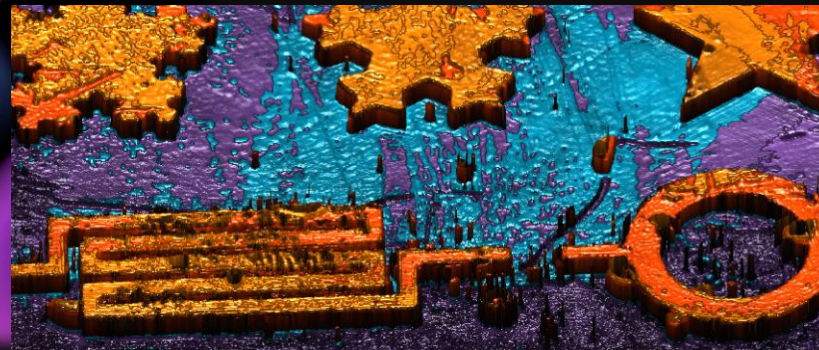


Tabletop-scale EUV coherent imaging using High Harmonic Light

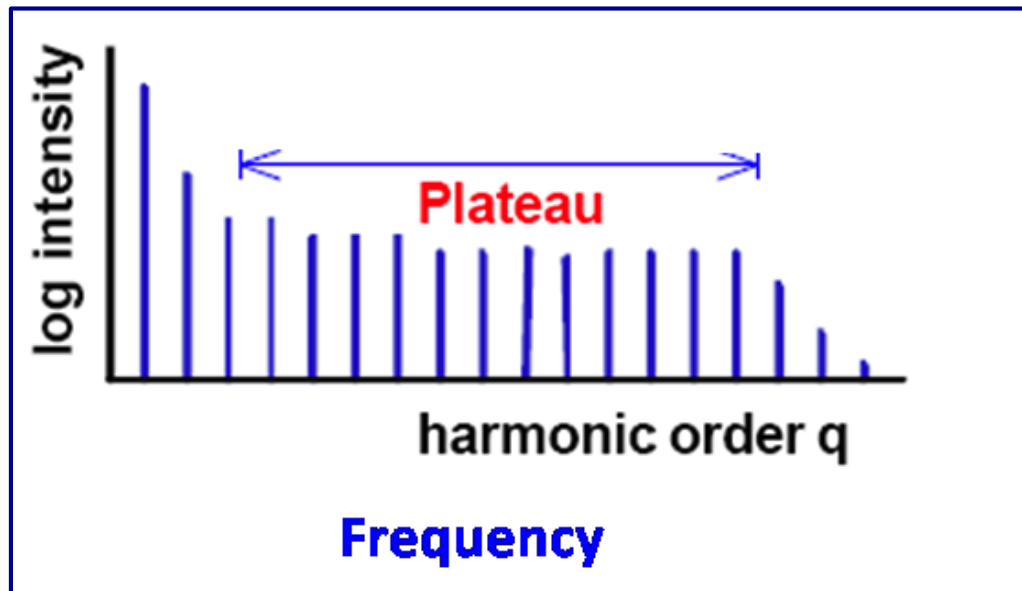
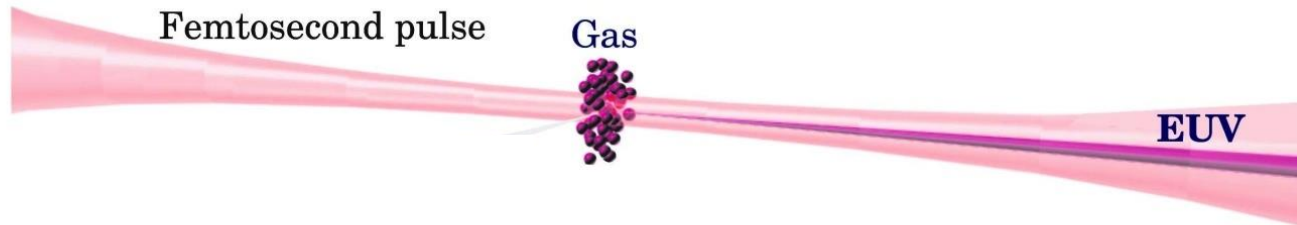
Henry C. Kapteyn
KMLabs Inc. and JILA



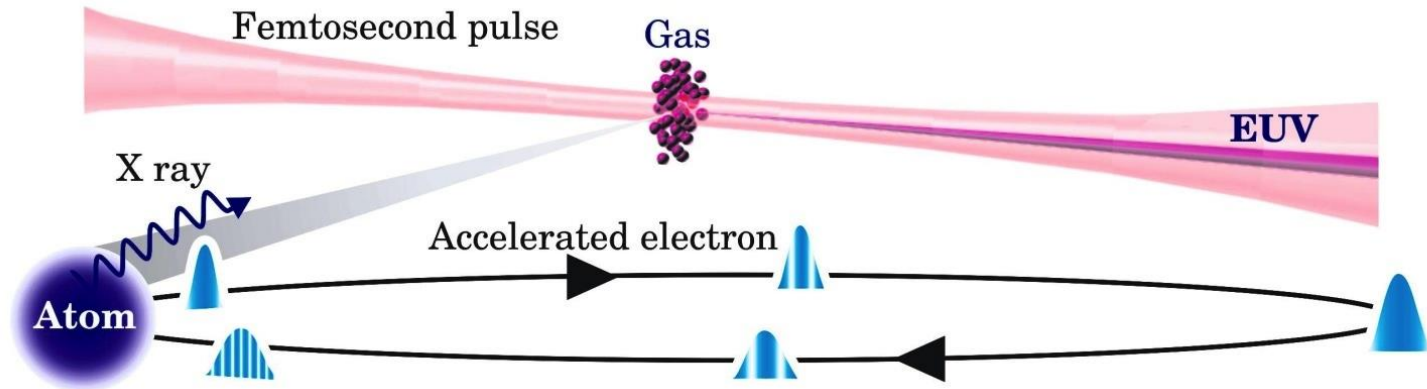
- Tabletop coherent EUV light sources – high-order harmonic generation.
- Revolution in coherent imaging: 14 nm spatial resolution @13.5nm.
- Progress in commercial “tabletop x-ray laser” light sources– **the KMLabs XUUS₄TM**.



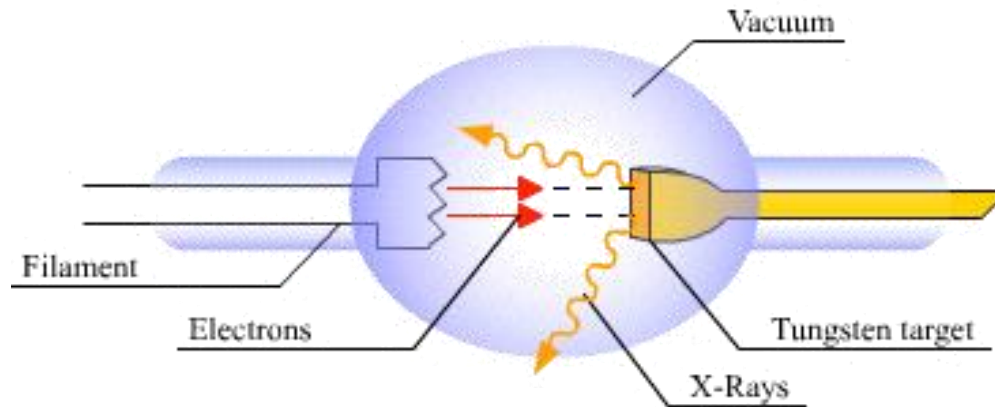
- Take a few-cycle ($\sim 10^{-14}$ sec) laser pulse, focus to $\sim 10^{14}$ W cm⁻²:

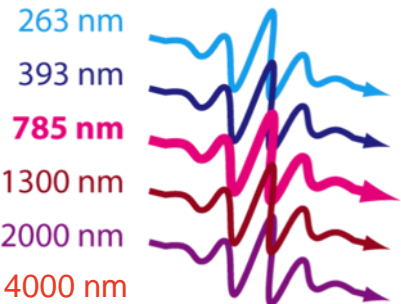


- **High Harmonic Generation**

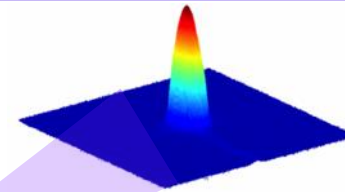


- **Röntgen X-ray Tube**

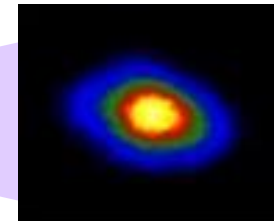
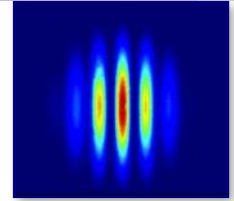




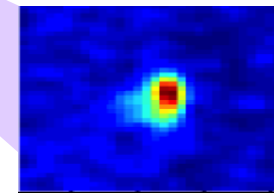
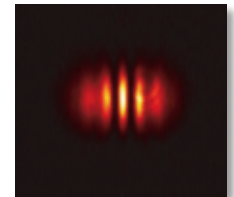
High pressure waveguide



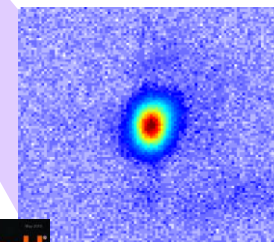
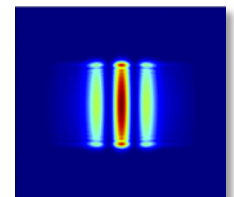
30nm HHG beam (2002)



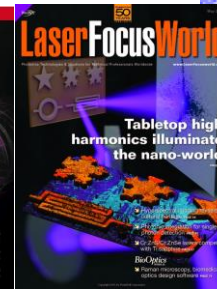
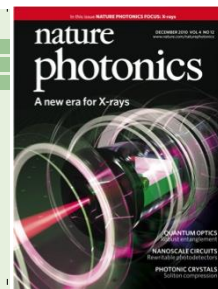
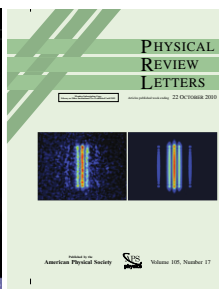
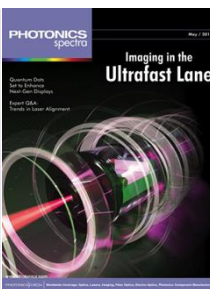
13nm HHG beam (2004)



3nm HHG beam (2010)



1nm HHG beam (2012)



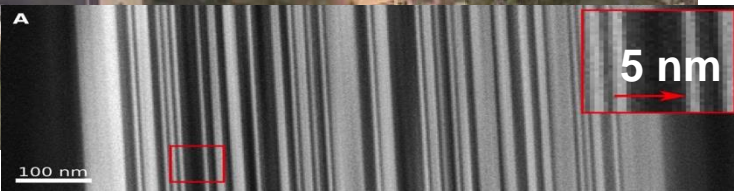
Science **280**, 1412 (1998)
Science **297**, 376 (2002)
Science **336**, 1287 (2012)
Science **348**, 530 (2015)
Science **350**, 1225 (2015)

Facility scale

- Synchrotron and free electron lasers
- **EUV to 12 keV (EUV to hard X-rays)**
- Nano to femto time resolution
- **High flux**
- Tunable
- **Facility scale** beamline w/support

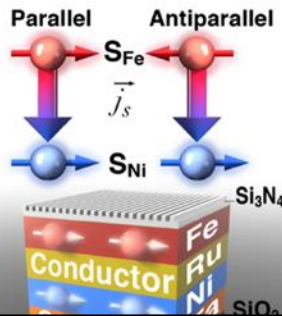
Tabletop

- High harmonic sources
- **mid-IR to 1 keV (EUV to soft X-rays)**
- Sub-femtosecond time resolution
- **Lower flux at higher hv**
- Hyperspectral
- **Tabletop** for easy student/industry access

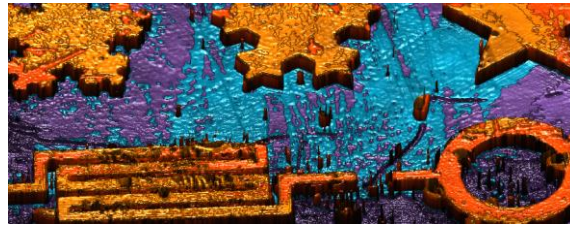


Spin scattering and transport

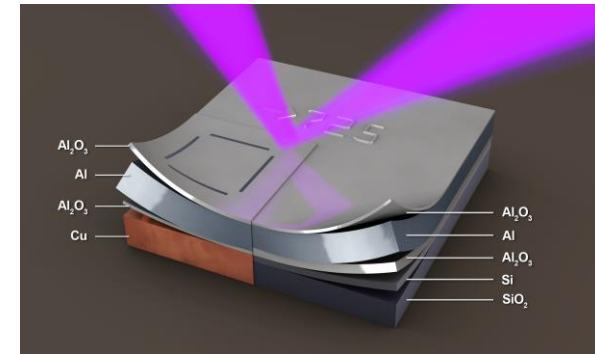
PNAS **109**, 4792 (2012)
 Nat. Comm. **3**, 1037 (2012)
 PRL **110**, 197201 (2013)
 arXiv:1401.4101 (2014)



