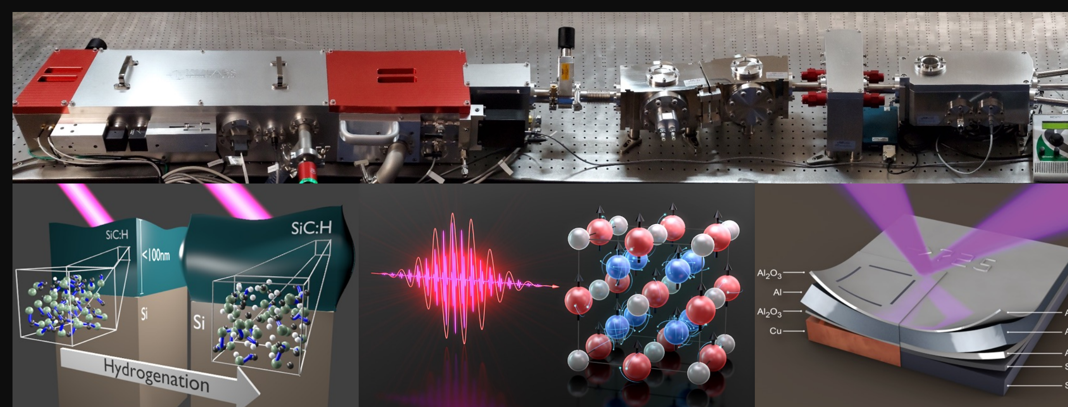
Putting High Harmonic Quantum Technologies to Work

Margaret Murnane, The Kapteyn-Murnane Group, KMLabs Inc.
Alumni and Collaborators



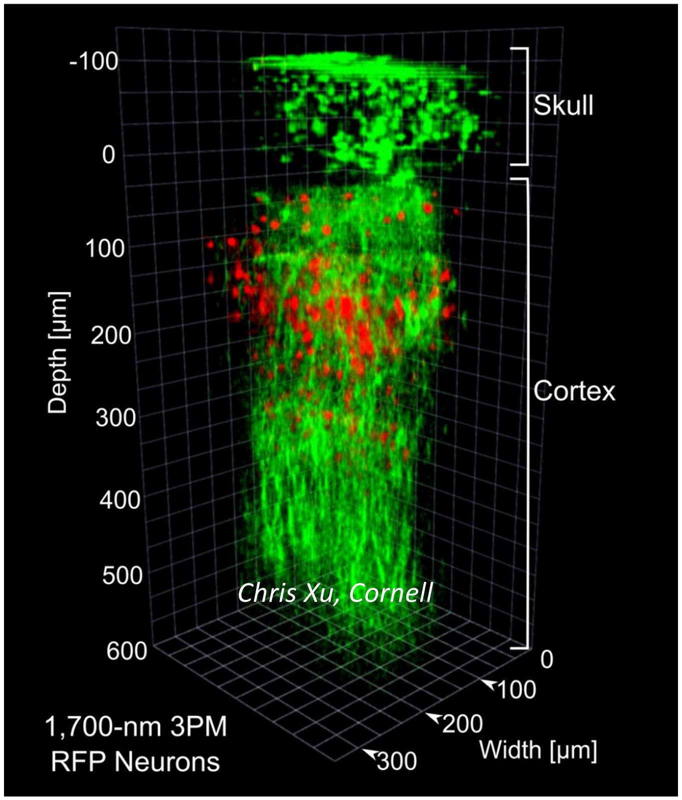
JILA NIST CU STROBE KMLABS inc. LEADING IN ULTRAFAST

Putting ultrafast lasers to work

- **The largest applications of ultrafast lasers**
  - **Multiphoton microscopy**
  - **Precision laser micromachining**

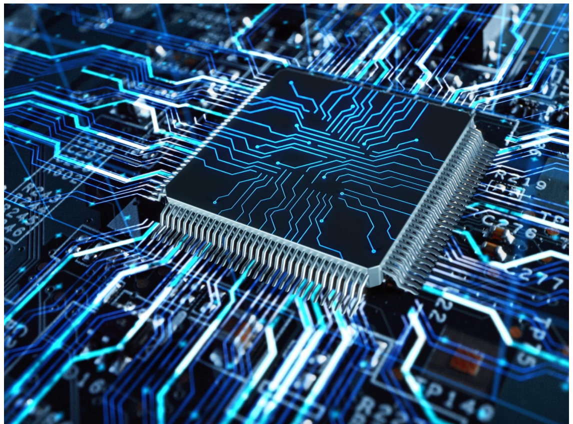
- **Are enabled by ultrafast…**

- **But are NOT ultrafast measurements**



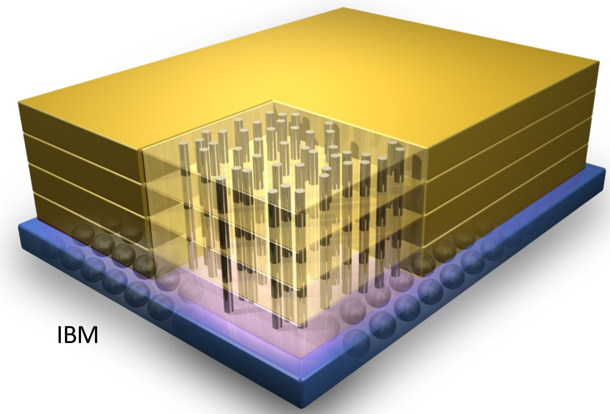
High harmonic sources are following the same trajectory

Nanostructured materials have very different properties than bulk



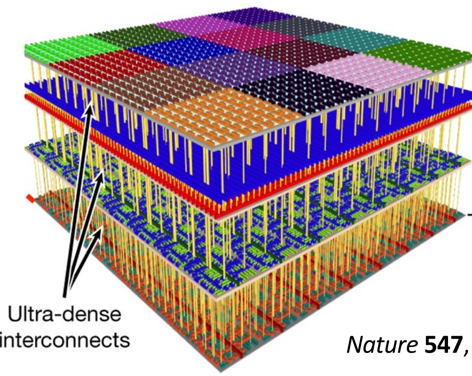
iStock/gorodenkoff

2.5 & 3D packaging



IBM

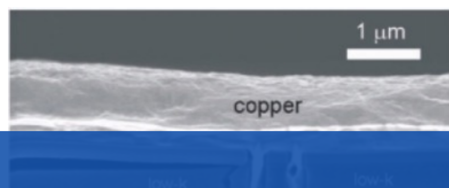
Novel architectures



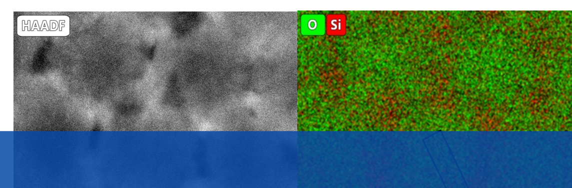
Nature **547**, 74 (2017)

Thermal

Mechanical



Structural/Interface



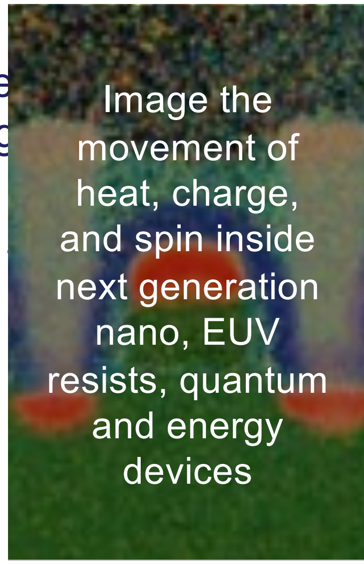
Electrical/Magnetic

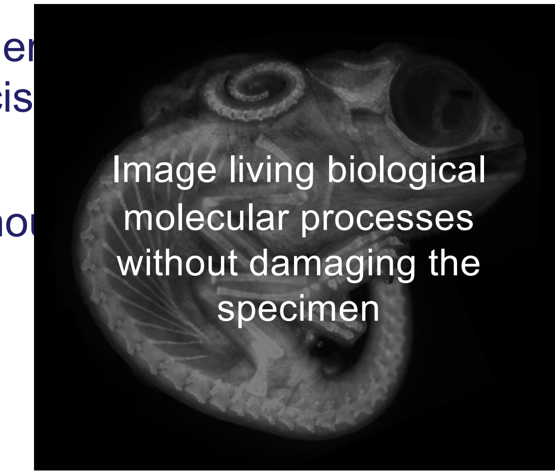
Critical need for better functional metrologies

More accuracy, more resolution, low dose, versatility, predictive, buried/interfaces/3D, nanoscale

The Nano, Bio & Materials Challenge

- **Conventional methods struggle to characterize *functional* nanoscale ($i.e. \lesssim 100\ nm$) properties**



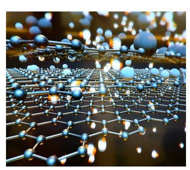


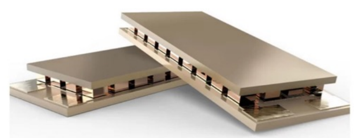
- **Challenge:** New e… o, quantum and ener… nake, measure and integ… atomic-scale precis… with structure

- **Challenge:** Need … pies that work withou… r labelling

Integrated circuits

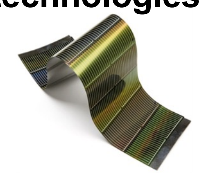
Photonic, 2D, quantum materials



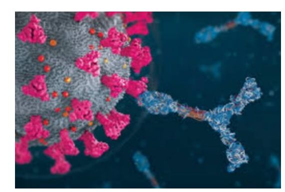


Nano-engineered thermoelectrics

Solar technologies



Bio



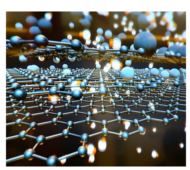
The Nano, Bio & Materials Challenge

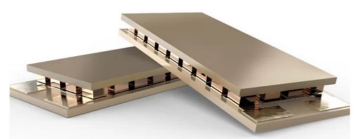
- **Conventional methods struggle to characterize *functional* nanoscale ($i.e. \lesssim 100\ nm$) properties**
- **Challenge:** New energy-efficient nano, quantum and energy devices require that we make, measure and integrate materials, with atomic-scale precision — and correlate function with structure
- **Challenge:** Need functional microscopies that work without cutting, coating, freezing or labelling

High harmonic sources enable new methods that can see better and see things as they happen!!

