High resolution Hard X-ray Microscopy at the Advanced Photon Source:

Current capabilities and Future Thrust

Materials Science with Coherent Nanobeams
at the Edge of Feasibility

June 27, 2011

Jörg Maser

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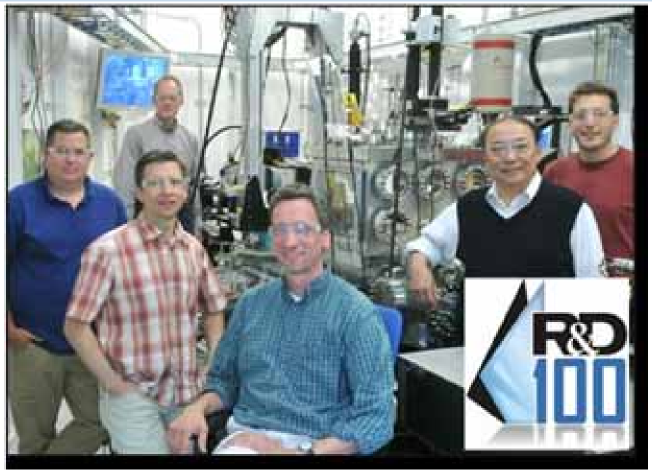
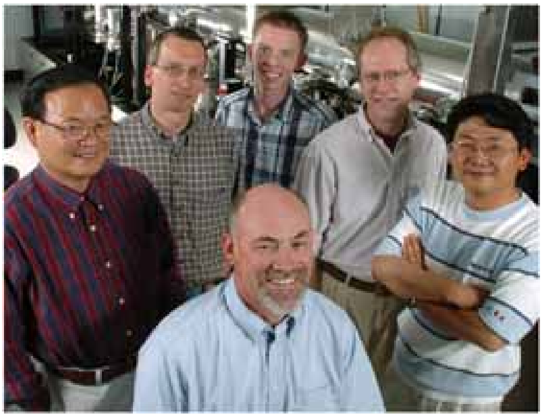
Collaborators



Hard X-ray Nanoprobe (CNM/APS)

R. P. Winarski, M. V. Holt[*], R. P. Winarski[+], V. Rose, D. Carbaugh[**], P. Fuesz, G. B. Stephenson[*$]



Multilayer Laue Lens

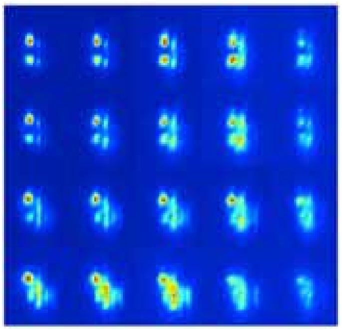
H. Yan[$$], V. Rose**, R. Conley[$$], H. C. Kang[$], A. T. Macrander**, R. Conley[$$], D. Shu[**], L. Assoufid**, B. Shi**

In Situ Nanoprobe

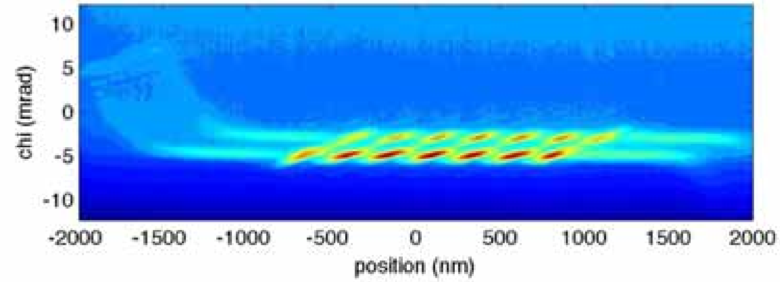
B. Lai, T. Buonassisi, D. Barton, W. Chiu, S. Darling, E. Ingall, J. Kang, K. Kemner, G. Mitchell, P. Monteiro, C. Murray, T. Rajh, V. Rose, Z. Cai,1 I. McNulty, L. Finney, C. Jacobsen, S. Vogt

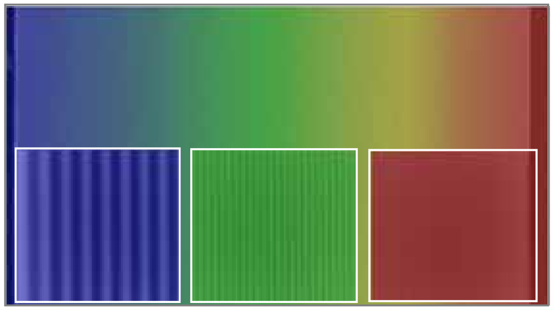
[*] CNM, **APS, [$] MSD, [$$] NSLS-II, [2] MIT, [3] Dow Chemical Company, [4] University of Connecticut, [5] Georgia Institute of Technology, [6] UC Berkeley, [7] IBM

Overview



- Motivation: High Resolution Imaging Approaches
- The Hard X-ray Nanoprobe – an Analytical X-ray Microscope
- Towards Nanofocusing of Hard X-rays: Multilayer Laue Lenses
- Outlook: the In-Situ Nanoprobe





I) High resolution characterization techniques

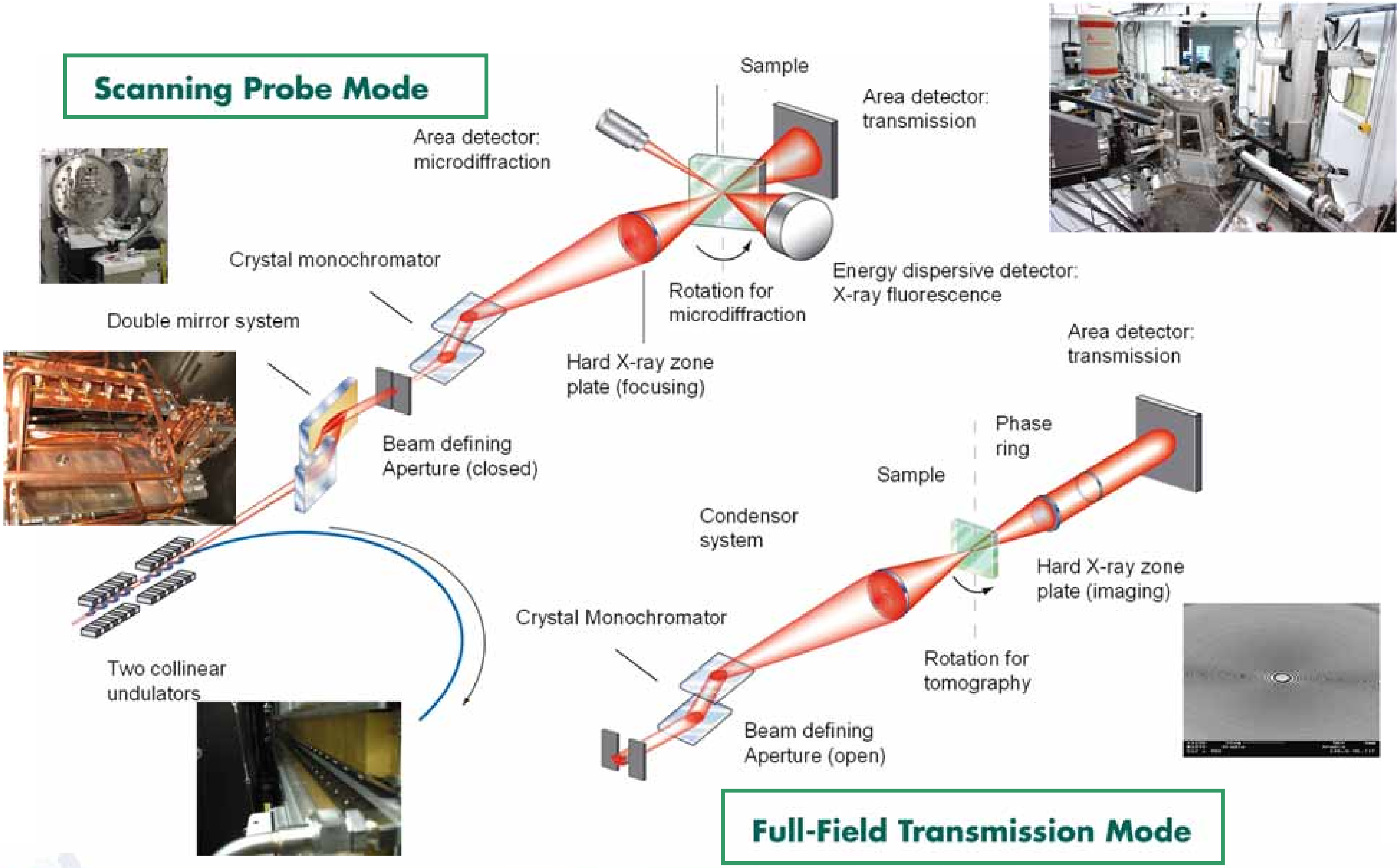
- Scanning Probe Microscopy: atomic-resolution surface characterization
  - Near-field optical microscopy, Atomic force/magnetic force/piezo force microscopy….