Quantitative imaging at λ limit



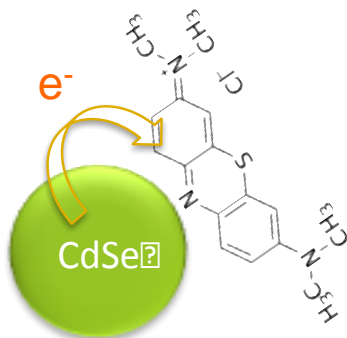
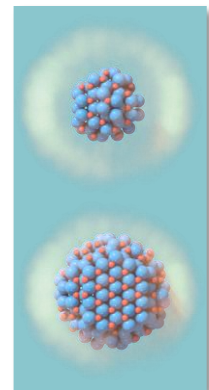
Elastic properties, dopants, ellipsometry, interfaces



Nanoletters, in press (2016)
 Submitted (2016)
 PRB **85**, 195431 (2012)
 no Letters **11**, 4126 (2011)

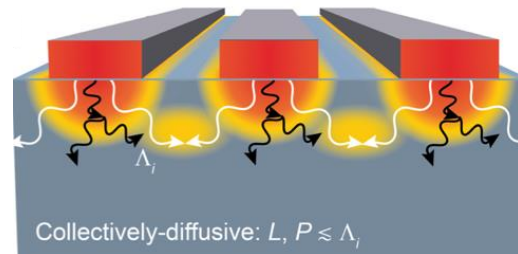
jila.colorado.edu/kmggroup

Charge transport in nano, energy science



Nano Lett. **13**, 2924 (2013)
 JACS **137**, 3759 (2015)

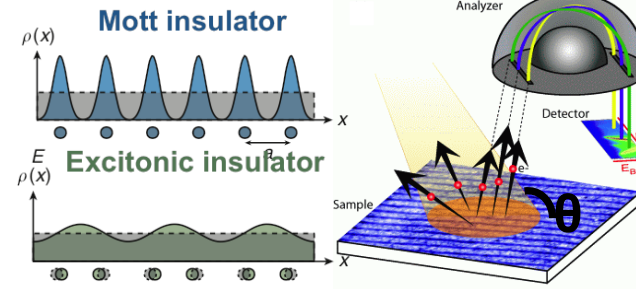
Nanoscale energy transport



Nature Mat. **9**, 26 (2010)
 PNAS **112**, 4846 (2015)



Electronic properties: full band structure (ARPES)



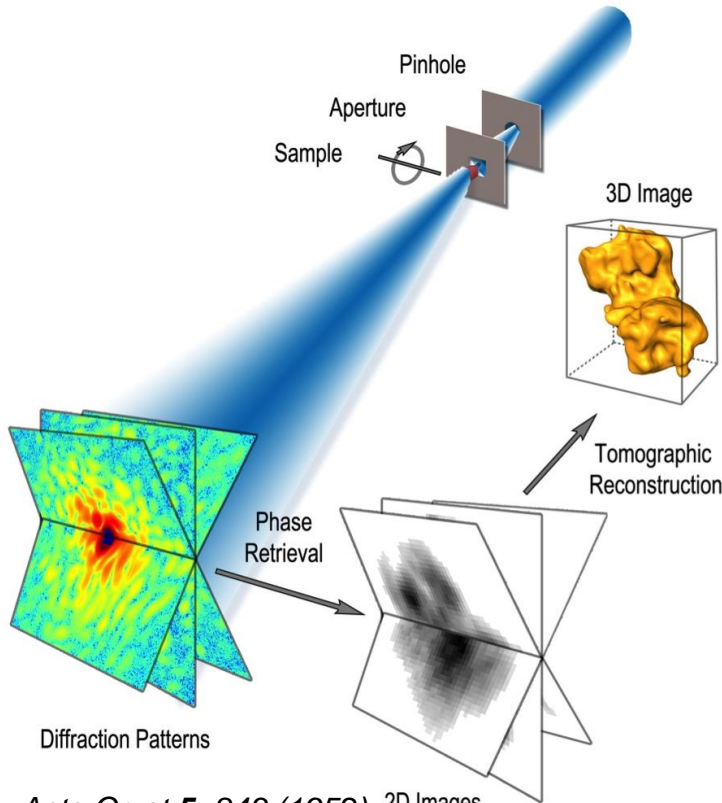
Nature **471**, 490 (2011)
 Nat. Comm **3**, 1069 (2012)
 PRL **112**, 207001 (2014)
 PRB **92**, 041407(R) (2015)
 Science, in press (2016)



REVIEW

Beyond crystallography: Diffractive imaging using coherent x-ray light sources

Jianwei Miao,^{1*} Tetsuya Ishikawa,² Ian K. Robinson,^{3,4} Margaret M. Murnane⁵

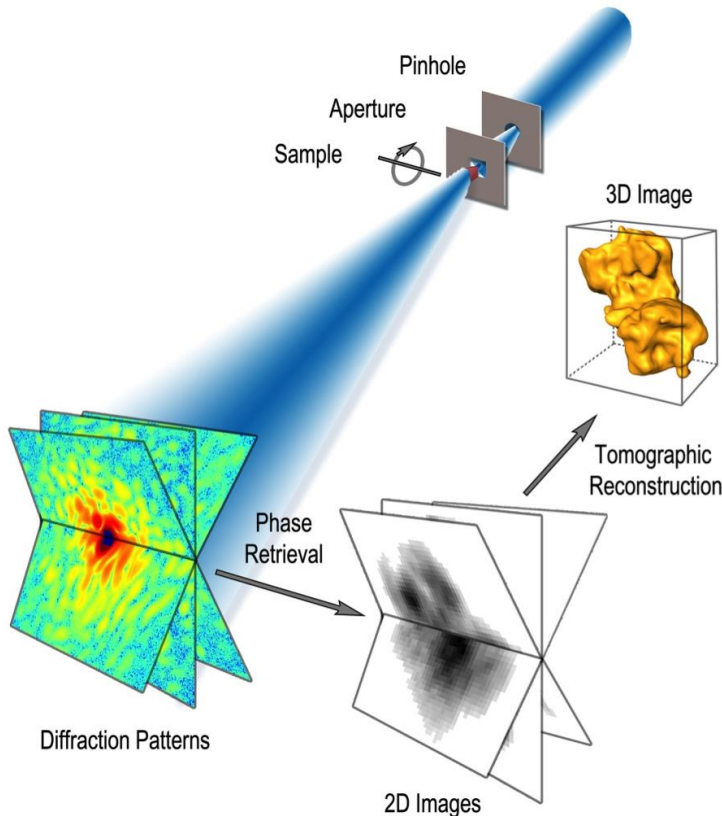


- **Diffraction-limited imaging $\approx \lambda/2NA$**
- Image thick samples in 3D
- Inherent contrast for X-rays
- **Phase and amplitude image contrast**
 - Transmission or reflection
- Robust to vibrations
- **Most photon-efficient form of imaging!**

Sayre, *Acta Cryst* **5**, 843 (1952) 2D Images
 Fienup, *Opt. Lett.* **3**, 27 (1978)
 Miao et al., *Nature* **400**, 342 (1999)
 Miao et al., *Science* **348**, 530 (2015)

Initial approaches to CDI (until 2011)

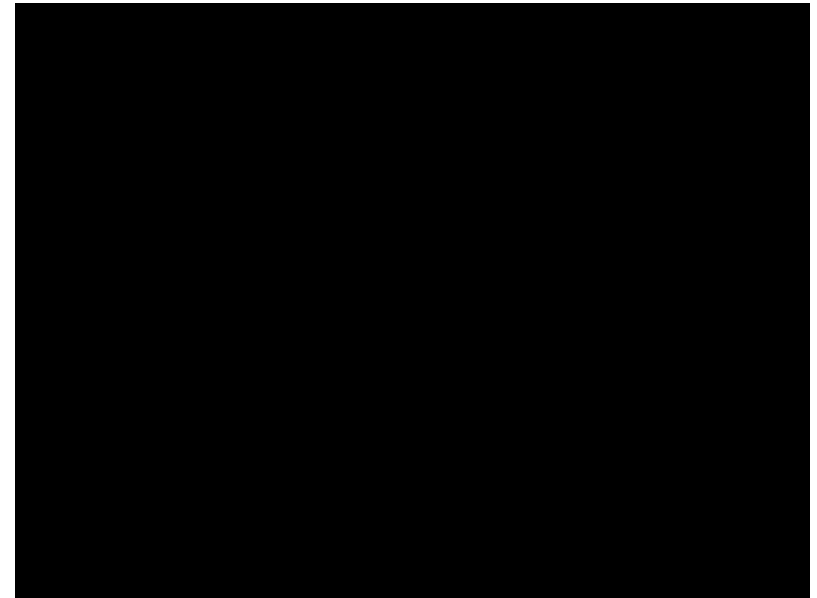
- Required isolated sample or beam
- Transmission mode only



Fienup, *Opt. Lett.* **3**, 27 (1978)
 Miao *et al.*, *Nature*, **400**, 342 (1999)

Advanced CDI (2016)

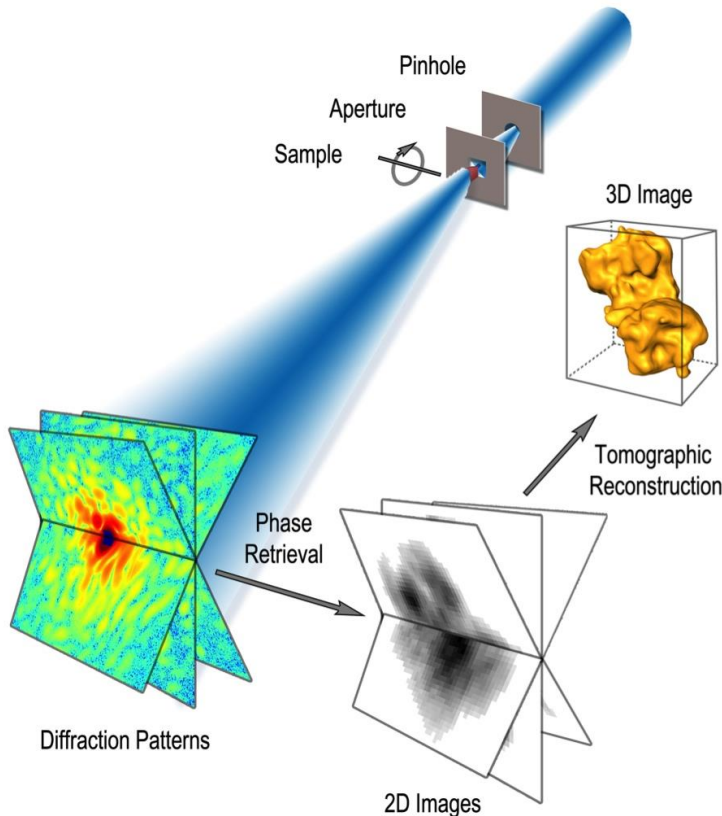
- Ptychographic CDI with overlapping beams
- Robust reflection and transmission modes
- Absolute interface structure determination
- 3D structure w/o tilting or sectioning
- Hyperspectral, multibeam, direct retrieval



Rodenburg *et al.*, *PRL* **98**, 034801 (2007)
 Thibault *et al.*, *Science* **321**, 379 (2008)
 Maiden *et al.*, *Ultramicroscopy* **109**, 1256 (2009)

Initial approaches to CDI (until 2011)

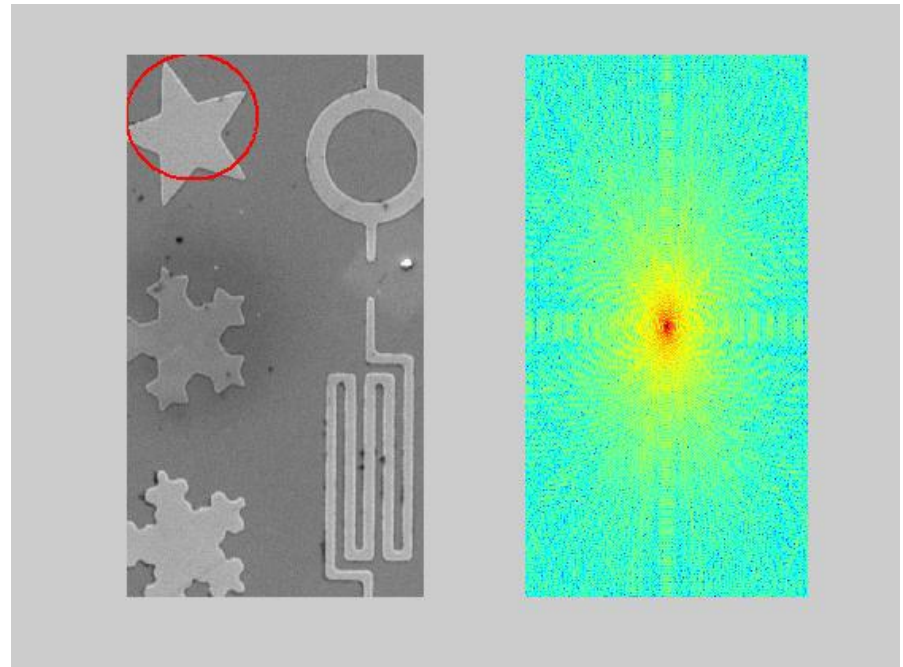
- Required isolated sample or beam
- Transmission mode only



