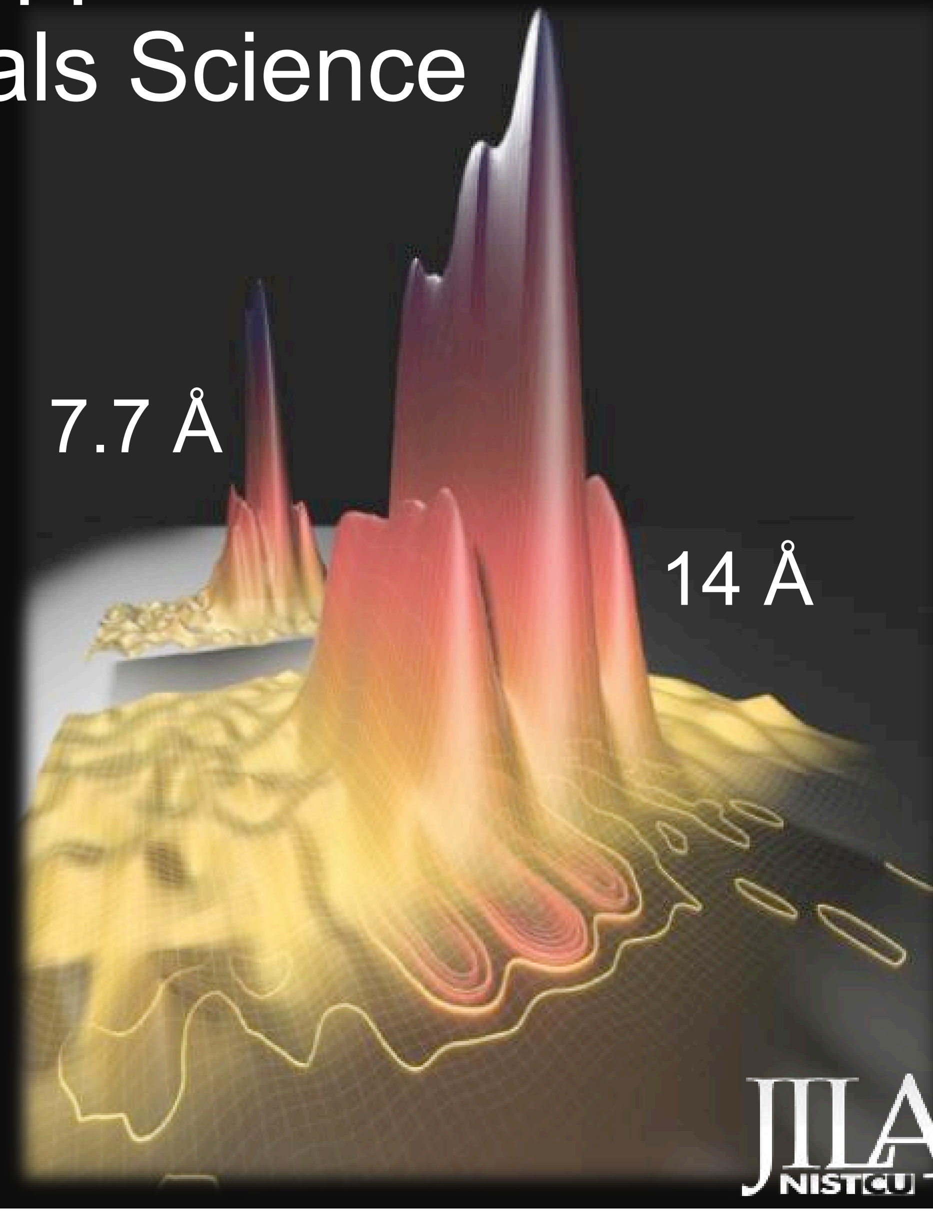
Bright Coherent Ultrafast X-Ray Beams on a Tabletop and Applications in Nano and Materials Science



Surprising new science

I. Nonlinear optics at the extreme

—Efficiently combine **>5000** mid-IR laser photons

—Bright **keV** x-rays from tabletop lasers

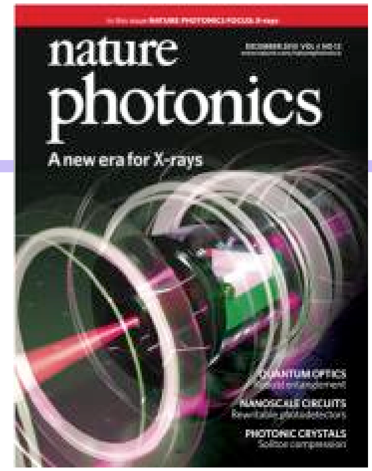
—Bright tabletop hard x-ray beams? Zeptosecond pulses?

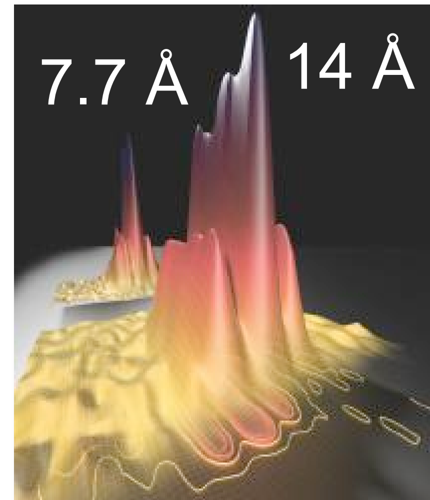
II. Probing the nanoworld at the space-time limits

—Capture coupled spin/charge/phonon/photon dynamics

—Imaging at the wavelength limit

—Applications in nano science, nanotechnology, energy, materials, bio science and engineering

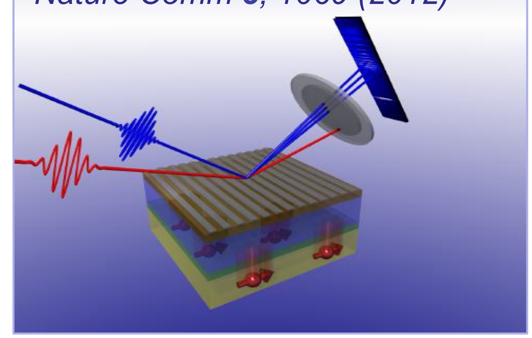




Science ***336***, *1287 (2012)*

Nature Comm ***3***, *1037 (2012)*

Nature Comm ***3***, *1069 (2012)*



Excellent students and collaborators

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Cornell

Tom Silva, Justin Shaw, Hans Nembach
NIST

Stefan Mathias, Martin Aeschlimann, Claus Schneider
Kaiserslautern and Julich

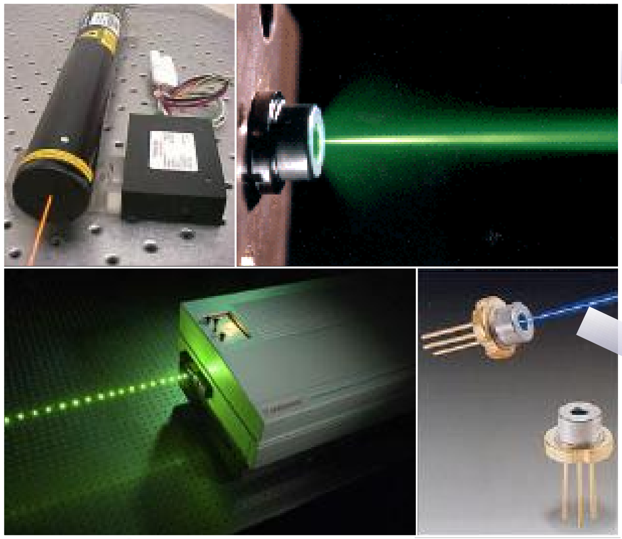
Michael Bauer
Kiel University

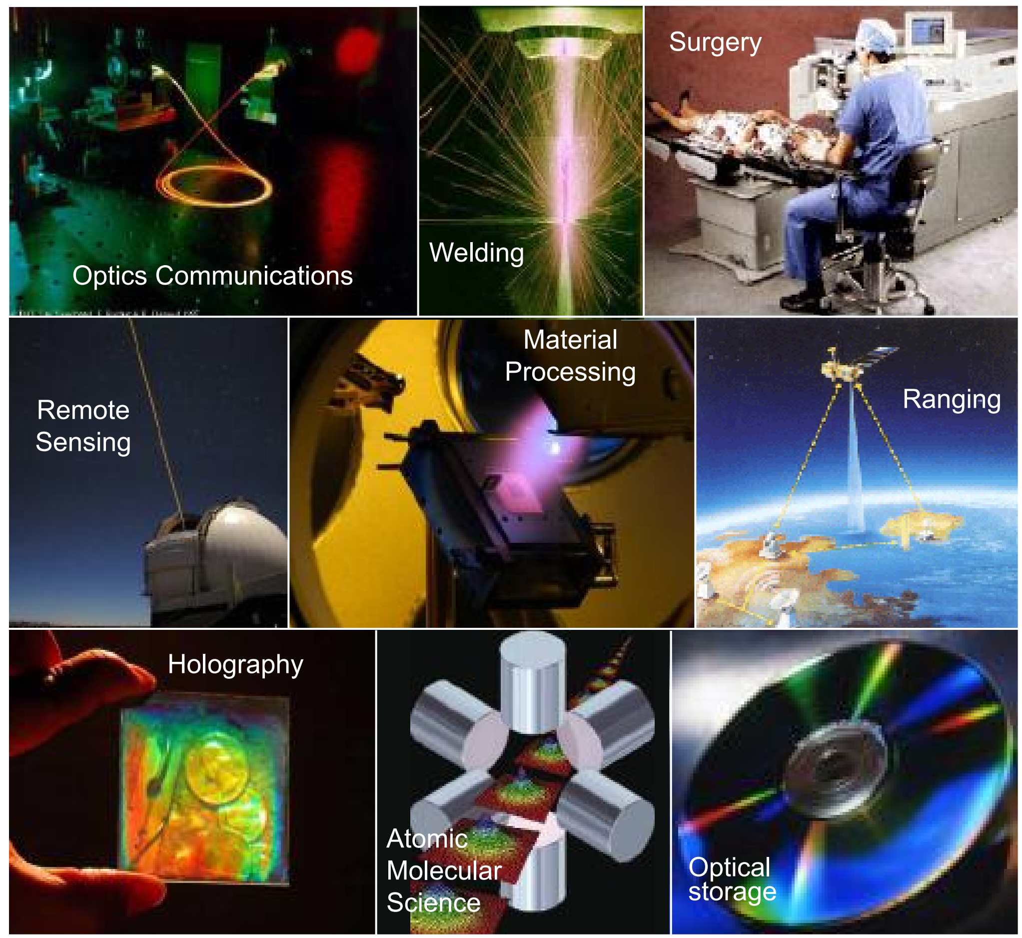
Keith Nelson
MIT

Tamar Seideman, Sai Ramakrishna
Northwestern

Xiao-Min Tong
Tsukuba University

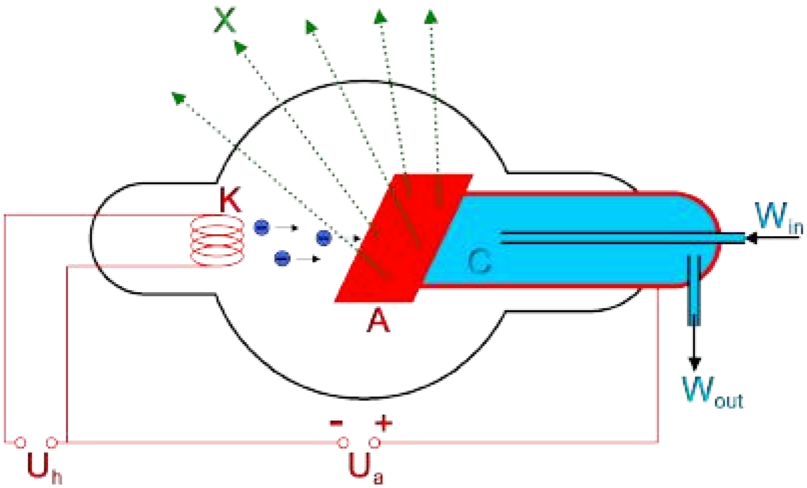
Visible laser light benefits society



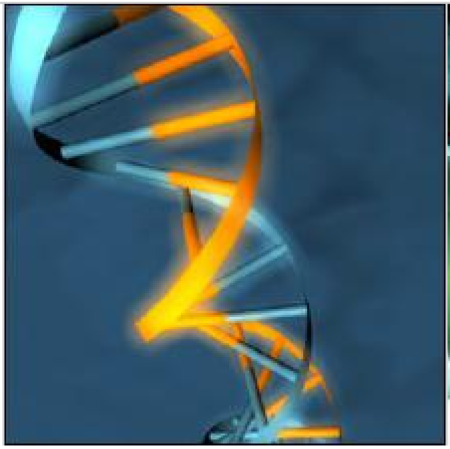


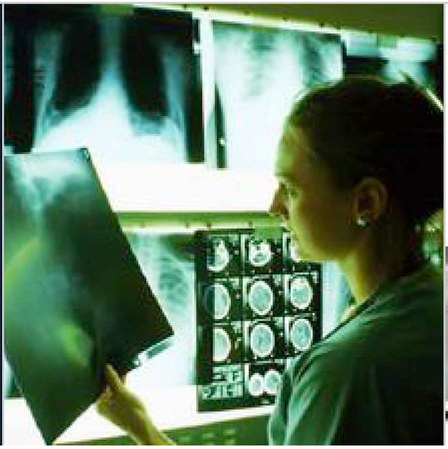
X-ray light also benefits society

Wilhelm Roentgen

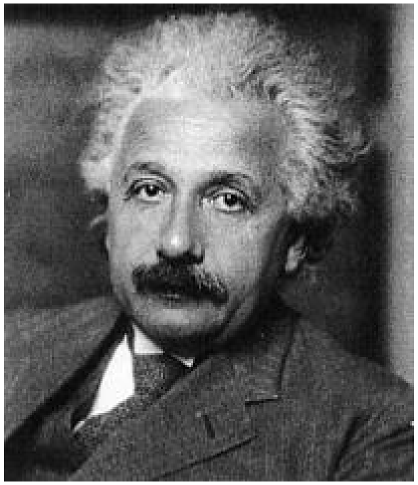


X-ray tube





X-ray lasers and free electron lasers

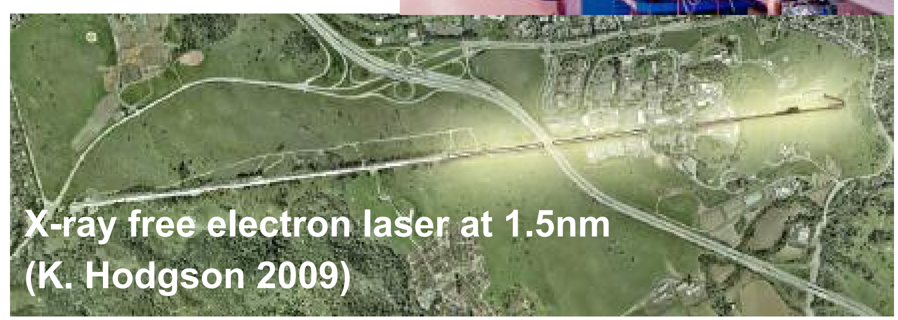


Spontaneous emission / Stimulated emission

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

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

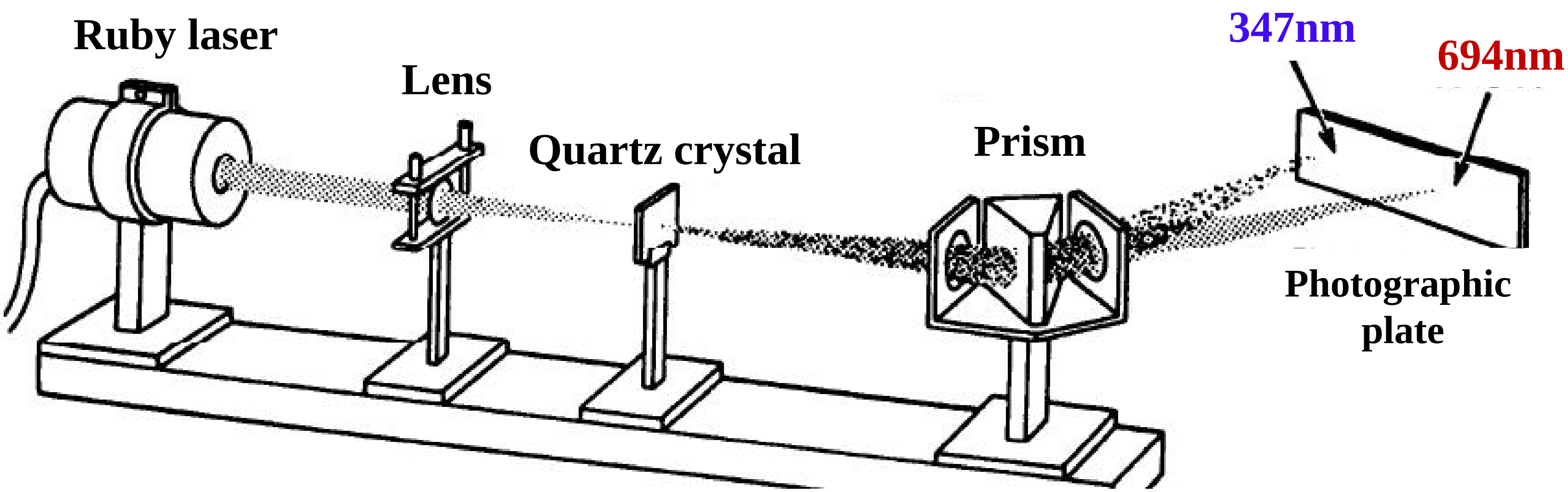
Soft x-ray laser at 20nm
(D. Matthews 1985)



X-ray free electron laser at 1.5nm
(K. Hodgson 2009)

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

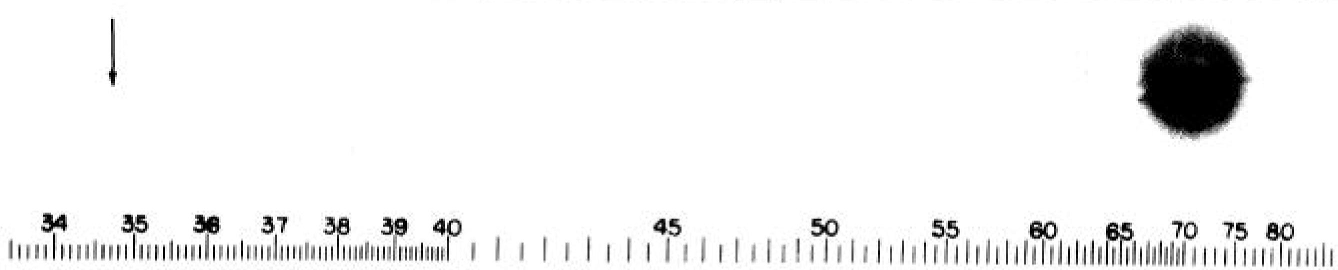
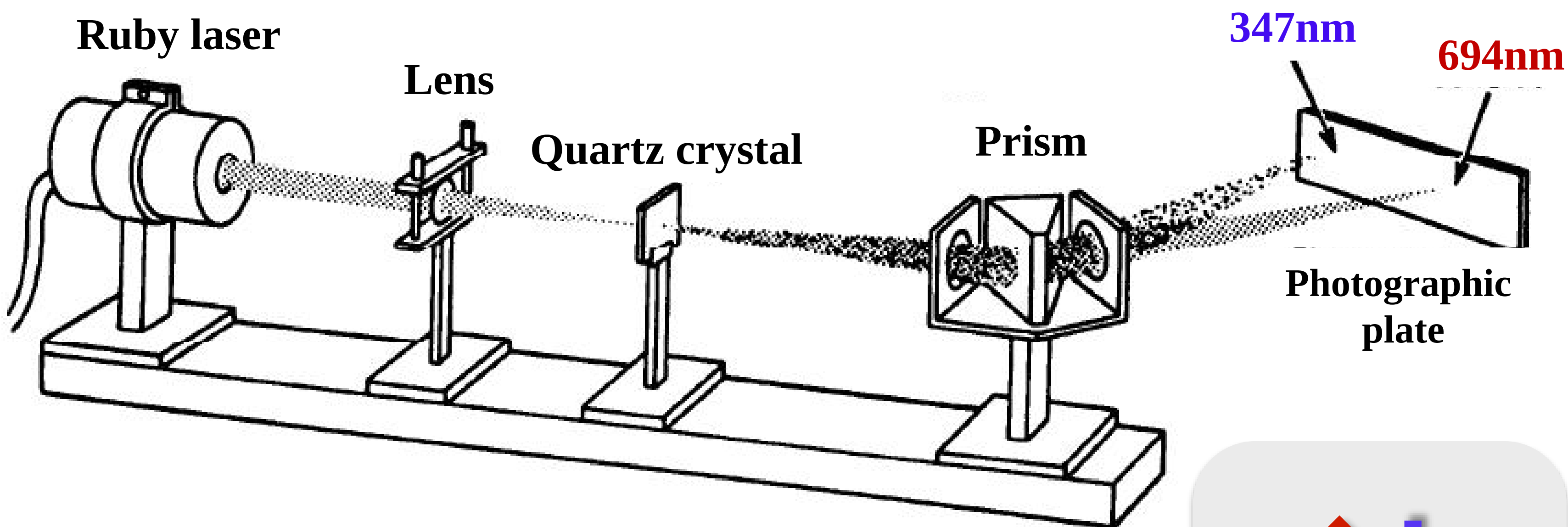
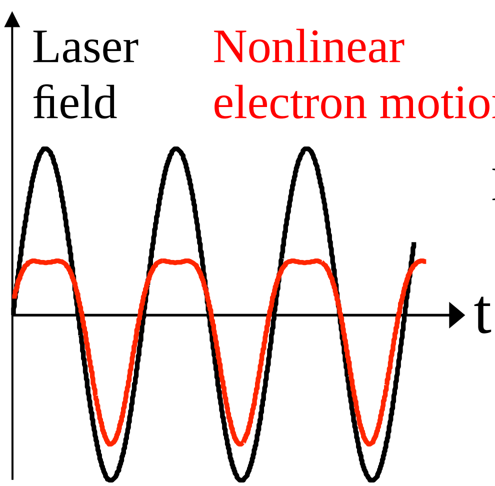


FIG. 1. A direct reproduction of the first plate in which there was an indication of second harmonic. The wavelength scale is in units of 100 A. The arrow at 3472 A indicates the small but dense image produced by the second harmonic. The image of the primary beam at 6943 A is very large due to halation.

The birth of Nonlinear Optics – second harmonic generation

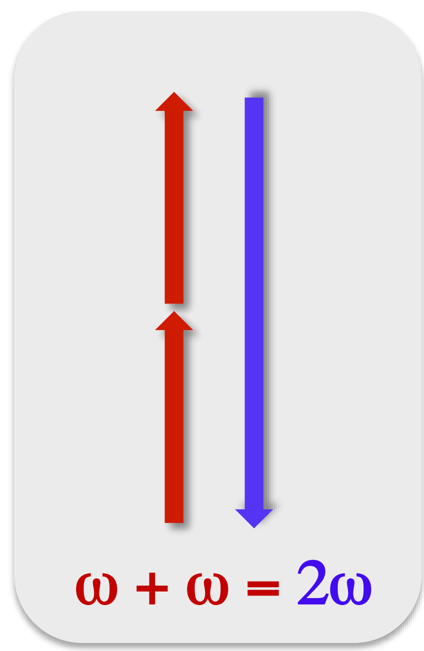
P.A. Franken et al, PRL 7, 118 (1961)



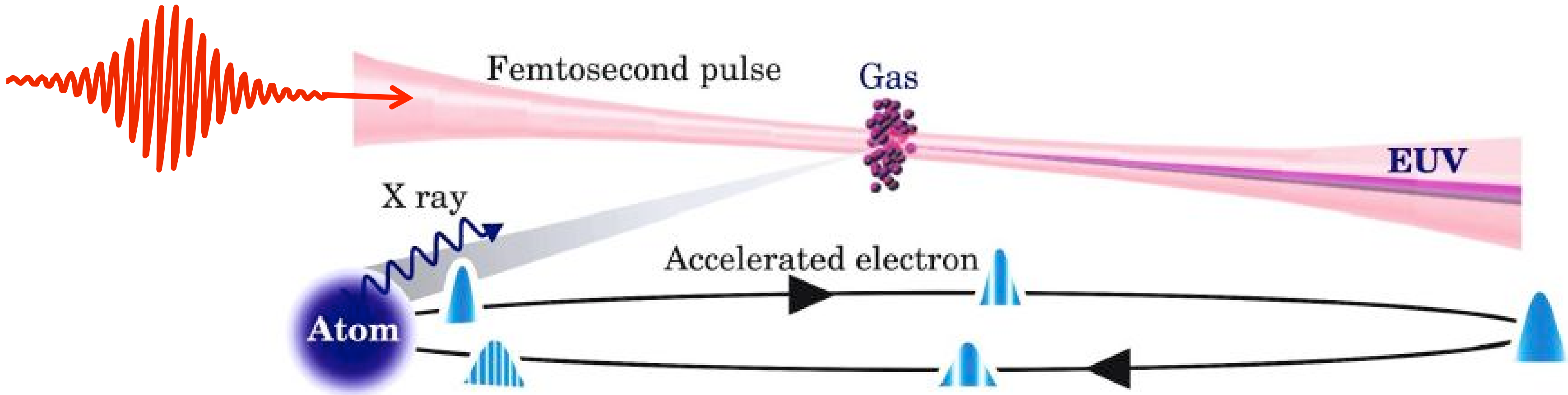


$E_P(\omega)=E_0^{i\omega t}$

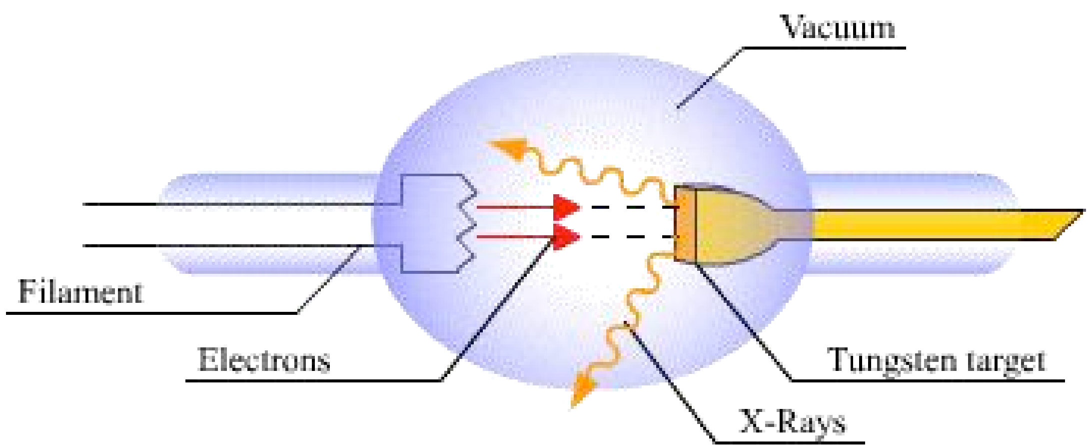
$d(\omega)=a_1e^{i\omega t} + a_2e^{i2\omega t} + \ldots$



High harmonics - coherent version of X-Ray tube

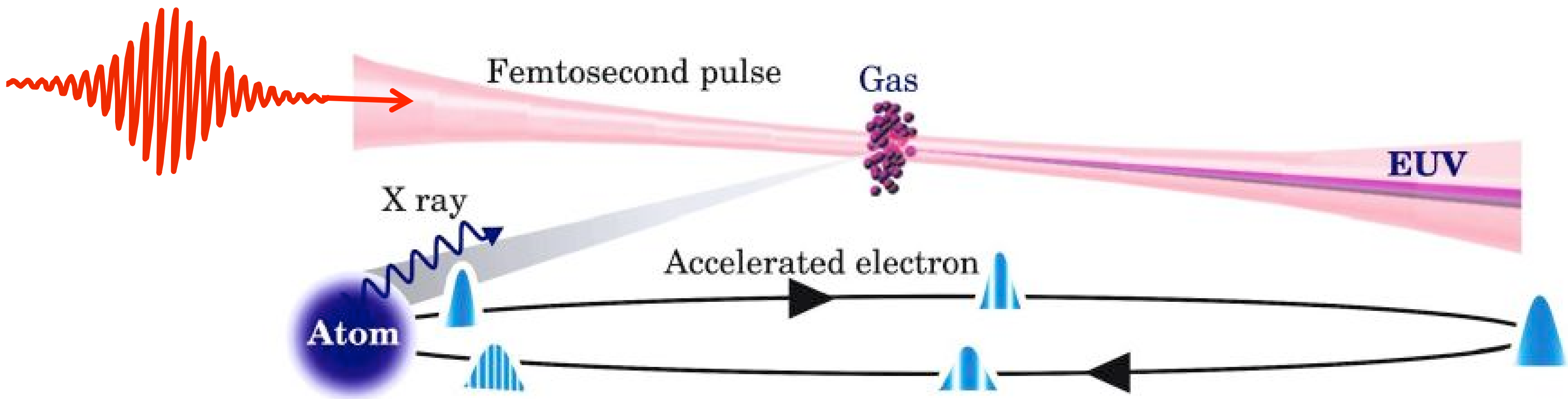


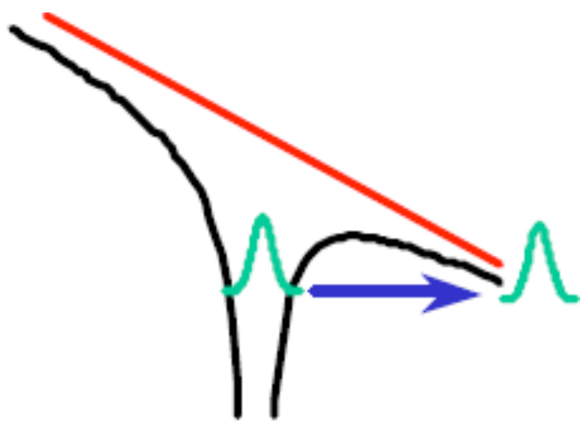
High Harmonic Generation *(McPherson et al, JOSA B **4**, 595 ('87); Ferray et al, J Phys B **21**, L31 ('88))*



