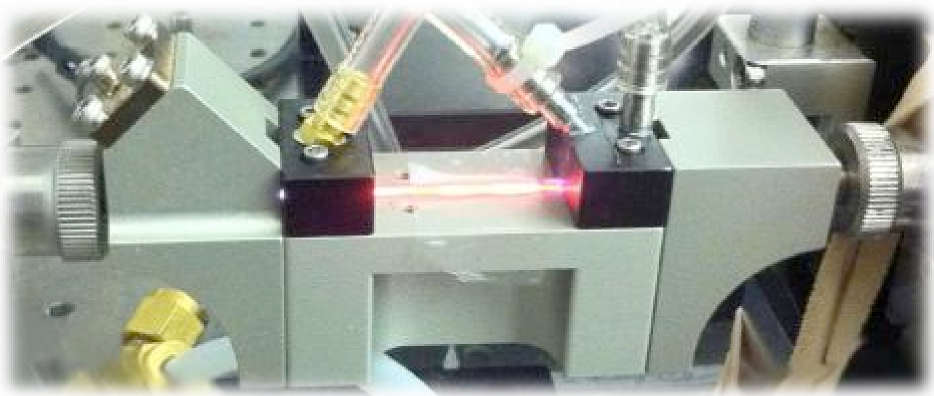
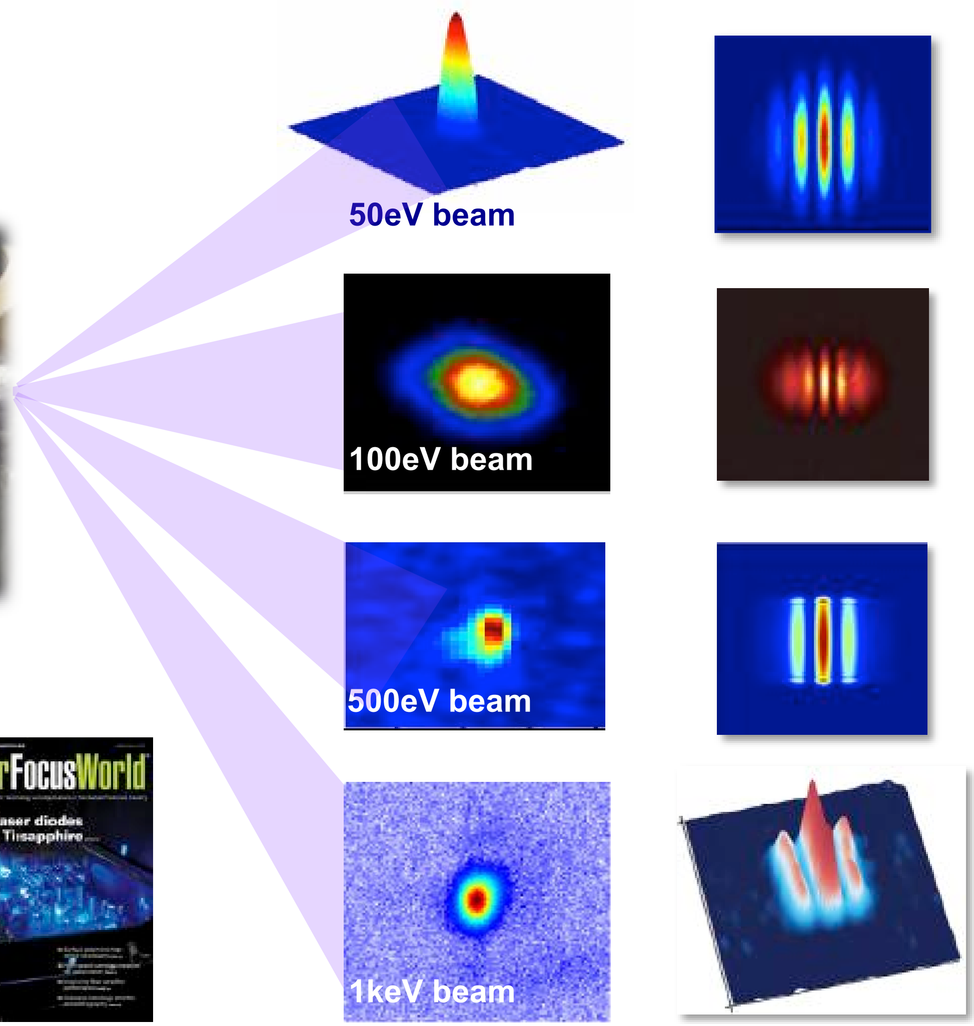
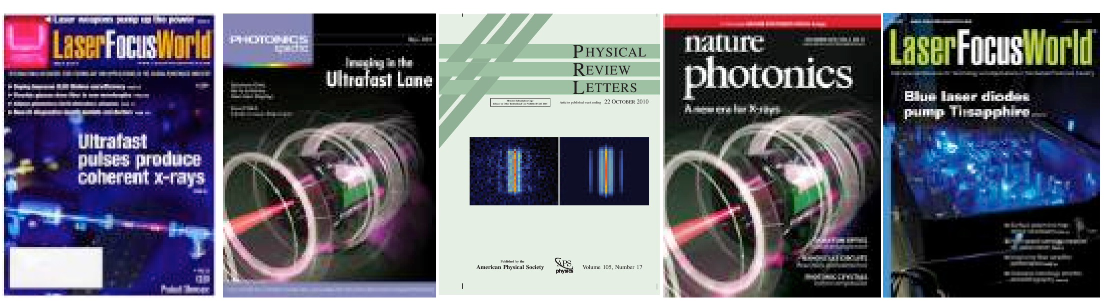
Nanoscale Materials Metrology using Coherent EUV High Harmonic Beams

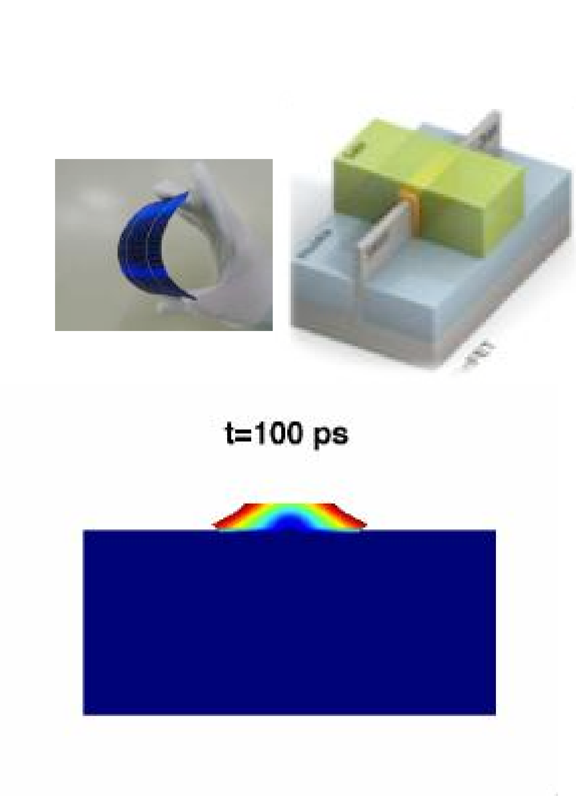




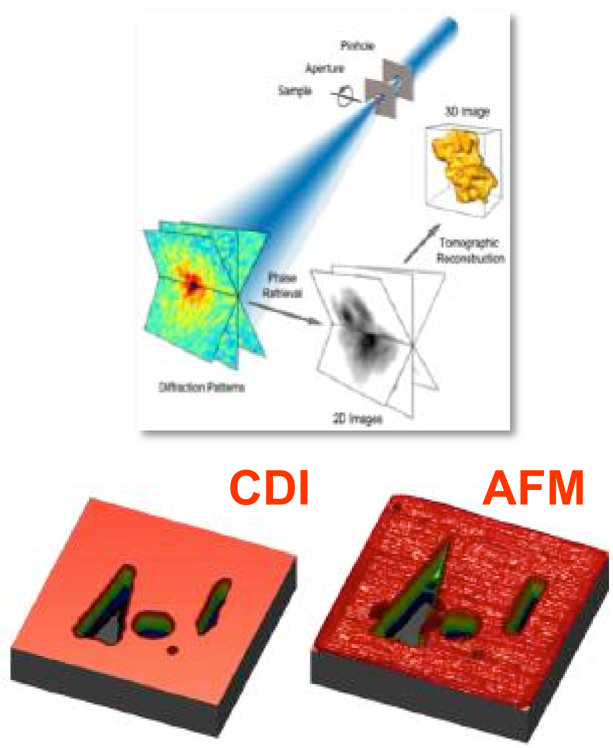


Nanoscale Materials Metrology using Coherent EUV High Harmonic Beams

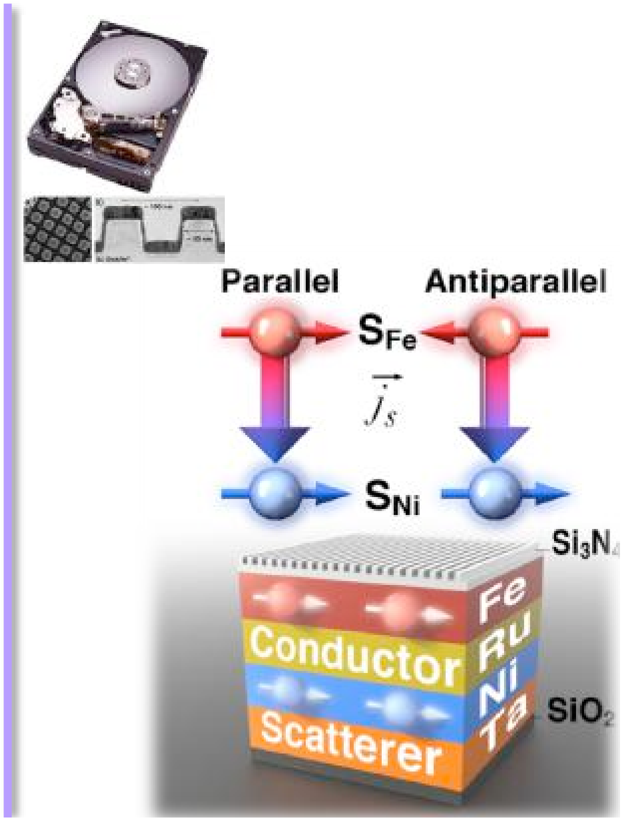
Acoustic nanometrology



Nanoscale coherent imaging

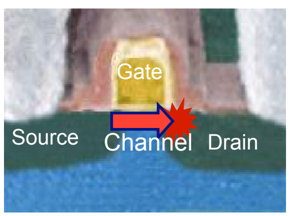


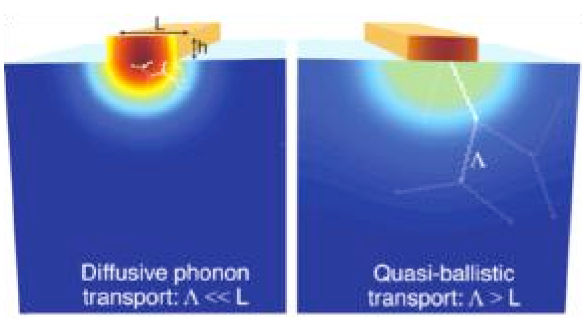
Nanoscale spintronics



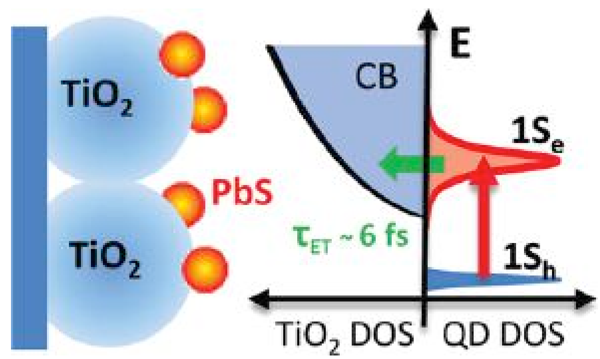
Nanoscale Materials Metrology using Coherent EUV High Harmonic Beams

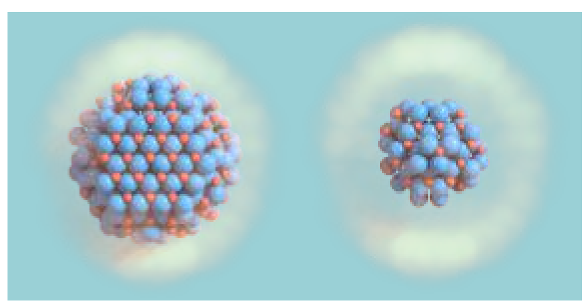
Nanoscale heat flow



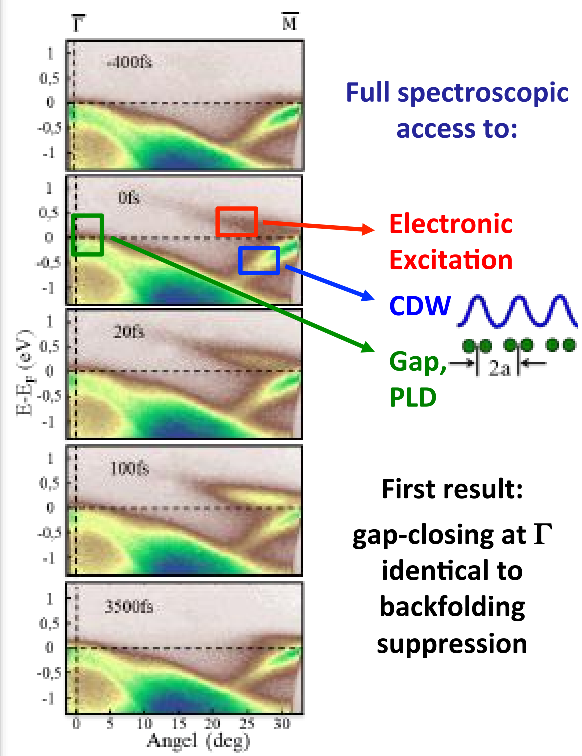


Photoemission quantum dots





Photoemission solids



Full spectroscopic access to:

First result:
gap-closing at Γ identical to backfolding suppression

Students and Collaborators

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JILA, University of Colorado at Boulder

Andrius Baltuška
Technical University Vienna

Eric Anderson, Eric Gullikson, Yanwei Liu, Farhad Salmassi
LBL

Marie Tripp and Sean King
Intel

Bruce Gurney, Olav Hellwig
Hitachi

Stefan Mathias, Martin Aeschlimann, Claus Schneider
Kaiserslautern and Julich

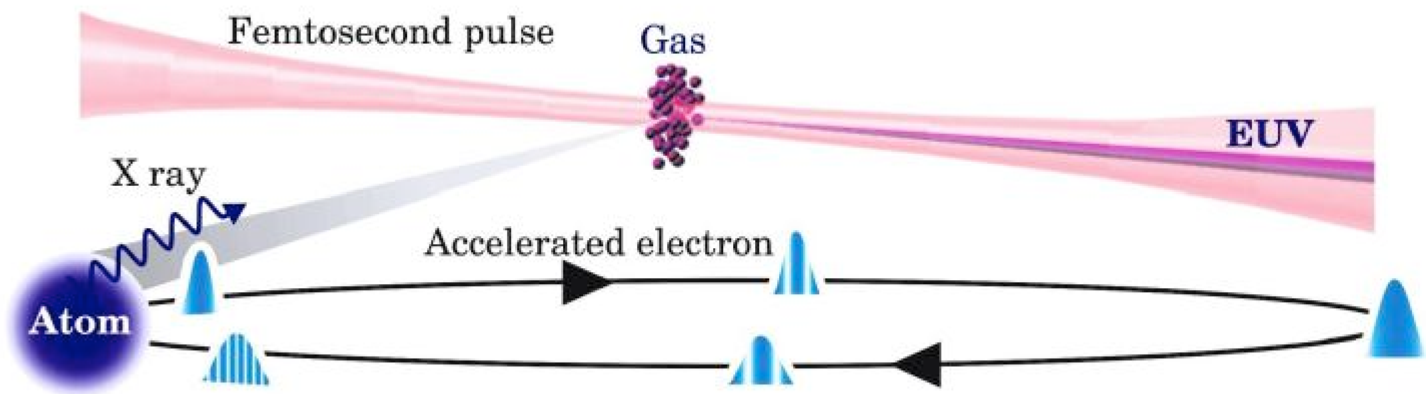
Tom Silva, Justin Shaw, Hans Nembach
NIST