Fienup, *Opt. Lett.* **3**, 27 (1978)
Miao *et al.*, *Nature*, **400**, 342 (1999)

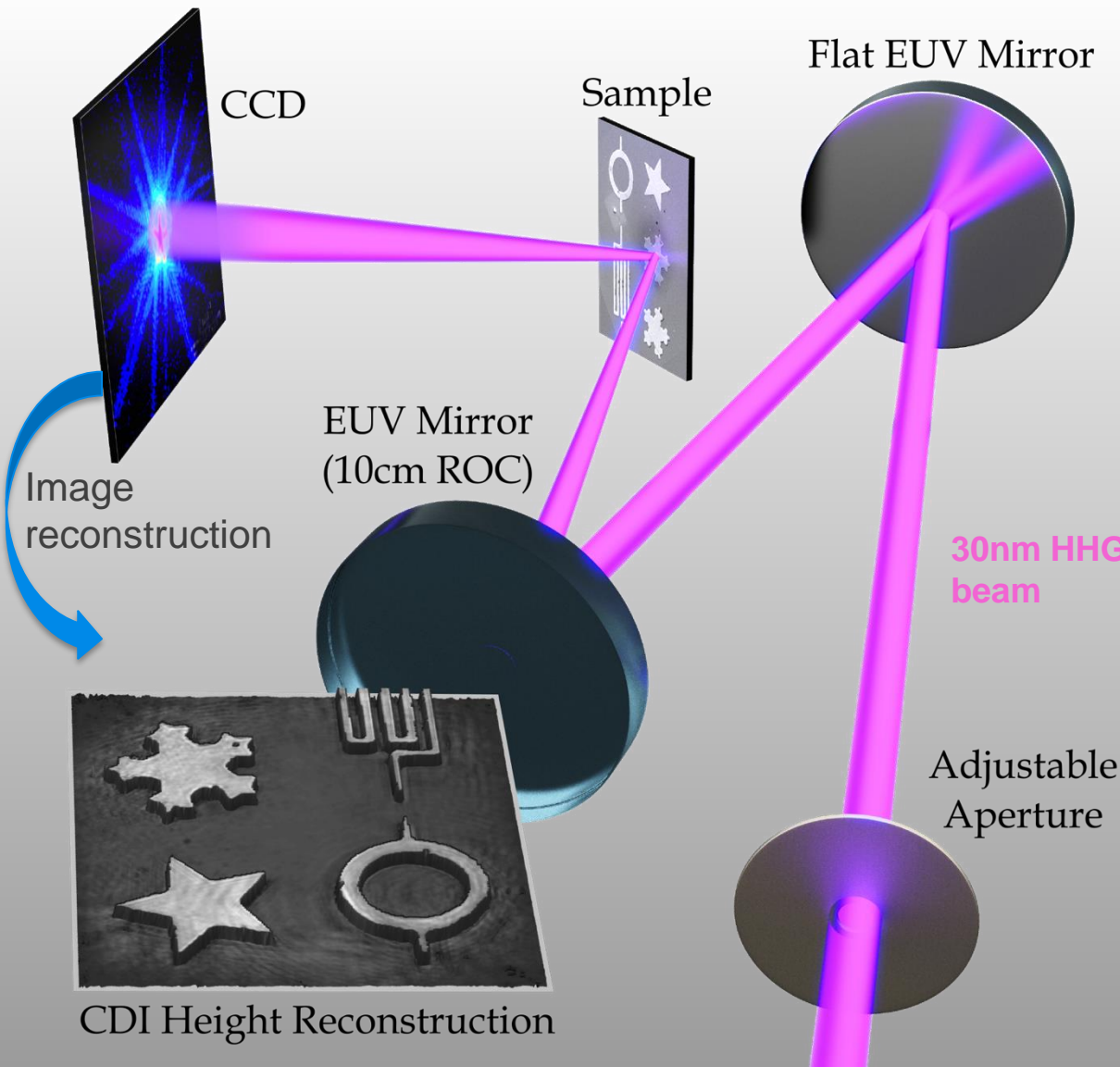
Advanced CDI (2016)

- Ptychographic CDI with overlapping beams
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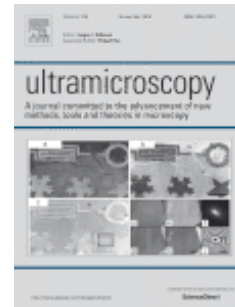
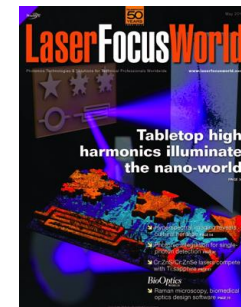
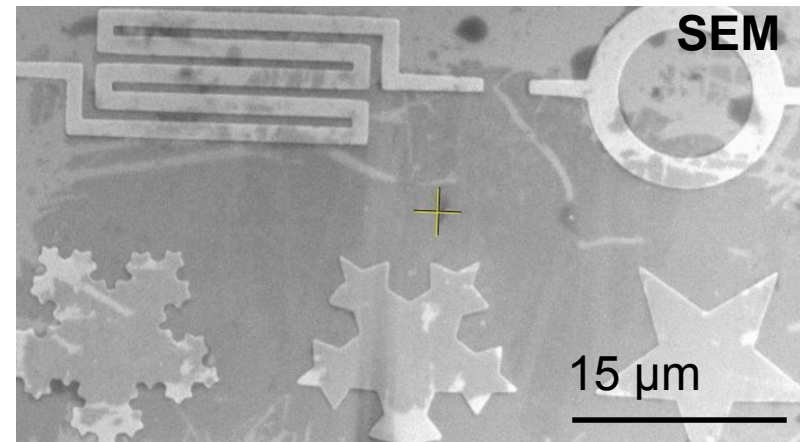
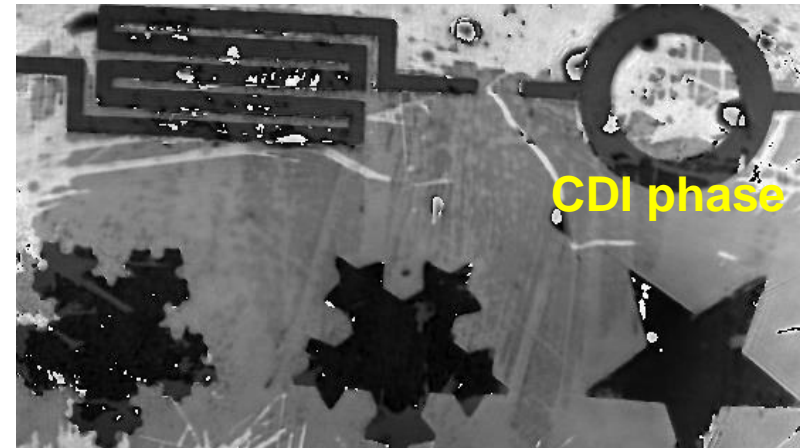
Rodenburg *et al.*, *PRL* **98**, 034801 (2007)
Thibault *et al.*, *Science* **321**, 379 (2008)
Maiden *et al.*, *Ultramicroscopy* **109**, 1256 (2009)

General tabletop reflection-mode full field microscope

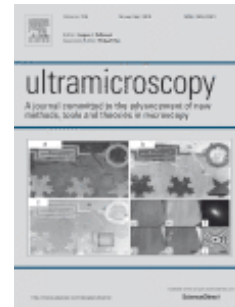
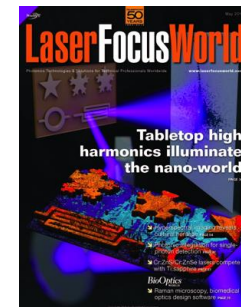
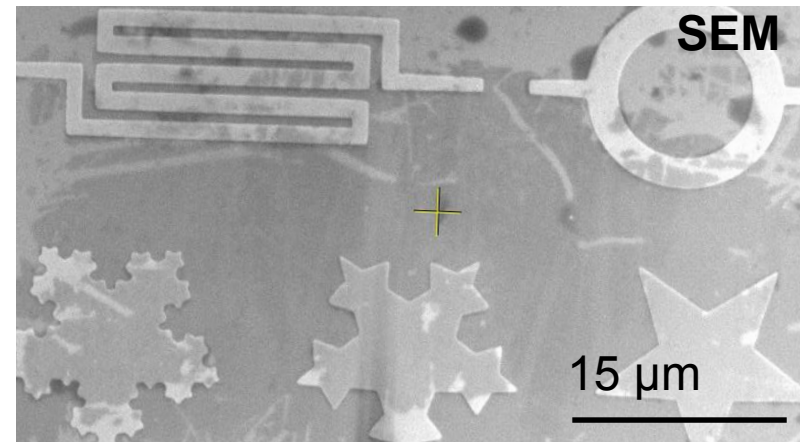
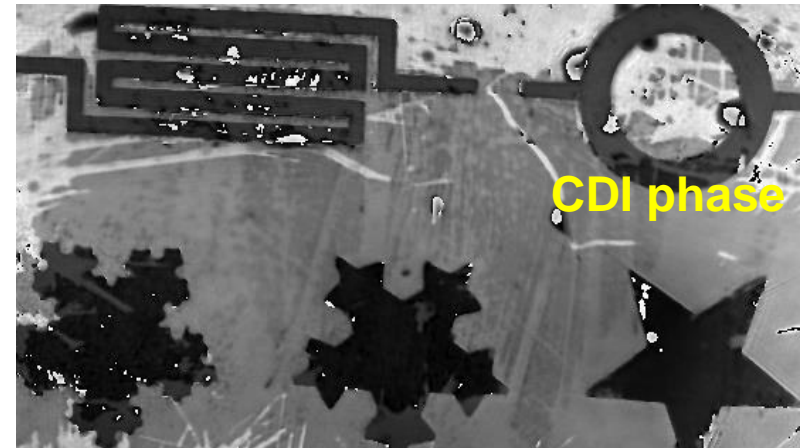


- Full field image of extended objects
- Arbitrary angle of incidence with tilted-plane correction
- Algorithm can correct for imperfect scanning stages
- Can use multiple colors and beams for elemental, chemical, spin contrast
- Reflection and transmission
- Limits in spatial/temporal resolution, speed, not known

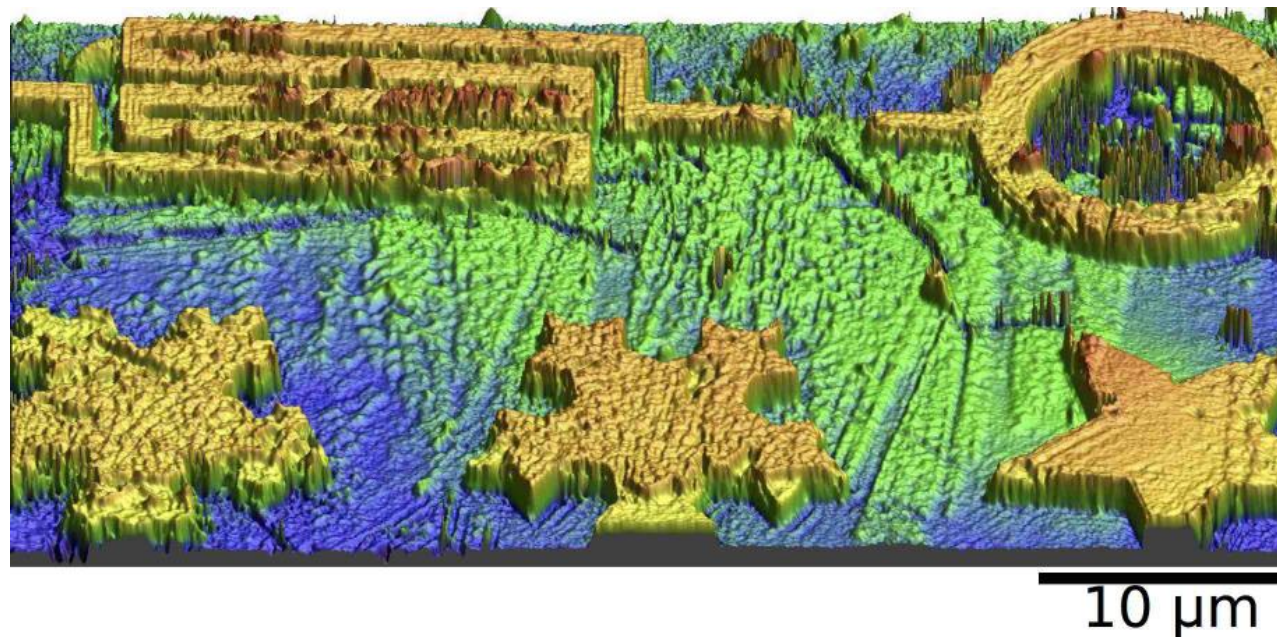
- Better contrast images than JILA SEM
 - phase contrast, element-specific reflectance
- 3D imaging: spatial resolution 1.3λ horizontal (<40 nm), $<5\text{\AA}$ profile height
- <1 minute HHG exposure time for full image (old laser, bad optics)
- Less damage than AFM or SEM
- Unlimited working distance
- Faster detector readout needed
 - <1 min exposure; >90 min readout
- New cluster image reconstruction, detectors, and lasers being implemented as KMLabs / JILA collaboration