Integrated circuits

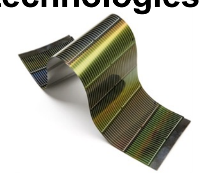
Photonic, 2D, quantum materials



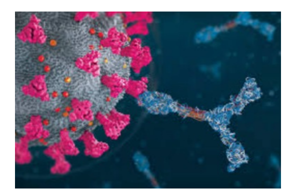


Nano-engineered thermoelectrics

Solar technologies

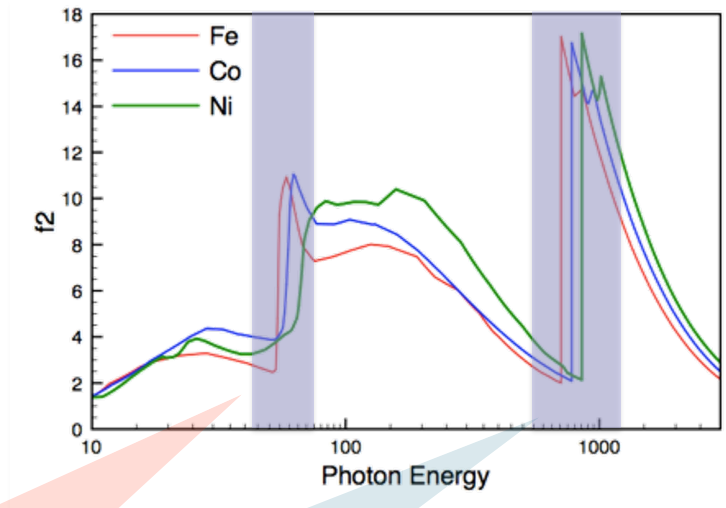


Bio

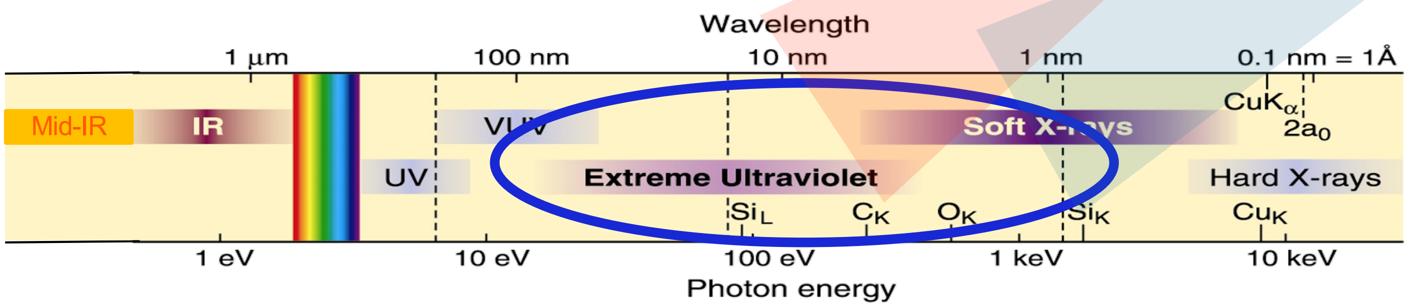


Coherent Extreme-UV and X-ray light → capture nanoscale processes

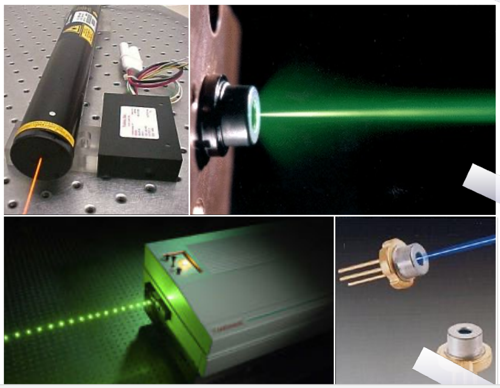
- Wavelengths $\lambda \sim 1 - 100\ nm;\ \ h\nu \sim 10 \rightarrow 1000\ eV$
- Elemental, chemical, magnetic specificity
- Non-destructive, low-dose compared with SEM, TEM
- Image with few-nm resolution
- Capture dynamics relevant to function (~fs)

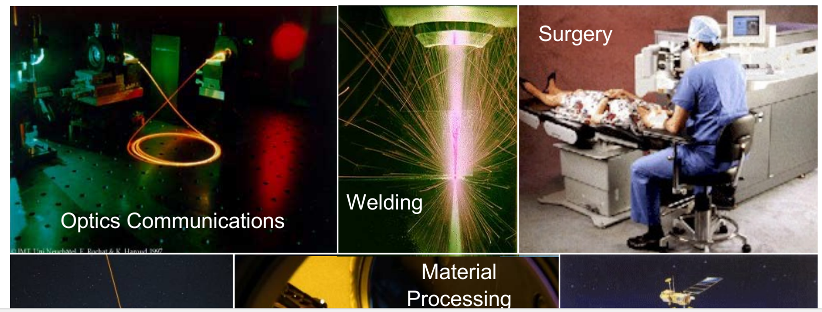


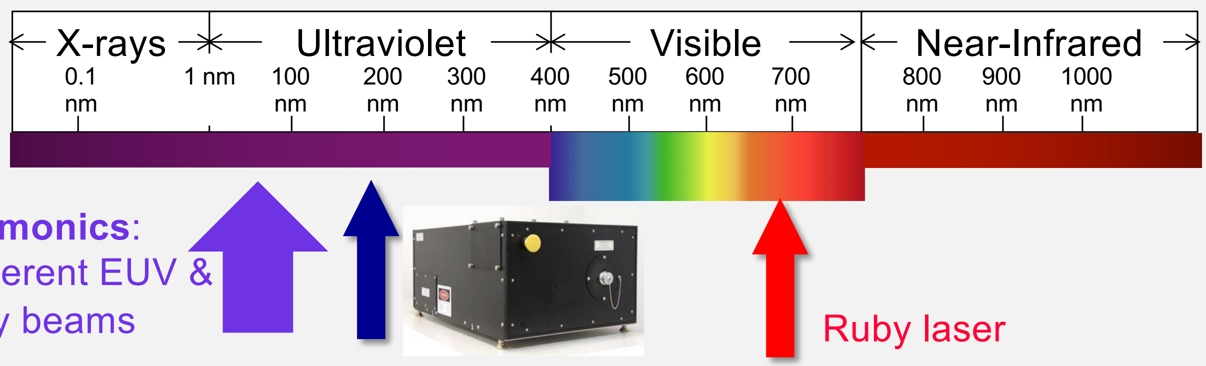
Requires coherence



Tabletop scale lasers in widespread use





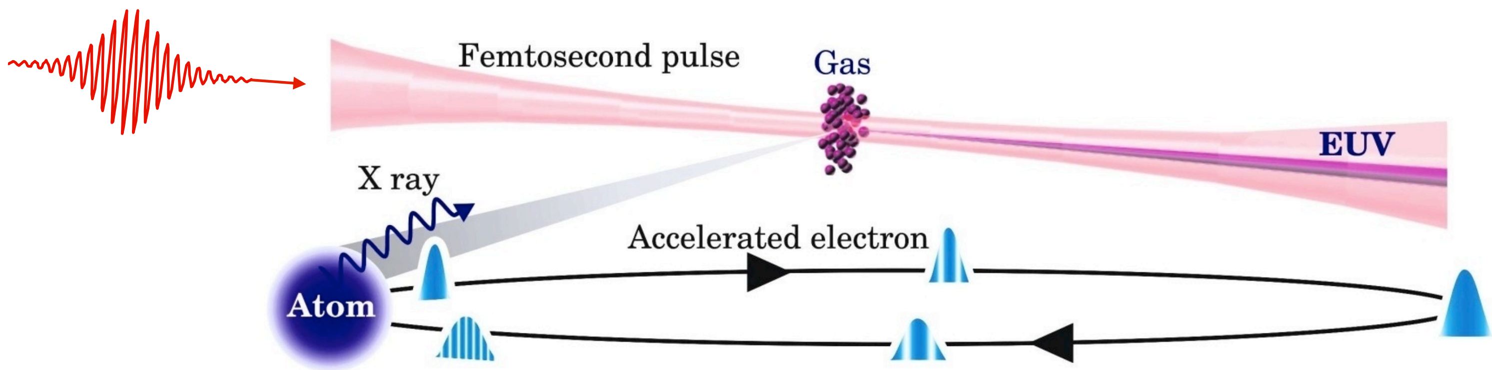


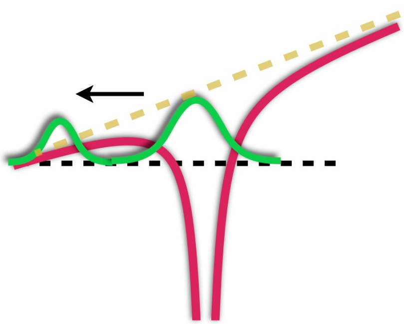
High harmonics: Quantum-coherent EUV & soft X-ray beams

Excimer lasers

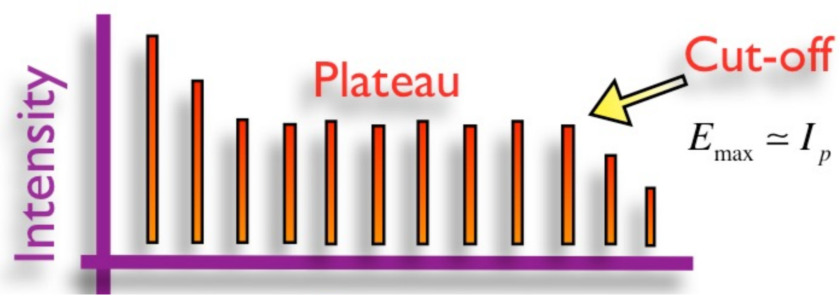
Ruby laser

High harmonic generation

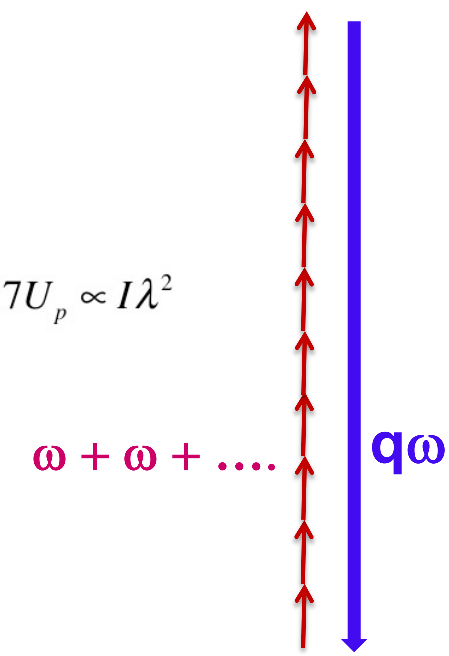




Tunnel ionization

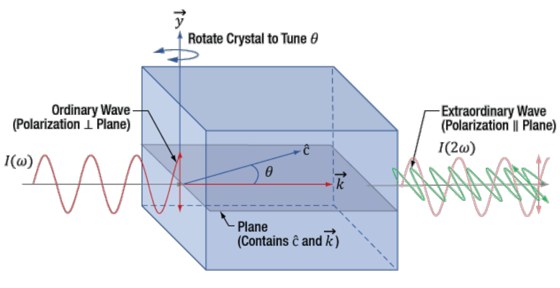


$$E_{\max} \simeq I_p + 3.17U_p \propto I\lambda^2$$

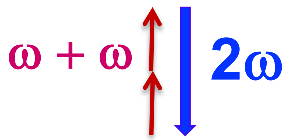


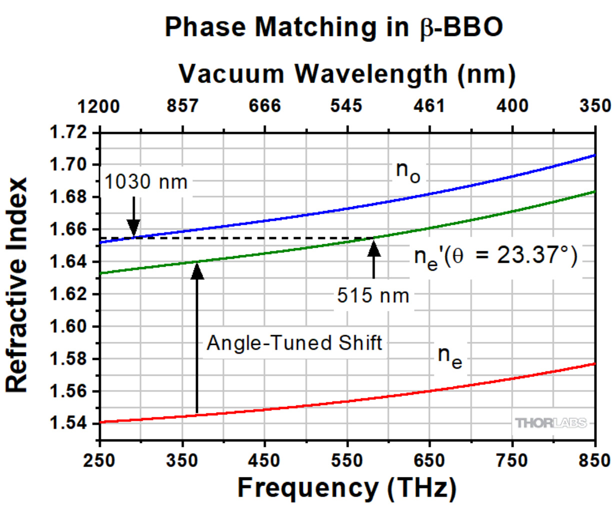
Bright high harmonic emission - *phase matching in time domain*

