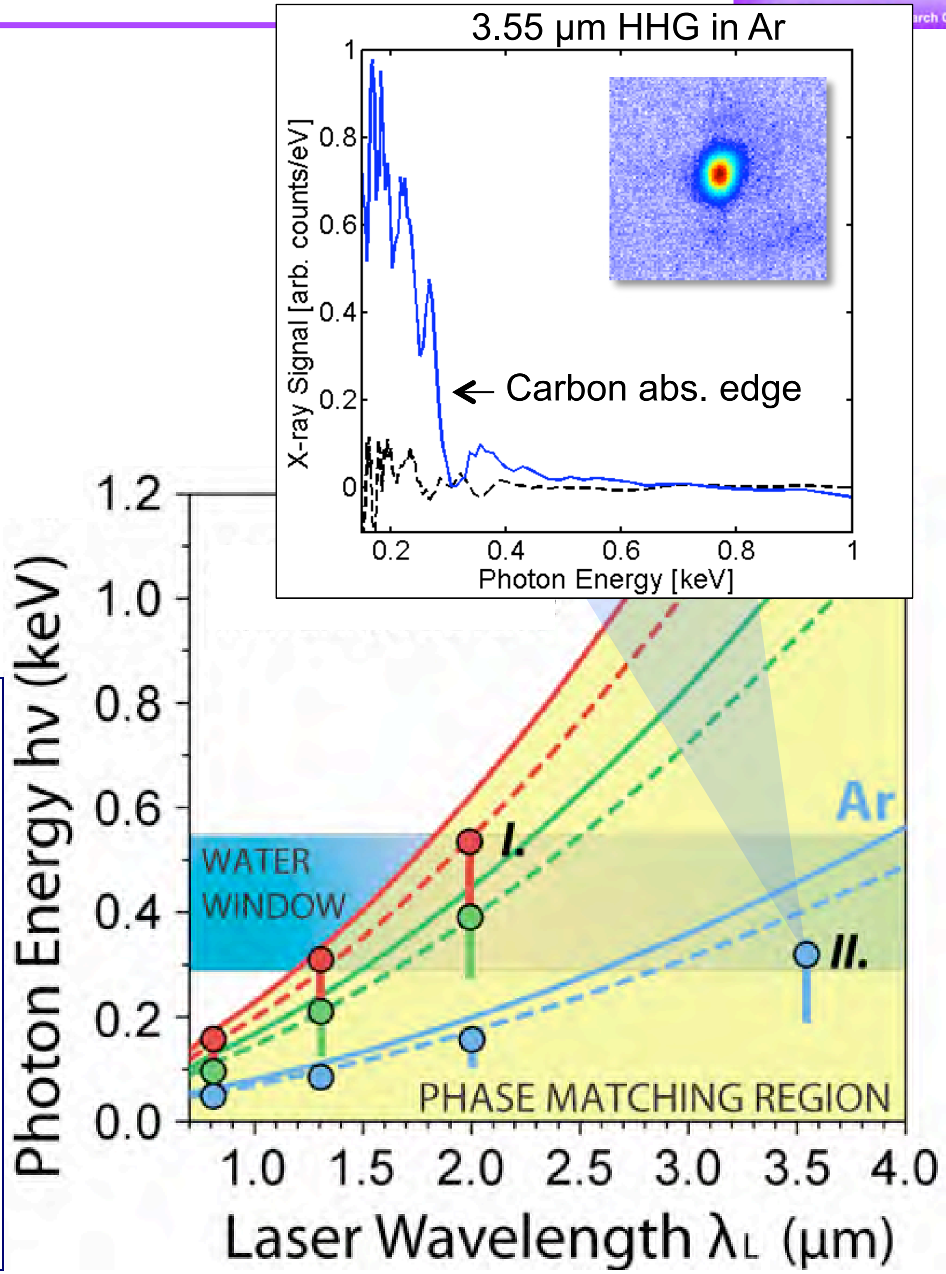
PNAS ***106****, 10516 (2009)*
Nature Photonics to be pub (2010)

First high harmonics driven by 3.55 µm light

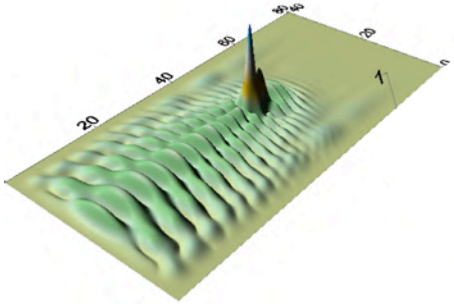


Andrius Baltuška
Audrius Pugzlys
Giedrius Andriukaitis
Tadas Balciunas
Oliver D. Mücke

- Observed first UV/EUV/SXR high-order harmonics driven by 3 µm lasers
- Magnetic field effects do not reduce HHG yield
- Route to zeptosecond keV harmonics using mid-IR driving lasers at 3.55 µm!



Science at the timescale of the electron – so what?

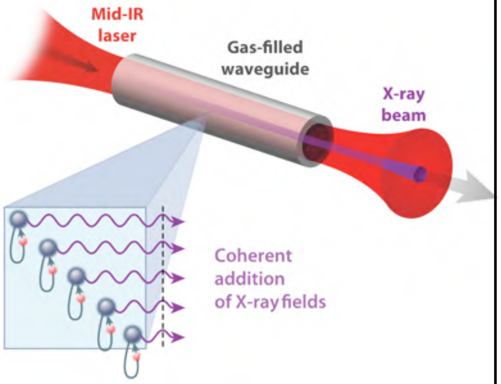


I. Bright coherent x-ray beams on a tabletop

—Ultrafast lasers can manipulate electrons on their fundamental timescale to implement a coherent version of the x-ray tube

—Bright coherent tabletop beams at > 0.5 keV

—Duration of 10 attoseconds – soon zeptoseconds!



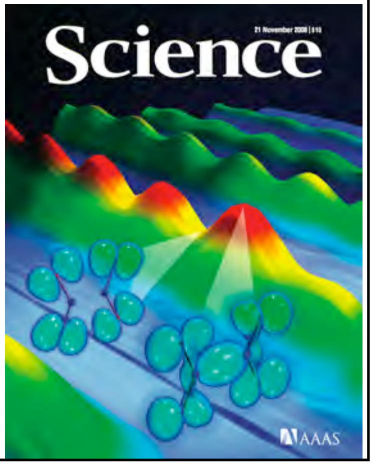
II. Ultrafast x-rays are an ideal probe of the nanoworld

—Image reactions at the level of electrons

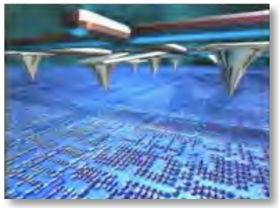
—Understand energy/charge transport at the nanoscale

—Capture correlated electron dynamics (spin, molecules, materials)

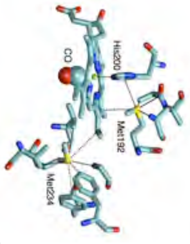
—Elemental and chemical nanoprobe of thick samples



Applications of ultrafast coherent x-rays



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



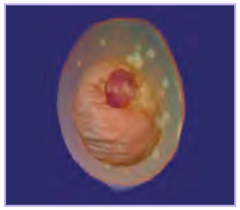
EXAFS/PEEM

Image reactions at the level of atoms and electrons

Understand and control function

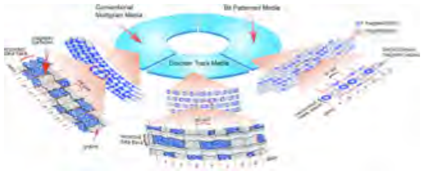
METROLOGY FOR EUV LITHOGRAPHY

BIO-IMAGING

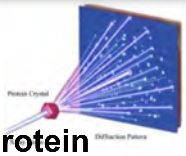


Cellular and materials tomography (10nm, elemental)