- Electron microscopy: atomic resolution structure/composition characterization
  - TEM, cross sectional TEM, STEM, SEM

- X-ray microscopy: nanoscale resolution structure/composition characterization

  - Properties:
    - Good penetration
    - Insensitive to electric/magnetic fields, environments → in-situ studies
    - Elemental/chemical/phase/strain sensitivity
    - Good time resolution
  - X-ray Scattering: nanosize characterization in environments/fluids
    - Particle size distributions (SAX), Non-uniform surface/interface features (Diffuse scattering)
    - Imaging of isolated, non-periodic systems (coherent)
  - X-ray Imaging:
    - Non-period complex structures with composition/phase/strain sensitivity
    - Use of secondary signals (fluorescence/scattering) from small volumes

CNM/APS Hard X-ray Nanoprobe: Combined Analytic and Imaging Mode



Scanning Probe Mode

Sample

Area detector: transmission

Area detector: microdiffraction

Crystal monochromator

Rotation for microdiffraction

Energy dispersive detector: X-ray fluorescence

Double mirror system

Hard X-ray zone plate (focusing)

Beam defining Aperture (closed)

Two collinear undulators

Area detector: transmission

Phase ring

Sample

Condensor system

Hard X-ray zone plate (imaging)

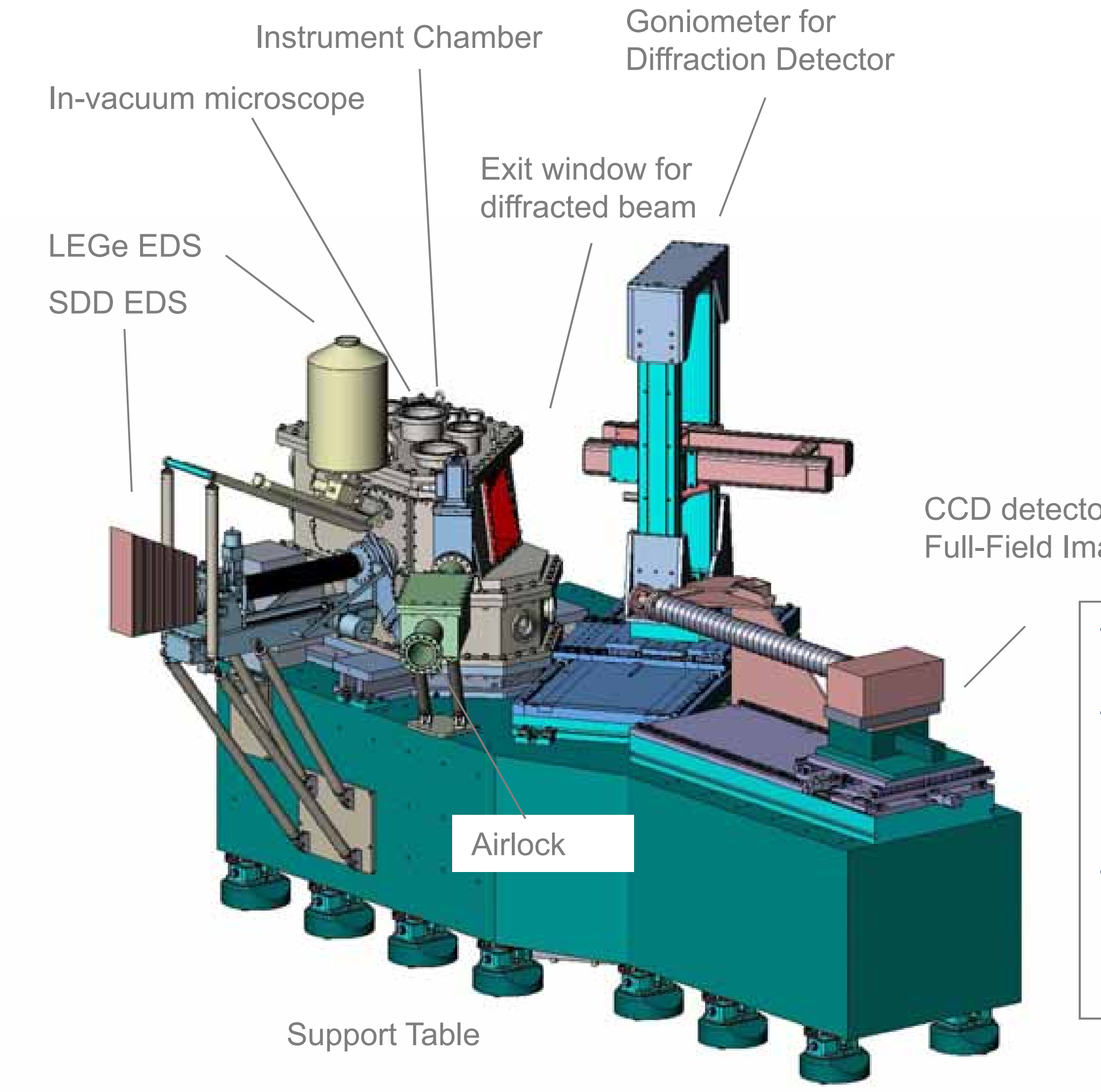
Crystal Monochromator

Rotation for tomography

Beam defining Aperture (open)

Full-Field Transmission Mode

Concept: Integration of 3D Imaging and Analytic Mode



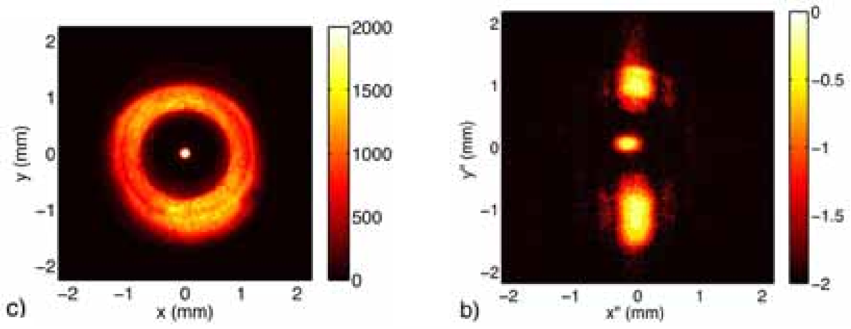
- Scientific Thrust:
  - Materials characterization at the nanoscale
- Capabilities:
  - Photon Energy: 3 – 30 keV
  - Spatial resolution: 30 nm (TXM)
  - 40 nm (scanning)
- Techniques:
  - Diffraction
  - x-ray fluorescence,
  - tomographic transmission imaging

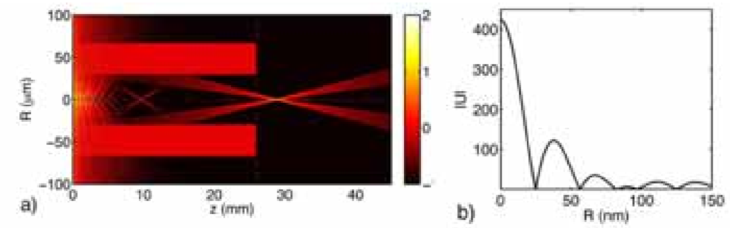
Overall concept and Scanning/Encoding Mechanism: ANL

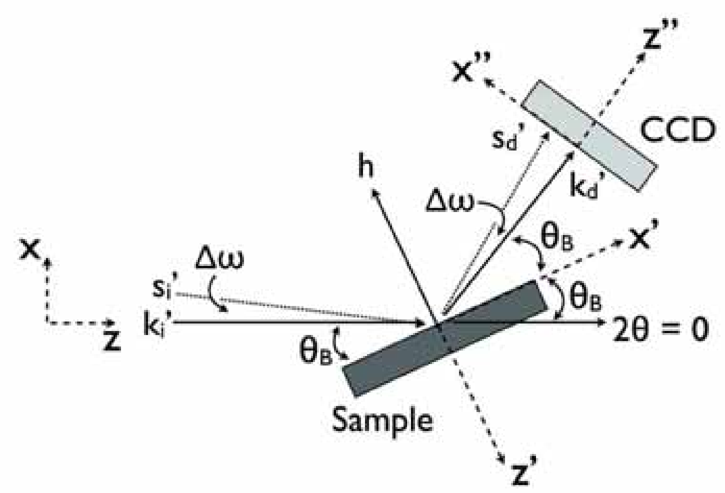
Engineering Design, Controls, TXM technology and Fabrication: XRADIA, Inc