Visible NLO: phase matching in spatial domain

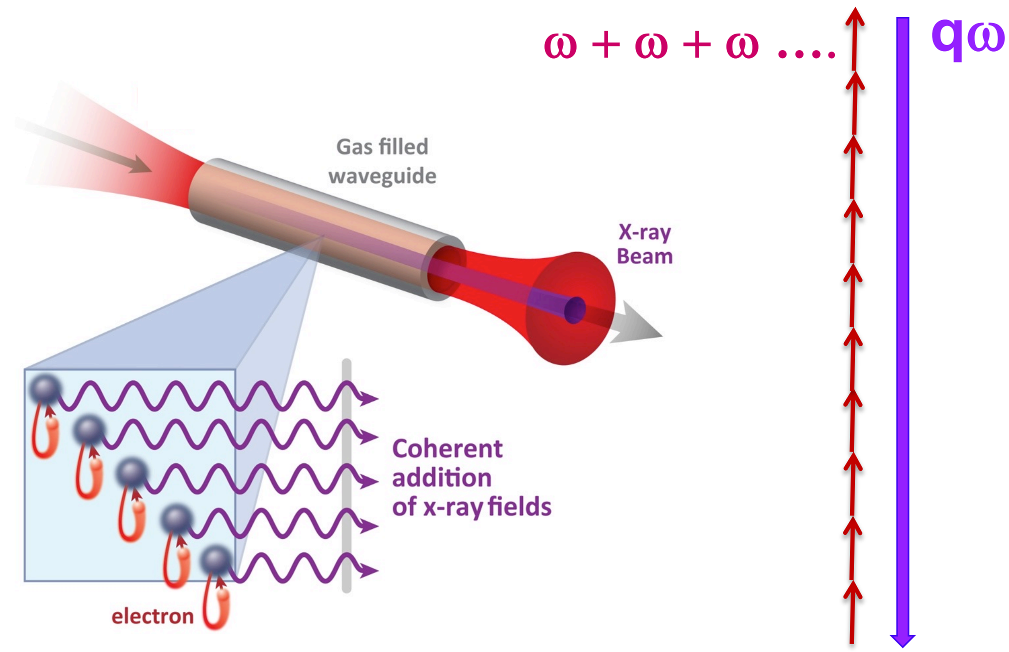


Type-I Phase Matching Using a β-BBO Crystal





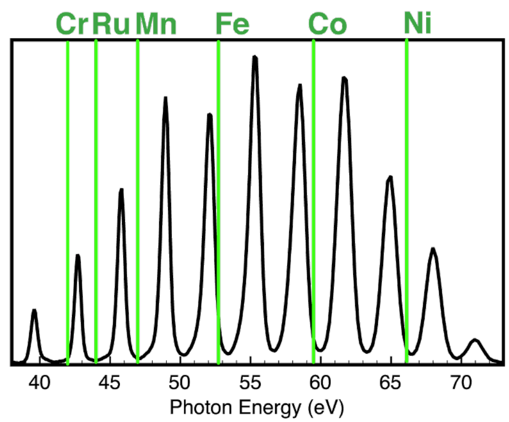
Extreme nonlinear optics: phase matching in time domain

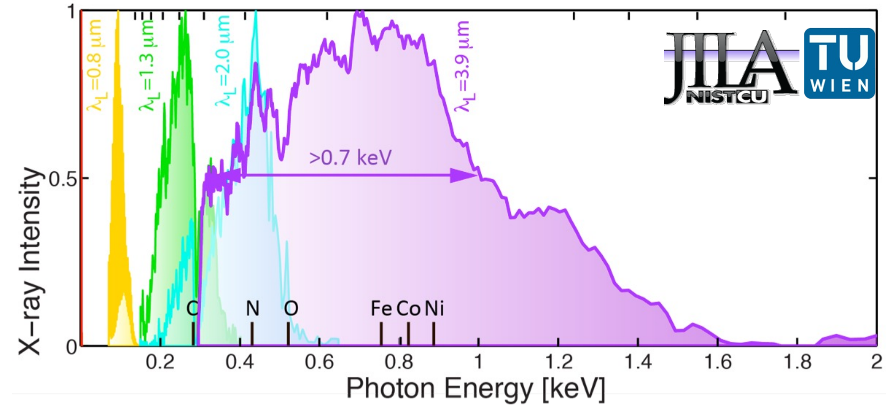


Laser and x-ray phase velocities are matched at the speed of light ONLY during part of the laser pulse

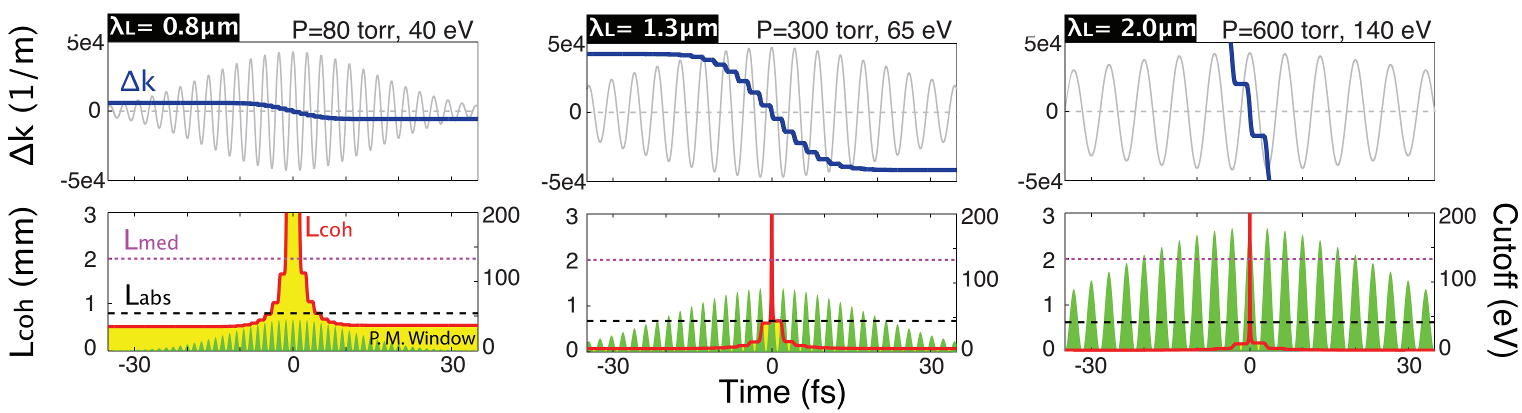
*Rundquist Ph.D. Thesis (1998); Science **280**, 1412 (1998)*

Phase matching window sculpts the HHG in time and frequency





Near-IR laser → **Mid-IR laser**

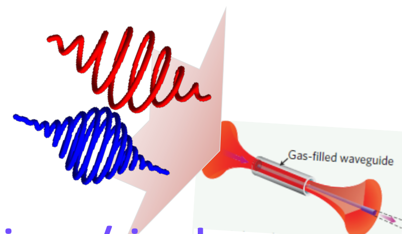


Rundquist Ph.D. Thesis (1998)
*Rundquist et al., Science **280**, 1412 (1998)*
*Bartels et al., Science **297**, 376 (2002)*
*Thomann et al., Opt. Exp. **17**, 4611 (2009)*
*Chen et al., PNAS **111**, E2361 (2014)*

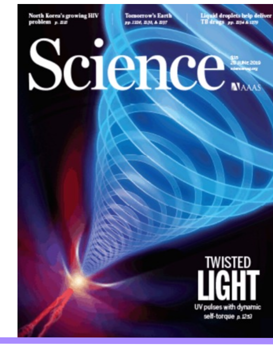
Tabletop high harmonic sources → new quantum technology enabling *exquisite* control over X-ray light

- 12 octave supercontinuum or isolated peaks
- Full temporal coherence: femto to atto to zepto
- Full spatial coherence to keV
- Bright linear, circular and OAM beams
- Unique light source better than we dreamed of!
- **Exquisite control over X-ray light using lasers**

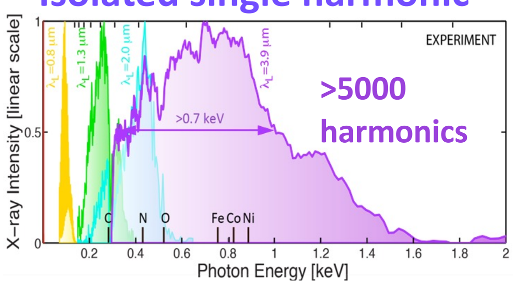
Science **364**, *9486 (2019); Nat. Photon.* **13**, *123 (2019); Science Ad.* **2**, *e1501333 (2016)*
PNAS **112**, *14206 (2015); Nature Photonics* **9**, *99 (2015); Nature Photon.* **9**, *743 (2015)*

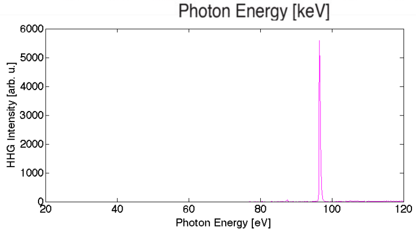


Linear/circular
OAM/SAM shaped



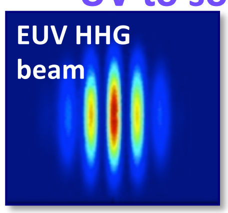
Broad coherent continuum OR Isolated single harmonic