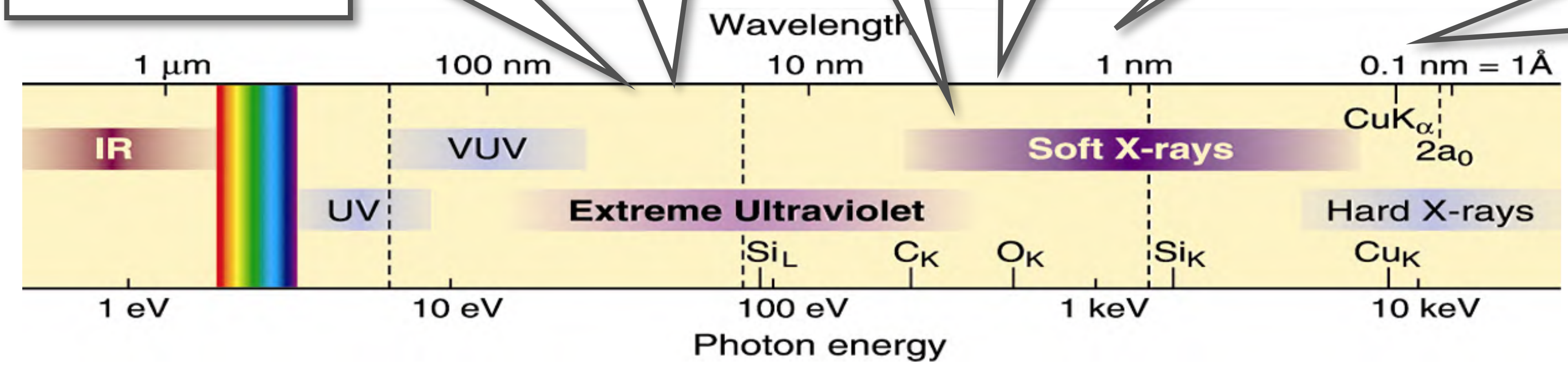
NANOSCIENCE (< 10nm)



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



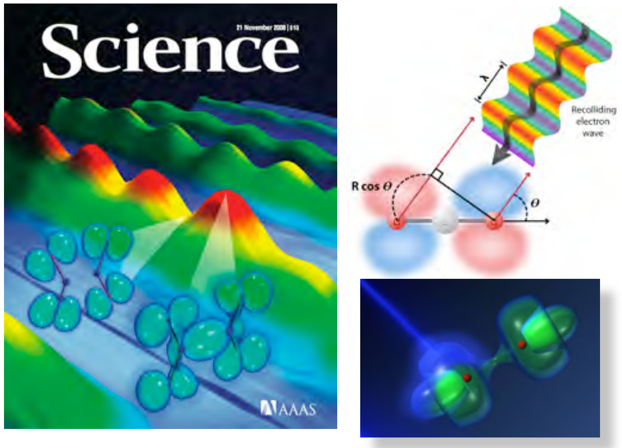
Protein crystallography



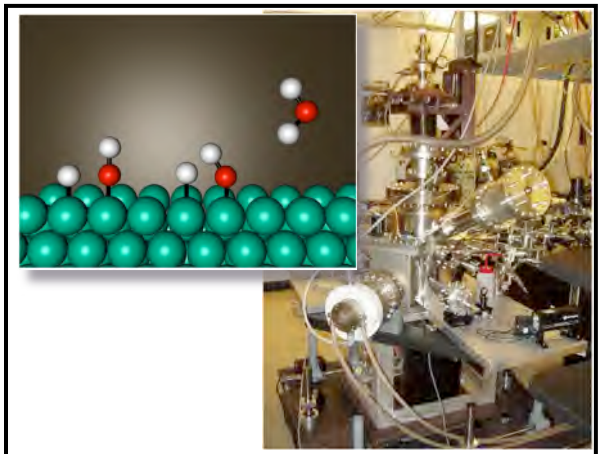
Soft x-rays are ideal probes of nanoworld:
- Penetrate thick objects and image small features
- Elemental and chemical specificity if HHG can extend to x-ray absorption edges
- Applications to date limited to ≈ 100 eV

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

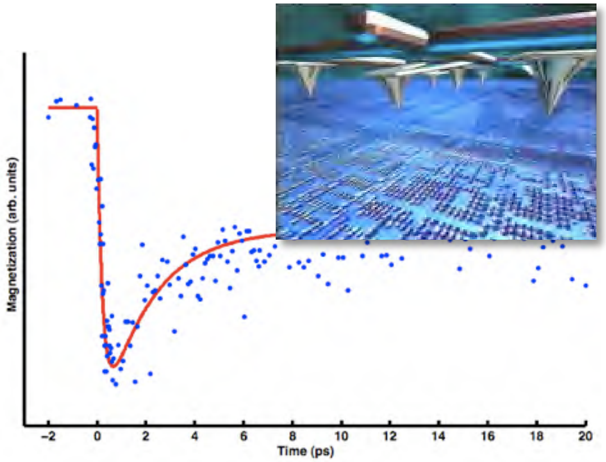
An NSF Engineering Research Center



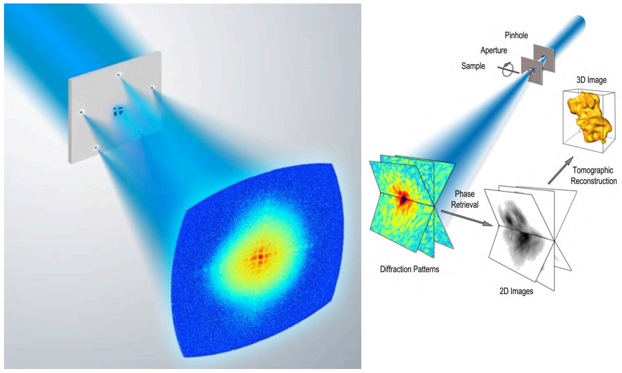
Molecular imaging: image changing electronic orbital and molecular structure



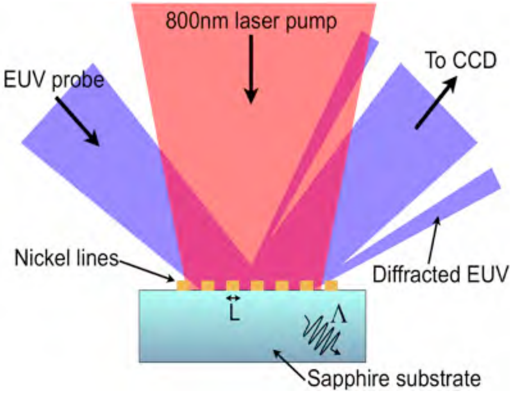
Surface science: probe electronic dynamics on catalysts, photovoltaics



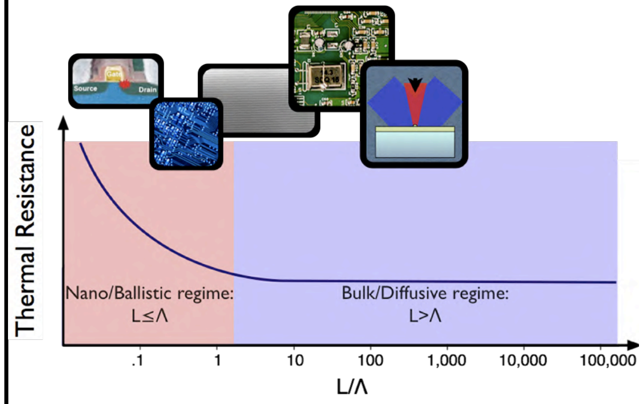
Magnetics: Probe nanodomains, magnetic dynamics



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

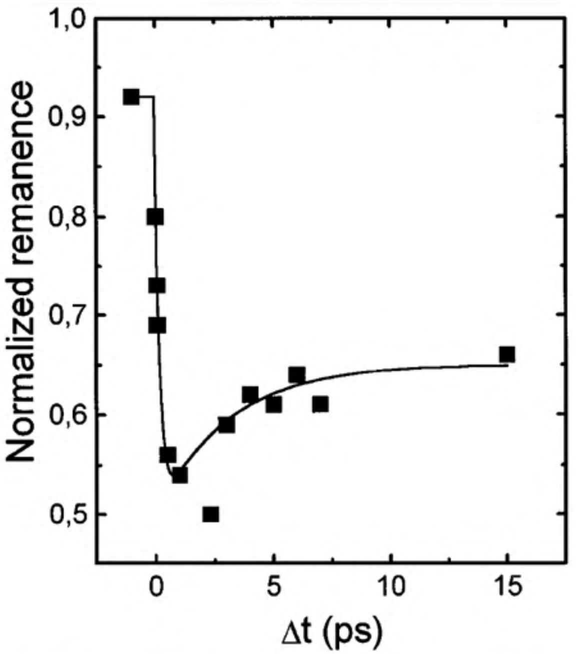


High frequency acoustic metrology: Characterize thin films, interfaces, adhesion



Nanothermal transport: probe heat flow in nanostructures

Ultrafast coherent magnetism – faster, denser, more efficient?