Röntgen X-ray Tube

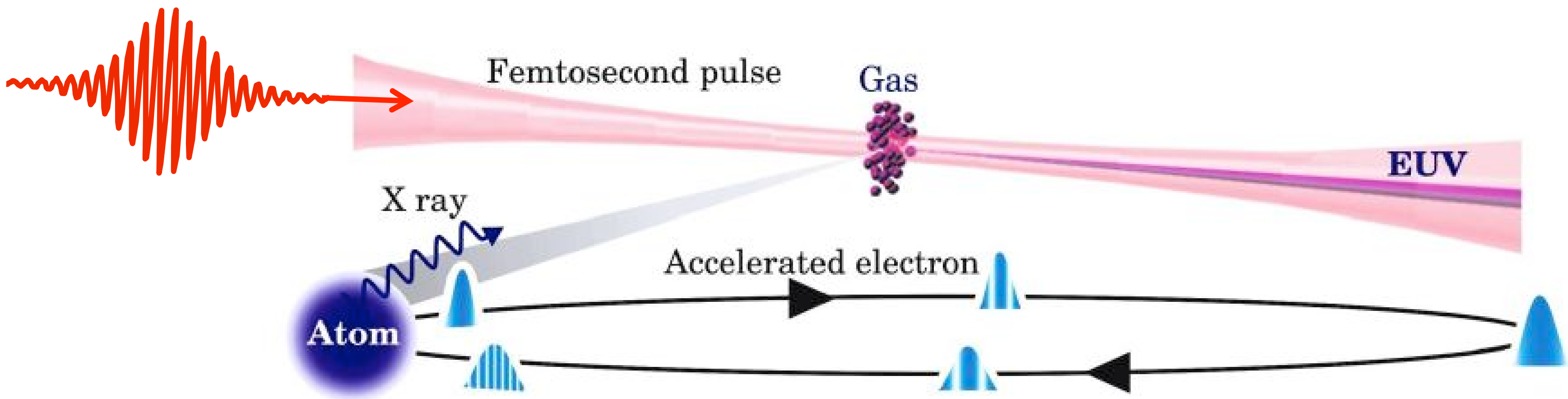
Extreme nonlinear optics - high harmonic generation

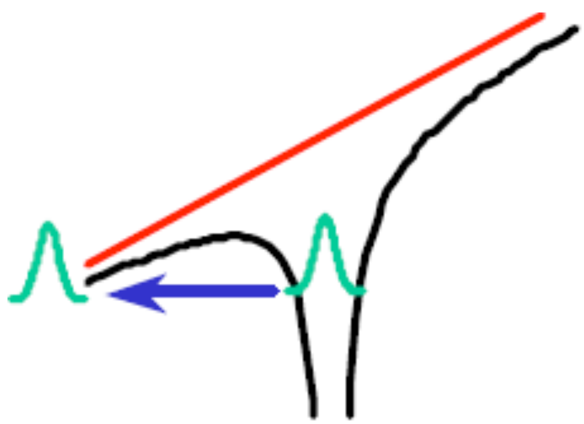




*Corkum, PRL **71**, 1994 (1993)*
Kulander, Schafer, Krause, SILAP Proceedings, 95 (1992-3)
*Kuchiev, JETP **45**, 404 (1987)*

Extreme nonlinear optics - high harmonic generation





*Corkum, PRL **71**, 1994 (1993)*
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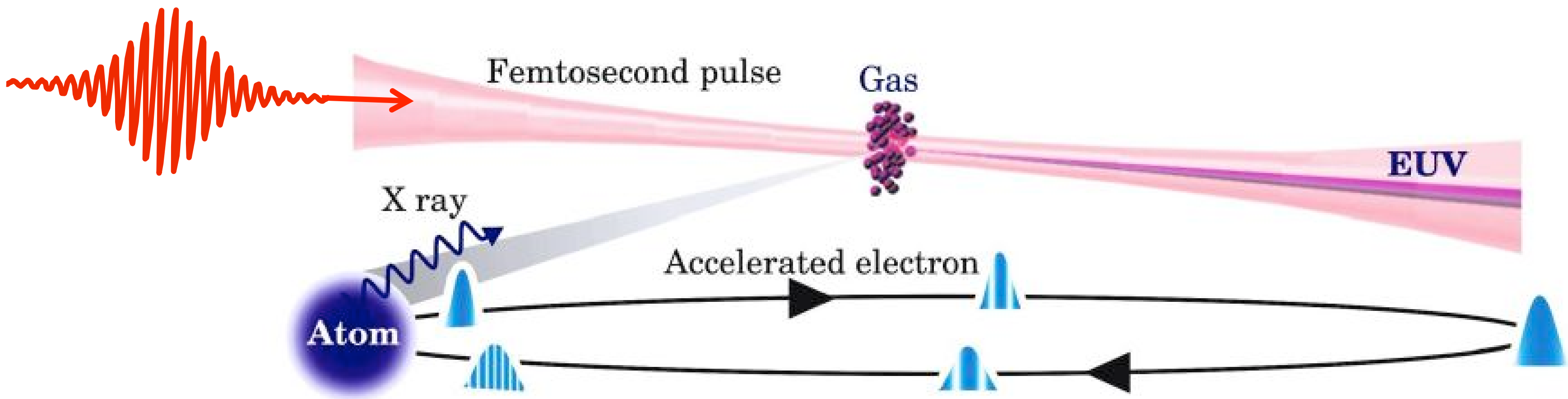
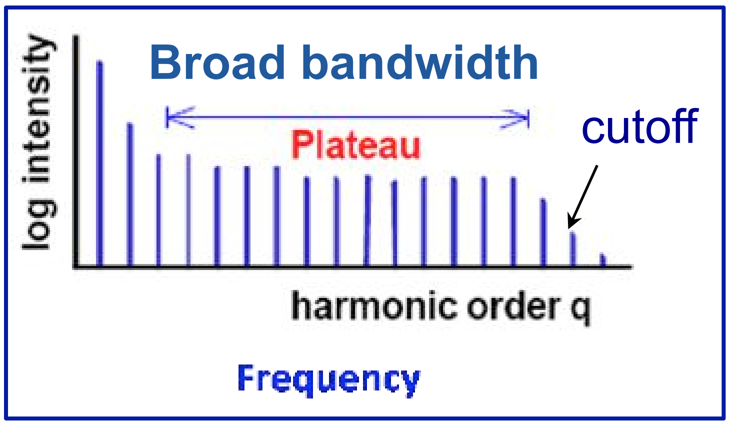
*Kuchiev, JETP **45**, 404 (1987)*

High harmonic generation – microscopic physics





Harmonics from single atom

$$h\nu_{cutoff} = I_p + 3.2U_p \approx I_L\lambda^2$$

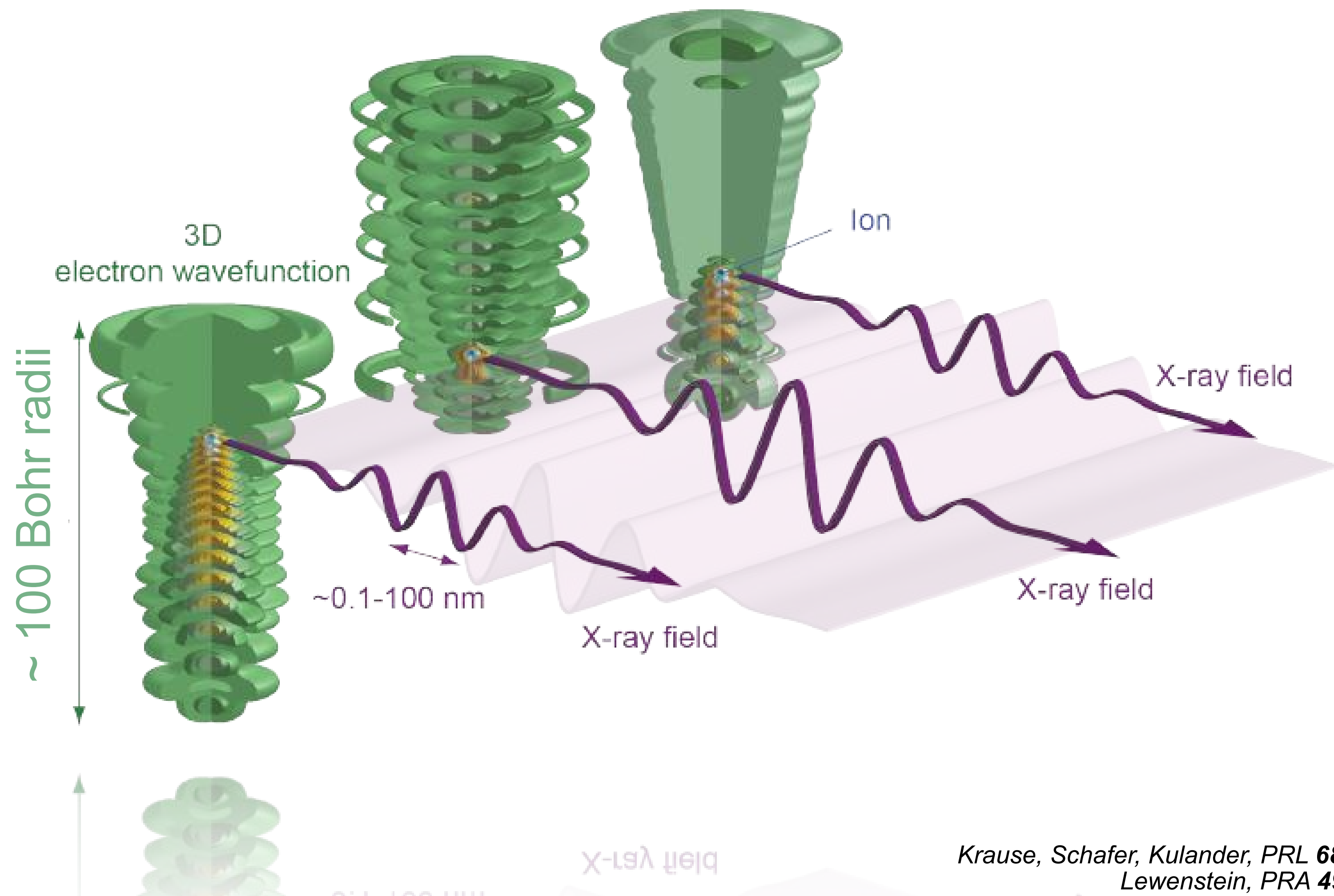
ionization potential (I_p) — energy of e^- (U_p)

*Corkum, PRL **71**, 1994 (1993)*
Kulander, Schafer, Krause, SILAP Proceedings, 95 (1992-3)
*Kuchiev, JETP **45**, 404 (1987)*

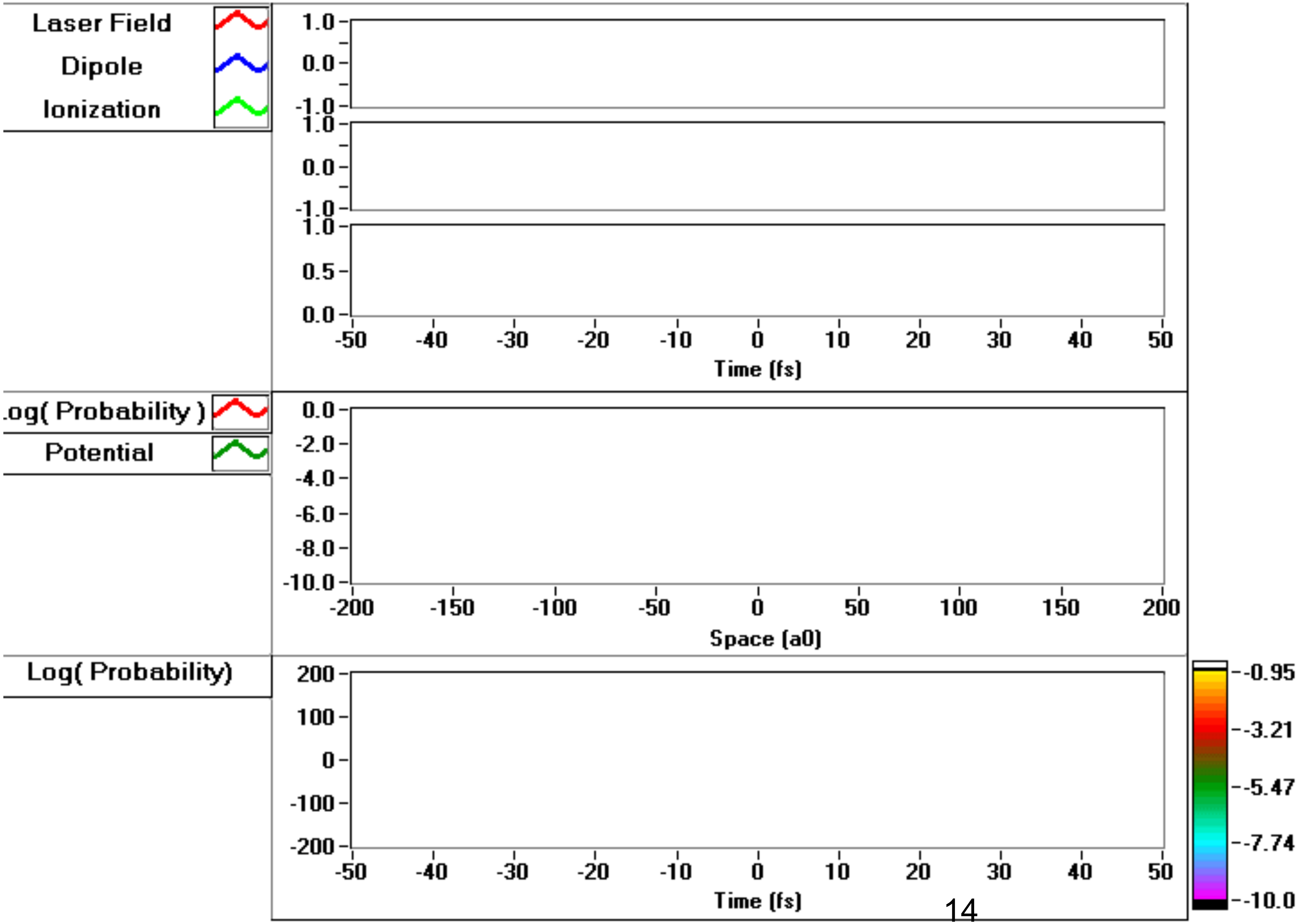
High Harmonic Generation – single atom quantum picture

$$x(t) = i\int_0^t dt'\int d^3\bar{p}\, d_x^*\left[\bar{p}-\bar{A}(t)\right] e^{-iS(p,t,t')} E(t') d_x\left[\bar{p}-\bar{A}(t')\right] + c.c.$$

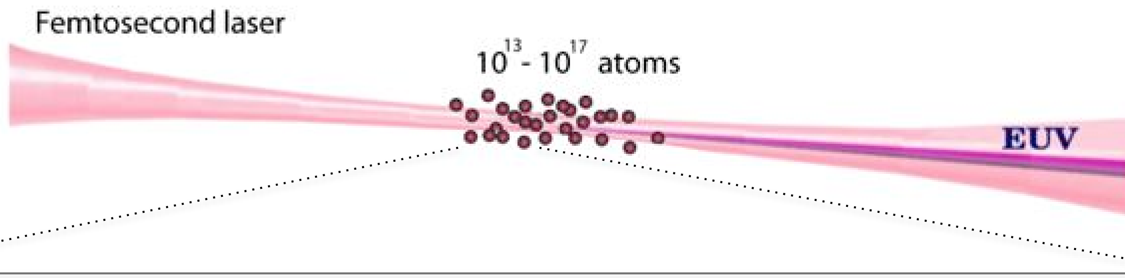
Tunnel ionization **Propagation** **Recombination**

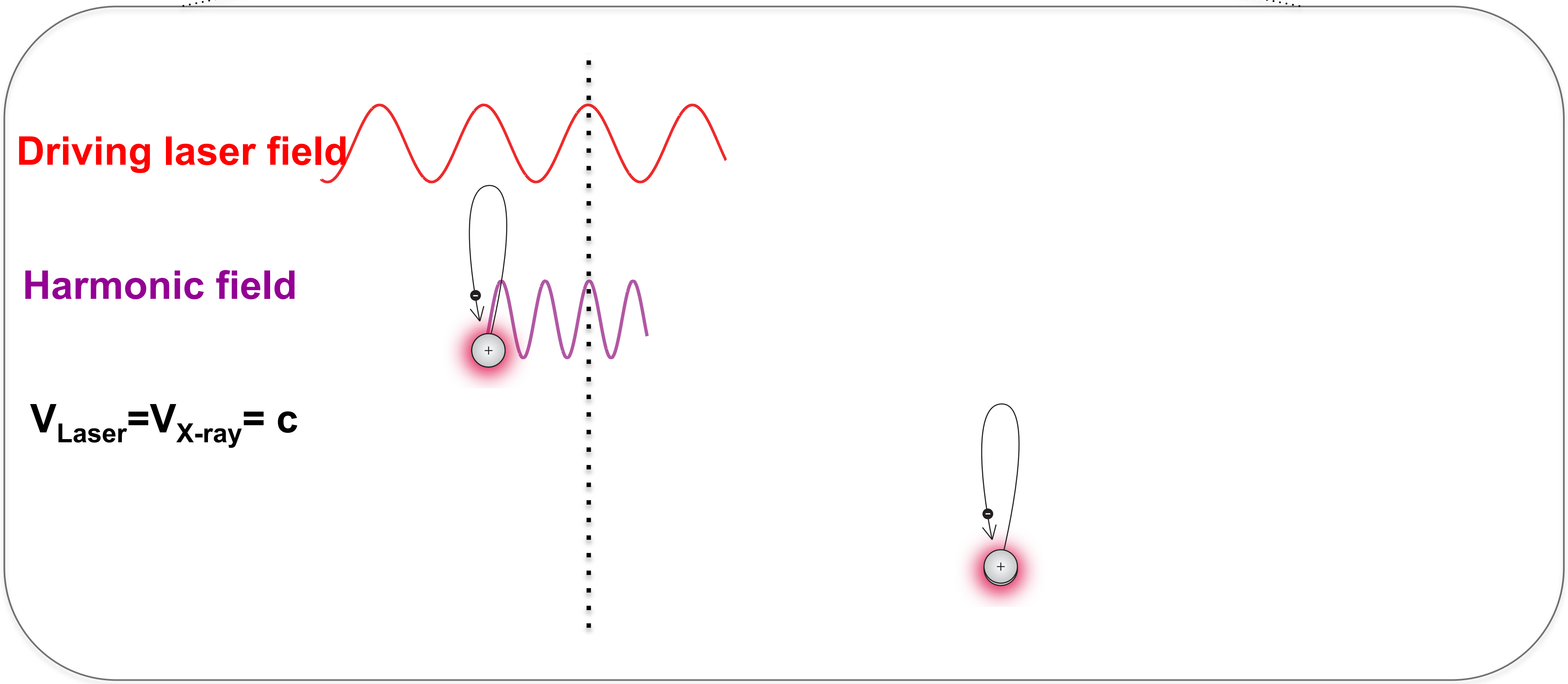


*Krause, Schafer, Kulander, PRL **68**, 3535 (1992)*
*Lewenstein, PRA **49**, 2117 (1994)*

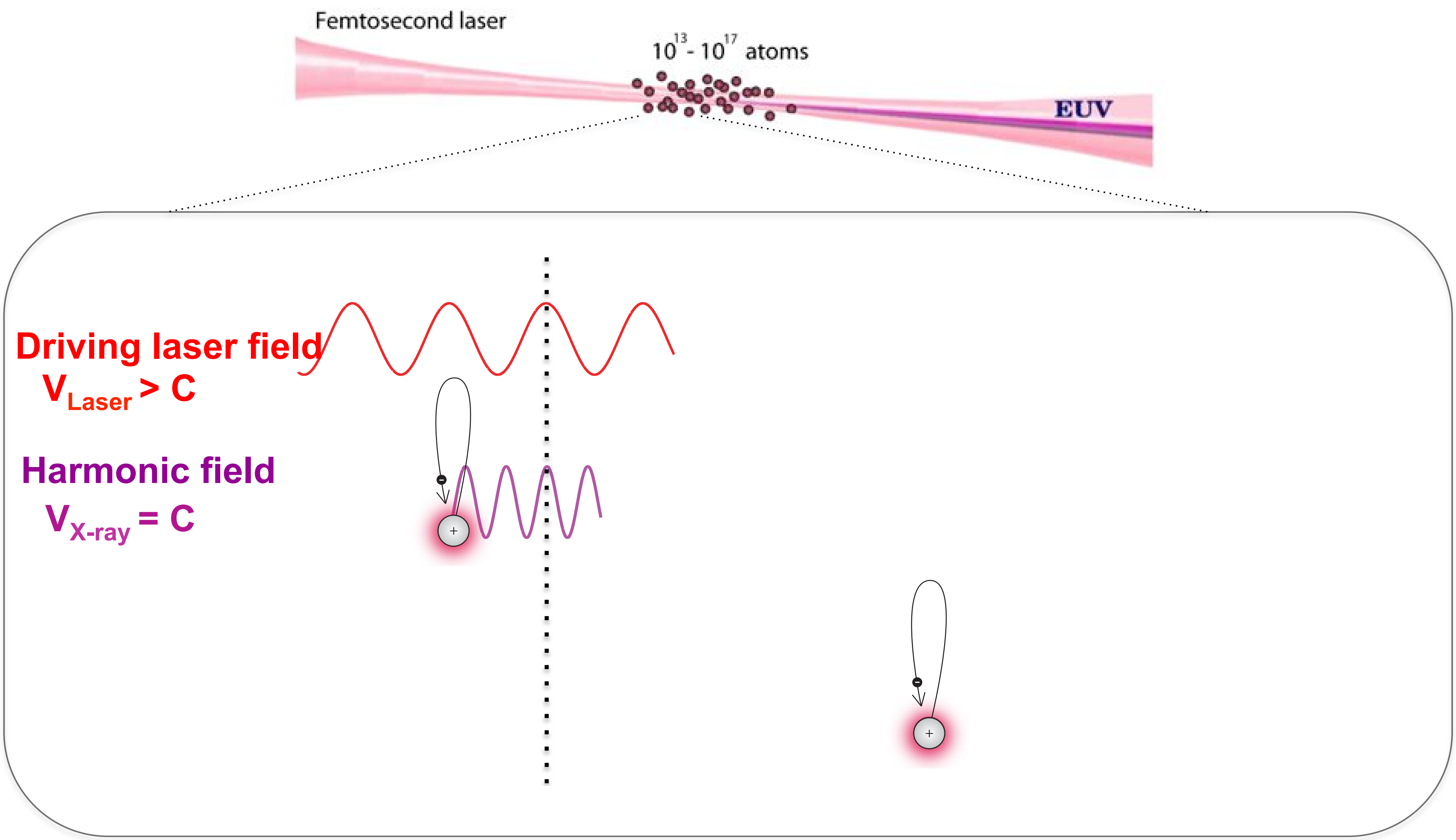

Laser Field
Dipole
Ionization
Time (fs)
Log(Probability)
Potential
Space (a0)
Log(Probability)
Time (fs)
-0.95
-3.21
-5.47
-7.74
-10.0

Challenge for HHG – macroscopic phase matching

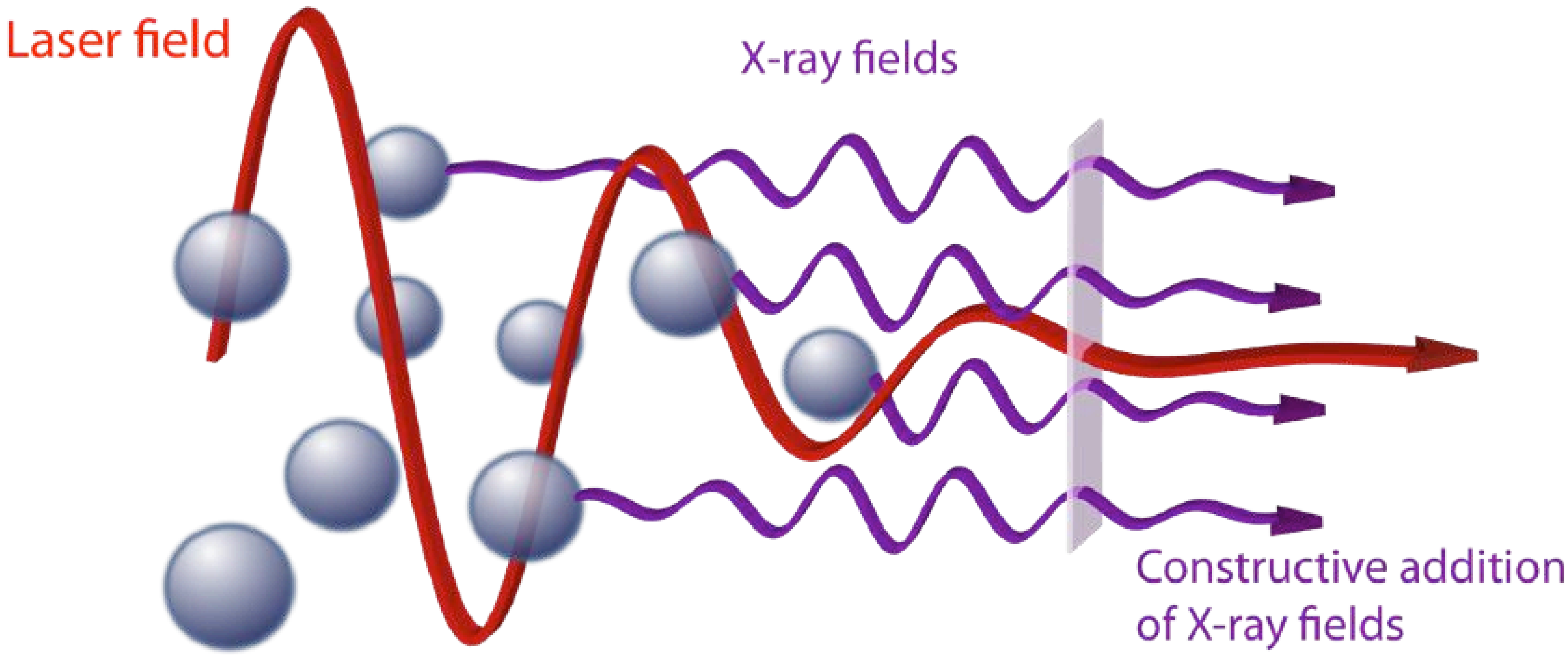


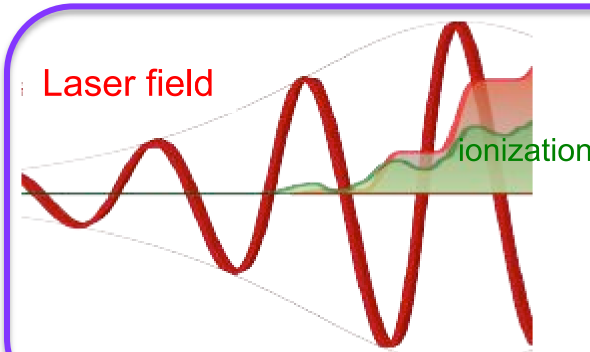


Challenge for HHG – macroscopic phase matching



Efficient HHG requires phase velocity matching





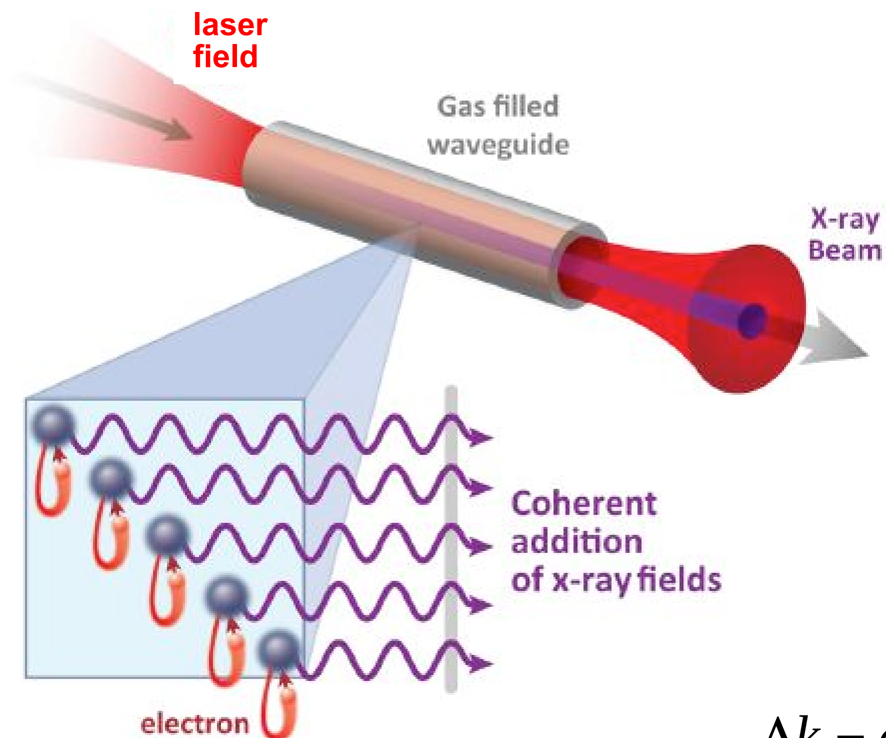
$V_{Laser} > C$

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

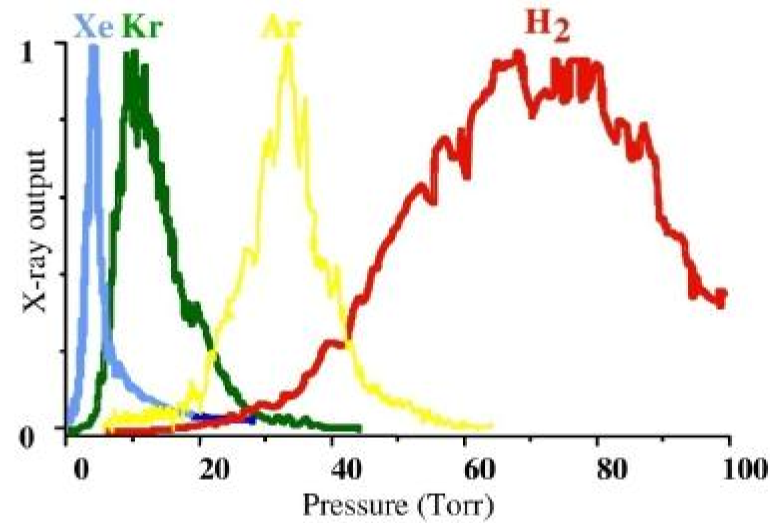
- Refractive index (phase velocity) of laser is time dependent!