- Better contrast images than JILA SEM
 - phase contrast, element-specific reflectance
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 - <1 min exposure; >90 min readout
- **New cluster image reconstruction, detectors, and lasers being implemented as KMLabs / JILA collaboration**



Quantitative CDI: height/composition/tomography maps



50 YEARS

May 2015

LaserFocusWorld
Photonics Technologies & Solutions for Technical Professionals Worldwide

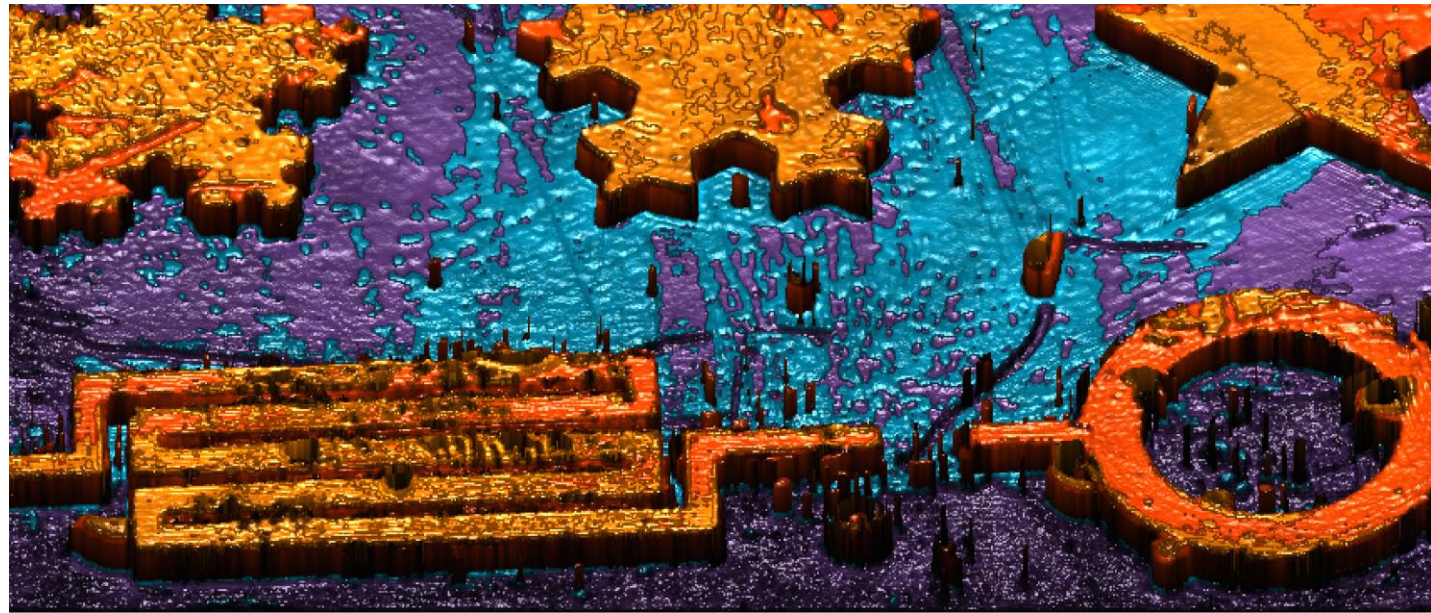
www.laserfocusworld.com

Tabletop high harmonics illuminate the nano-world

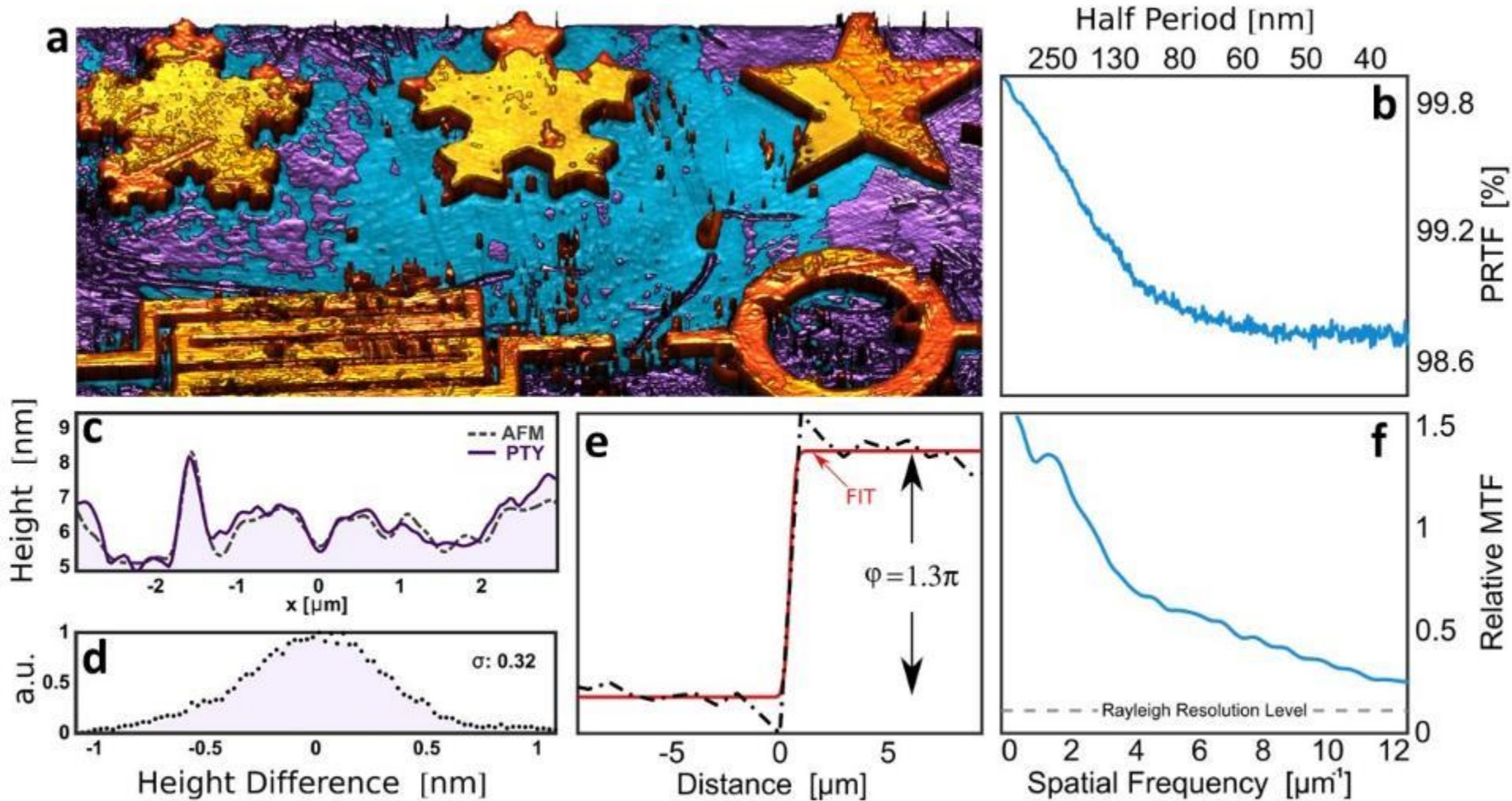
- Hyperpectral imaging reveals collateral heritage PAGE 54
- Photonic integration for single-photon detection PAGE 71
- Cr:ZnS/Cr:ZnSe lasers compete with Ti:sapphire PAGE 47

BioOptics
Raman microscopy, biomedical optics design software PAGE 71

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Determining the spatial resolution – 3 approaches