Beaurepaire et. al PRL ***76****, 4250 (1996)*

PRL 99, 047601 (2007) PHYSICAL REVIEW LETTERS week ending 27 JULY 2007

All-Optical Magnetic Recording with Circularly Polarized Light

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Institute for Molecules and Materials, Radboud University Nijmegen, Toernooiveld 1, 6525 ED Nijmegen, The Netherlands
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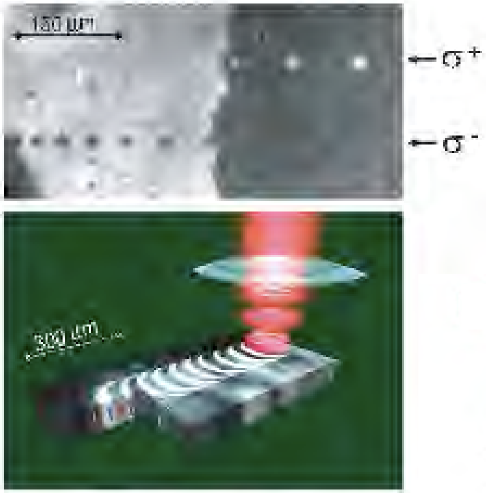
(Received 2 March 2007; published 25 July 2007)



nature physics
ARTICLES
PUBLISHED ONLINE: 31 MAY 2009; CORRECTED ONLINE: 16 JUNE 2009 | DOI: 10.1038/NPHYS1285

Coherent ultrafast magnetism induced by femtosecond laser pulses

Jean-Yves Bigot*, Mircea Vomir and Eric Beaurepaire

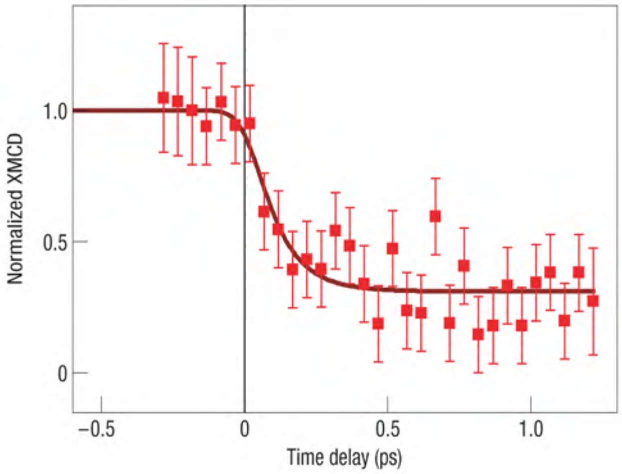
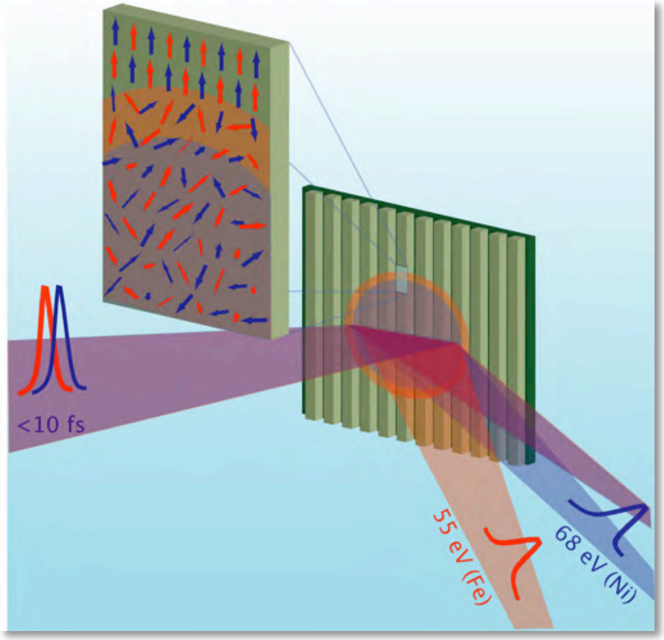
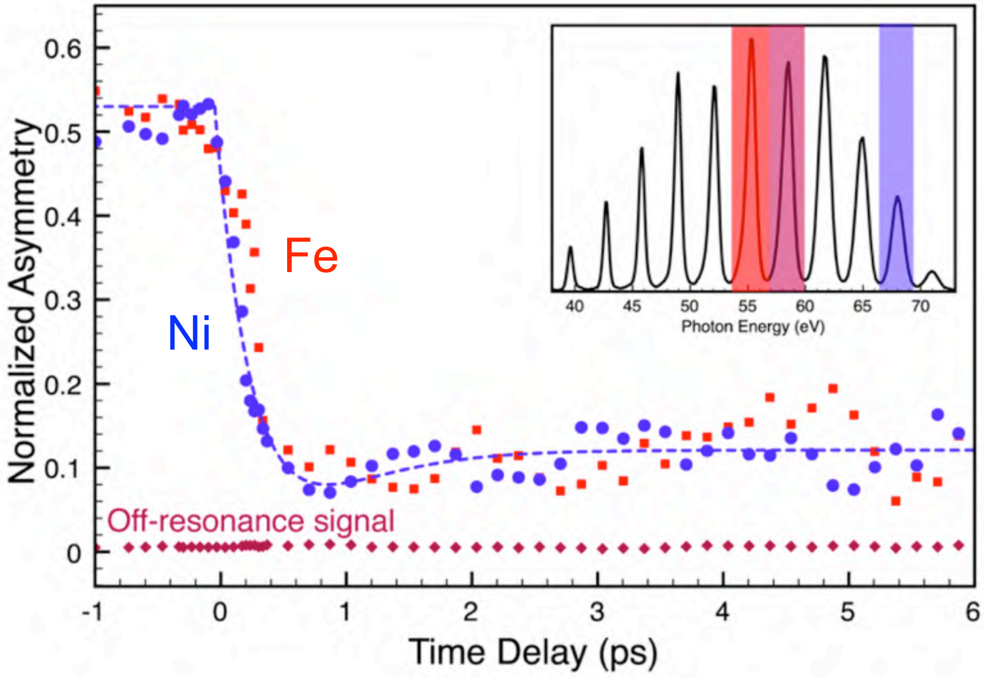


Figure 3 Femtosecond evolution of Ni electronic and magnetic structure.