Pressure tuned phase velocity matching

- 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**

Science ***280****, 1412 (1998)*
Science ***297****, 376 (2002)*

Pressure tuned phase velocity matching

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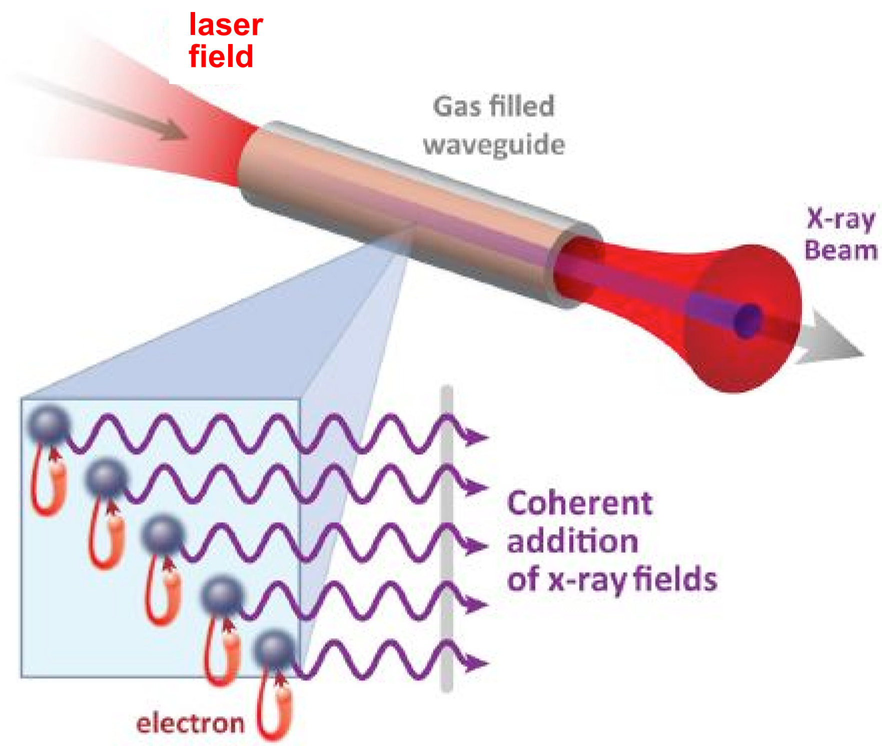
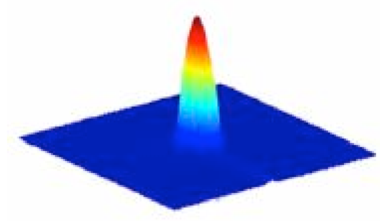
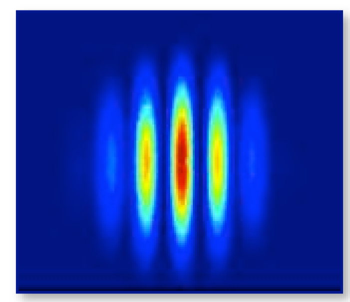






Fully coherent, laser-like, extreme ultraviolet high harmonic beams

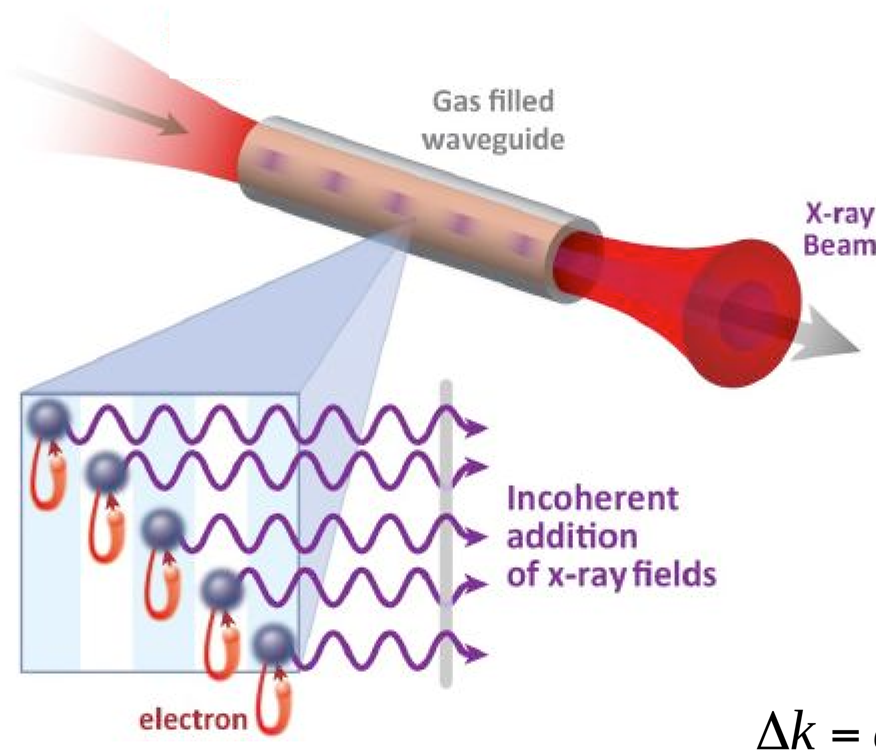
$V_{Laser}=V_{X\text{-}ray}=C$

Science ***280****, 1412 (1998)*
Science ***297****, 376 (2002)*

Limits of phase matching

- Turning up laser intensity creates plasma that speeds up laser phase velocity
- Defines critical ionization/photon energy above which phase matching impossible (150eV)

$$h\nu_{Single\ atom\ cutoff} \propto I_L \lambda_L^2$$



$V_{laser} \neq C$

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

Waveguide **Neutrals** **Plasma**

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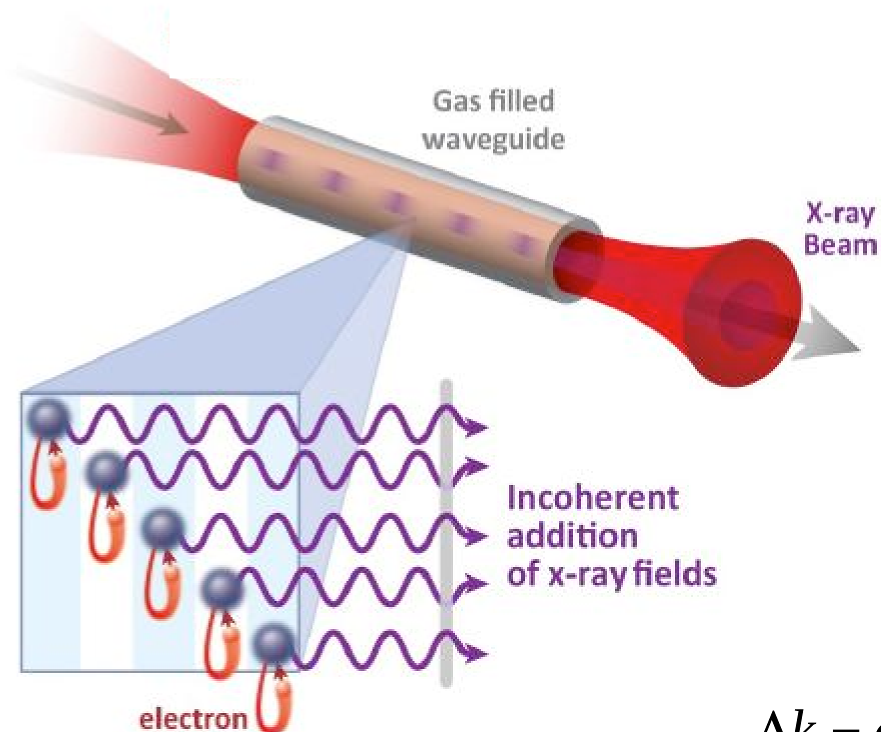
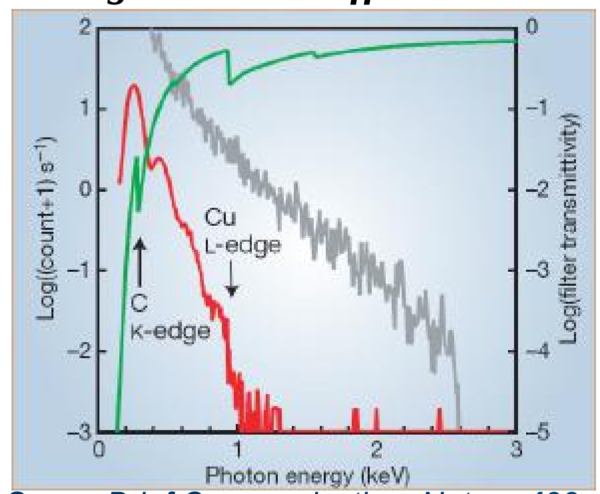


$V_{laser} \neq C$



Seres, Brief Communication, Nature ***433****, 596 (2005)*

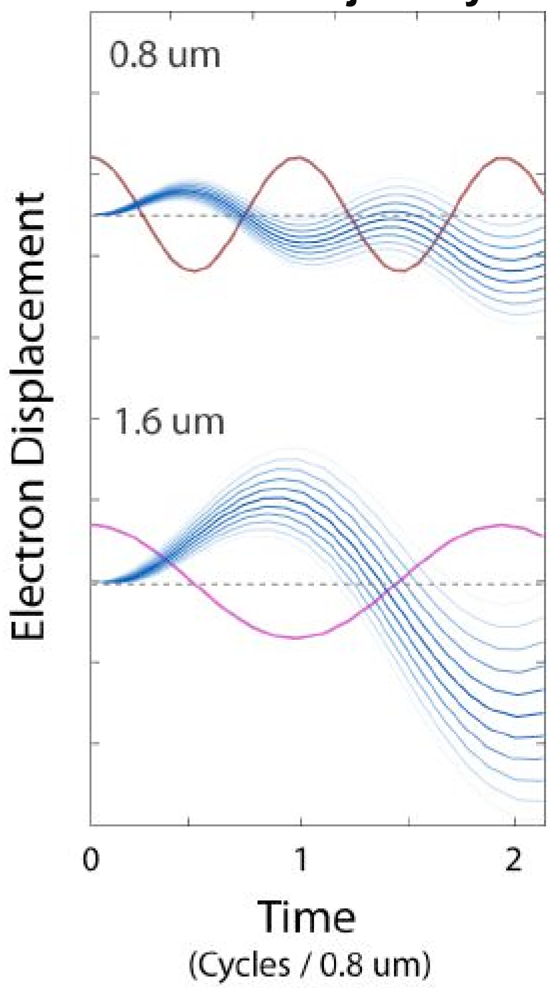
$$\Delta k = q\left\{\left(\frac{u_{11}^2 \lambda_0}{4\pi a^2}\right) - P\left((1-\eta)\frac{2\pi}{\lambda_0}\Delta\delta - \eta\left[N_{atm} r_e \lambda_0\right]\right)\right\}$$

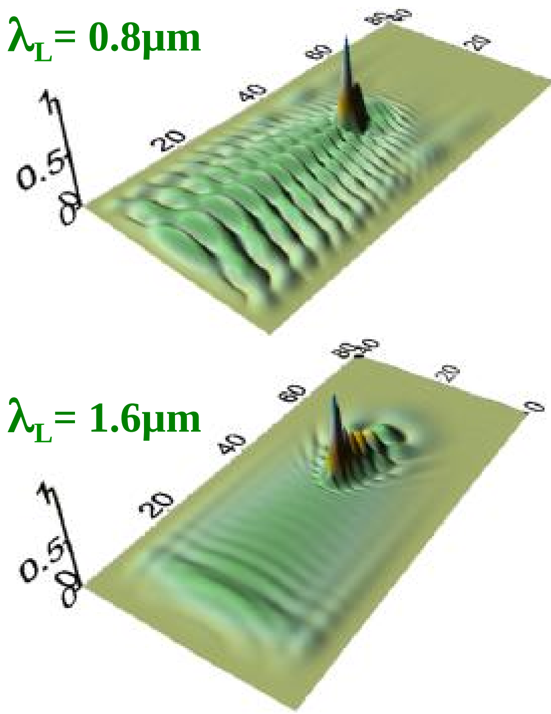
Waveguide **Neutrals** **Plasma**

High harmonic generation using long wavelength lasers

Single atom HHG: $h\nu_{Single\ atom\ cutoff} \propto I_L \lambda_L^{2}$

Electron trajectory





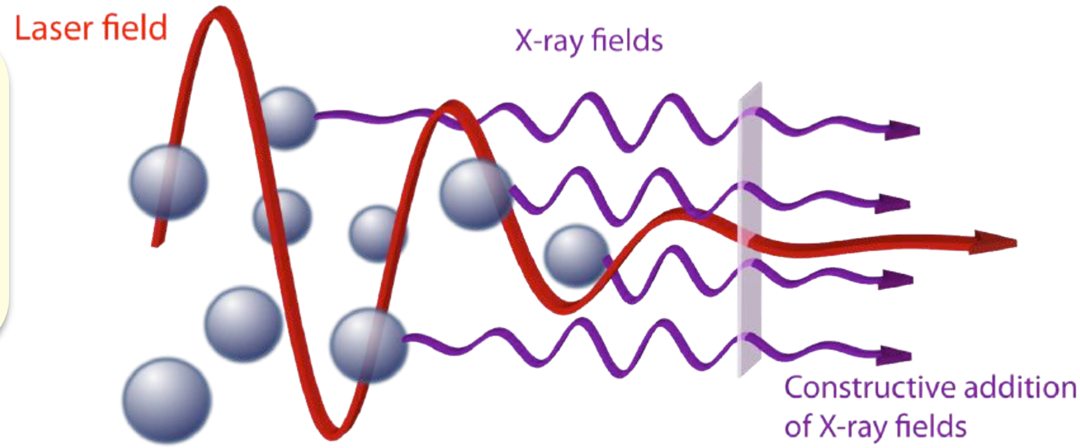
Electron wavepacket spreading due to quantum diffusion means single atom yield scales $\propto \lambda_L^{-5.5}$

Sheehy, Schafer, Gaarde, PRL ***83****, 5270 (1999)*
Tate et al.,PRL ***98****, 013901 (2007)*
Schiessl et al., PRL ***99*** *253903 (2007)*
Frolov et al., PRL ***100****, 173001 (2008)*

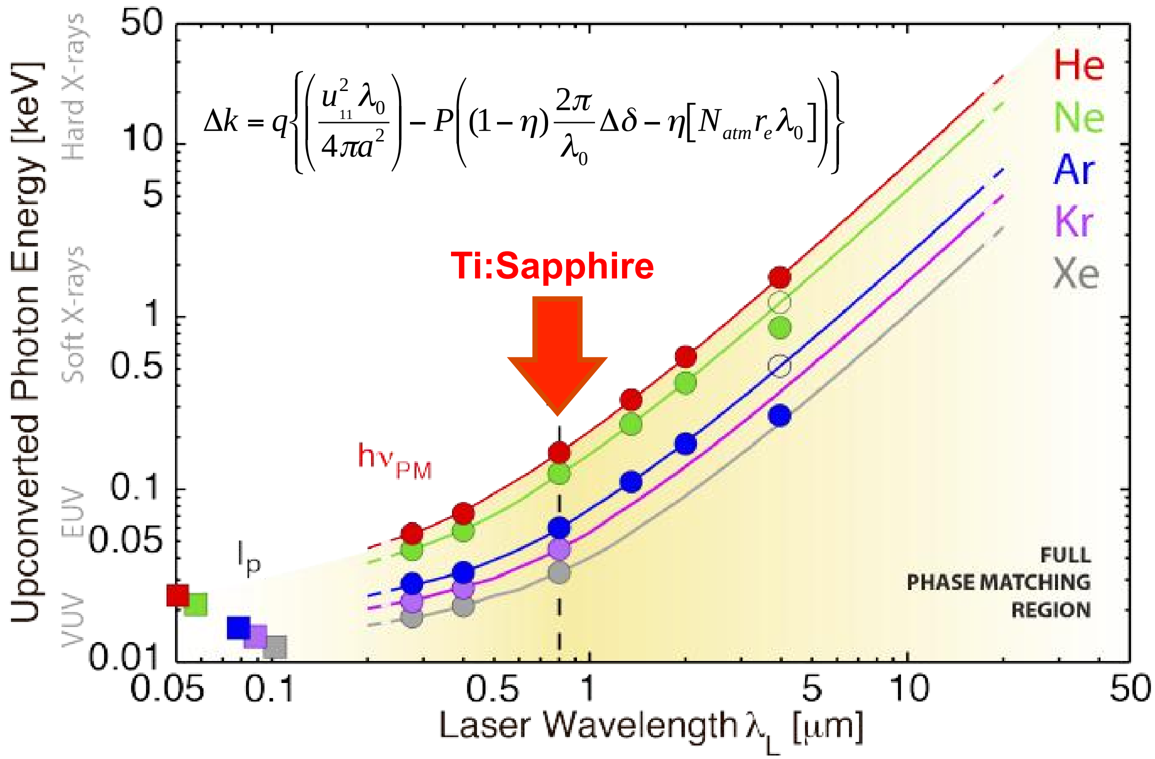
Phase matching in mid-IR overcomes low single-atom yield!

$$h\nu_{Single\text{-}atom\ cutoff} \propto I_L \lambda_L^{2}$$

$$h\nu_{Phase\ matched\ cutoff} \propto I_L \lambda_L^{1.7}$$



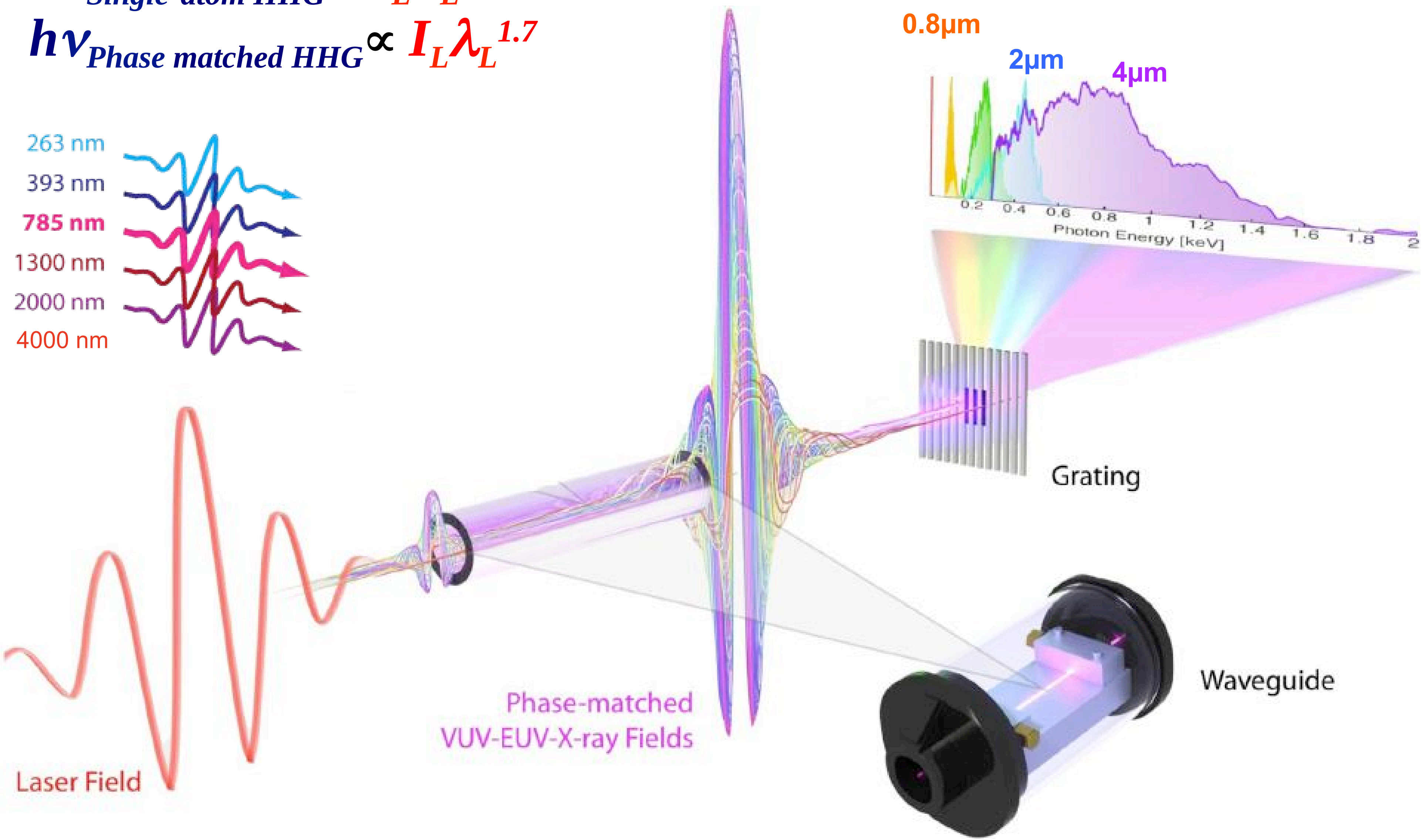
- Mid-IR driving lasers extend HHG phase matching to > keV
- **Counterintuitive finding:** MIR phase matching can overcome low single-atom yield since gas pressure and transparency increase!



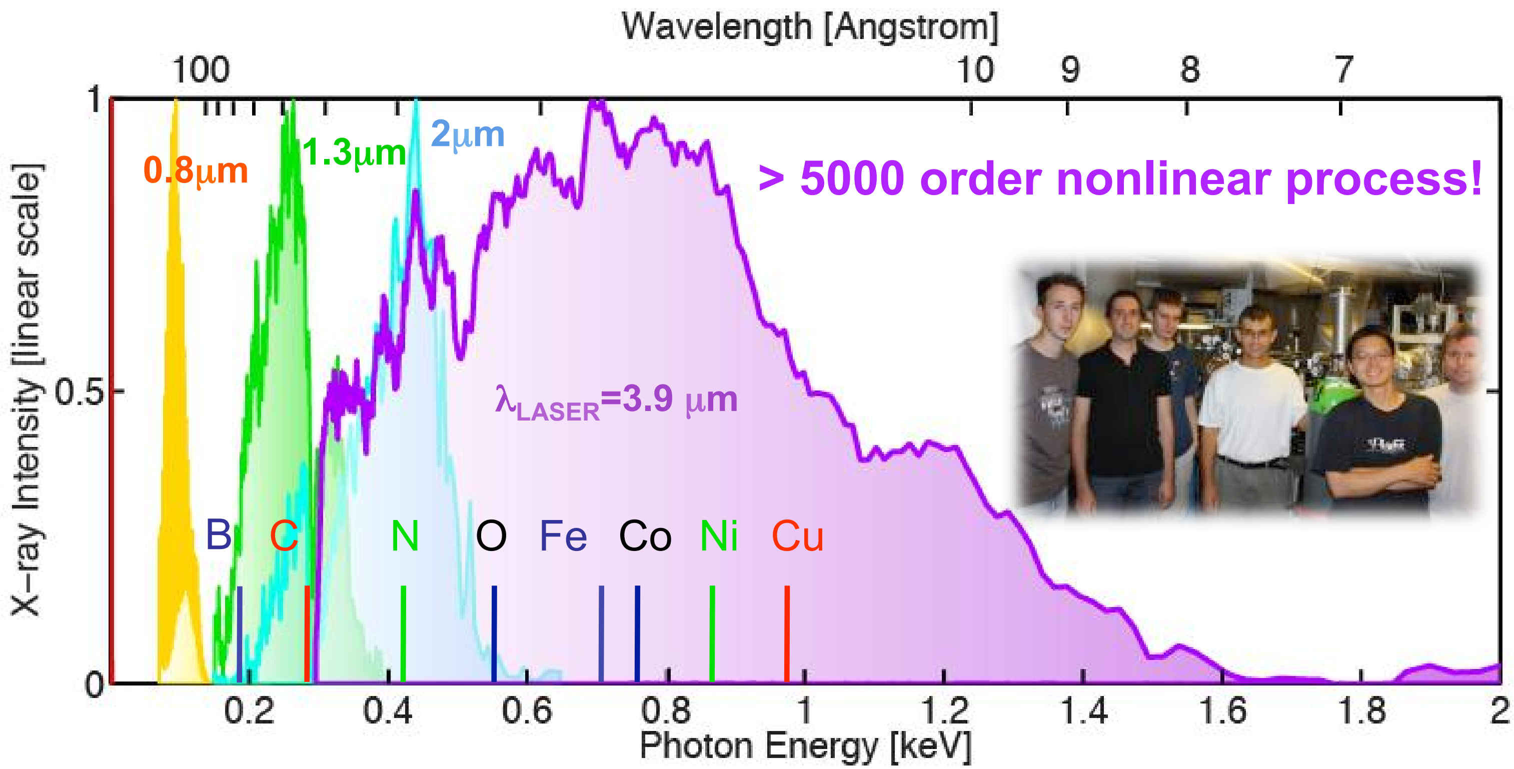
Extending bright high harmonics into the soft x-ray region

$h\nu_{Single\ atom\ HHG} \propto I_L \lambda_L^{2}$

$h\nu_{Phase\ matched\ HHG} \propto I_L \lambda_L^{1.7}$



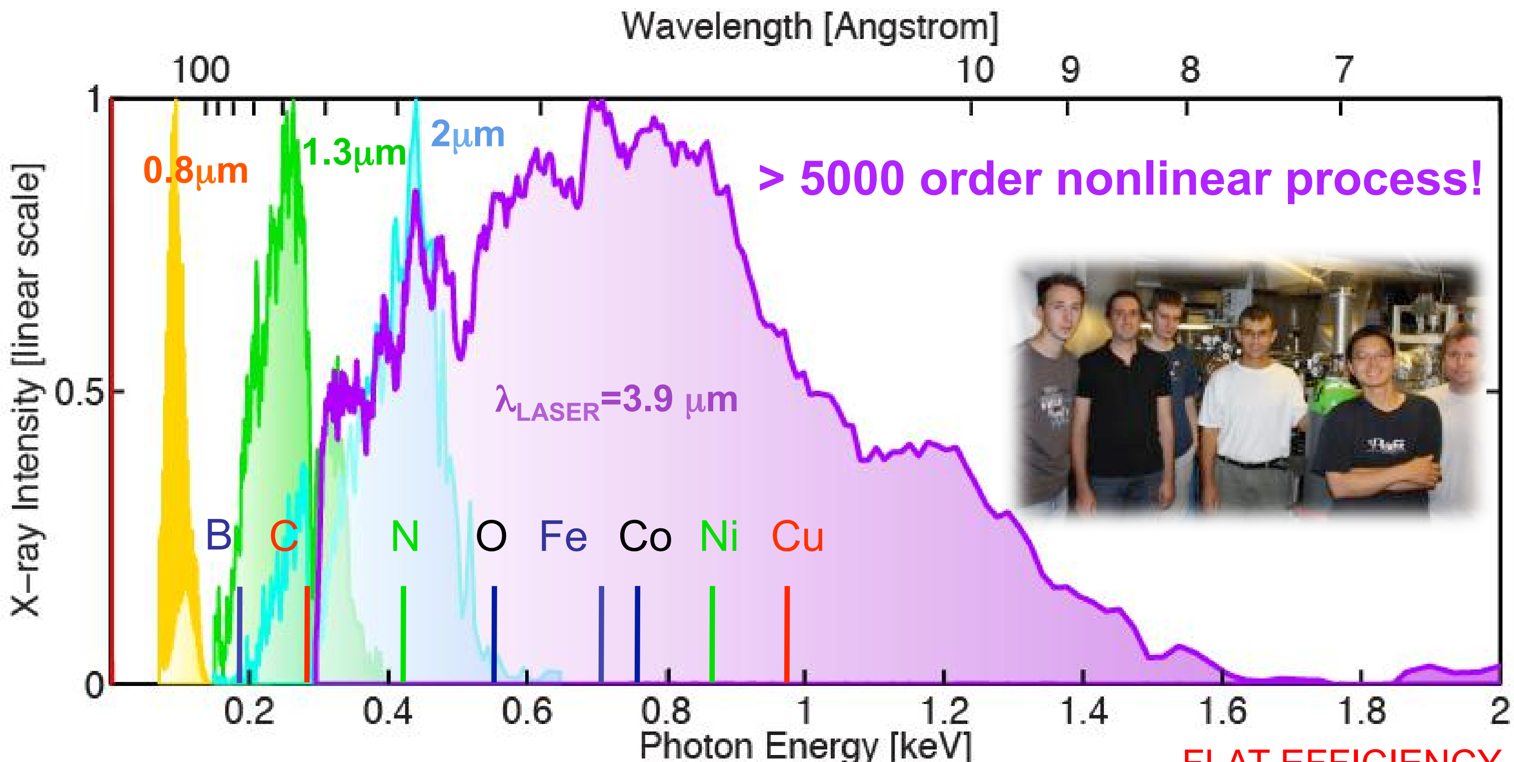
Coherent x-ray supercontinuum reaching 8Å



