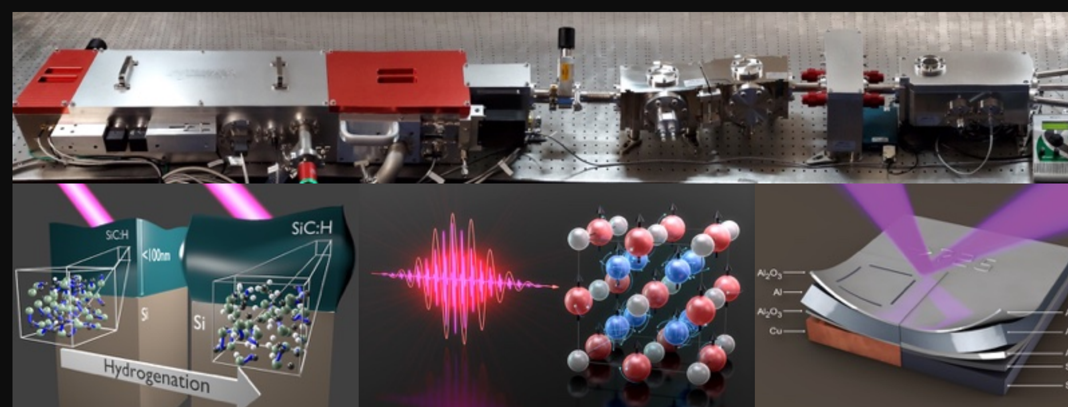
Tabletop X-Ray Lasers: *Revealing Spin, Charge and Phonon Dynamics and their Interactions*

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



JILA NIST CU
STROBE
KMLABS inc.
LEADING IN ULTRAFAST

JILA
CU Boulder and NIST

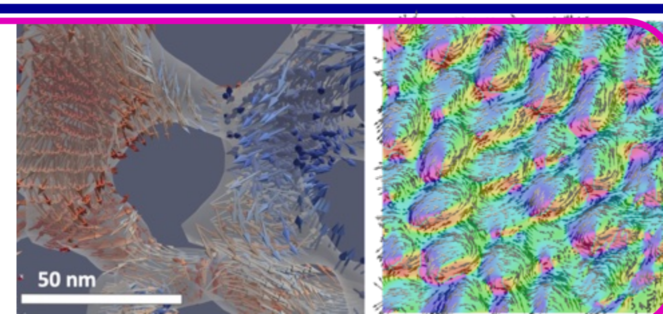
STROBE
BUILDING THE MICROSCOPES OF TOMORROW

Putting High Harmonic Quantum Technologies to Work

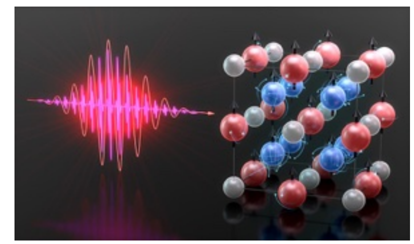
Kapteyn-Murnane Group, Alumni, KMLabs, Collaborators

Outline

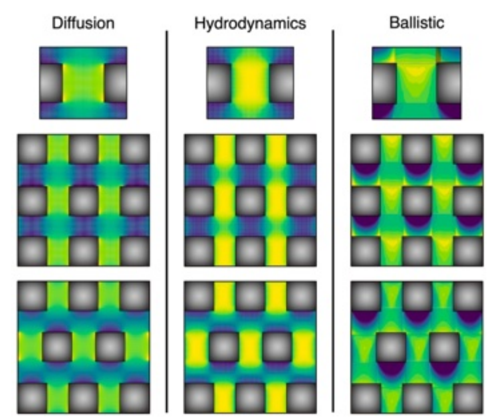
- **Lecture 1 – Brief intro to the physics of High Harmonics (Nobel 2023) and coherent imaging**



- **Topic 2 – Nanoscale phonon transport** (with Dr. Brendan McBennett, NIST)



- **Topic 3 – Understanding spin and many-body electron-phonon dynamics** via EUV MOKE and ARPES



Timescales of this talk: picoseconds and femtoseconds

- 1000,000,000,000,000 femtoseconds = 1 second
- 1000,000,000,000,000 seconds = 30 million years

1000,000,000,000 picoseconds = 1 second

1000,000,000,000 seconds = 30 thousand years

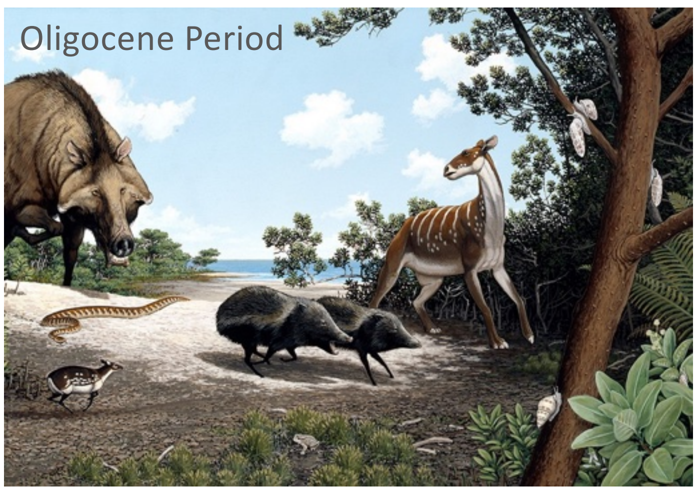
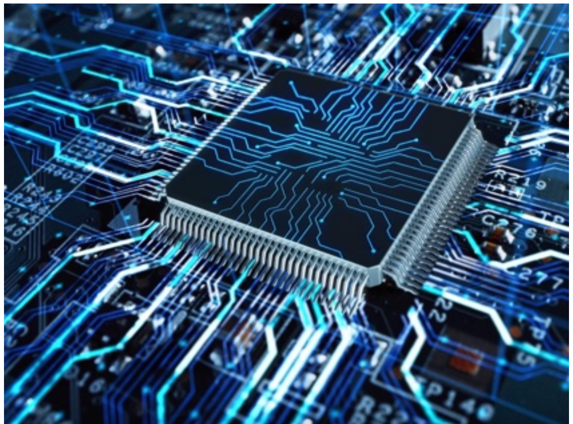


Image credit: www.floridamuseum.ufl.edu

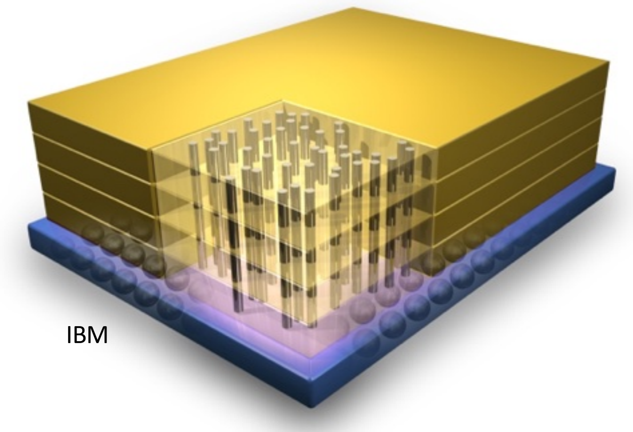
Image credit: André Houot

Nanostructured materials have very different properties than bulk



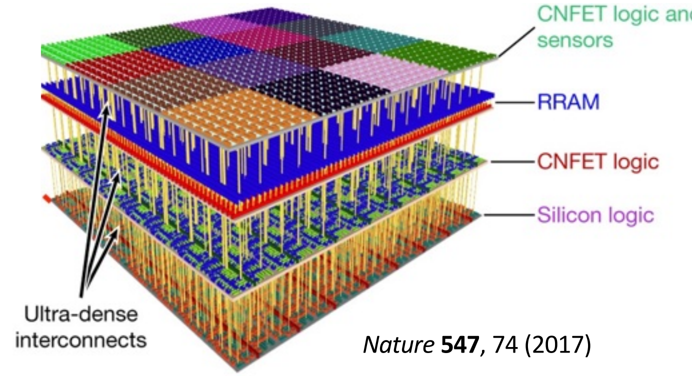
iStock/gorodenkoff

2.5 & 3D packaging



IBM

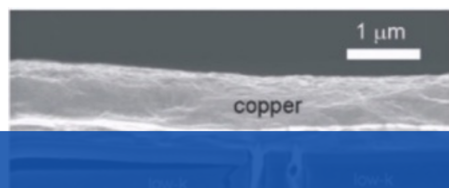
Novel architectures



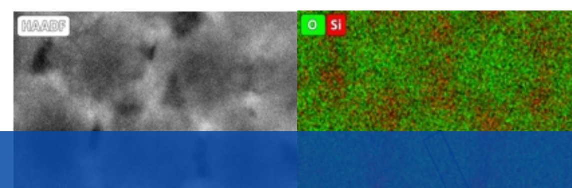
Nature **547**, 74 (2017)

Thermal

Mechanical



Structural/Interface

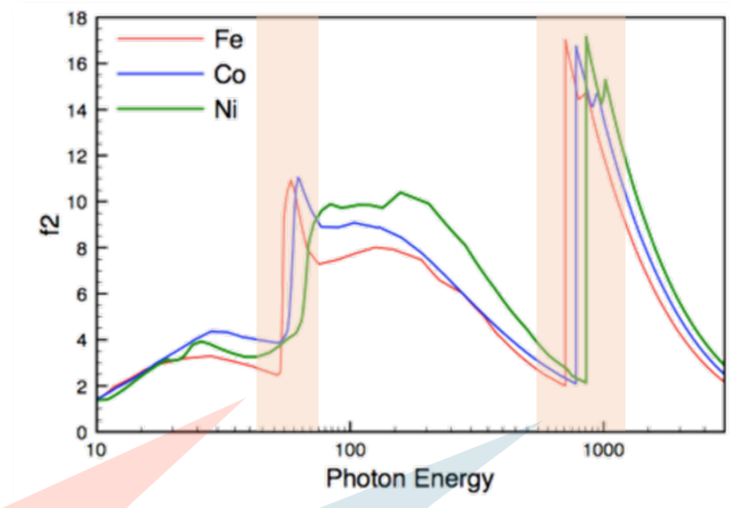


Electrical/Magnetic

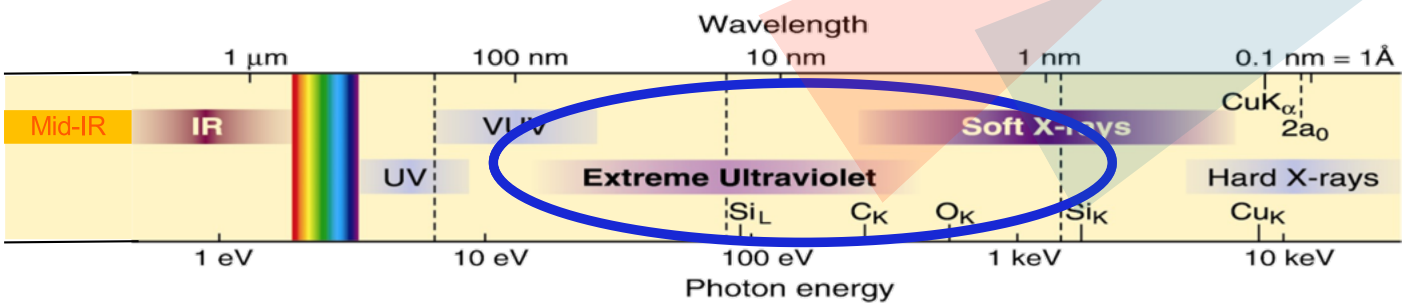
Critical need for better functional metrologies

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

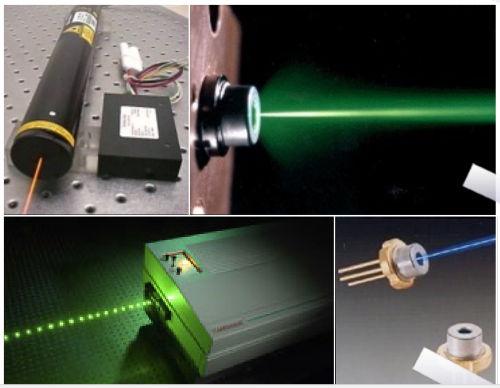
- Wavelengths $\lambda \sim 1 - 100\ nm;\ \ h\nu \sim 10 \rightarrow 1000\ eV$
- Elemental, chemical, magnetic specificity
- Non-destructive, low-dose compared with SEM, TEM
- Image with few-nm resolution
- Capture dynamics relevant to function (~fs)

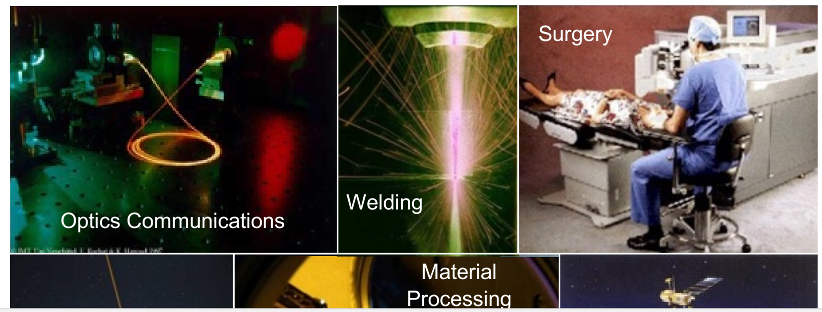


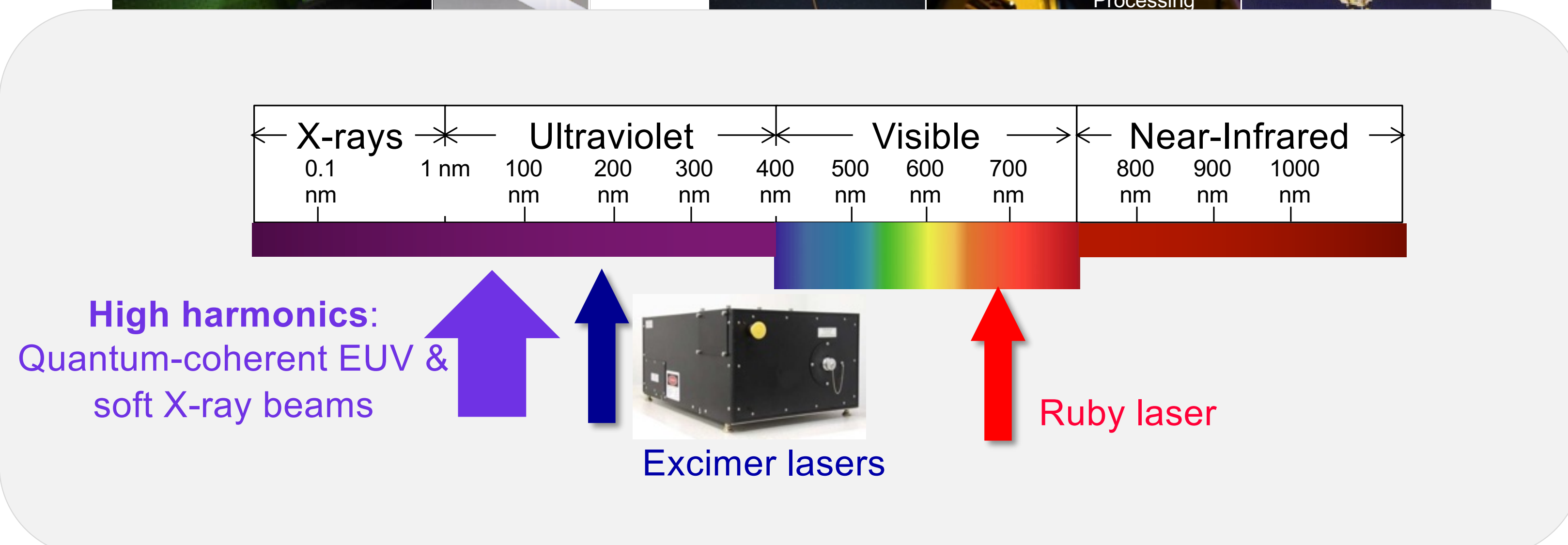
Requires coherence



Tabletop scale lasers in widespread use







Discovery of high harmonics of light: 1987

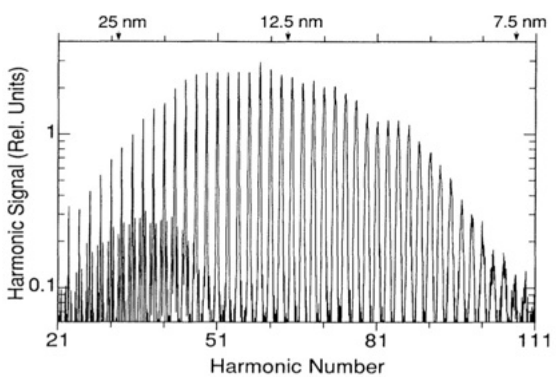
Studies of multiphoton production of vacuum-ultraviolet radiation in the rare gases

A. McPherson, G. Gibson, H. Jara, U. Johann, T. S. Luk, I. A. McIntyre, K. Boyer, and C. K. Rhodes

Department of Physics, University of Illinois at Chicago, P.O. Box 4348, Chicago, Illinois 60680

Received November 1, 1986; accepted December 18, 1986

Measurements of the vacuum-ultraviolet (<80-nm) radiation produced by intense ultraviolet (248-nm) irradiation (10^{15}–10^{16} W/cm^2) of rare gases have revealed the copious presence of both harmonic radiation and fluorescence from excited levels. The highest harmonic observed was the seventeenth (14.6 nm) in Ne, the shortest wavelength ever produced by that means. Strong fluorescence was seen from ions of Ar, Kr, and Xe, with the shortest wavelengths observed being below 12 nm. Furthermore, radiation from inner-shell excited configurations in Xe, specifically the $4d^95s5p \rightarrow 4d^{10}5s$ manifold of Xe^{7+} at ~17.7 nm, was detected. These experimental findings, in alliance with other studies concerning multielectron processes, give evidence for a role of electron correlations in a direct nonlinear process of inner-shell excitation.



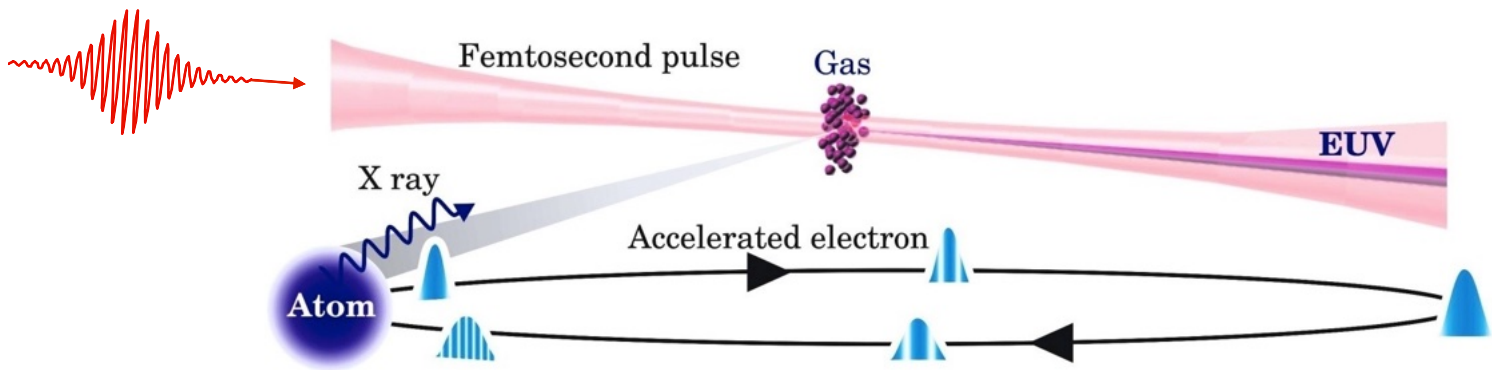
How are they emitted?