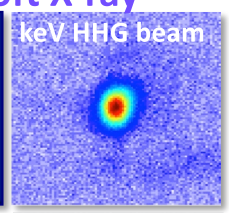


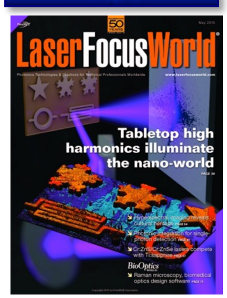


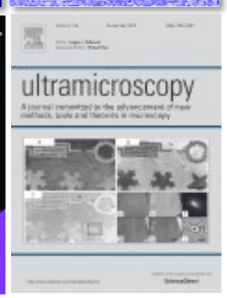
Science **336**, *1287 (2012); Science* **350**, *1225 (2015)*

Full spatial coherence: UV to soft X-ray



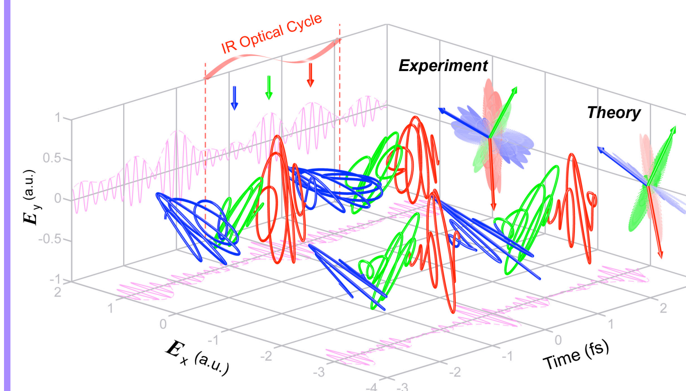


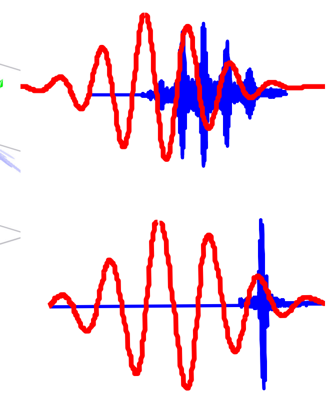




Science **297**, *376 ('02); Science* **348**, *530 ('15)*

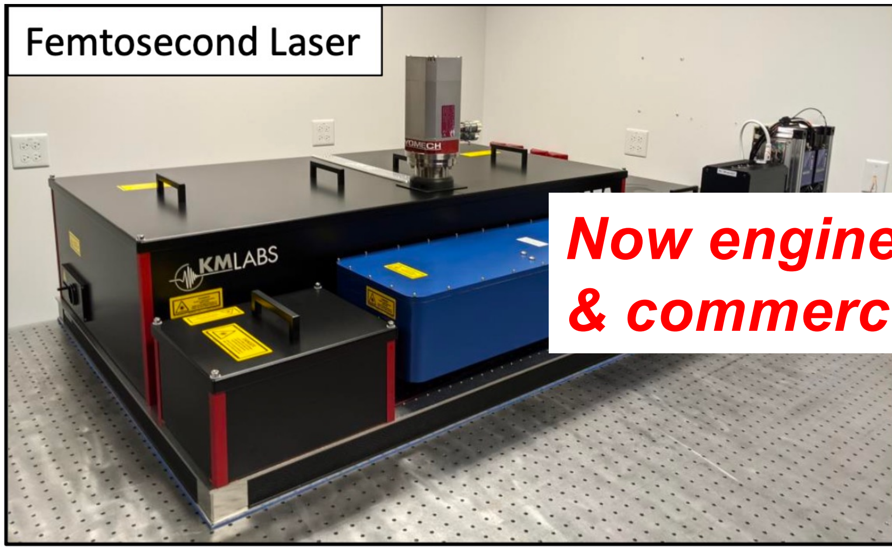
Attosecond pulses and pulsetrains: UV to soft X-ray



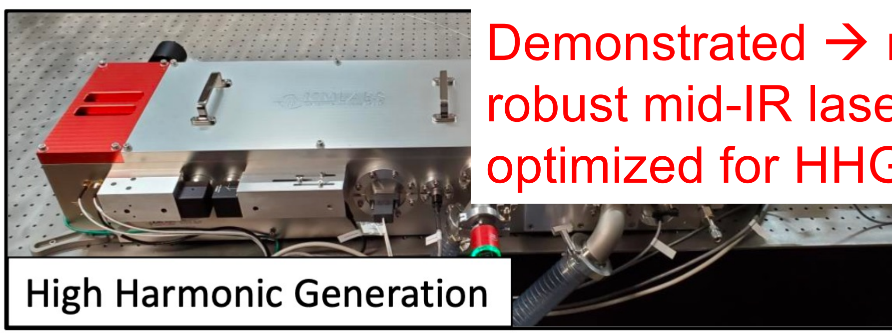


PRL **111**, *033002 (2013); PNAS* **111**, *E2361 (2014); PRL* **120**, *093002 (2018); Optica* **7**, *832 (2020)*

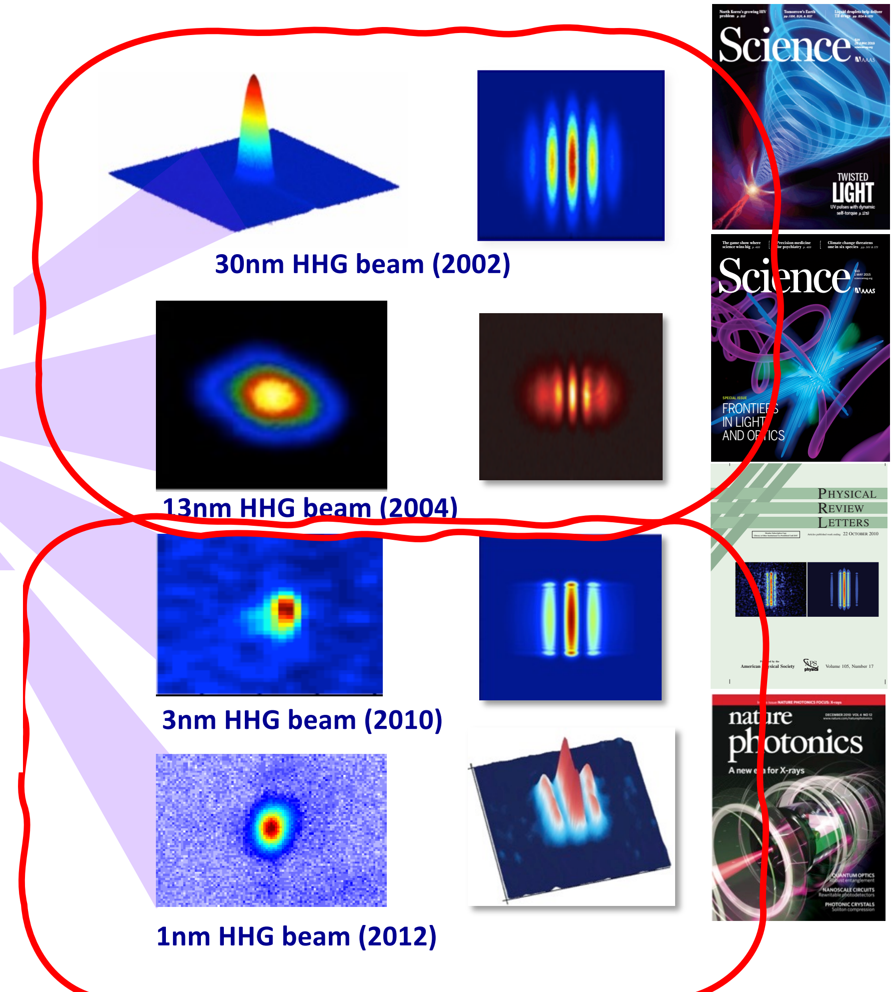
Putting EUV High Harmonic Generation to Work



Now engineered & commercial



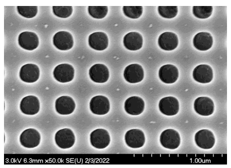
Demonstrated → need robust mid-IR lasers optimized for HHG



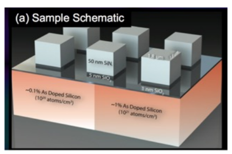
Rundquist et al., Science ***280****, 1412 (1998)*
Bartels et al., Science ***297****, 376 (2002)*
Popmintchev et al., Science ***336****, 1287 (2012)*

EUV metrologies: nondestructive, ultrasensitive, functional metrologies

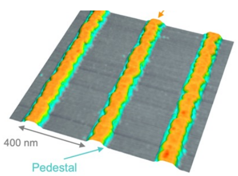
INDUSTRY Collaborations



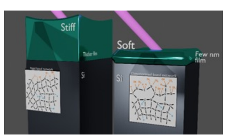
3M
Metamaterials
Thermal materials



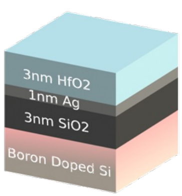
imec KMLABS LEADING IN ULTRAFAST
ARPES, CDI, 2D



imec SAMSUNG
Nanoelectronics

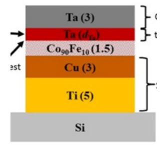


intel
Thin films

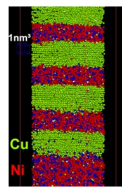


Ball Aerospace THE UNIVERSITY OF ARIZONA
Detectors

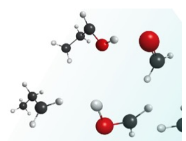
NATIONAL LAB Collaborations



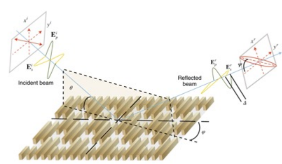
NIST
Spintronic



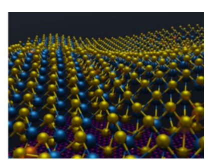
NIST
Atom Probe



Argonne NATIONAL LABORATORY
Combustion



NIST
Nanoelectronics



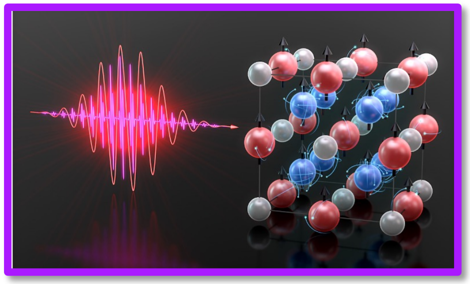
GORDON AND BETTY MOORE FOUNDATION
Quantum, Bio, Low dose

Unique HHG advantages

- Coherence: diffraction limit
- Short wavelength: high resolution
- Short pulses: functional imaging
- Ultrastable phase: ~pm sensitivity
- Shot noise limited metrology
- Penetrate buried layers, interfaces
- Focus to small <μm spot size
- Element specificity
- Nondestructive & low dose
- Broad access for R&D