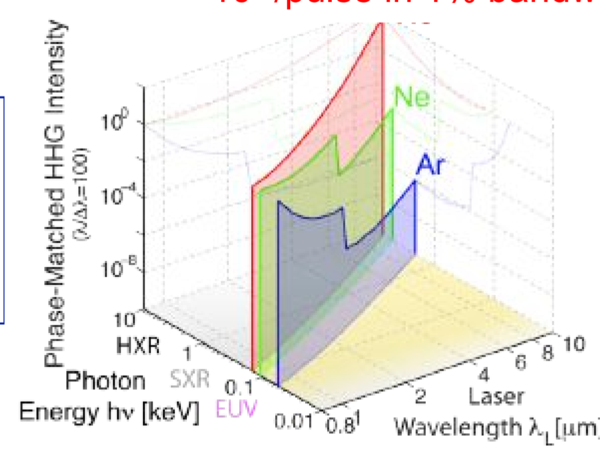
- Bright coherent tabletop keV x-rays for first time
- Near theoretically-limited (absorption-limited) efficiency to keV! (> 10^{-5}/pulse in 1% bandwidth)

Popmintchev et al., Prov. US Patent (2008);
CLEO Postdeadline (2008);
Opt. Lett. ***33****, 2128 (2008);*
PNAS ***106****, 10516 (2009);*
Nature Photonics ***4****, 822 (2010).*
Chen et al., PRL ***105****, 173901 (2010).*
Popmintchev et al., CLEO Postdeadline (2011).
Science ***336****, 1287 (2012).*

Coherent x-ray supercontinuum reaching 8Å



FLAT EFFICIENCY INTO keV!
10^{-5}/pulse in 1% bandwidth



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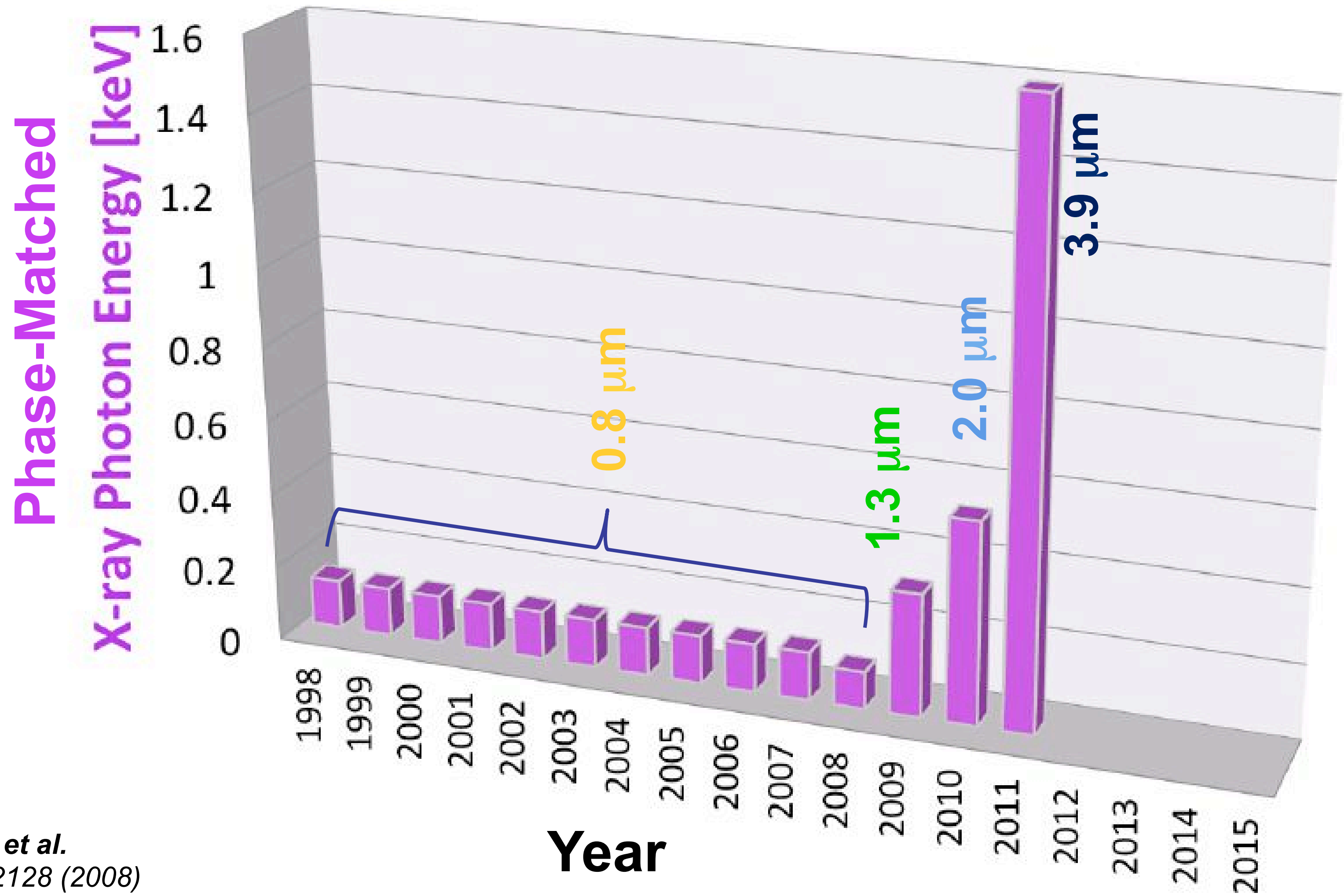
- Near theoretically-limited (absorption-limited) efficiency to keV! (> 10^{-5}/pulse in 1% bandwidth)

Dramatic progress in bright tabletop high harmonics

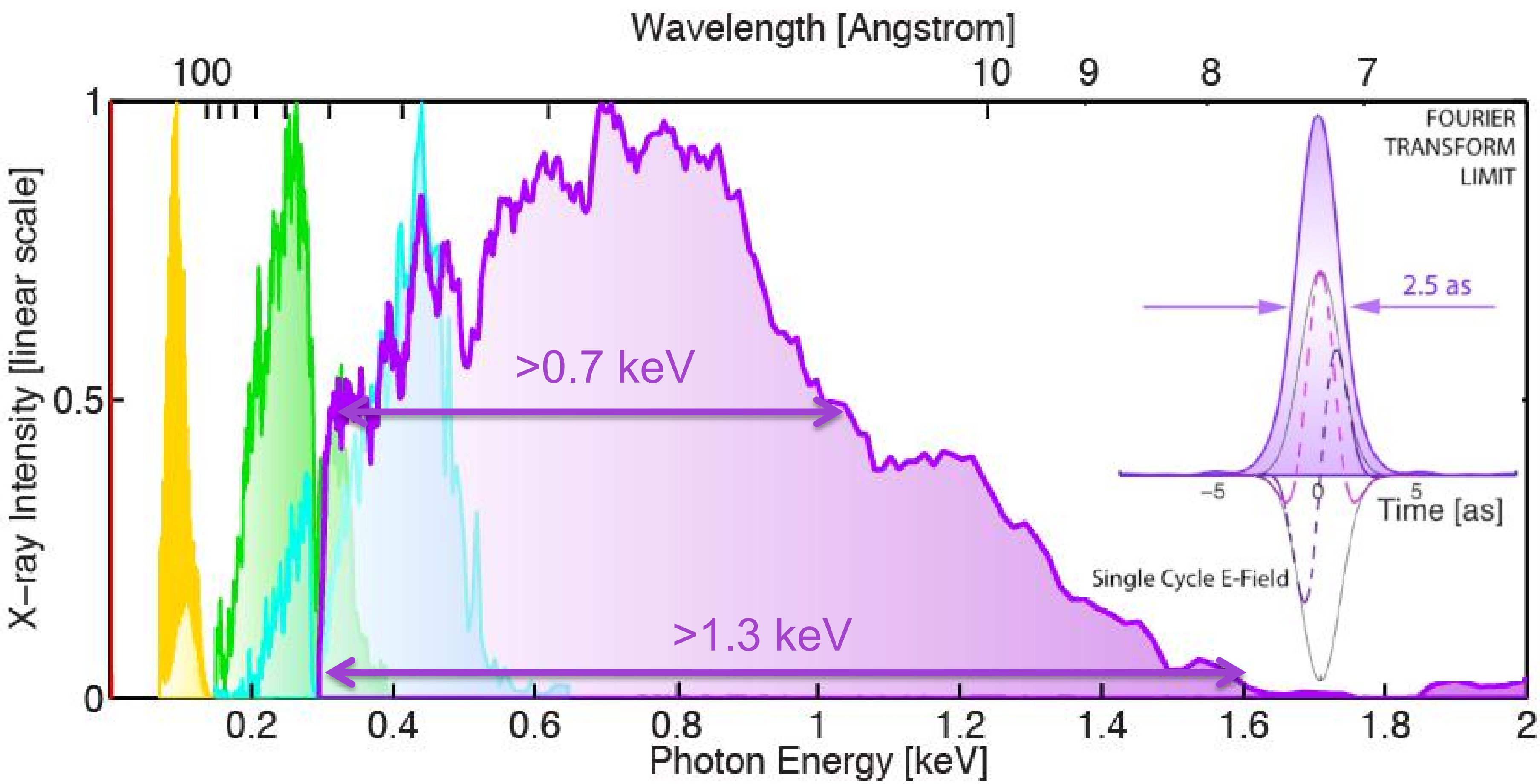


Popmintchev et al.
*Opt. Lett. **33**, 2128 (2008)*
*PNAS **106**, 10516 (2009)*
*PRL **105**, 173901 (2010)*
*Nature Photonics **4**, 822 (2010)*
*Science **336**, 1287 (2012)*

Broadest supercontinuum to date - 2.5 as pulses!

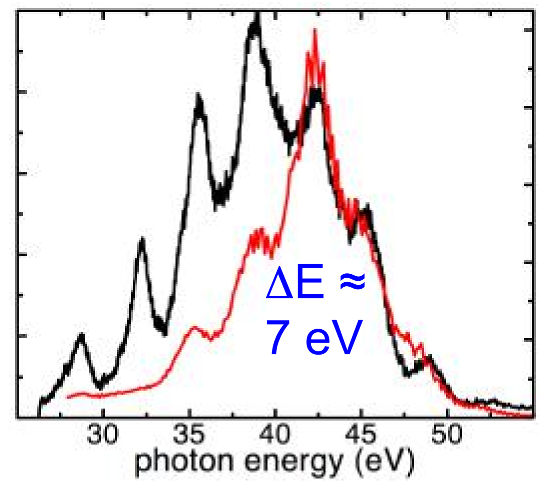
Uncertainty principle -

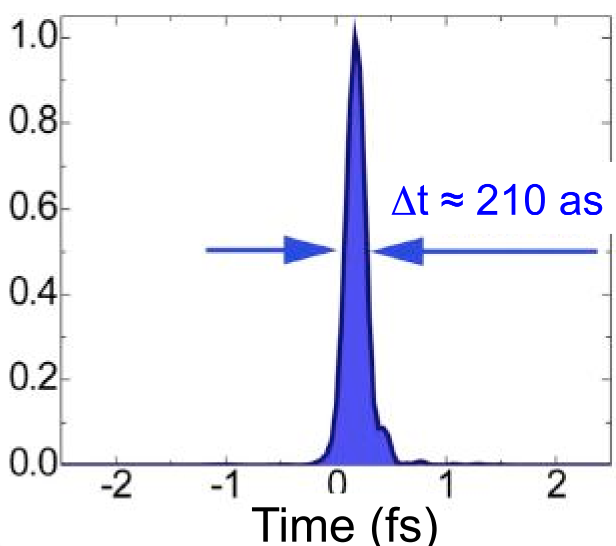
$\tau_{X\text{-}ray}^{FWHM}[as]\ \Delta E_{X\text{-}ray}[keV] \sim 1.8$



Bright **EUV** and **soft x-ray** attosecond pulses

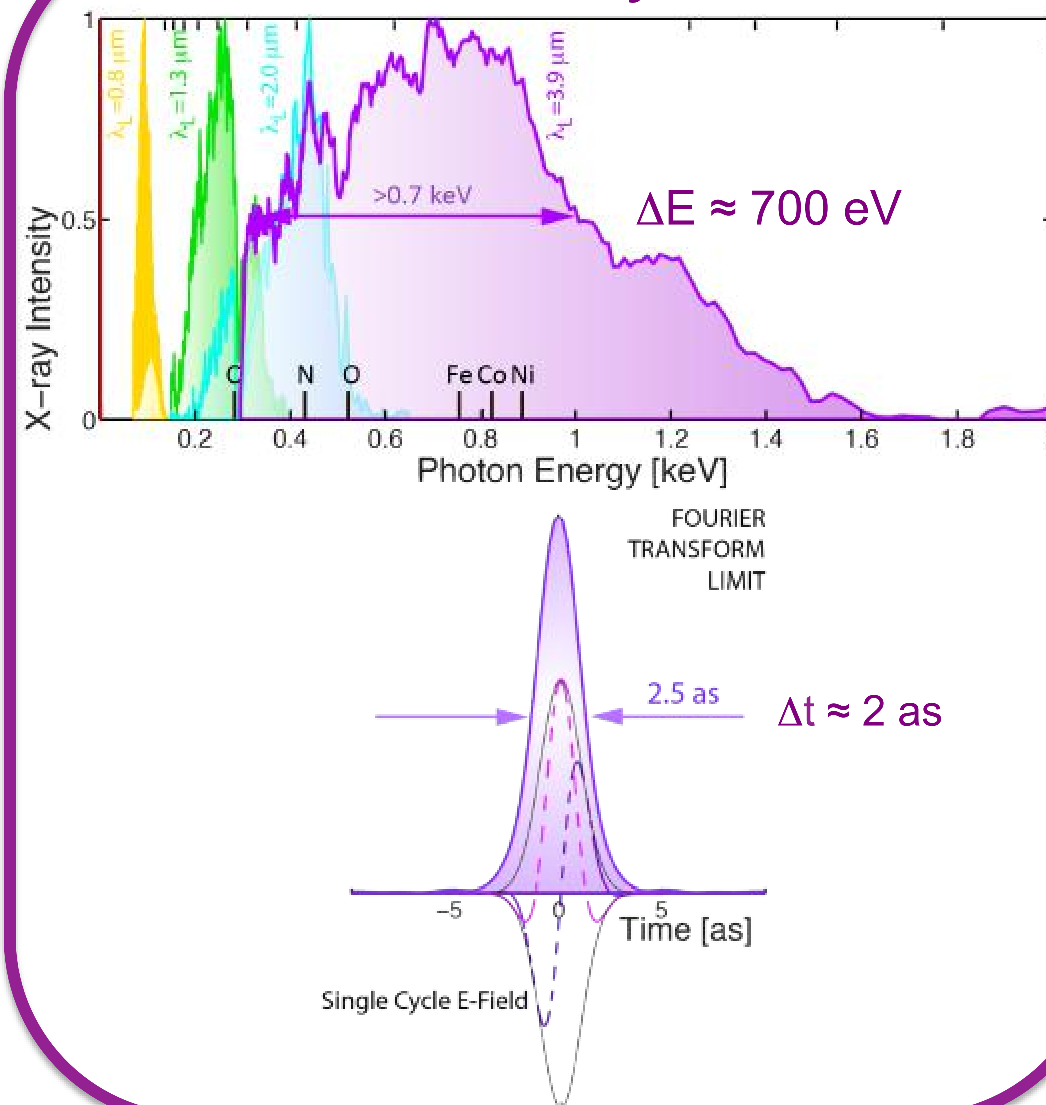
EUV





Opt. Express ***17****, 4611 (2009)*

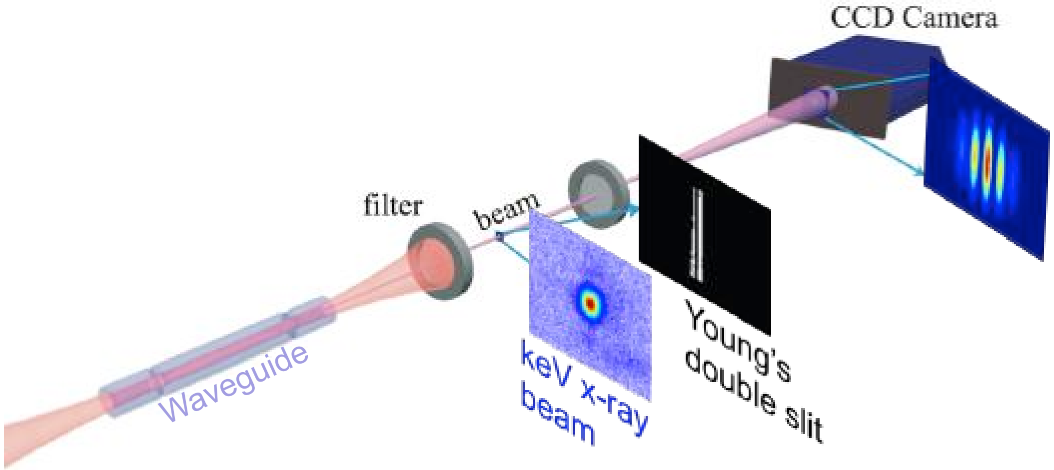
Soft x-ray

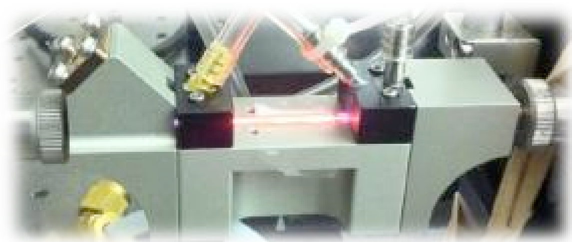


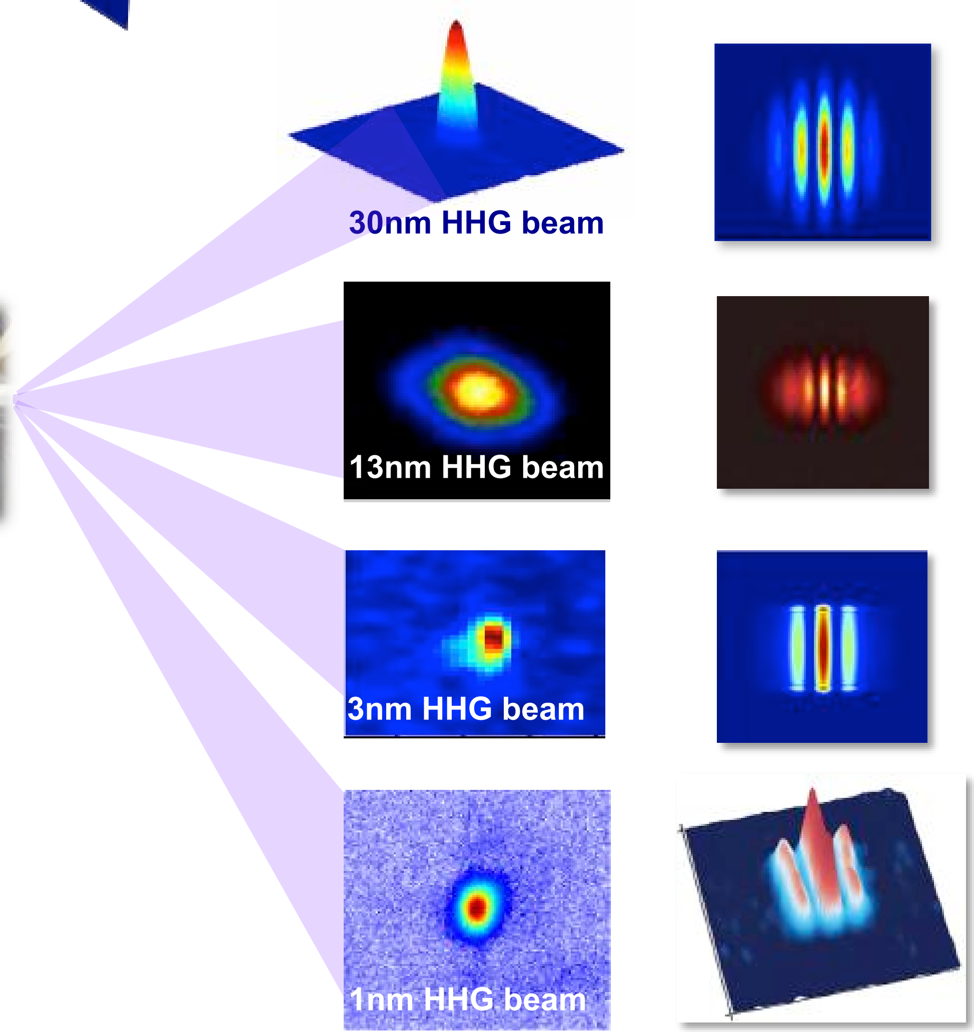
Science ***336****, 1287 (2012)*

Coherent laser-like x-ray beams from 1 – 30 nm

$$\theta \approx \lambda/d$$







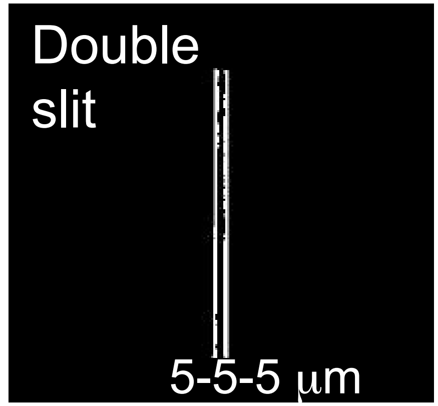
*Young, Philos. Trans. R. Soc. XCII **12**, 387 (1802)*
*Bartels et al, Science **297**, 376 (2002)*
*Zhang et al, Optics Letters **29**, 1357 (2004)*
*Popmintchev et al, Nature Photonics **4**, 822 (2010)*
*Chen et al, PRL **105**, 173901 (2010)*
*Popmintchev et al, Science **336**, 1287 (2012)*

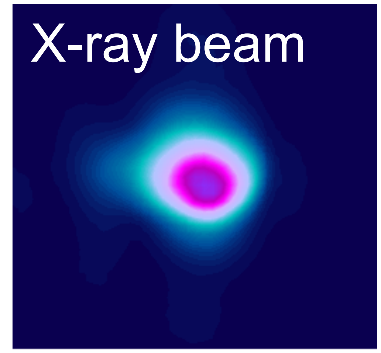
Bright, tabletop, coherent keV X-ray beams

Incoherent X-ray Supercontinua

Roentgen, Nature (1896).

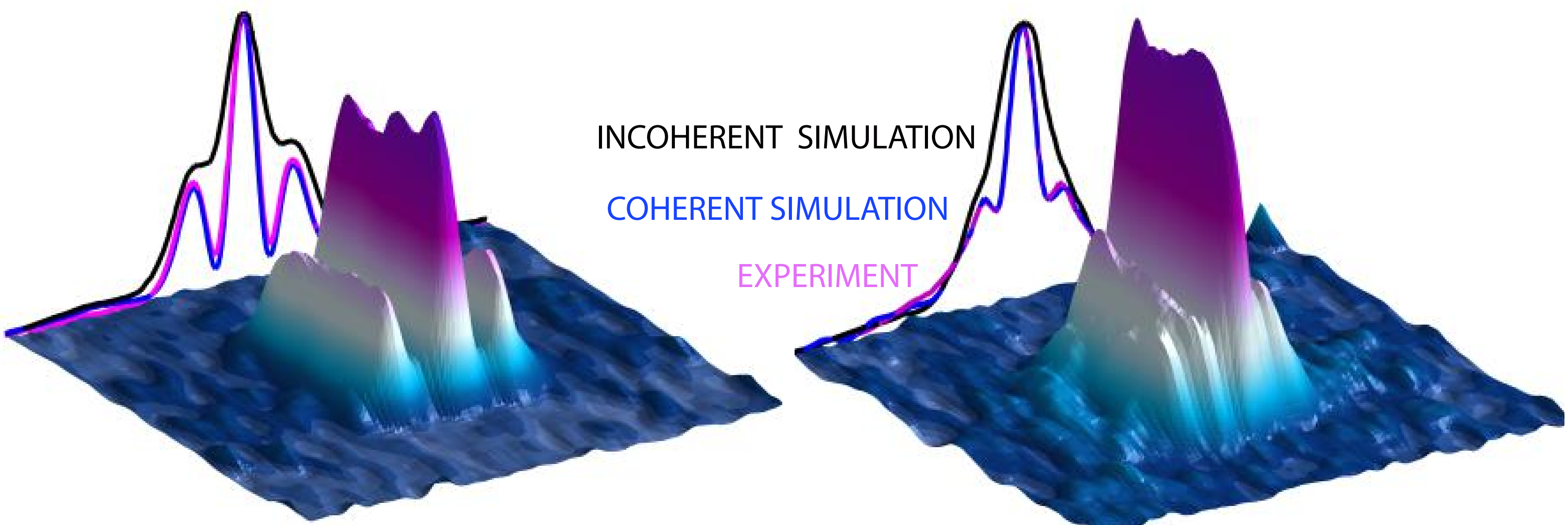
Coherent X-ray Supercontinua





Ne 14-43 Å

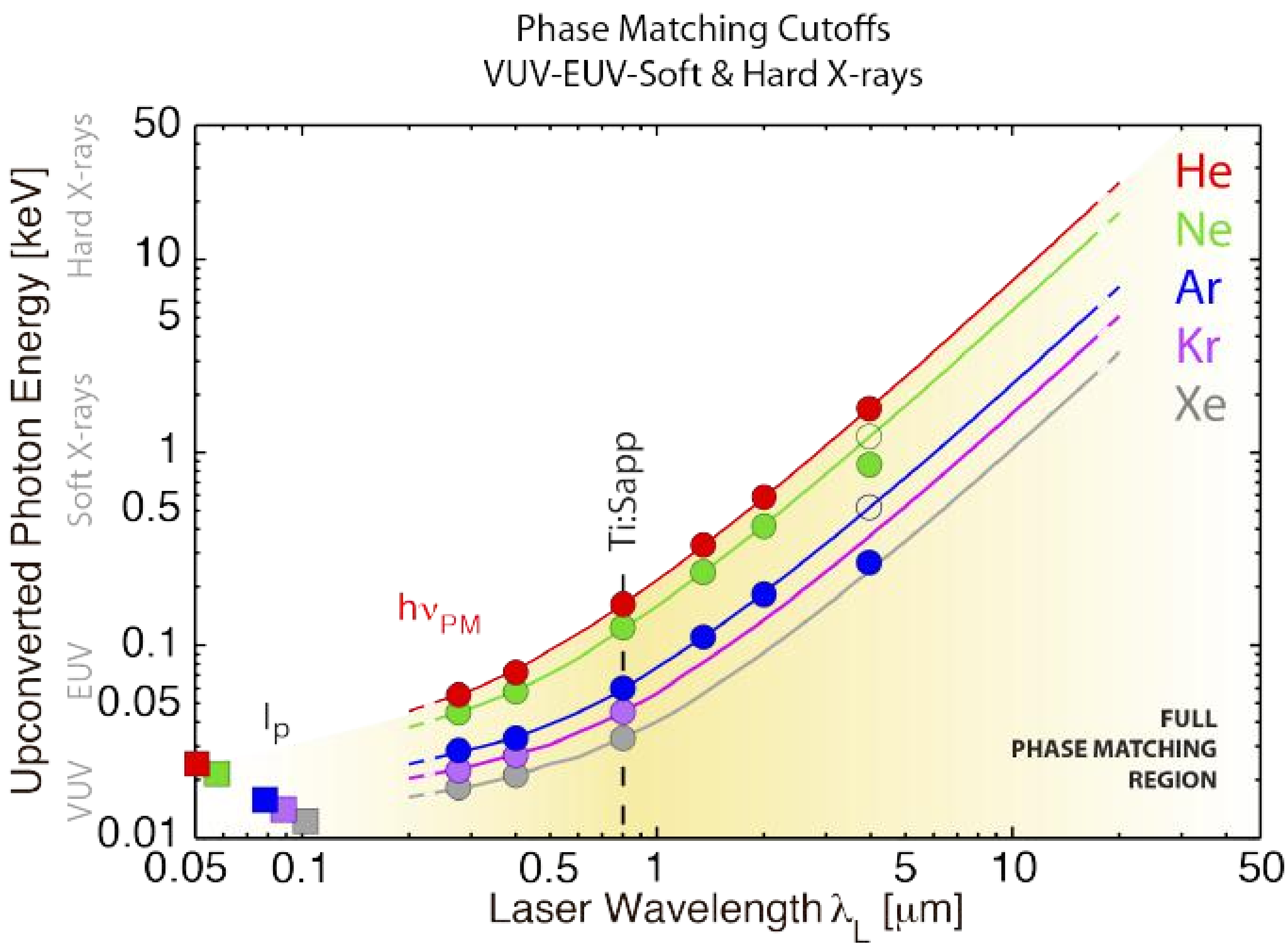
He 7.7-43 Å

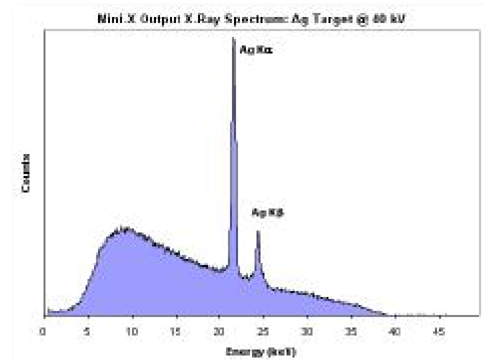


*Popmintchev et al., Science **336**, 1287 (2012).*

Limits of high harmonic generation?