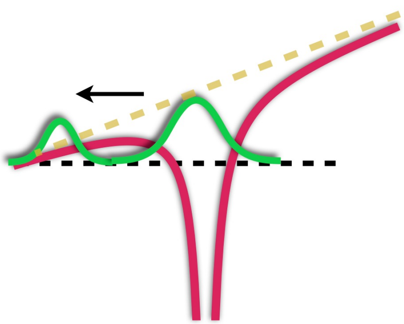
Advice – ignore since likely nothing new…

More than 100 harmonics of near-infrared light!!

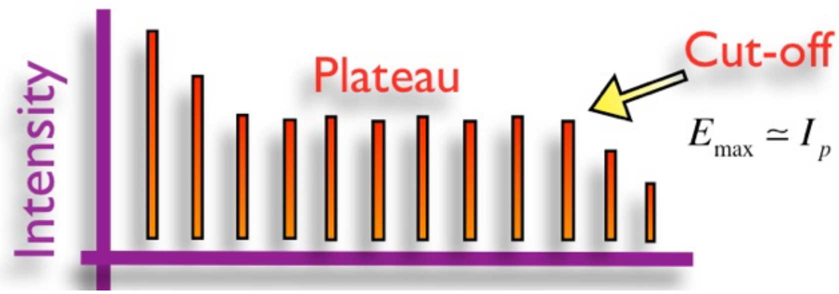
McPherson, et al, JOSA B 4, 595 (1987)
Ferray et al., J Phys B 21, L31 (1988)
Macklin et al, PRL 70, 766 (1993)

High harmonic generation

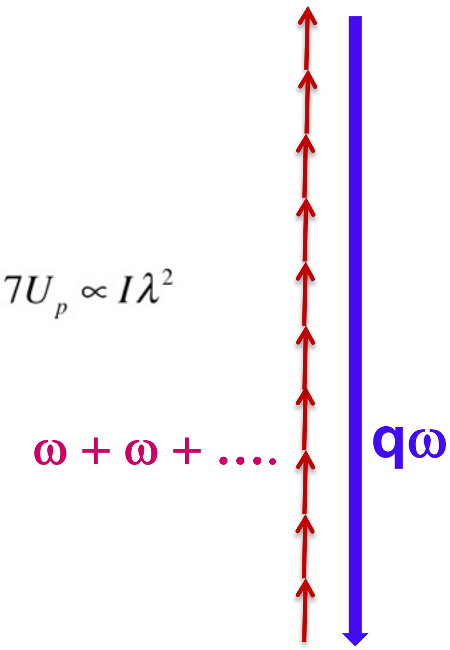




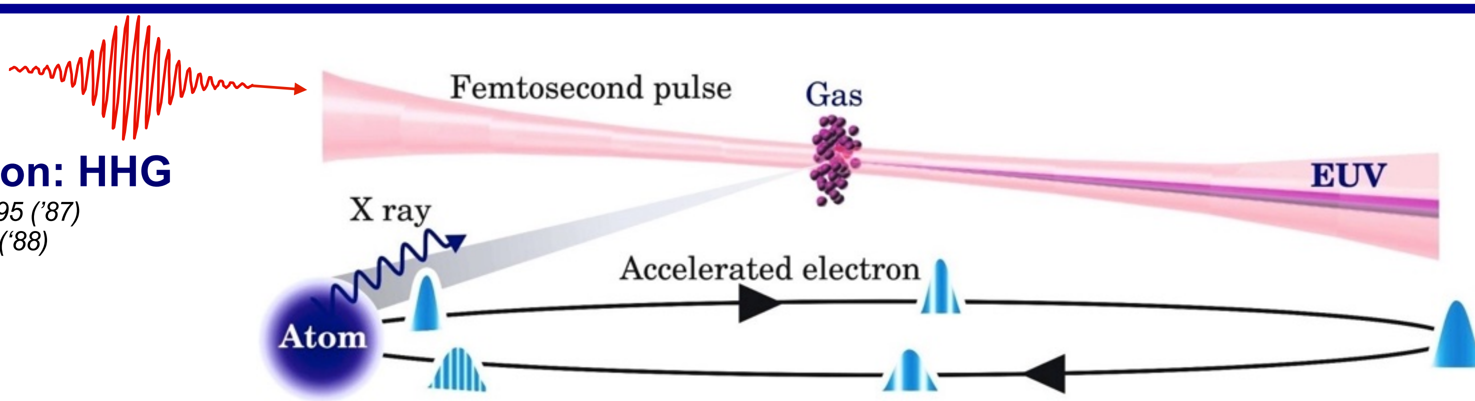
Tunnel ionization



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



High harmonic generation: nanoscale quantum antenna

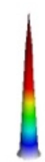


New observation: HHG

McPherson, JOSA B 4, 595 ('87)
Ferray, J Phys B 21, L31 ('88)

Quantum nature of HHG

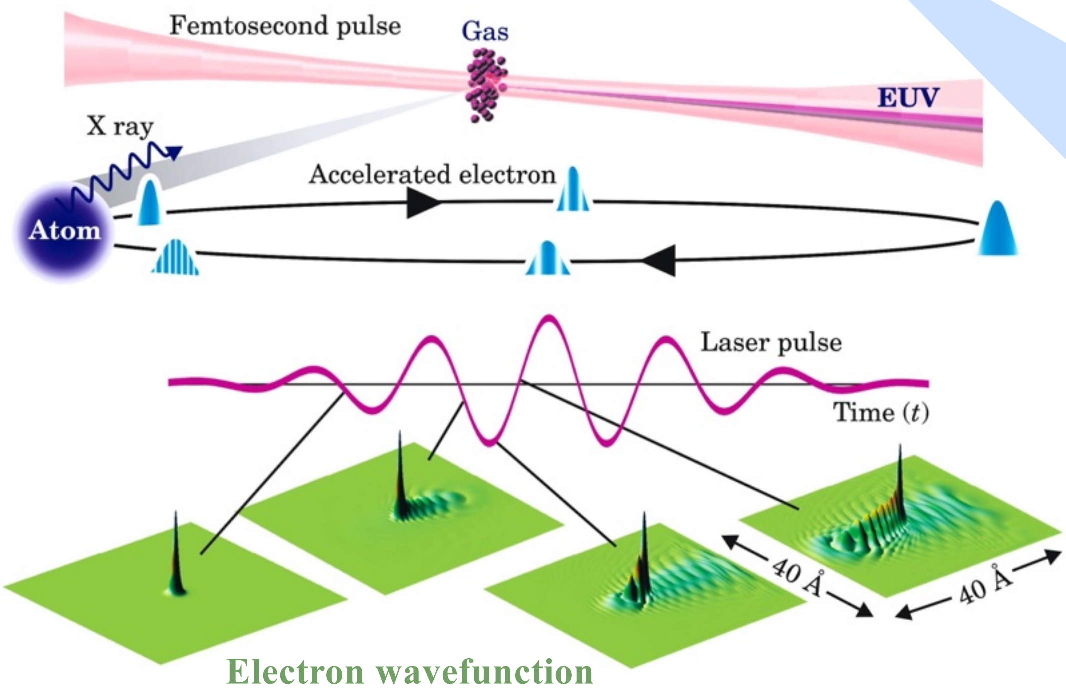
*Krause, Schafer, Kulander, PRL **68**, 3535 (1992)*
*Lewenstein, PRA **49**, 2117 (1994)*



High harmonic generation: Quantum phase of radiating electron

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

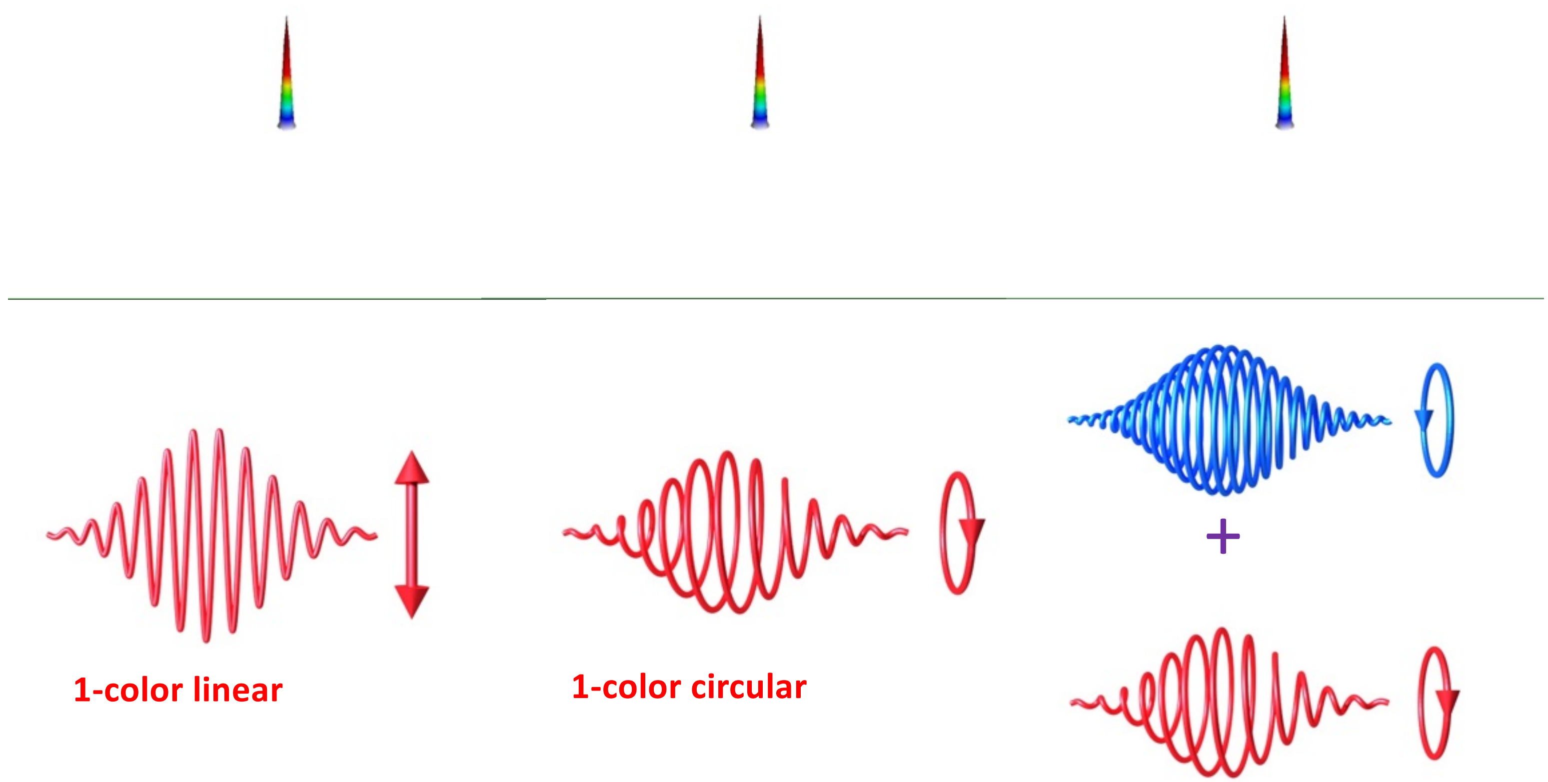
Tunnel ionization — Propagation — Recombination

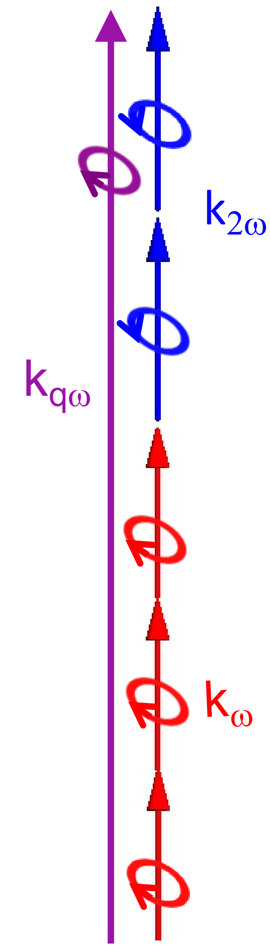


$$\varphi_{EUV} \propto \frac{2\pi}{h} I_{laser} \lambda^2 \tau$$

$$>> 2\pi!$$

Quantum sculpting: circularly polarized harmonics

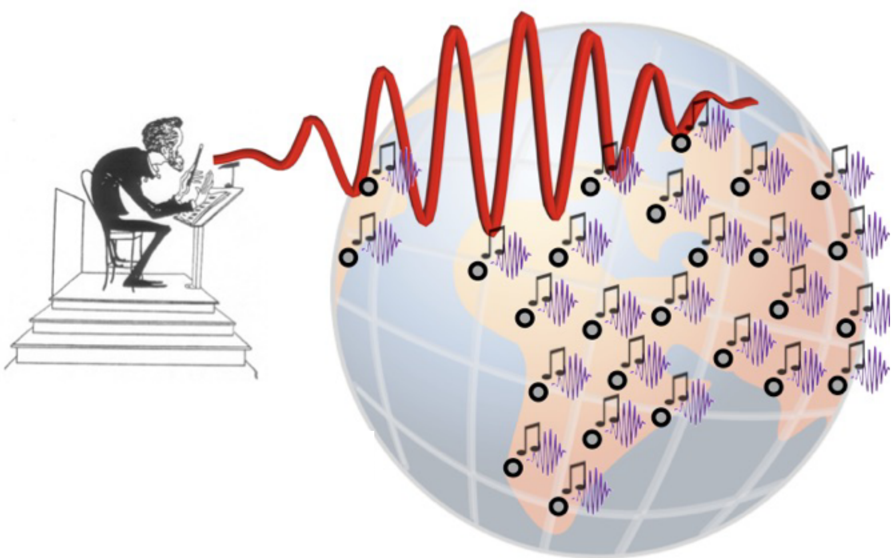




*PRA **51**, R3414 (1995); PRA **52** 2262 (1995)*
*Nature Photonics **9**, 99 (2015); PNAS **112**, 14206 (2015)*
*Nature Photonics **9**, 743 (2015); Science **348**, 530 (2015)*

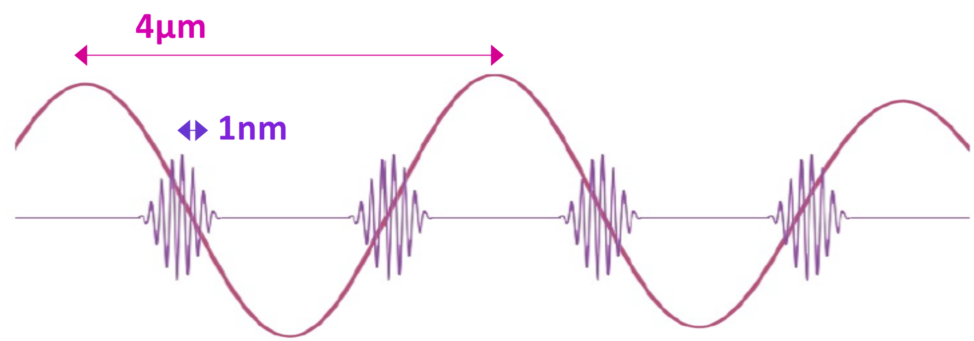
Bright HHG beams → synchronize emission from trillions of atoms

The IR laser is a quantum conductor, directing a trillion atomic musicians!



... each atom in the gas as a musician, emitting attosecond pulses at the rate dictated by the conductor.

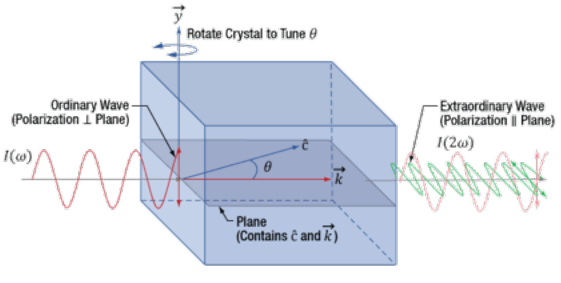
Image courtesy of Carlos Hernandez-Garcia



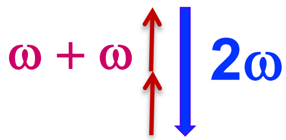
- Laser wavelength: 800 - 4000nm
- X-ray wavelength: 1 – 30nm
- X-ray fields must add in phase to sub-Å spatial resolution and sub-attosecond (10^{-18} s) temporal resolution
- Nature does this passively due to the quantum nature of strong field ionization!!