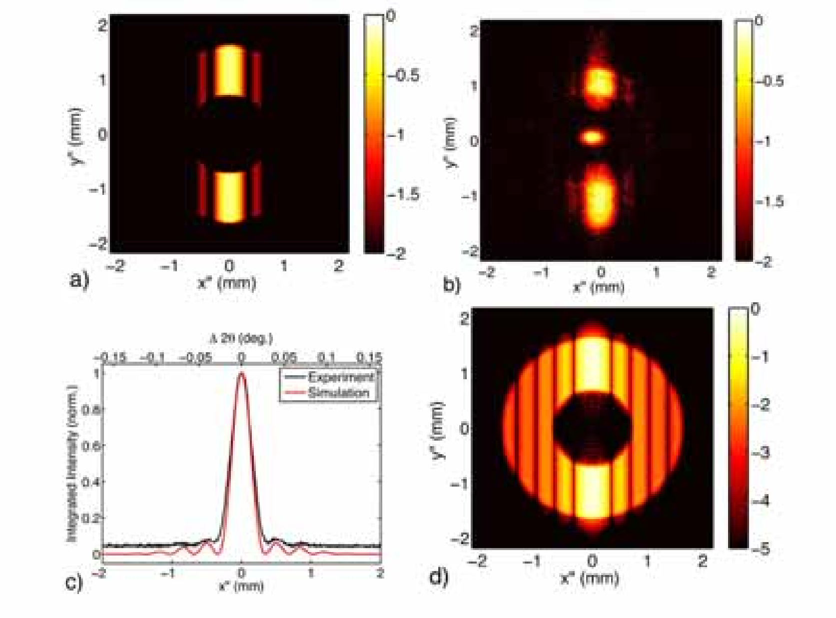
Nanodiffraction of strained SOI heterostructures

Characterization of nanobeam
diffraction with high numerical aperture



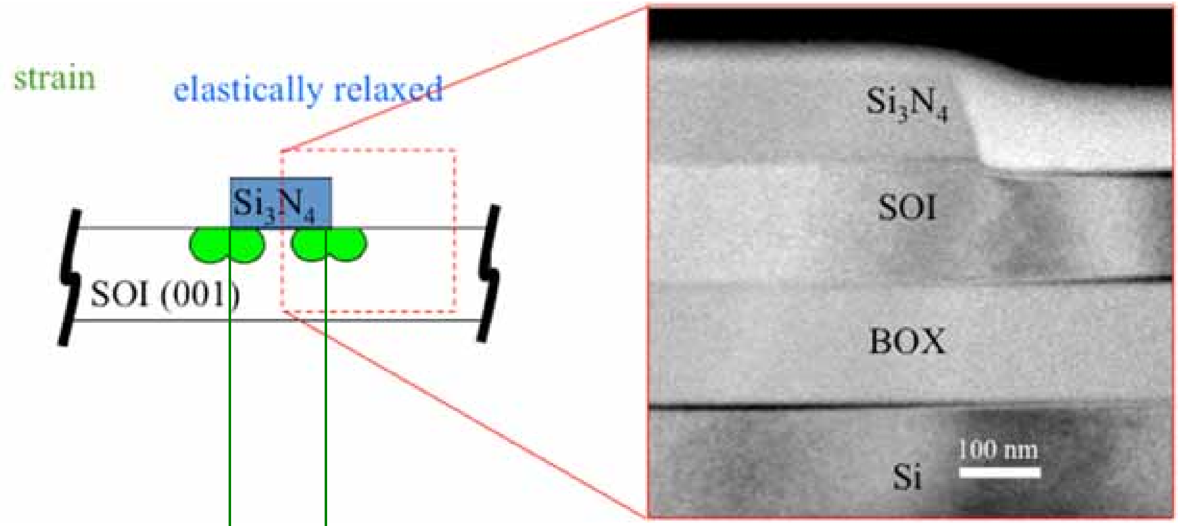




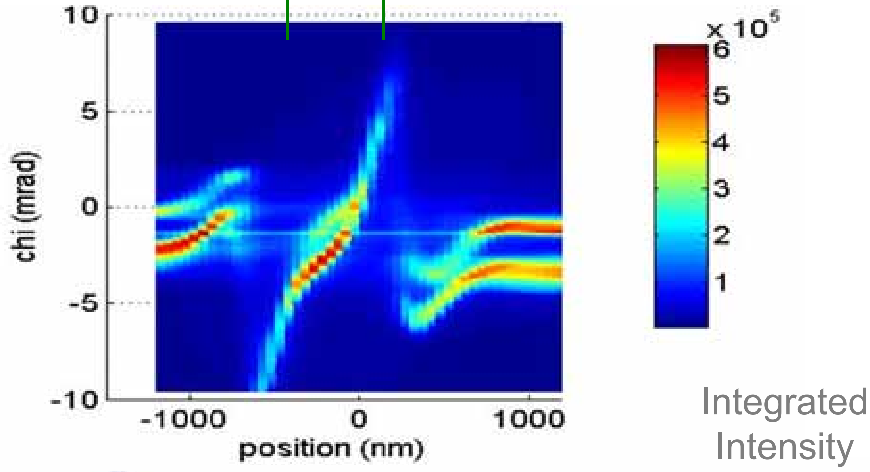


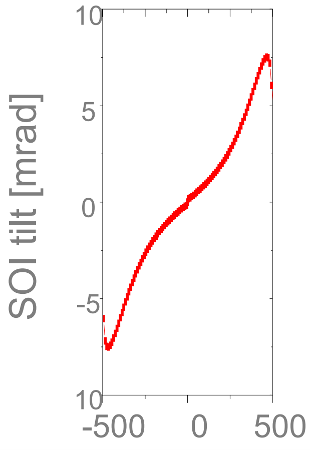
Andrew Ying, Braxton Osting, I.C. Noyan, Conal E. Murray, Martin Holt and Jörg Maser.
J. Appl. Crystallogr. 43 (3), 587-595 (2010).

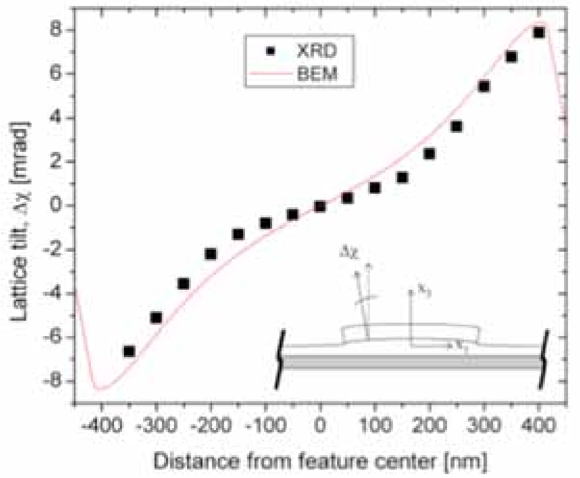
Strained silicon for CMOS applications



- antisymmetric lattice tilt distribution observed
- high degree of strain at feature edges
- tilt maxima @ ± 7.5 mrad







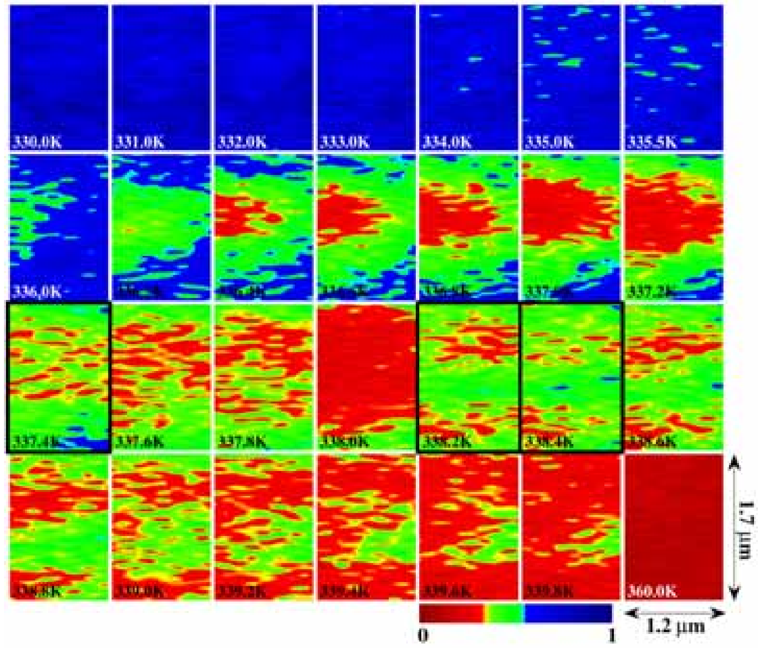
Conal E. Murray, A. Ying, S. M. Polvino, I. C. Noyan, M. Holt, and J. Maser, Journ. Appl. Phys. ***109****, 083543 (2011)*

Structural Transitions in VO_2

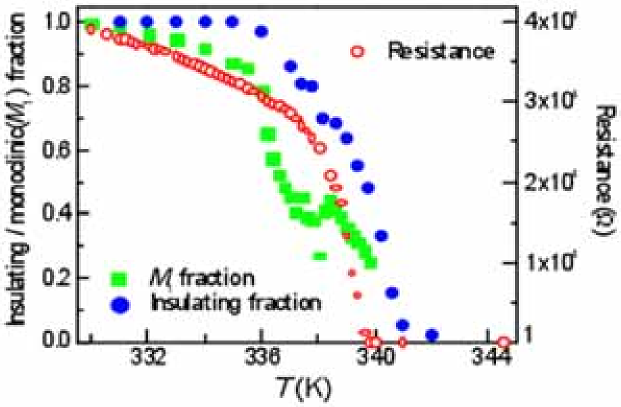
Study metal-to-insulator transition - correlation between structural and electronic phase transitions?