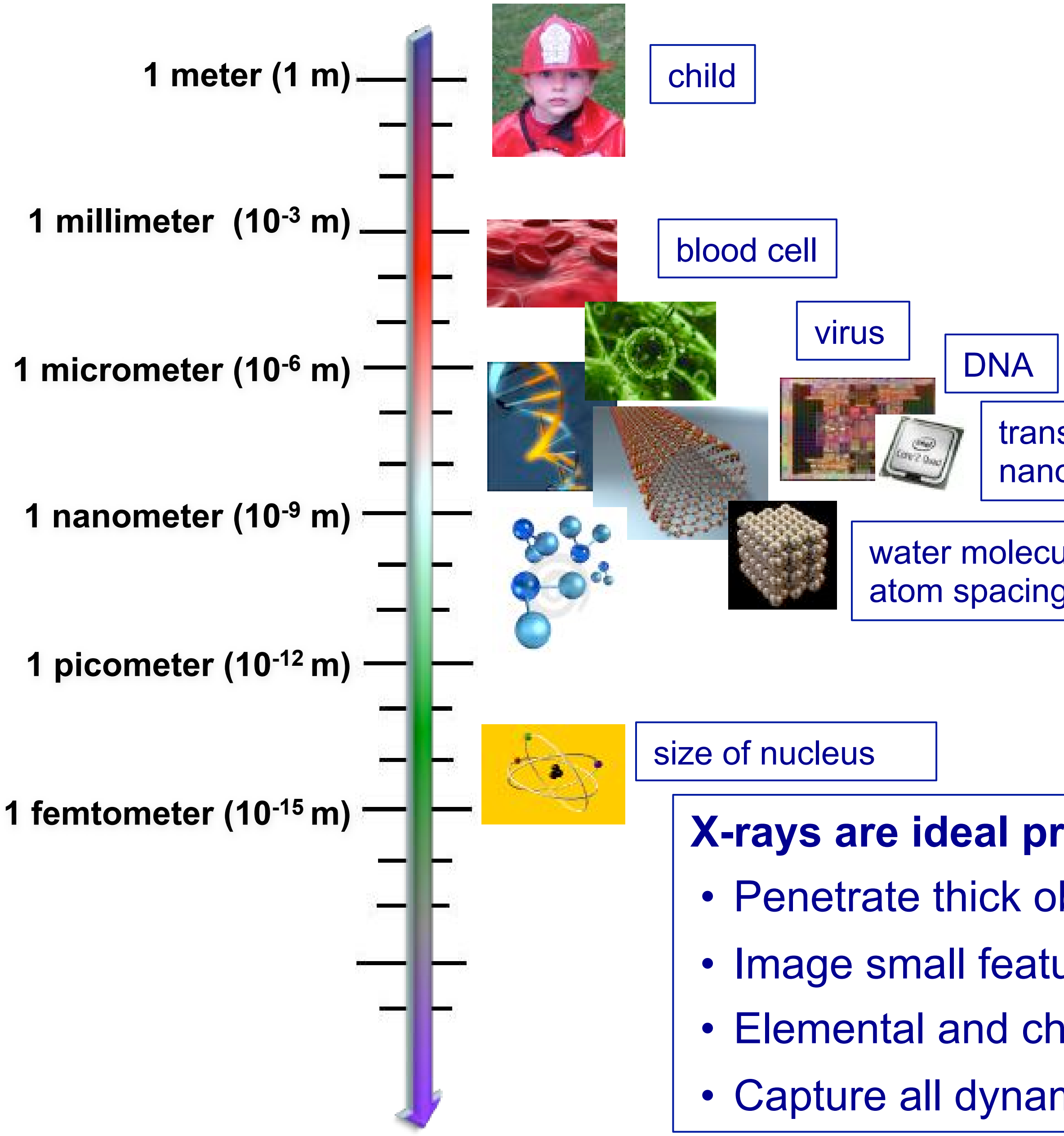
- He driven by 20 μm mid-IR lasers may generate bright 25 keV beams
- ≈ ½ million order phase matched nonlinear process!

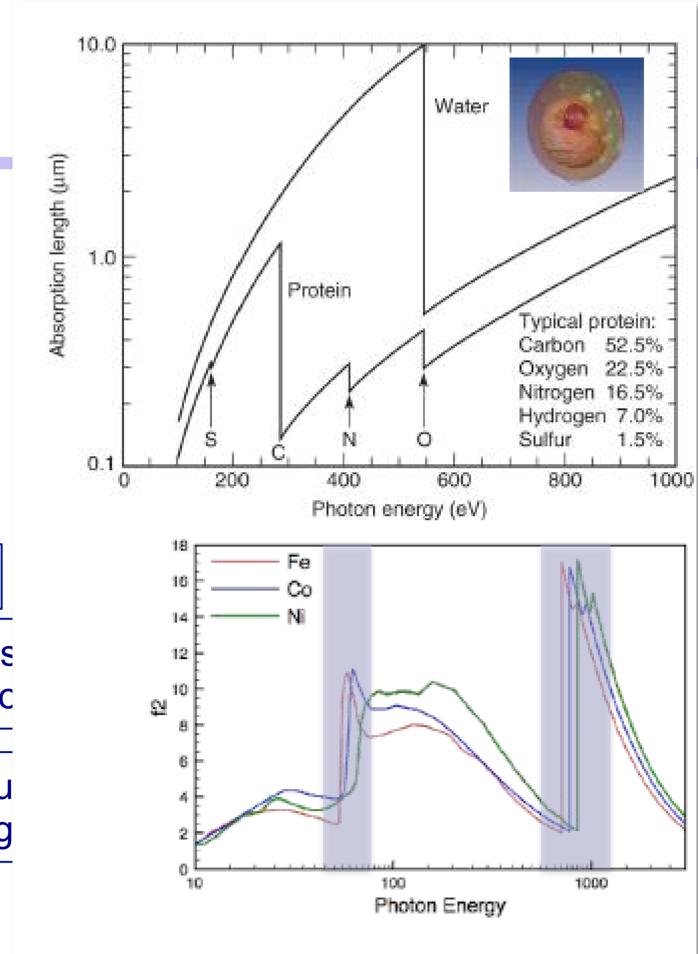




Coherent x-ray tube

The power of x-rays

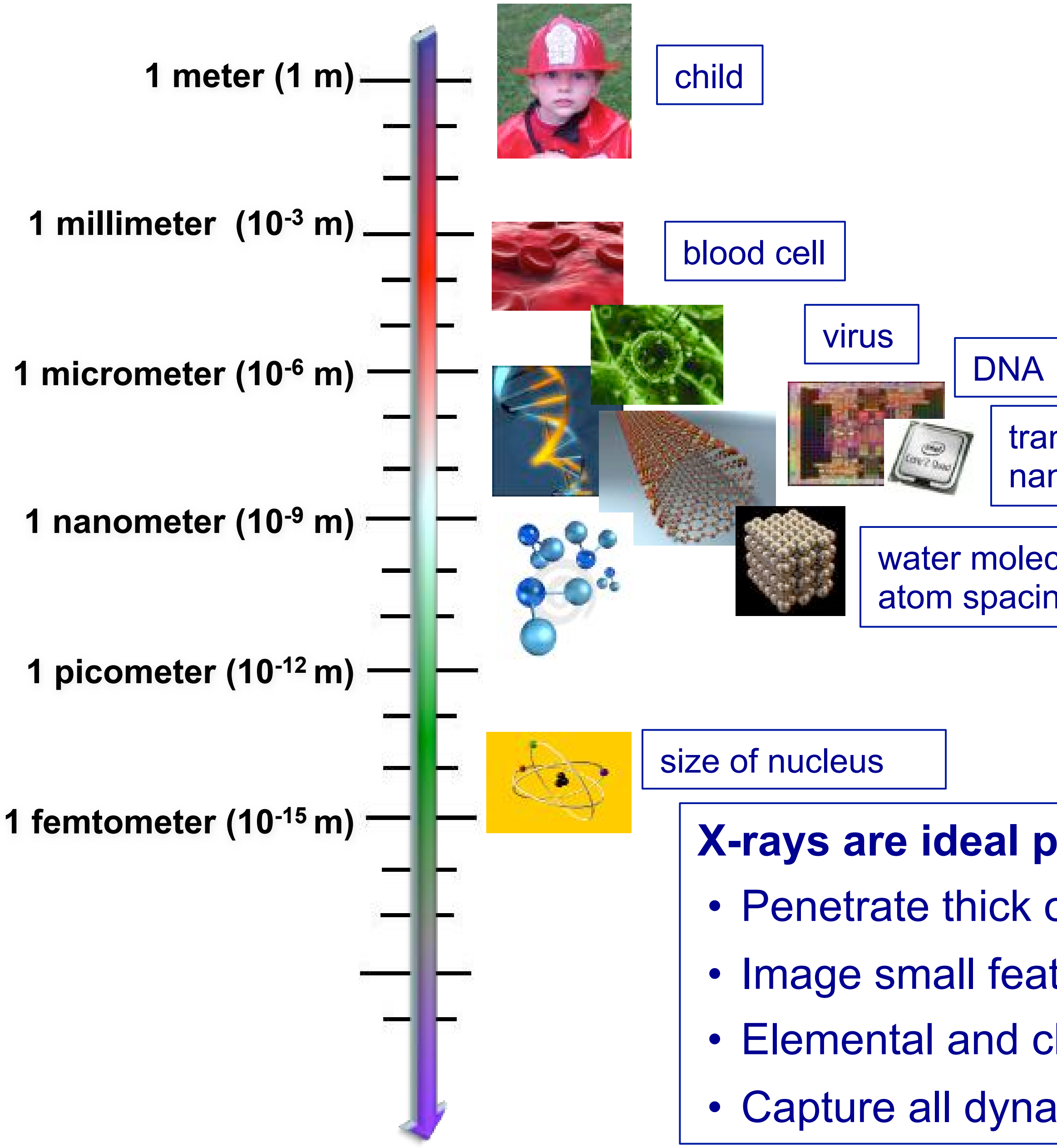


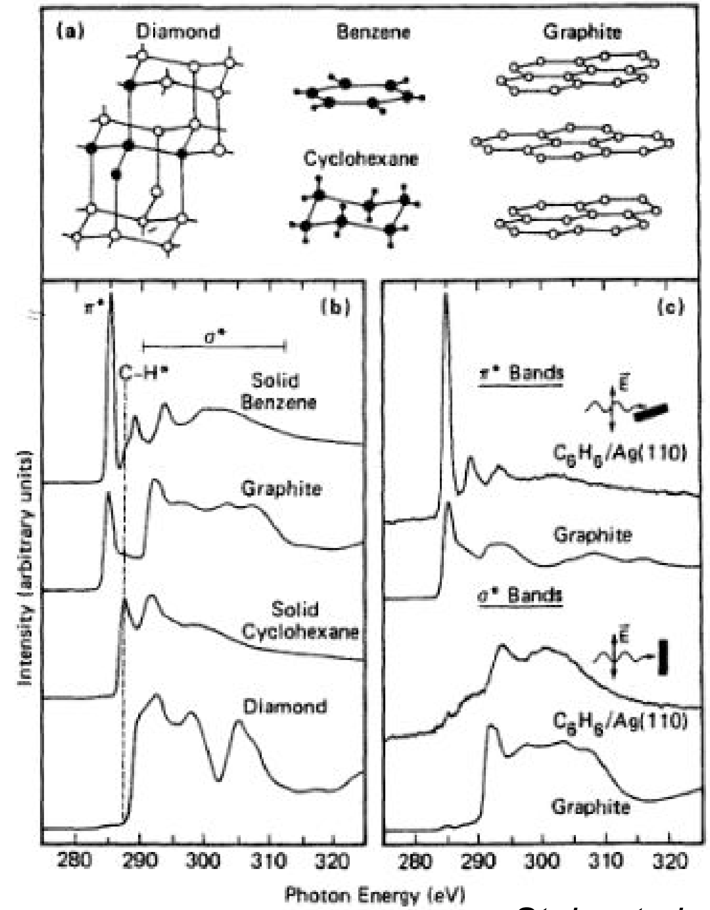


X-rays are ideal probes of the nanoworld:

- Penetrate thick objects
- Image small features
- Elemental and chemical specificity
- Capture all dynamics relevant to function

The power of x-rays



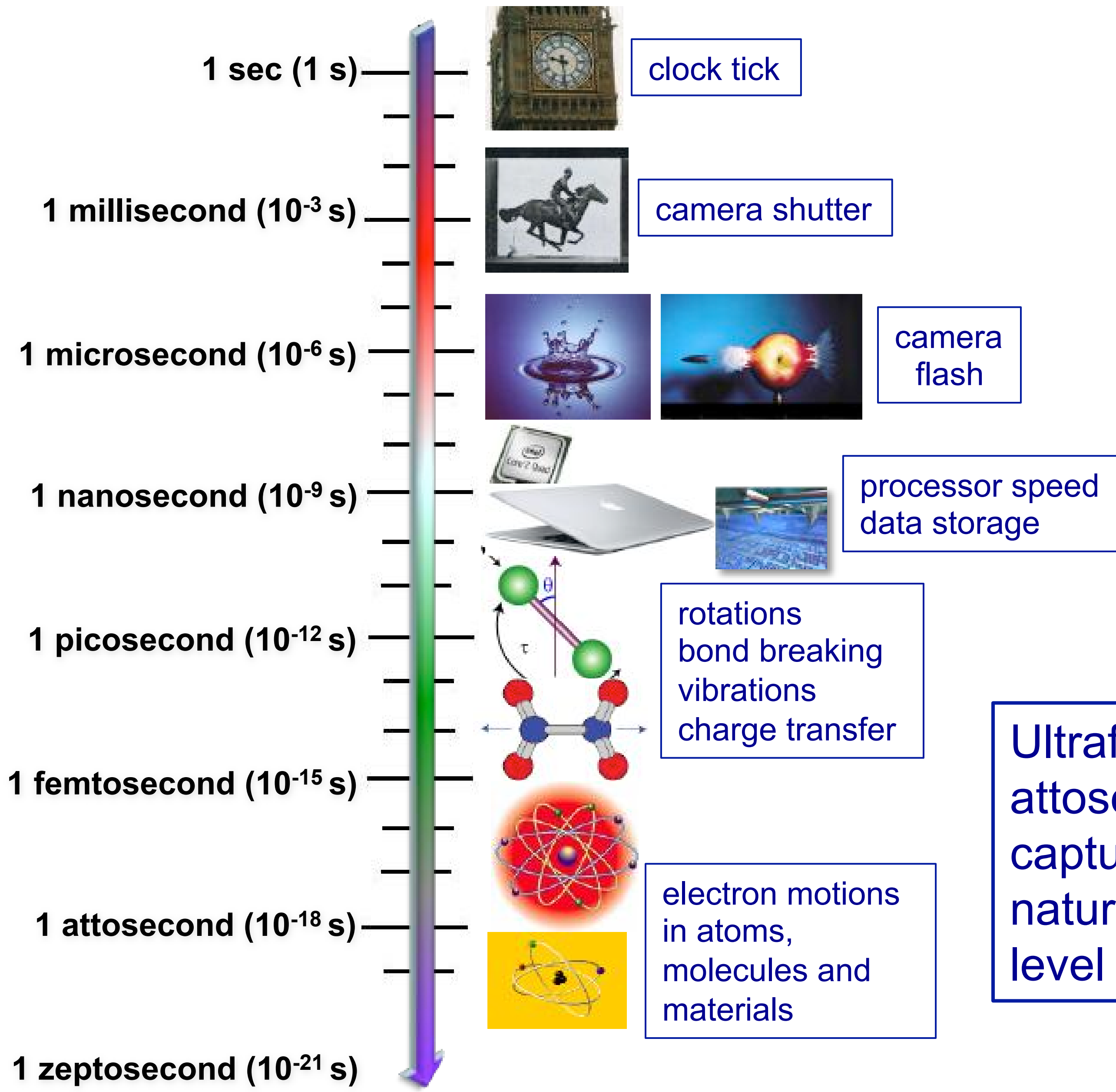


Stohr et al.

X-rays are ideal probes of the nanoworld:

- Penetrate thick objects
- Image small features
- Elemental and chemical specificity
- Capture all dynamics relevant to function

The power of ultrafast x-rays



Ultrafast femtosecond to attosecond x-ray pulses can capture all motion in the natural world, even at the level of electrons

Understanding the fastest processes in nature

- Charge transfer in catalytic/photovoltaic systems – 1 fs and longer
- Phase change in materials – 2 fs and longer
- Ultrafast spintronics – fs and longer
- Control electron-ion motions in chemical reactions – 1 fs and longer
- X-ray induced processes – 50 as and longer
- Strong field physics – zeptoseconds and longer

Electron dynamics

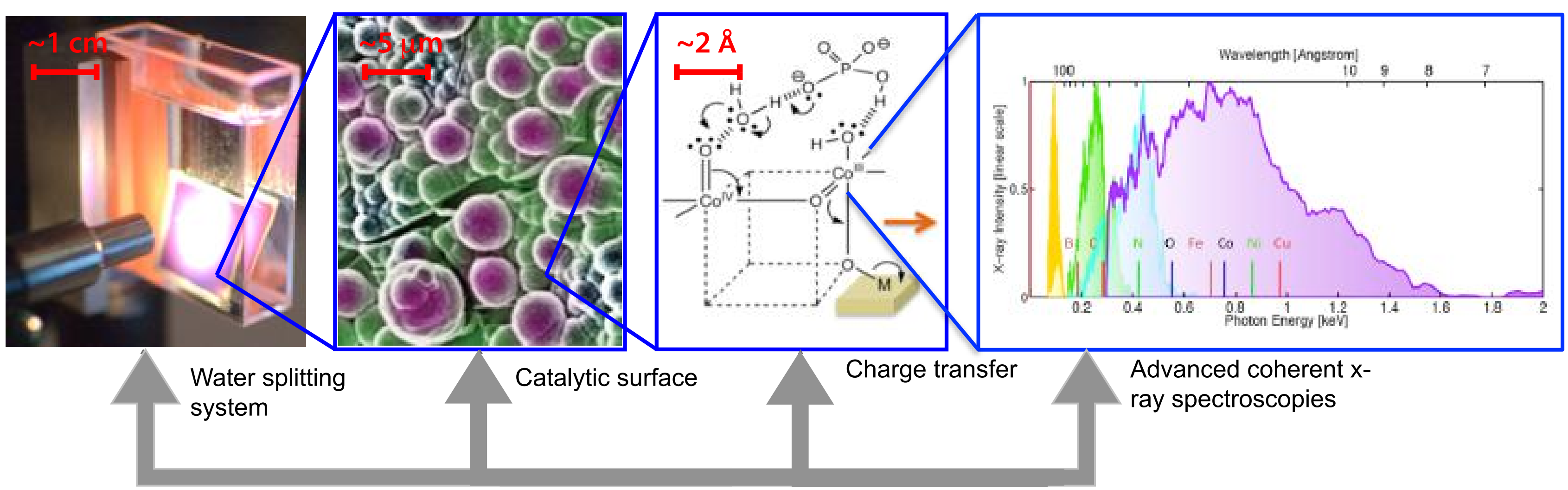


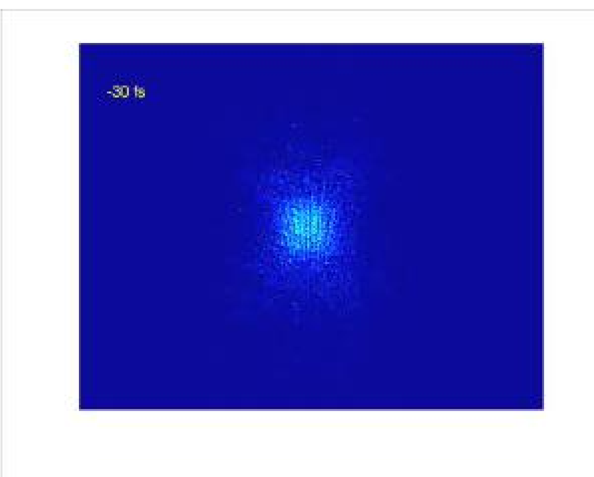
Image charge transfer in complex systems relevant to energy, catalysis using coherent x-ray spectroscopy spanning many elemental absorption edges simultaneously

Understanding the fastest processes in nature

- Charge transfer in catalytic/photovoltaic systems – 1 fs and longer
- Phase change in materials – 2 fs and longer
- Ultrafast spintronics – fs and longer
- Control electron-ion motions in chemical reactions – 1 fs and longer
- X-ray induced processes – 50 as and longer
- Strong field physics – zeptoseconds and longer

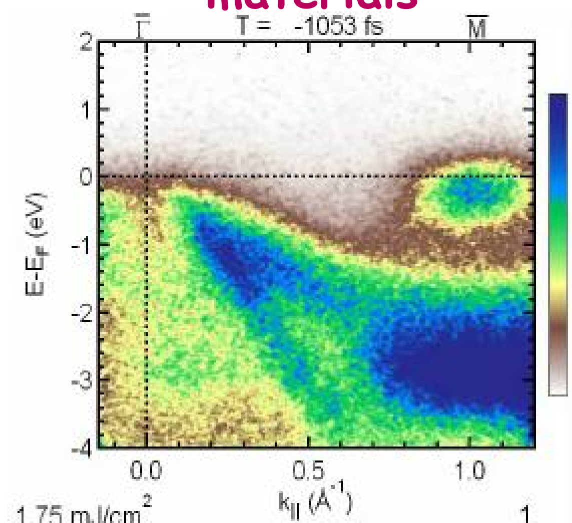
Electron dynamics

Bond breaking in molecules



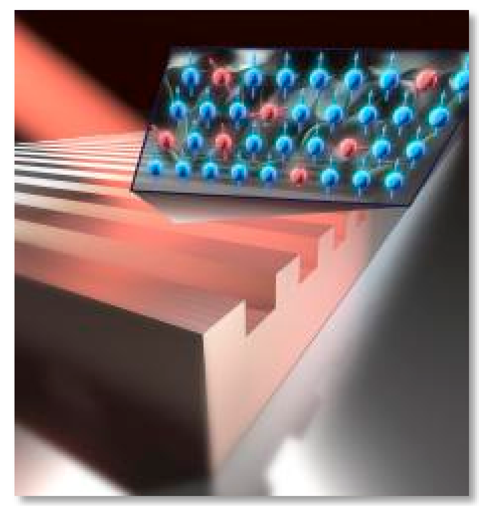
PNAS ***107***, *20219 (2010)*

Phase changes in materials



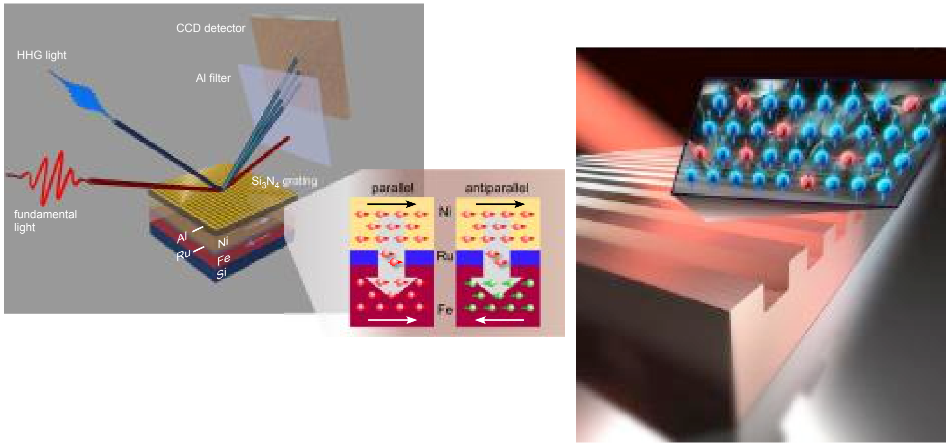
Nature ***471***, *490 (2011)*
Nature Comm ***3***, *1069 (2012)*

Ultrafast spintronics

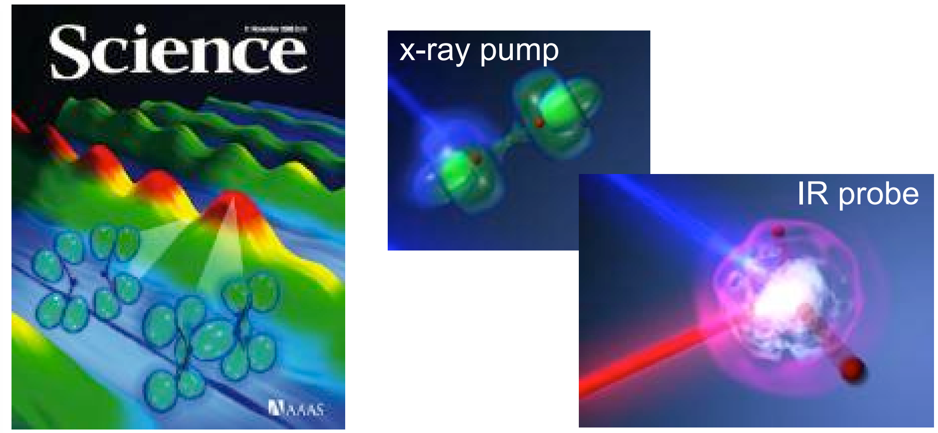


PNAS ***109***, *4792 (2012)*
Nature Comm ***3***, *1037 (2012)*

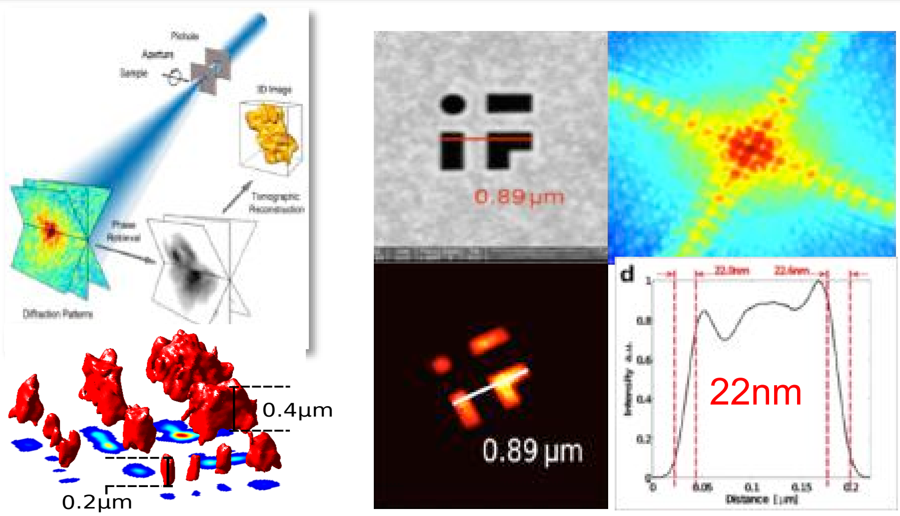
Capturing nanoscale dynamics using high harmonics



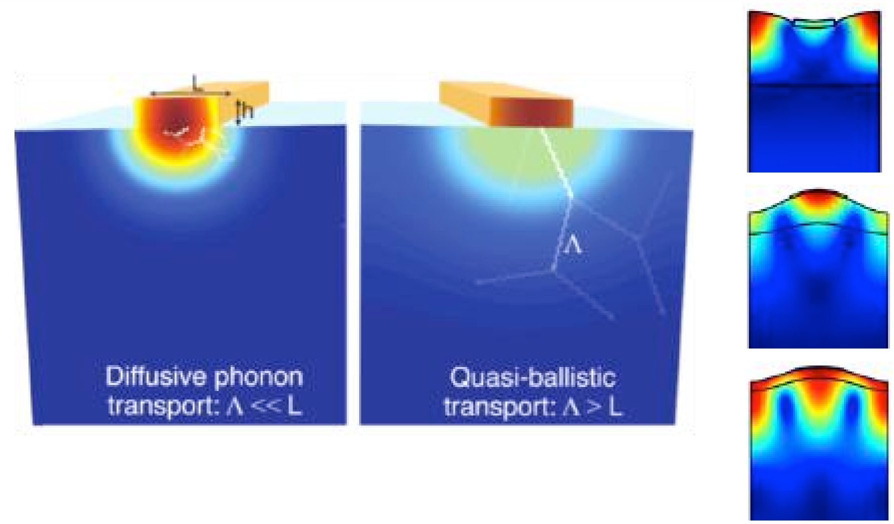
Capture charge-spin-phonon dynamics at multiple sites: *(Nature **471**, 490 (2011), PNAS **109**, 4792 (2012); Nature Comm **3**, 1037 (2012); Nature Comm **3**, 1069 (2012))*