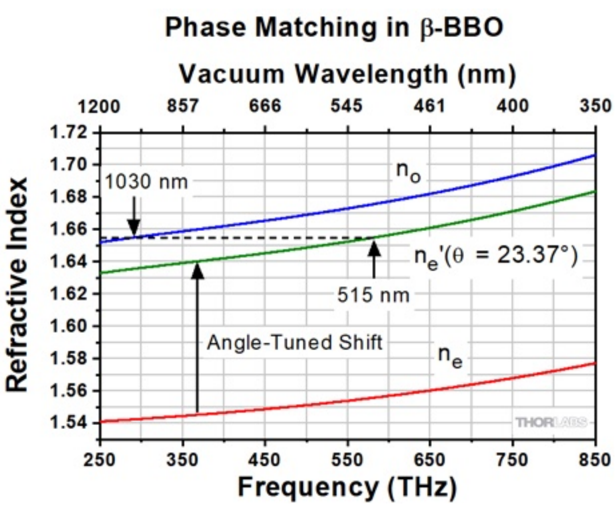
Bright high harmonic emission - *phase matching in time domain*

Visible NLO: phase matching in spatial domain

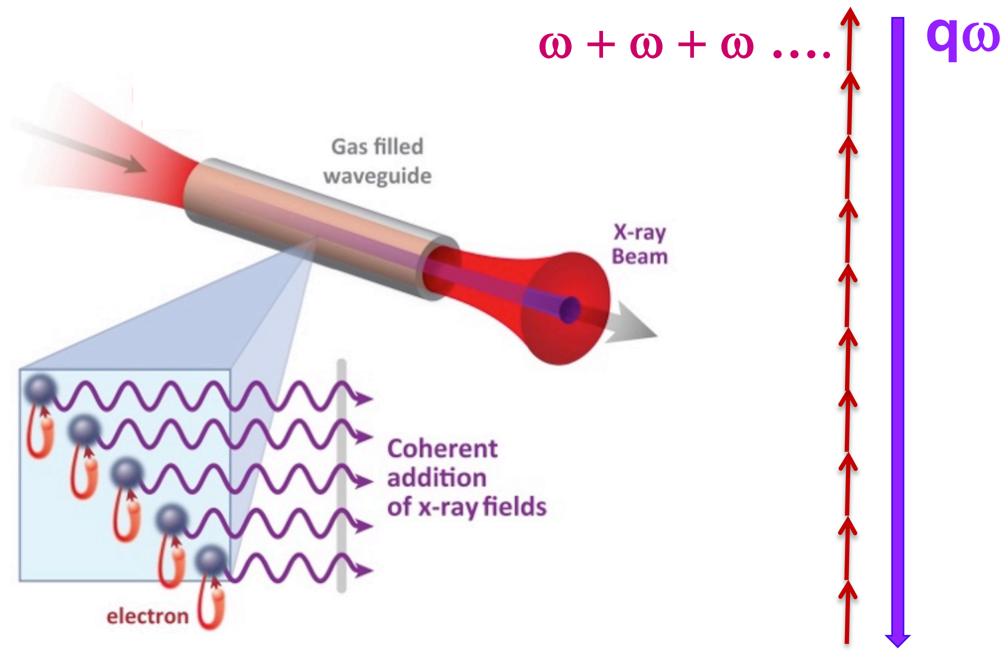


Type-I Phase Matching Using a β-BBO Crystal





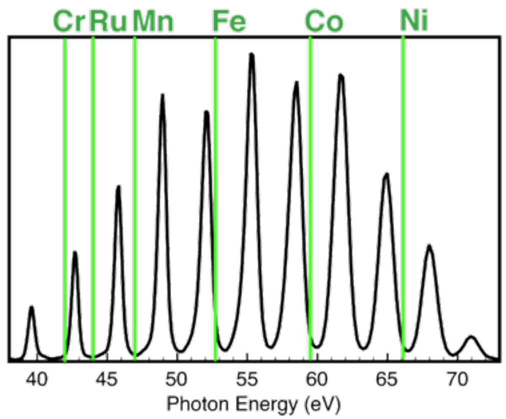
Extreme nonlinear optics: phase matching in time domain

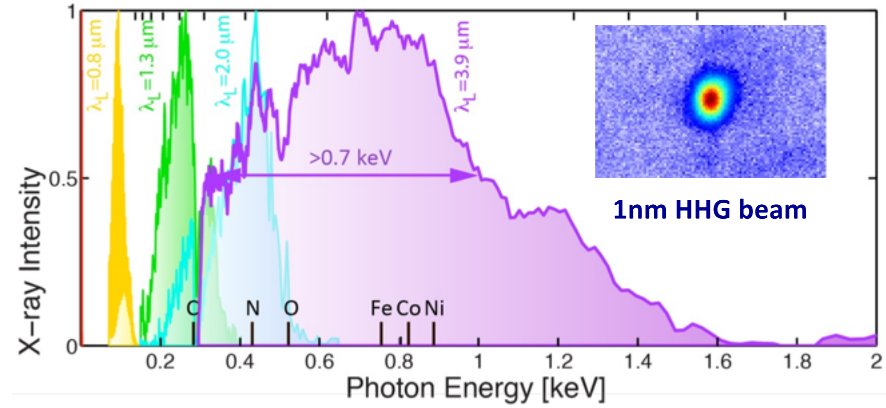


Laser and x-ray phase velocities are matched at the speed of light ONLY during part of the laser pulse

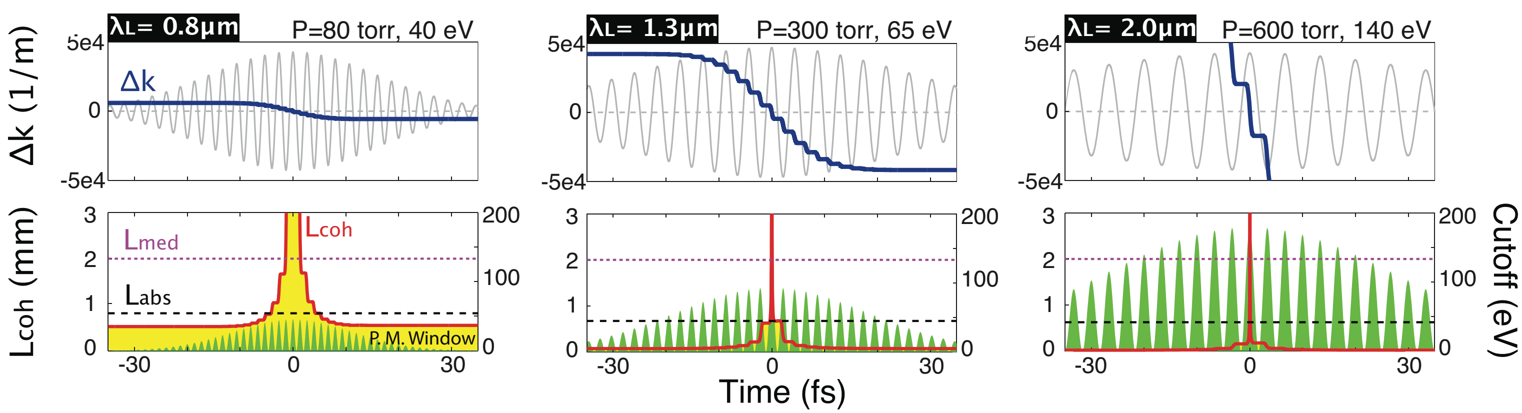
Rundquist Ph.D. Thesis (1998); Science ***280****, 1412 (1998)*

Phase matching window sculpts the HHG in time and frequency





Near-IR laser → **Mid-IR laser**



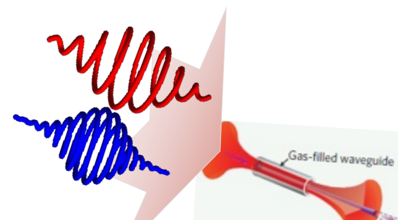
Rundquist Ph.D. Thesis (1998)
*Rundquist et al., Science **280**, 1412 (1998)*
*Bartels et al., Science **297**, 376 (2002)*
*Popmintchev et al., Science **336**, 1287 (2012)*
*Chen et al., PNAS **111**, E2361 (2014)*

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

JILA CU Boulder and NIST

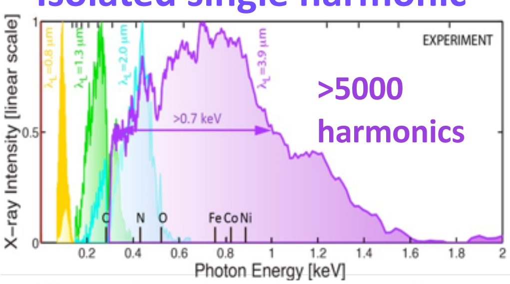
- 12 octave supercontinuum or isolated peaks
- Full temporal coherence: femto to atto to zepto
- Full spatial coherence to keV
- Bright linear, circular and OAM beams
- Unique light source better than we dreamed of!
- **Exquisite control over X-ray light using lasers**

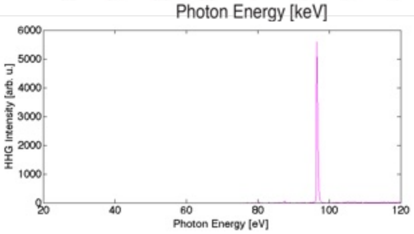
Science **364**, 9486 (2019); *Nat. Photon.* **13**, 123 (2019); *Science Ad.* **2**, e1501333 (2016) *PNAS* **112**, 14206 (2015); *Nature Photonics* **9**, 99 (2015); *Nature Photon.* **9**, 743 (2015)



Linear/circular OAM/SAM shaped

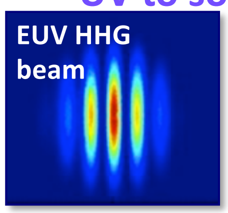
Broad coherent continuum OR Isolated single harmonic

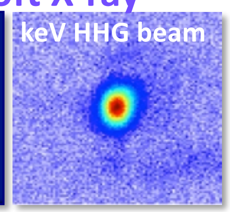


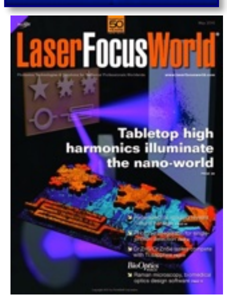


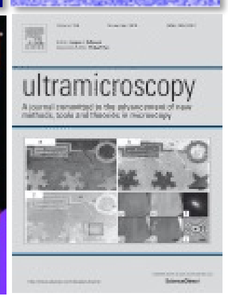
Science **336**, 1287 (2012); *Science* **350**,1225 (2015)

Full spatial coherence: UV to soft X-ray



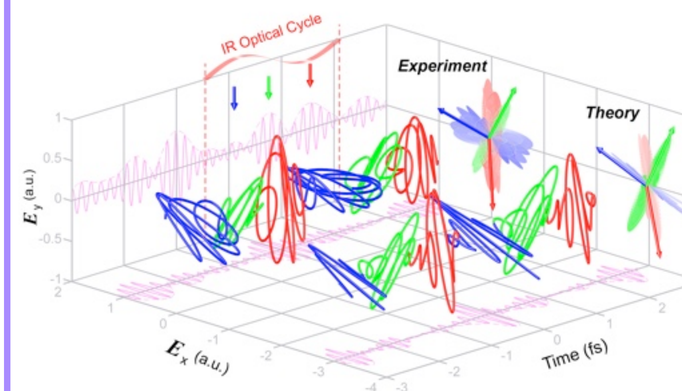


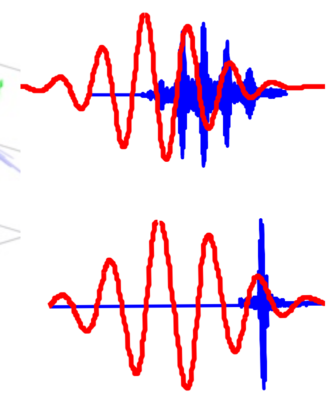




Science **297**, 376 ('02); *Science* **348**, 530 ('15)

Attosecond pulses and pulsetrains: UV to soft X-ray



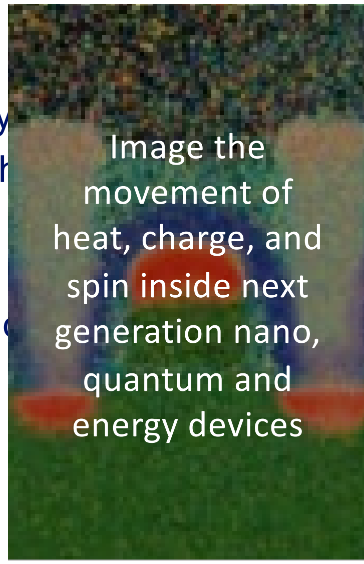


PRL **111**, 033002 (2013); *PNAS* **111**, E2361 (2014); *PRL* **120**, 093002 (2018); *Optica* **7**, 832 (2020)

JILA CU Boulder and NIST

The Nano, Bio & Materials Challenge

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

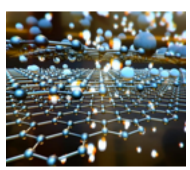


- Challenge: New energy … um and energy devices … e and integrate materials, with … on — and correlate fun…

- Challenge: Need functi… t work without cutting, …

Integrated circuits

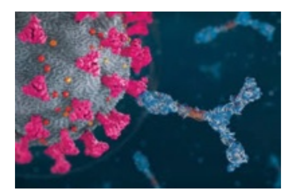
Photonic, 2D, quantum materials



Nano-engineered thermoelectrics

Solar technologies

Bio



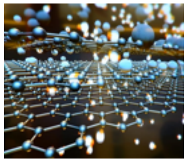
The Nano, Bio & Materials Challenge

- 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 can see better and see things as they happen!!

Integrated circuits

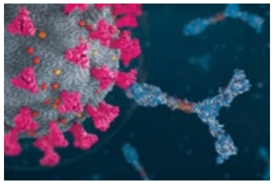
Photonic, 2D, quantum materials



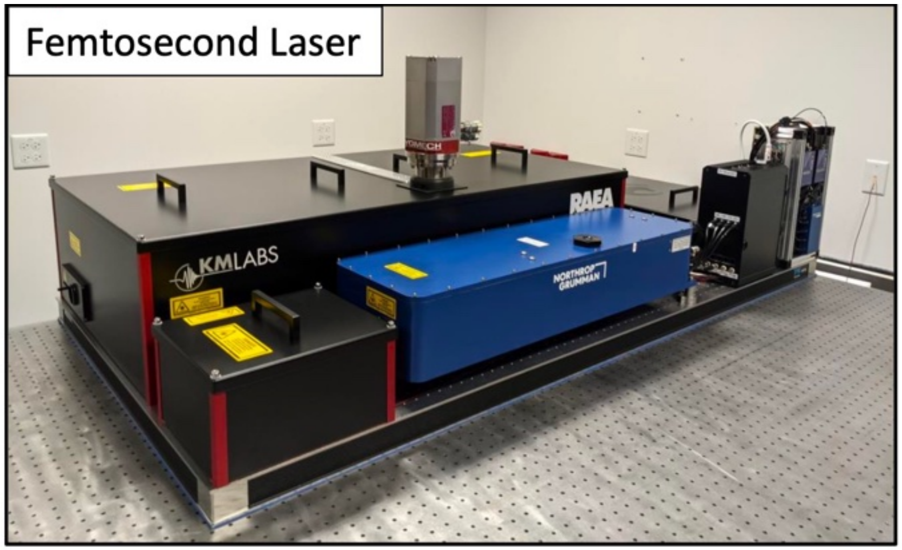
Nano-engineered thermoelectrics

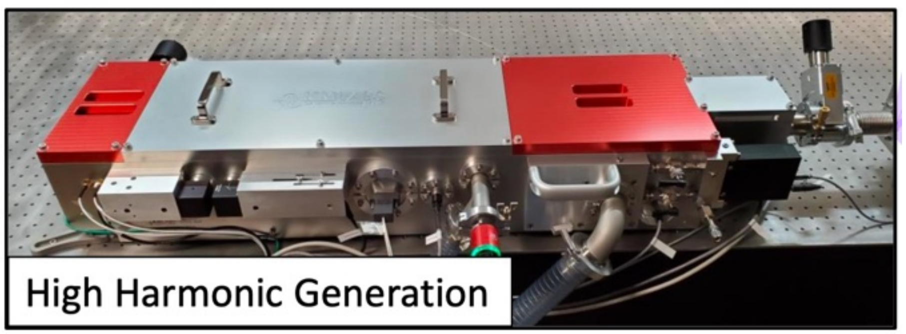
Solar technologies

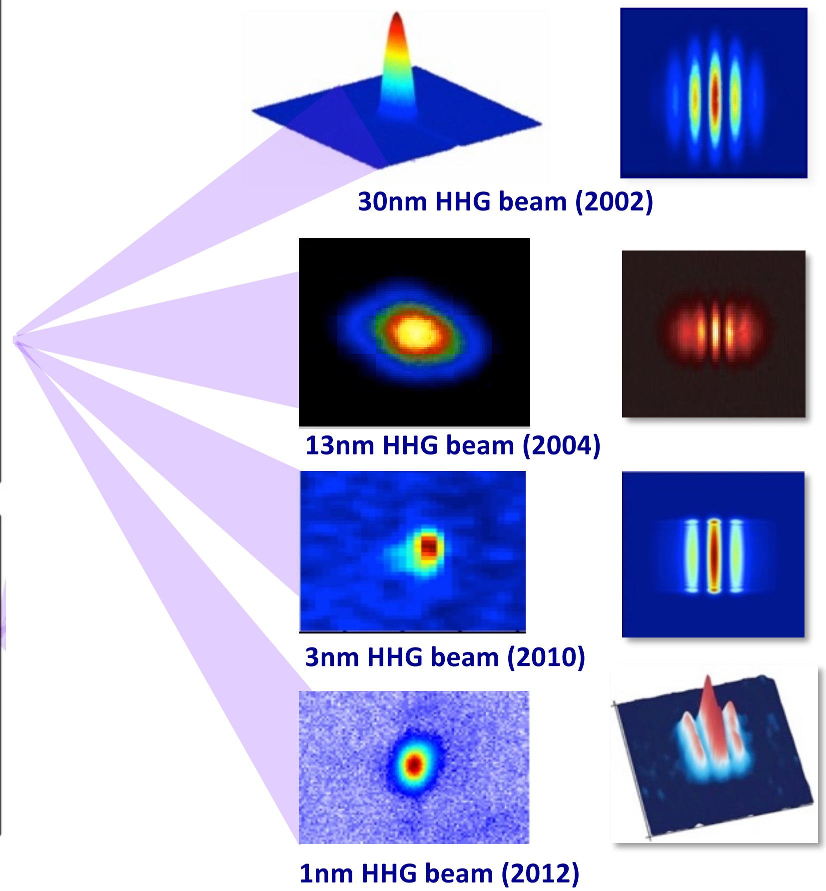
Bio

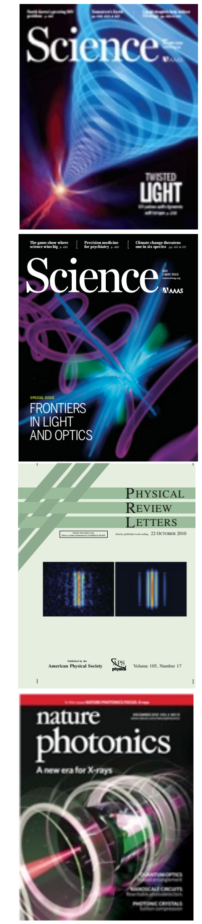


High Harmonic Generation: laser-like temporal & spatial coherence



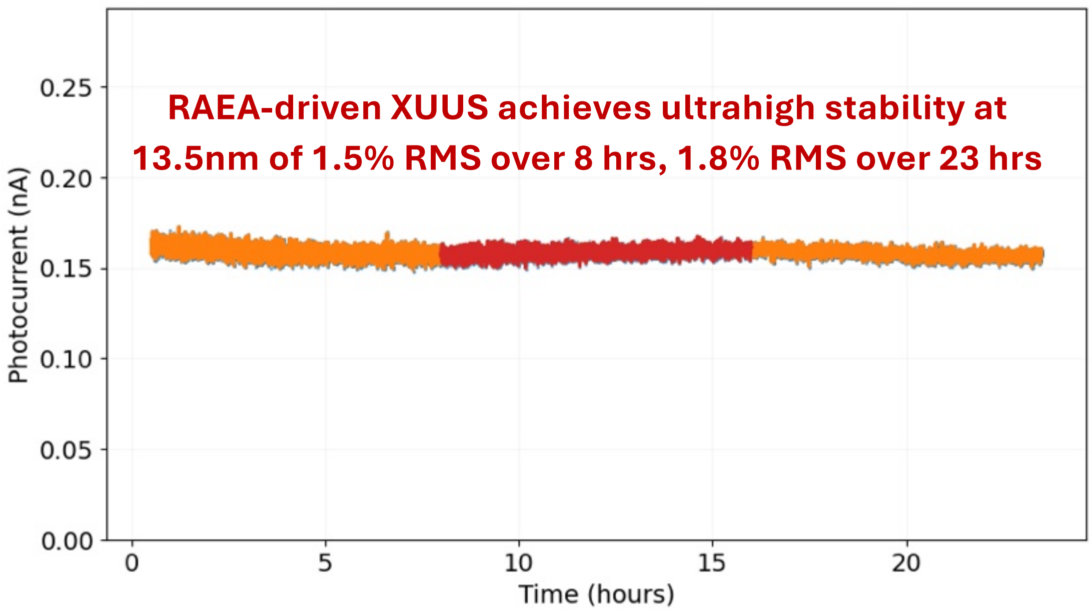






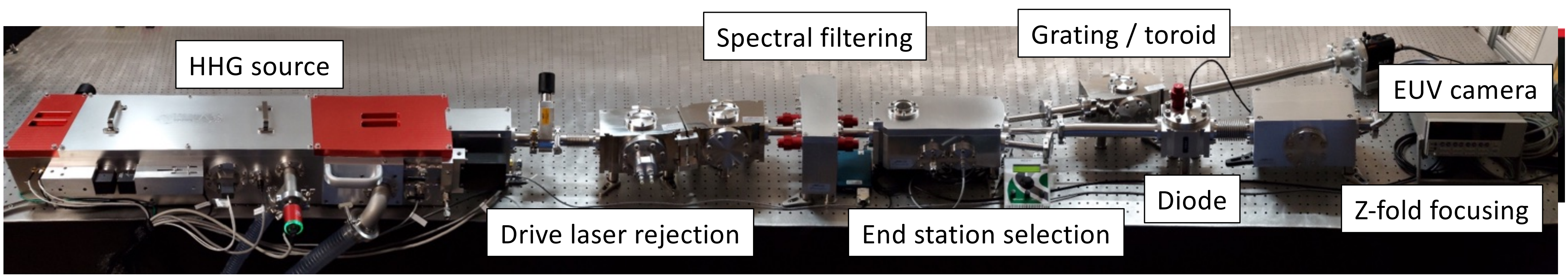
*Rundquist et al., Science **280**, 1412 (1998)*
*Bartels et al., Science **297**, 376 (2002)*
*Popmintchev et al., Science **336**, 1287 (2012)*