HHG metrologies

→ nondestructive, functional imaging at high resolution, element specificity



Element-resolved spin dynamics

Magnetic

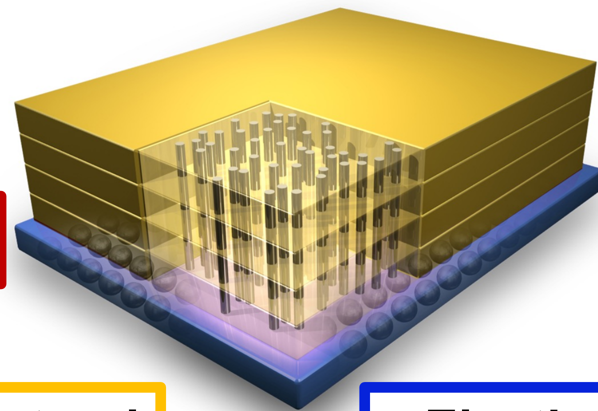
Electronic

Adv. Energy Mater. 1402180 (2015)

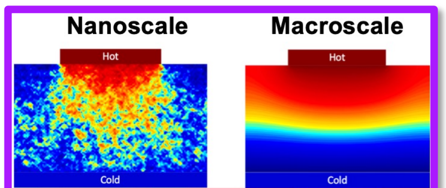
Cleaning Recipe	$E_F - E_V$ (eV)	Band Bending (meV)	Surface Band Structure
NH_4OH	0.6	180	*Upward*
$H_2O_2 + NH_4OH$	0.4	130	*Upward*
NH_4OH + 3A° Cu	0.2	100	*Downward*

Band bending, interfacial fields

Transport

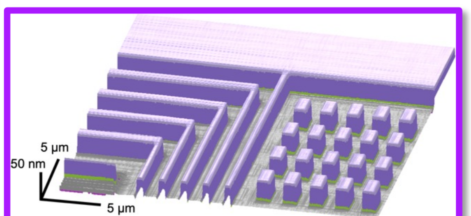


Interfacial



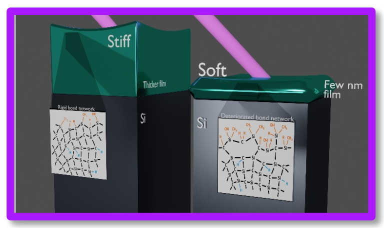
New understanding of nanoscale heat flow

Structural

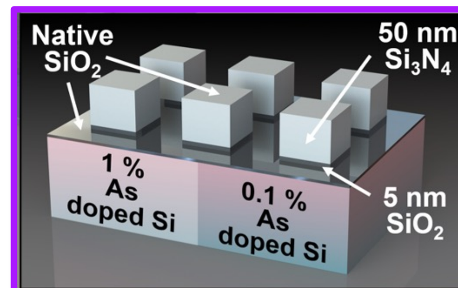


Sub-wavelength EUV imaging

Elastic

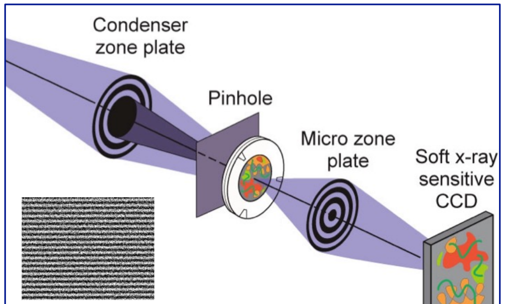


Ultrathin films



Compositional, space & depth-resolved imaging

Coherent diffraction imaging – a revolution in short wavelength imaging

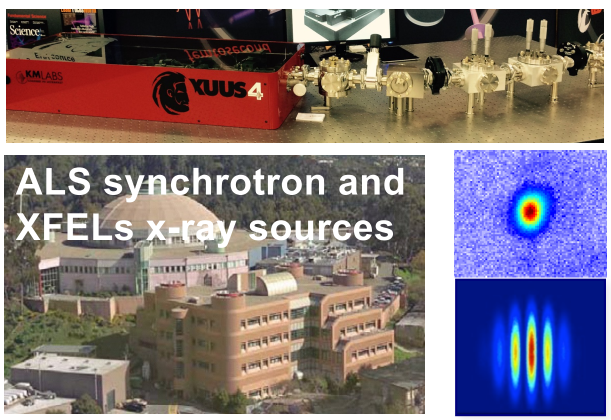


Nature ***435,*** *1210 (2005)*

Traditional X-ray microscopes

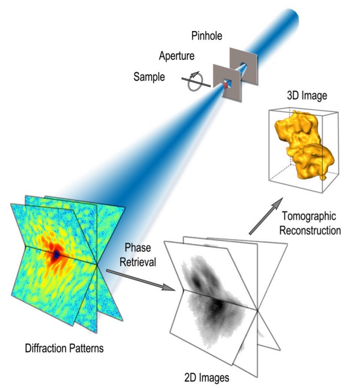
- Limited by lossy, imperfect, optics
- ~10x diffraction limited imaging

Coherent EUV/SXR sources



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

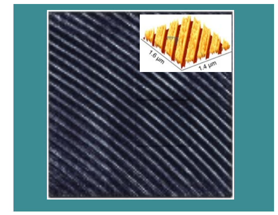
Coherent imaging



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

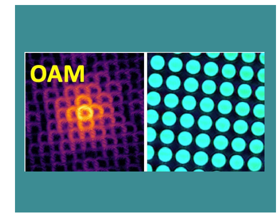
Fundamentally new imaging

2017: 1st sub-λ EUV imaging



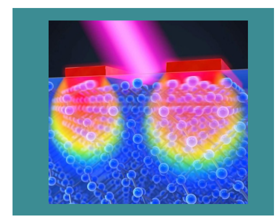
Nat. Photon. **11**, 22 (2017)

2023: Periodic samples



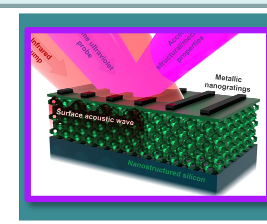
Optica ***10****, 1245 (2023)*

2021: Nano-hotspot



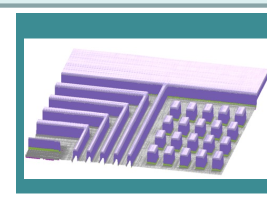
PNAS ***118****, 2109056118 (2021)*

2023: General transport



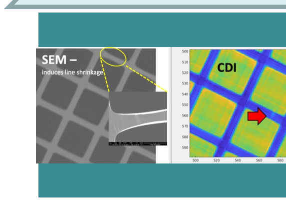
Nano Lett. ***23****, 2129 (2023)*

2021: Nondestructive Maps



Science Adv. ***7****, 9667 (2021)*

2023: Low-dose OAM

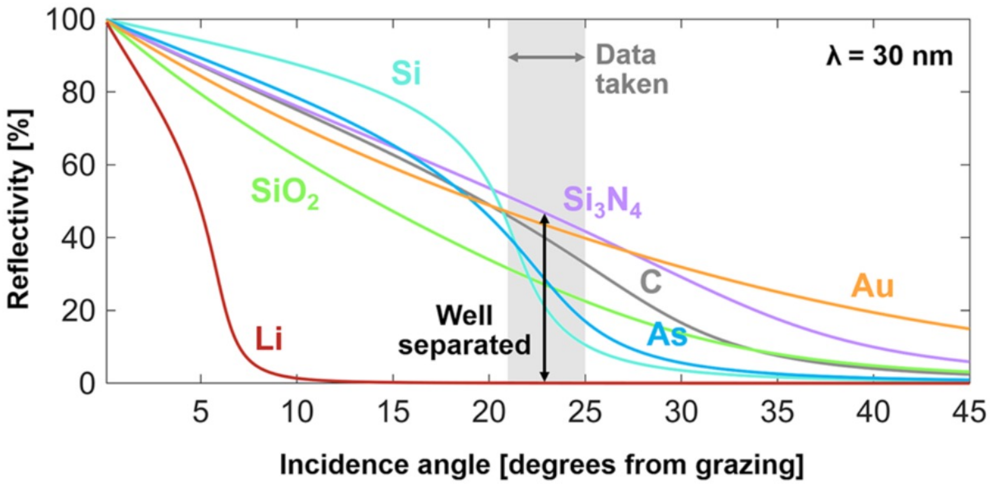


Optica ***10****, 1245 (2023)*

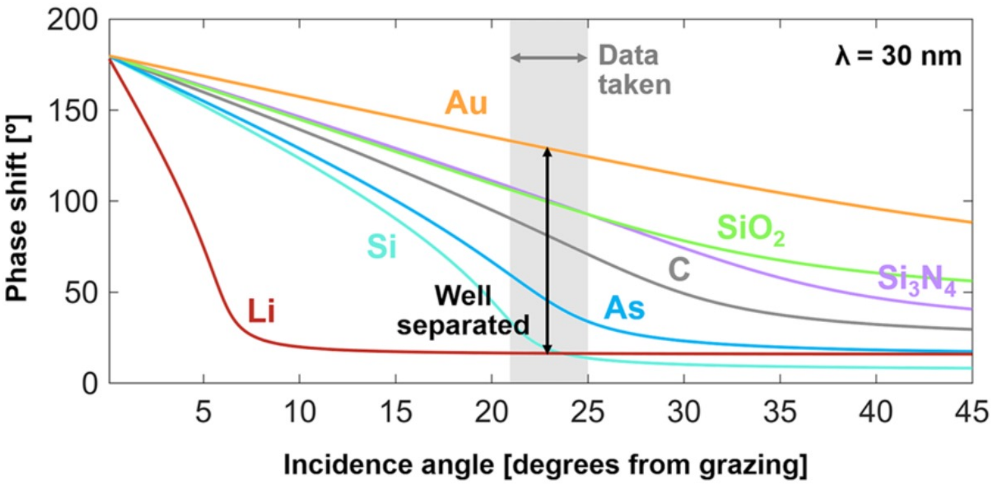
Ptychography in reflection

- Most materials still reflect in the EUV ($\lambda \geq 30\ nm$), with amplitude and phase contrast
- Can build a reflection-mode microscope in the EUV
- Most soft X-ray microscopes work in transmission