Coupled electron-nuclear dynamics in molecules: (*Science **317**, 1374 (2007), Science **322**, 1081 (2008), Nature Phys. **8**, 232 (2012), PRL **109** , 073004 (2012)*)

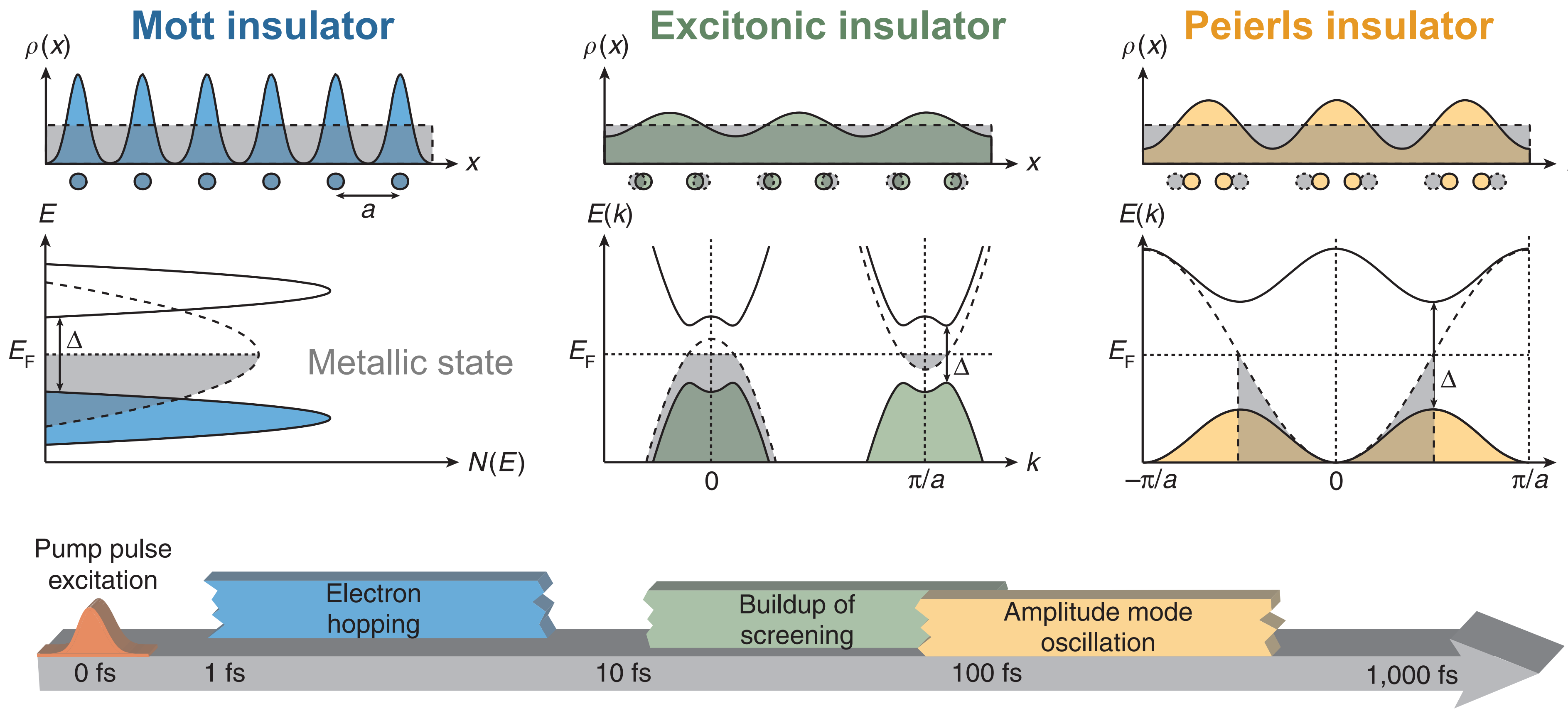


Nanoscale imaging: Record tabletop 22nm resolution *(Op. Ex. **19**, 22470 ('11); **17**, 19050 ('12); Nature **463**, 214 (2010))*



Nanoscale energy transport: probe nanoscale energy/strain flow *(Nature Materials **9**, 26 (2010); Nano Letters **11**, 4126 (2011); PRB **85**, 195431 (2012))*

Understand correlated interactions of charge/phonons/photons

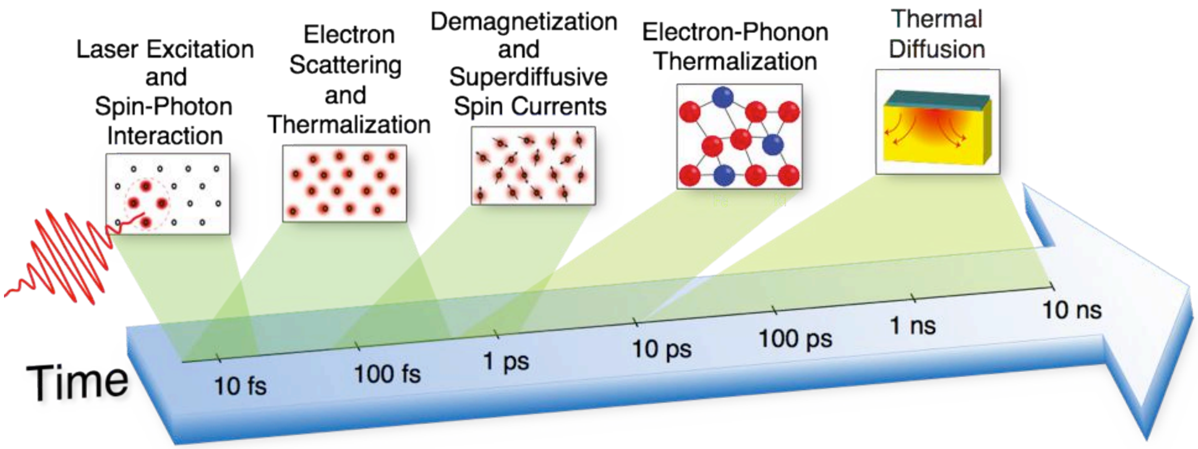


- Separation of timescales allows one to learn about nature of interactions in insulators

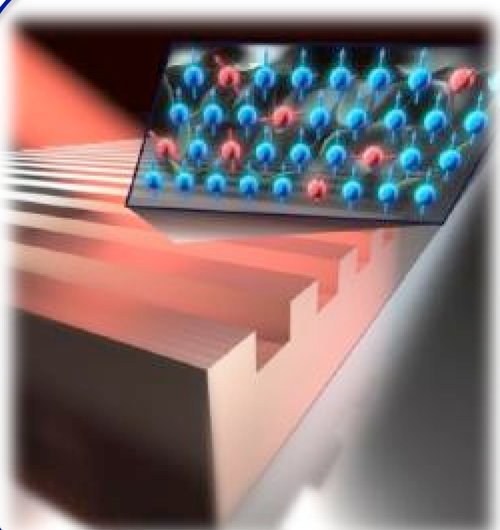
- Measure the melting times of electronic order parameters to identify the dominant interaction in a charge-density-wave material

*Nature **471**, 490 (2011); Nature Comm **3**, 1069 (2012)*

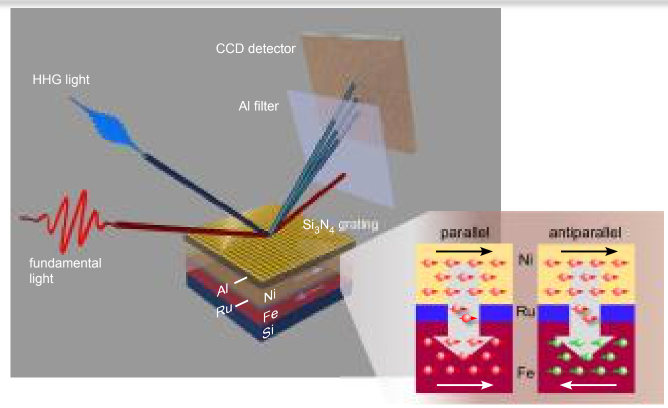
Surprising ultrafast spin dynamics



- No complete microscopic theory of magnetism exists on fs time scales
- High harmonics enable ultrafast, element-specific, spin dynamics to be probed at multiple sites simultaneously



Even in a strongly exchange-coupled Fe-Ni ferromagnetic alloy, the dynamics of the individual spin sub-lattices can be different on timescales faster than that characteristic of the exchange interaction energy (10 – 80 fs)



Large, superdiffusive, spin currents can be launched by a femtosecond laser through magnetic multilayers, to enhance or reduce the magnetization of buried layers, depending on their relative orientation

PUBLICATIONS

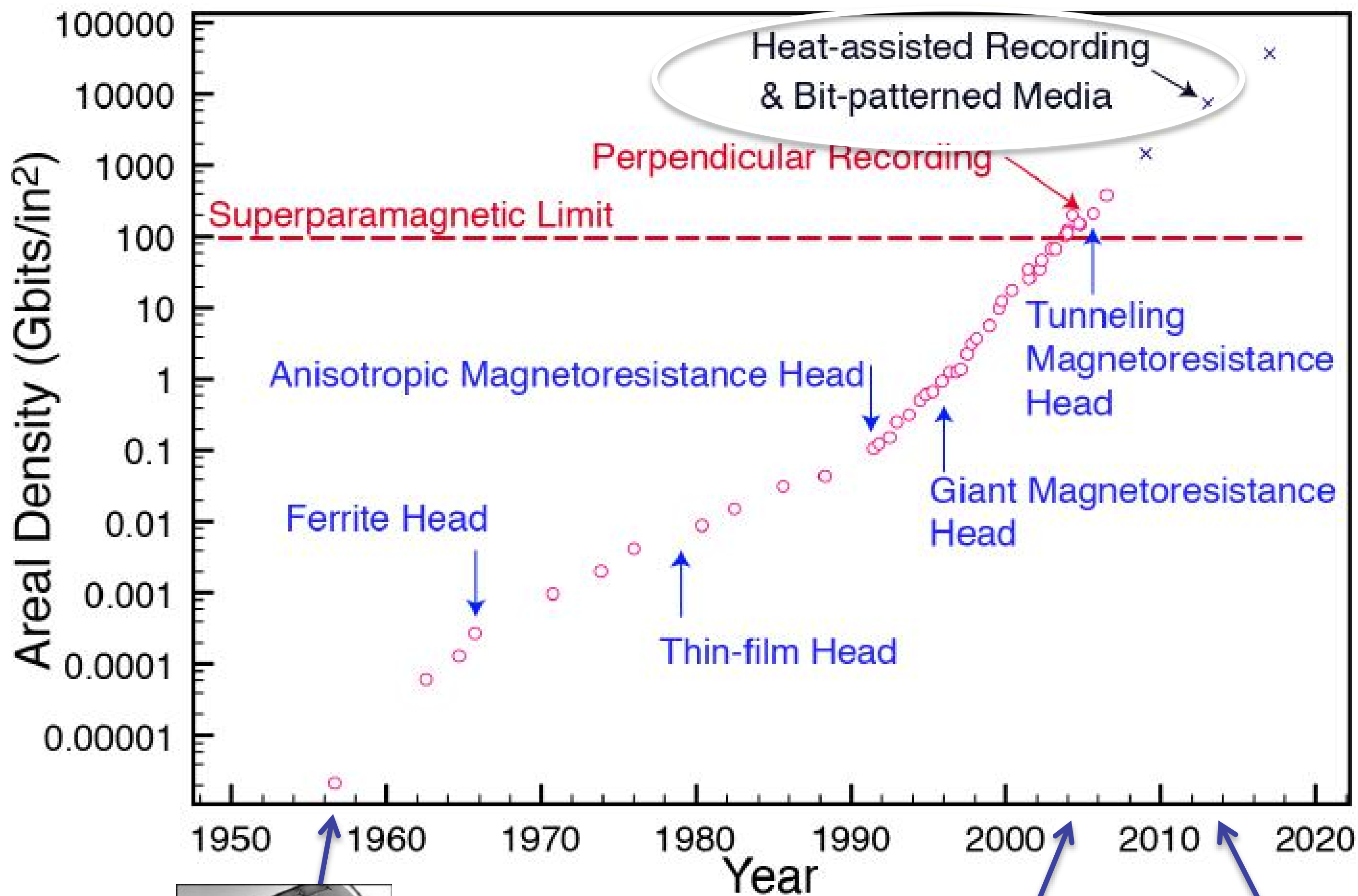
*PRX **2**, 011005 (2012)*
*PNAS, **109**, 4792 (2012)*
*Nature Commun. **3,** 1037 (2012)*

NEWS ARTICLES ABOUT WORK

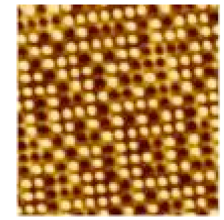
*Physics **5**, 11 (2012)*
*Physics Today **65** (5), 18 (2012)*
*Physik Journal **11**, Nr. 6, page 26 (2012)*

Jülich Forschungszentrum
Technische Universität Kaiserslautern

Exponential growth in data storage – zetabytes/yr!



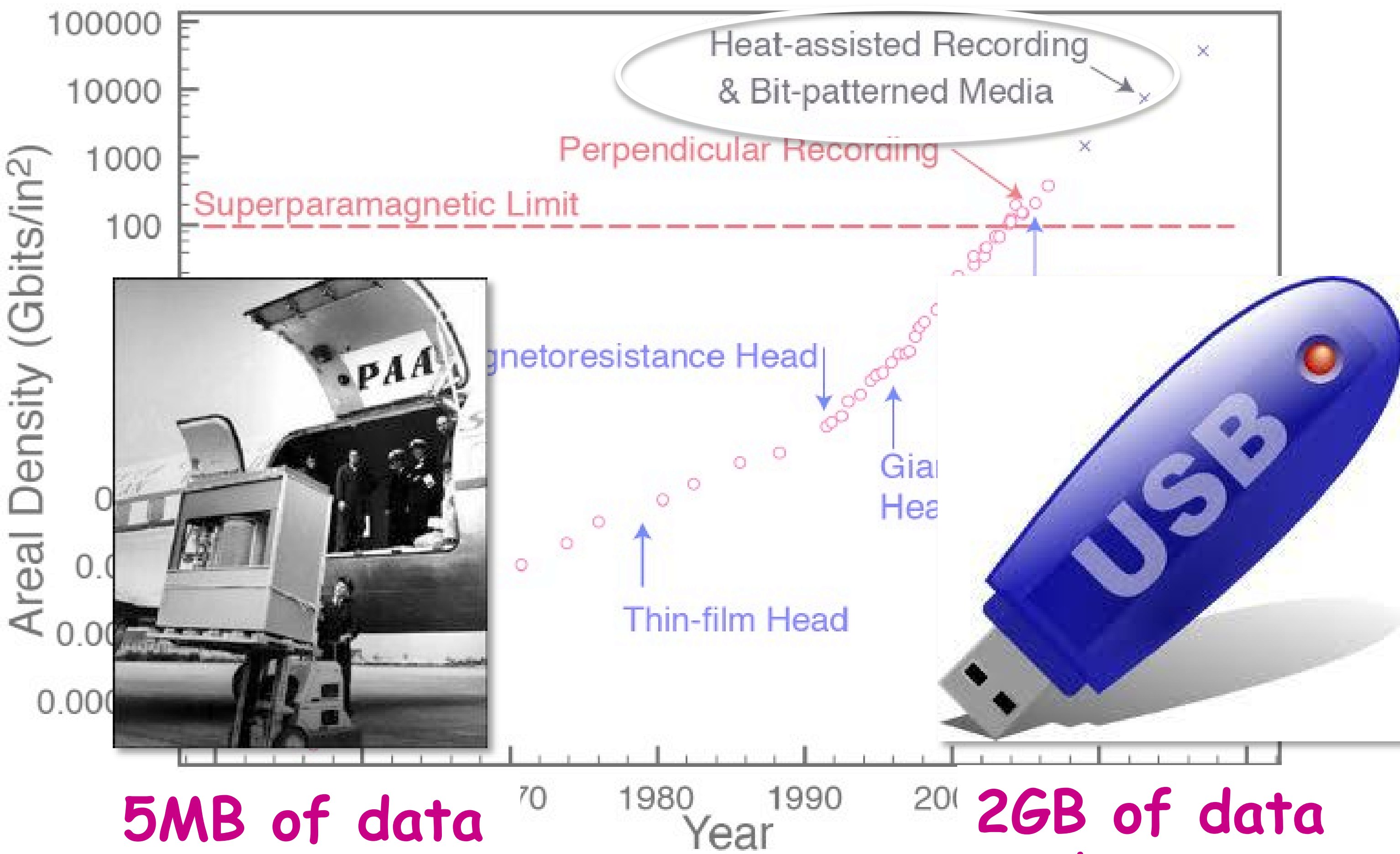
Shaw et al., PRB **78**, 024414 (2008)



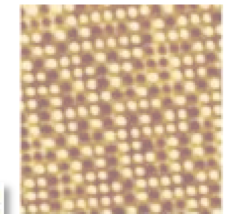
The Nobel Prize in Physics 2007

Albert Fert, Peter Grünberg

Exponential growth in data storage – zetabytes/yr!



Shaw et al., PRB **78**, 024414 (2008)

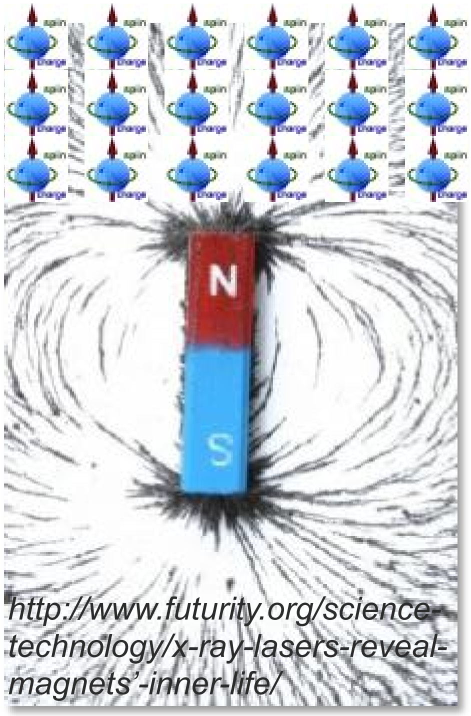


5MB of data at $10,000 a megabyte!

The Nobel Prize in Physics
Albert Fert, Peter Grünberg

2GB of data at $1 a gigabyte!

Ferromagnetism

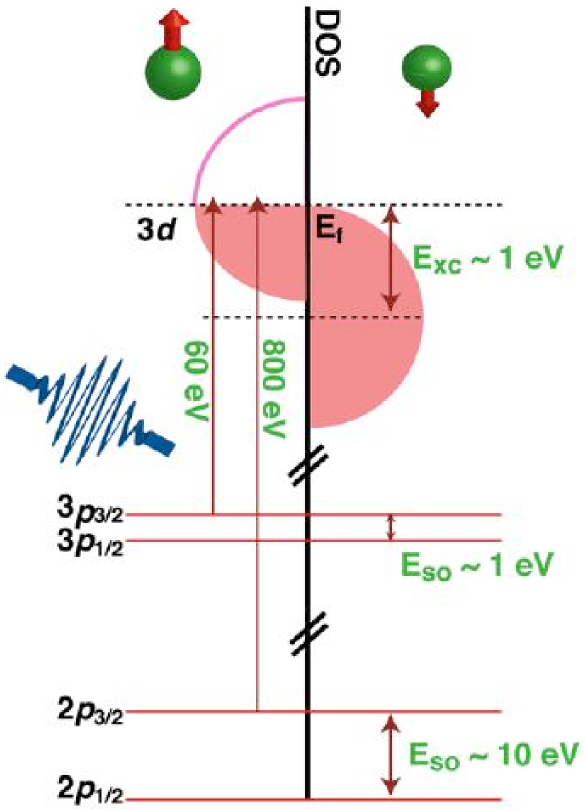


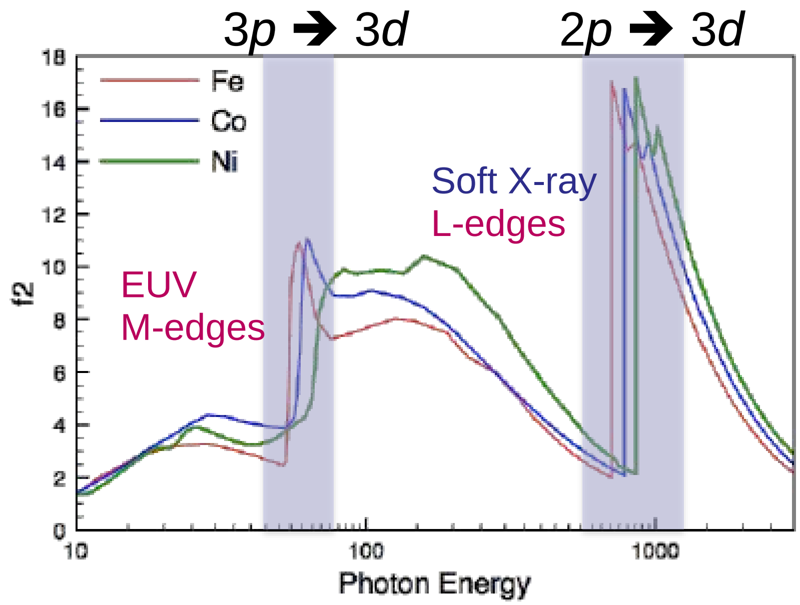
http://www.futurity.org/science-technology/x-ray-lasers-reveal-magnets'-inner-life/

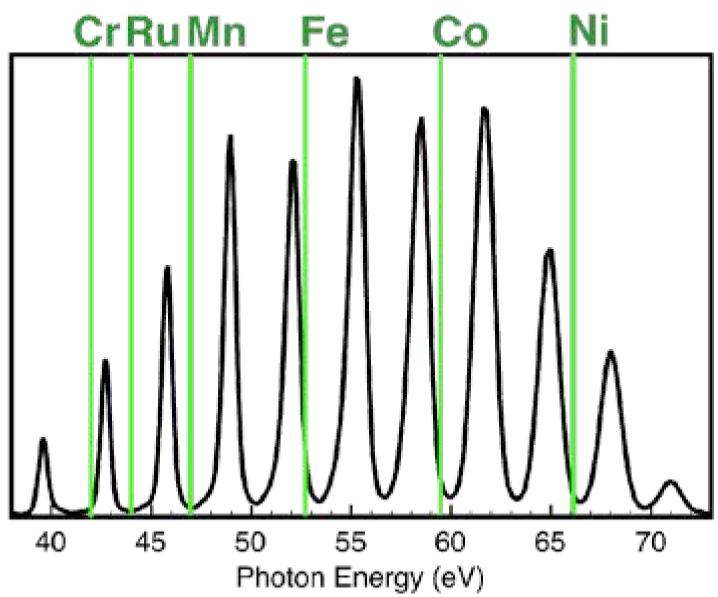
- Magnetism exists because all of the "spins" in a magnet line up to point in same direction due to exchange interaction
- Generally, metals are complex because collection of mobile electrons interacting one another - many body problem without complete theoretical model

m↓l	-2	-1	0	1	2	S	m_{exp}
$_{26}$**Fe,**	↑↓	↓	↓	↓	↓	2	2.216
$_{27}$**Co,**	↑↓	↑↓	↓	↓	↓	3/2	1.715
$_{28}$**Ni,**	↑↓	↑↓	↑↓	↓	↓	1	0.616

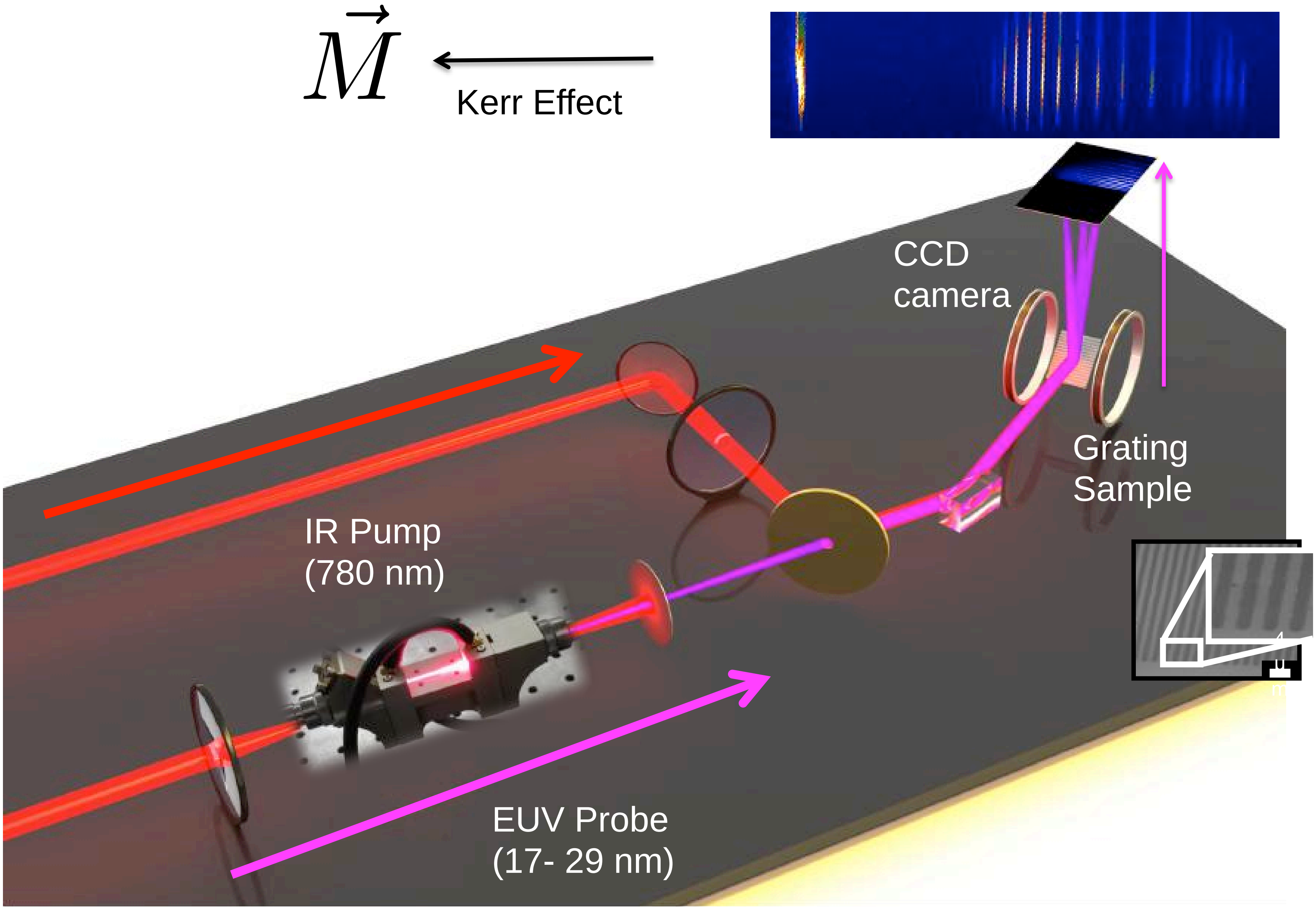
How can we measure the magnetic state?