Carmen Menoni
CSU

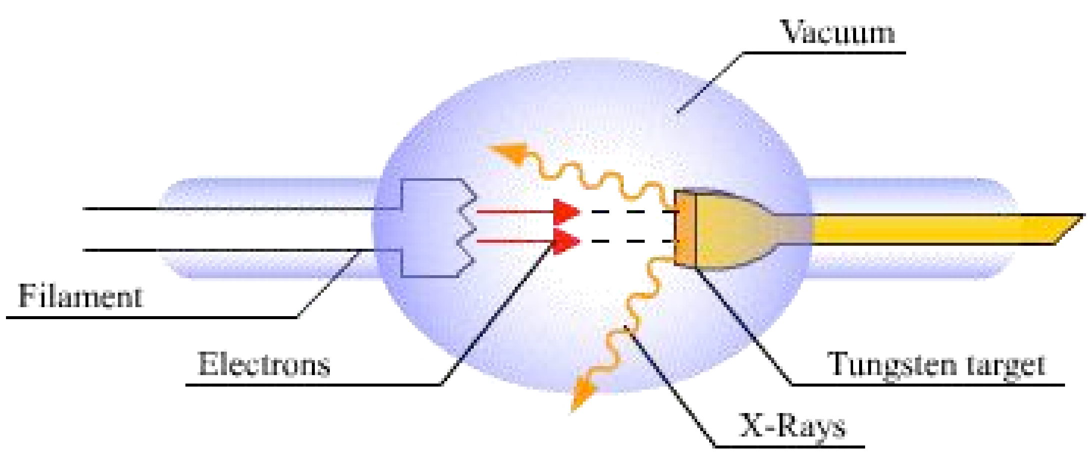
Bill Schlotter
SLAC

National Security Science and Engineering Faculty Fellowship Competition

High harmonics - coherent version of X-Ray tube

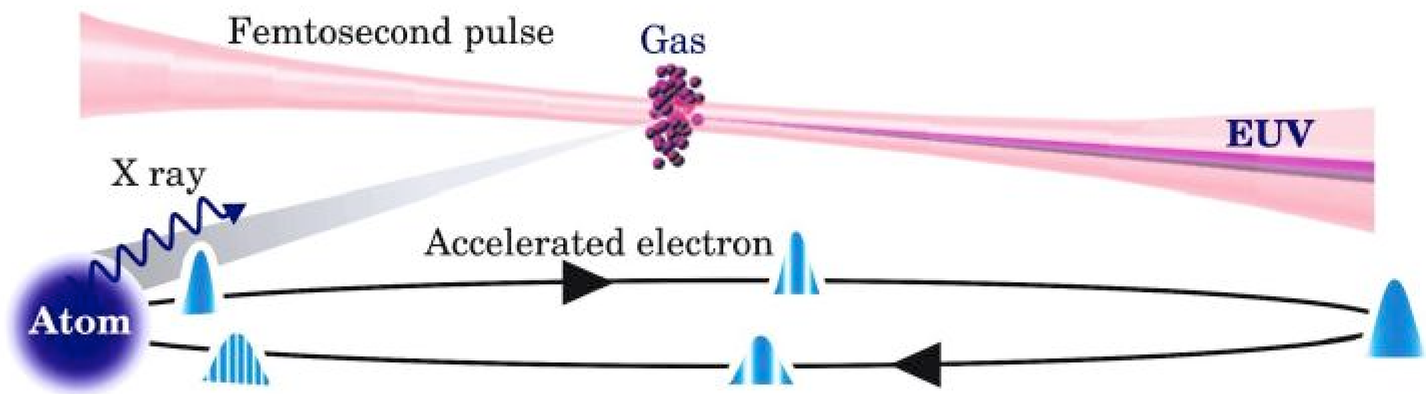


High Harmonic Generation



Röntgen X-ray Tube

High harmonics - coherent version of X-Ray tube



High Harmonic Generation

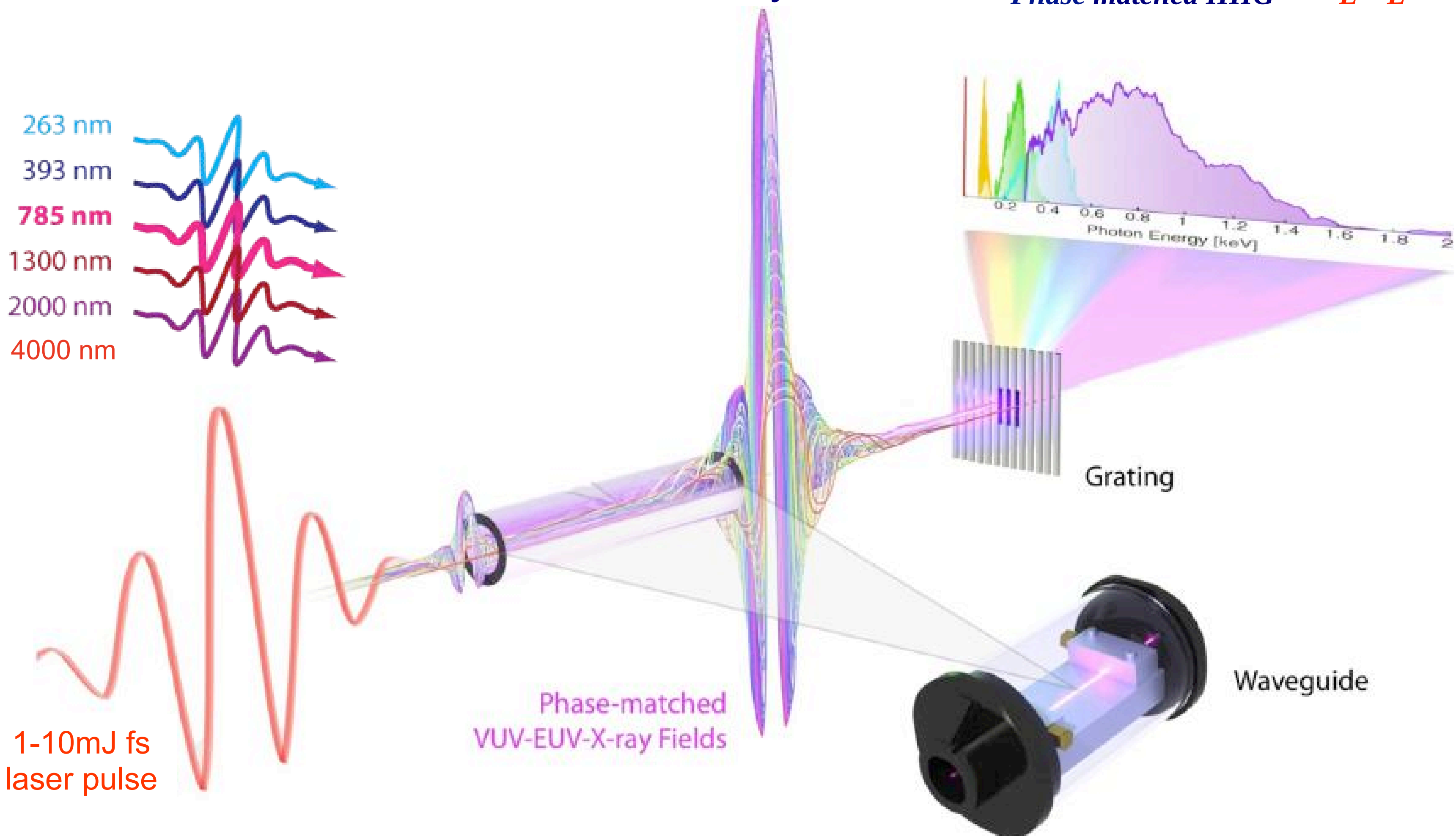
Extreme nonlinear optics

Optimizing high harmonic sources

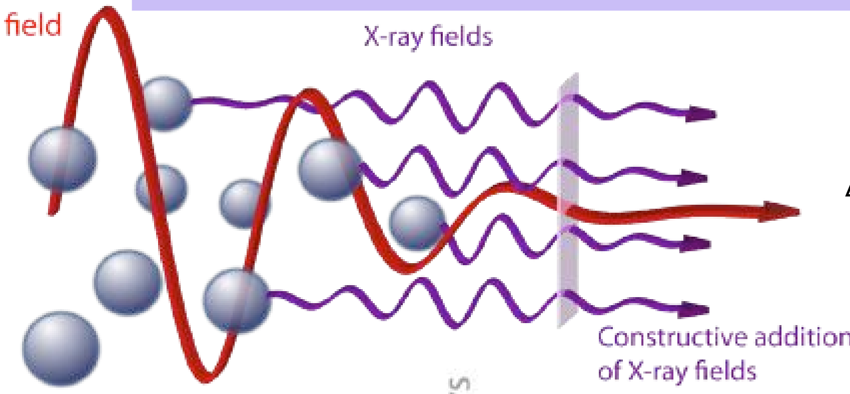
What driving laser wavelength and gas pressure to use for bright HHG?

1. Need bright single atom emission: $h\nu_{Single\ atom\ HHG} \propto I_L \lambda_L^{2}$

2. Need coherent addition from many atoms: $h\nu_{Phase\ matched\ HHG} \propto I_L \lambda_L^{1.7}$

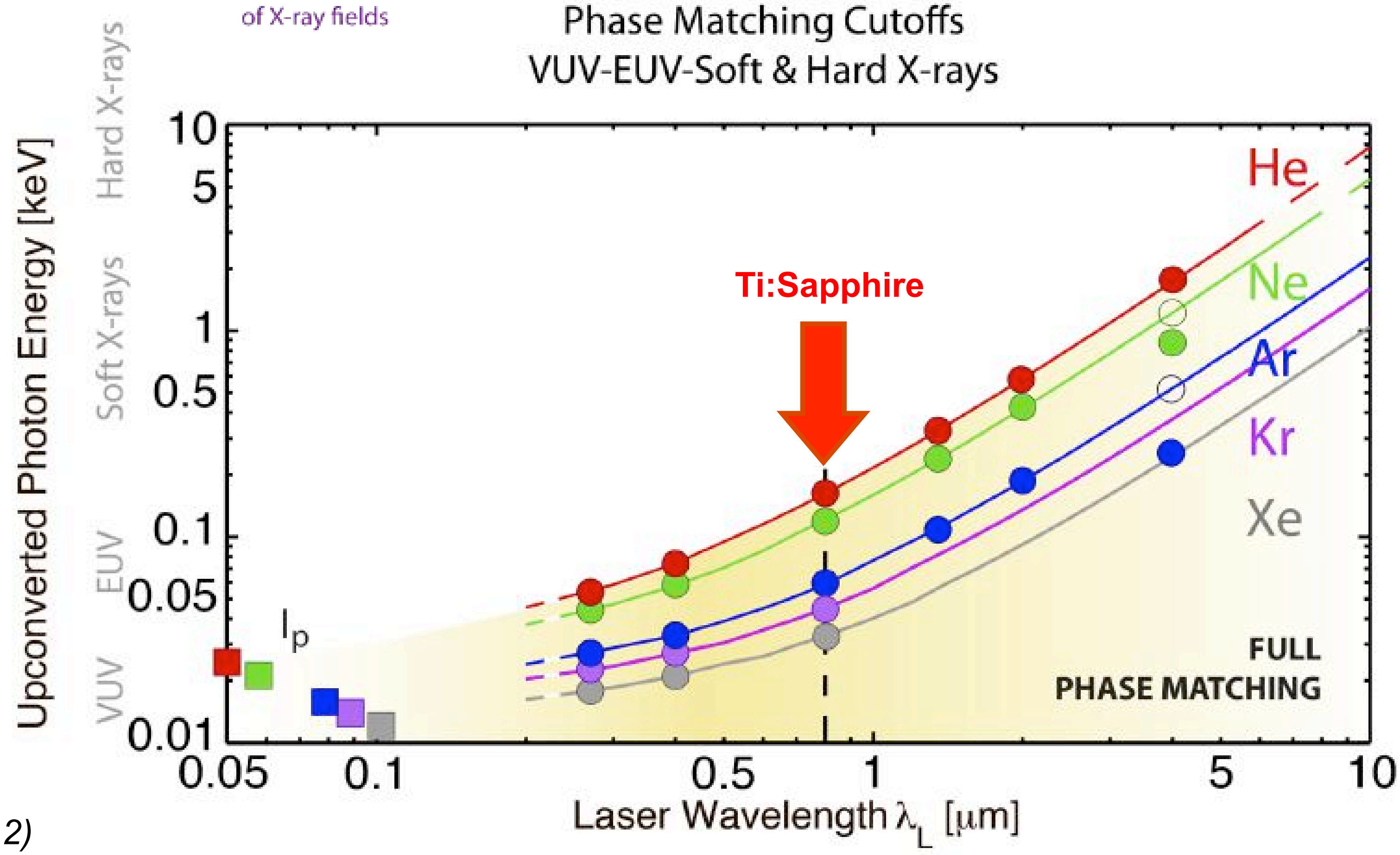


Bright HHG emission driven by ultrafast lasers



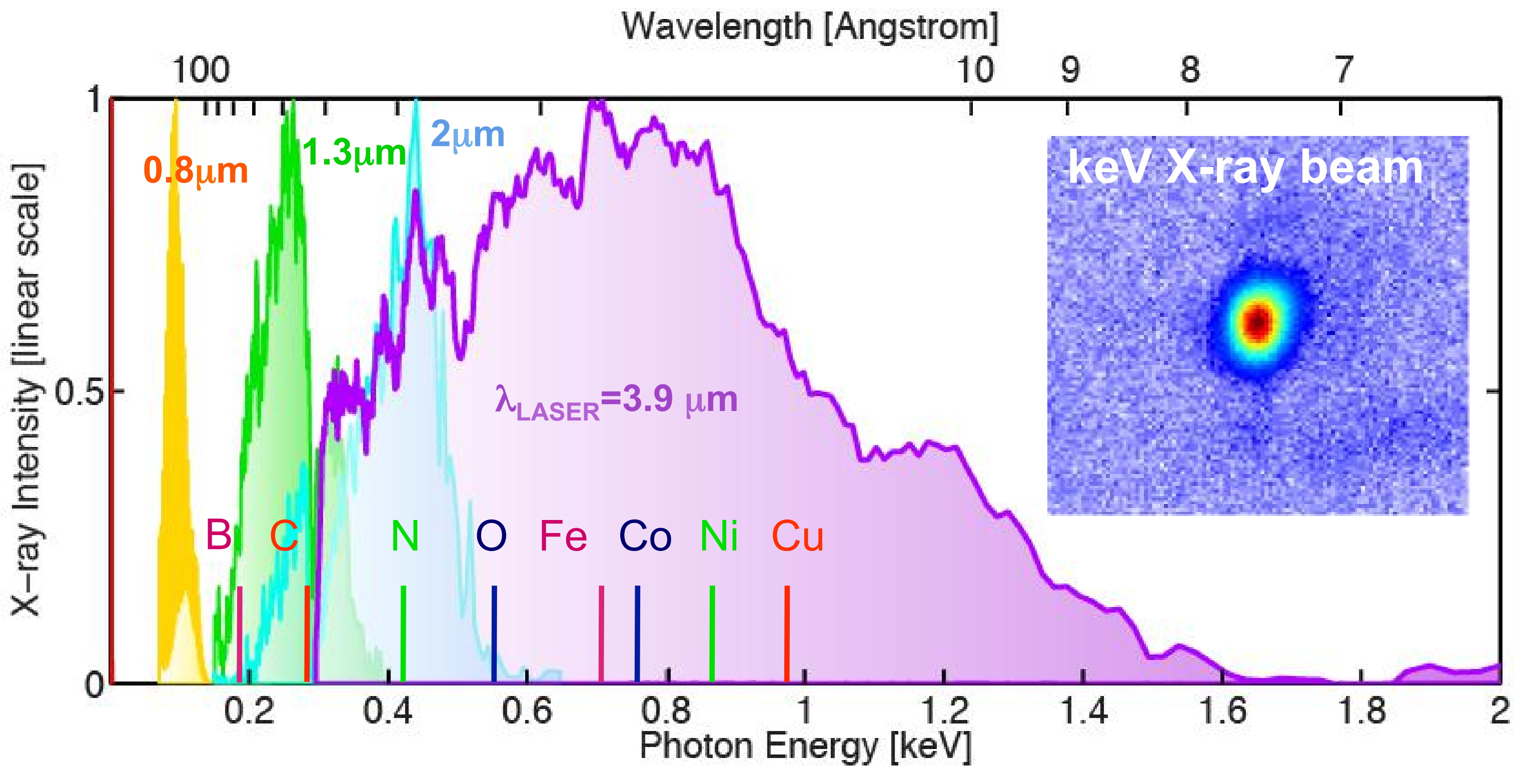
$$\Delta k = q\left\{\left(\frac{u_{11}^2 \lambda_0}{4\pi a^2}\right) - P\left((1-\eta)\frac{2\pi}{\lambda_0}\Delta\delta - \eta\left[N_{atm} r_e \lambda_0\right]\right)\right\}$$

- For **keV** HHG, need mid IR lasers
- For **EUV** HHG, want 0.8µm lasers
- For **VUV** HHG, want UV lasers



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

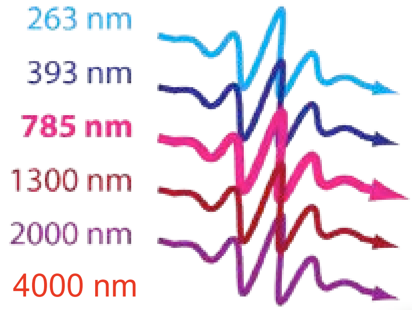
Unique source – coherent supercontinuum to 8Å

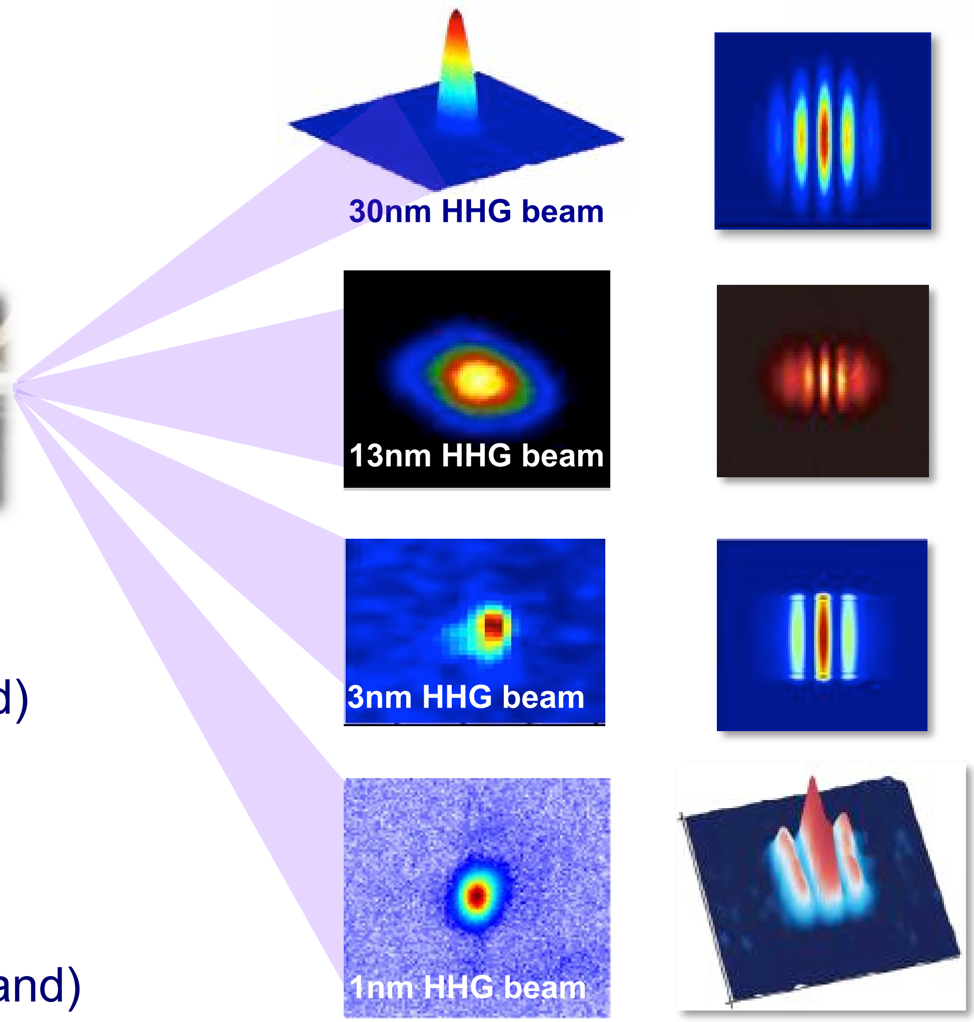


- Bright coherent tabletop keV X-rays for first time
- Highest nonlinear and phase matched process at > 5000 orders
- Phase matching bandwidth ultrabroad since $v_{X\text{-rays}} \approx c$
- Coherent spectrum spans many elemental x-ray edges