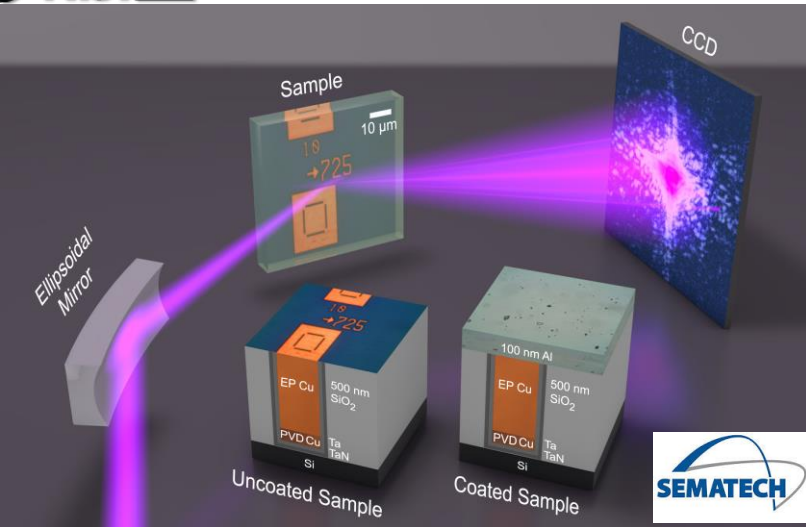


1. Comparison with AFM

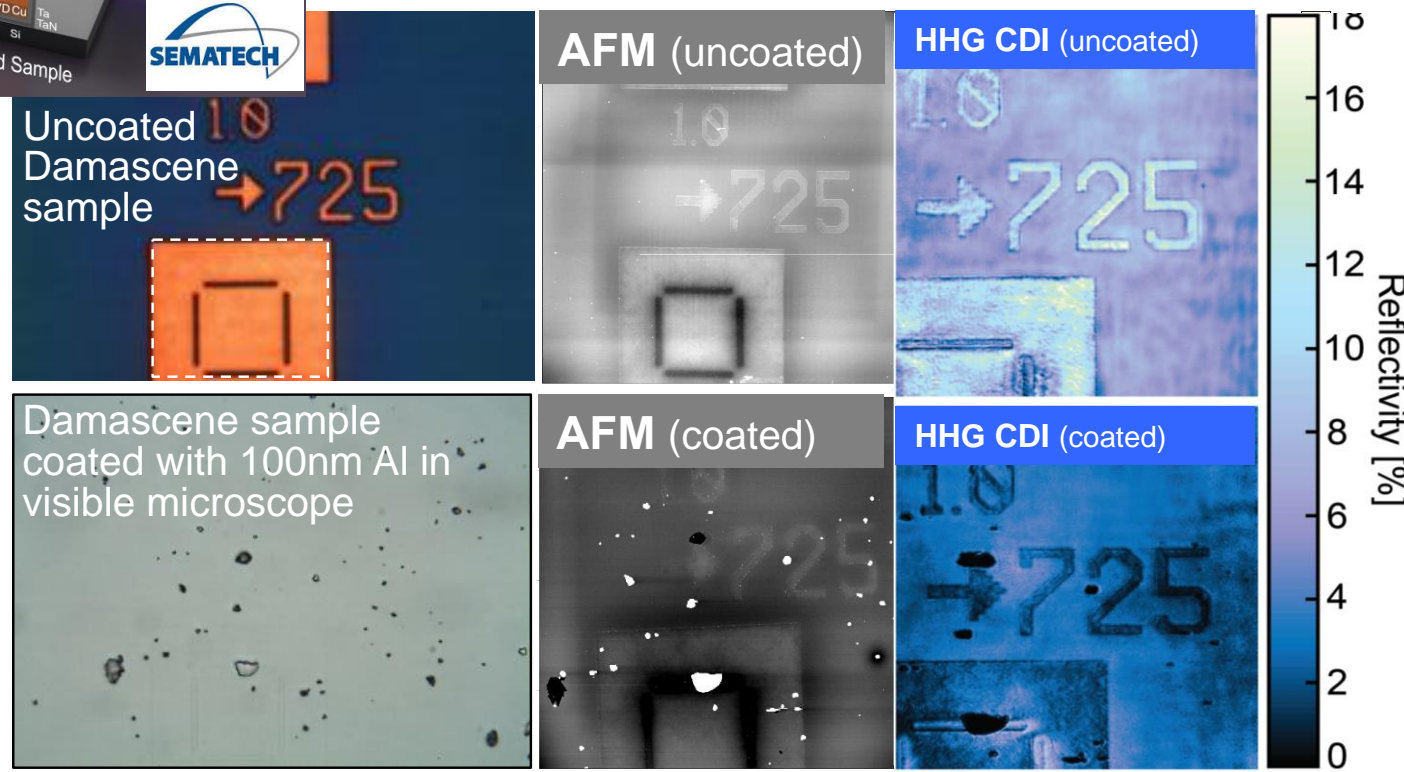
2. Lineout at edge

3. Spatial frequencies

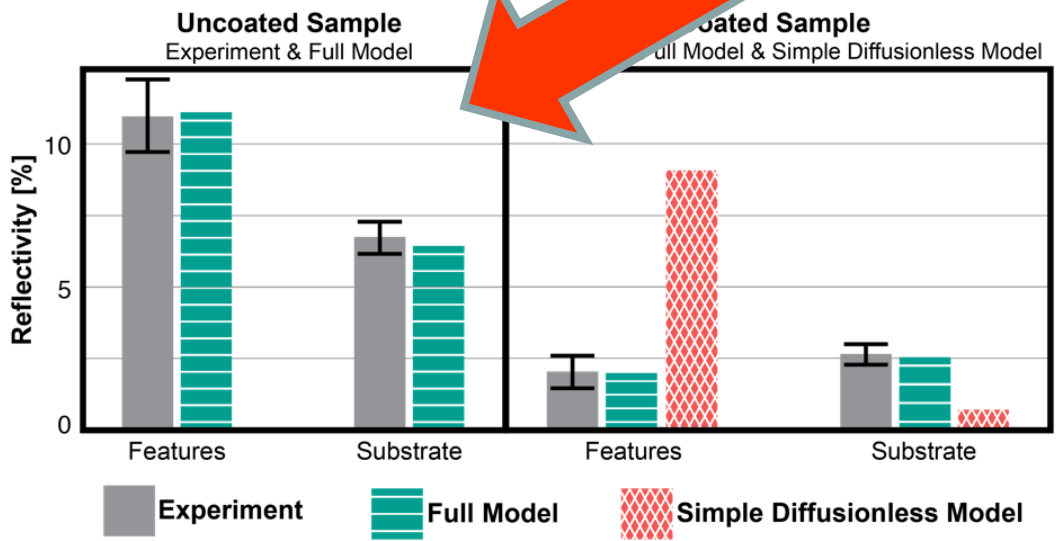
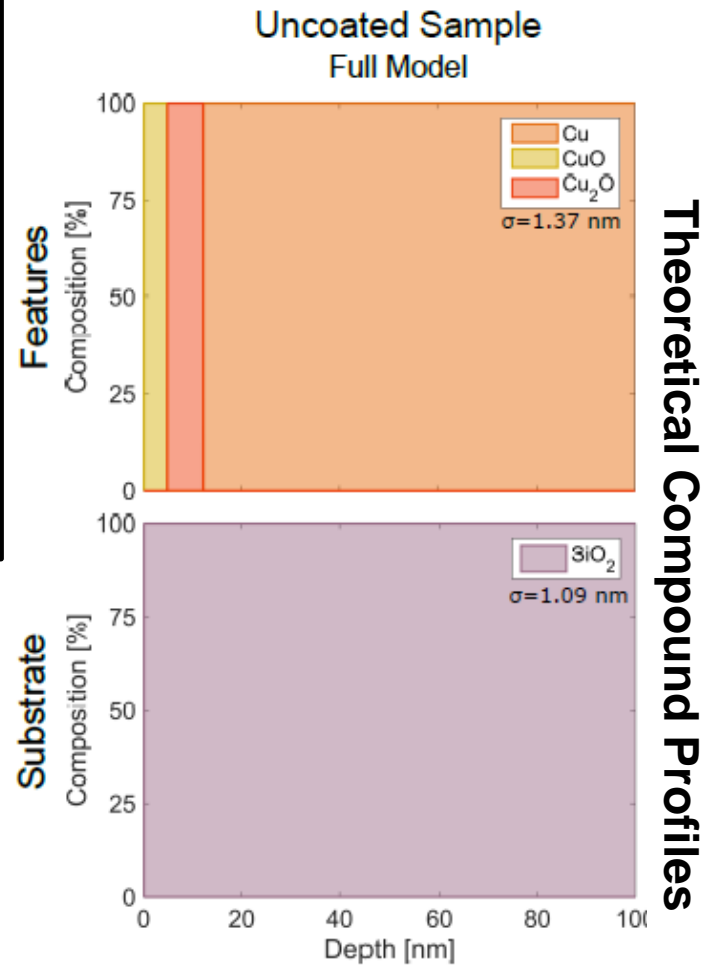
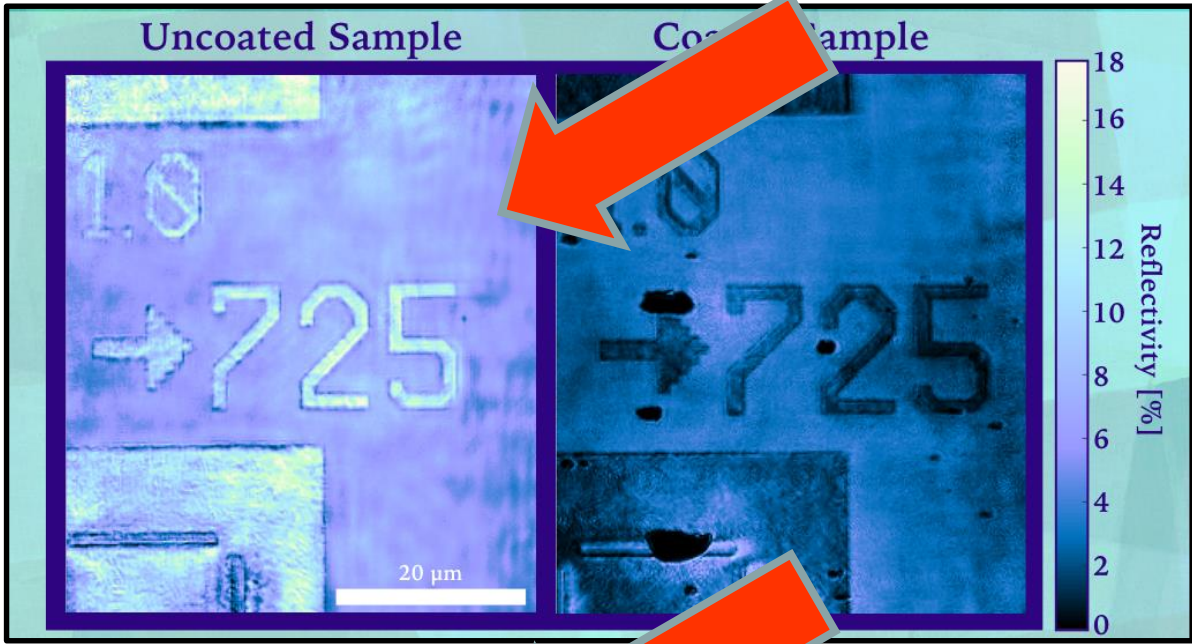
Seeing through buried layers and interfaces



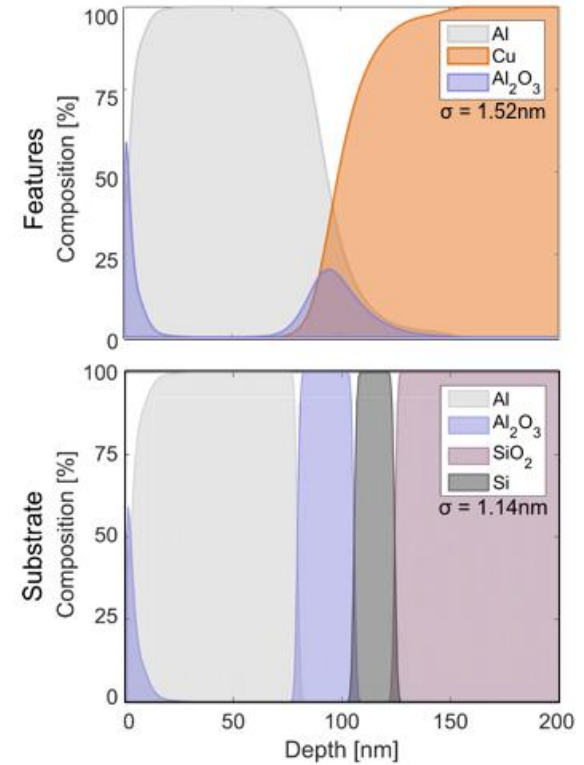
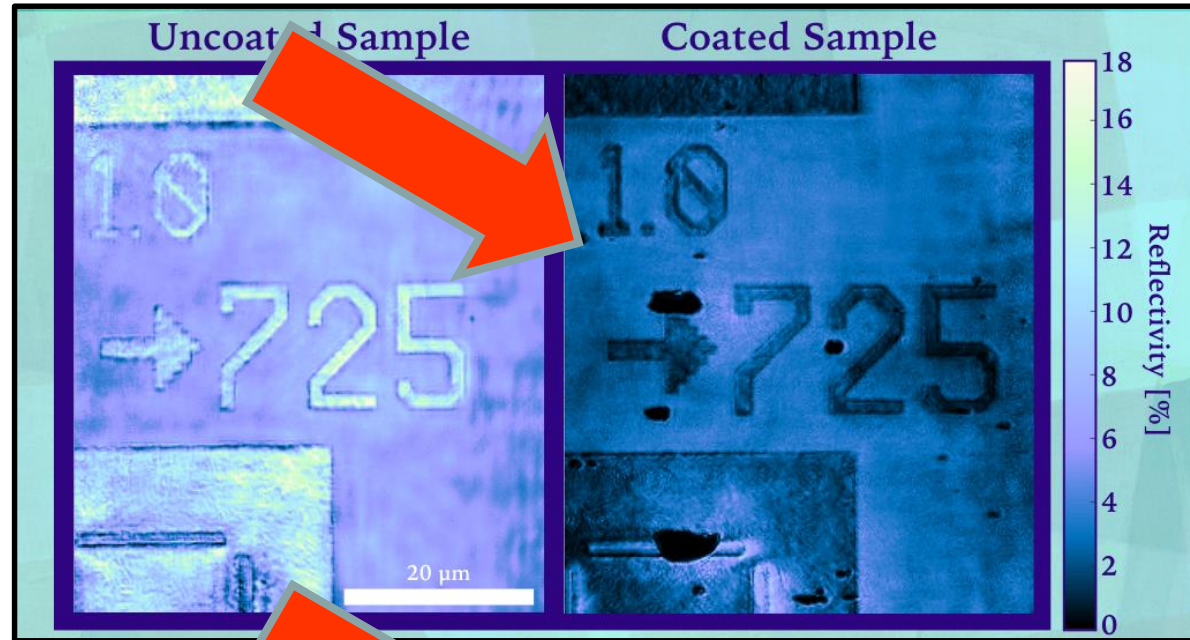
- **CDI amplitude** image enables imaging of elemental composition **through** 100nm of Al
- Quantitative non-destructive imaging of elemental and interfacial properties due to changes in EUV reflectivity
- Identified interdiffusion of Al into Cu, and formation of thin Al oxide layer on SiO₂