Stamm, et al., Nature Mat. ***6****, 740 (2007)*

Ultrafast element-selective demagnetization dynamics

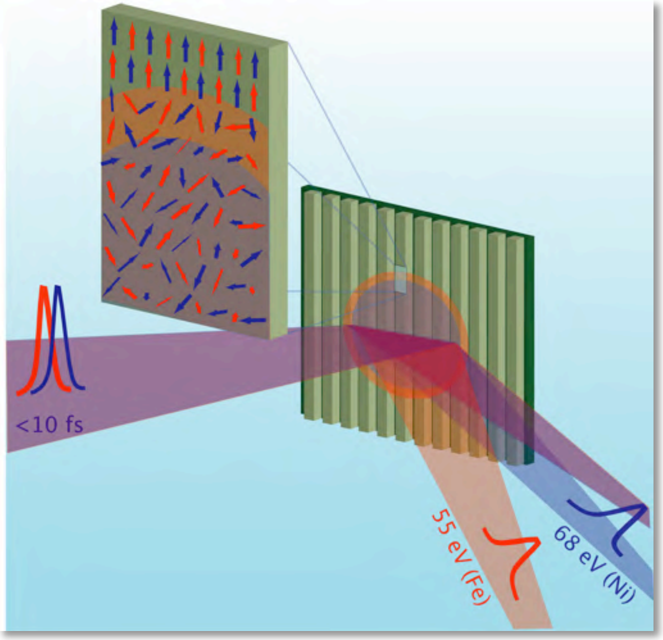


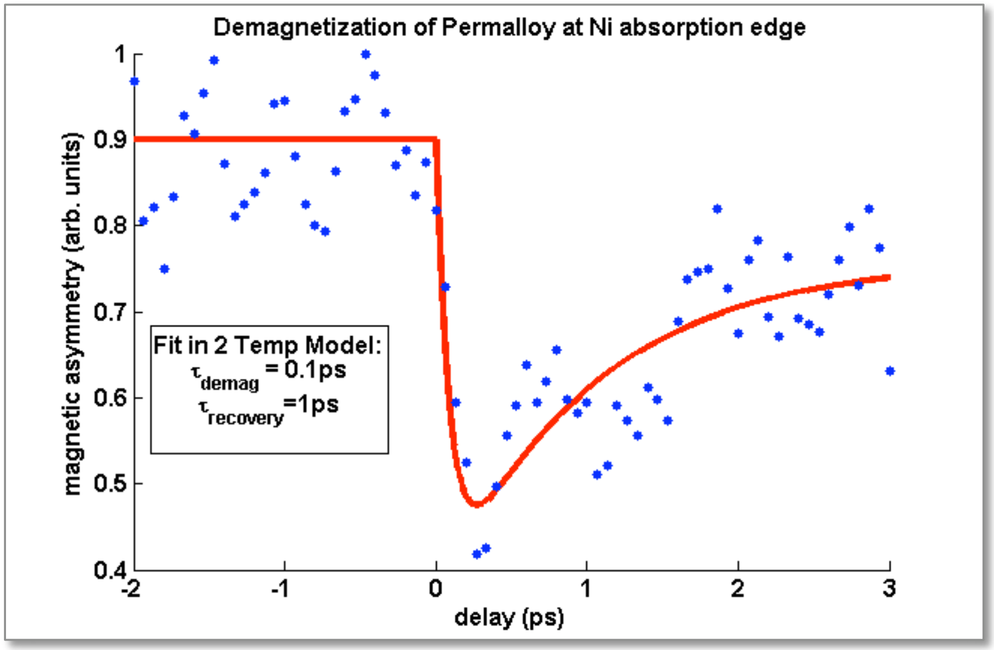


- Demagnetize permalloy using IR laser, probe using M-edge harmonics
- Highest time resolution ≈ 55fs and elemental specific measurement
- First result: Fe and Ni in Permalloy both decay within ≈ 400 fs since they are strongly exchanged coupled even during non-adiabatic heating
- Next steps – attosecond, domain imaging, L- edges etc.
- Collaboration with NIST, Kaiserslautern, Julich

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

Ultrafast element-selective demagnetization dynamics





- Demagnetize permalloy using IR laser, probe using M-edge harmonics
- Highest time resolution ≈ 55fs and elemental specific measurement
- Second result: Fe and Ni in Permalloy both decay within ≈ 100 fs at lower sample temperatures
- Next steps – attosecond, domain imaging, L- edges etc.

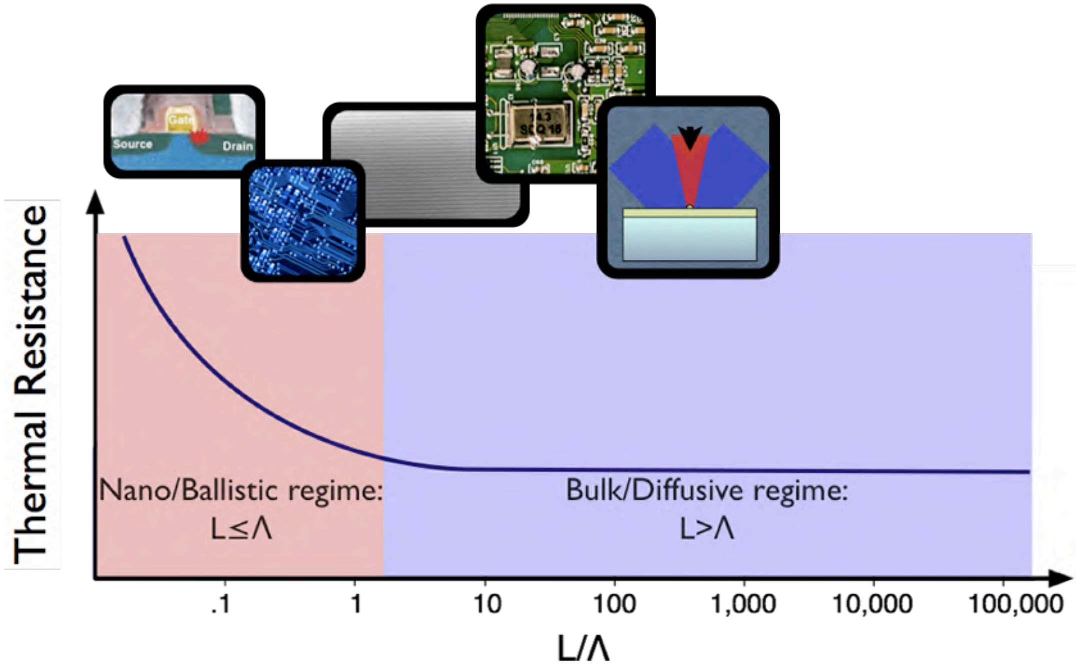
Phys. Rev. Lett. ***103****, 257402 (Dec 2009)*

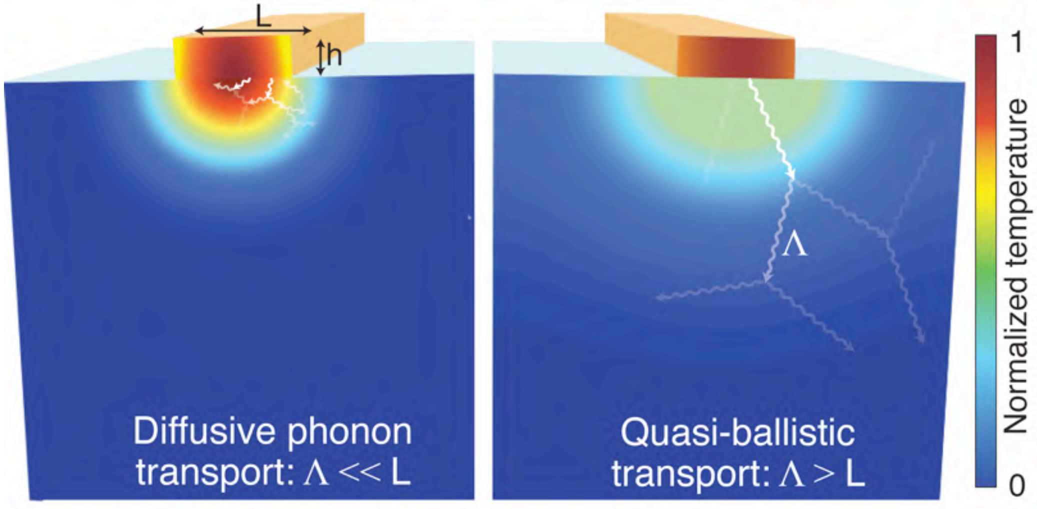
Understanding nanoscale heat flow

- 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))