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

Bright coherent beams from UV to keV





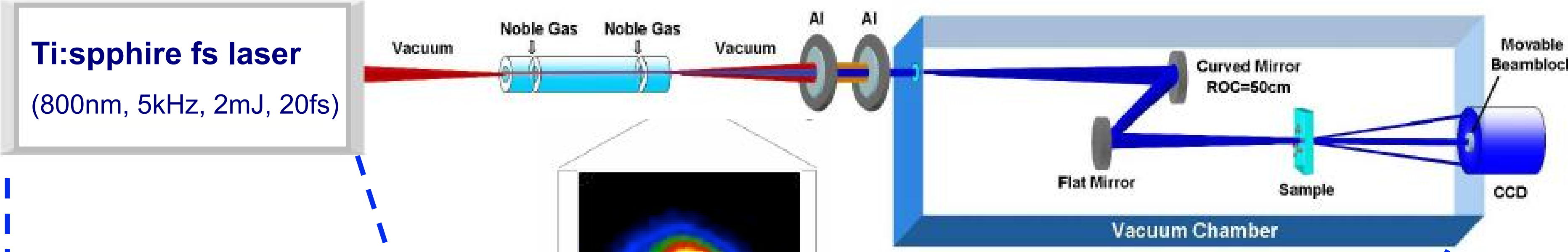
Current conversion efficiency:
10-50 eV: 10^{-3} – 10^{-4}/eV (per 1% band)
50-100 eV: 10^{-5} – 10^{-6} /eV
300-1000 eV: 10^{-6} – 10^{-7}/eV

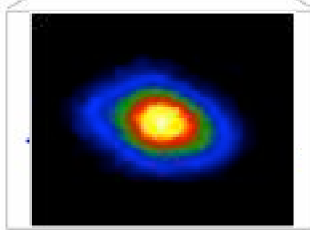
Laser powers: 10 – 50W
EUV power: μW – 0.5mW (per 1% band)

Limit not known: Increases in efficiency and photon energy very likely

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

Robust, simple, HHG setup in EUV

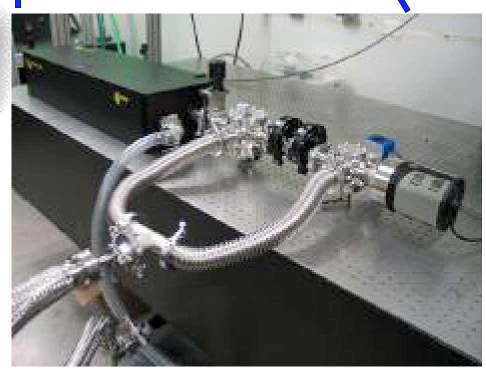




X-ray beam

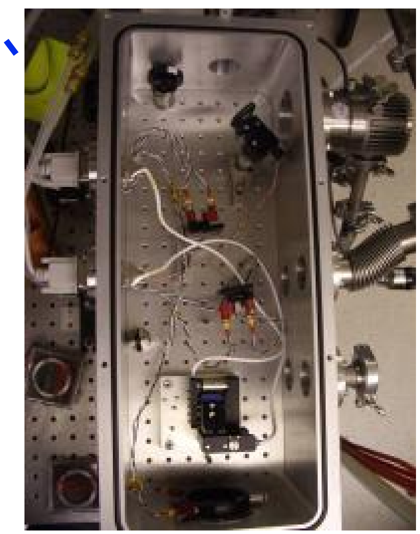
High average power femtosec laser system

+



EUV HHG

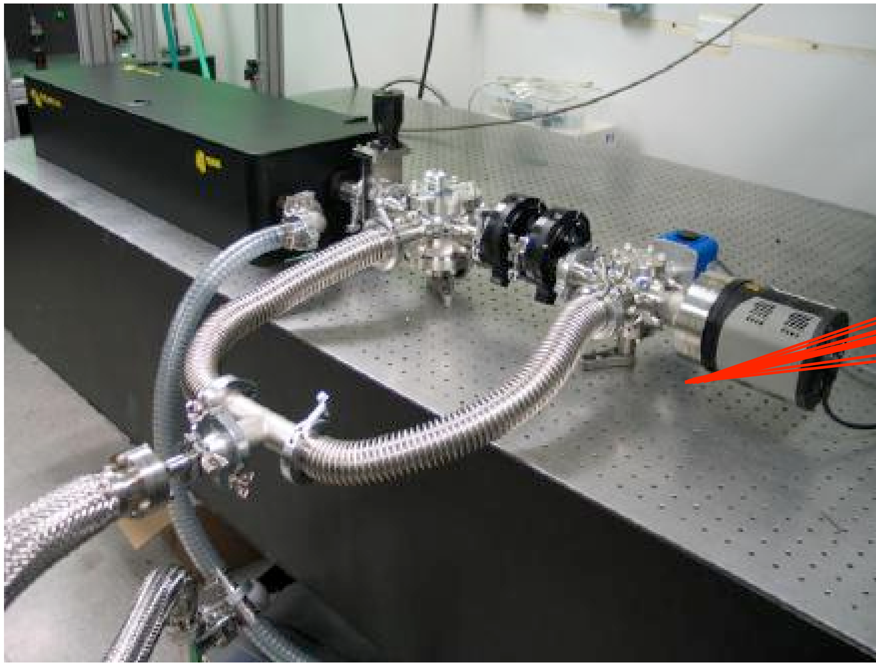
+

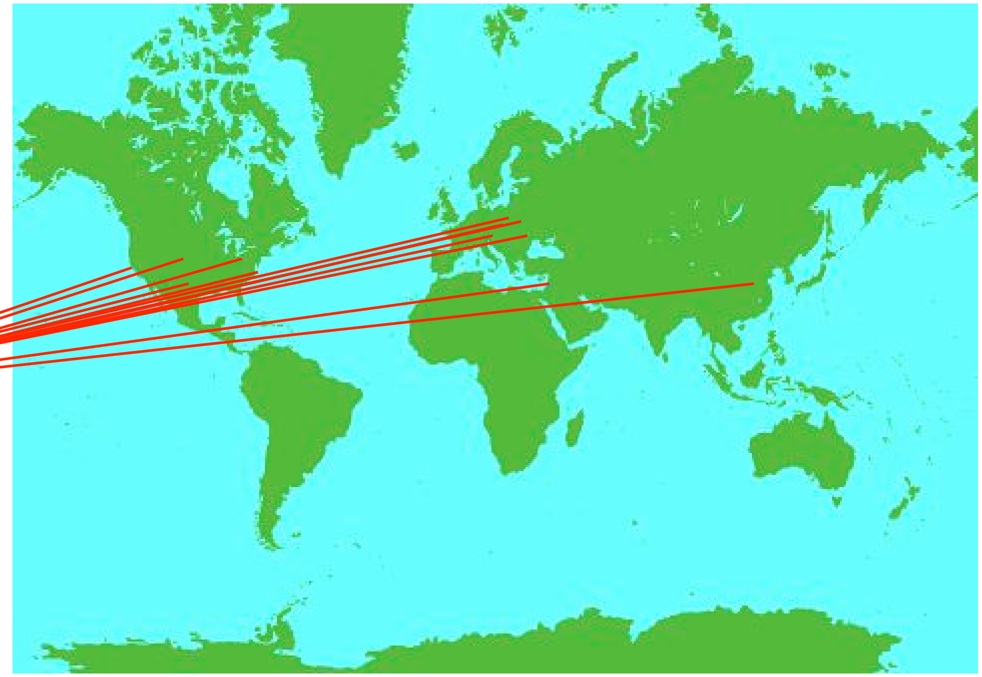


Coherent diffractive microscope

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

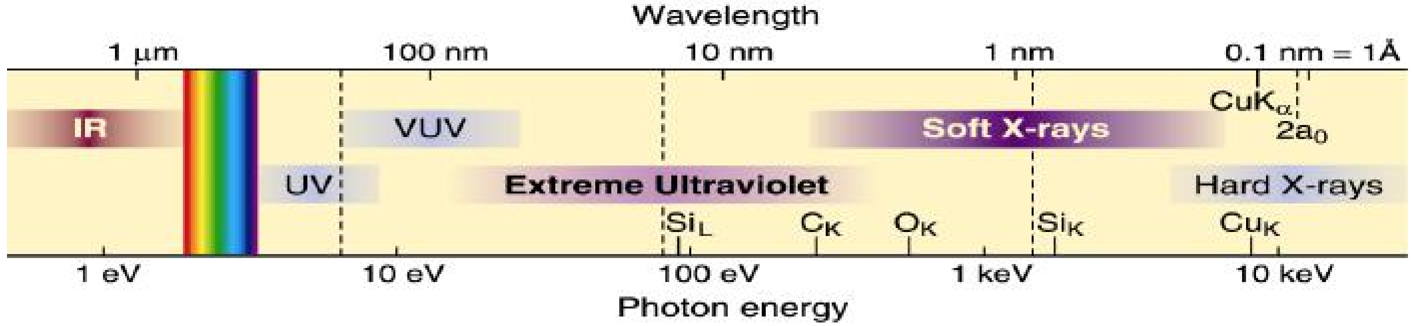
Tested in many research labs worldwid



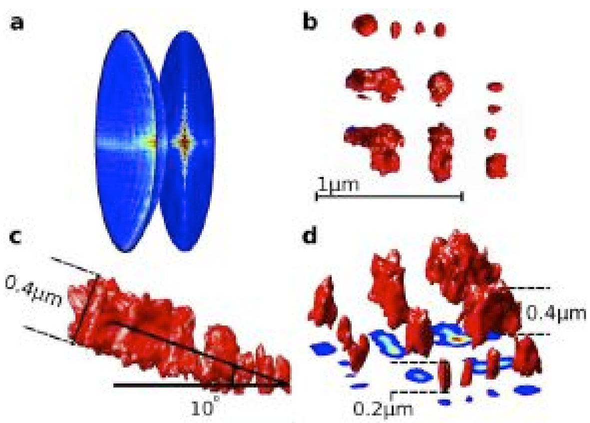


- First commercial ultrafast coherent EUV source
- Operated at CLEO exhibit in May 2009
- Commercial, integrated, UHV-compatible system installed in Germany (4), Israel (2), MIT(1), Caltech (1), China (1) and Bulgaria (1) for applications in materials science
- Used successfully by many groups

Unique ultrafast coherent tabletop X-ray source

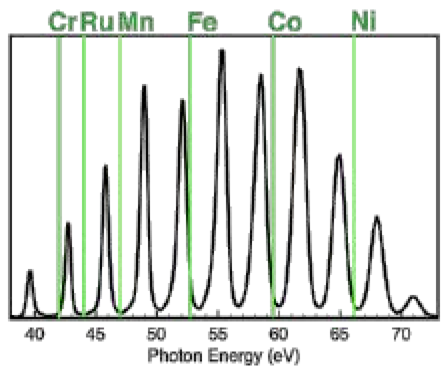


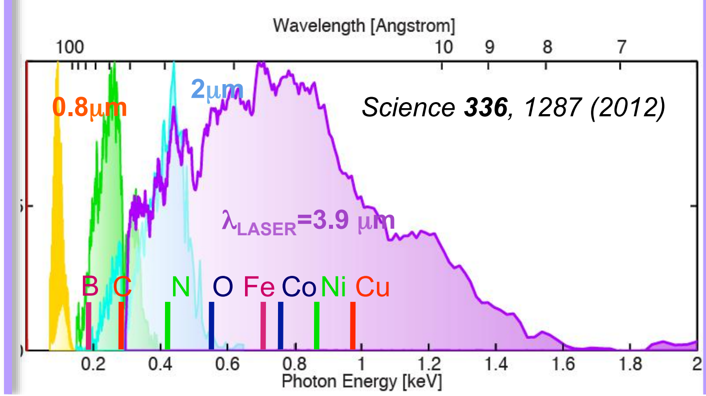
SPATIAL: 3D near-λ imaging



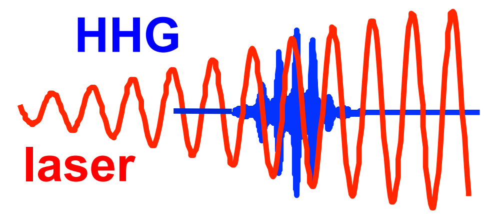
*Nature **463**, 214 (2010)*
*Optics Express **19**, 22470 (2011)*

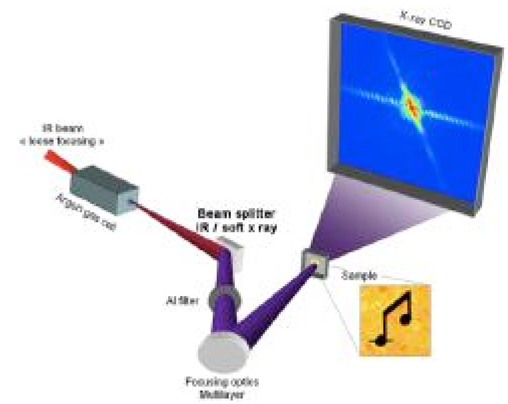
SPECTRAL: Elemental and chemical specificity





TEMPORAL: Ultrafast, single shot

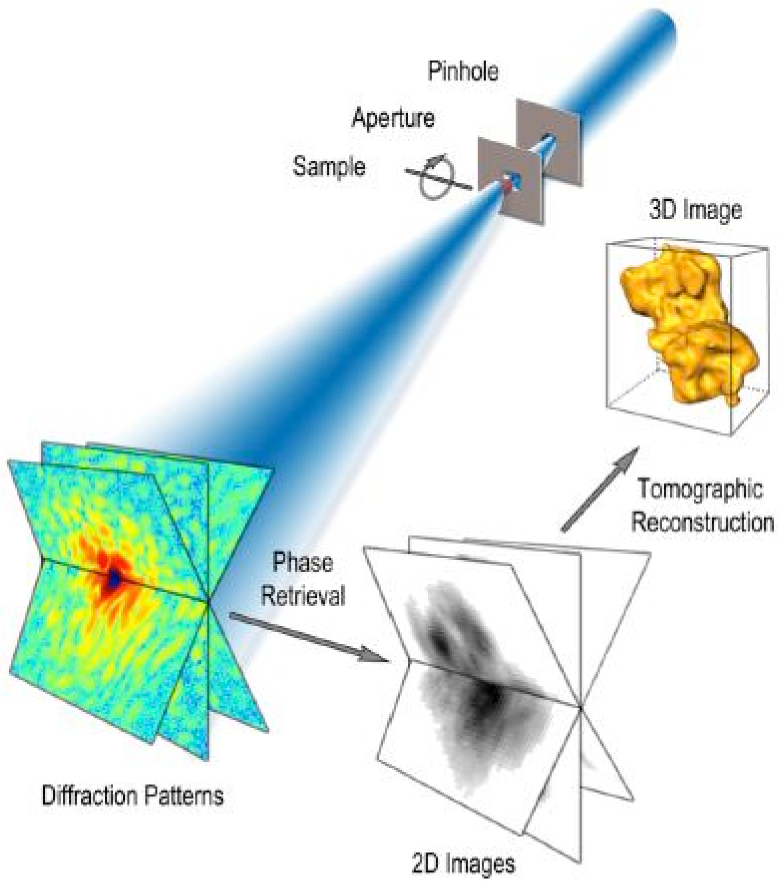


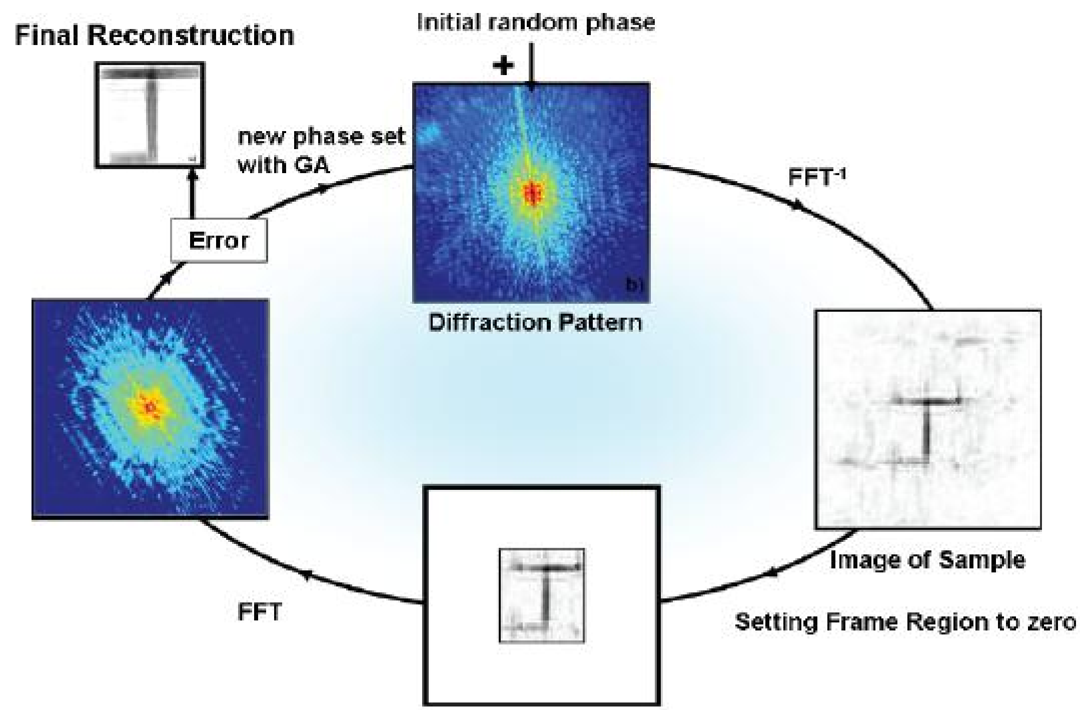


*Phys. Rev. Lett. **103**, 028104 (2009)*
*Nature News and Views **460**, 1088 (2009)*