Putting EUV High Harmonic Generation to Work: Flux & Stability



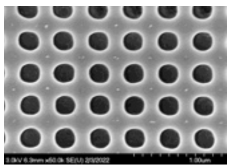
BRIGHT, ULTRASTABLE, CONTROLLED

- EUV intensity & phase
- Adjustable EUV beam shape and focus
- Accurate beam pointing
- Stable and adjustable spectrum
- Minimal nonlinear fluctuations
- High EUV to VUV flux: μW to tens of mW



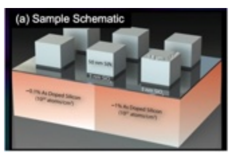
EUV metrologies: nondestructive, ultrasensitive, functional metrologies

INDUSTRY Collaborations



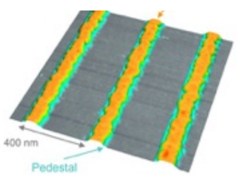
3M

Metamaterials
Thermal materials



imec KMLABS LEADING IN ULTRAFAST

ARPES, CDI, 2D

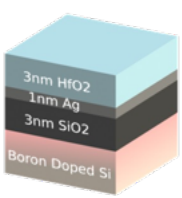


imec SAMSUNG

Nanoelectronics

intel

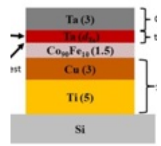
Thin films



Ball Aerospace THE UNIVERSITY OF ARIZONA

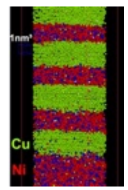
Detectors

NATIONAL LAB Collaborations



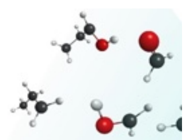
NIST

Spintronic



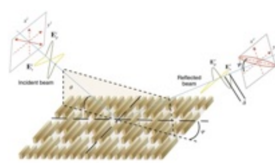
NIST

Atom Probe



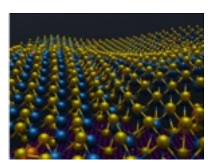
Argonne NATIONAL LABORATORY

Combustion



NIST

Nanoelectronics



GORDON AND BETTY MOORE FOUNDATION

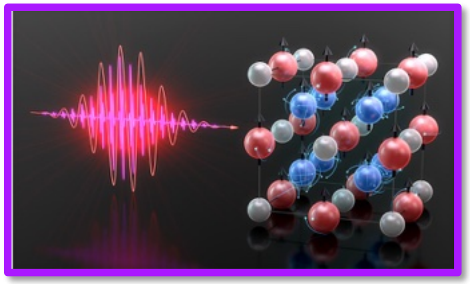
Quantum, Bio, Low dose

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 tabletop R&D

HHG metrologies

→ nondestructive, functional imaging at high resolution, element specificity



Element-resolved spin dynamics

Magnetic

Electronic

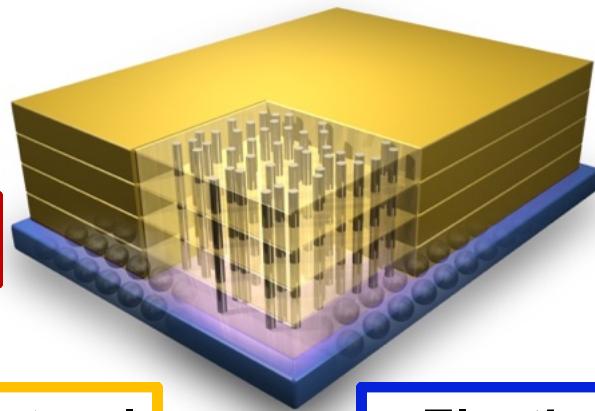
Adv. Energy Mater. 1402180 (2015)

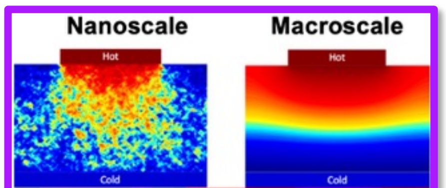
Cleaning Recipe	$E_F - E_V$ (eV)	Band Bending (meV)	Surface Band Structure
NH_4OH	0.6	180	Upward
$H_2O_2 + NH_4OH$	0.4	130	Upward
NH_4OH + 3A° Cu	0.2	100	Downward

Band bending, interfacial fields

Transport

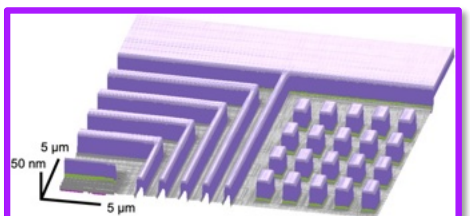
Interfacial





New understanding of nanoscale heat flow

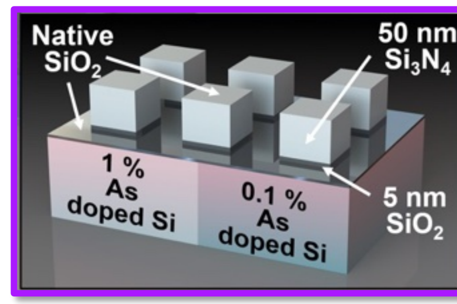
Structural



Sub-wavelength EUV imaging

Elastic

Ultrathin films



Compositional, space & depth-resolved imaging

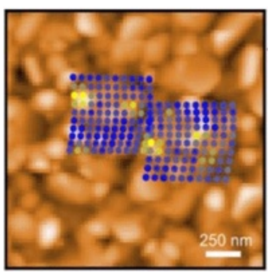
STROBE
BUILDING THE MICROSCOPES OF TOMORROW

Pushing Nanoscale Imaging Science to the Quantum Limits to address materials/nano/quantum/bio challenges

Hardware Toolbox

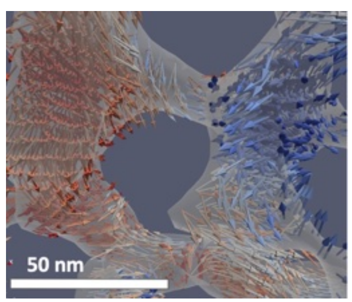
Computational Toolbox

New microscopes and low-dose methods



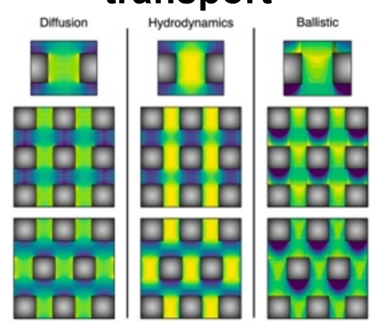
Science Advances (2024)
Optica (2023)

3D spin texture



Nature Nanotech (2023)

New predictive transport

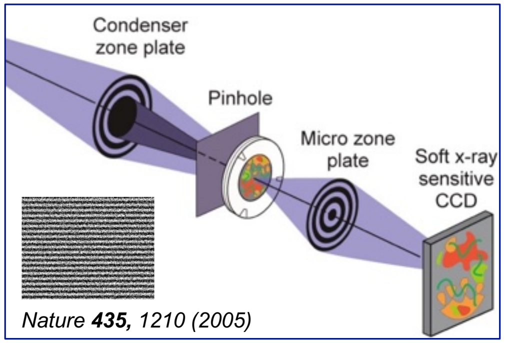


In press (2025)

Better vaccines

Zhou Lab, UCLA (2020-)

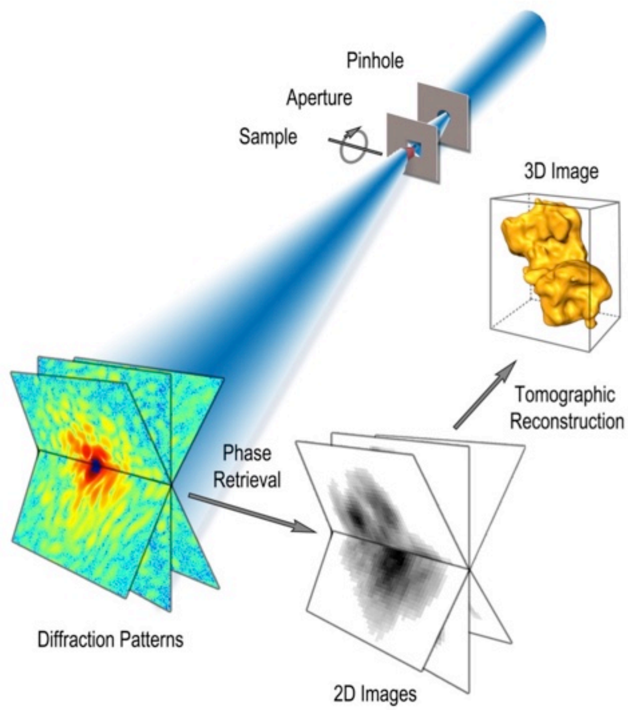
Coherent diffraction imaging – a revolution in short wavelength imaging



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

Traditional X-ray microscopes

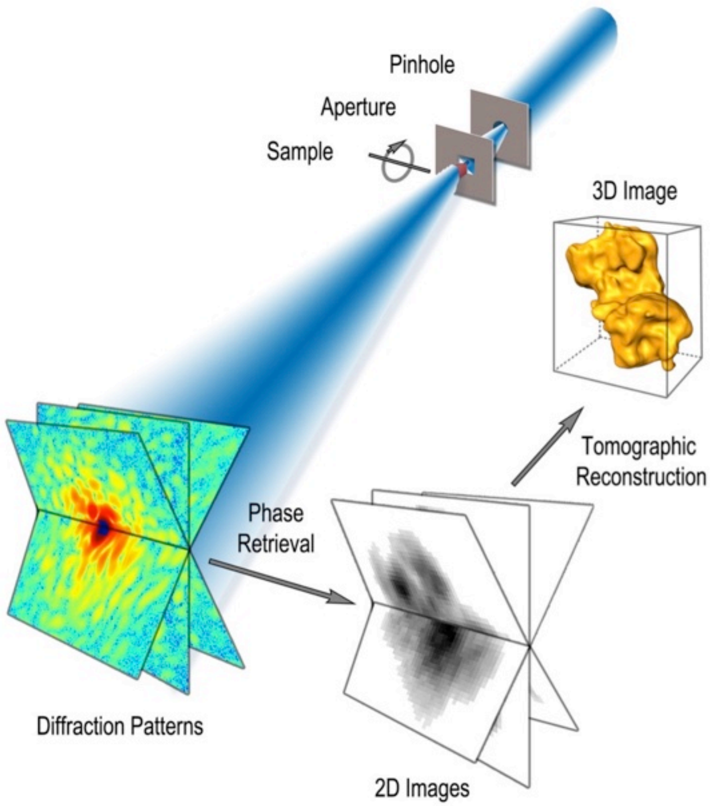
- Limited by lossy, imperfect, optics
- ~10x diffraction limited imaging



- **Diffraction-limited imaging ≈ λ/2NA**
- Inherent contrast of EUV and x-ray light
- Phase and amplitude image contrast
- Robust to vibrations
- MOST photon-efficient form of imaging!

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

Coherent lensless imaging – revolution in X-ray imaging



Acta Cryst. (1952). **5**, 843

Some implications of a theorem due to Shannon. By D. SAYRE, *Johnson Foundation for Medical Physics, University of Pennsylvania, Philadelphia 4, Pennsylvania, U.S.A.*

(*Received* 3 *July* 1952)

Shannon (1949), in the field of communication theory, has given the following theorem: If a function $d(x)$ is known to vanish outside the points $x = \pm a/2$, then its Fourier transform $F(X)$ is completely specified by the values which it assumes at the points $X = 0, \pm 1/a, \pm 2/a, \ldots$. In fact, the continuous $F(X)$ may be filled in merely by laying down the function $\sin \pi aX/\pi aX$ at each of the above points, with weight equal to the value of $F(X)$ at that point, and adding.

Now the electron-density function $d(x)$ describing a single unit cell of a crystal vanishes outside the points $x = \pm a/2$, where a is the length of the cell. The reciprocal-lattice points are at $X = 0, \pm 1/a, \pm 2/a, \ldots$, and hence the experimentally observable values of $F(X)$ would suffice, by the theorem, to determine $F(X)$ everywhere, if the phases were known. (In principle, the necessary points extend indefinitely in reciprocal space, but by using, say, Gaussian atoms both $d(x)$ and $F(X)$ can be effectively confined to the unit cell and the observable region, respectively.)

For centrosymmetrical structures, to be able to fill in the $|F|^2$ function would suffice to yield the structure, for sign changes could occur only at the points where $|F|^2$ vanishes. The structure corresponding to the $|F|^2$ function is the Patterson of a single unit cell. This has twice the width of the unit cell, and hence to fill in the $|F|^2$ function would require knowledge of $|F|^2$ at the half-integral, as well as the integral h's. This is equivalent to a statement made by Gay (1951).

I think the conclusions which may be stated at this point are:

1. Direct structure determination, for centrosymmetric structures, could be accomplished as well by finding the sizes of the $|F|^2$ at half-integral h as by the usual procedure of finding the signs of the F's at integral h.

2. In work like that of Boyes-Watson, Davidson & Perutz (1947) on haemoglobin, where $|F|^2$ was observed at non-integral h, it would suffice to have only the values at half-integral h.

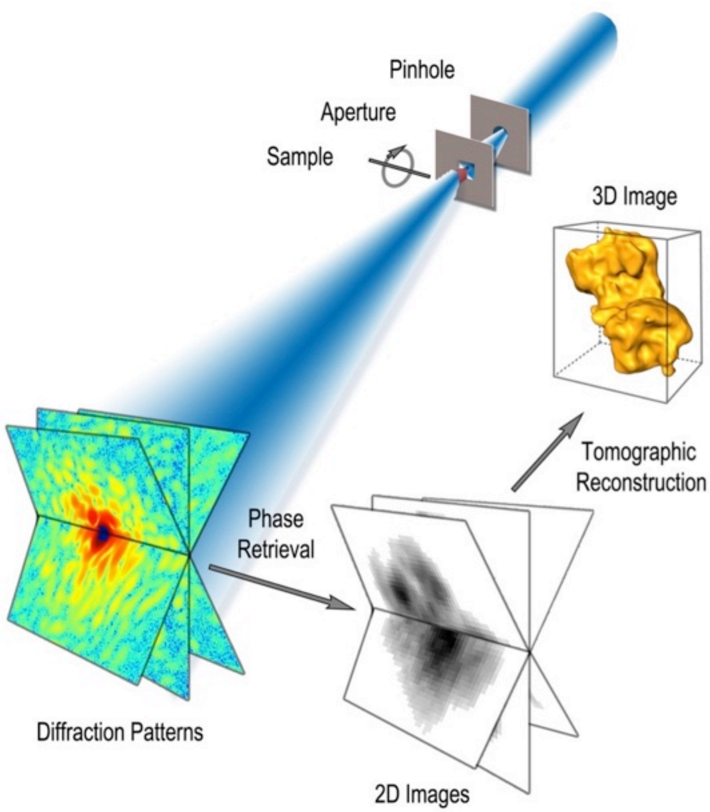
The extension to three dimensions is obvious.

References

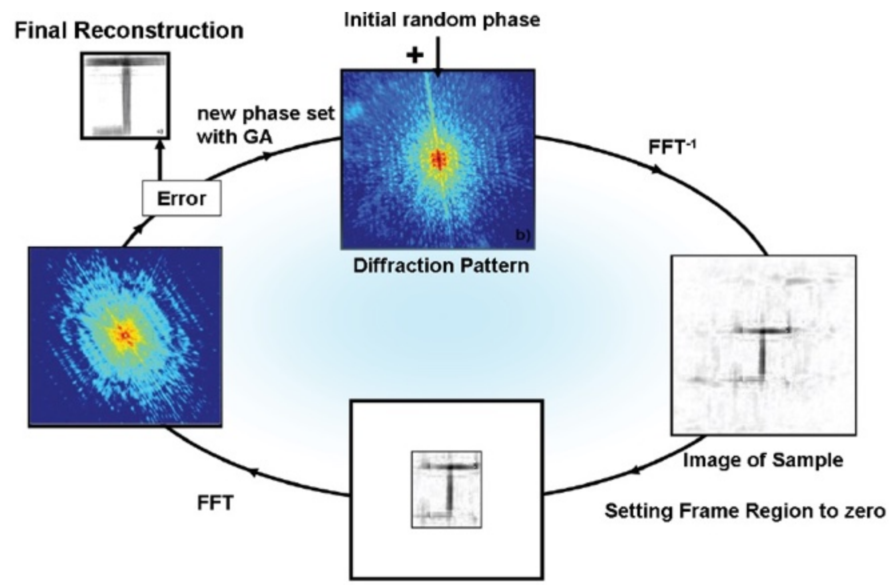
BOYES-WATSON, J., DAVIDSON, E. & PERUTZ, M. F. (1947). *Proc. Roy. Soc.* A, **191**, 83.
GAY, R. (1951). Paper presented at the Second International Congress of Crystallography, Stockholm.
SHANNON, C. E. (1949). *Proc. Inst. Radio Engrs., N.Y.* **37**, 10.