Variation in the intensity of the Bragg peak from the M1 phase of VO2, θ =29.58°.

Higher intensities (dark blue) indicate the presence of the M1 phase, lower intensities (red) represent the rutile phase, and intermediate intensities (green color) indicates coexistence of M1 and rutile phases.



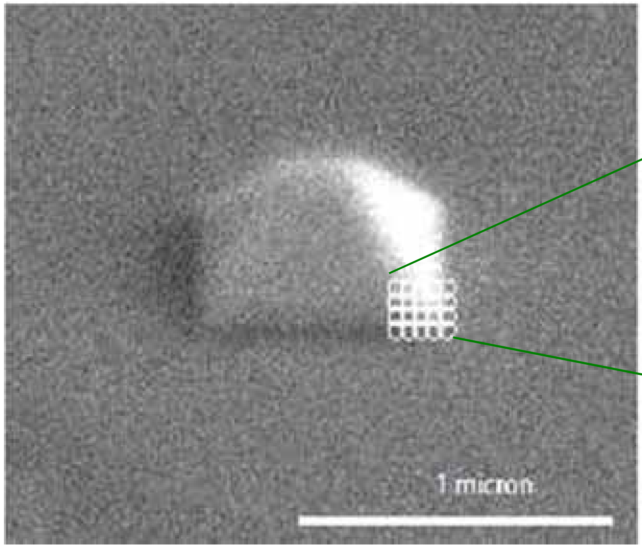
Non-monotonic growth of metallic “puddles”



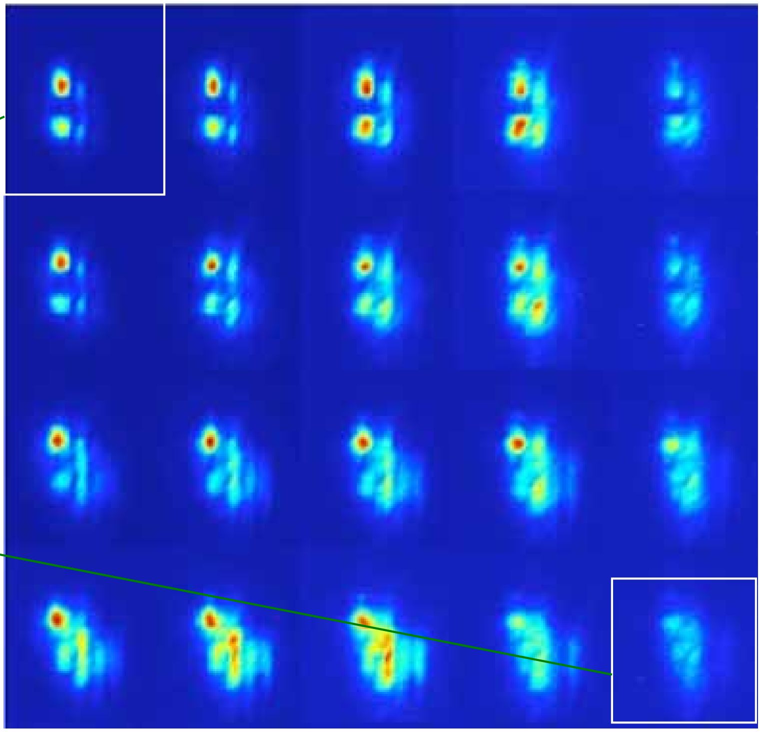
Fraction of monoclinic M_1 phase, resistance of VO_2 vs. temperature

M. M. Qazilbash, A. Tripathi, Bong-Jun Kim, Hyun-Tak Kim, Z. Cai, M. V. Holt, J. M. Maser, F. Keilmann, O. G. Shpyrko, and D. N. Basov. Phys. Rev. B 83, 165108 (2011)

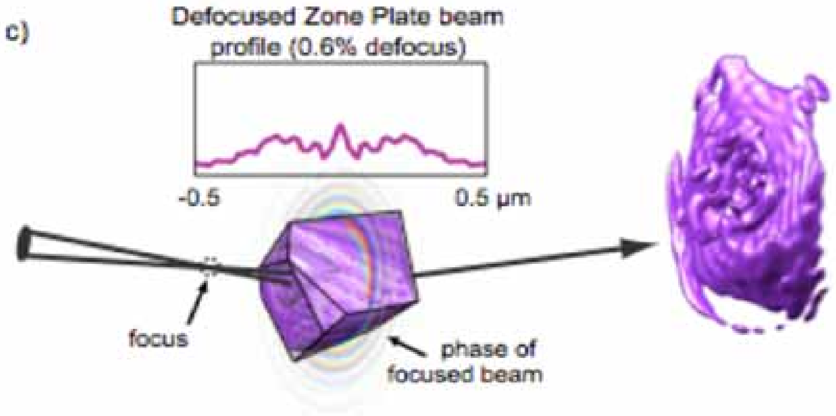
Bragg coherent diffraction in Bragg geometry: from Bi_2O_3 nanoislands



Bi_2O_3 ß-phase nanopyramid (SEM).

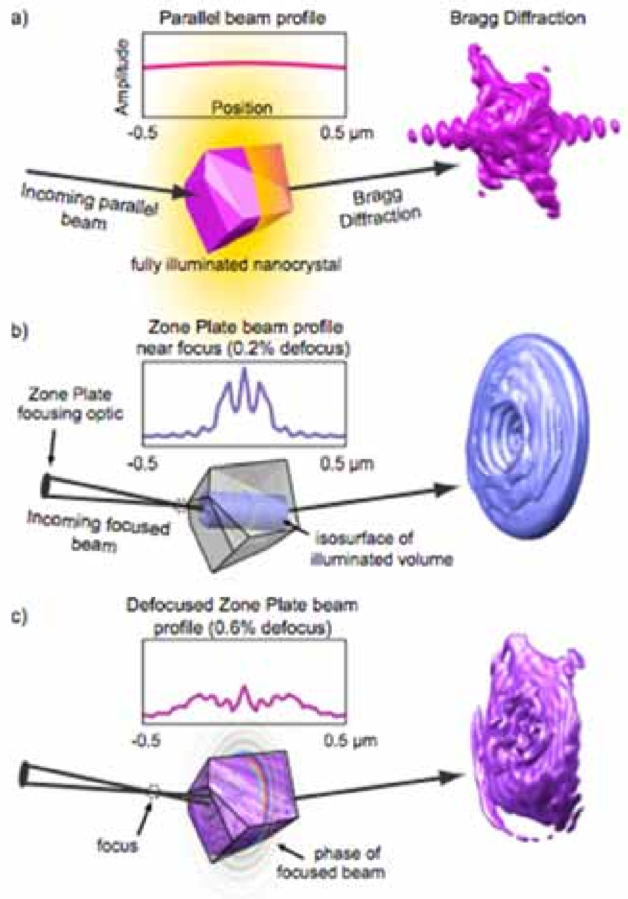


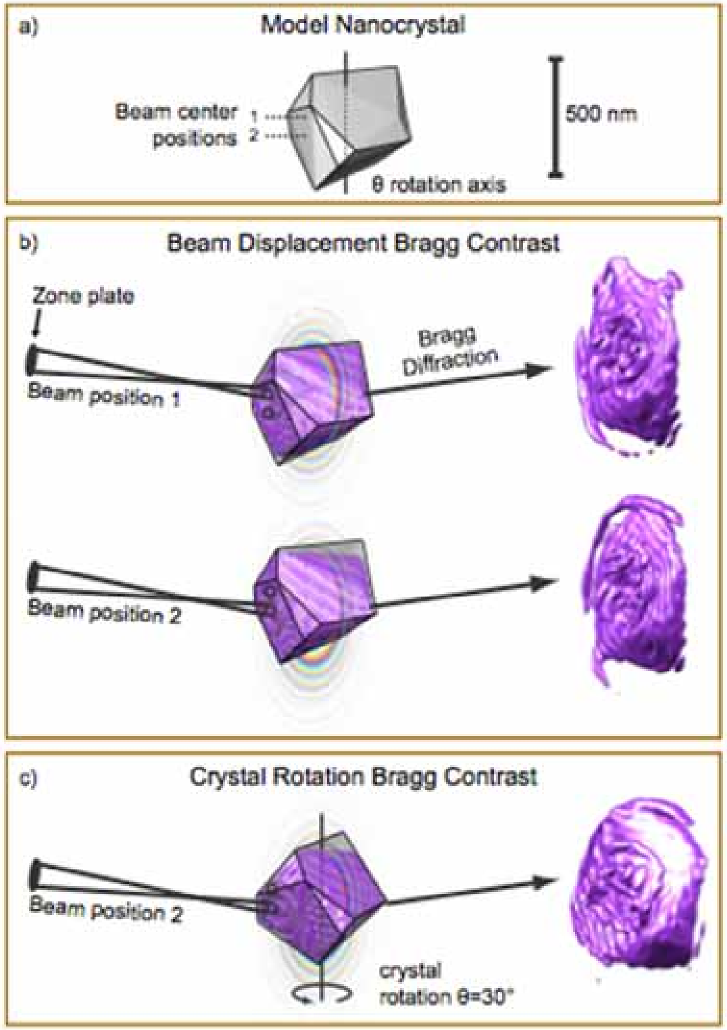
D.L Proffit (MSD-ANL & Northwestern U.); G.-R. Bai, D.D. Fong, T.T. Fister, S. Hruszkewycz, M.J. Highland, P.M. Baldo, P.H. Fuoss, J.A. Eastman (MSD-ANL); T.O. Mason (Northwestern U.)