Coherent Diffractive X-Ray Imaging (XCDI)

- **Diffraction-limited imaging ≈ $\lambda/2NA$**
- Image thick samples
- Inherent contrast of x-rays
- Robust, insensitive to vibrations
- Needs a coherent beam and isolated sample

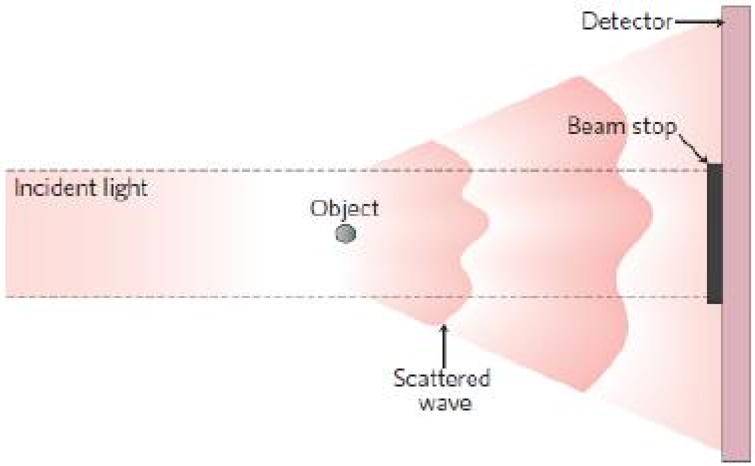




Sayre, Acta Cryst ***5****, 843 (1952)*
Miao et al., Nature ***400****, 342 (1999)*

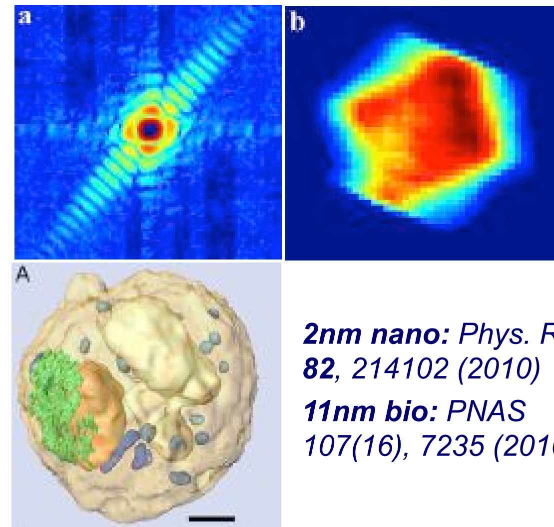
EUV and X-ray Microscopy

Coherent Diffractive Imaging (CDI)



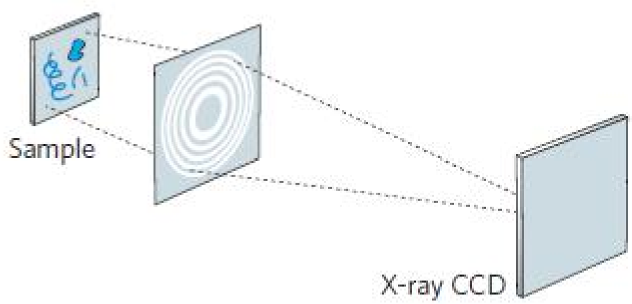
*Miao et al., Nature **400**, 342 (1999)*
*Chapman and Nugent, Nature Photonics, **4**, 833 (2010)*

- ✓ λ limited resolution (≈2nm)
- ✓ Robust to vibrations



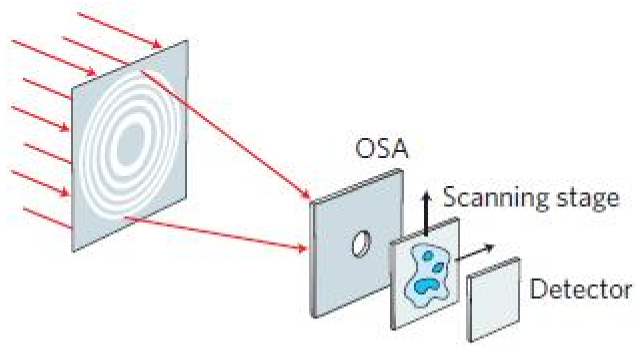
***2nm nano:** Phys. Rev. B **82**, 214102 (2010)*
***11nm bio:** PNAS 107(16), 7235 (2010)*

Full-field Zone Plate Microscopy



*Sakdinawat and Attwood, Nature Photonics, **4**, 840 (2010)*

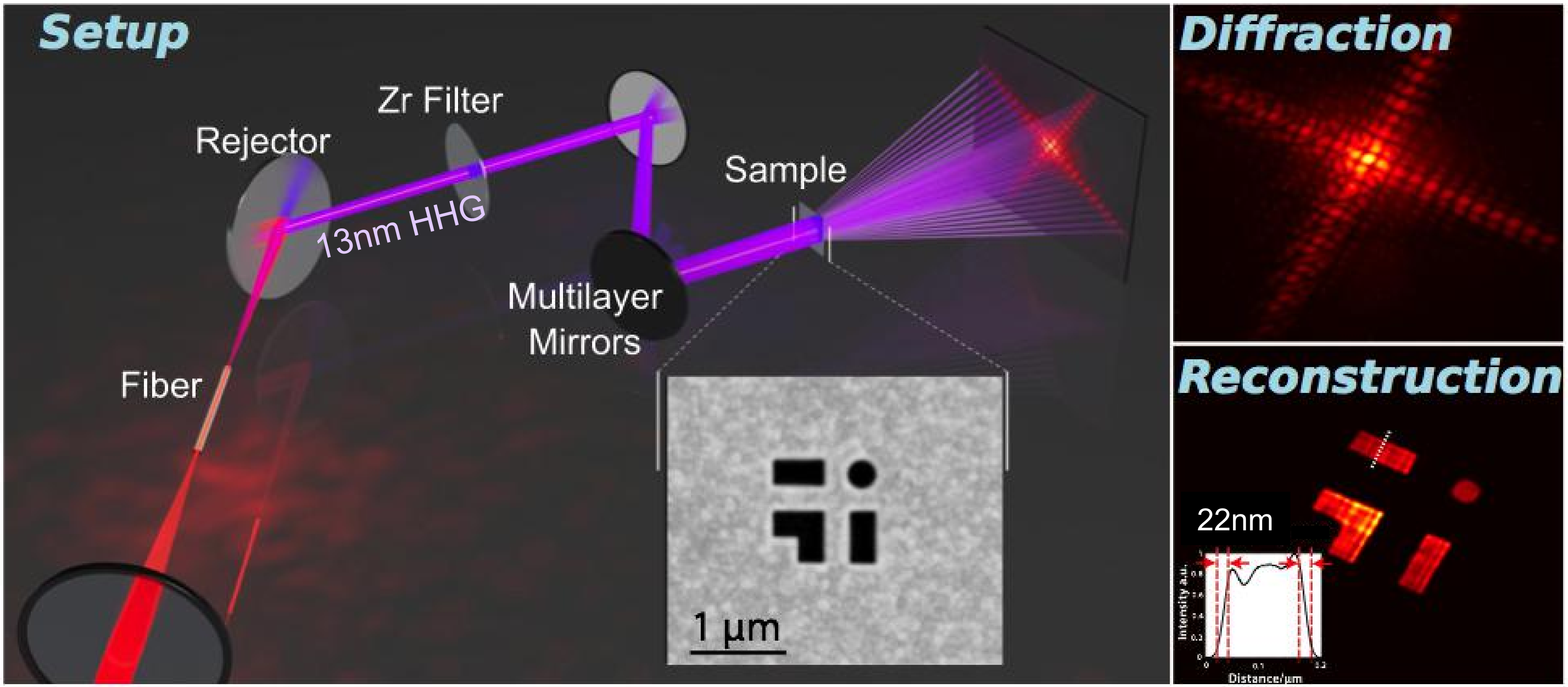
Scanning Transmission X-ray Microscopy



- ✓ Well established (≈13nm)
- ✓ Real image

Record tabletop light microscope: 22nm resolution

NA ≈ 0.6 – 0.8

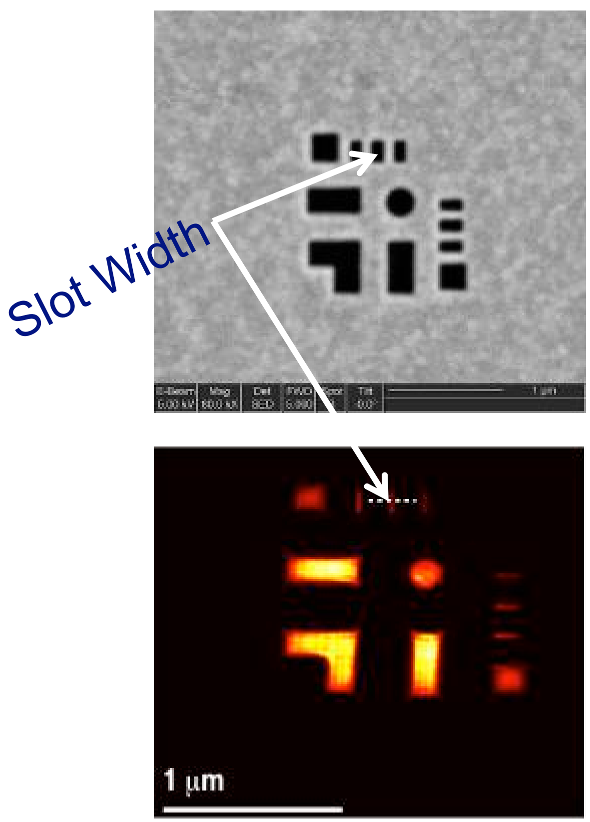


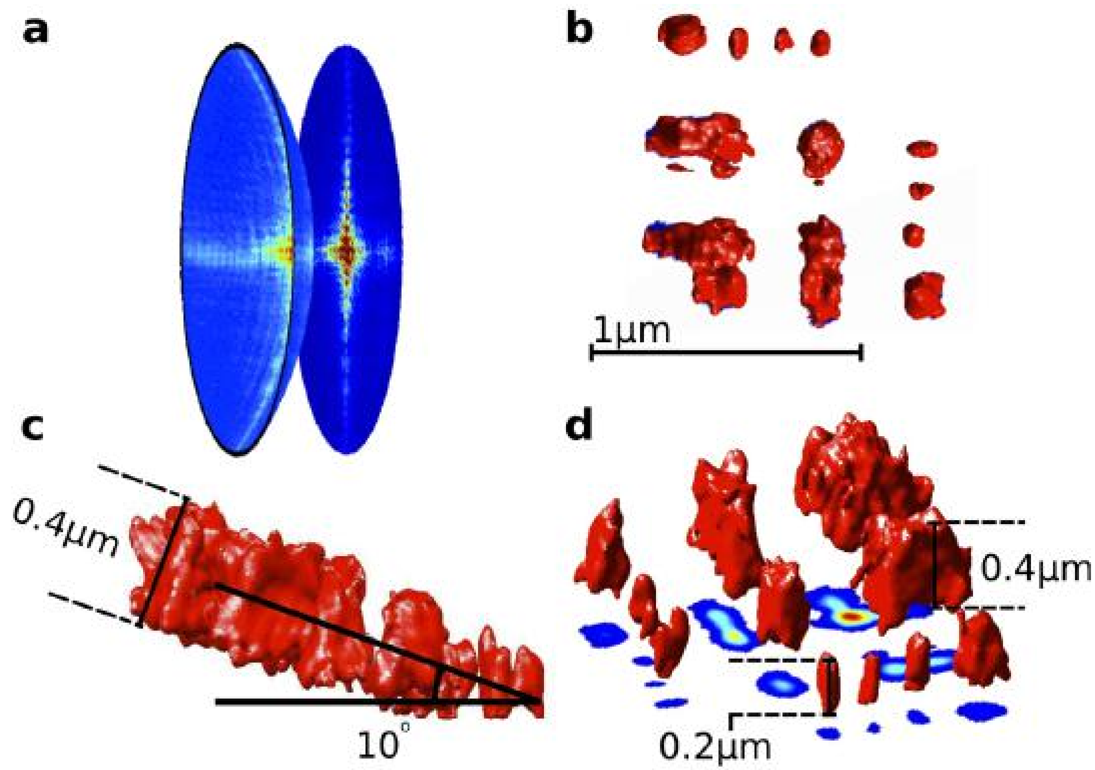
PRL ***99****, 098103 (2007); Nature* ***449****, 553 (2007); PNAS* ***105****, 24 (2008);*
Nature Photon. ***2****, 64 (2008); OL* ***34****, 1618 (2009); Optics Express* ***19****, 22470 (2011)*

Three-dimensional structure determination from a single view

NATURE | Vol 463 | 14 January 2010

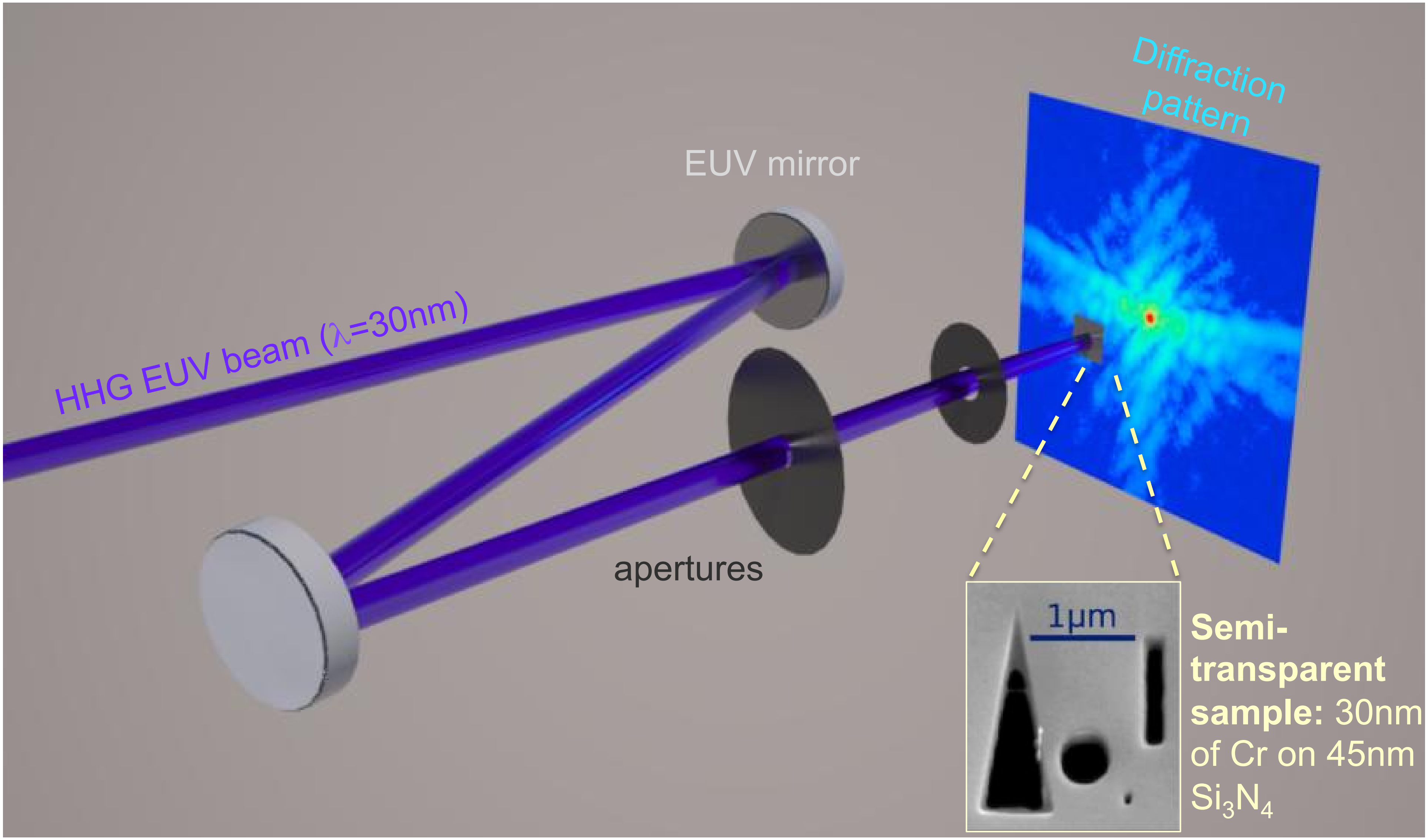
Kevin S. Raines[1,2], Sara Salha[1,2], Richard L. Sandberg[4,5], Huaidong Jiang[1,2], Jose A. Rodríguez[3], Benjamin P. Fahimian[1,2], Henry C. Kapteyn[4,5], Jincheng Du[6,7] & Jianwei Miao[1,2]