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

Coherent lensless imaging – revolution in X-ray imaging



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

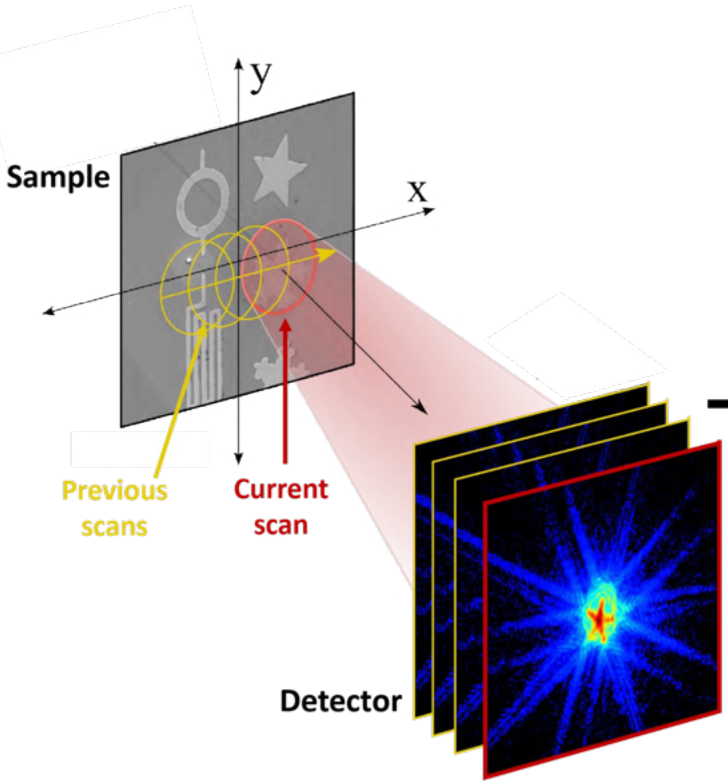


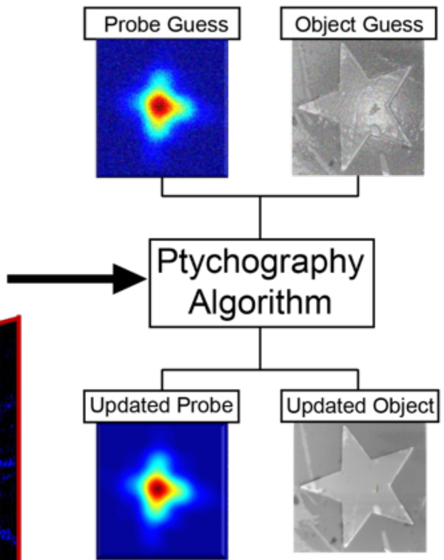
*Sandberg et al., PRL **99**, 098103 (2007)*

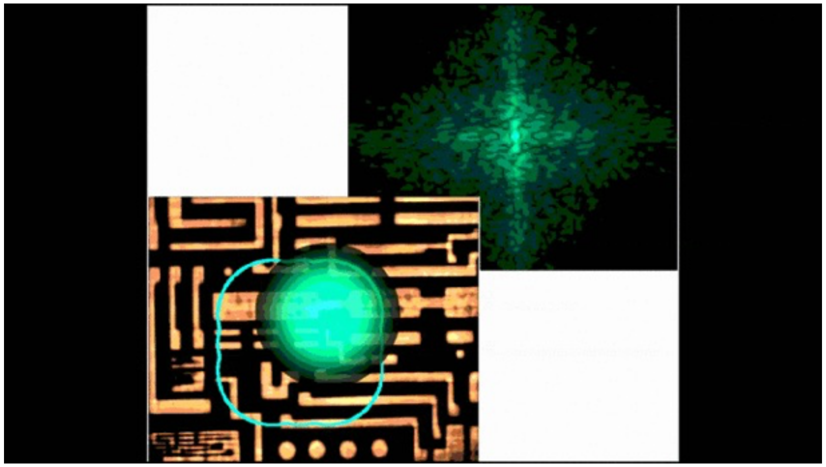
- **Diffraction-limited imaging ≈ λ/2NA**
- Inherent contrast of EUV and x-ray light
- Phase and amplitude image contrast
- Robust to vibrations
- MOST photon-efficient form of imaging!

CDI *Ptychography*

- *Scan* beam over sample
  - Redundant data from overlapping illumination areas ensures accurate phase retrieval and imaging

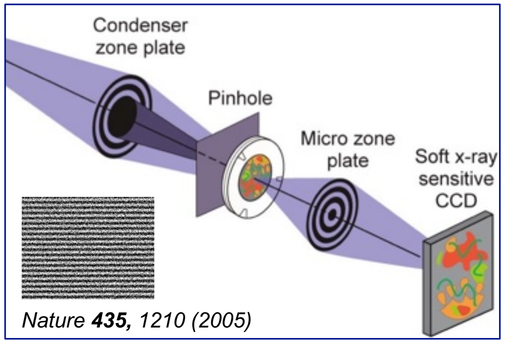






*Rodenburg et al., PRL **98**, 034801 (2007)*
*Thibault et al., Science **321**, 379 (2008)*
*Maiden et al., Ultramicroscopy **109**, 1256 (2009)*

Coherent diffraction imaging – a revolution in short wavelength imaging



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

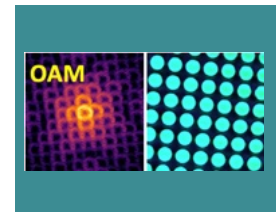
Traditional X-ray microscopes
- Limited by lossy, imperfect, optics
- ~10x diffraction limited imaging

Fundamentally new imaging

2017: 1st sub-λ EUV imaging

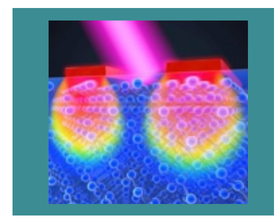
Nat. Photon. **11**, 22 (2017)

2023: Periodic samples



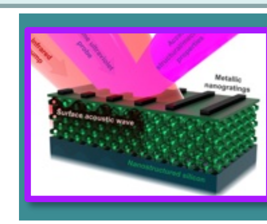
Optica ***10****, 1245 (2023)*

2021: Nano-hotspot



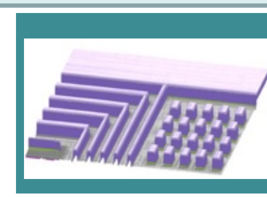
PNAS ***118****, 2109056118 (2021)*

2023: General transport



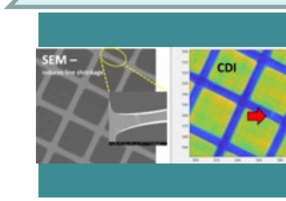
Nano Lett. ***23****, 2129 (2023)*

2021: Nondestructive Maps



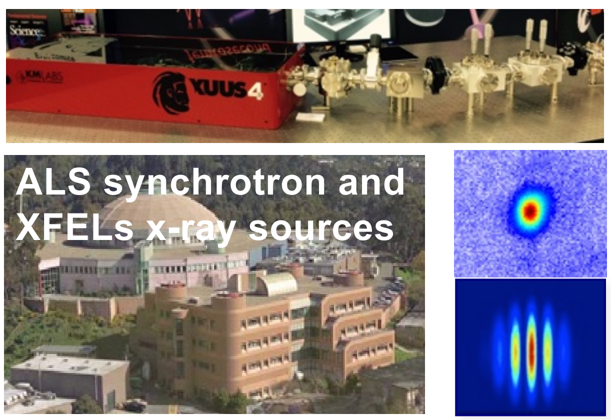
Science Adv. ***7****, 9667 (2021)*

2023: Low-dose OAM

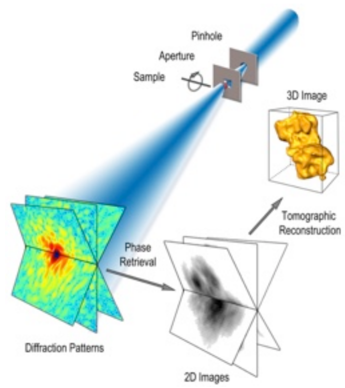


Optica ***10****, 1245 (2023)*

Coherent EUV/SXR sources



Coherent imaging

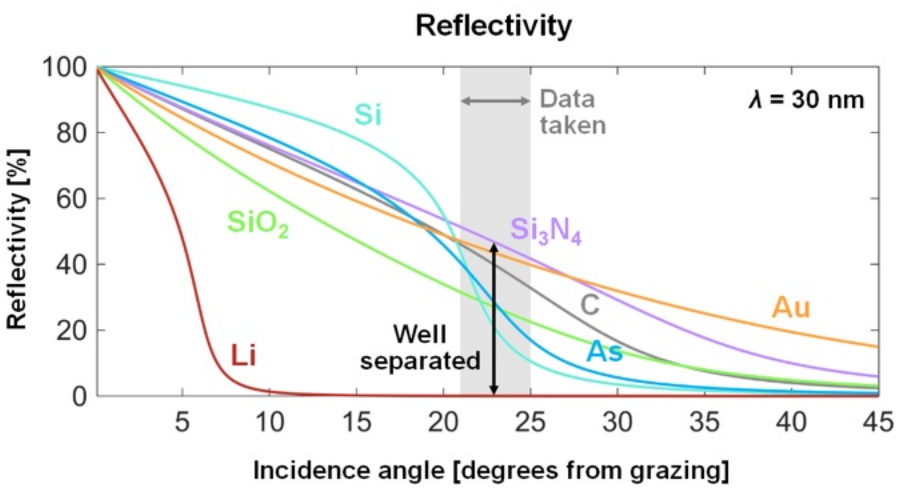


Miao et al., Nature ***400****, 342 (1999)*

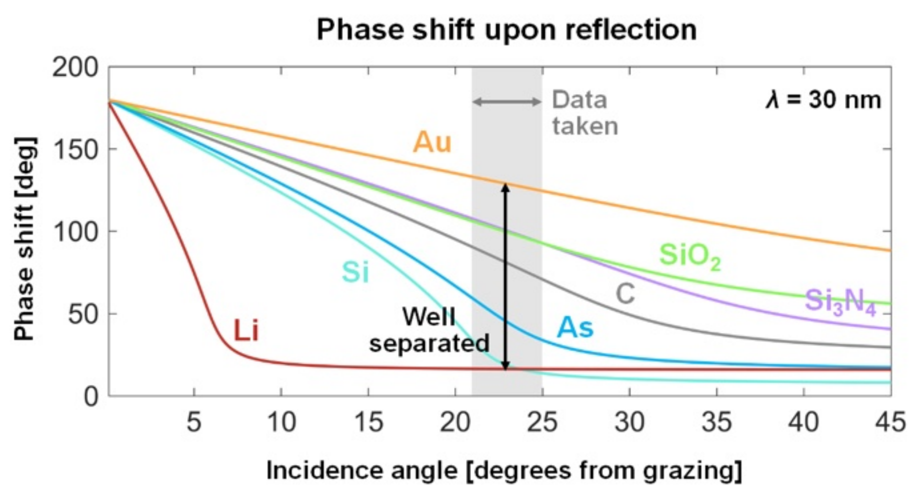
Ptychography in reflection in the EUV

- Most materials 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

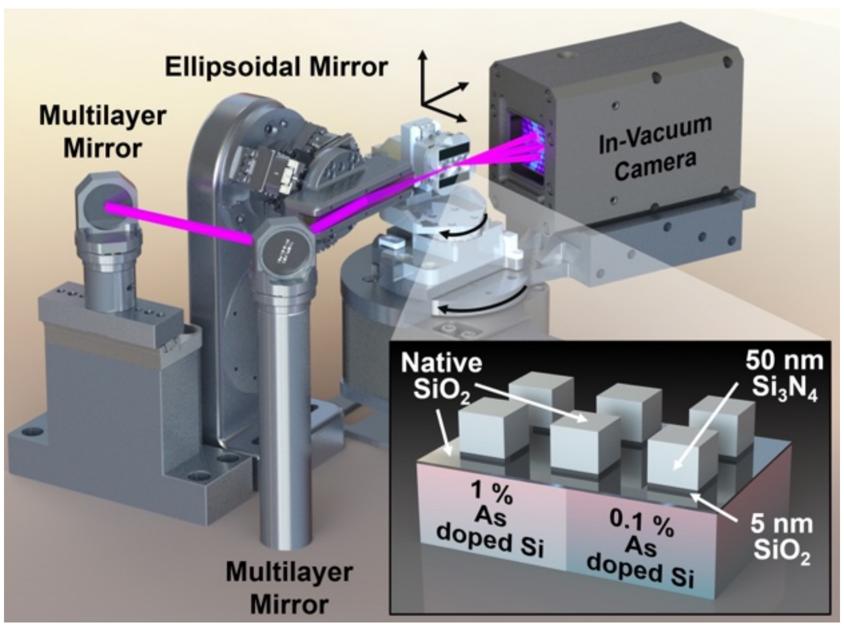


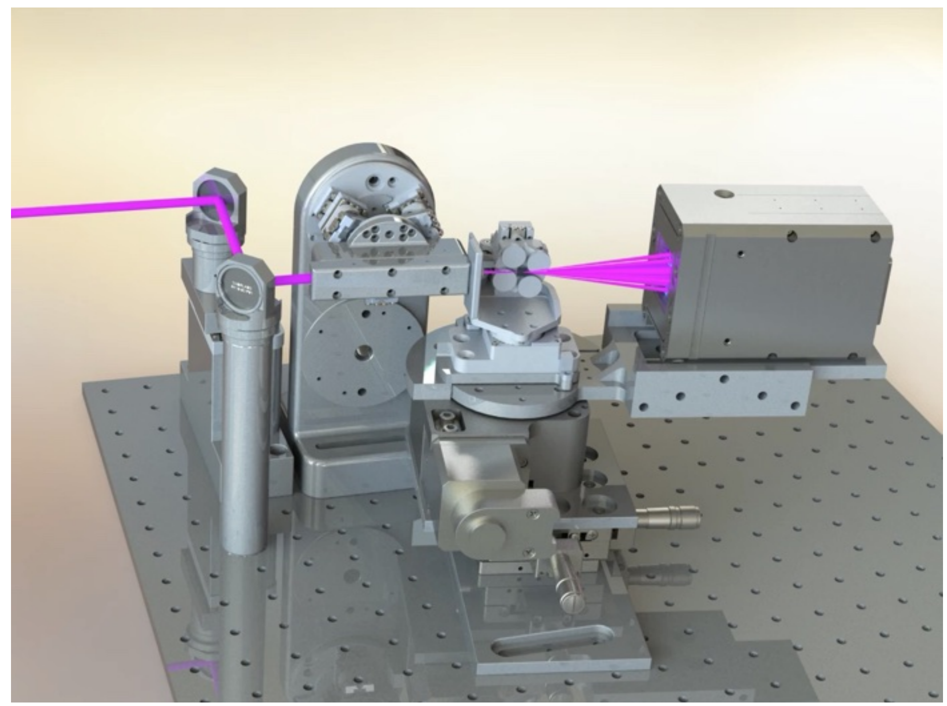
Tanksalvala et al., Sci. Advances ***7****, 9667 (2021)*
Esashi et al., RSI ***94****, 123705 (2023)*

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

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

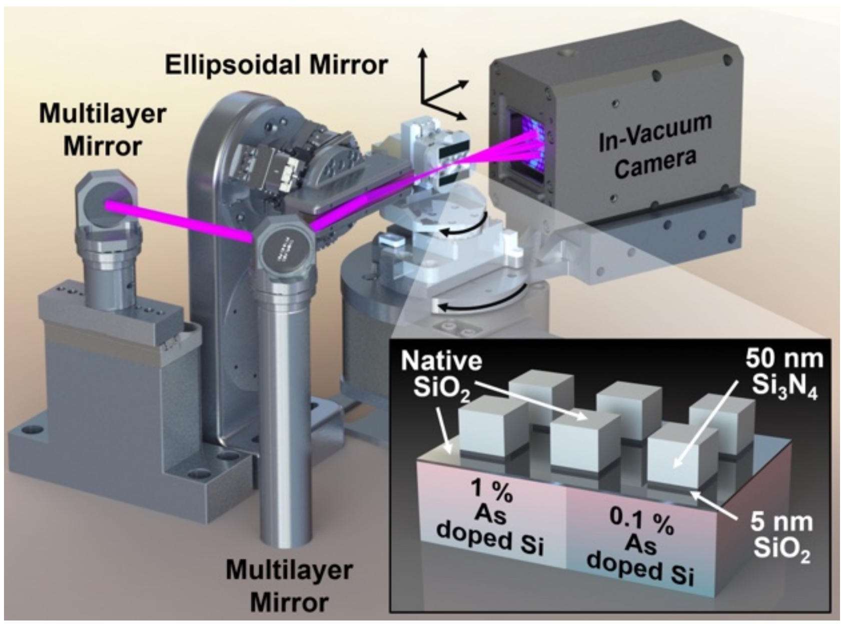




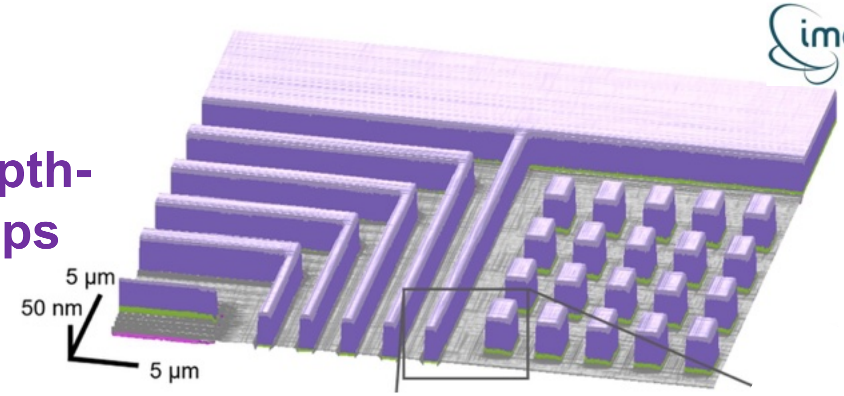
Phase-sensitive nondestructive EUV Imaging reflectometer

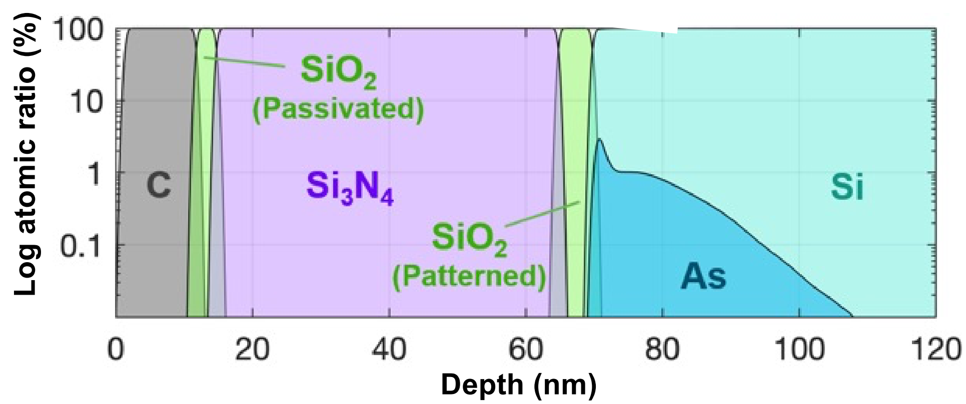
- Unique non-destructive spatial and depth-resolved compositional mapping
- Extract interfaces, layers, dopant profiles, composition of challenging semiconductor test samples