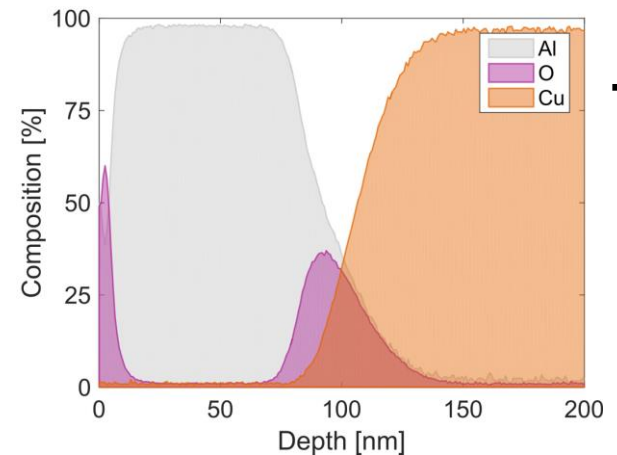
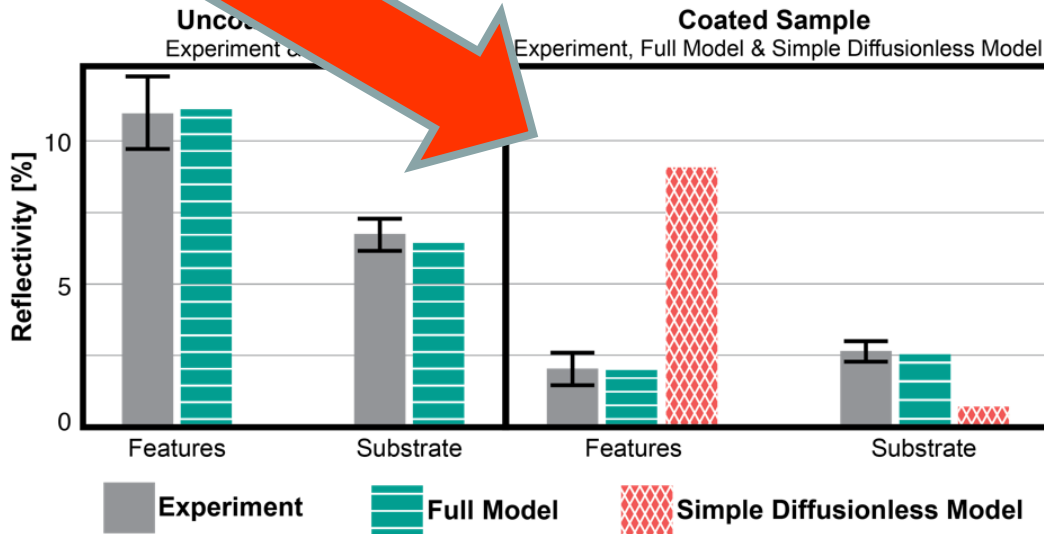
Reflectivity of uncoated damascene shows oxide layer



Reflectivity of coated damascene shows interdiffusion – should be able to measure doping profiles

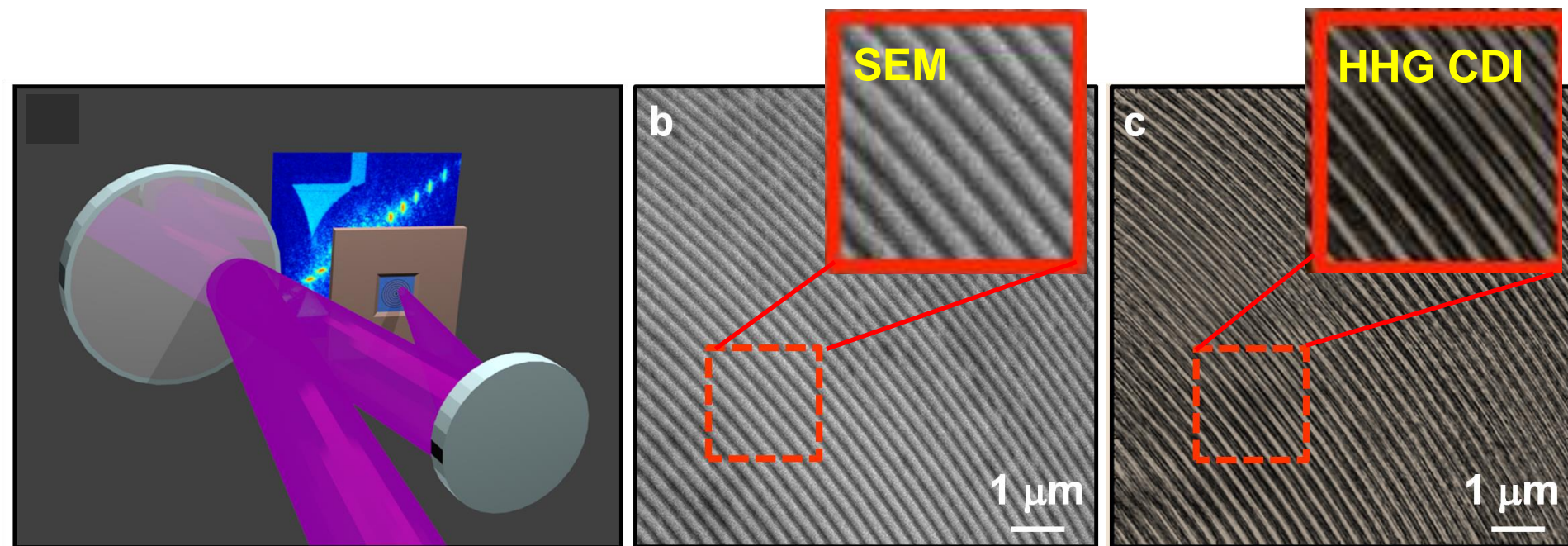


Theoretical Profiles

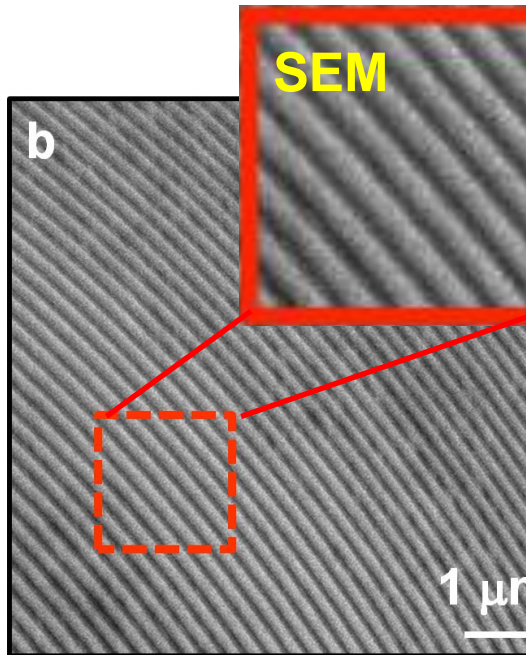
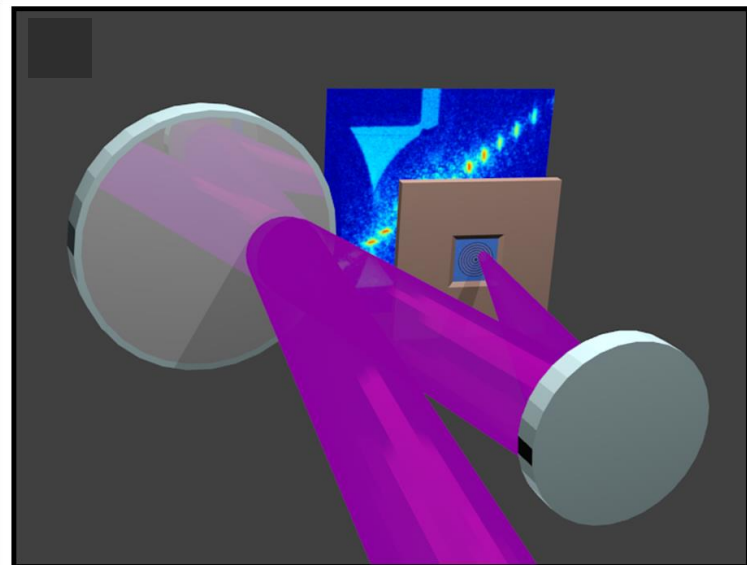
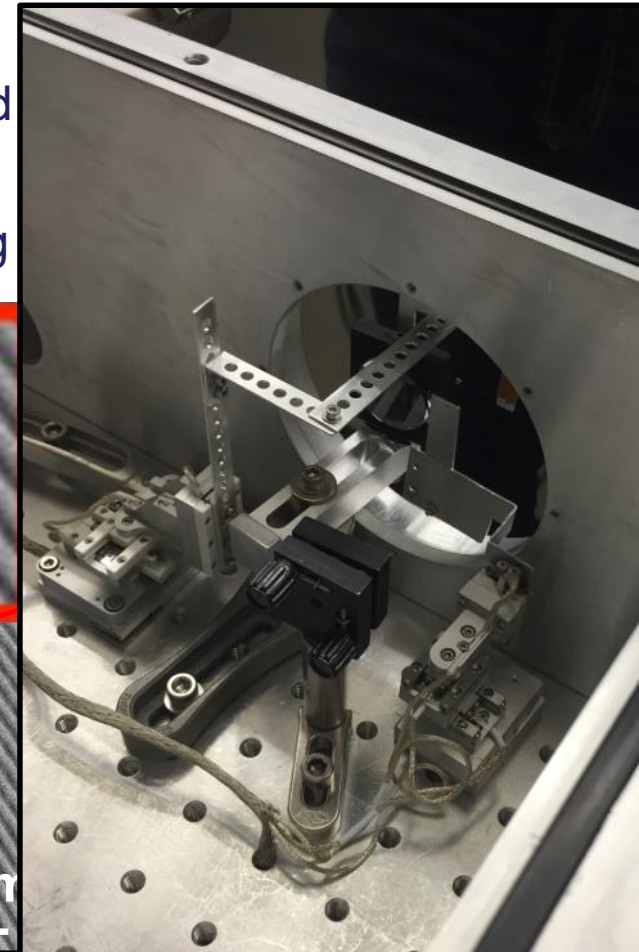


Auger Sputter Depth Profile

- **Using $\lambda = 13.5$ nm, spatial resolution of 14 nm**
 - Spatial resolution **1.04 λ** (PMMA zone plate sample)
 - Record spatial resolution for this wavelength
 - Requires ultrastable engineered HHG XUUS source
- **Not yet resolution or speed limited**
 - Exposure time ~ 10 sec/ μm^2
 - Orders of magnitude increase in speed possible

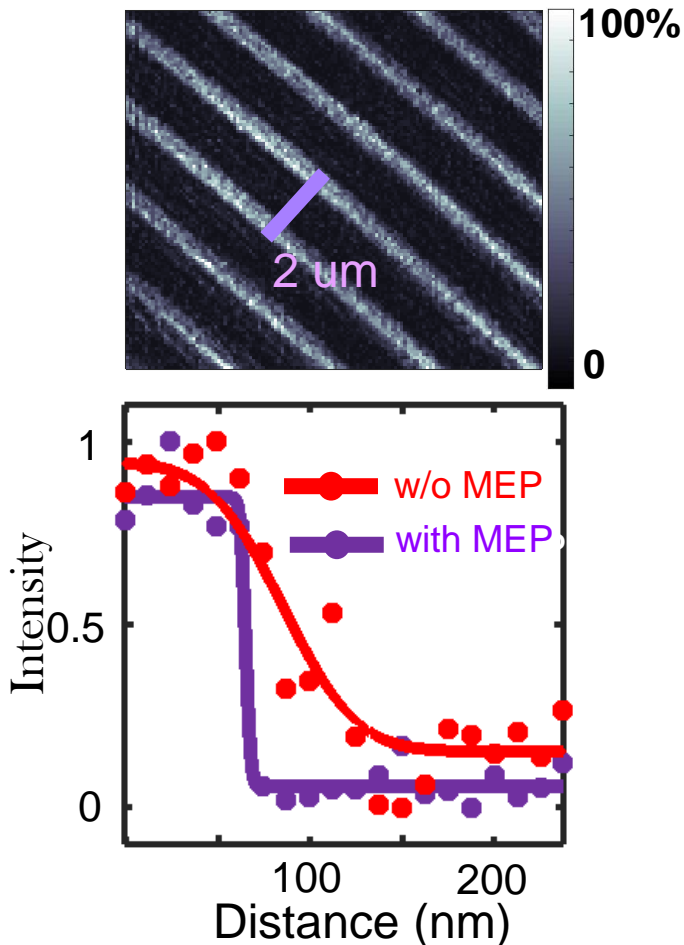


- **Using $\lambda = 13.5$ nm, spatial resolution of 14 nm**
 - Spatial resolution **1.04 λ** (PMMA zone plate sample)
 - Record spatial resolution for this wavelength
 - Requires ultrastable engineered HHG XUUS source
- **Next Steps**
 - Use single-stage, >20W average power cryocooled
 - Optimize HHG scheme (optimized XUUS)
 - Improve resolution to sub-10nm– simply by moving
 - Reflective geometry