Nature ***463***, *214 (2010)*
Optics Express ***19***, *22470 (2011)*

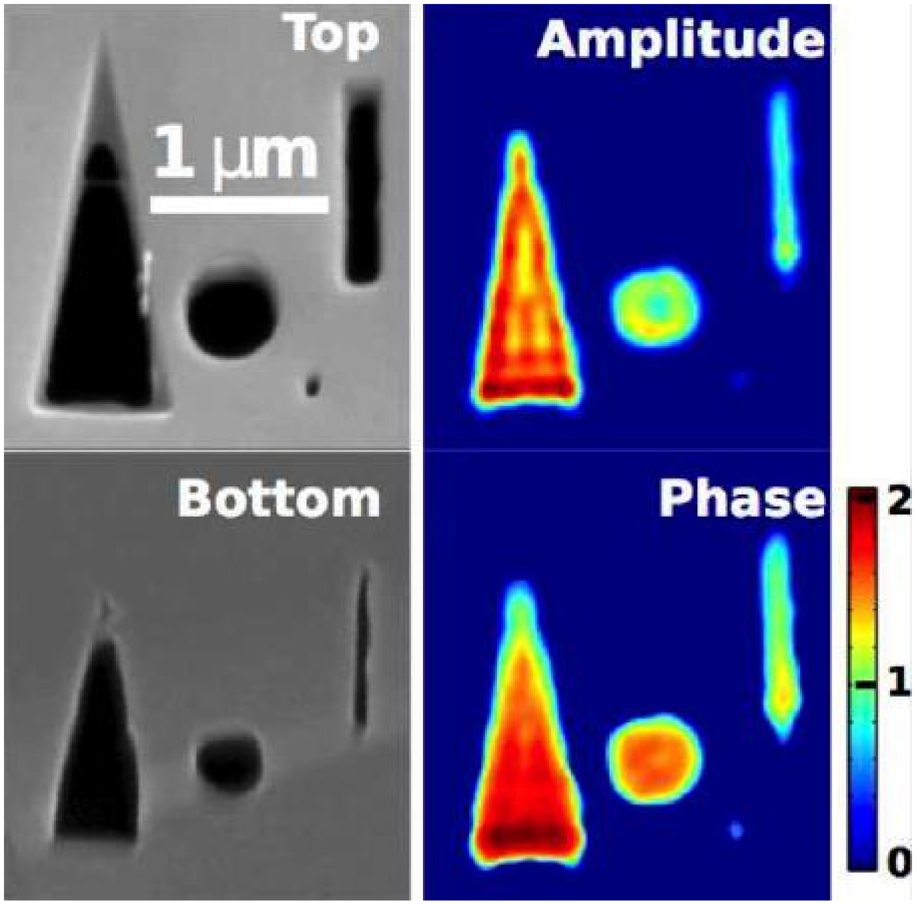
Scanning non-isolated transmission mode CDI



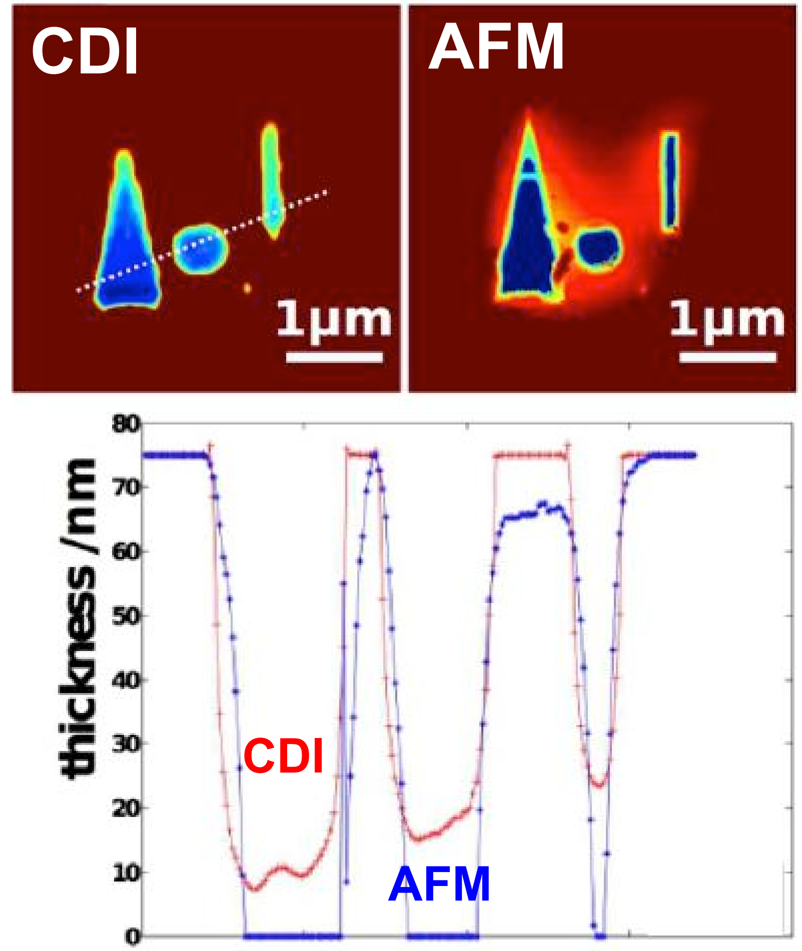
*Keyhole CDI: Abbey et al., Nat. Phys., **4**, 394 (2008)*

*Optics Express **20**, 19050 (2012)*
Submitted (2013)

Retrieved images: comparison with SEM and AFM



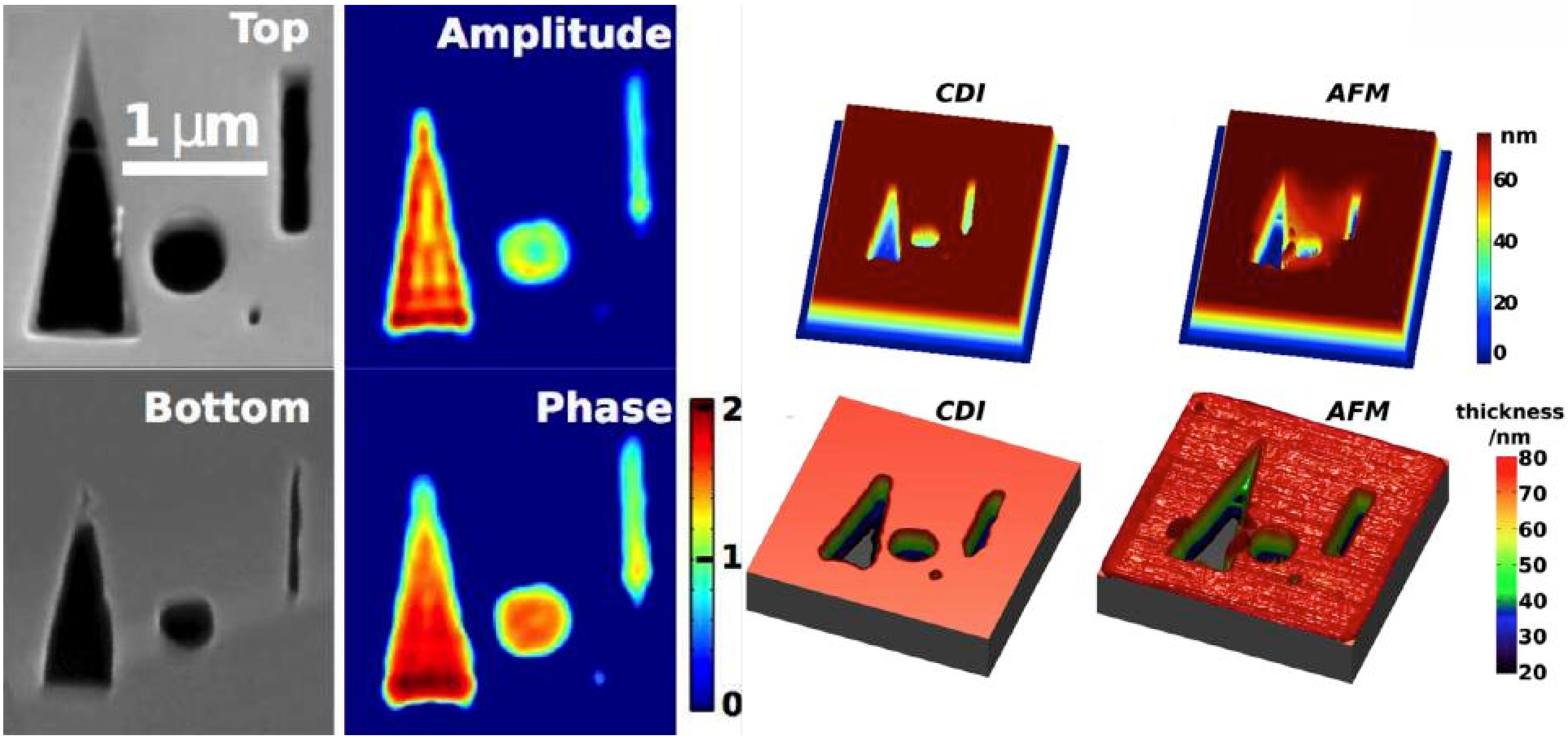
SEM CDI



Thickness map

- Semi-transparent background – can extract thickness
- 50nm hole not completely drilled through: 48nm (CDI) vs 52nm (AFM)

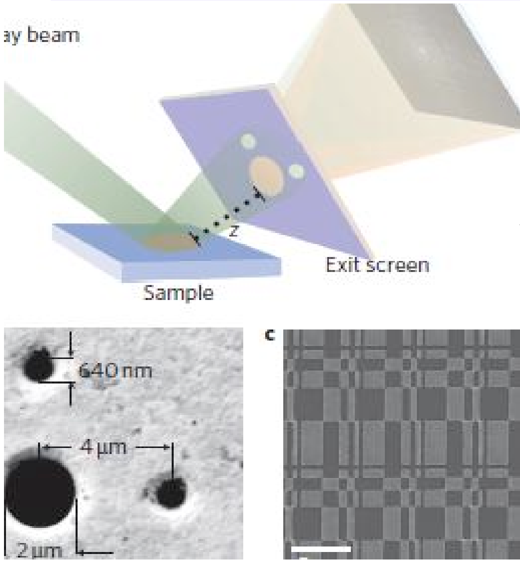
Retrieved images: comparison with SEM and AFM



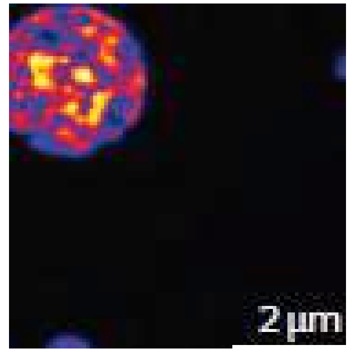
SEM **CDI** **CDI** **AFM**

3D profiling

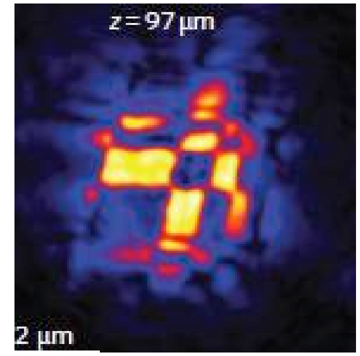
First reflection mode coherent imaging



Nature Photon. **5**, *243 (2011)*

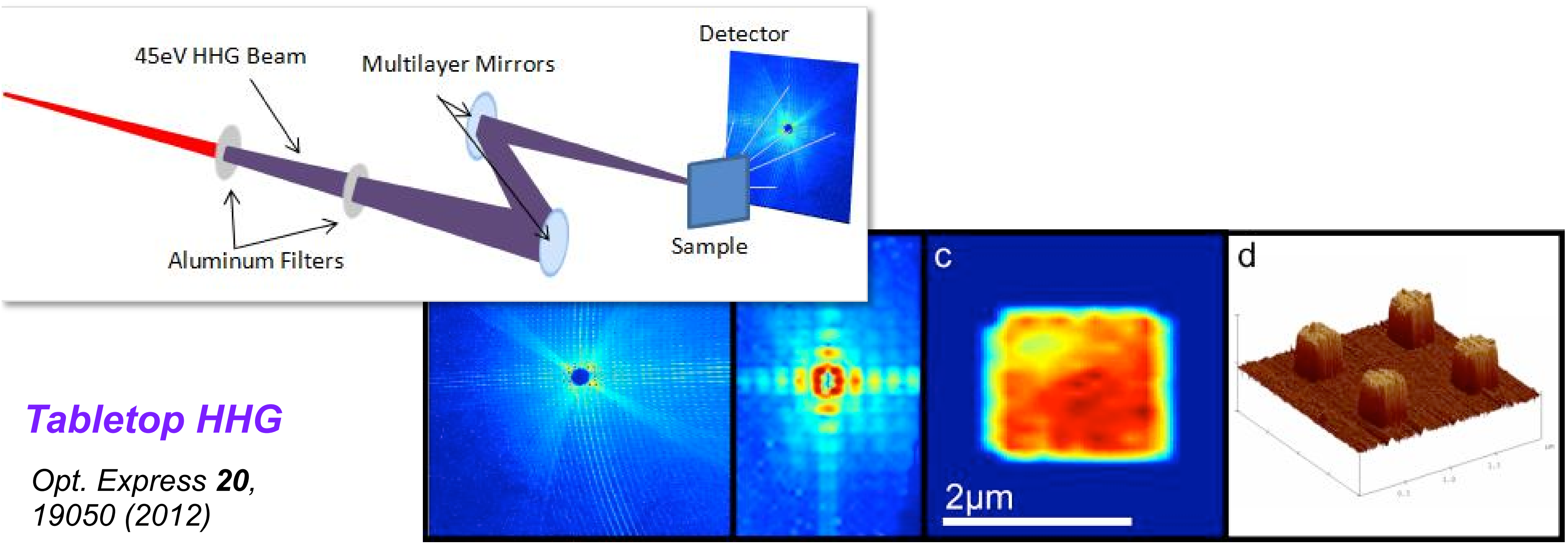


Reconstruction at Exit Screen



Field at sample

ALS

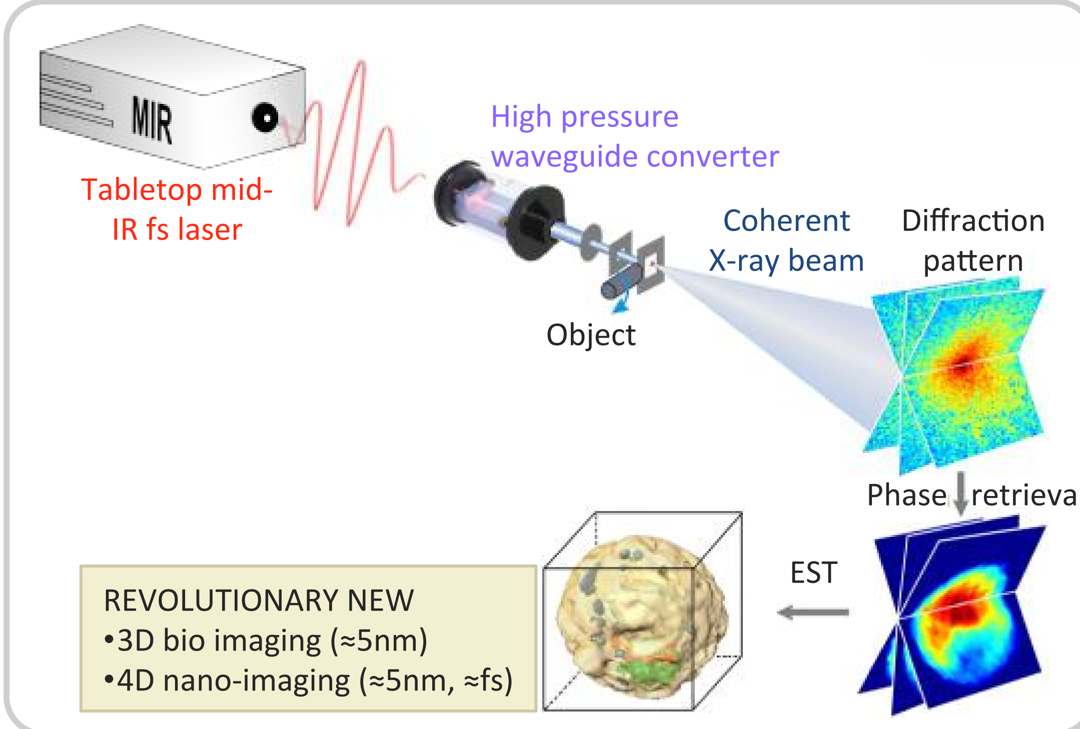


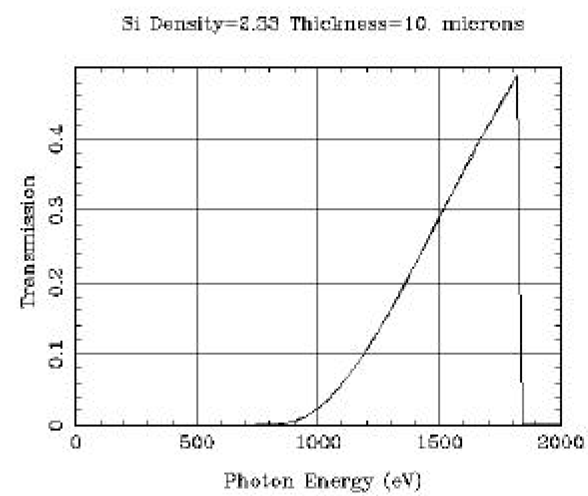
Tabletop HHG

Opt. Express **20**, *19050 (2012)*

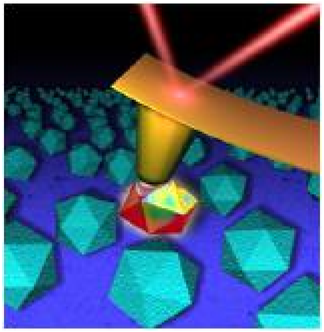
Future plans for coherent imaging

- General scanning reflection mode coherent microscope (2013)
- Shorter wavelengths to increase the NA, spatial resolution ≈ 5nm, and 3D imaging of thick samples
- Advanced low-dose EST algorithms *(Miao, Nature **483**, 444 (2012))*
- Rate limiting step – need 3μm lasers and advanced detectors