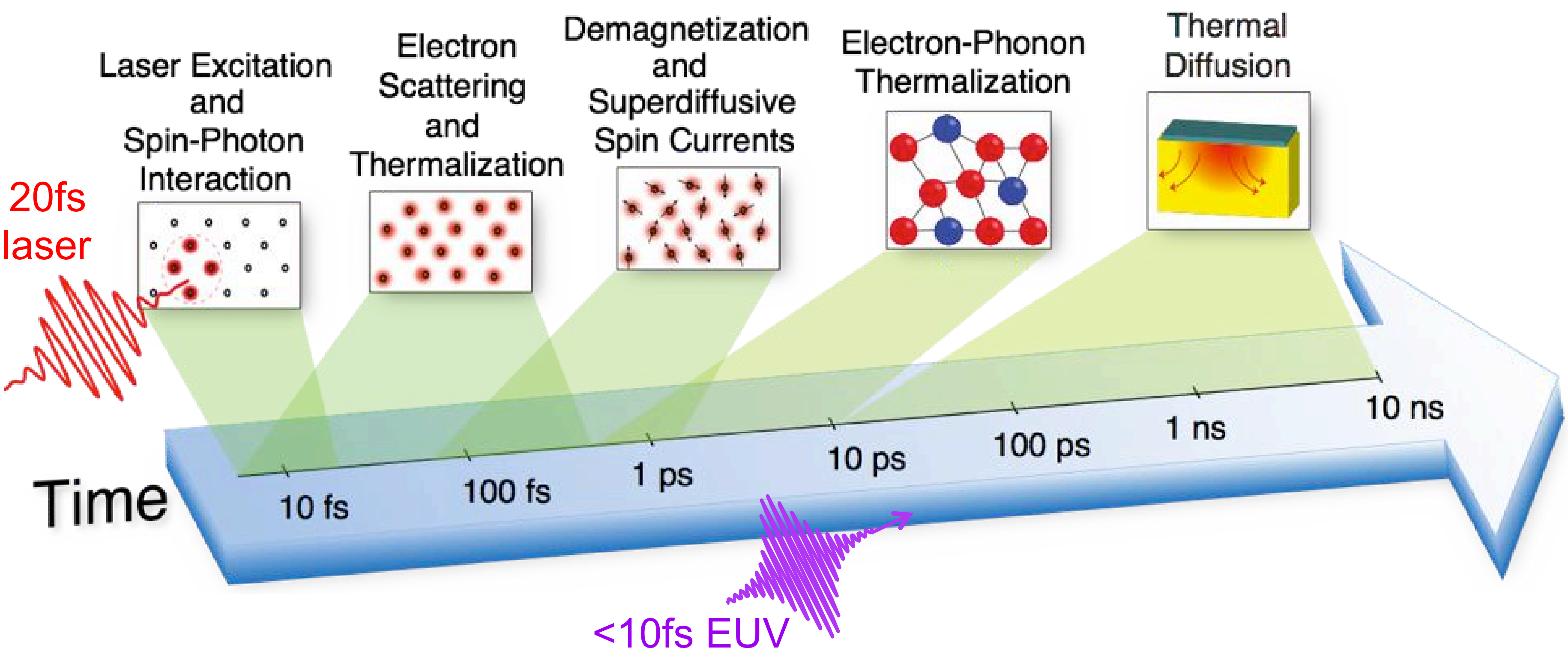




Experimental setup

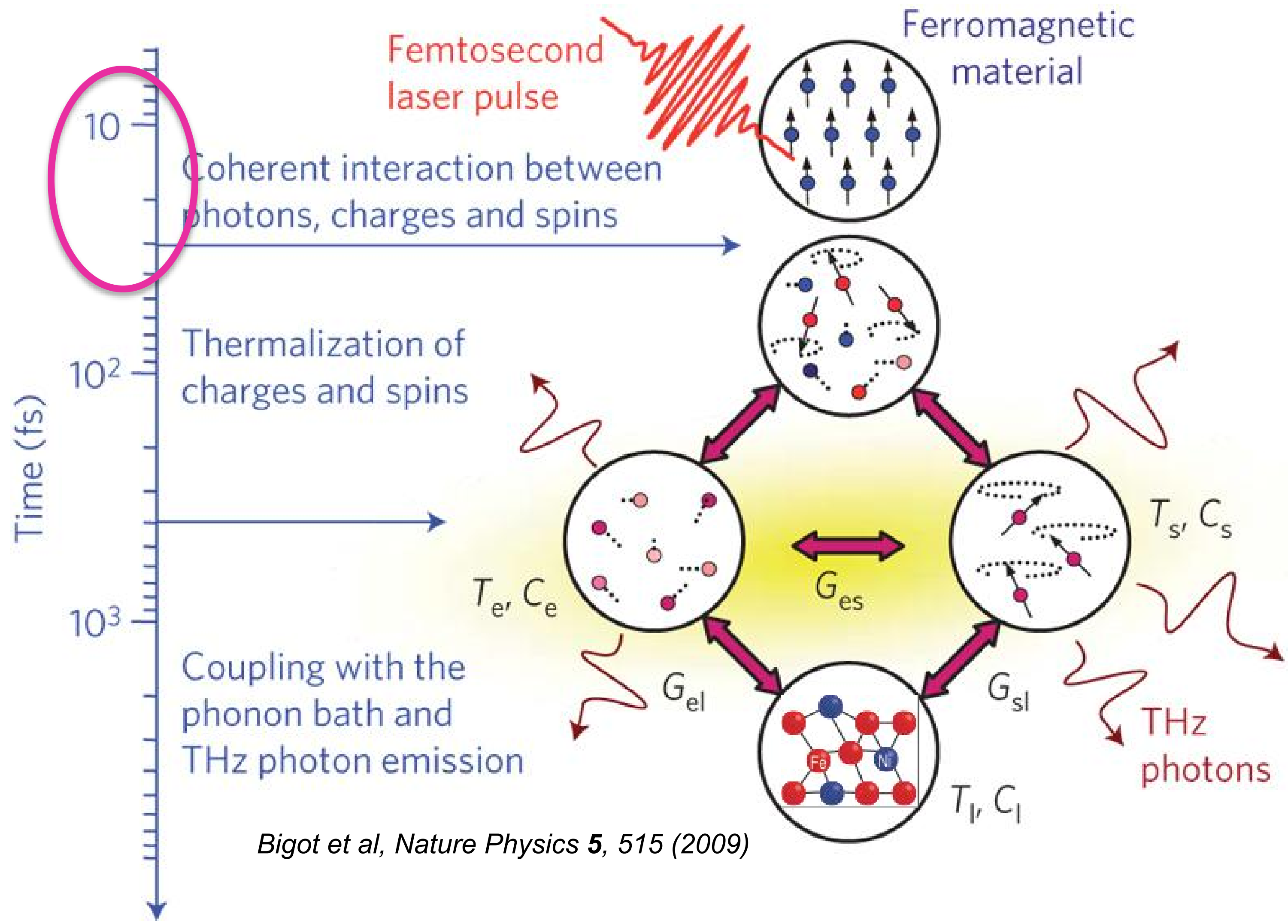


No complete microscopic theory of magnetism exists on fs time scales



- Excite electrons in material using 20fs 800nm pulse
- Probe dynamics using sub-10fs high harmonics
- Capture element-specific, spin dynamics at multiple sites simultaneously
- Separation of timescales allows one to learn about interactions and nature of magnetism on the fastest timescales

Coupled dynamics of spins/charge/photons dominate on < 100fs times



Bigot et al, Nature Physics ***5****, 515 (2009)*

Nature of magnetic signal under hot debate………

additional temperature dependence of χ_m due to a (possibly) reduced magnetization. While there is some evidence that the magnetization can be reduced within a few ps, on the time scale below ≈2 ps a thermodynamic description fails and therefore a direct determination of the magnetization from Kerr measurements is not possible in the fs time scale.

H. Regensburger et al, PRL 61, 14 716, 2000

nature physics

LETTERS

PUBLISHED ONLINE: 14 JUNE 2009 | DOI: 10.1038/NPHYS1315

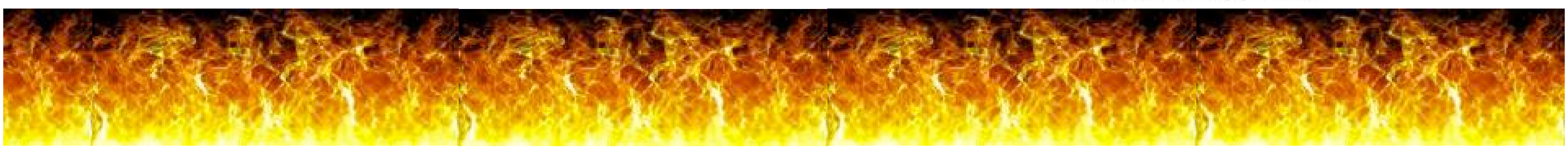
Paradigm of the time-resolved magneto-optical Kerr effect for femtosecond magnetism

G. P. Zhang[1*], W. Hübner[2], Georgios Lefkidis[2], Yihua Bai[3] and Thomas F. George[4]

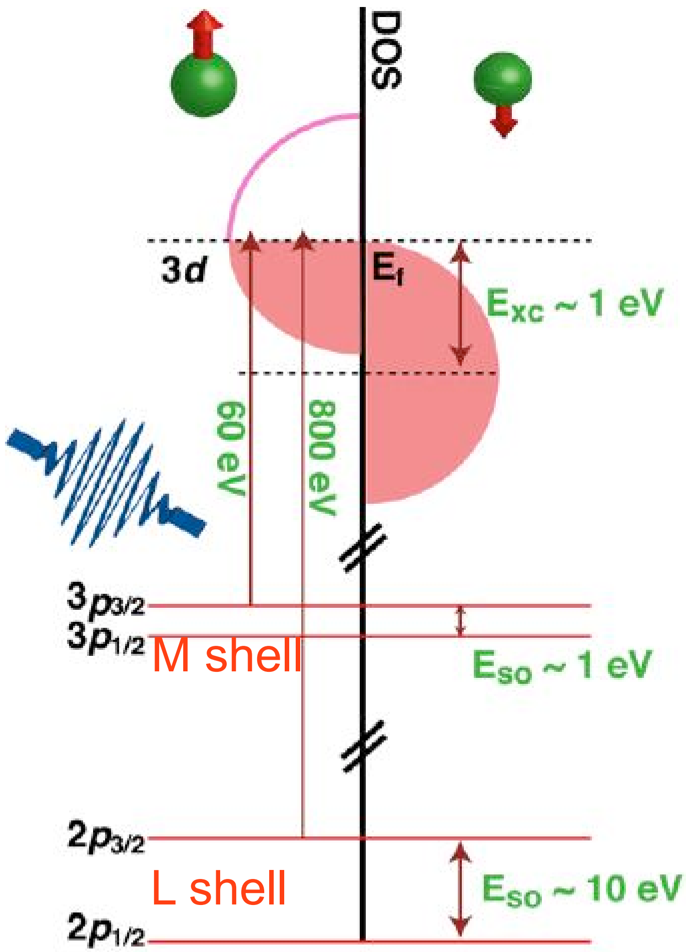
correspondence

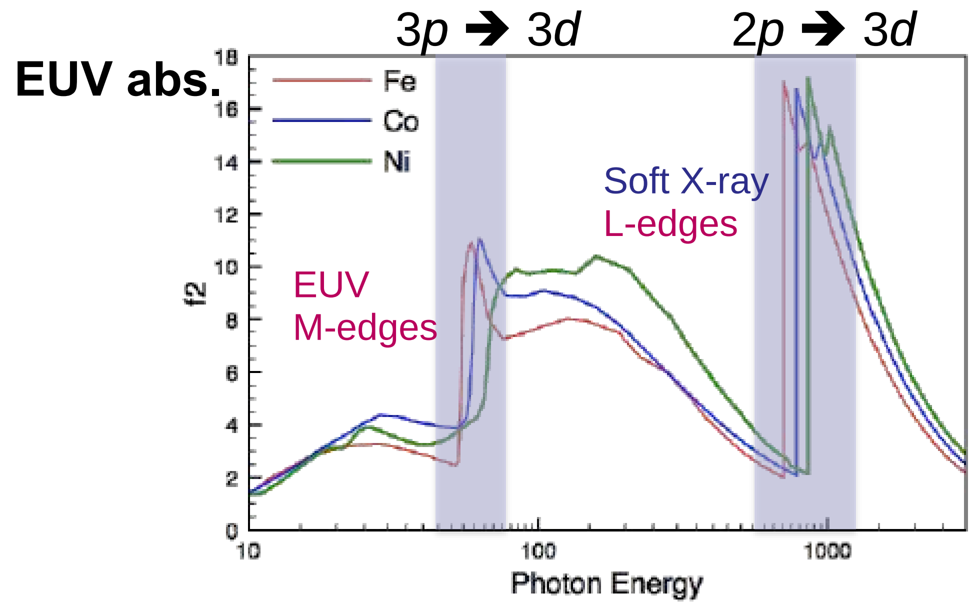
Is the controversy over femtosecond magneto-optics really solved?

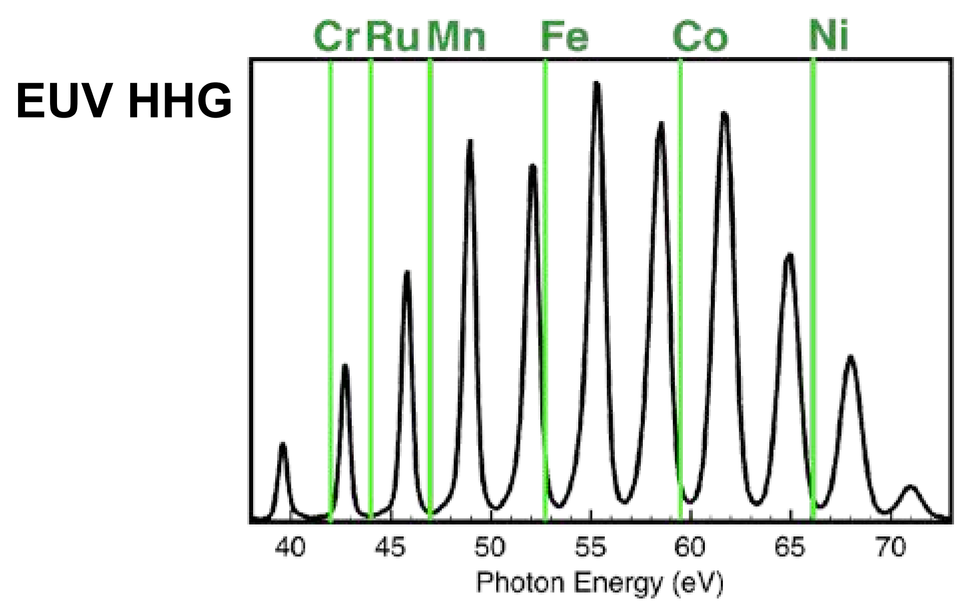
Karel Carva[1,2*], Marco Battiato[1] and Peter M. Oppeneer[1]



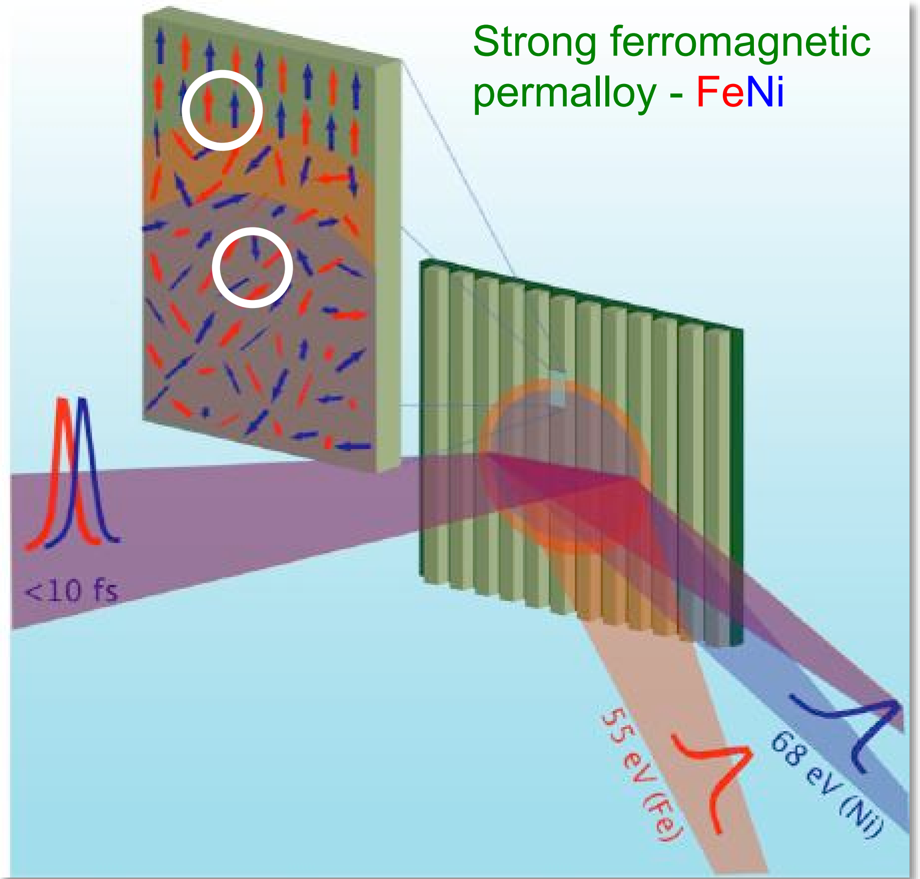
Measuring the magnetic state using EUV-MOKE

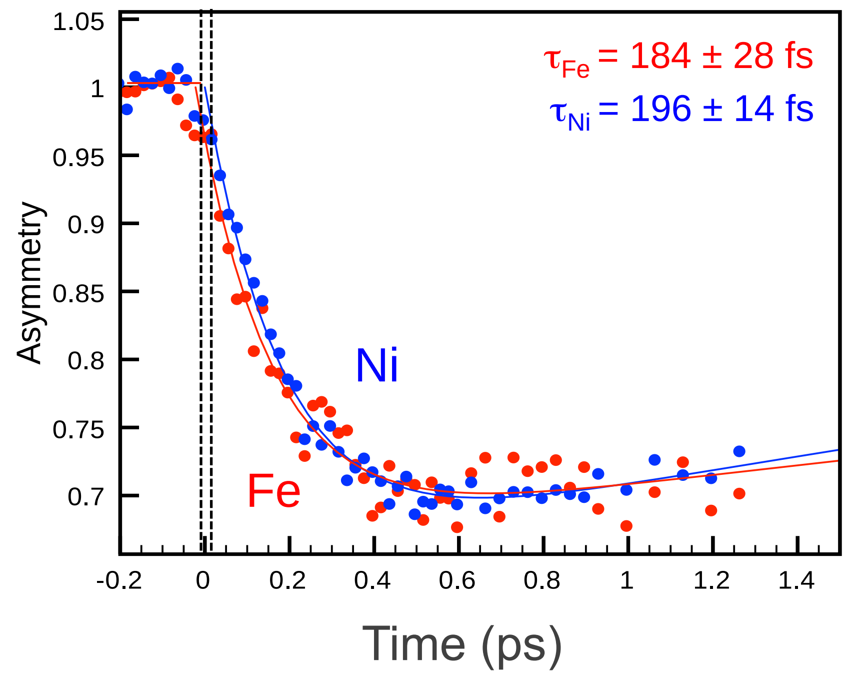






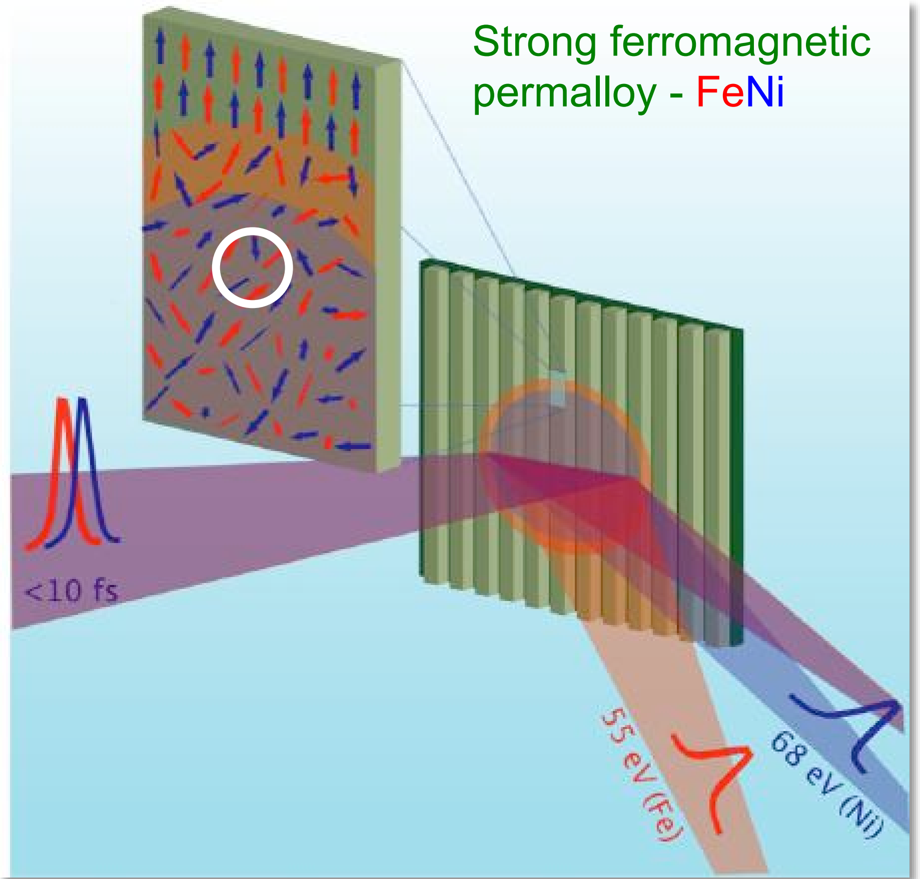
How fast can we destroy the magnetic state?

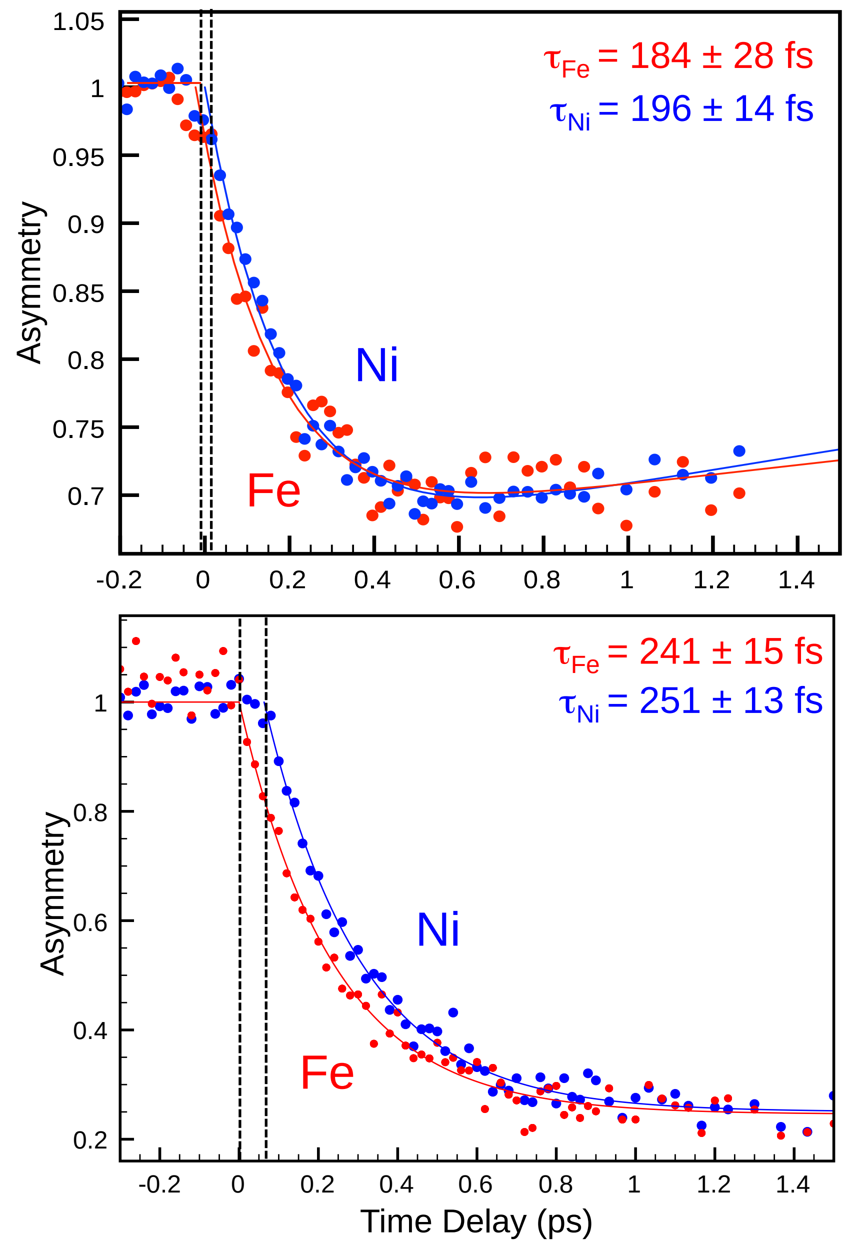




*PNAS, **109**, 4792 (2012); Physics Today **65** (5), 18 (2012); Physik Journal **11**, Nr. 6, page 26 (2012)*

How fast can we destroy the magnetic state?

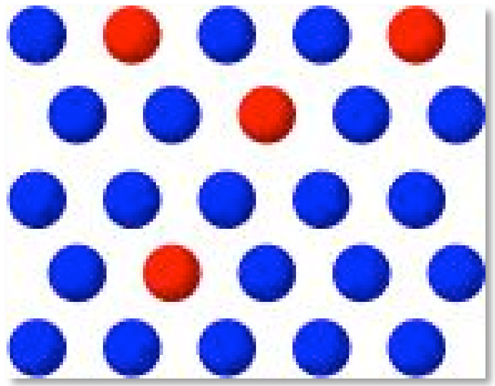


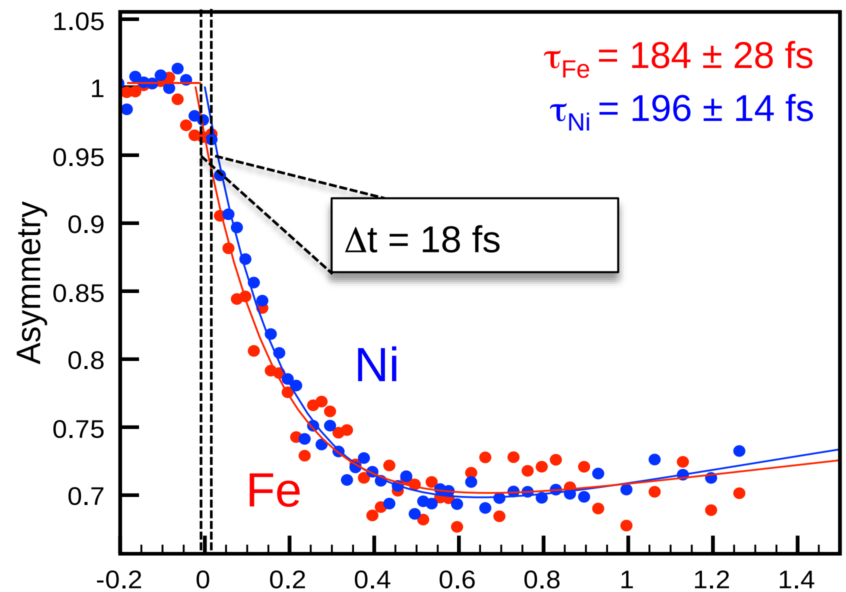


PNAS, ***109****, 4792 (2012); Physics Today* ***65*** *(5), 18 (2012);*
Physik Journal ***11****, Nr. 6, page 26 (2012)*

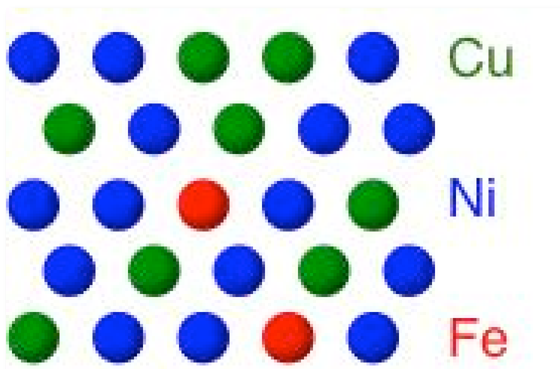
How fast can we destroy the magnetic state?