S. Hruszkewycz, M. V. Holt, A. Tripathi, J. Maser, P. H. Fuoss, Optics Letters, Vol. 36, Issue 12, pp. 2227-2229 (2011)

Bragg coherent diffraction from Bi_2O_3 nanoislands





S. Hruszkewycz, M. V. Holt, A. Tripathi, J. Maser, P. H. Fuoss, Optics Letters, Vol. 36, Issue 12, pp. 2227-2229 (2011)

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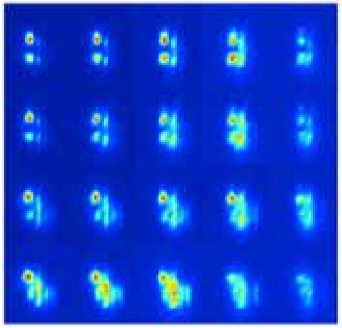
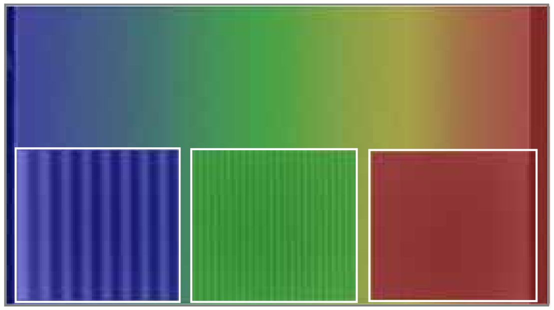
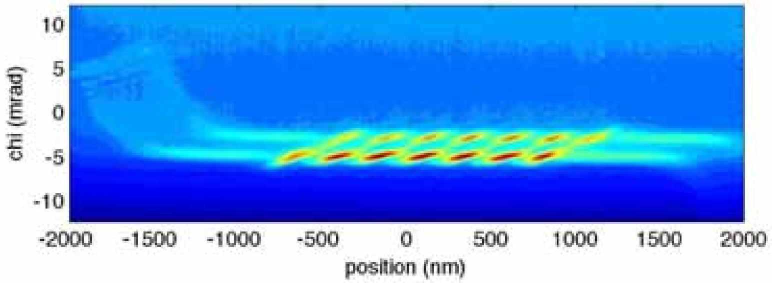
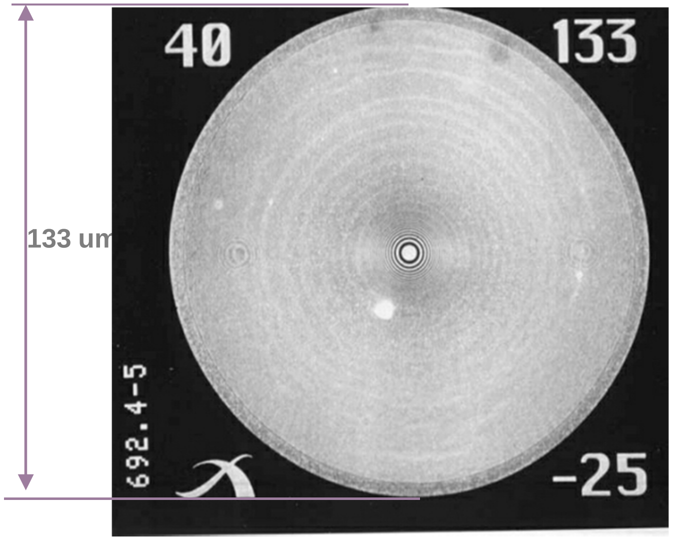


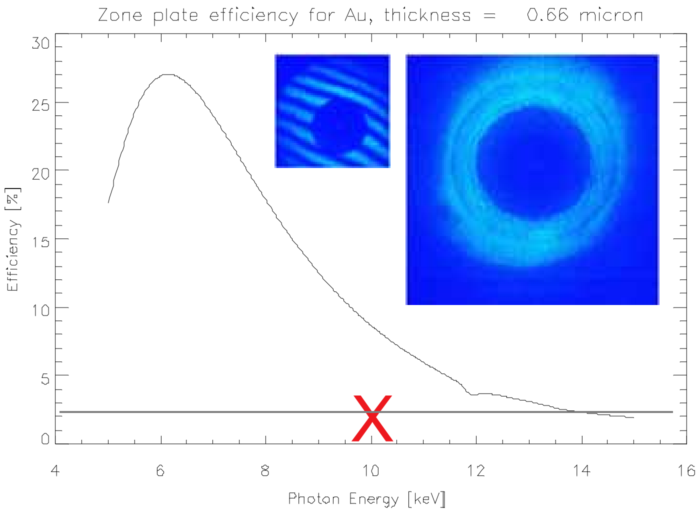




Diffractive Optics for Focusing and Imaging Optics

- Spatial resolution: ~ 15 nm for soft x-rays (CXRO/BESSY),
  ~ 30 nm – 40 nm for hard x-rays (APS TXM, CNM HXN, 2008)





Critical Parameter: Aspect ratio A = t/dr$_n$

dr$_N$ = 24 nm: AR ! 100:1, E = 10 keV

E-beam lithography: AR ≅ 10:1 – 15:1

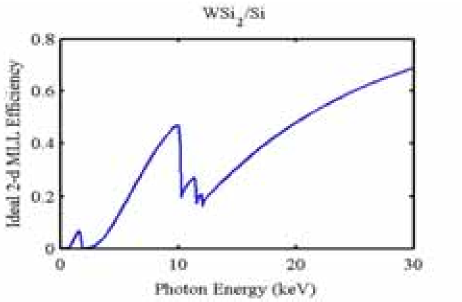
Currently fabricated, XRADIA:

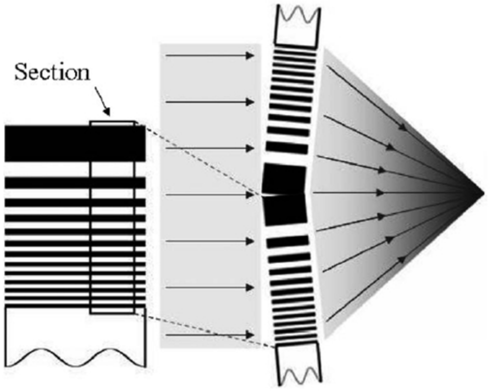
single stack zone plate, t = 300 nm

two stacked 24 nm ZP, t = 660 nm, η = 2%

Path to high resolution and high efficiency:

Multilayer Laue Lens



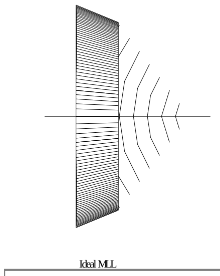


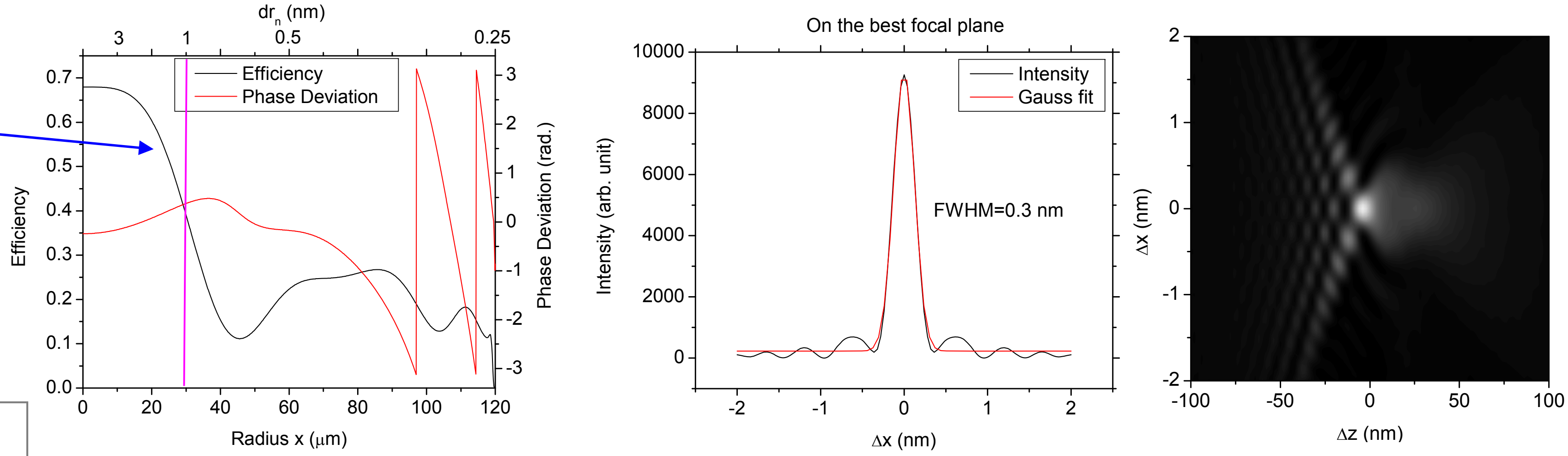
Courtesy G. B. Stephenson

Understanding the Ultimate Resolution Limit

Takagi-Taupin Description of Dynamical Diffraction

Reduced Efficiency at dr_N ~ 1.5 nm





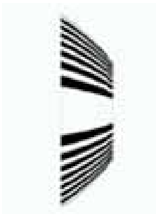
Wedged MLL, straight zones

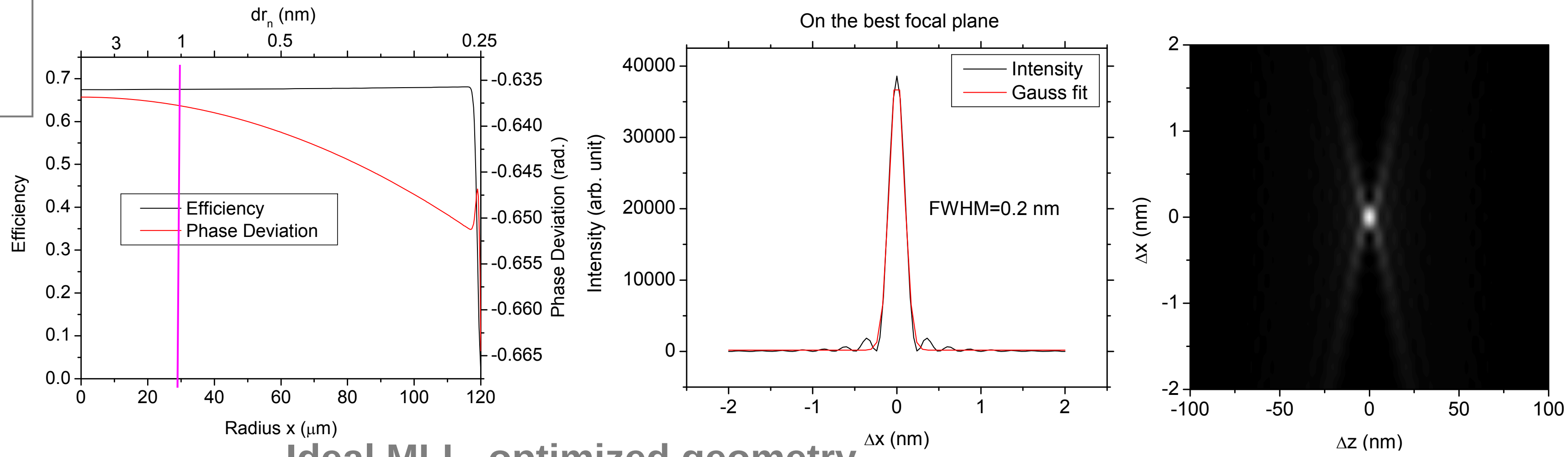
dr_N = 2.5 Å

E = 19.5 keV

D = 120 um

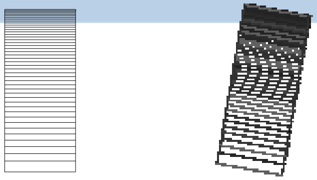
WSi_2/Si





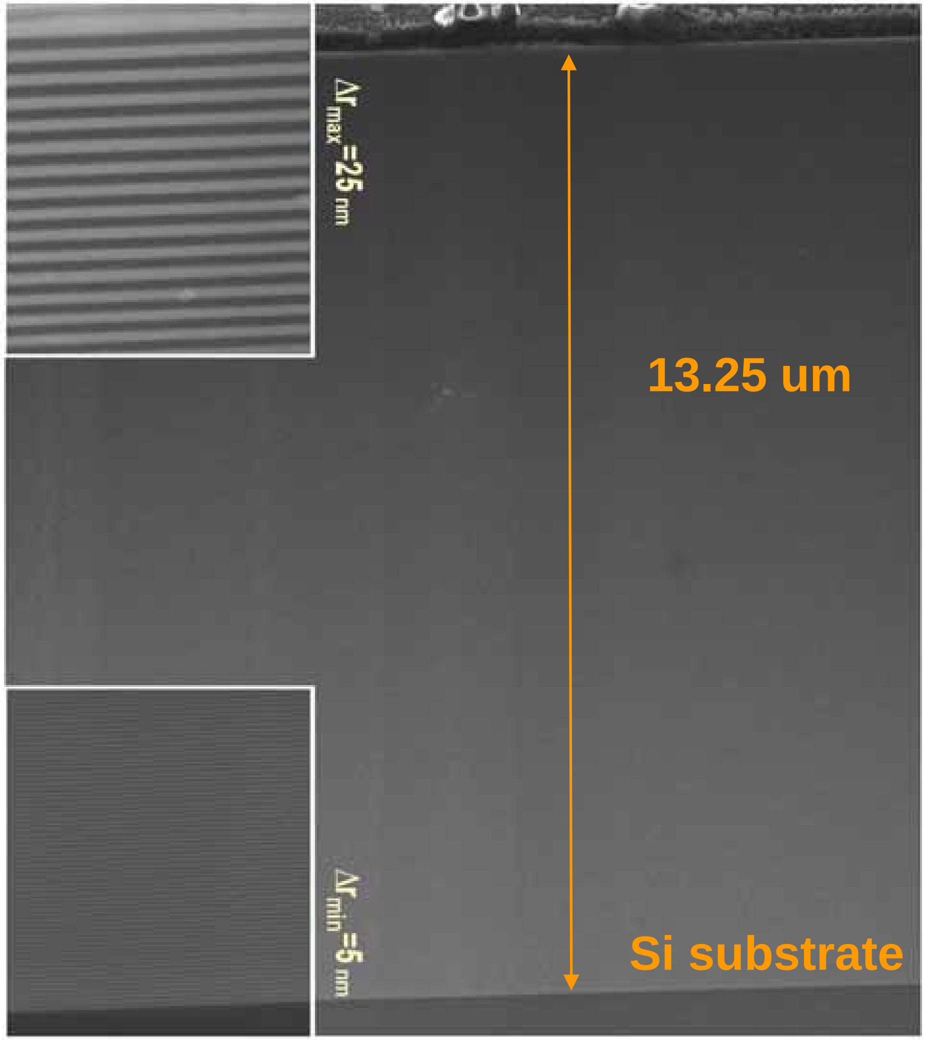
Ideal MLL, optimized geometry

Hanfei Yan et al. (PRB, 2007)



Partial flat/tilted Multilayer Laue Lens

- **Example for dr_N = 5 nm structure:**
  - Material: WSi_2/Si
  - dr_N = 5 nm (2d = 10 nm)
  - r_n = 16.5 um (13.3 um deposited)
  - N = 1653 (~ 1588 deposited)
  - Usable aperture fraction: 40%
- **Challenges:**
  - Large deposition thickness requires low stress: → WSi_2/Si
  - High accuracy of layer placement:
    - $f = (2\,r_n/\lambda)\cdot dr_n(r_n)$
  - Thinning/Polishing: avoid distortions of structure