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

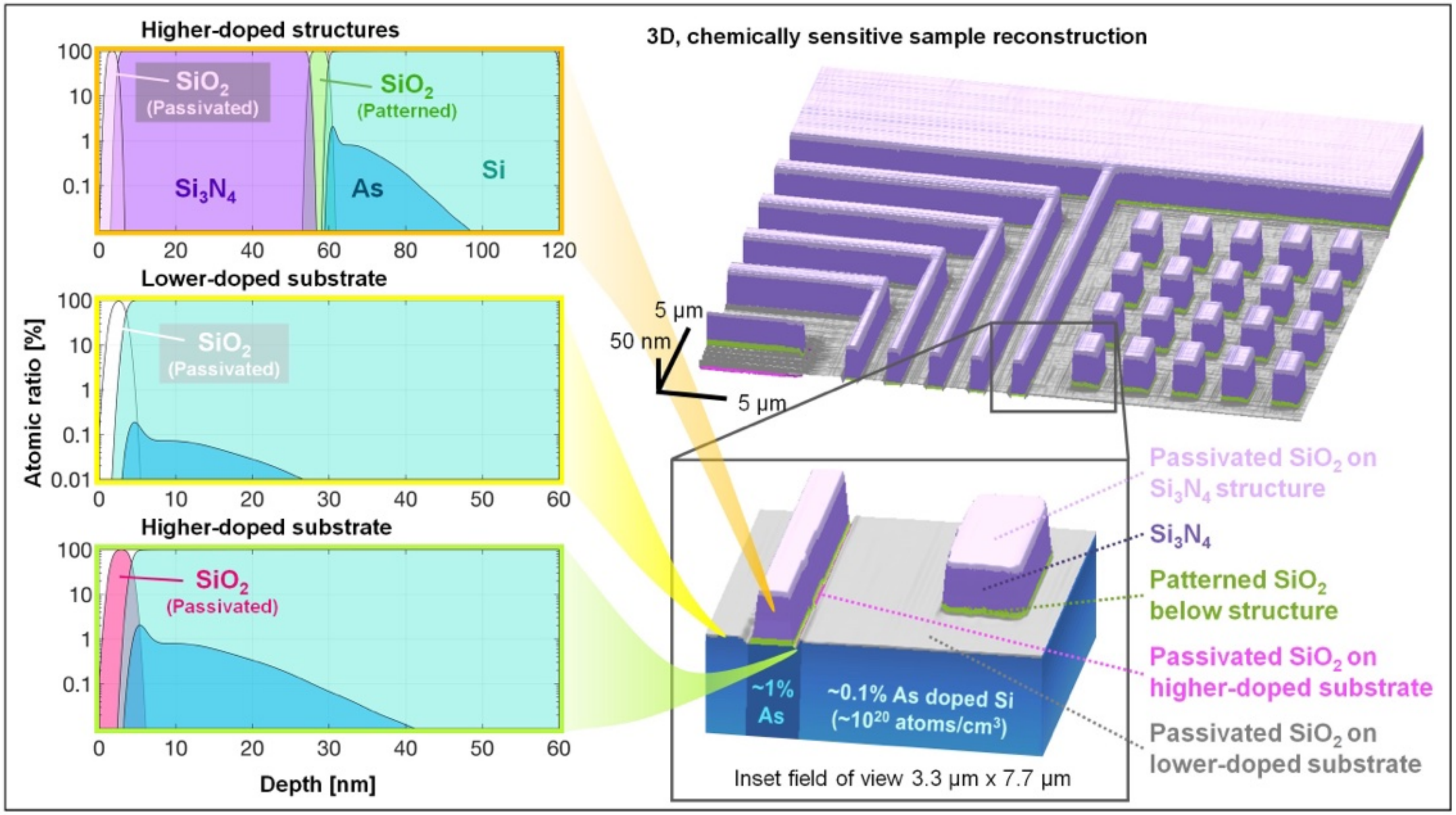


Spatial & depth-resolved maps



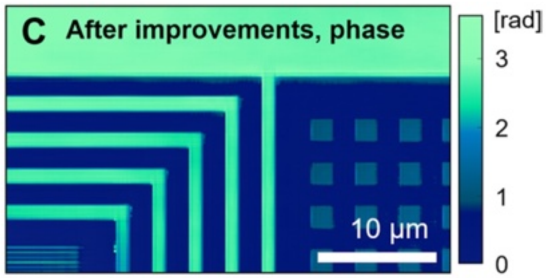


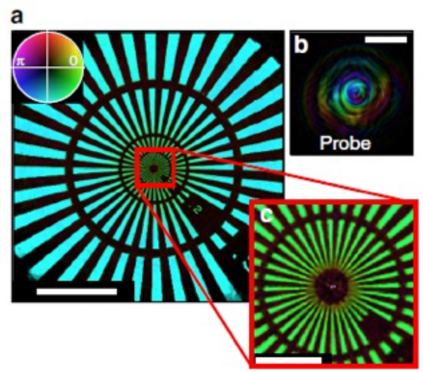
Non-destructive Dopant Profiling

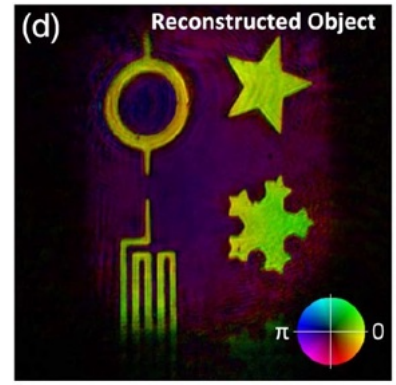


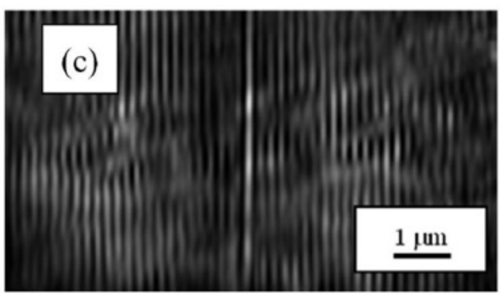
Tanksalvala et al., Sci. Advances ***7****, 9667 (2021)*

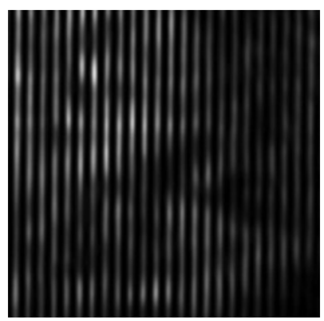
Friends and Enemies of Ptychographic Reconstruction

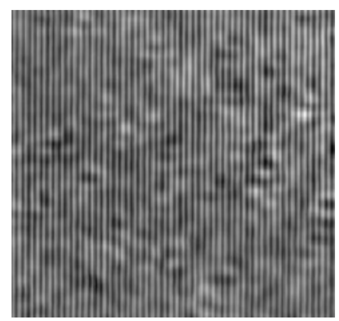


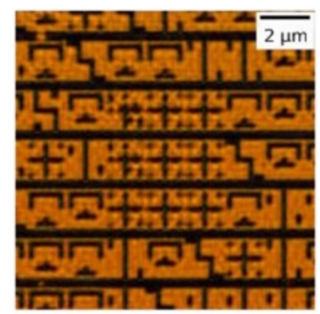




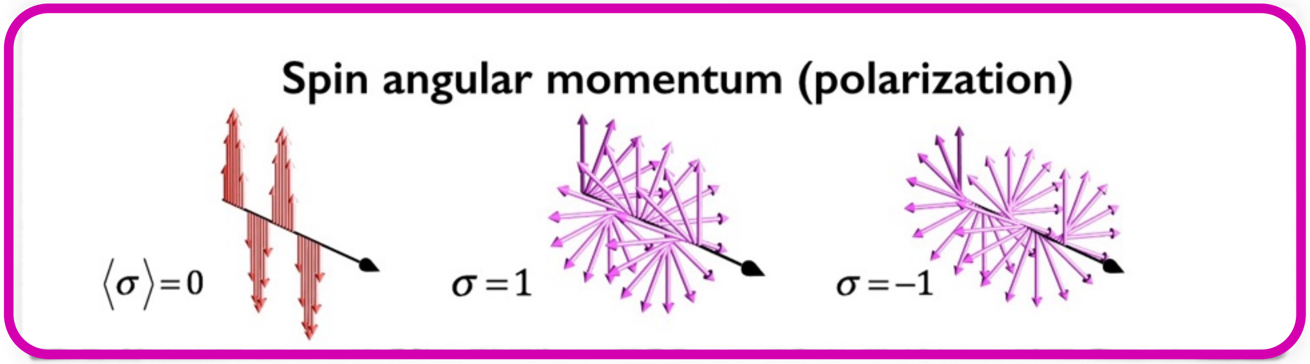


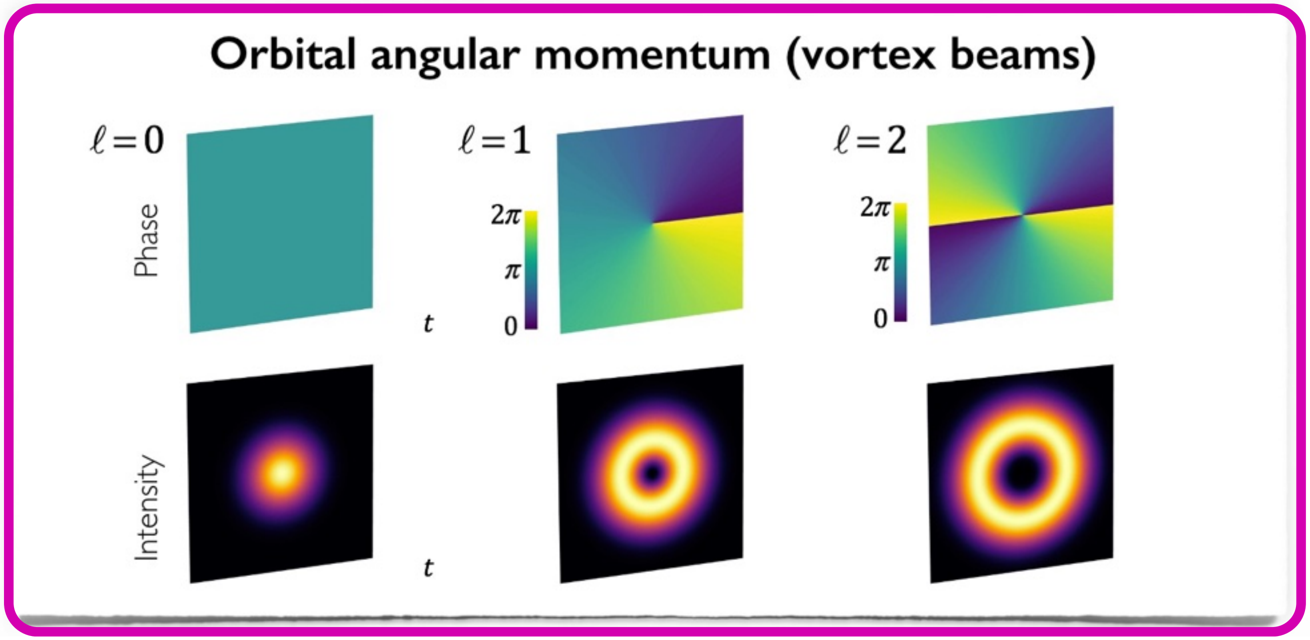


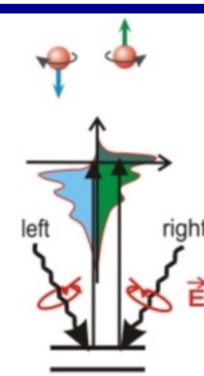




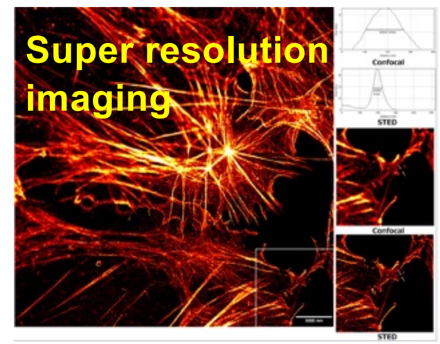
Spin and orbital angular momentum of light





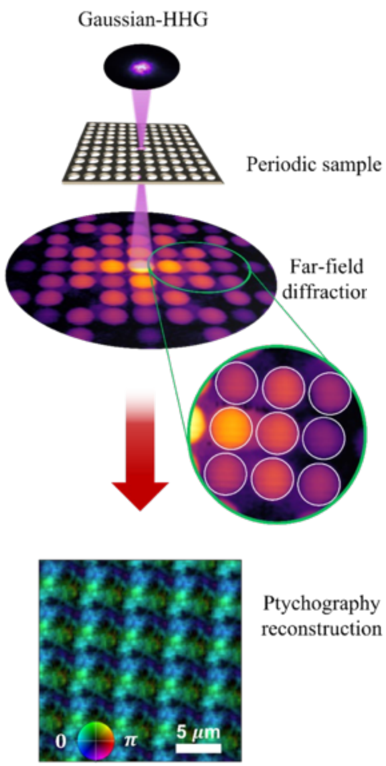


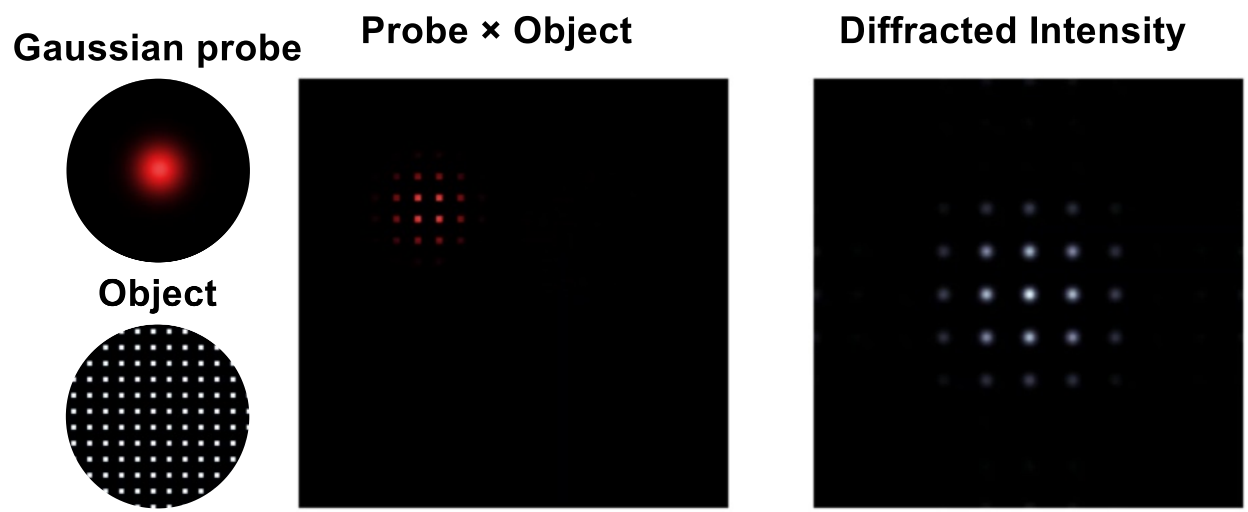
X-ray magnetic circular dichroism



High fidelity low-dose CDI of near-periodic structures

- Highly-periodic samples are challenging

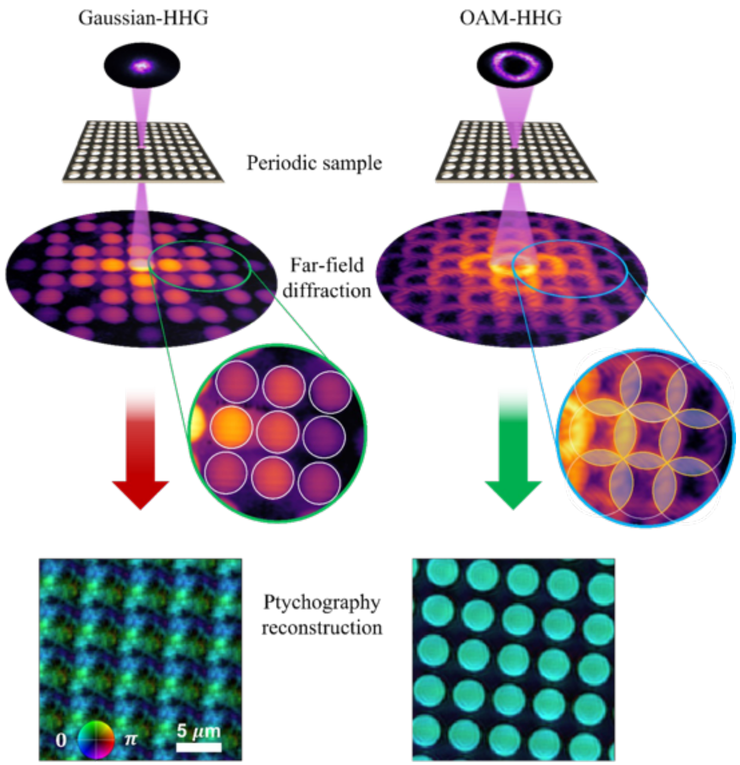


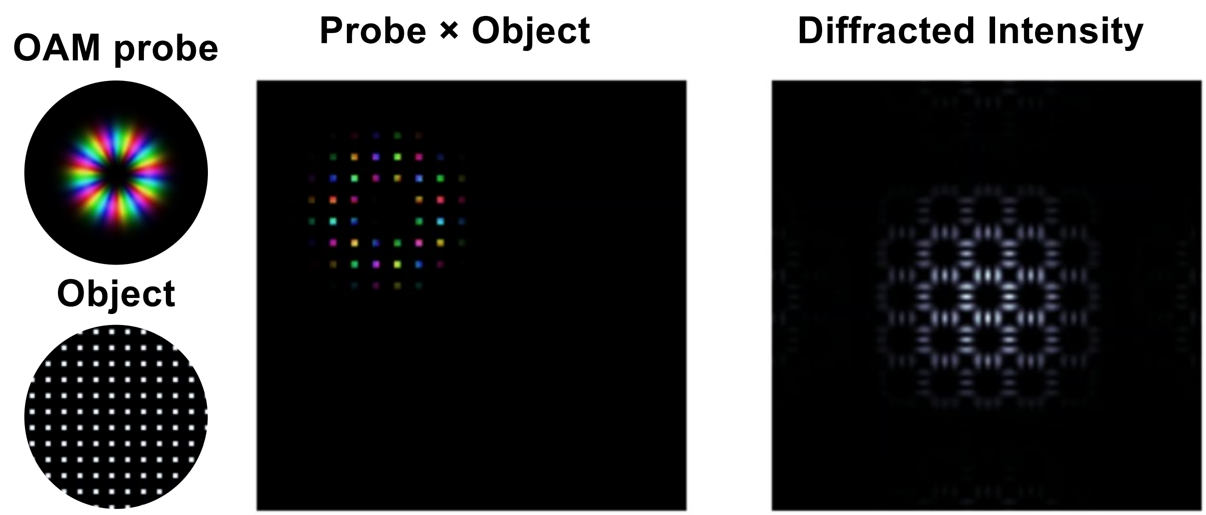


*Wang et al., Optica **10**, 1245 (2023); Wang et al., Optics in 2023*

High fidelity low-dose CDI of near-periodic structures

- Highly-periodic samples are challenging
- Solution: ***engineered*** illumination

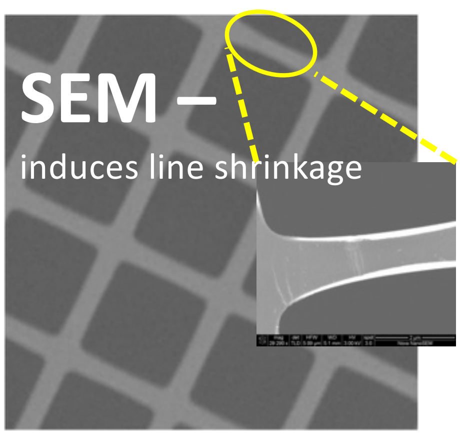


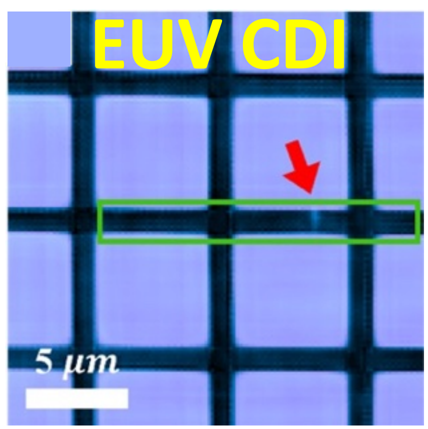


*Wang et al., Optica **10**, 1245 (2023); Wang et al., Optics in 2023*

High fidelity low-dose CDI of near-periodic structures

- Highly-periodic samples are challenging
- Solution: ***engineered*** illumination
- Non-destructive and low dose
- Applications in defect inspection