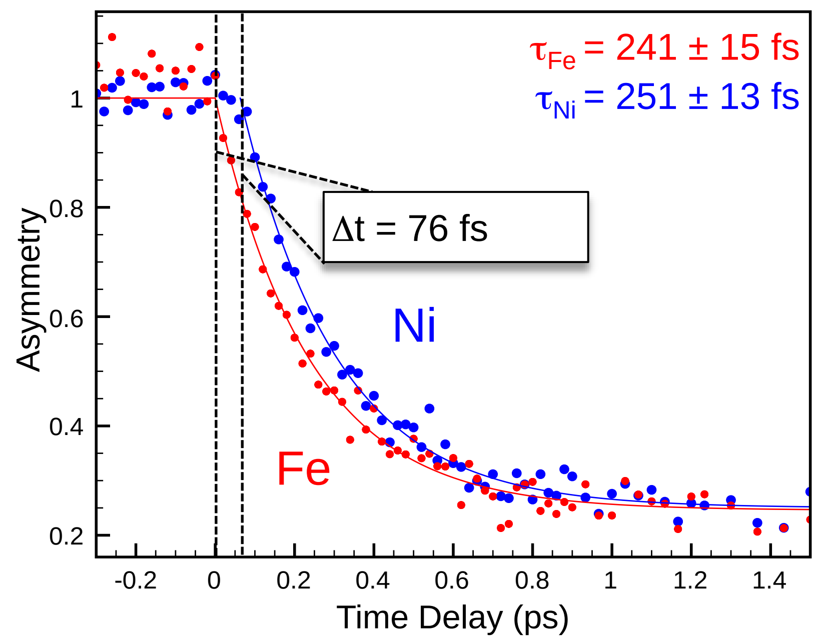
Permalloy
Strong
Exchange
T_c = 850K





Cu-doped Permalloy
Weak
Exchange
T_c = 400K





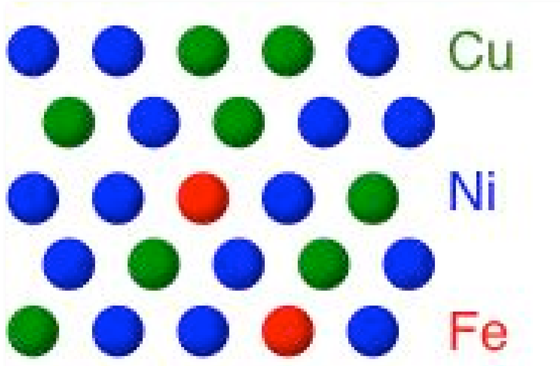
*PNAS, **109**, 4792 (2012); Physics Today **65** (5), 18 (2012); Physik Journal **11**, Nr. 6, page 26 (2012)*

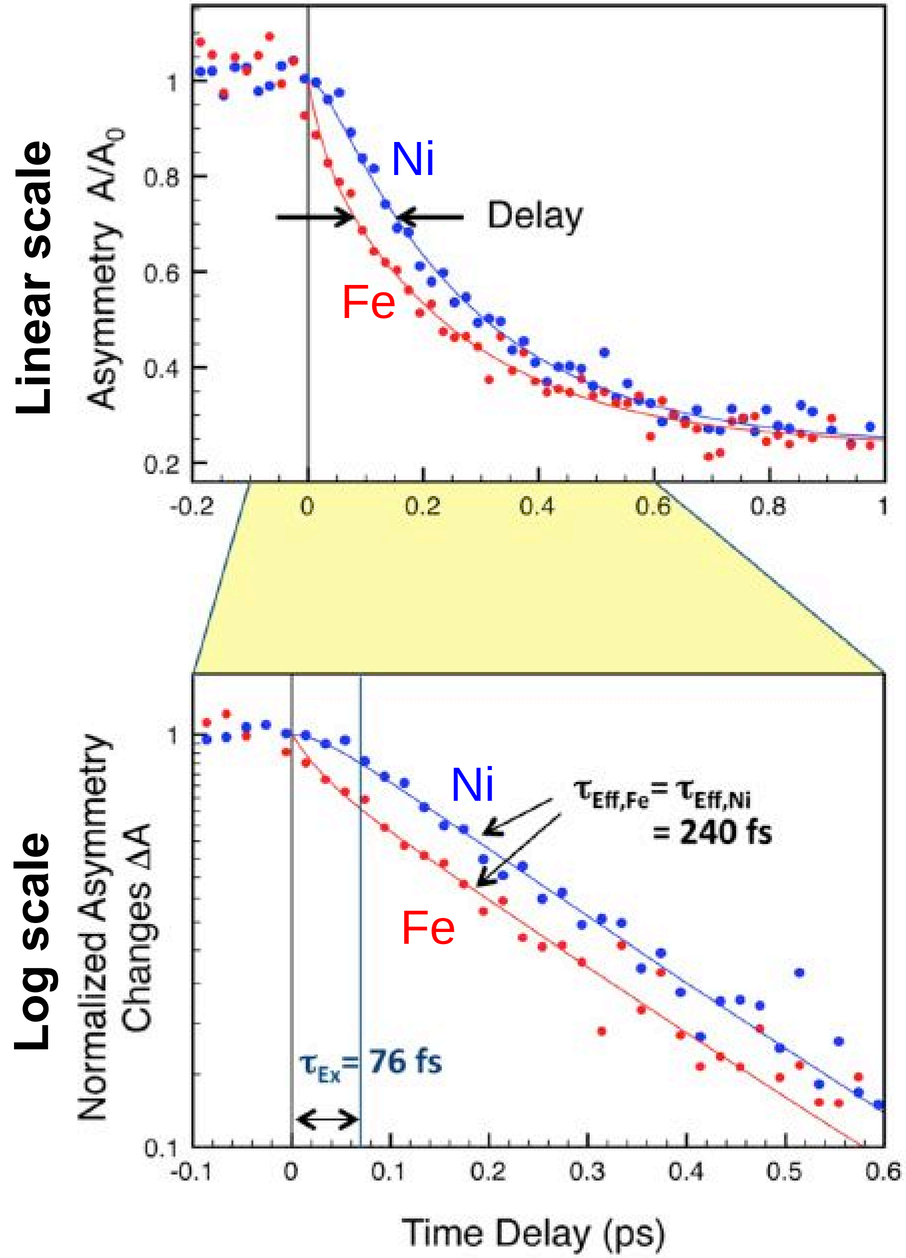
Characteristic **time lag** for ferromagnetic coupling to re-establish

Cu-doped Permalloy

Weak Exchange

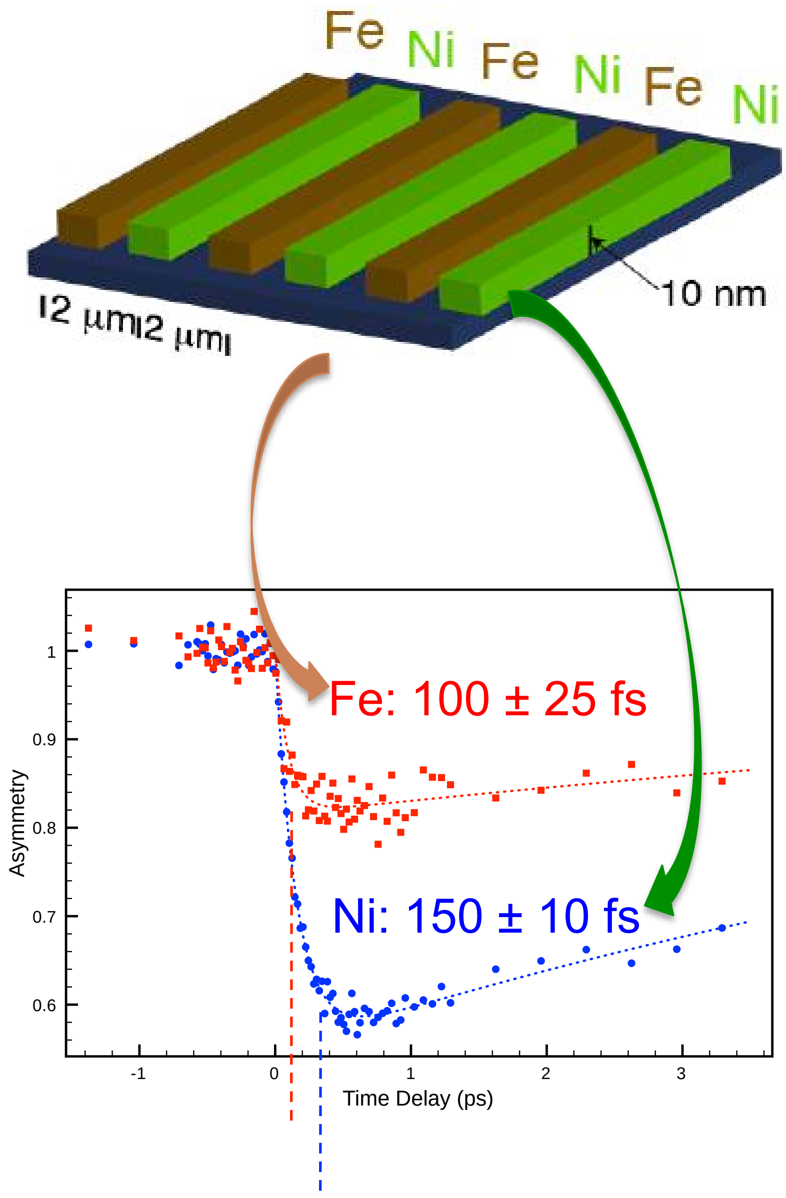
T_c = 400K





*PNAS, **109**, 4792 (2012); Physics Today **65** (5), 18 (2012); Physik Journal **11**, Nr. 6, page 26 (2012)*

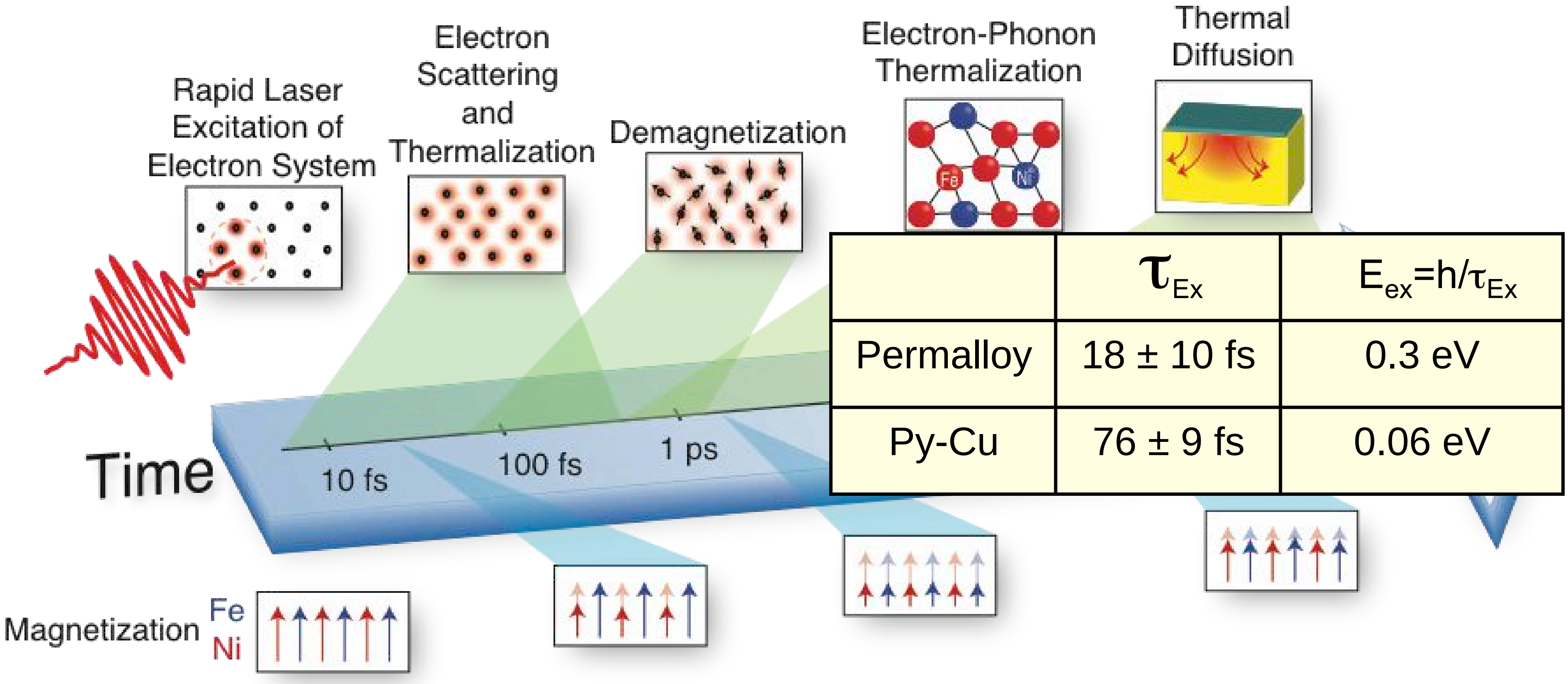
Demagnetization timescales different for pure Fe and Ni



- Measure characteristic demagnetization times for elemental Fe and Ni accurately for first time
- Fe demagnetizes faster than Ni since nanoscale spin-flip scattering processes are faster!
- In the alloy, since the de-magnetization timescales are the **SAME** after a characteristic time lag, we can observe how the quantum exchange interaction influences dynamics

Phys. Rev. Lett. ***103****, 257402 (2009); PRX* ***2****, 011005 (2012) ; Physics* ***5****, 11 (2012); PNAS,* ***109****, 4792 (2012)*

Extracting the exchange interaction timescale

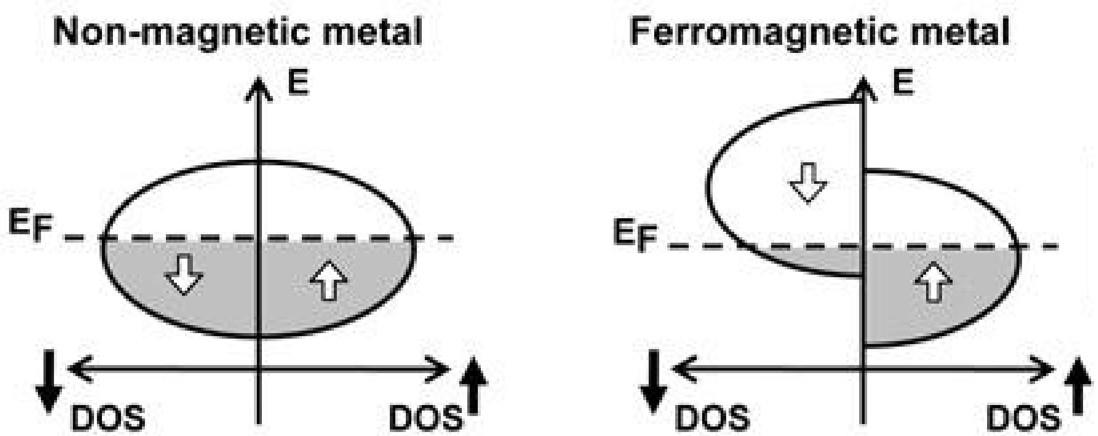


	τ_{Ex}	$E_{ex}=h/\tau_{Ex}$
Permalloy	18 ± 10 fs	0.3 eV
Py-Cu	76 ± 9 fs	0.06 eV

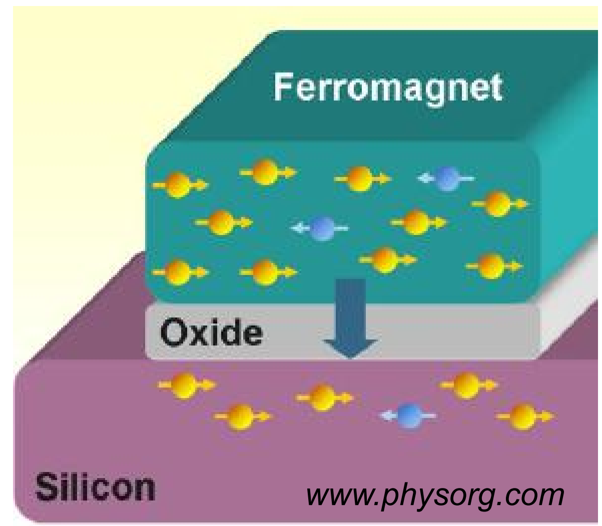
- On timescales shorter than the quantum exchange interaction time, Fe spins randomize faster than Ni spins due to faster spin-flip scattering
- Quantum exchange interaction means that Fe spins will drag the Ni spins after some time lag corresponding to “exchange time” τ_{ex}

Mathias et al., PNAS, ***109****, 4792 (2012)*
Physics Today, May 2012; Physik Journal 26 (2012)

Spintronics – spin electronics - for efficient nanoelectronics

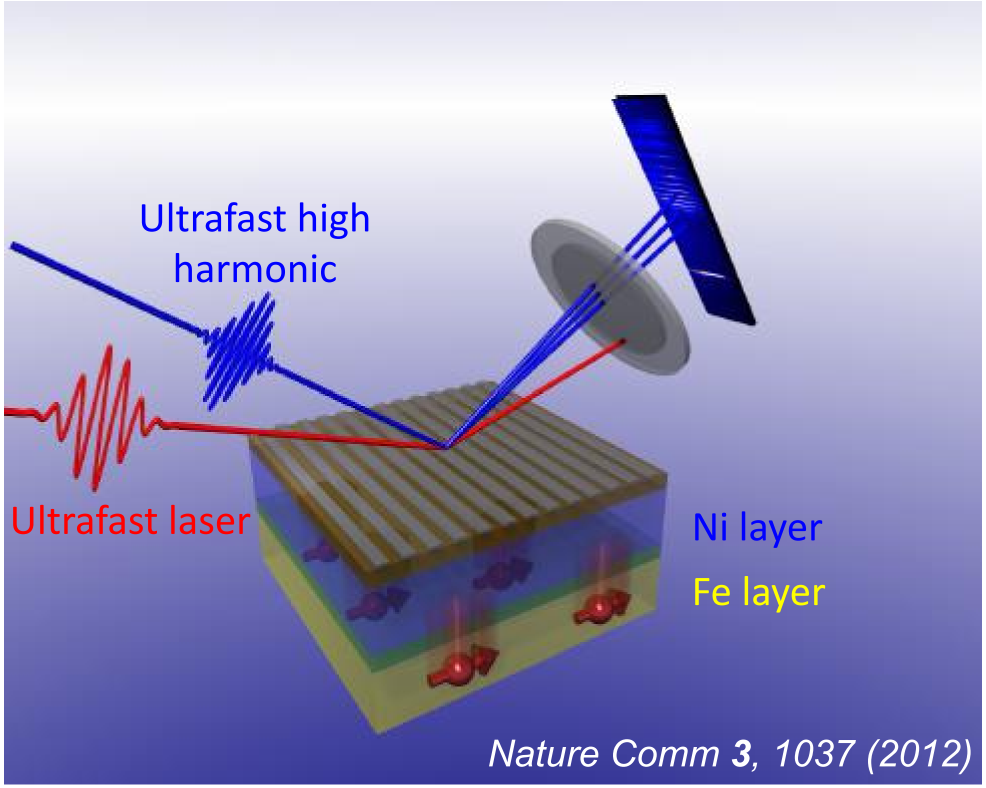


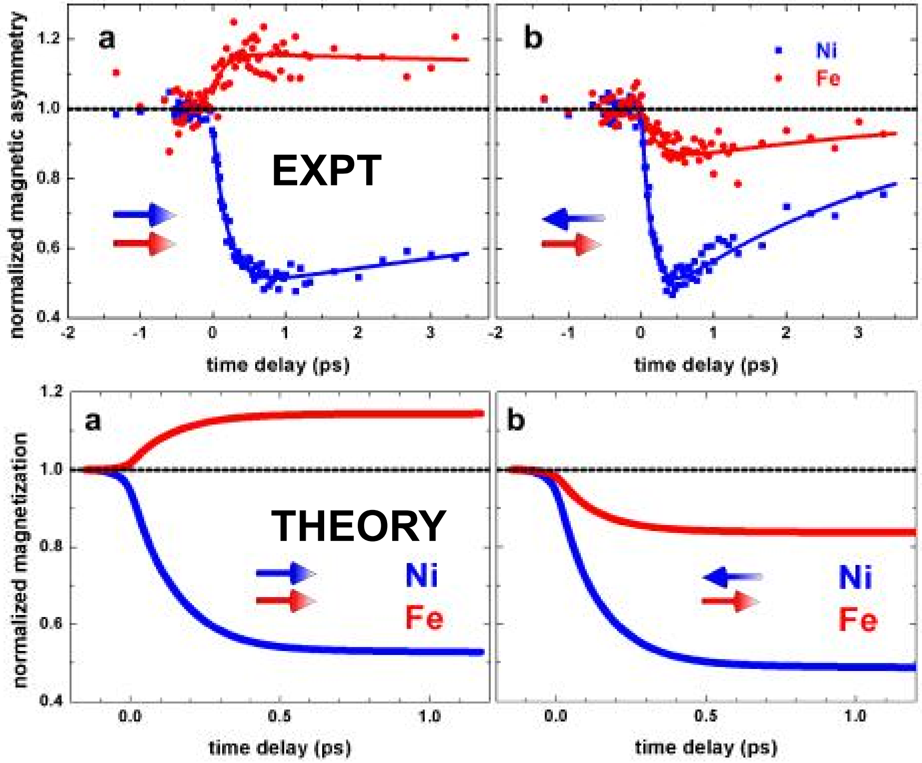
http://asdn.net/asdn/electronics/spintronics.shtml



- Energy dissipation from electrical current is a major roadblock in nanoelectronics
- Encoding data in electron spin, rather than charge, may reduce energy requirements
- Most transport properties depend on the density of states near the Fermi level
- Spin asymmetry in the density of states allows ferromagnets to generate, manipulate, and detect spin

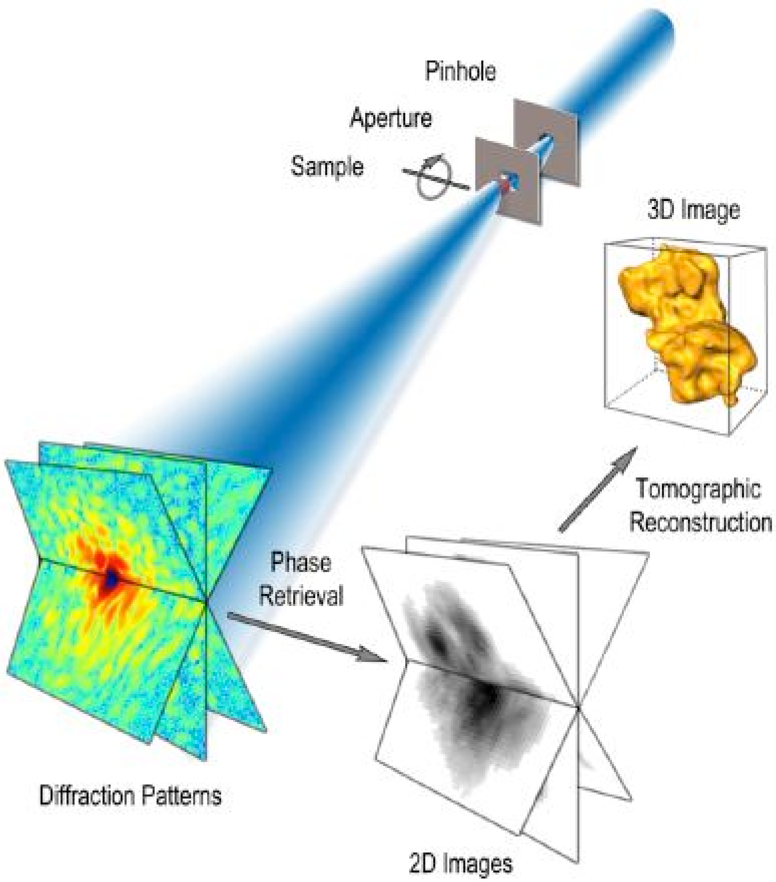
Ultrafast spin transport can enhance magnetization



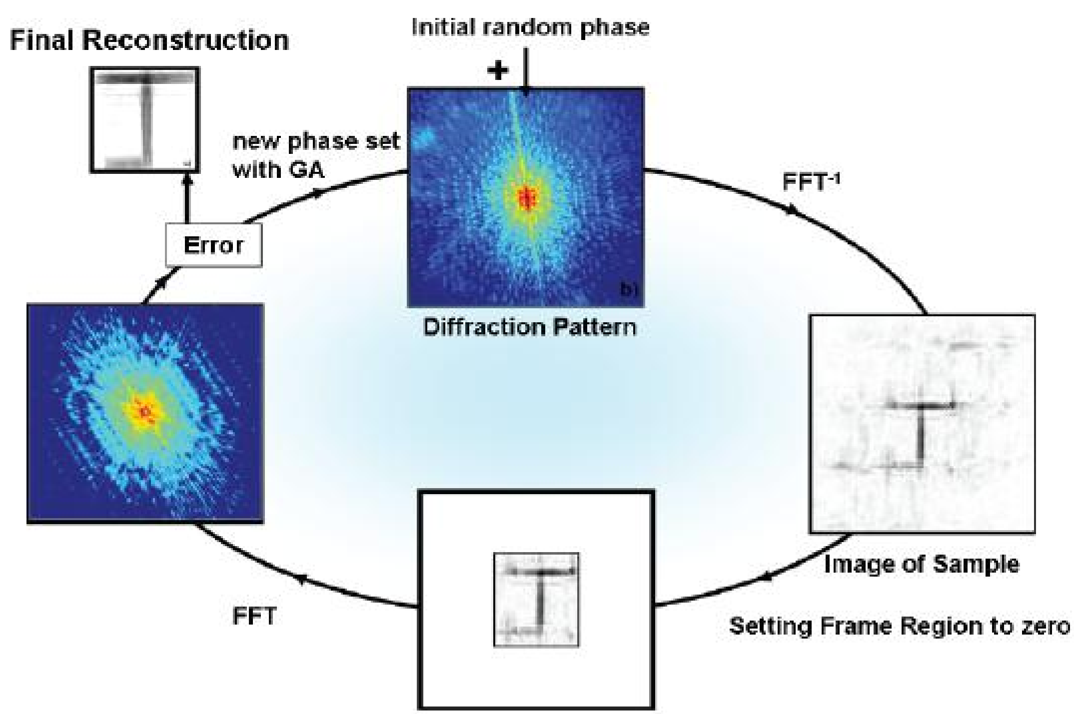


- Sample – ferromagnetic Ni-Fe layers separated by a Ru spacer layer
- Can demagnetize Ni very rapidly using fs laser
- Launch large spin current from Ni to Fe to **increase** or decrease magnetization depending on initial orientation!
- Need fast x-rays to capture spin dynamics in multiple layers simultaneously

Coherent Diffractive Nano-Imaging

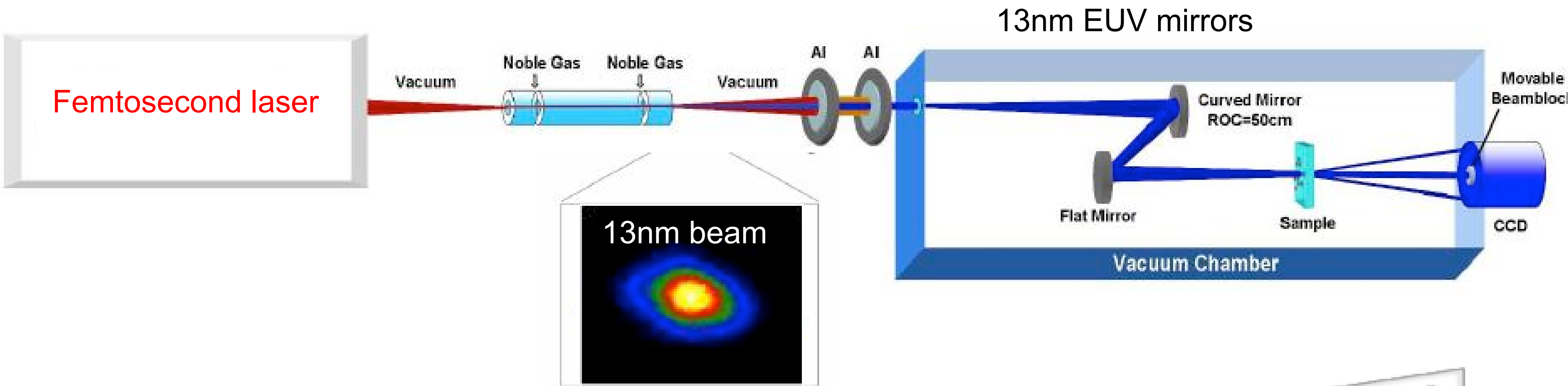


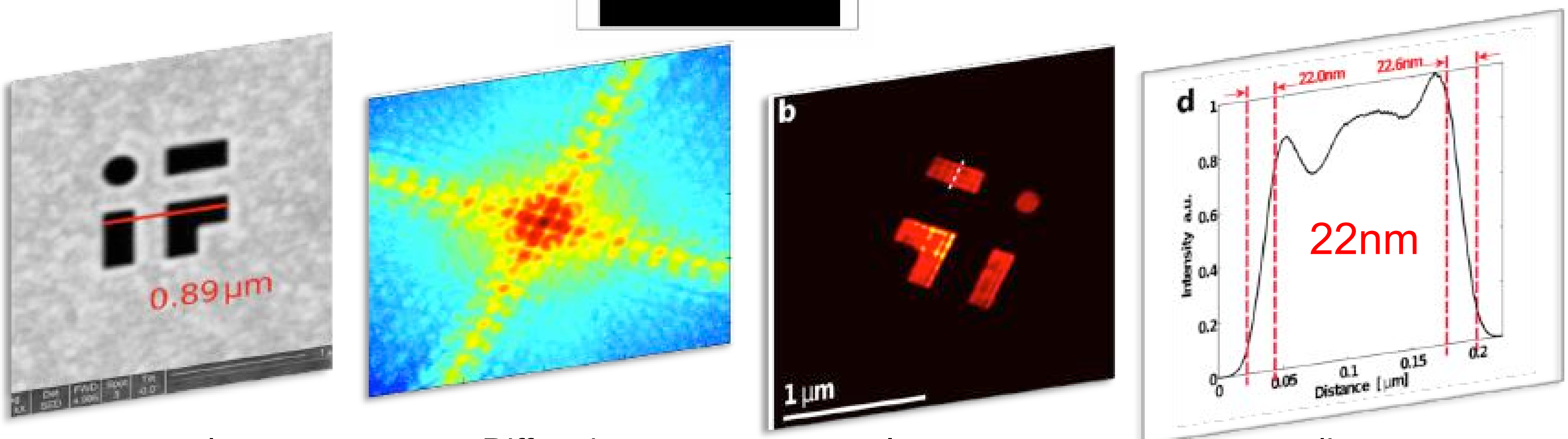
- No aberrations - diffraction-limit in theory
- Image thick samples
- Inherent contrast of x-rays
- Robust, insensitive to vibrations
- Requires a coherent beam and an isolated sample



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

Record tabletop light microscope: 22nm resolution using HHG





sample
(SEM image)

Diffraction
pattern

Image
reconstruction

lineout

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PRL ***99****, 098103 (2007); Nature* ***449****, 553 (2007); PNAS* ***105****, 24 (2008);*
Nature Photon. ***2****, 64 (2008); OL* ***34****, 1618 (2009); Optics Express* ***19****, 22470 (2011)*

Future

- Take attosecond electron rescattering physics, discovered over 20 years ago, to generate coherent x-rays
- Now have ultrafast coherent soft x-ray beams on a tabletop, and excellent prospects for hard x-ray beams from lasers
- Ultrafast x-rays and lasers can capture and control function in the nanoworld at the space time limits relevant to function
- Table-top microscopes, nanoprobes and x-ray imaging with unprecedented spatial and temporal resolution