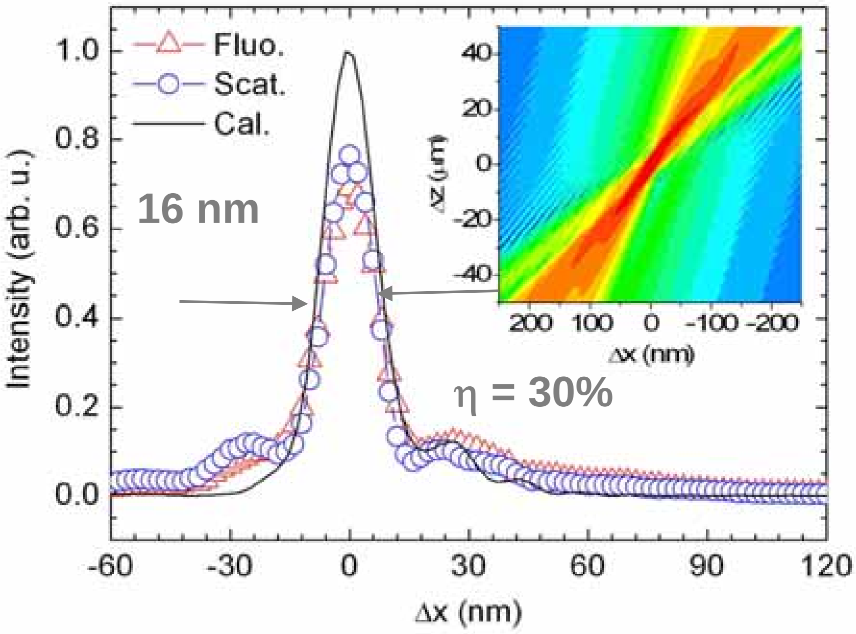


Courtesy R. Conley, C. Liu, A. Macrander

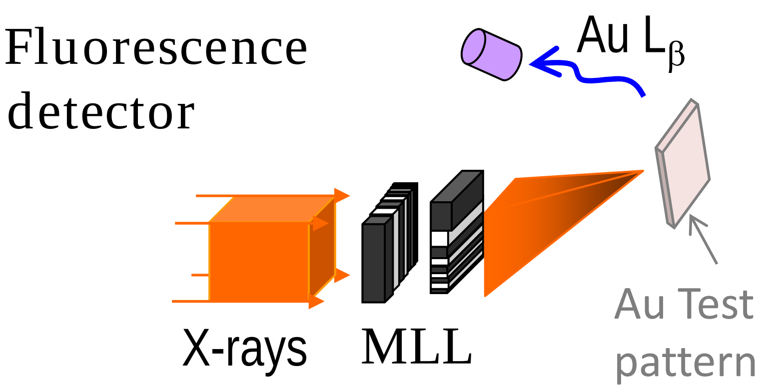
X-ray focusing with tilted MLL sections (1D, 2D), dr_N = 5 nm

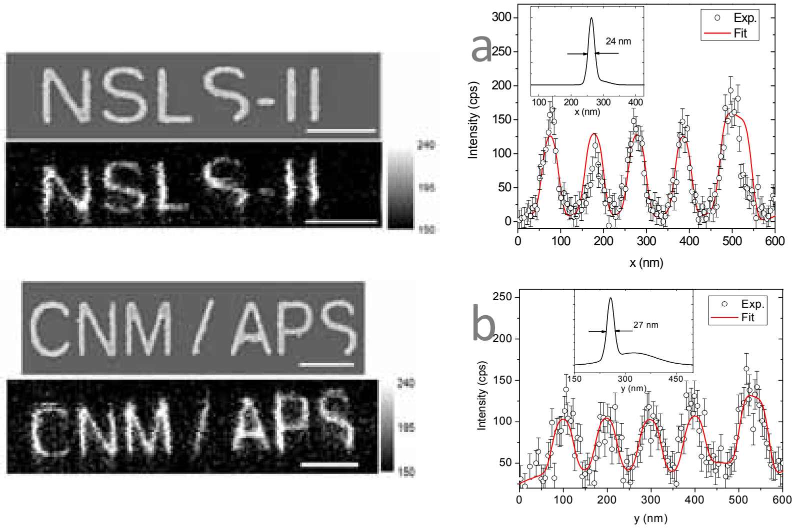
- **Parameters:**
  - drN = 5 nm structure:
  - Deposited: 13.3 um of 30 μm
  - → diffraction limit: 12.5 nm

Photon Energy: 19.5 keV



1D Efficiency: 30% at 19.5 keV





Measured Spot size 25 x 27 nm, 12 keV

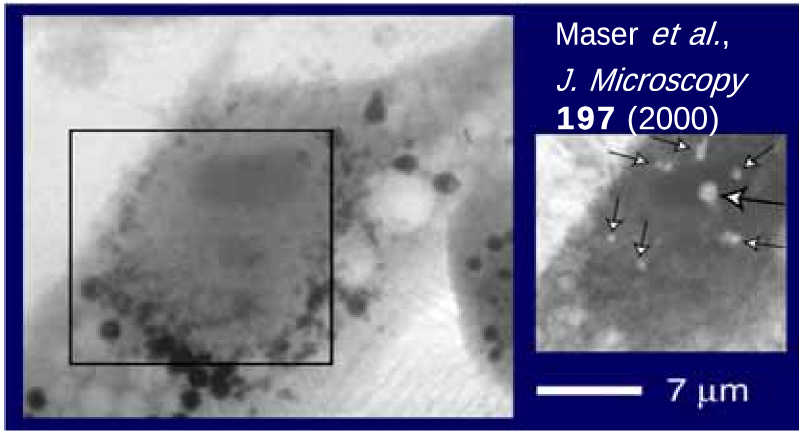
2D Efficiency: 17% at 19.5 keV

Kang et al, APL 92, 221114 (2008)

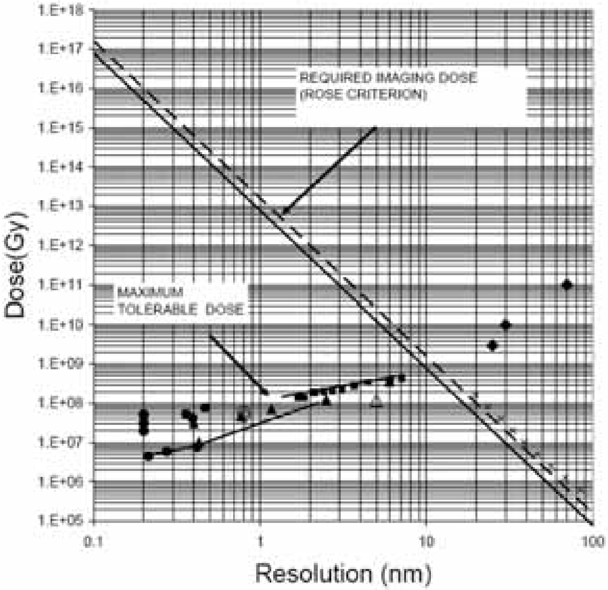
Yan et al, submitted, Optics Express

Experimental Considerations: Radiation Damage

- Bio-medical sciences: Dose limit ~10^{10} Gy

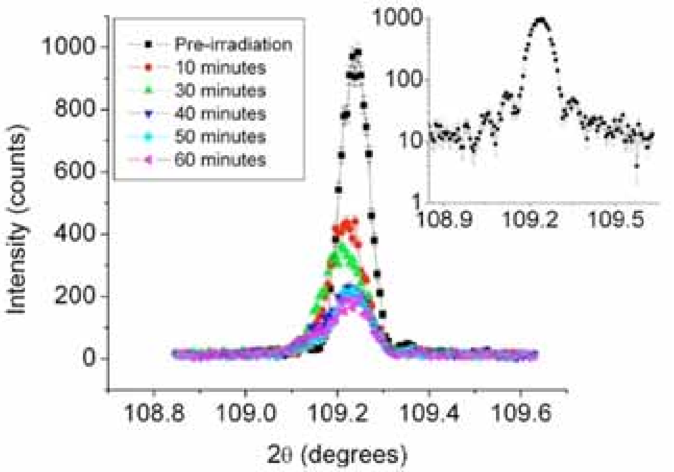


Maser *et al.*, *J. Microscopy* **197** (2000)

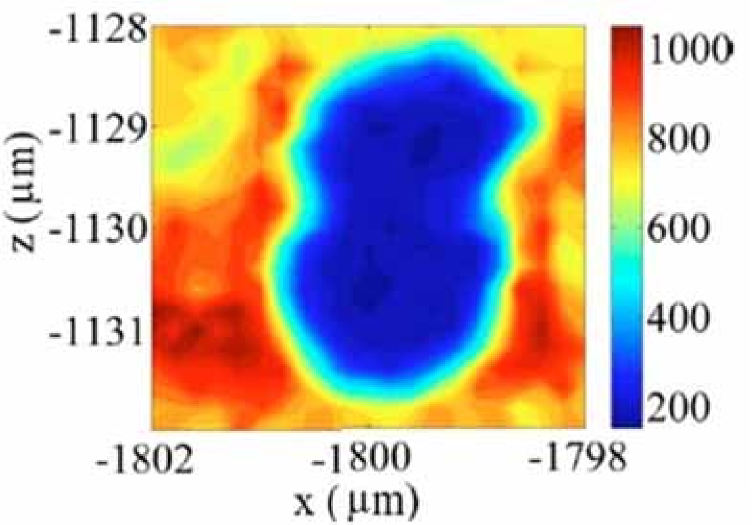


M. Howells et al. 2005

- Materials science: Dose limit?