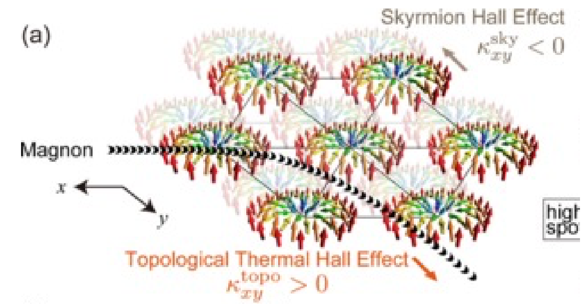


Drs. Bin Wang & Nathan Brooks

Wang et al., Optica ***10****, 1245 (2023); Wang et al., Optics in 2023*
Ph.D. Theses of Bin Wang, Nathan Brooks

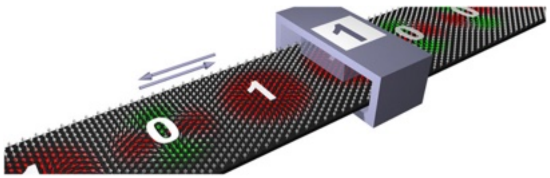
Magnetic Skyrmions (3D topological spin textures)

Hall effect in Skyrmion Lattice



Akazawa et al., Phys. Rev. Research 4, 043085 - 2022

Memory Device

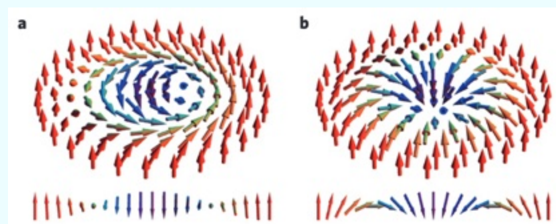


Hoffmann et al., *Front. Phys.* 9:769873 (2021)

Magnetic Skyrmion

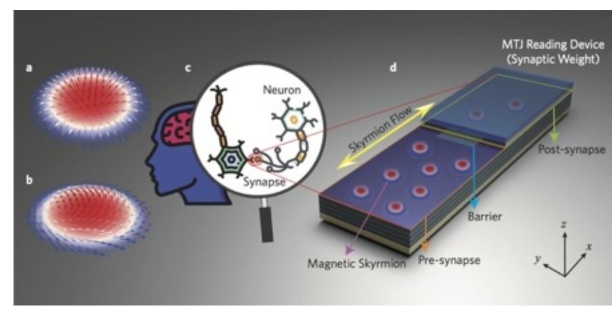
- Quasiparticle of 2D magnetic spin structure
- Topologically protected, robust to noise

Bloch-type Néel-type



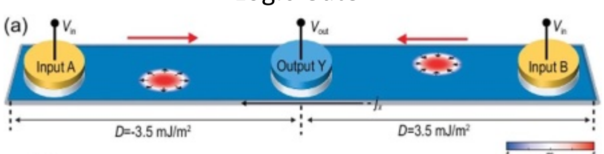
Kézsmárki et al., Nature Mater 14, 1116–1122 (2015)

Neuromorphic Computing



Chen et al., Phys. Rev. Applied 14, 014096 - 2020

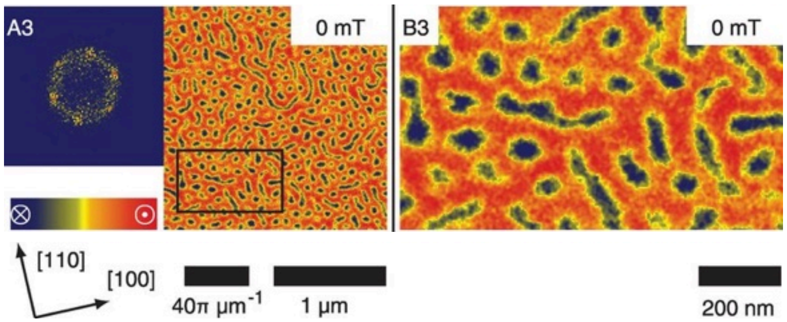
Logic Gate



Yu et al., National Science Review, Volume 9, Issue 12, 2022, nwac021

Current Approaches for Imaging Skyrmions

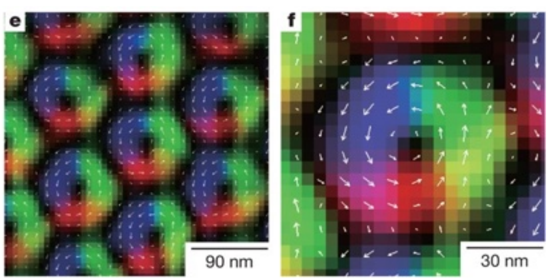
Magnetic Force Microscope



P. Milde et al., Science340,1076-1080 (2013)

- 2D (surface only)
- Magnetic tip
- Pixel size ~ 20nm

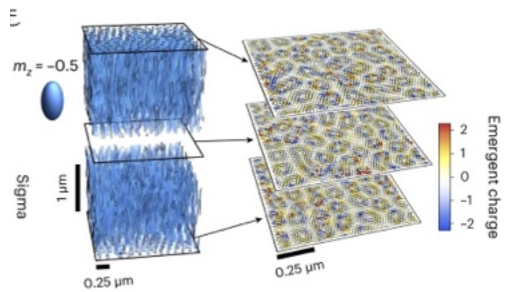
Lorentz Transmission Electron Microscope



Yu et al., Nature 465, 901–904 (2010)

- 2D (bulk projection)
- Electron beam
- Pixel size ~ 5nm

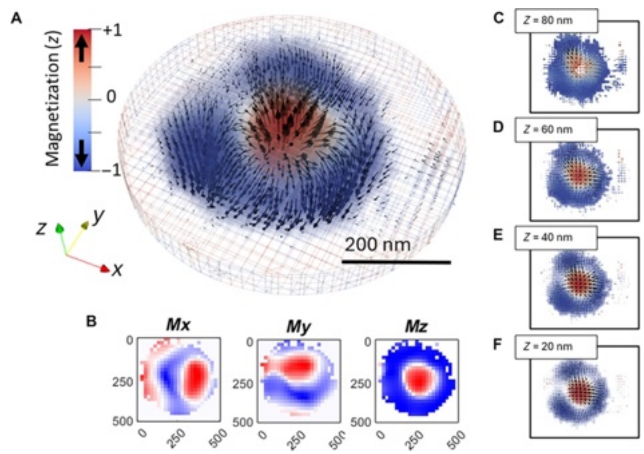
Small Angle Neutron Scattering Tomography



Henderson et al., Nat. Phys. 19, 1617–1623 (2023)

- 3D
- Neutron beam
- Voxel size ~ 15nm

X-Ray Laminography (through STXM) + Tomography



Raftrey et al., Sci. Adv.10, eadp8615 (2024)

- 3D
- Soft X-Ray
- Voxel size ~ 20nm

Start with imaging topological magnetic monopoles

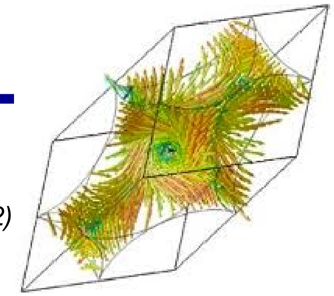
- Topological magnetic monopoles (TMM), also known as hedgehogs or Bloch points, are 3D nonlocal spin textures that are robust to thermal and quantum fluctuations due to their topology

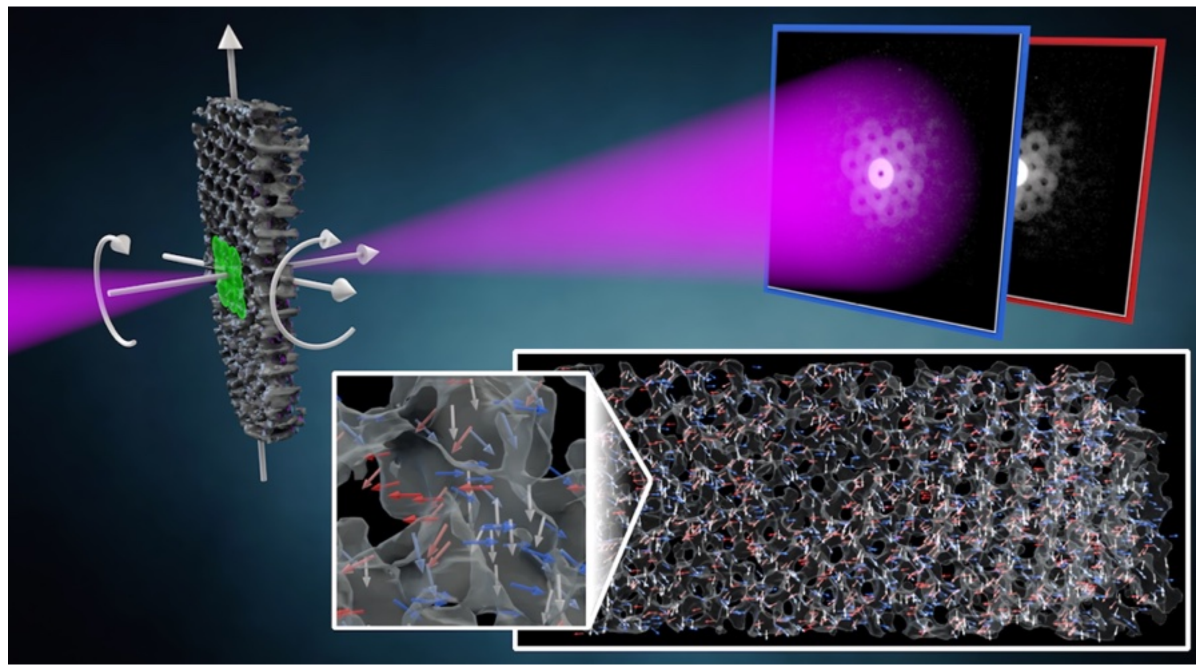
- Predicted by Han and Crespi in 2002

- We used a new 3D soft x-ray vector ptychography to map TMM and anti-TMM pairs and interactions for the first time. The spatial resolution achieved (~10 nm) is comparable to the magnetic exchange length of transition metals.

- How does it work?

Han, Crespi, PRL ***89****, 197203 (2002)*

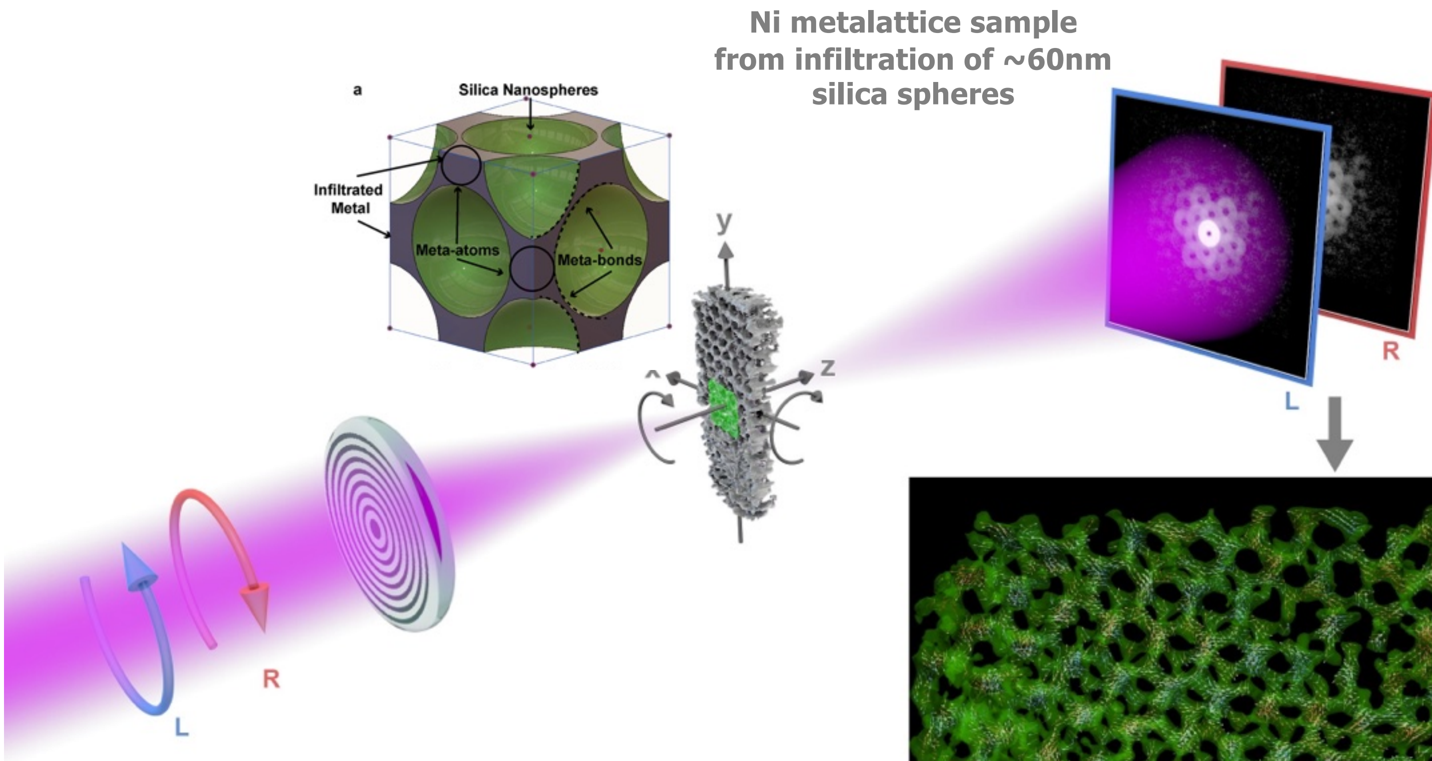




Direct observation of 3D topological spin textures and their interactions using soft x-ray vector ptychography, Rana et al., Nature Nanotech. **18**, 227 (2023). 10.1038/s41565-022-01311-0

University of Colorado Boulder JILA UCLA STROBE NSF SCIENCE AND TECHNOLOGY CENTER PennState

3D static imaging of spin texture in metalattices



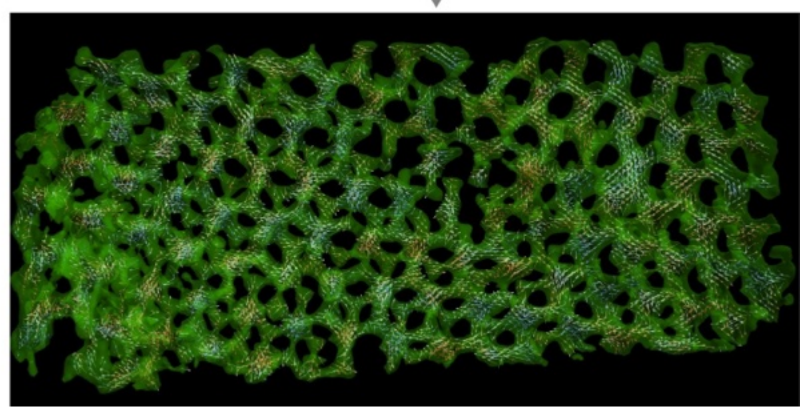
Coherent X-ray beam (COSMIC)

3D vector CDI

- 192 Images @ each of 3 in-plane rotations and @ each of 33 tilt angles, for LCP & RCP light **(796,485 diff. patterns)**
- **Several TBs of data!!!**

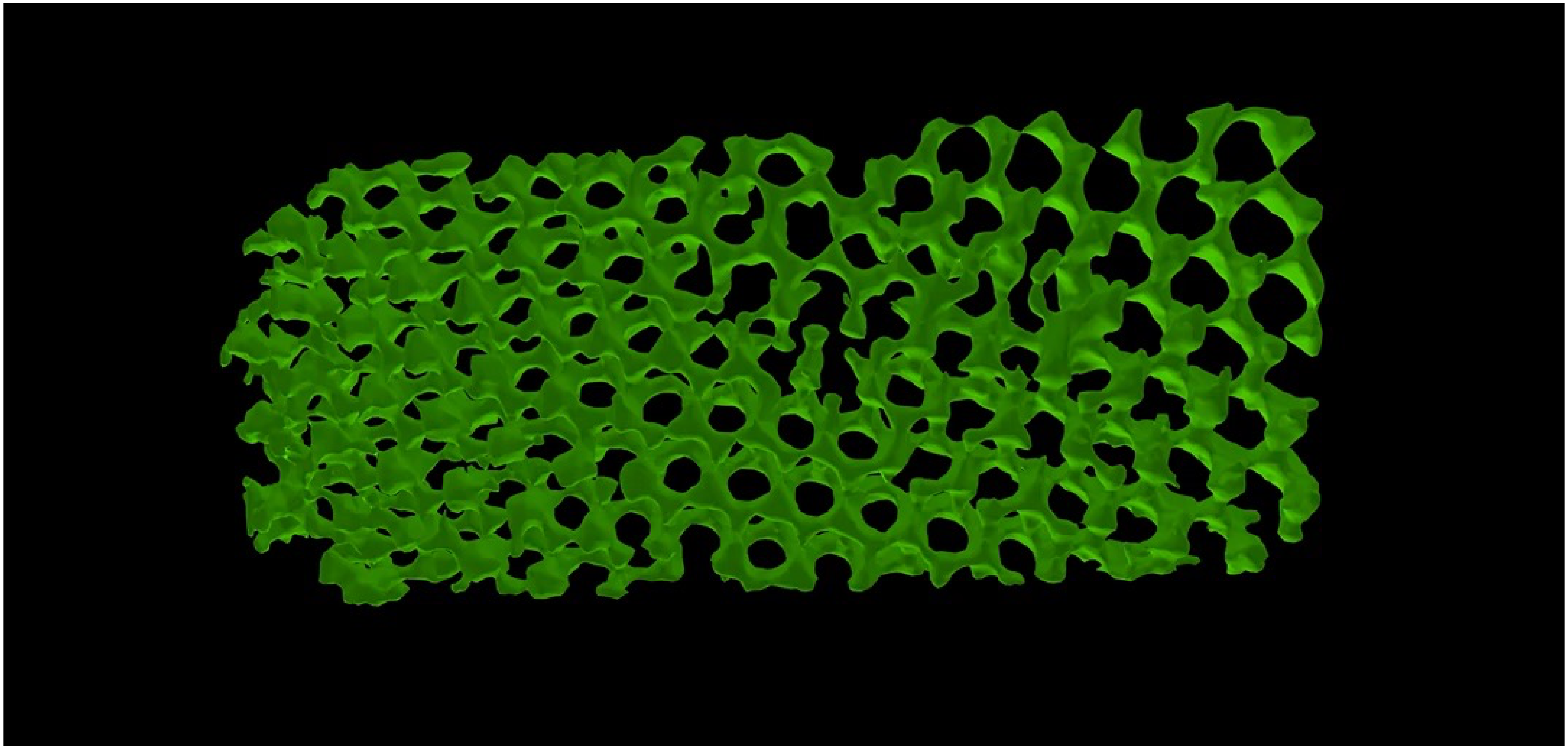
Ideal metalattice sample

- Topological magnetic monopoles
- Electronic/structural & magnetic signals
- Badding group synthesis



Green – full density reconstruction
Blue-red – z component magnetization reconstruction