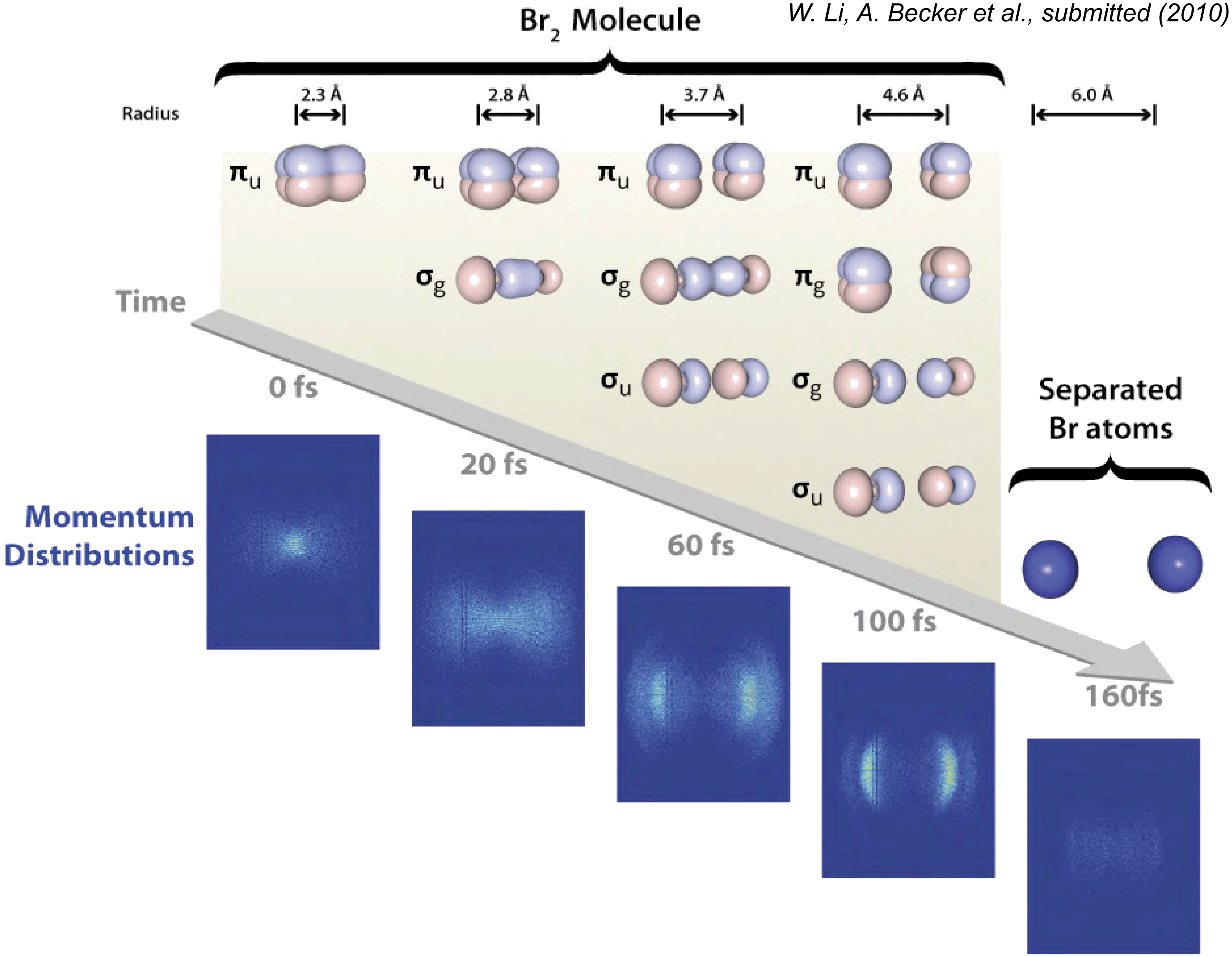


Following entire valence shell electron density rearrange as a bond breaks

EUV

An NSF Engineering Research Center

W. Li, A. Becker et al., submitted (2010)



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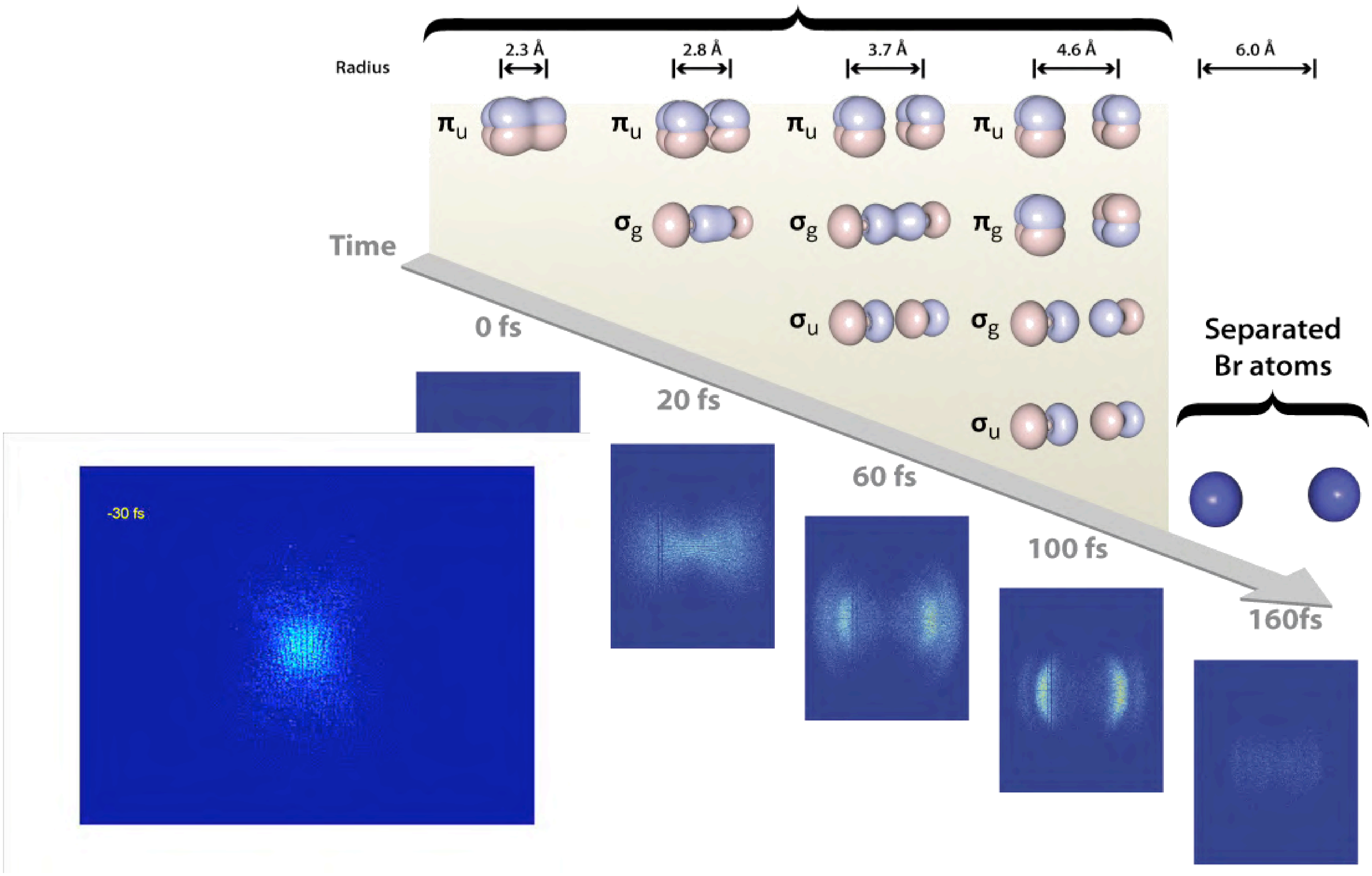
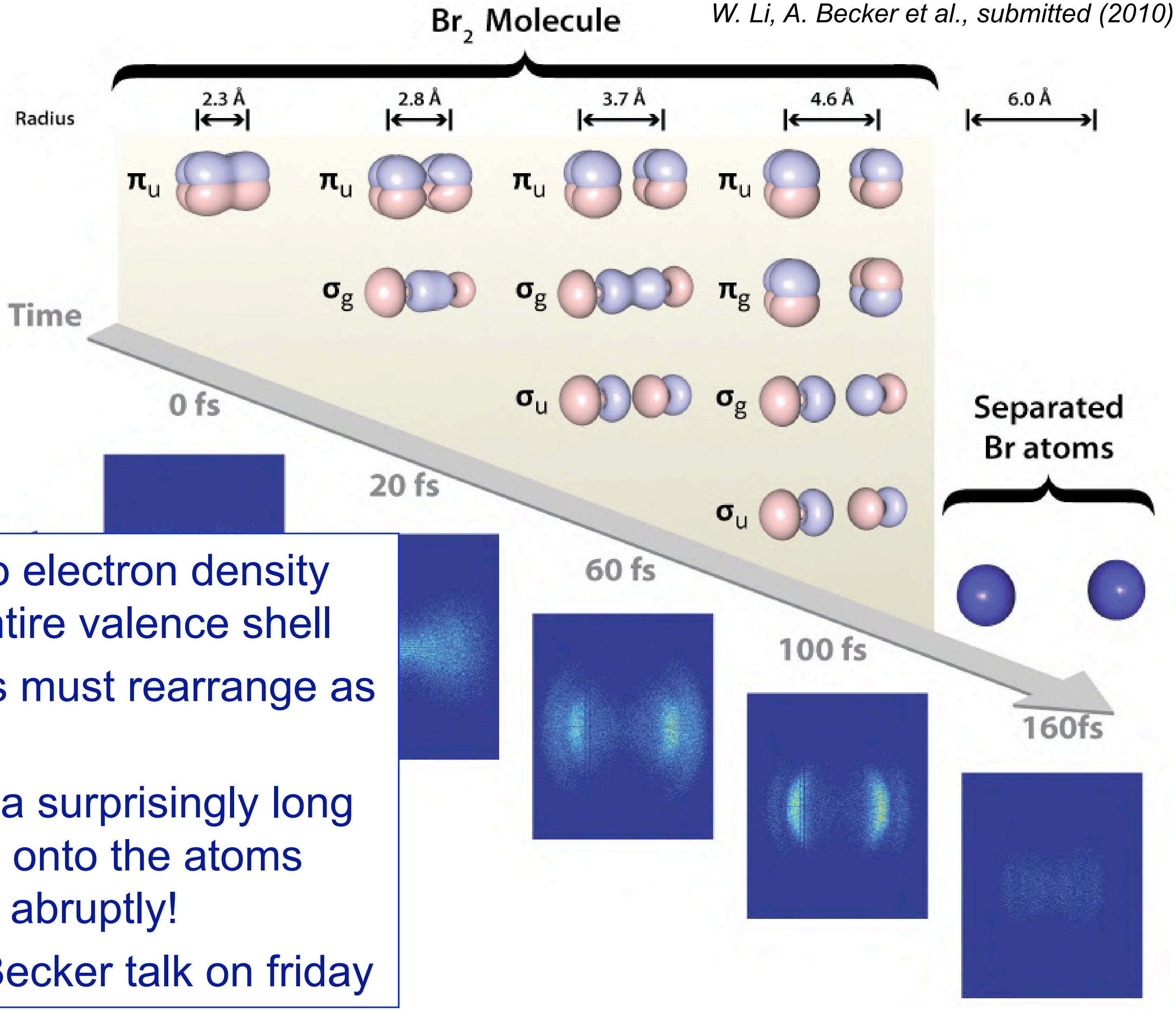