Reflectivity vs Angle in the EUV



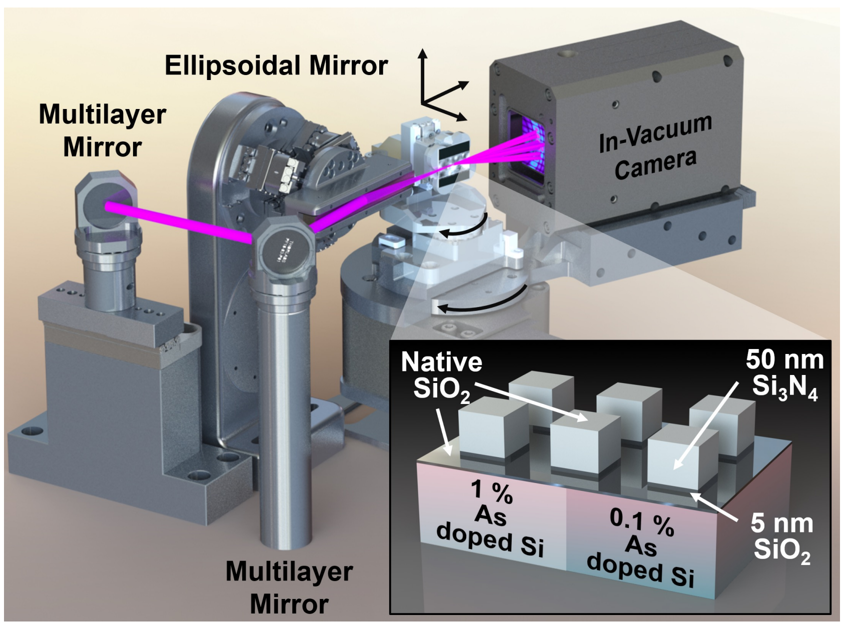
Phase Shift vs Angle in the EUV

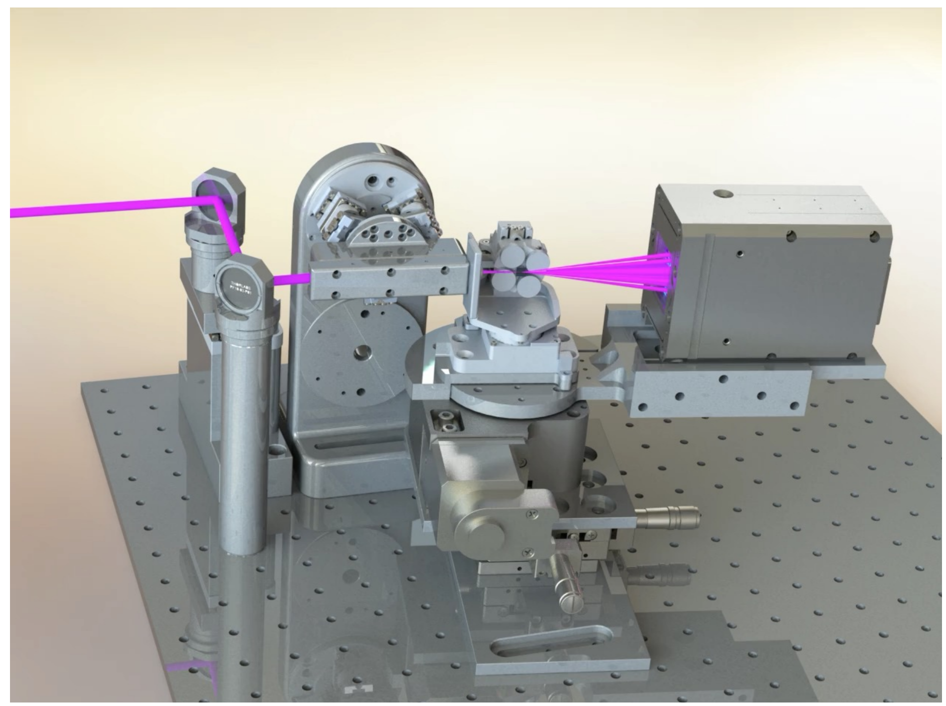


Phase-sensitive nondestructive EUV Imaging reflectometer

- Unique non-destructive spatial and depth-resolved compositional mapping
- Extract interfaces, layers, dopant profiles, composition of challenging semiconductor test samples (from imec, Samsung, 3M)

Tanksalvala et al., Sci. Advances ***7****, 9667 (2021)*
Esashi et al., RSI ***94****, 123705 (2023)*





STROBE — BUILDING THE MICROSCOPES OF TOMORROW

Phase-sensitive nondestructive EUV Imaging reflectometer

- Unique non-destructive spatial and depth-resolved compositional mapping
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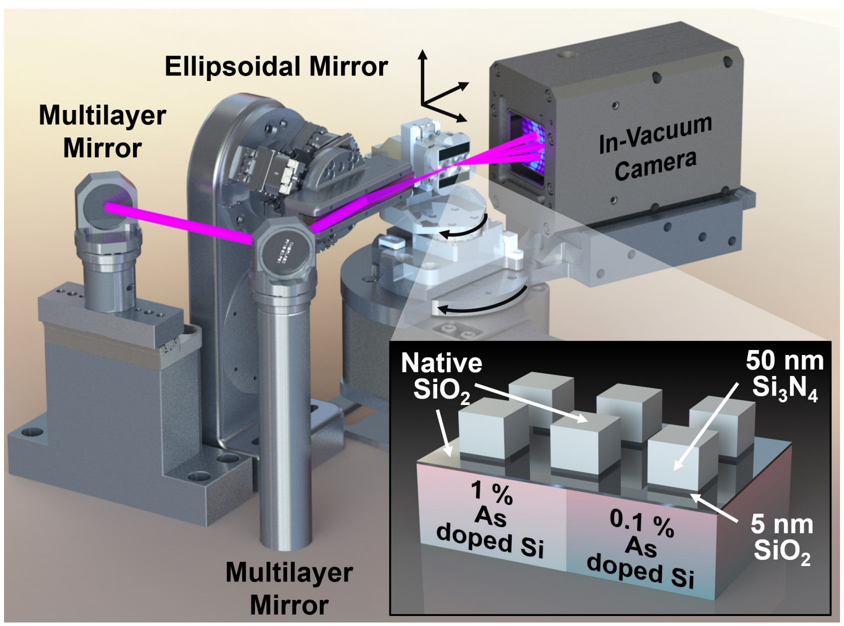
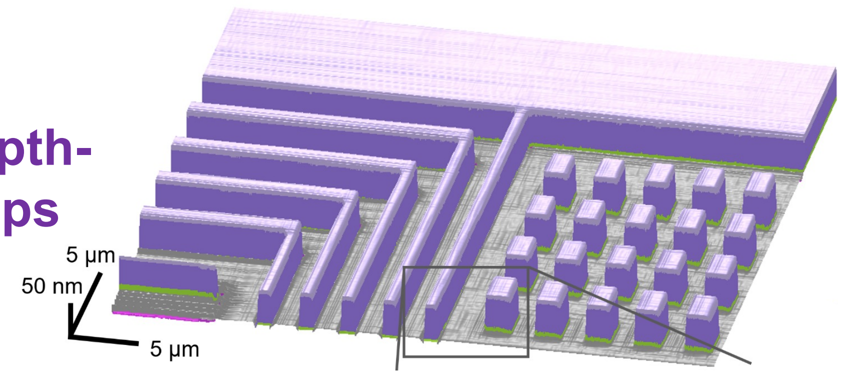
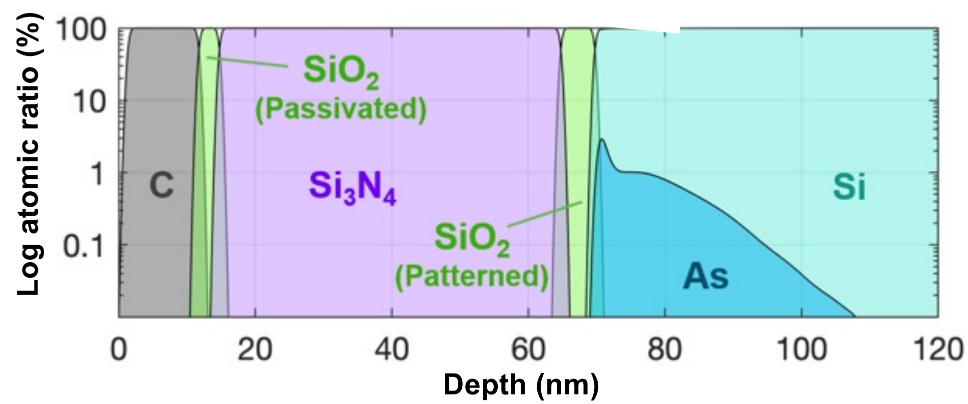
Tanksalvala et al., Sci. Advances ***7****, 9667 (2021)*
Esashi et al., RSI ***94****, 123705 (2023)*



Spatial & depth-resolved maps





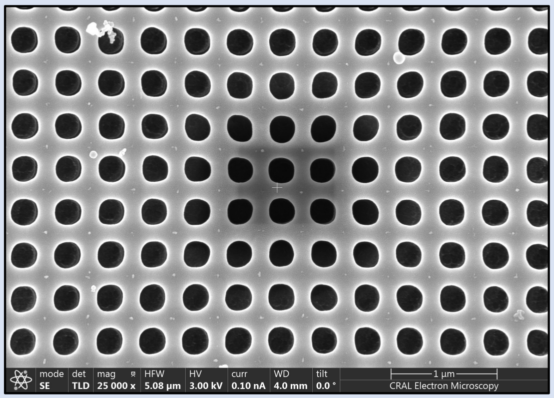
STROBE
BUILDING THE MICROSCOPES OF TOMORROW

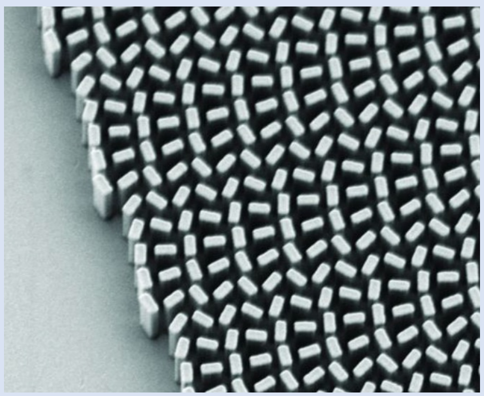
Low dose imaging of polymer metamaterials

3M

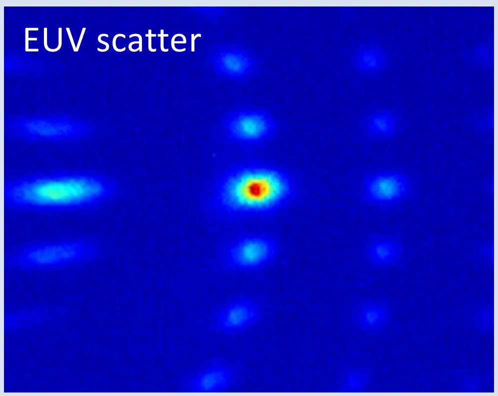
Metamaterials

Engineered subwavelength nanostructures with precisely controlled optical properties





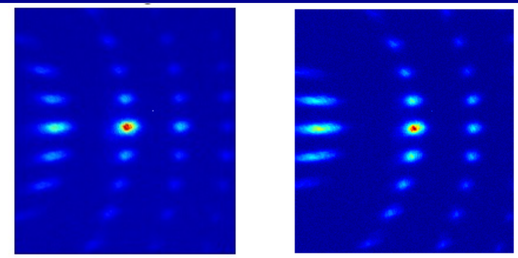
Khorasaninejad et al. Science 352, 6290 (2016)