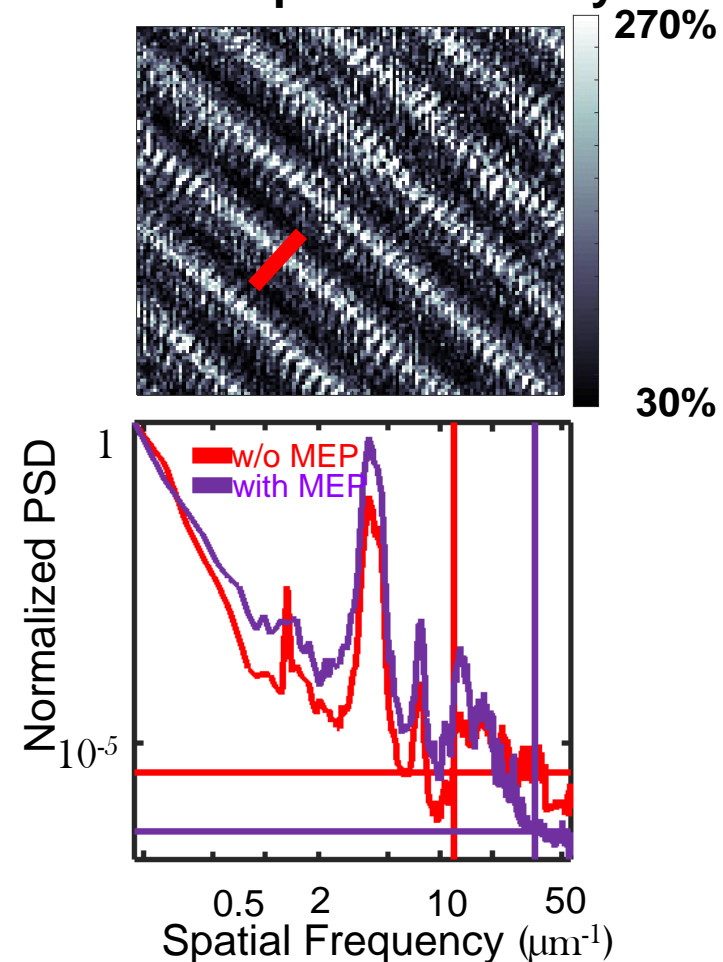


- **NA:** Supports 14.4 nm Resolution
- **Lineout:** Supports 14.4 nm Resolution
- **PSD:** Supports 14.2 nm Resolution: improve resolution to sub-10nm

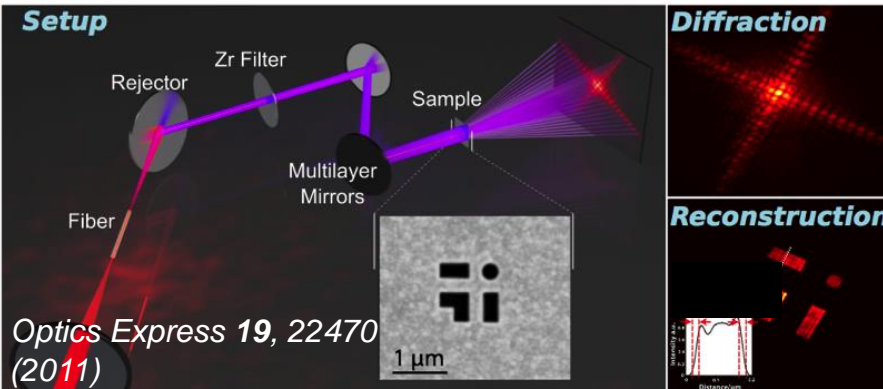
Lineouts



Power Spectral Density



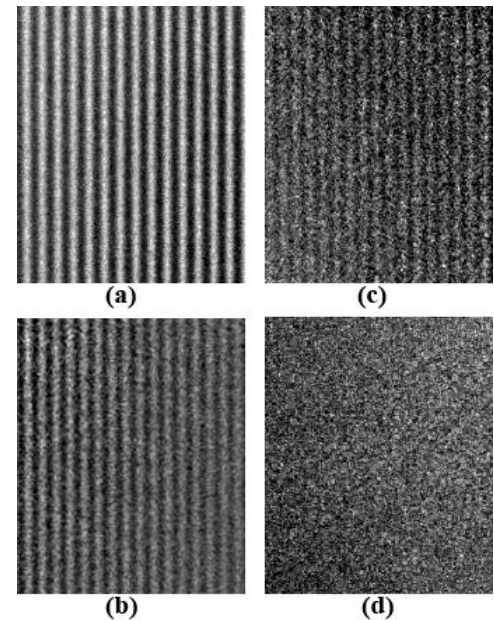
TABLETOP HHG COHERENT IMAGING



2011 HHG Results

- Toy sample
- Simple CDI algorithm
- 22nm spatial resolution

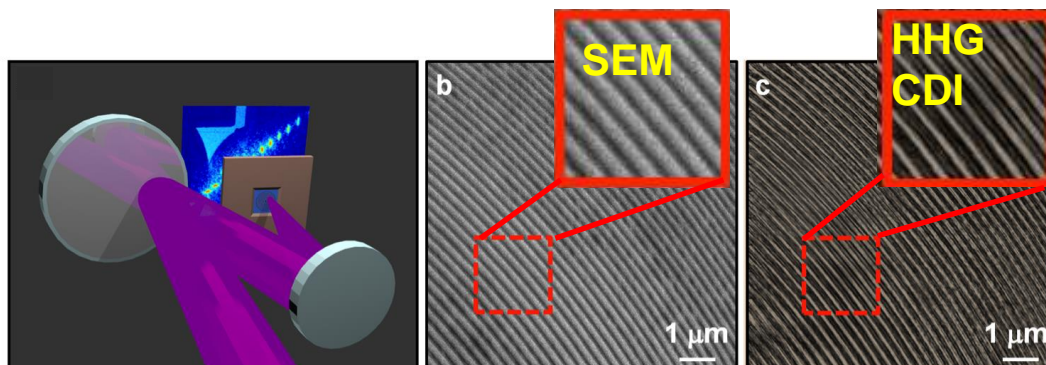
FACILITY-SCALE ZONE PLATE IMAGING



Chao et al. *Optics Express* 17, 17669 (2009)

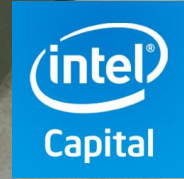
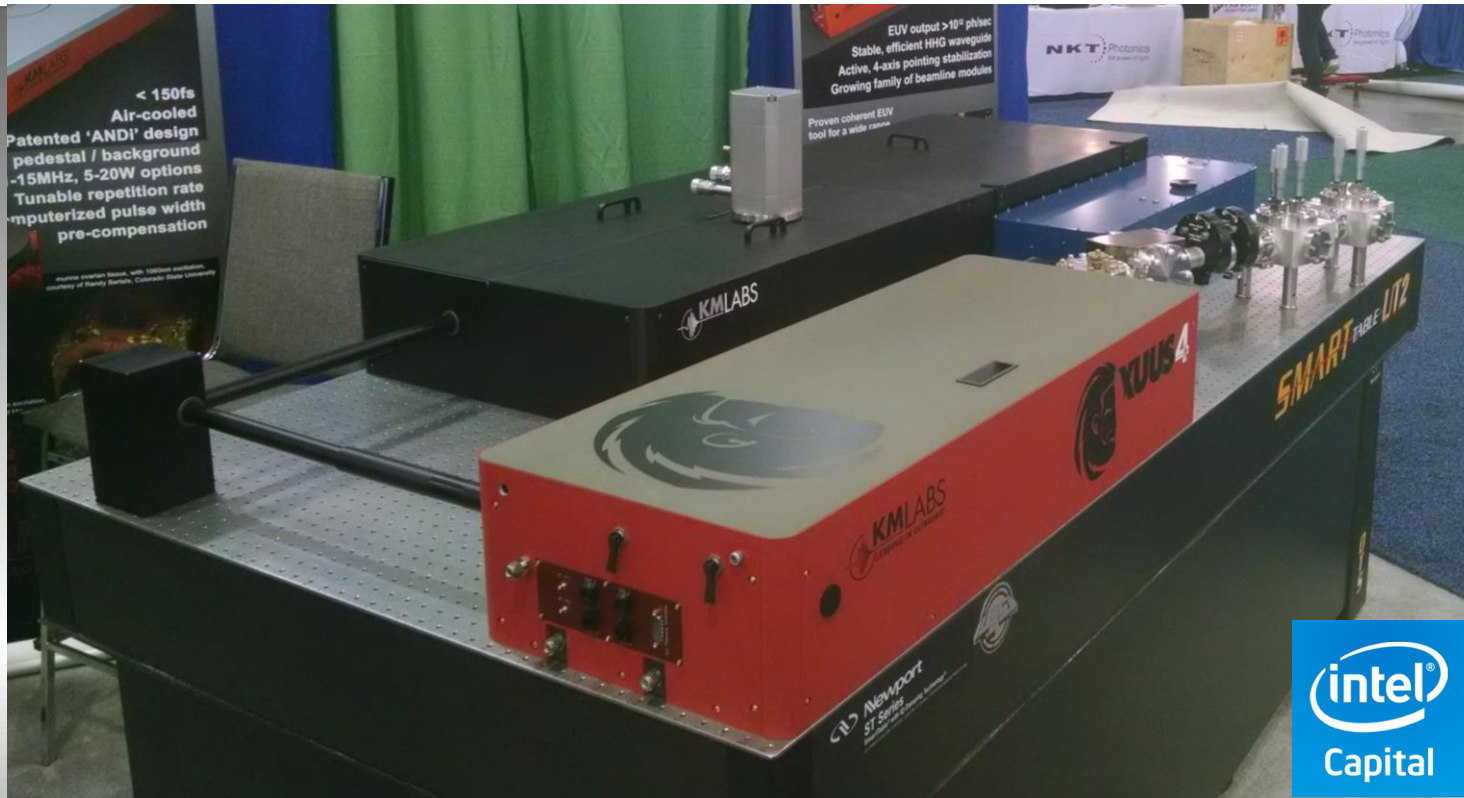
New Record 13.5nm Imaging Results (2016)

- Full field, high contrast ptychography
- **New record 14 nm resolution (1.04λ)**
- Can increase spatial resolution; extend to reflection mode



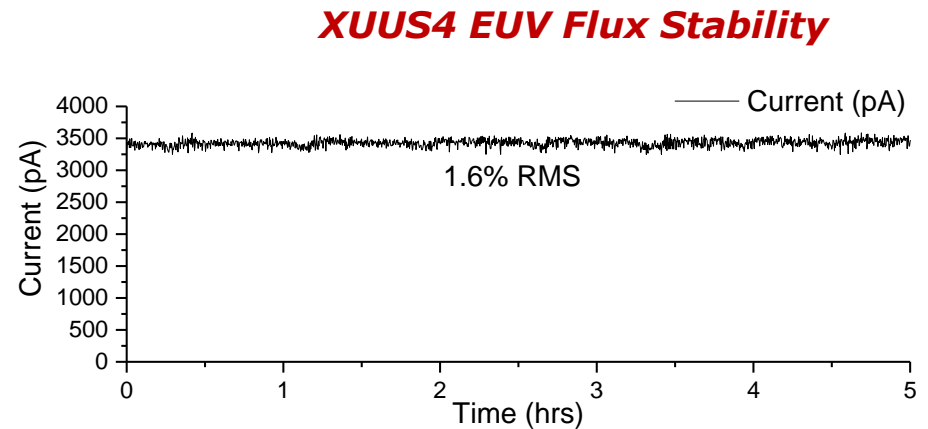
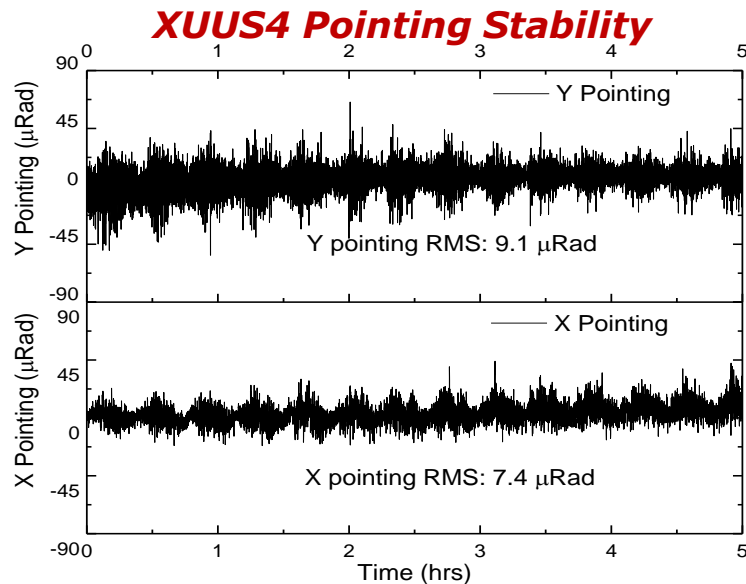
Synchrotron Source

- Zone plate image, 12nm resolution
- Used 2nm illumination



- Optimized for high average-power, high rep-rate, drive lasers: 1 to >200kHz
- Complete HHG XUUS source and beamline
- Active input laser beam stabilization – 4 axis control
- Ultrastable HHG beam intensity, wavefront, beam
- Temperature stabilized breadboard
- Stable, industrial optical mounting
- Complete software control