Following entire valence shell electron density rearrange as a bond breaks

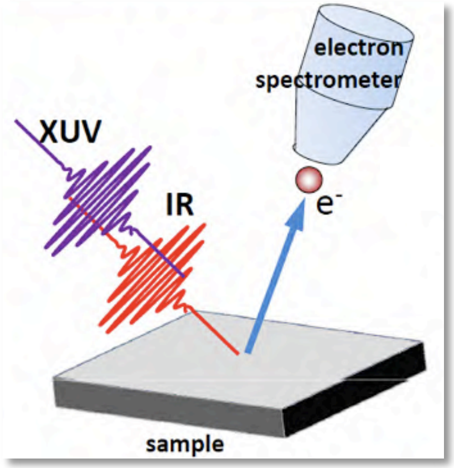
W. Li, A. Becker et al., submitted (2010)



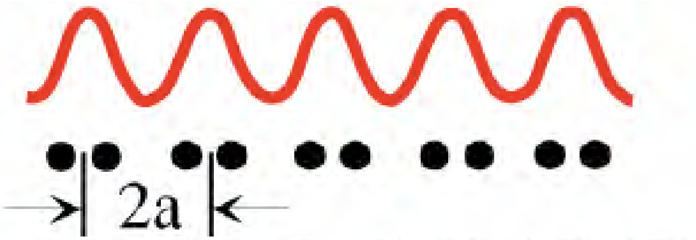
- SFI sensitive to electron density dynamics of entire valence shell
- Many electrons must rearrange as bond breaks!
- Electrons take a surprisingly long time to localize onto the atoms and it happens abruptly!
- See Andreas Becker talk on friday

How fast can we switch the electronic state of a solid?

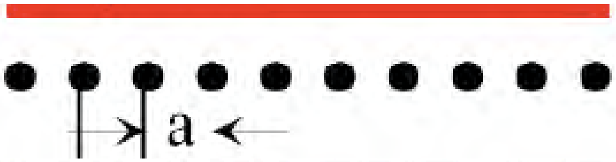
- $TiSe_2$ undergoes a photo-induced phase transition
- Probe entire band structure using angle-resolved HHG photoemission
- **Prof. Michael Bauer, University of Kiel**

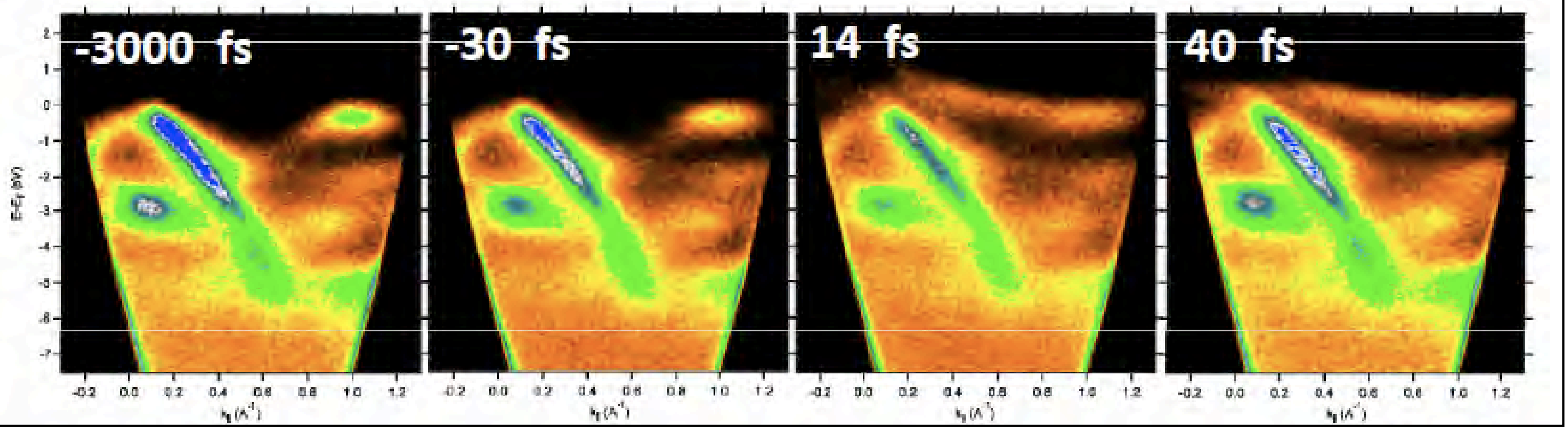


T < 200K: insulating charge density wave state



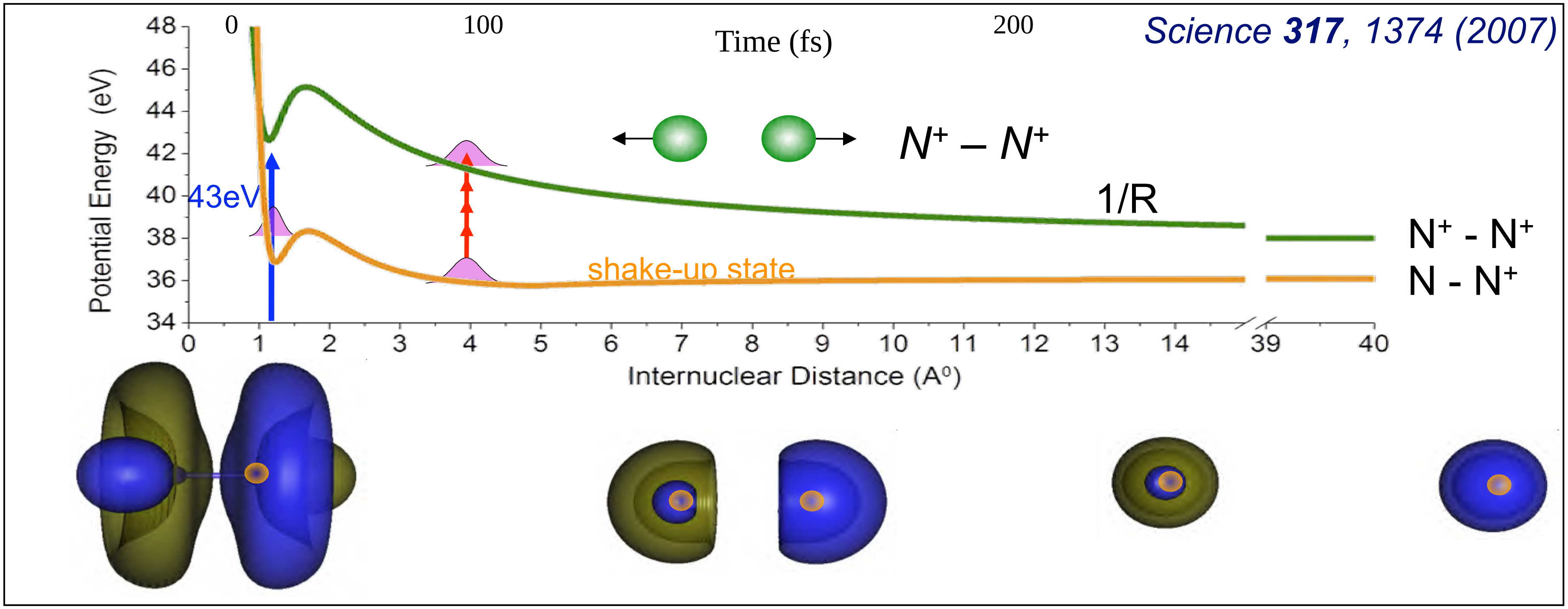
T < 200K: normal metallic phase





Radiation Femtochemistry

- Expt #1: Decay of N_2^+ excited by 43eV
  - Can identify the Rydberg dissociative states
  - Can follow dynamics as system changes from symmetric to 2-center
  - Theory by Xiao-Min Tong