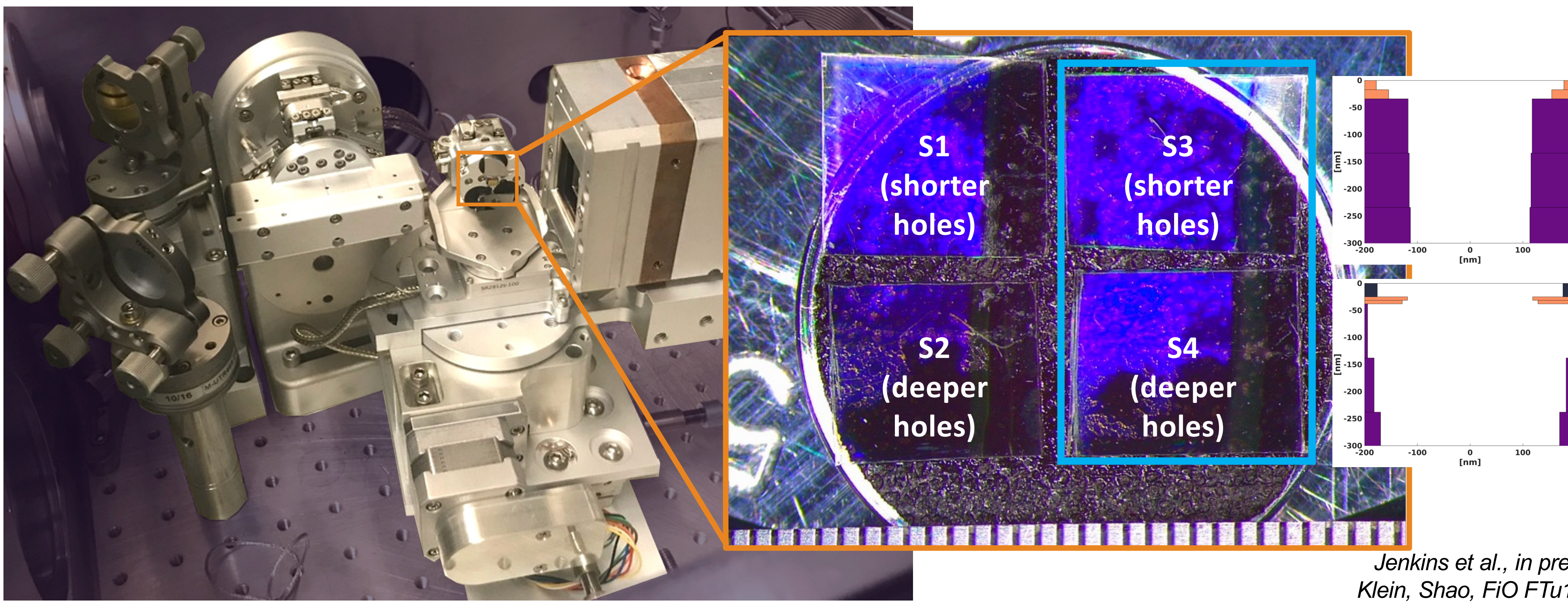
Challenge: Beam damage from electron imaging, and sample cutting & coating

Nondestructive EUV scatterometry of polymers

- HHG scatterometry is fast, no sample prep, non-destructive
- Extract layer thicknesses, densities, top-surface geometry
- Verified with AFM and very specialized transmission electron microscopy

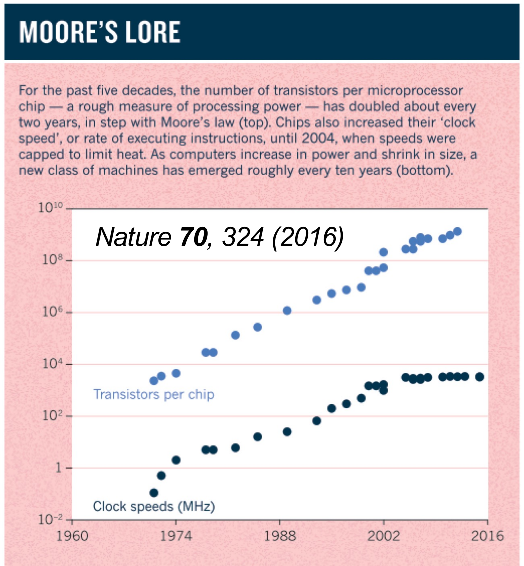


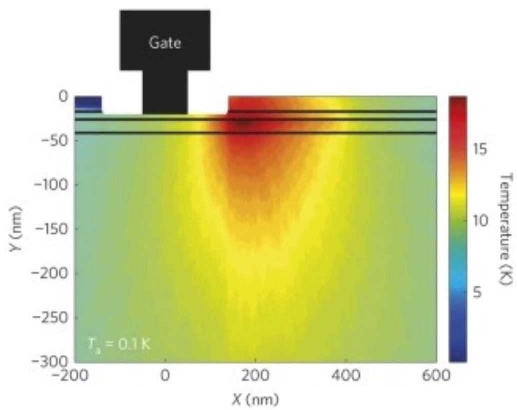
EUV diffraction patterns



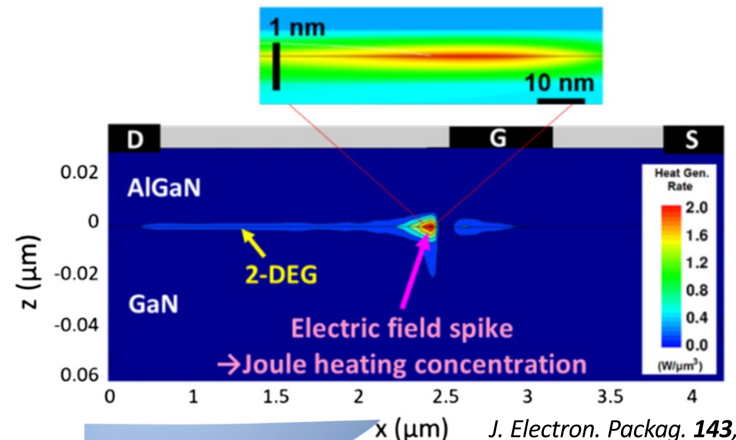
Jenkins et al., in prep. (2024)
Klein, Shao, FiO FTu1B (2024)

Nanoscale Thermal Transport: Transistor clock rates, wide bandgap power electronics





Nat. Mater. **14**, 187 (2015)



J. Electron. Packag. **143**, 020804 (2021)

Quantum devices

Electric vehicles

Communication

No general models to describe nanoscale heat transport

Theory and measurements off by 100x, over-predict lifetime, under-predict noise floor

Nat. Comm. **5**, 4015 (2014)

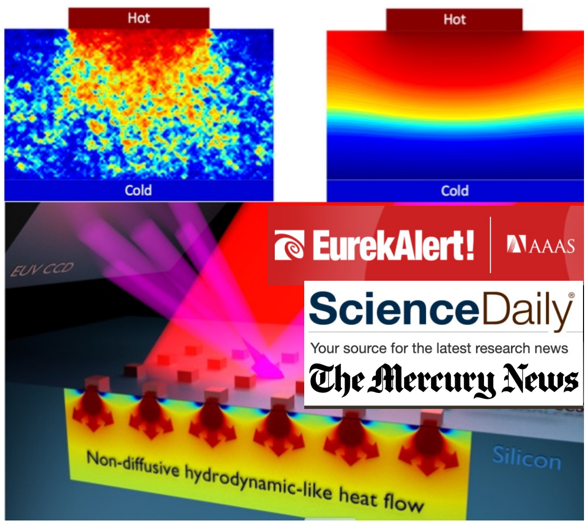
STROBE
BUILDING THE MICROSCOPES OF TOMORROW

First predictive and general understanding of heat transport in nanostructured semiconductors!

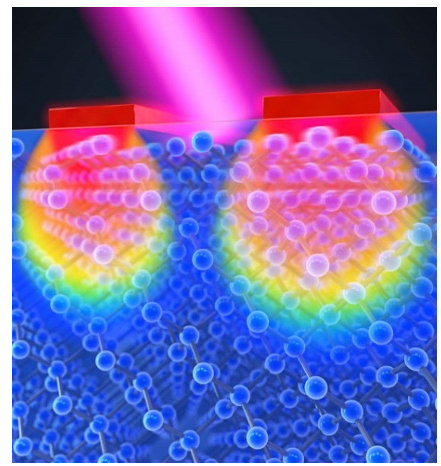
Ann and H.J. Smead
Aerospace Engineering Sciences
JILA UAB Universitat Autònoma de Barcelona

- <u>New findings</u>: Heat can be channeled at the nanoscale in 1D/2D geometries, rather than spreading diffusively
- <u>Achievement</u>: 1st comprehensive, predictive, understanding of 1D & 2D nanoscale heat transport
- <u>Significance</u>: Routes for thermal management in nano, quantum, energy devices

Nanoscale **Macroscale**



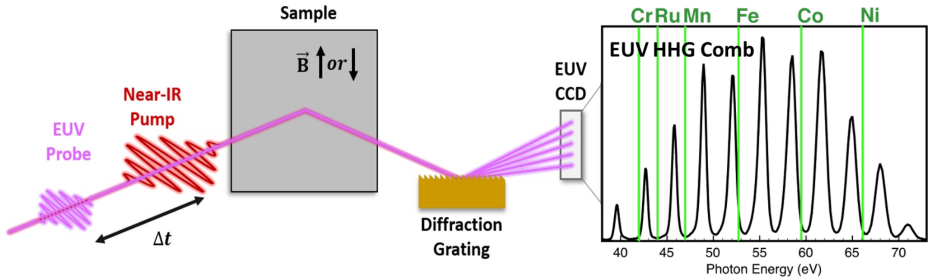
Mechanism: enhanced in-plane phonon scattering → enhanced cross-plane transport



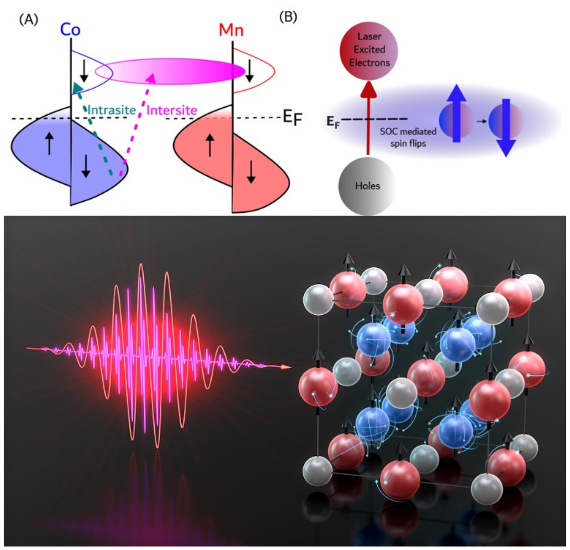
Karl et al., Science Advances ***4****, eaau4295 (2018); Frazer et al., Phys. Rev. Applied* ***11****, 024042 (2019)*
Frazer et al., Phys. Rev. Materials ***4****, 073603 (2020); Beardo, Knobloch et al., ACS Nano* ***15****, 13019 (2021)*
Honavar et al., PNAS ***118****, e2109056118 (2021); Knobloch et al., ACS AMI* ***14****, 41316 (2022); McBennett et al., Nano Letters* ***23****, 2129 (2023); submitted (2024)*