Nanoscale acoustic metrology of <100nm films



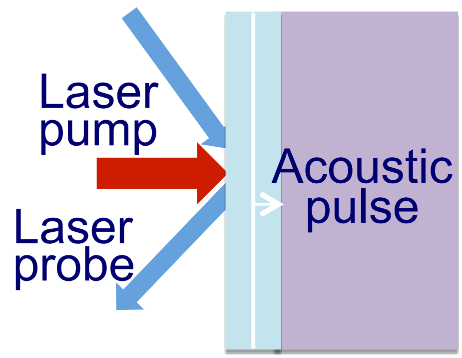
1. Nano-indentation

- ☑ Localized
- ☒ Destructive
- ☒ Substrate contribution

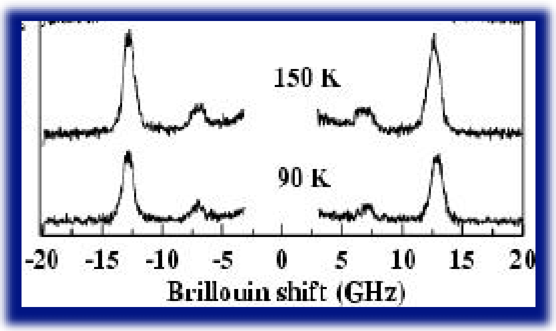
J. Mater. Res 24, 2960 (2009)

2. Optical photoacoustics

- ☑ Non-contact
- ☒ LAW only → assume v
- ☒ Limited to thick films



MRS Bulletin 31, 607 (2006)



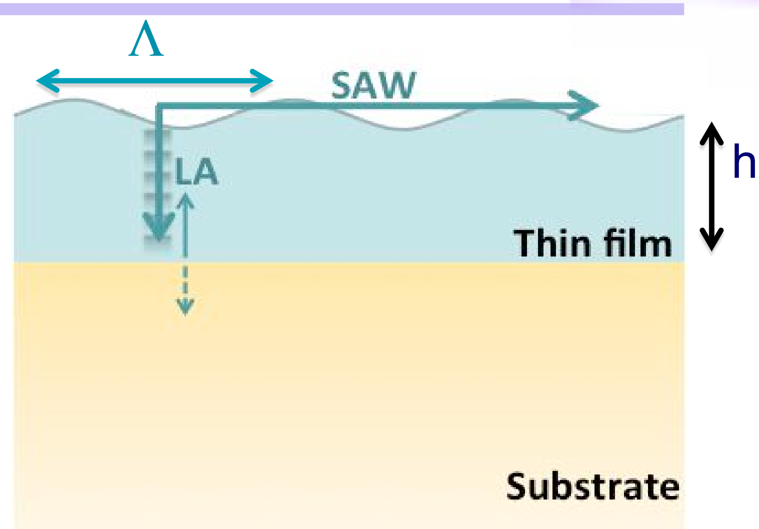
JAP 100, 013507 (2006)

3. Brillouin light scattering

- ☑ Both LA and TA modes
- ☒ Complex interpretation
- ☒ Sensitive to experimental accuracy

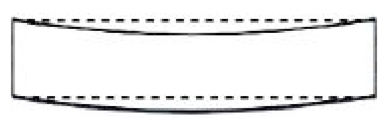
Advantages of EUV acoustic nano-metrology

- Acoustic waves propagate along (SAW) and into (LAW) surface: EUV x100 more sensitive than visible
- Penetration depth ≈ ζ~Λ/2π: EUV can characterize **nano**-mechanical elastic properties of < 10nm films
- Simultaneously extract Young's modulus and Poisson's ratio from LAW and SAW



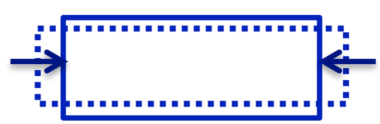
Young's modulus

$$E = c_{44}\left(\frac{3c_{11} - 4c_{44}}{c_{11} - c_{44}}\right)$$



Poisson's ratio

$$\nu = \frac{c_{11} - 2c_{44}}{2(c_{11} - c_{44})}$$

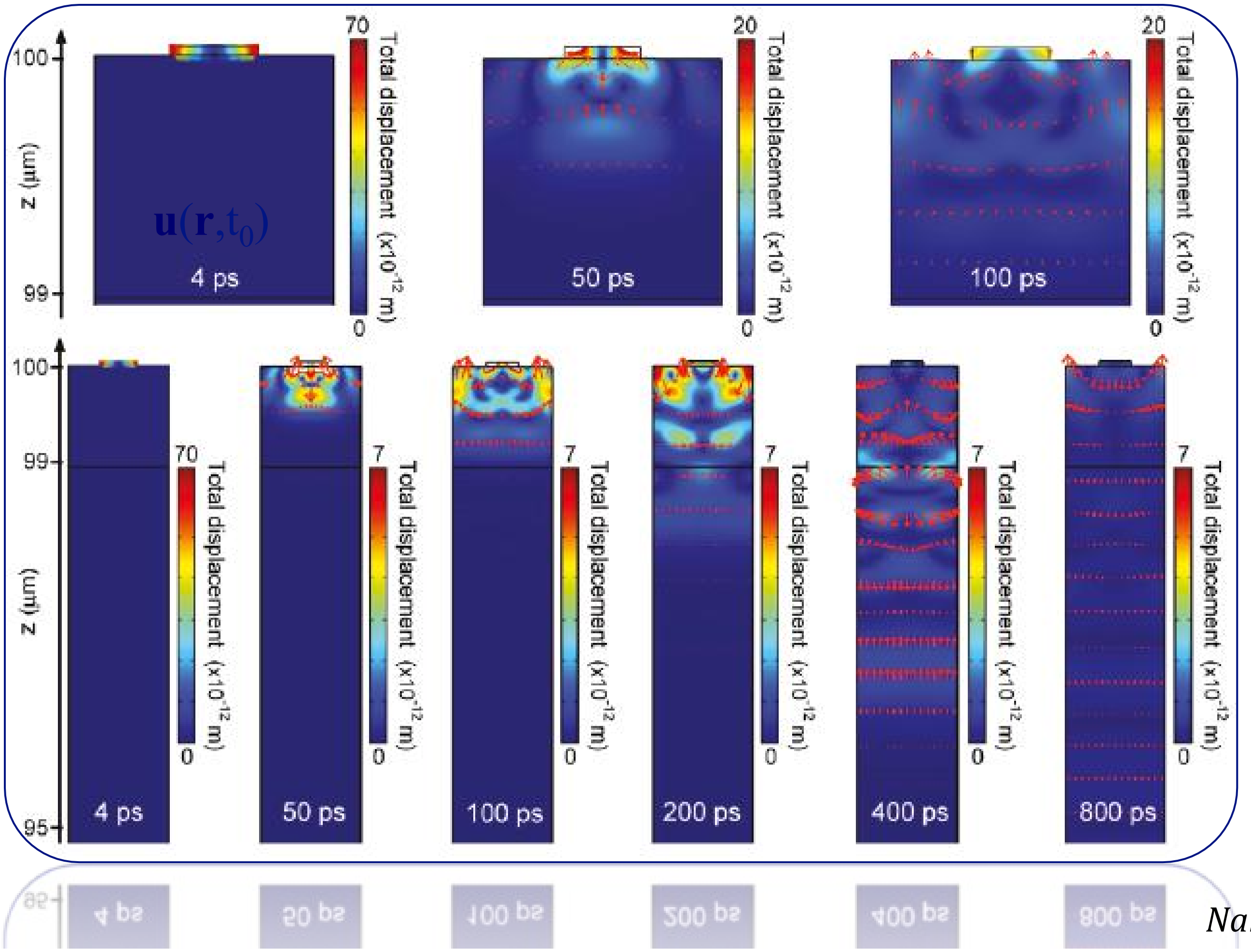


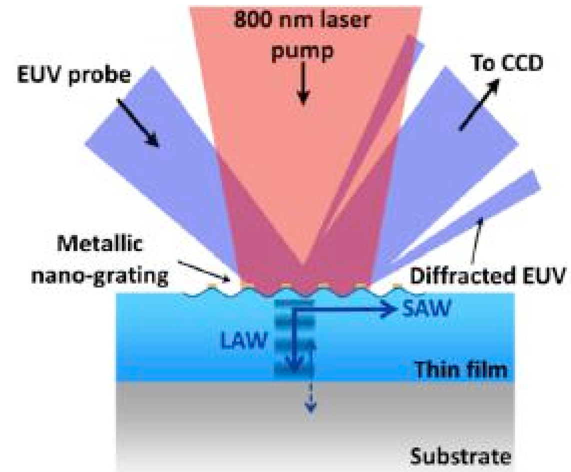
$$v_L = \sqrt{\frac{c_{11}}{\rho}}$$ Longitudinal acoustic wave

$$v_T = \sqrt{\frac{c_{44}}{\rho}}$$ Surface acoustic wave

Excitation of short wavelength SAWs

- Launch short-wavelength acoustic waves from IR impulsive excitation of nano-gratings (CXRO)
- Detect ≈ picometer displacements using coherent EUV beams

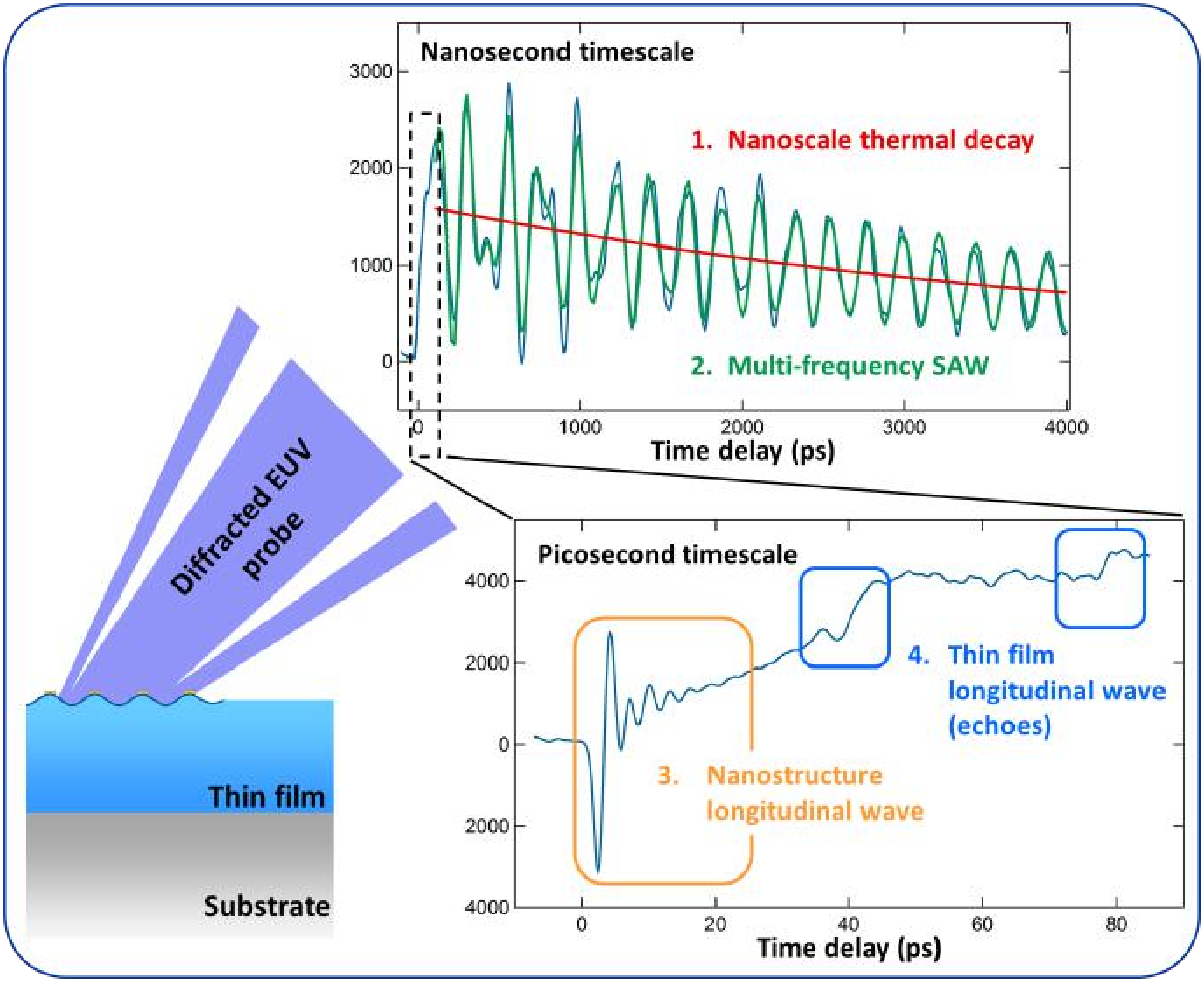




Nardi et al., Nano Lett. ***11****, 4126 (2011)*

Many components to diffracted EUV HHG signal

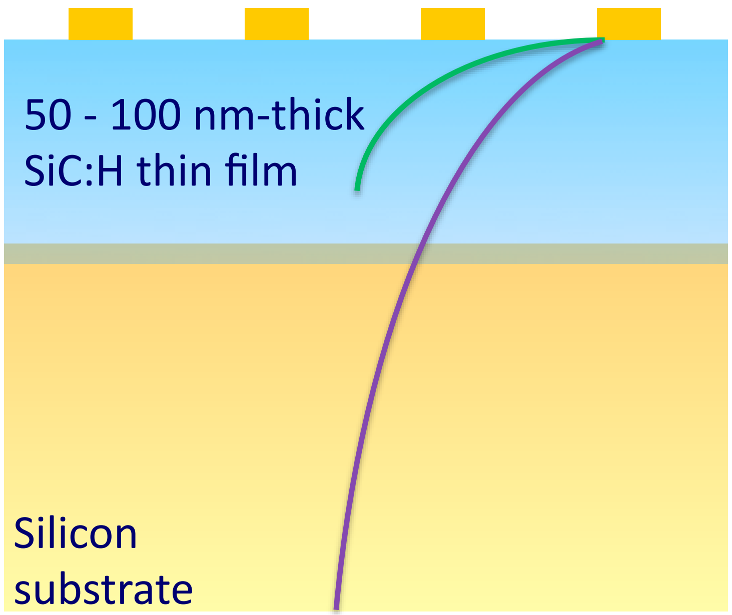
- Diffracted EUV signal from thin film sample has four components:



*Nano Letters **11**, 4126 (2011)*
*PRB **85**, 195431 (2012)*
*SPIE **8324** (2012)*

Why EUV needed

- Acoustic wave penetration depth $\zeta \sim \Lambda/\pi$
- Visible light will not diffract form short wavelength acoustic waves
- The measured transverse velocity changes for shorter wavelengths as the SAW is more and more confined within the thin film.



Short Λ SAW confined within film
v_T **= 2800 m/s (**consistent with SiC:H)

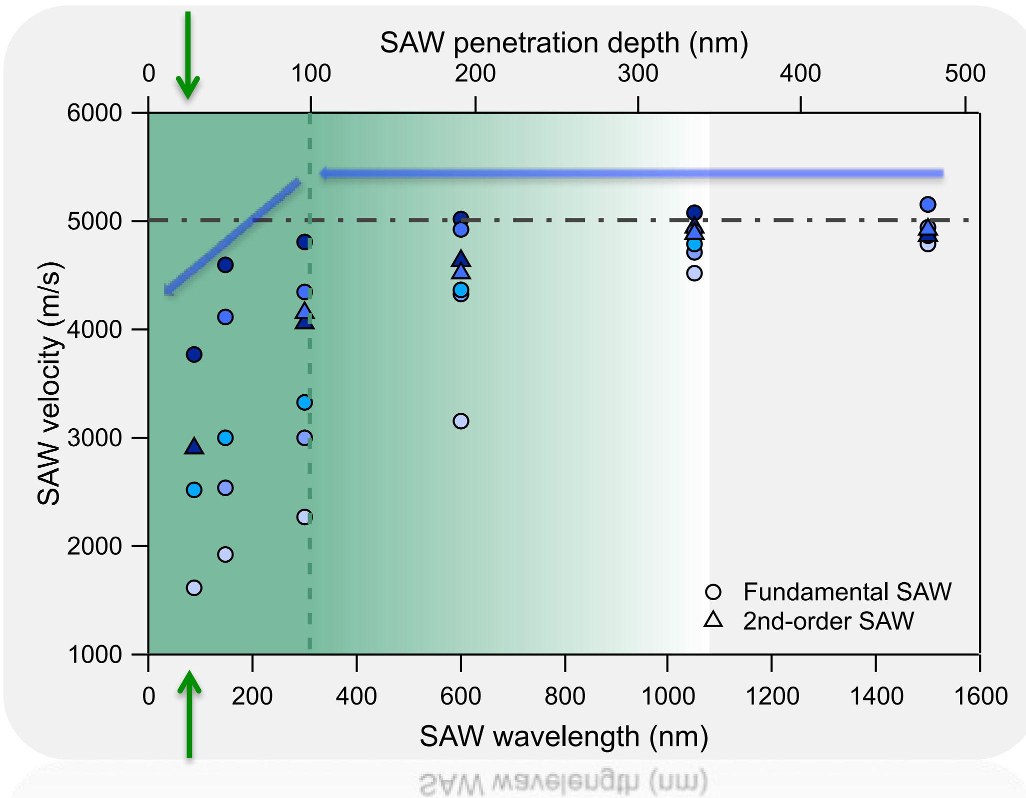
Medium Λ SAW evenly in film and substrate
v_T **= 4250 m/s (**consistent with SiC:H)