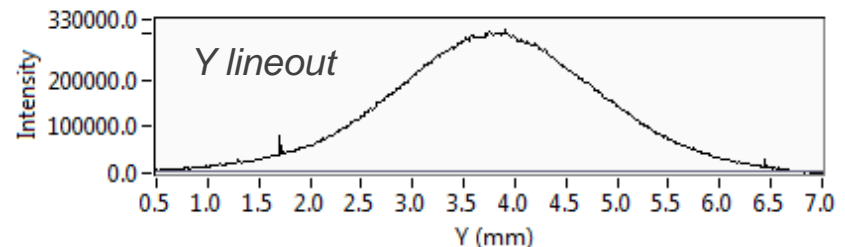
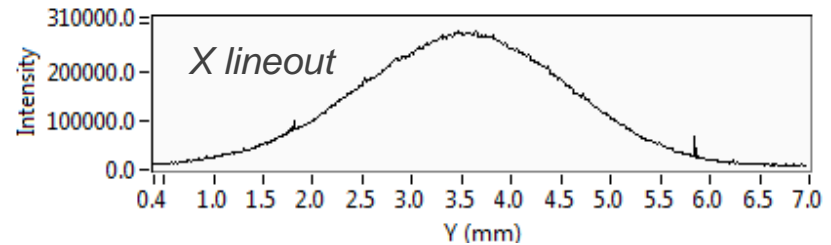
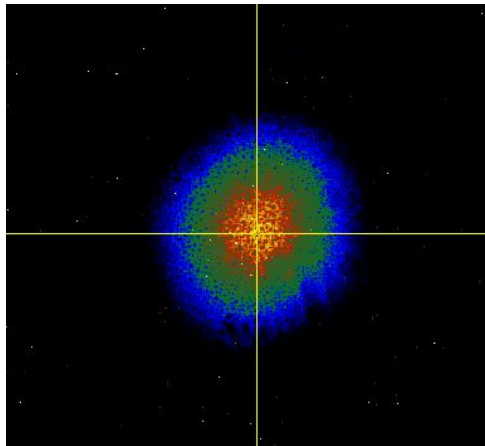
- Integrated HHG light source
 - Optimized for high average-power, high rep-rate, drive lasers: 1 to >100kHz
 - Cartridge waveguide: increased stability, performance, optimized at 13.5nm
 - Active input beam stabilization – 4 axis control
 - Ultrastable HHG beam intensity, wavefront, beam
 - Temperature stabilized breadboard
 - Stable, industrial optical mounting
 - Complete software control



Data for 5 harmonic orders peaked at 42 eV.

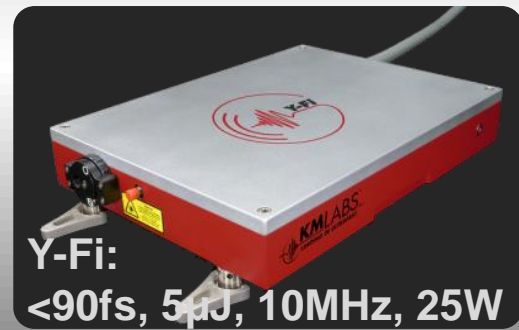
- Integrated HHG light source
 - Optimized for high average-power, high rep-rate, drive lasers: 1 to >100kHz
 - Cartridge waveguide: increased stability, performance, optimized at 13.5nm
 - Active input beam stabilization – 4 axis control
 - Ultrastable HHG beam intensity, wavefront, beam
 - Temperature stabilized breadboard
 - Stable, industrial optical mounting
 - Complete software control

XUUS4 EUV Beam

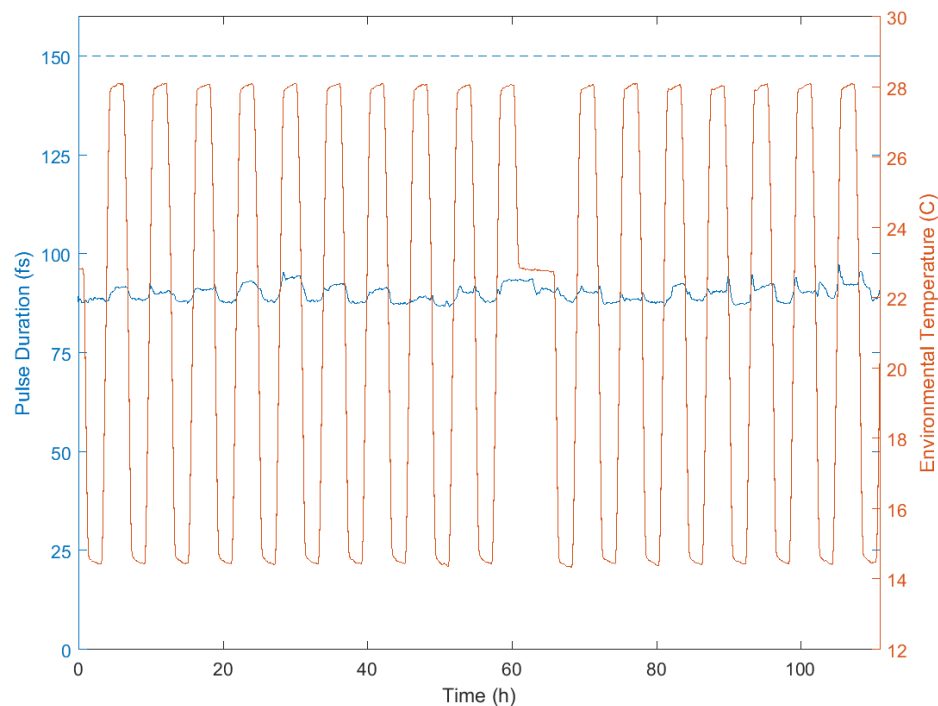
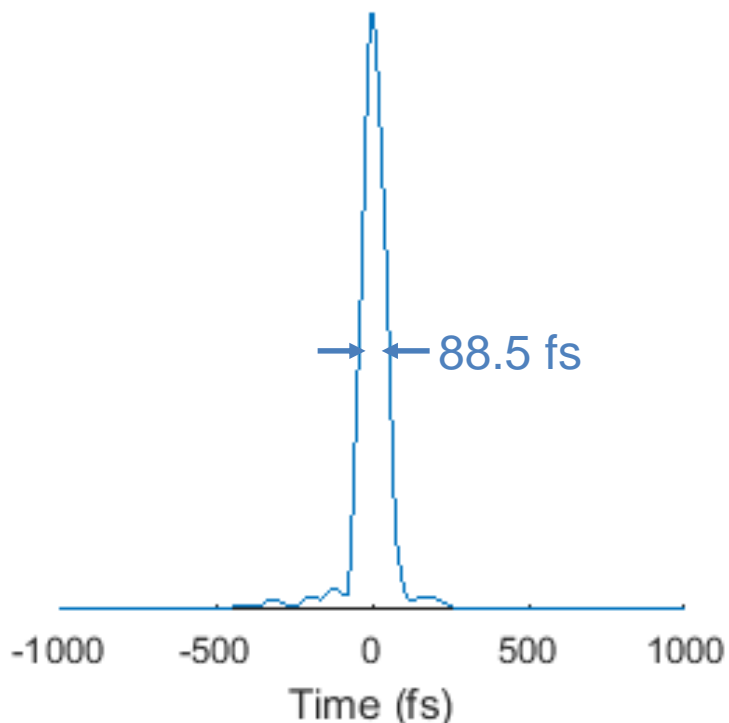


- KMLabs has developed a record 25W single stage Ti:sapphire system for science market
- Repetition rates from kHz to MHz
- $M^2 \sim 1.1$ – flawless Gaussian beam
- Unprecedented power and stability
- New XUUS_{4.2} will enable >10x increase in HHG flux in 10 – 20 nm region
- Compact >25W hybrid fiber lasers also under development
- $\approx 10^{-4} - 10^{-7}$ into 1 harmonic order

25W single stage fs laser



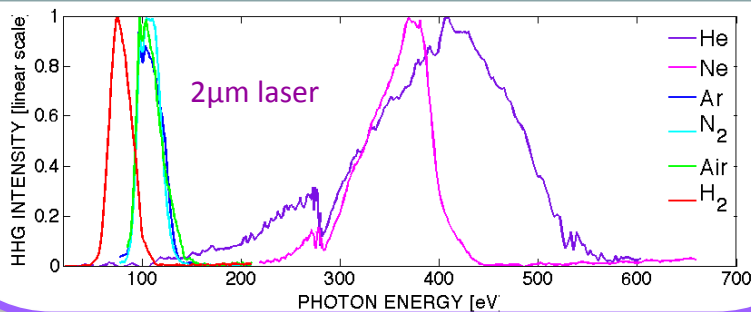
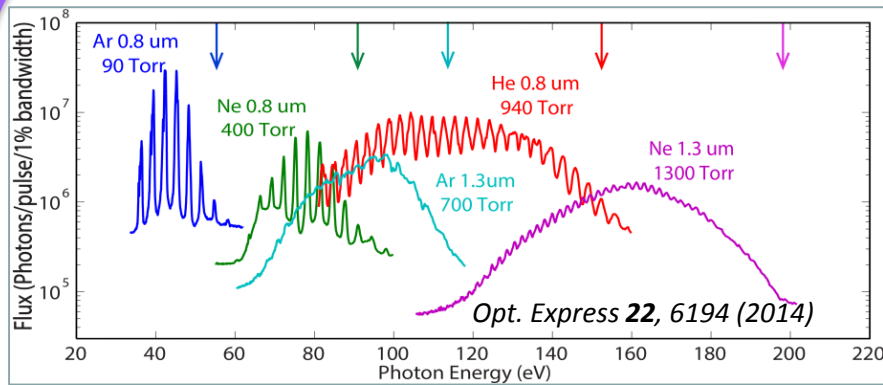
- Pulse duration over >100 hours of temperature cycling
- 90 ± 2.2 fs, <0.4% amplitude stability over 14°-28° C temp cycling
- Necessary “front end” system for future compact diode-pumped ultrafast systems



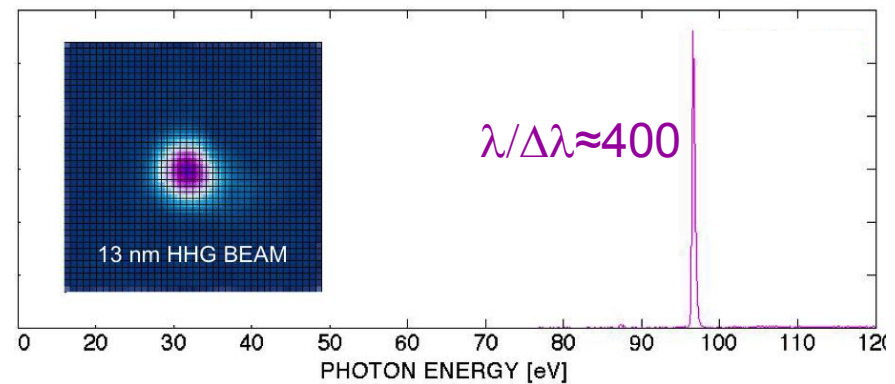
- Coherent diffractive imaging is rapidly establishing itself as the “gold standard” for EUV imaging
 - Large, redundant data set allows one to obtain a *full characterization* of how an object scatters incident light
 - i.e. everything you could *ever* know using light at that wavelength
 - With **NO** instrumental distortions/limitations
- Near future (JILA → KMLabs) versatile general purpose 13 nm microscope
- Broader applications of HHG EUV microscopy to support nanoscience Have been demonstrated and remain to be fully-developed
 - Interfaces
 - Mechanical properties (Young’s Modulus, Poisson ratio)
 - Magnetic properties
 - Dynamic behavior

- 10^{-4} to 10^{-6} into one harmonic order at 30nm depending on HHG scheme
- Using mid-IR lasers, supercontinua ideal for spectroscopy (NEXAFS, MOKE)
- Using UV lasers, isolated HHG peaks ideal for imaging and metrology
- Using 2W, 1kHz, 0.8 μ m laser, achieve 10^{10} photons/s/1% band @ 13nm
- Using 2W, 1kHz, 0.8 μ m laser, achieve 160nW, 1% band @ 13nm
- Using 2W, 1kHz, 0.27 μ m laser, achieve $>\mu$ W in $\lambda/\Delta\lambda\approx 400$ @ 13nm (still in research)
- Using 15W, 1kHz, laser, achieve $>15\mu$ W in 1 order @ 30nm

Near and mid-IR driven HHG



UV driven 13nm HHG



Electron imaging



Knowledge Transfer



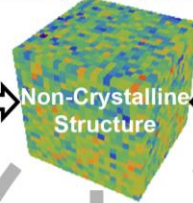
Education research, broadening participation

Capturing and monitoring individual atoms in 3D

Routine 3D atomic resolution structure of biological complexes

Functional 3D imaging of energy, magnetic and spintronic materials

Imaging various forms of energy flow and fields across interfaces



Multi-D Electron Microscopy

Ultrafast Imaging

Correlative Microscopy

Detector Algorithm Big Data
Super Resolution

Functional 3D X-ray Imaging

Advanced Optical Nano-Imaging

X-ray



Nano, correlative, h imaging



Underpinning technologies



Detectors, algorithms