Rana et al., Nature Nanotechnology ***18****, 227 (2023)*



Hedgehog pairs in bulk

Red points: Q = +1 hedgehogs in bulk

Blue points: Q = −1 anti-hedgehogs in bulk

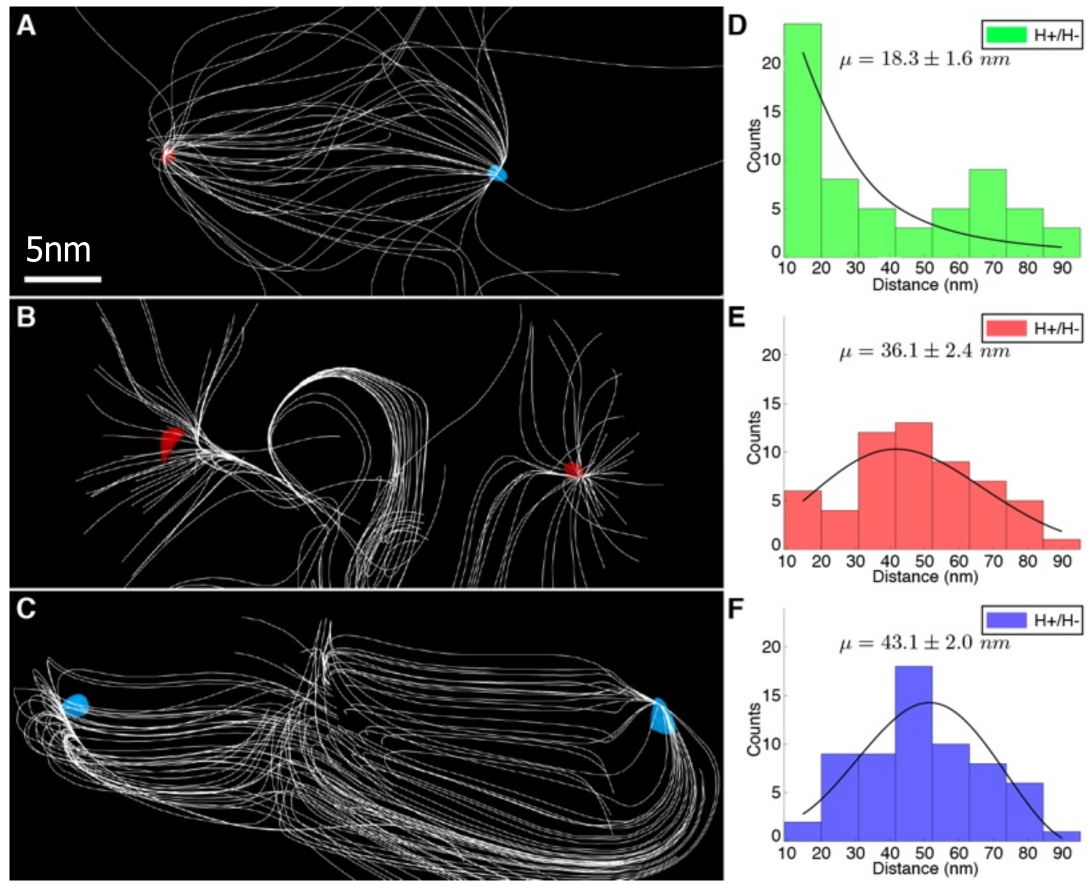
Hedgehog pairs with effective field

Nearest-neighbor distance statistics

Hedgehog Anti-Hedgehog Pair

Hedgehog Pair

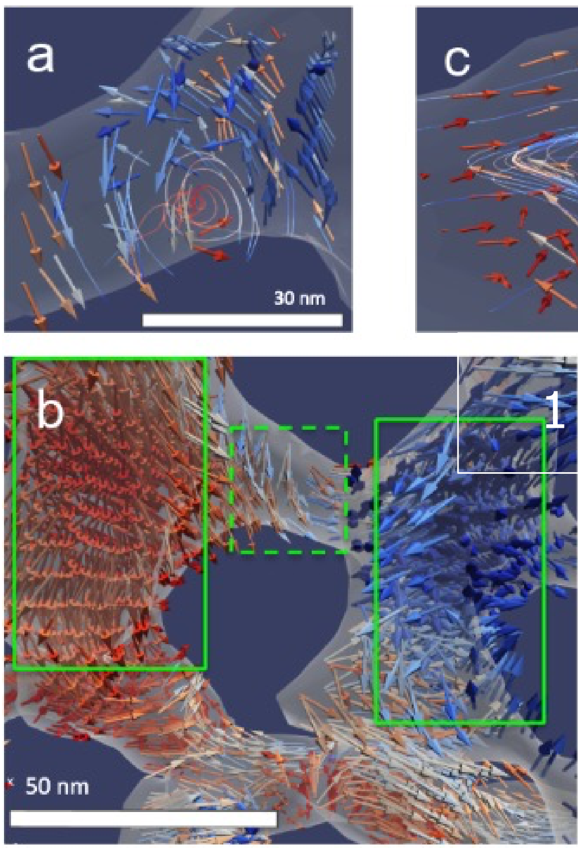
Anti-Hedgehog Pair

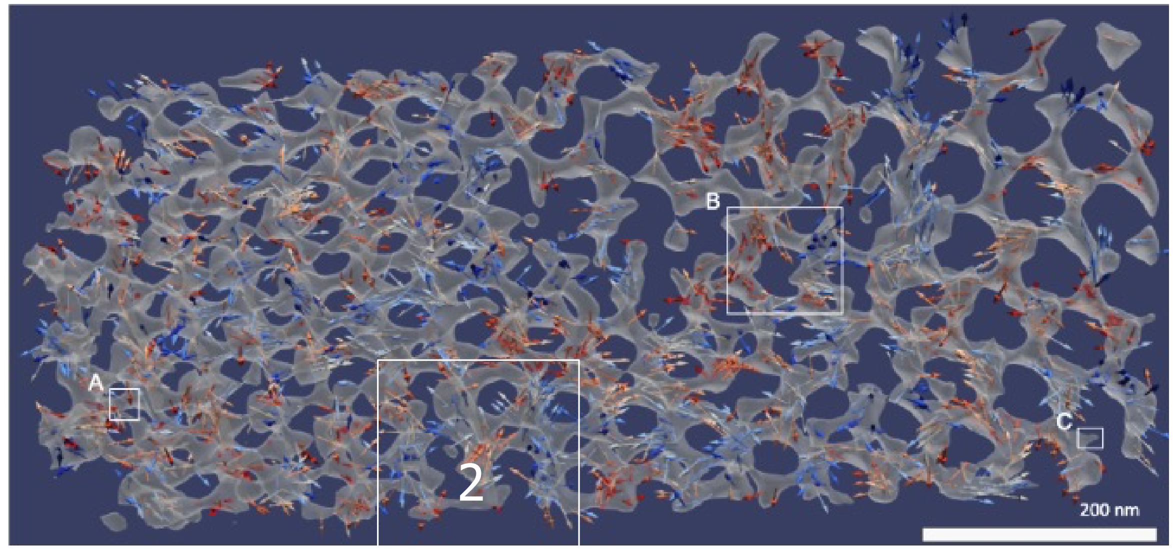


Rana et al., Nature Nanotechnology ***18****, 227 (2023)*

Capture magnetization vector & emergent magnetic field of TMMs and anti-TMMs

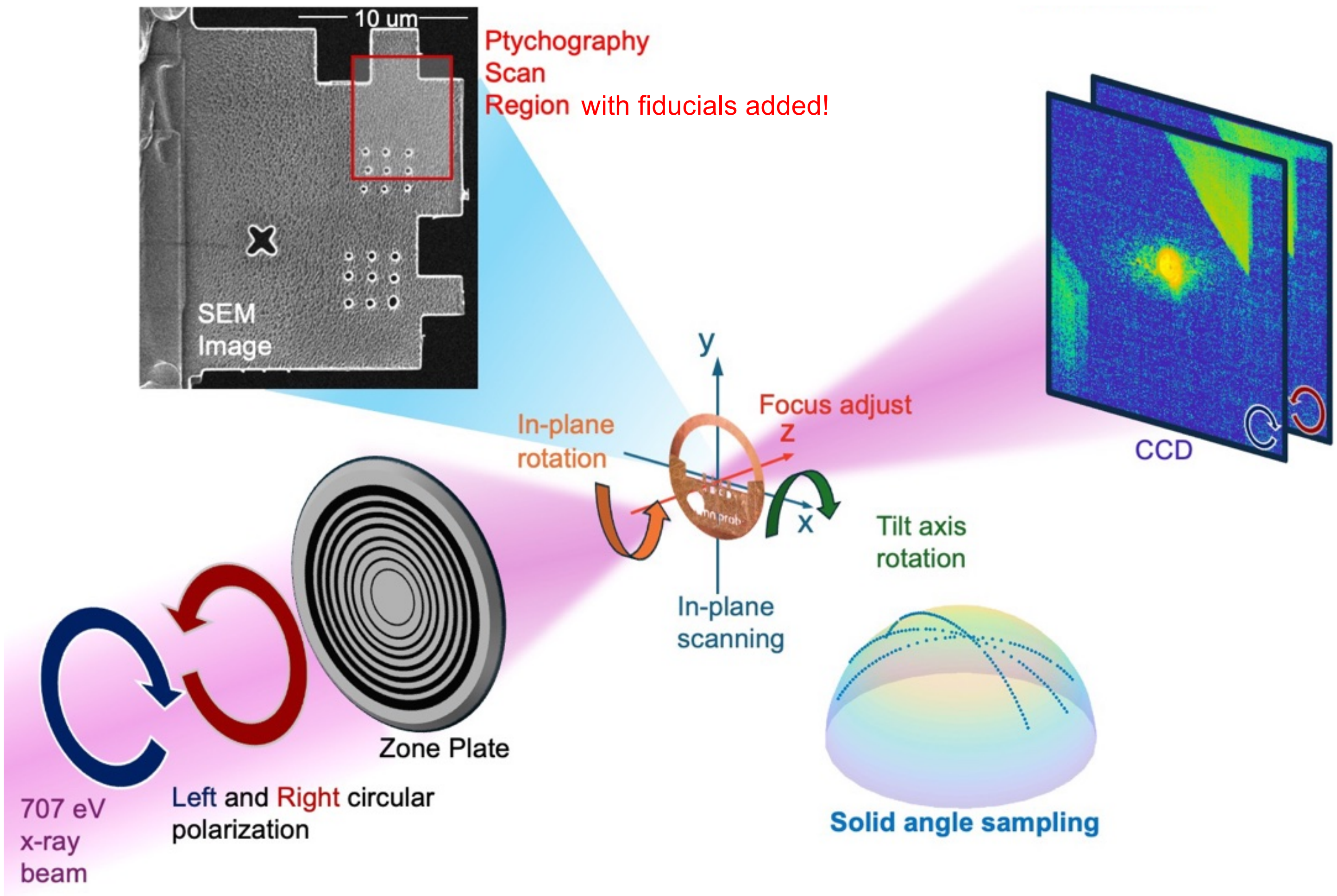
Rana et al., Nature Nanotechnology ***18****, 227 (2023)*





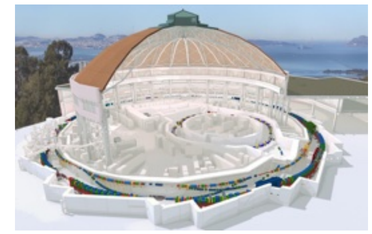
- **Lorentz TEM** can image the scalar 2D magnetization vector field
- **Vector ptycho-tomography** is the first method that can image the 3D magnetization vector field at the nanoscale without any prior knowledge or pre-assumed model.
- Highest spatial resolution 3D spin texture to date ~10nm
- Soft X-ray HHG can image dynamic spin textures

Challenge: Vector Ptycho-tomography of Skyrmions



Binnie et al., in prep (2025)

ALS synchrotron
COSMIC beamline



Kortright et al., Phys. Rev. B 62, 12216

X-ray Magnetic
Circular Dichroism (XMCD)

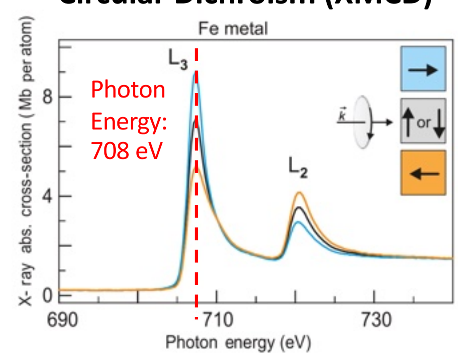
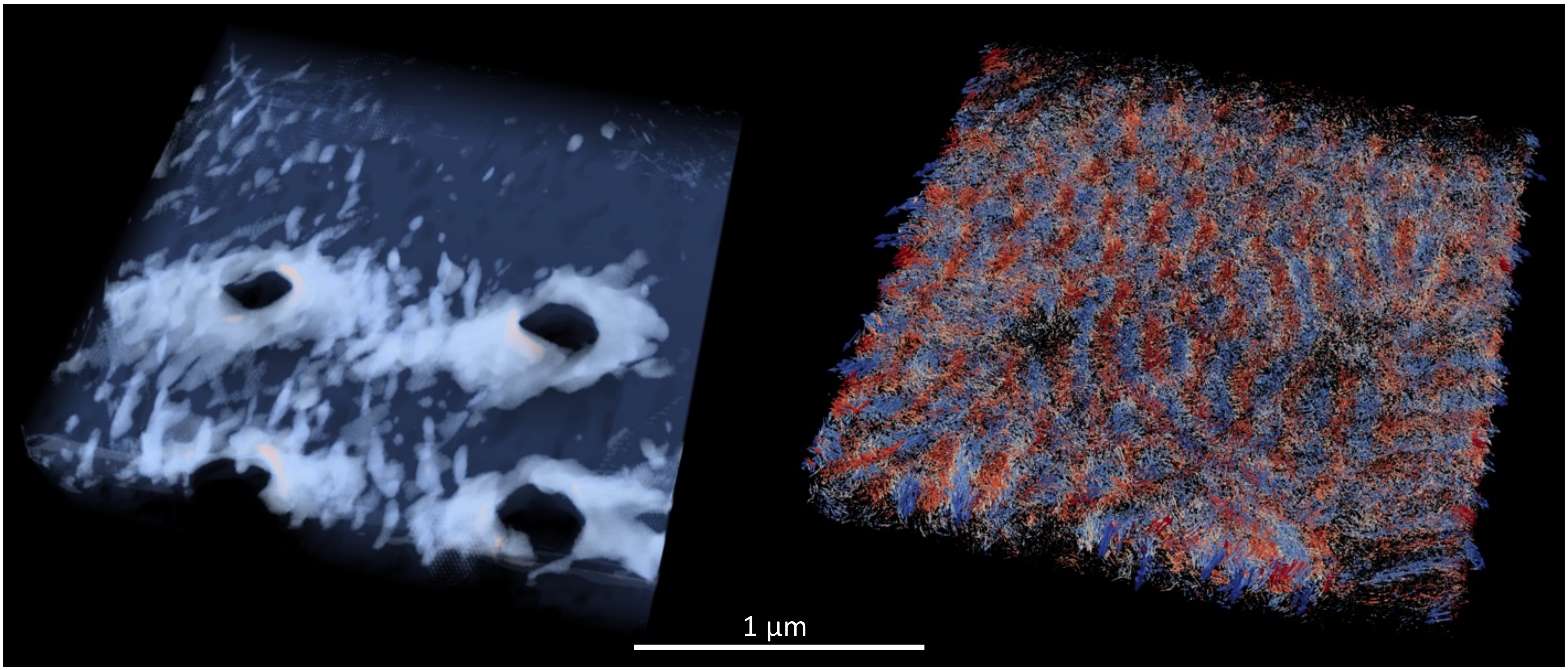


Image from Bonetti, J. Phys.: Condens. Matter 29 133004 (2017)

Vector Tomography of Dipole-Stabilized Skyrmions & Stripe Domains



Charge signal

Magnetic signal

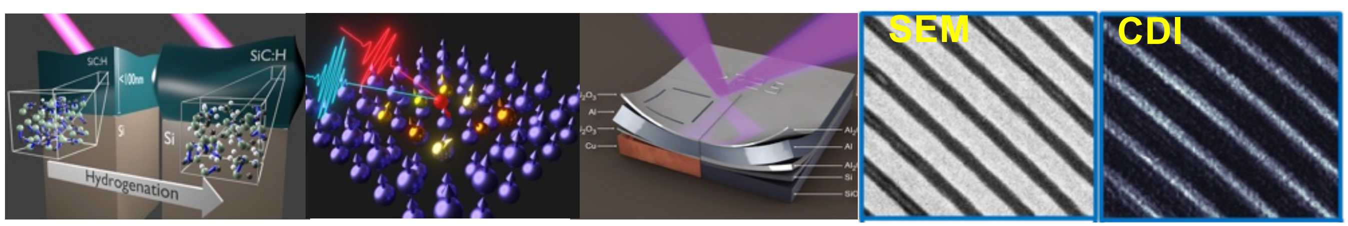
Binnie et al., in prep (2025)

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 being put to work in other fields
  - First sub-wavelength EUV imaging
  - Non-destructive maps of heterostructures for nanotechnologies
  - New understanding about nanoscale thermal transport
  - New understanding of spin and charge dynamics
  - Bright future – everything scales with the wavelength

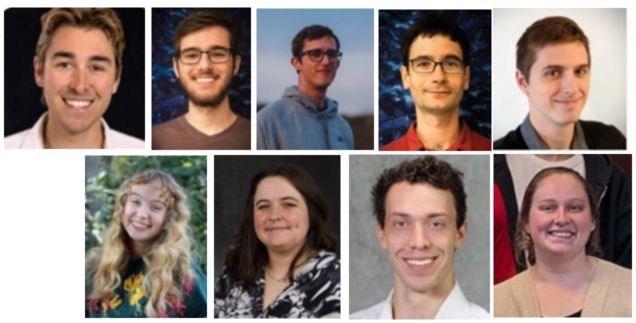


Light science **Materials Characterization** **Spin Dynamics** **ARPES** **Imaging**

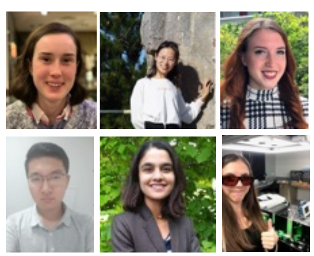
Team, collaborators and funders

Imaging & Materials Team
Dr. Nick Jenkins
Emma Nelson
Oliver Shao
Iona Binnie
Eric Feng
Clay Klein
Teddy Culman
Ben Shearer
Eric Feng
Yunhao Li
Jiayi Liu

Laser Team
Dr. Michaël Hemmer
Dr. Drew Morrill
Jeremy Thurston
Will Hettel
Greg Golba
Gabriella Seifert
Ivan Dickson
Emma Burton
Rachel Larson



Spin and Charge Dynamics Team
Anya Grafov, Dr. Na Li
Richa Sapkota, Dr. Shunye Gao



CU Boulder
Andreas Becker
Markus Raschke
Rahul Nandkishore
Mahmoud Hussein
Sadegh Yazdi
Steve George
Scott Diddams
Thomas Schibli

University of Salamanca
Carlos Hernandez-Garcia
Luis Plaja

UCLA
John Miao

UCSD
Eric Fullerton
Sergio Montayo

NIST
Justin Shaw
Hans Nembach

KMLABS: Liam Weiner, Travis Frazer, Autumn Wyatt

3M: Matt Atkinson, Matt Fray

SAMSUNG: Seungbeom Park, Wookrae Kim

Recent Graduates
Drs. Wenjing You, Yuka Esashi, Yingchao Zhang, Guan Gui (KLA)
Drs. Bin Wang, Chrissy Porter (ASML)
Drs. Michael Tanksalvala, Brendan McBennett (NIST)
Drs. Dan Carlson, Tika Kafle (Sandia)
Dr. Nathan Brooks (Academica Sinica, Taiwan)
Dr. Sinéad Ryan (Seagate)
Dr. Josh Knobloch (Utah State Univ.)