Long Λ SAW primarily in substrate
v_T **= 4900 m/s** (consistent with Si)

Results for many SiC:H on Si samples

- **Large-period gratings:** SAW velocity consistent with literature for Si
- **Short-period gratings:** slower velocities associated with softer film materials
- Detected 45nm SAWs - confined to 10nm depths

$\zeta \sim \Lambda/2\pi$



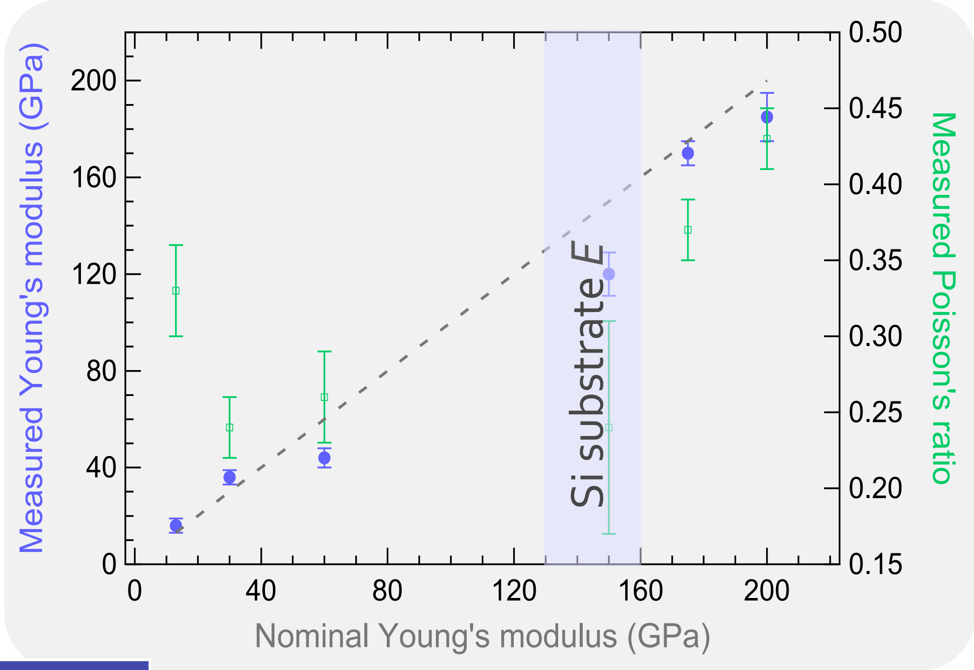
E = 200 GPa

Softer SiC:H films

E = 13 GPa

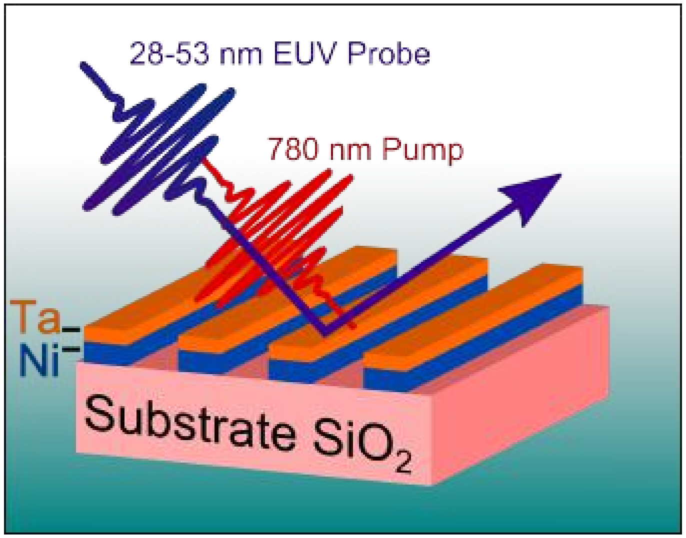
Extracting mechanical properties

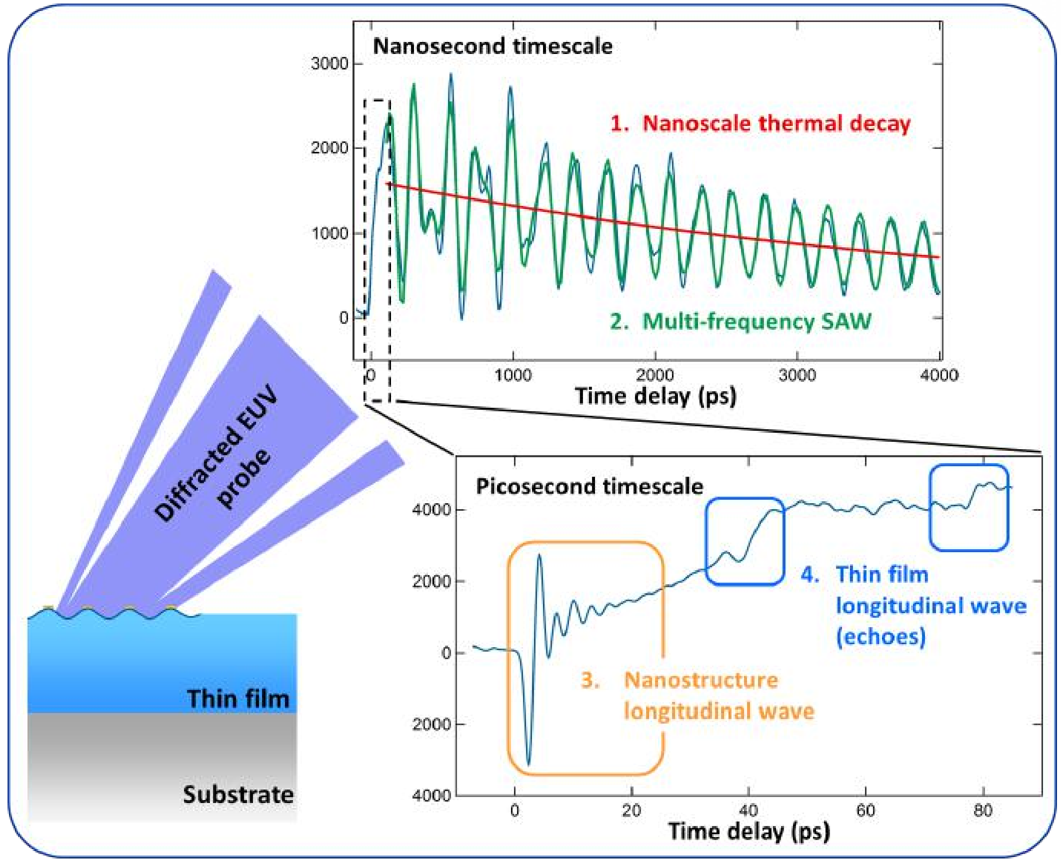
- Compare nominal and measured Young's modulus
- Measure Poisson's ratio using LAW
- Improvements in many aspects can significantly increase accuracy



Nominal E	Measured E	Measured ν
13 GPa (100nm)	16 ± 3 Gpa	0.33 ± 0.03
30 GPa (100nm)	36 ± 3 GPa	0.24 ± 0.02
60 GPa (100nm)	44 ± 4 GPa	0.26 ± 0.03
150 GPa (100nm)	120 ± 9 GPa	0.24 ± 0.07
175 GPa (100nm)	170 ± 5 GPa	0.37 ± 0.02
200 GPa (50nm)	185 ± 10 GPa	0.43 ± 0.02

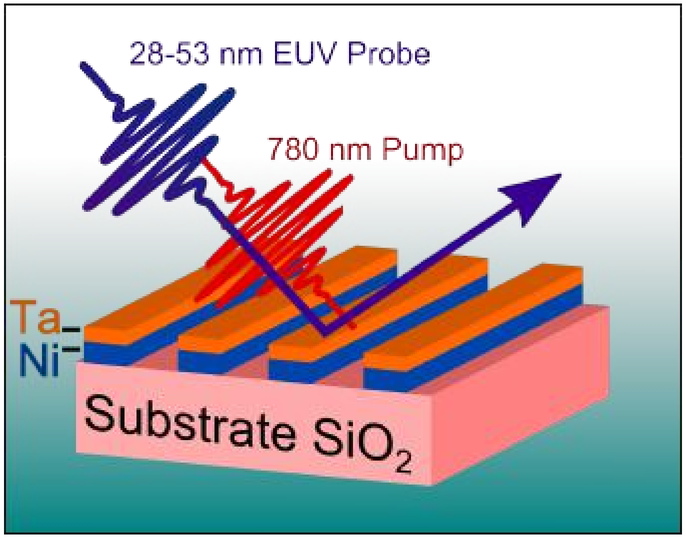
Ultrasensitive to additional mass loading

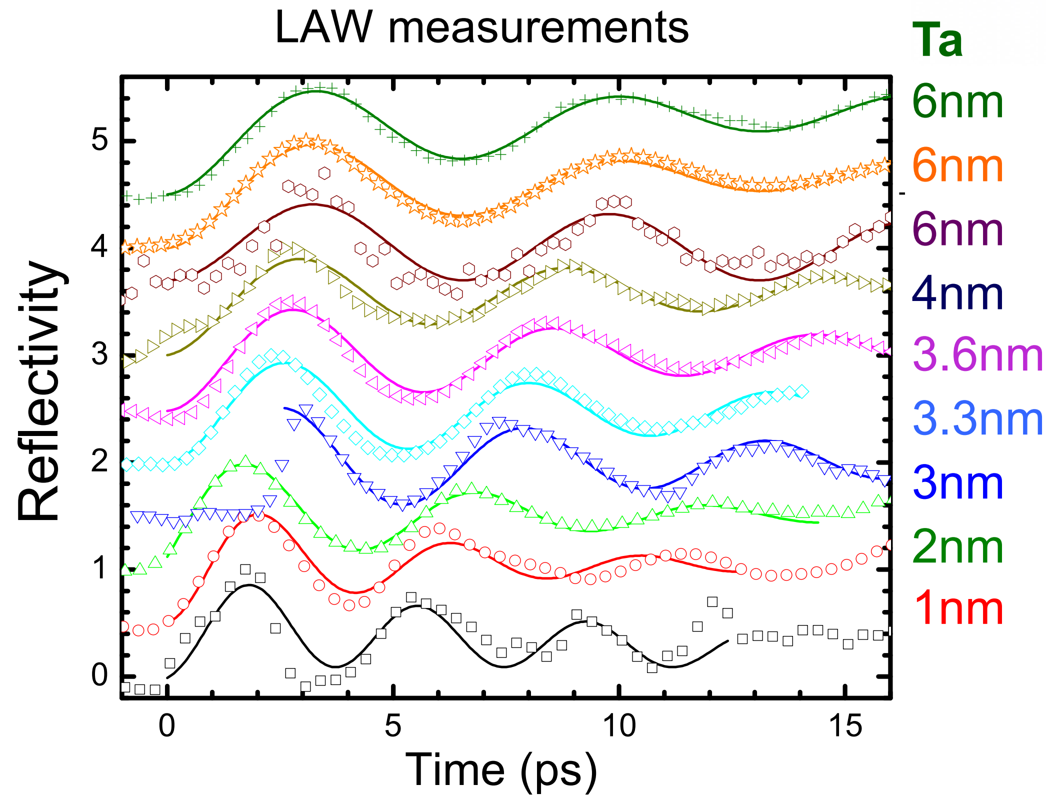


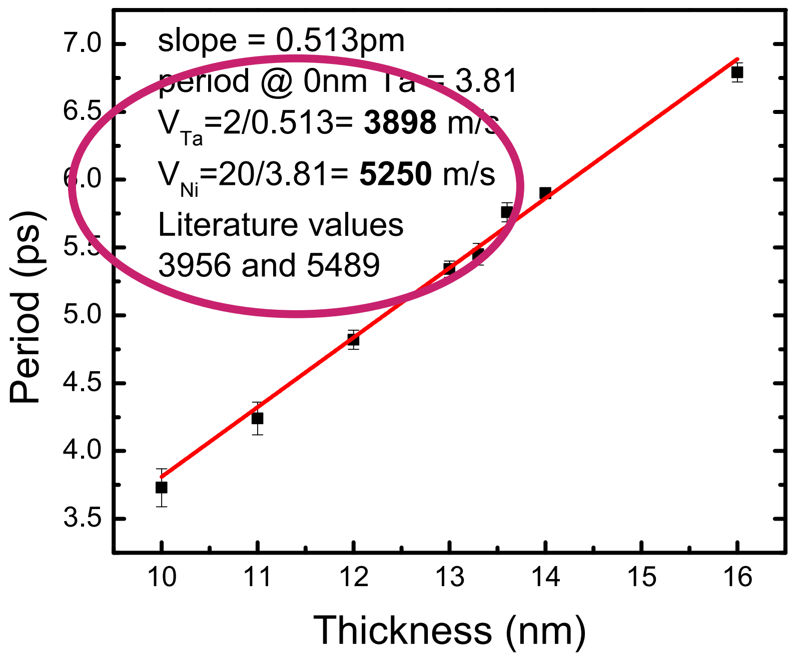


- Presence of additional 1nm layer on nanostructure is easily detected by difference in longitudinal wave velocity
- What is our detection limit?

Sub-monolayer sensitivity ≈ 0.2Å!

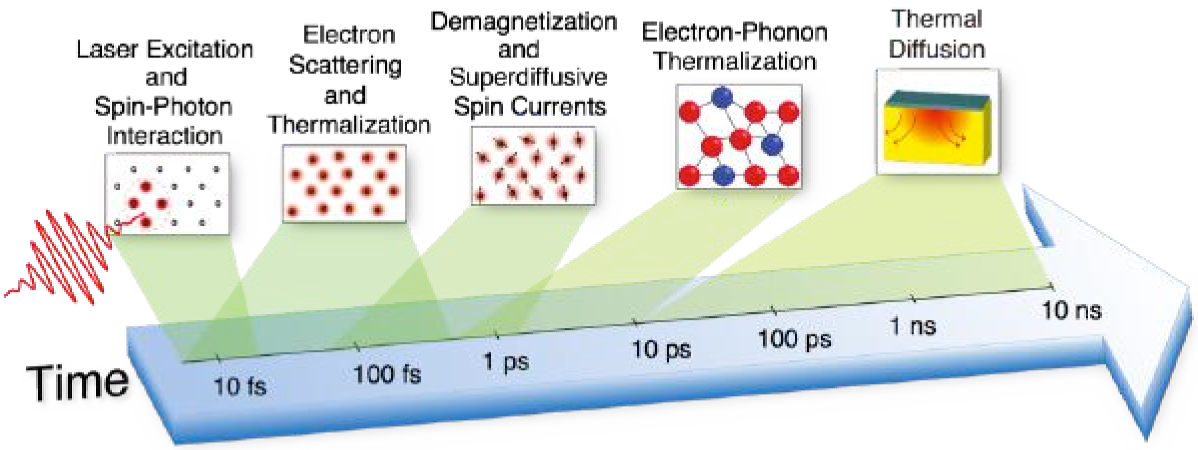




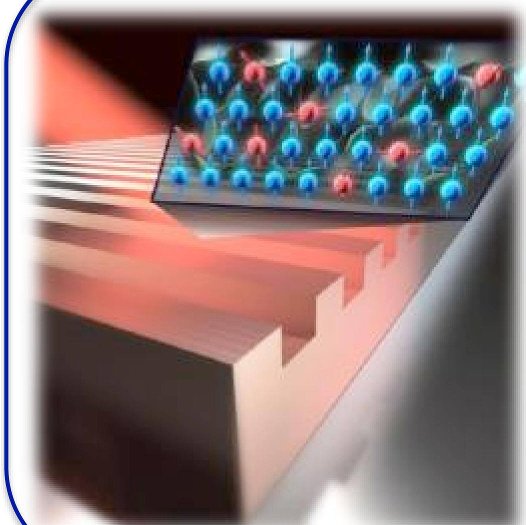


- Sensitive to pm deflection in holographic mode
- Retrieve layer-sensitive velocities
- **Next steps –** 1) extract film density and elastic properties of individual layers as film builds up, and 2) image acoustic/thermal propagation

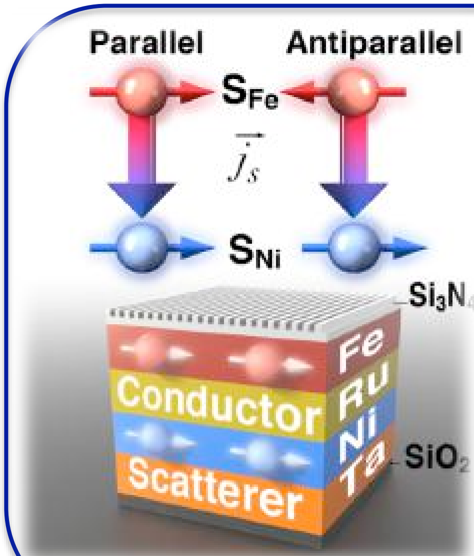
Surprising ultrafast spin dynamics



- 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)