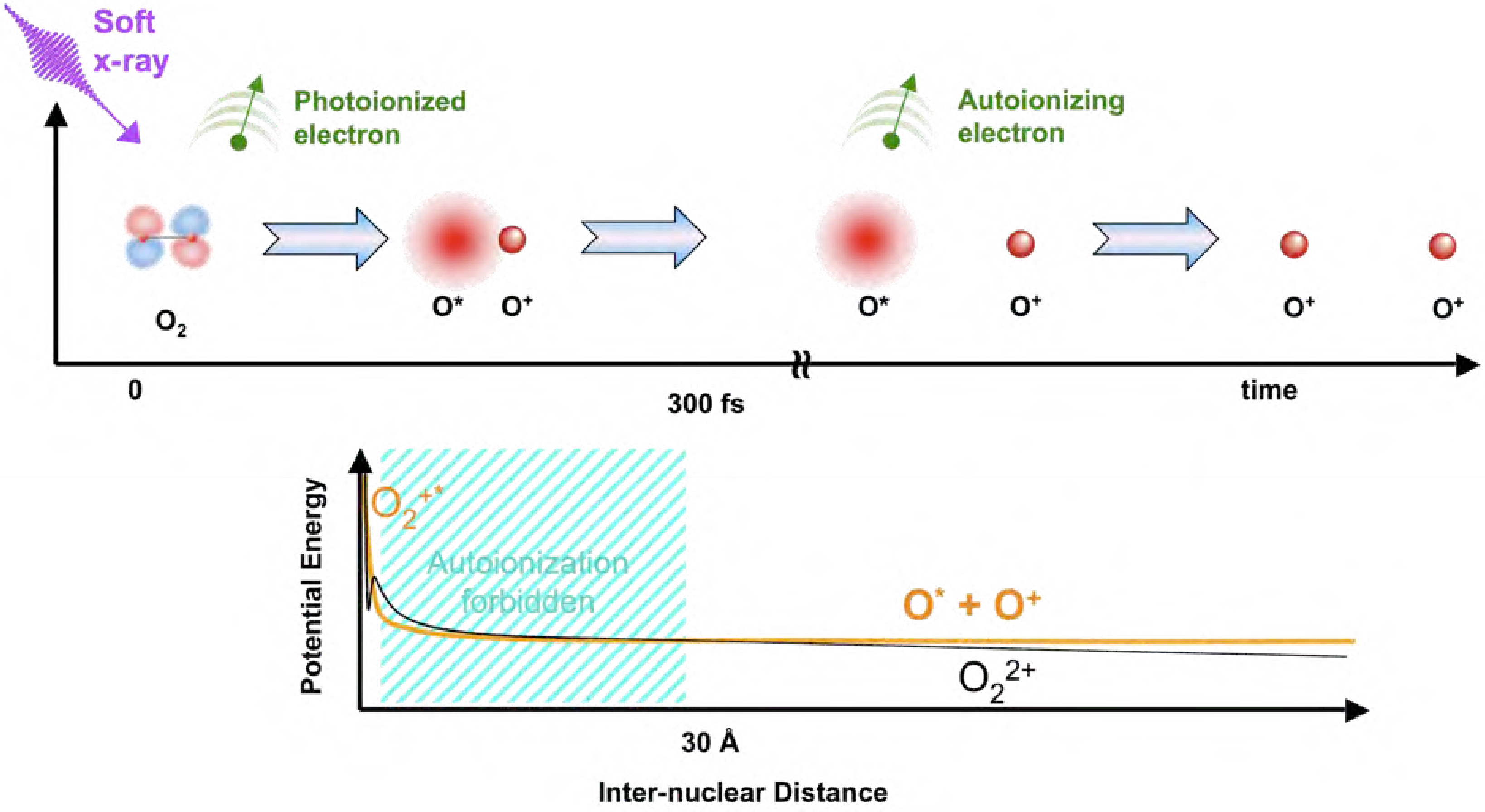


Science ***317***, *1374 (2007)*

R = 1.1 A^0 R = 3 A^0 R = 10 A^0

Radiation Femtochemistry

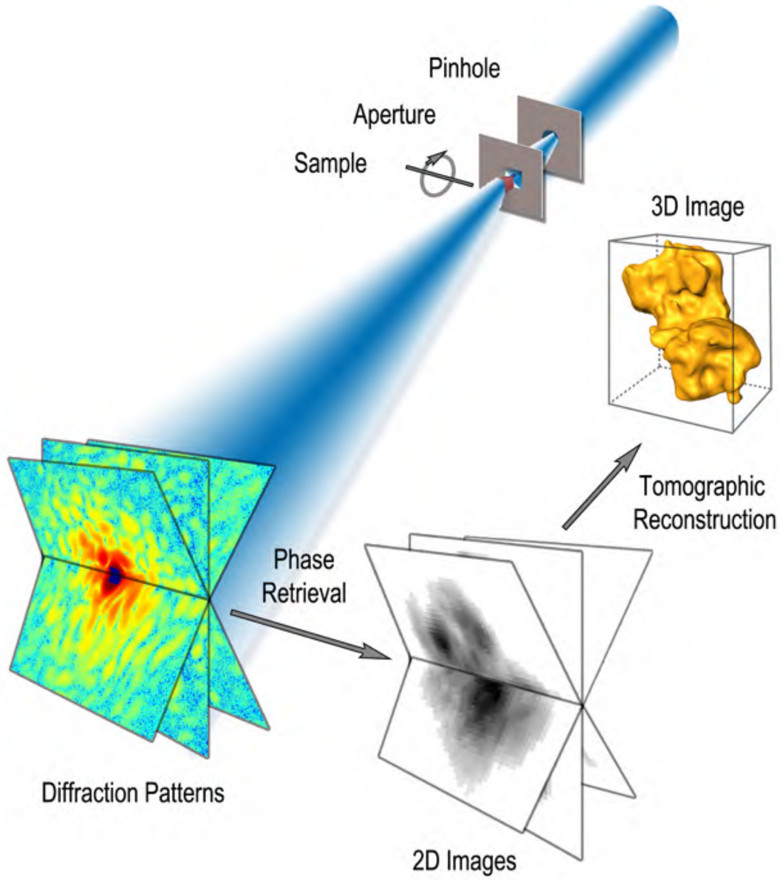
- Expt #2: Decay of O_2^+ excited by 43eV, theory by Robin Santra
  - Create long-lived superexcited states in O through Feschbach resonance
  - System still bound up to 30 Å