High harmonics probe the site-specific dynamic magnetic state

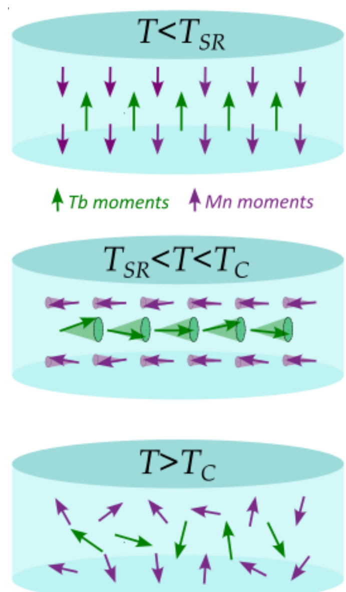
La Vorakiat et al., PRL **103**, 257402 (2009)
Turgut et al., PRL **110**, 197201 (2013)
Tengdin et al., Science Adv. **6**, eaaz1100 (2020)
Ryan et al., Science Adv. **9**, eadi1428 (2023)
Rana et al., Nature Nanotech **18**, 227 (2023)
Ryan et al., arXiv:2407.18894 (2024)



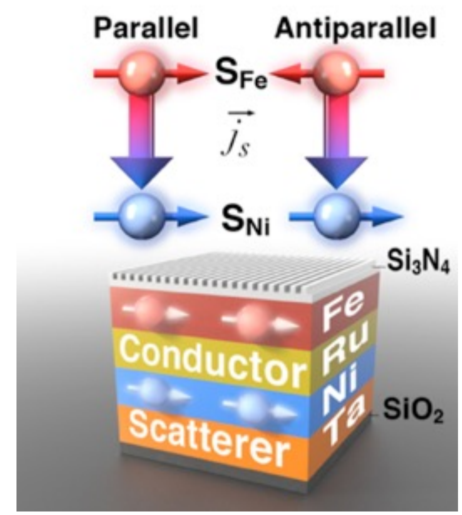
Spin transfer, Spin flips (~fs)



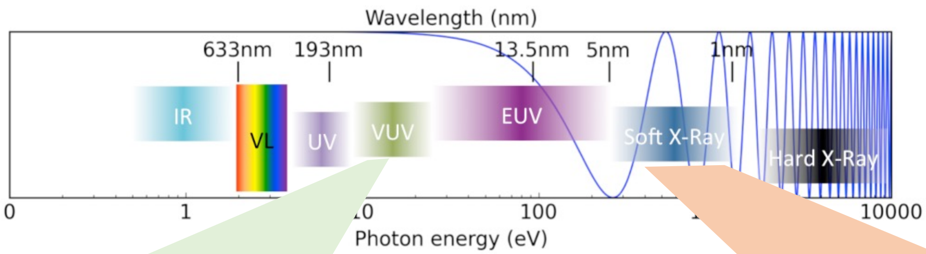
Spin reorientation (~ps)



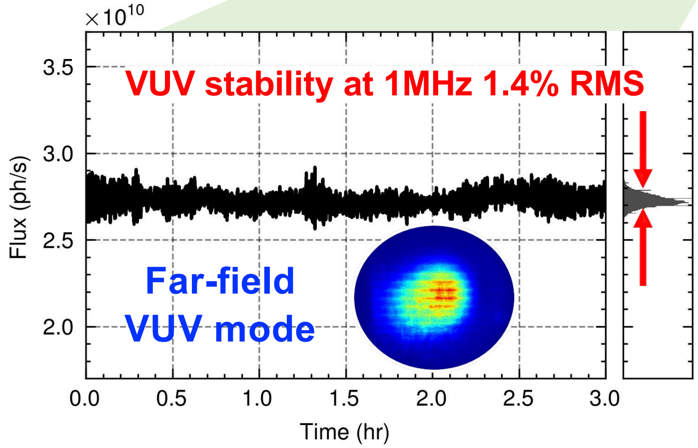
Spin transport (~fs)

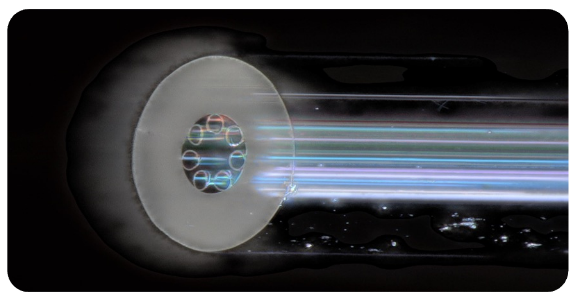


Expanding the spectrum: Soft X-ray and VUV



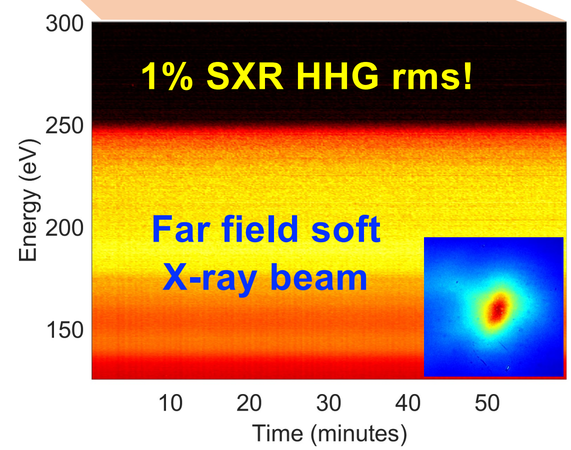
Narrowband VUV





Anti-resonant hollow-core fibers

Broadband SXR

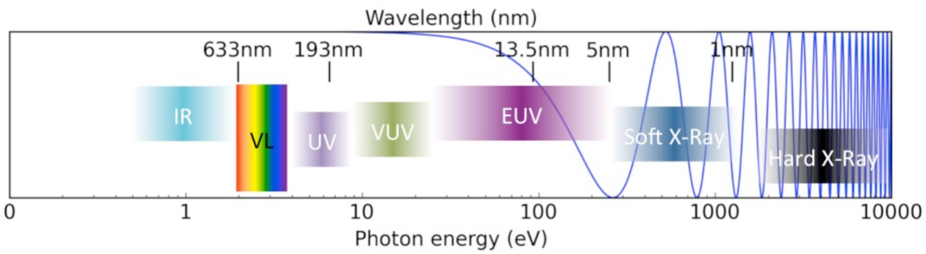


*Couch et al., Optica **7**, 832-837 (2020)*
Thurston et al., FIO FTu6E (2024); submitted (2024)

Hettel et al, FiO FM1E (2024); In prep. (2024)

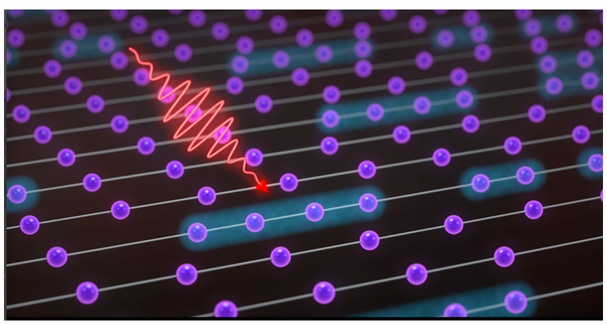
VUV spectral region – hard, neglected, but important!

VUV
~60-200 nm
~6-20 eV



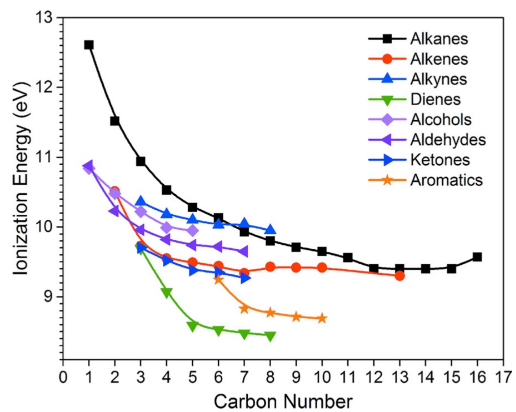
H. Kim, No. RWTH-2017-02814. Fachgruppe Physik (2016)

Photoelectron and photovoltage spectroscopies: electronic structure



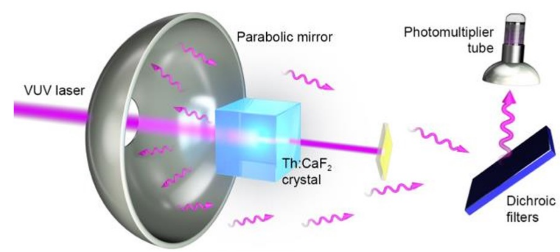
*Zhang et al., Nano Letters **23**, 8392 (2023)*

Access molecular ionization energies: combustion, hypersonics



*Zhou et al., J. Synchrotron Rad. **23**, 1035 (2016)*
*Couch et al., Proc. Combustion Institute **38**, 1737 (2021)*

VUV combs for nuclear clocks



^{229m}Th nuclear state
8.338 ± 0.24 eV (~148nm)

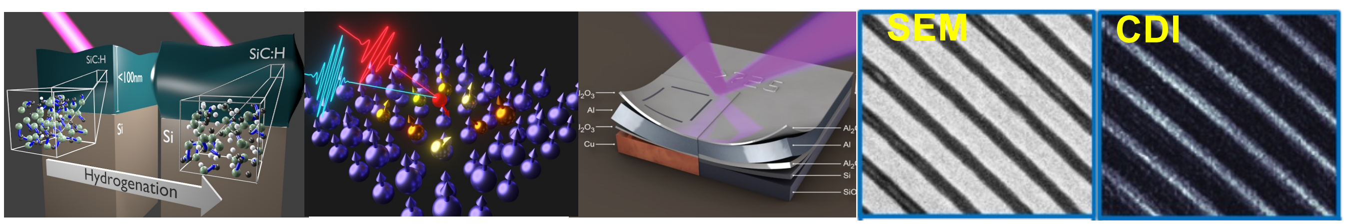
*Kraemer et al., Nature **617**, 706 (2023)*
*Zhang et al., Nature **633**, 63 (2024)*

Summary

- High harmonic sources are a unique quantum technology allowing exquisite control over EUV and soft X-ray light

- The limits of HHG technology are not yet known: 10keV, 50keV?

- HHG technology is already a useful tool
  - First sub-wavelength EUV imaging
  - Non-destructive maps of heterostructures for quantum technologies
  - New understanding about nanoscale thermal transport
  - New understanding of spin and charge dynamics
  - Distinguish electron-electron screening and scattering in quantum materials
  - Bright future – everything scales with the wavelength



Light science **Materials Characterization** **Spin Dynamics** **ARPES** **Imaging**