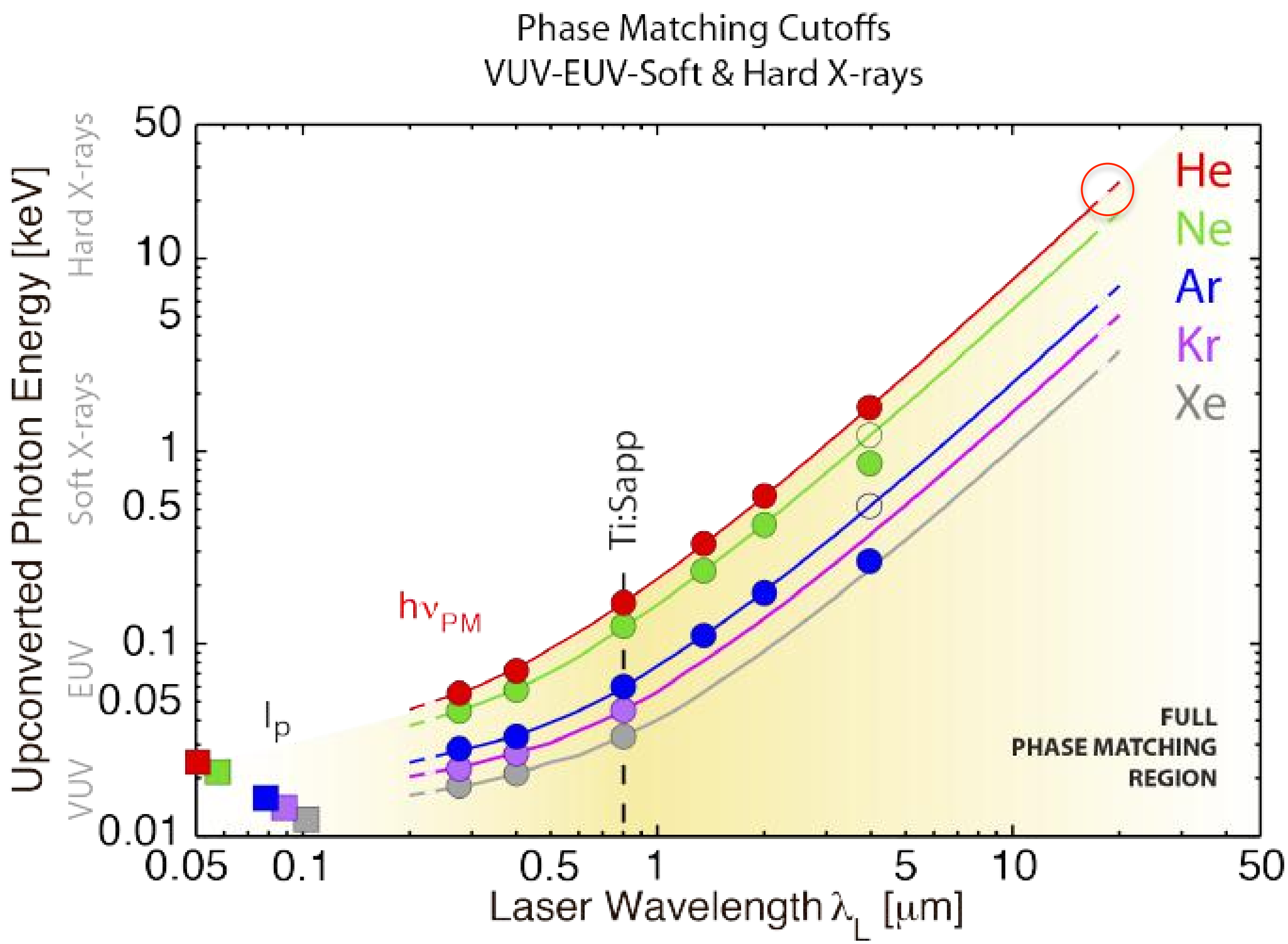
Large, superdiffusive, spin currents can be launched by a femtosecond laser through magnetic multilayers, to enhance or reduce the magnetization of buried layers, depending on their relative orientation

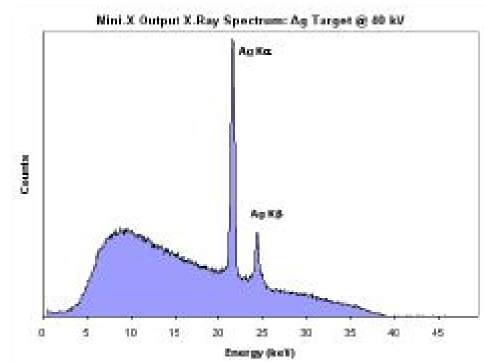
PUBLICATIONS
*PRX **2**, 011005 (2012)*
*PNAS, **109**, 4792 (2012)*
*Nature Commun. **3,** 1037 (2012)*

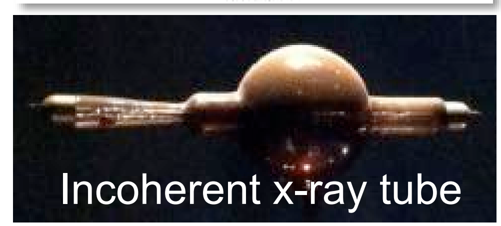
NEWS ARTICLES ABOUT WORK
*Physics **5**, 11 (2012)*
*Physics Today **65** (5), 18 (2012)*
*Physik Journal **11**, Nr. 6, page 26 (2012)*

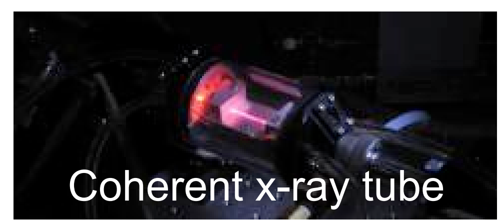
Limits of HHG not yet known!

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

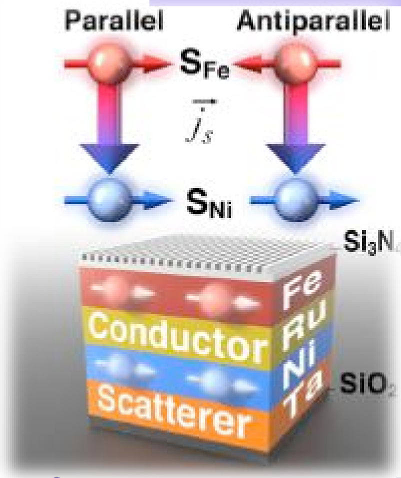




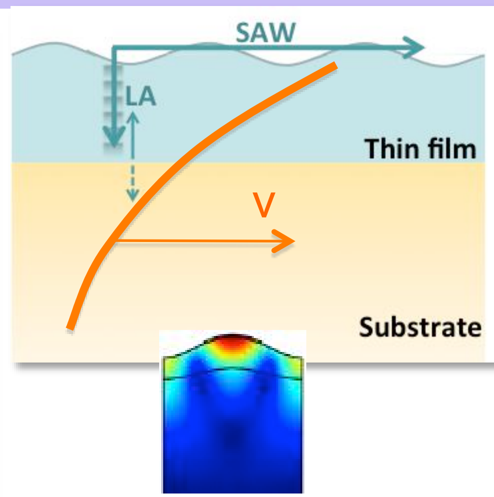


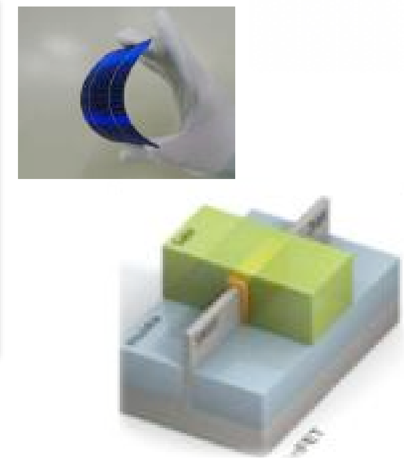


Unique new EUV nanoscience and nanometrology

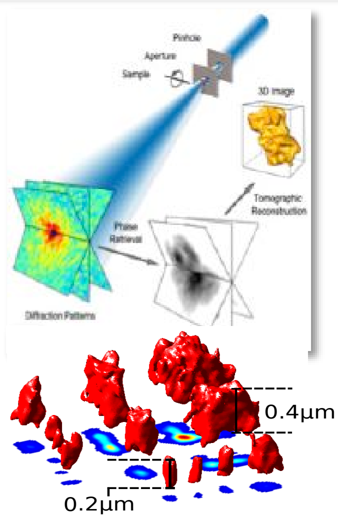


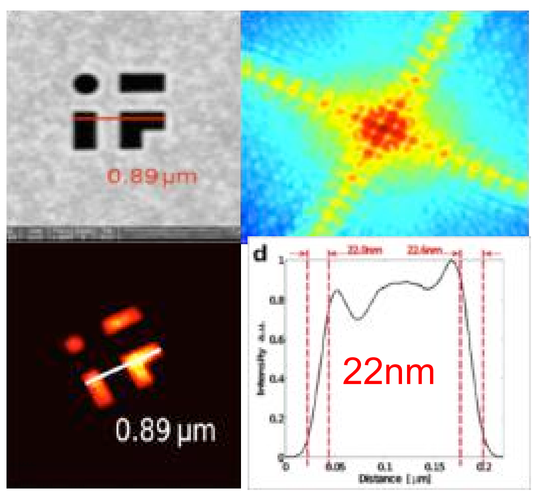
Ultrafast elemental-specific spintronics: *(Nature **471**, 490 (2011), PNAS **109**, 4792 (2012); Nature Comm **3**, 1037 (2012); Nature Comm **3**, 1069 (2012))*



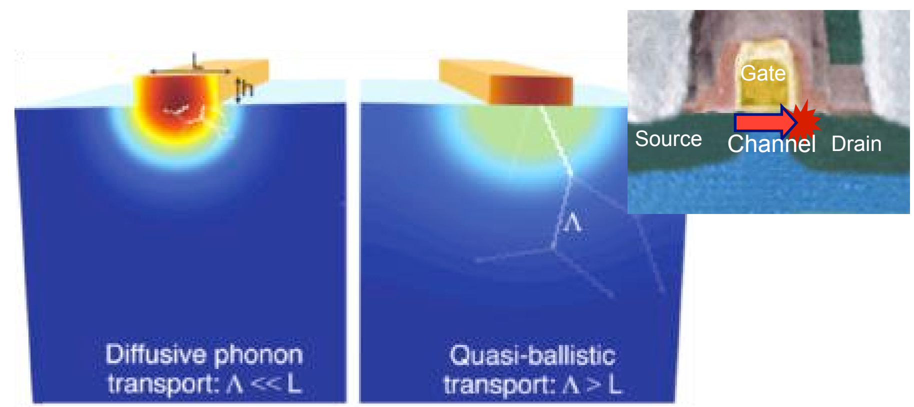


Acoustic nanometrology: thin film metrology *(Nano Letters **11**, 4126 (2011); SPIE, PRB (2012))*



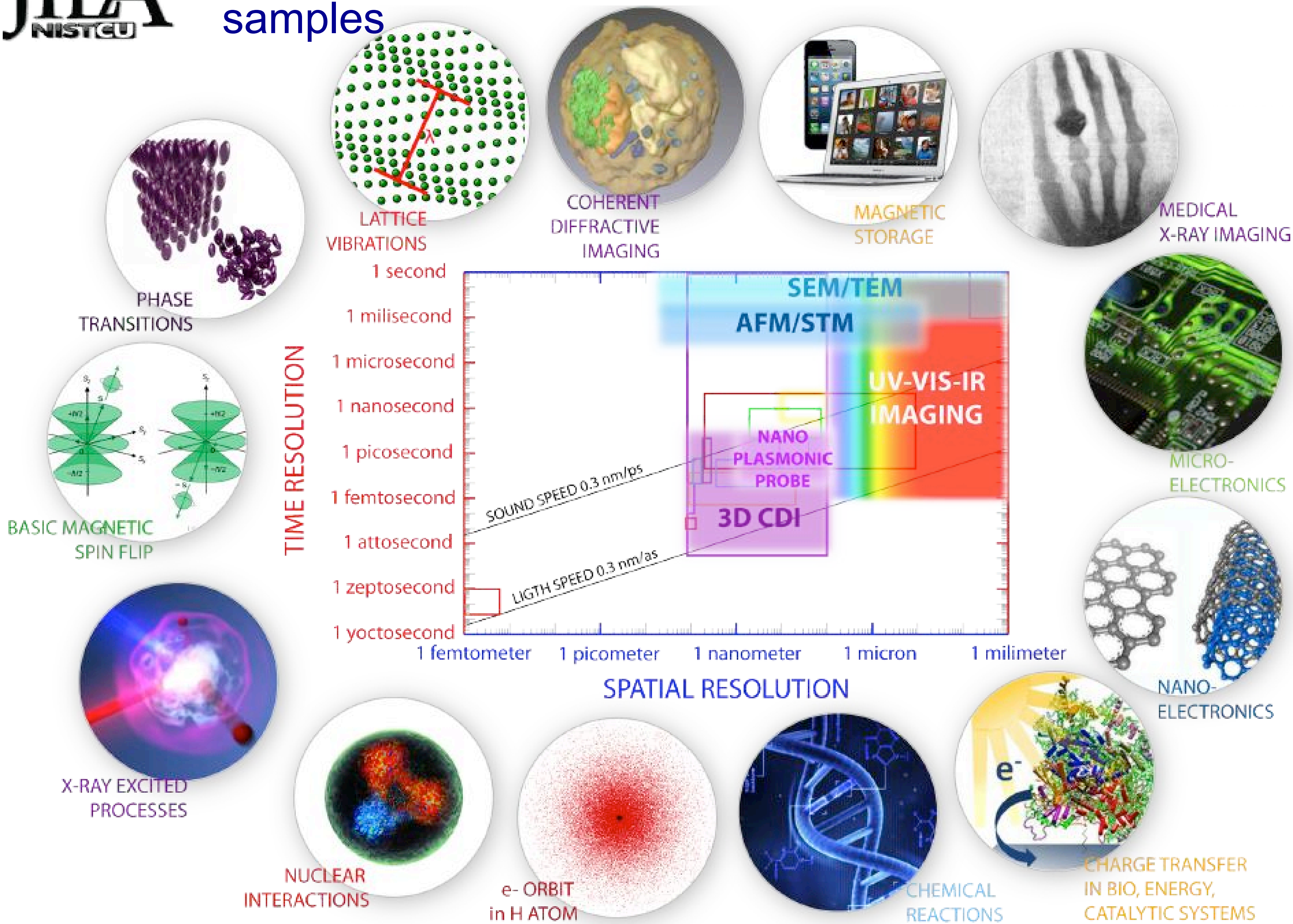


Nanoscale imaging: Record tabletop 22nm resolution *(Op. Ex. **19**, 22470 ('11); **17**, 19050 ('12); Nature **463**, 214 (2010))*



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

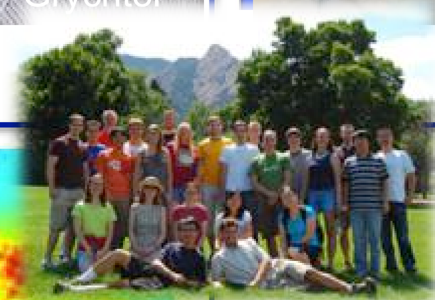
HHG – unique probe of nanoscale function in thick samples

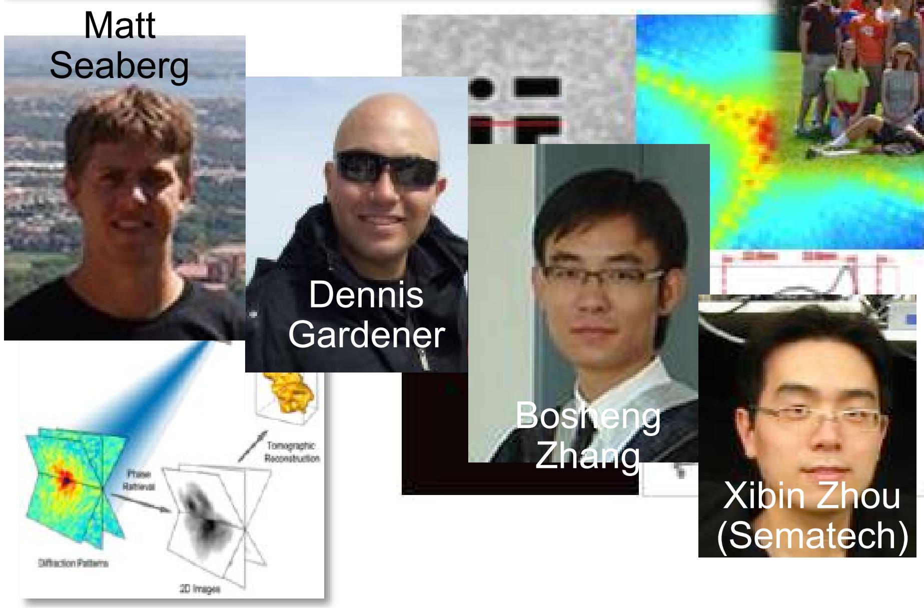


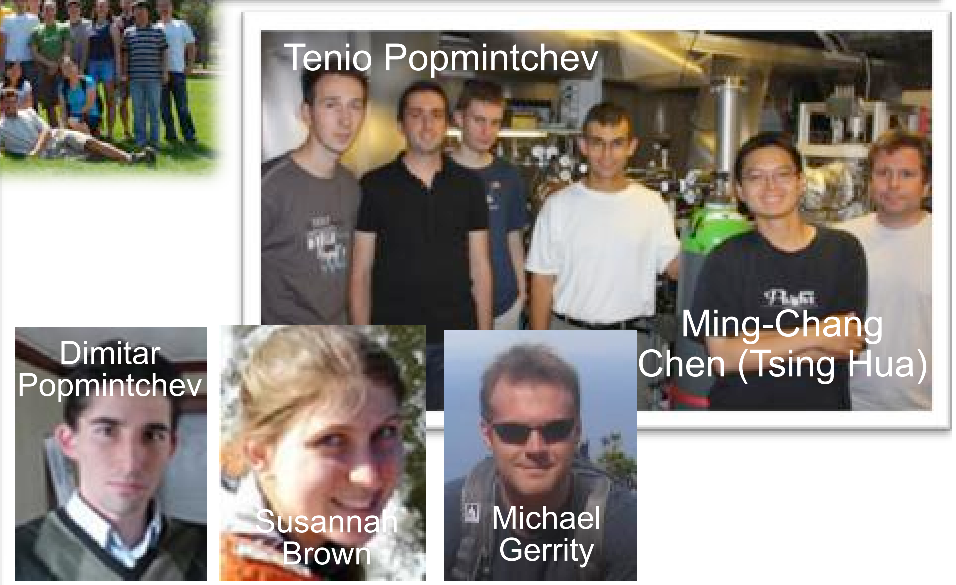
Students and postocs

Magnetic and material switching speeds, spin transport

Nanoscale acoustic metrology and energy transport







Nanoscale imaging

High harmonic sources