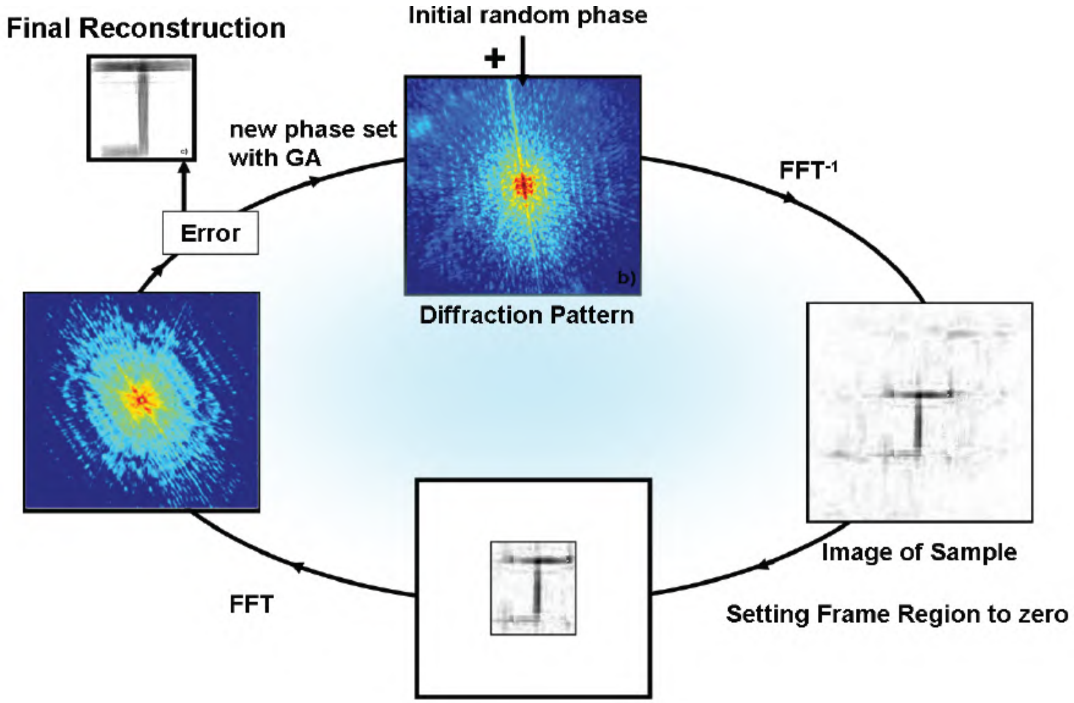


Sandhu et al., Science ***322****, 1081 (2008)*

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

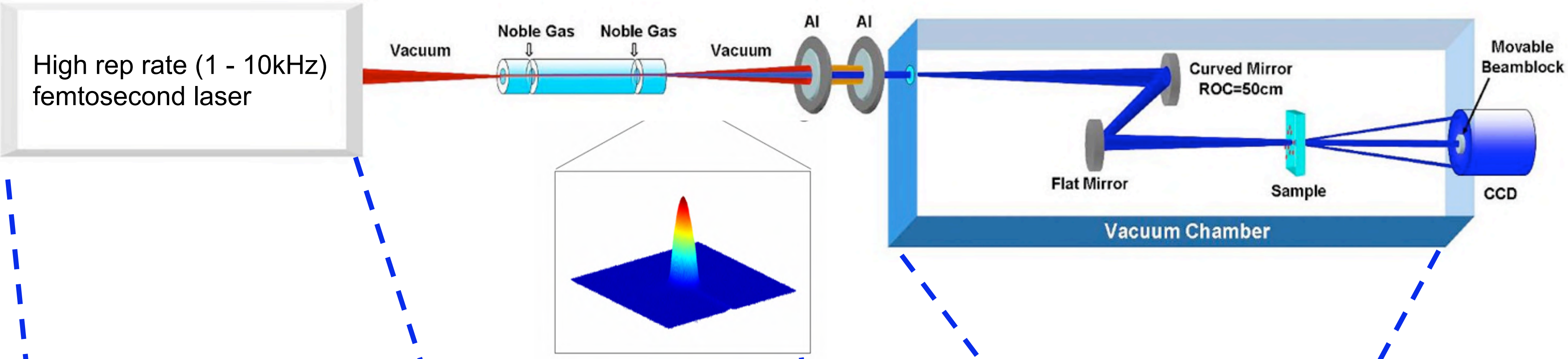




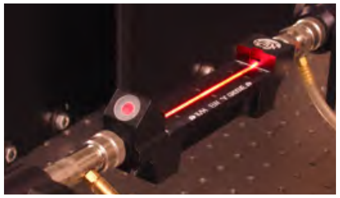
Sayre, Acta Cryst 5, 843 (1952)

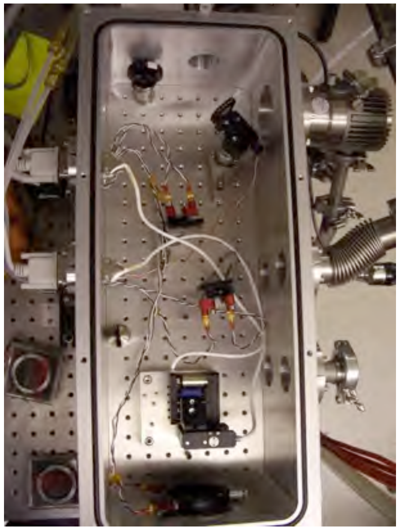
Miao et al., Nature 400, 342 (1999)

Lensless imaging using tabletop soft x-ray sources



X-ray beam

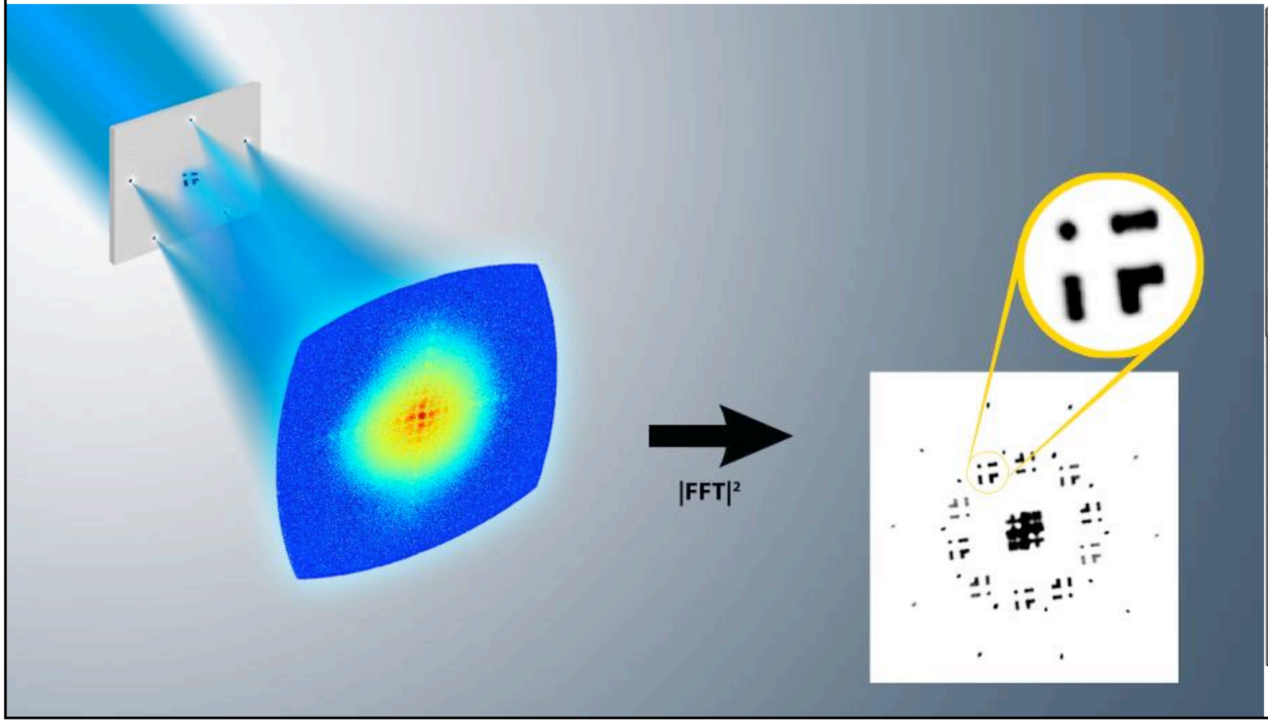


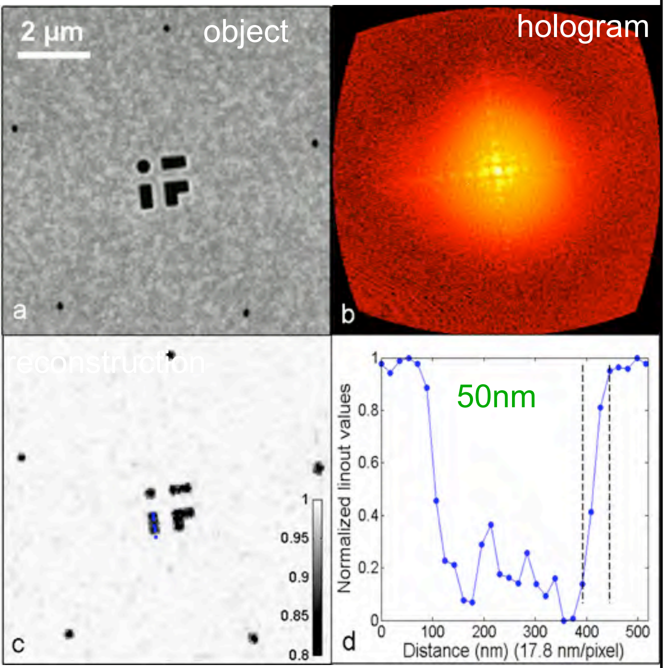


High average power fs laser system + High harmonic converter + Coherent diffractive microscope

Lensless coherent imaging –50nm resolution

- 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…….





Future

- Take attosecond electron rescattering physics, discovered just over 20 years ago, to generate coherent x-rays and electrons
- Now have coherent soft x-ray laser beams that span to 0.5 keV, with enough flux for expts., and with attosecond pulse duration, and with excellent prospects for hard x-ray laser beams on a tabletop
- Table-top microscopes, nanoprobes, nanomanipulation and x-ray imaging with unprecedented spatial and temporal resolution
- Thanks to NSF, DOE, DOD

Students and postdocs

STUDENTS *Paul Arpin, Susannah Brown, Tory Carr, Ming-Chang Chen, Michael Gerrity, Craig Hogle, Kathy Hoogeboom, Robynne Lock, Chan La-O-Vorakiat, Qing Li, Dimitar Popmintchev, Emrah Turgut, Matt Seaberg*
POSTDOCS *Xibin Zhou, Alon Bahabad, Predrag Ranitovic, Stefan Mathias, Tenio Popmintchev*