140 nm SOI Si(800) reflection Total dose: 10^8 Gy



Polvino *et al.*, APL **92,** (2008) APS sector 2 microdiffraction

Experimental considerations for sub-10 nm beams

- Experimental Parameters:
  - Photon Energy: 20 keV
  - Resolution: 5 nm
  - Required monochromaticity: $\Delta E/E = 10^{-5}$
- Focal length: f = 5 mm (D = 60 um @ 74 m)

- Depth of focus: DOF = 1 micron @ 20 keV

- Considerations for future approach:

  - Compact instrument ("Pocket Nanoprobe")
  - Fluorescence, CDI capabilities
  - Science Thrust:
    - defects in solar cells
    - dopants distributions in nanoelectronics
    - catalysis

Experimental Considerations for ~ 1 nm beams

- Experimental Parameters (e.g. ERL @ APS):
  - Photon Energy: 10 keV
  - Resolution: 1 nm
  - Required monochromaticity: $\Delta E/E = 10^{-5}$
- Focal length: f = 1 mm (D = 120 um @ 74 m)
- Depth of focus: DOF = 20 nm
- Power considerations
  - Example *($B = 5 \cdot 10^{21}$ Photons/s/mm^2/mrad^2/0.1%BW)*
    - Focused Photon Flux: $1.5 \cdot 10^{11}$ Photons/s ($\Delta E/E = 10^{-5}$)
    - Power absorbed by sample: $2 \cdot 10^{-5}$ W
    - Sample: Si, t = 1 um → PD = 10^7 W/mm^2

➔ Significant fundamental issues at 1 nm/ ERL brightness

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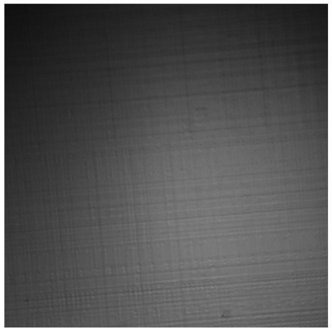
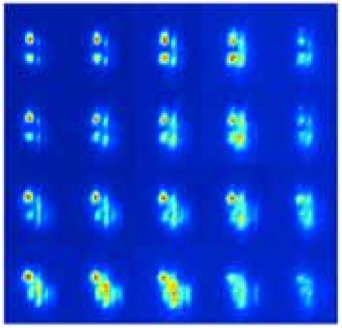
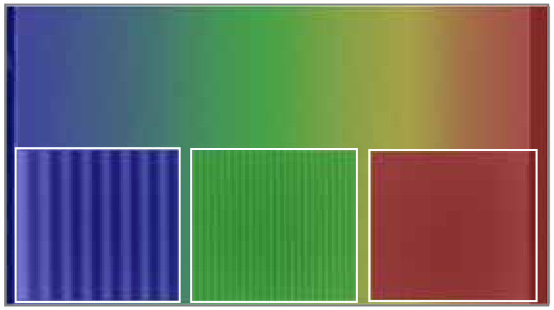
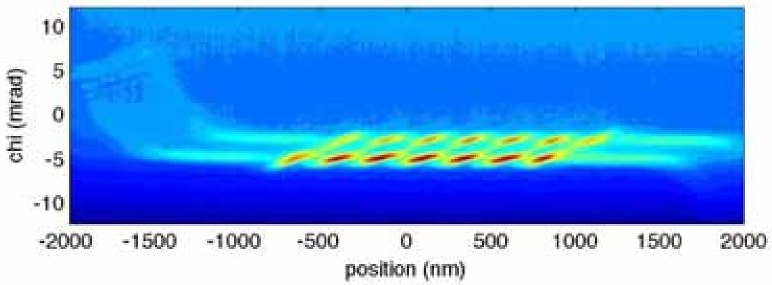








Next Generation Facility: In-situ Nanoprobe Beamline. Imaging hierarchical structures under Real conditions

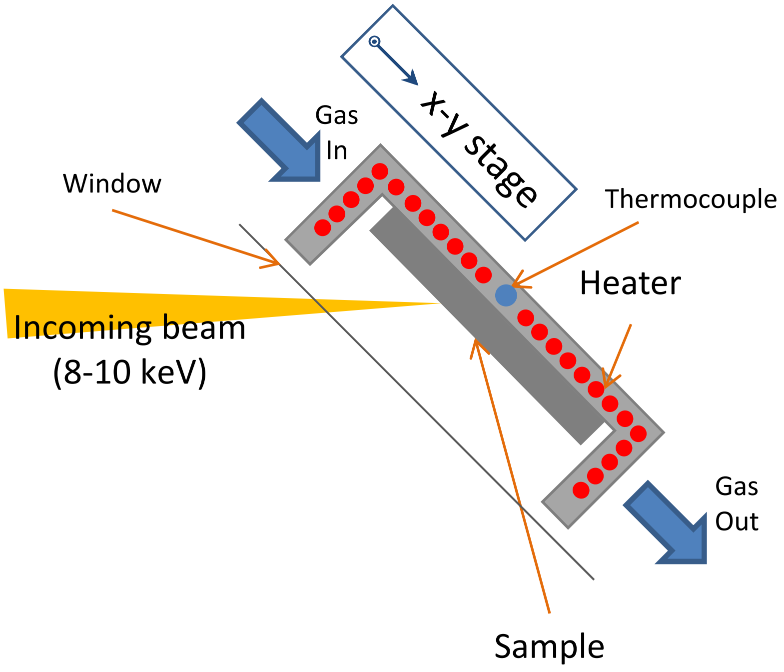
- Capabilities: Fast Fluorescence Imaging and Spectroscopy

- Mirror Optics for focusing to 50 nm
  - Accomodates 100x larger bandwidth than diffractive optics
  - 10x focusing efficiency compared to diffractive optics
  - Full spectroscopy
- MLL optics for focusing to 20 nm
  - Good efficiency for hard x-rays
- In-situ capabilities
  - Heating to 1000 ° , cooling to 90 K (LN2), sub-70K (He)
  - Flow of fluids and gases
- Detection channels
  - X-ray fluorescence for elemental mapping and spectroscopy
  - 2D/3D elemental imaging
  - Small angle coherent diffraction to detect defects

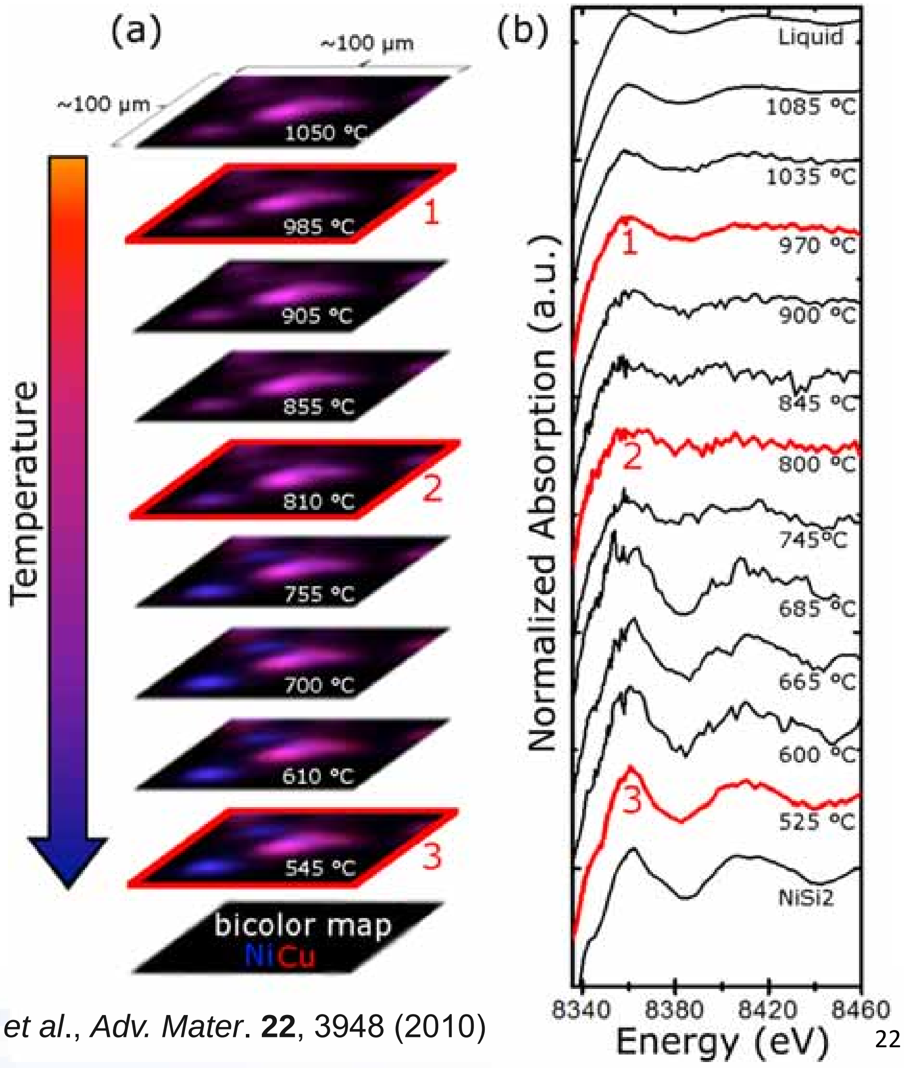
Jörg Maser,[1] Barry Lai,[1] Tonio Buonassisi,[2] David Barton,[3] Wilson Chiu,[4] Seth Darling,[1] Ellery Ingall,[5] Joo Kang,[3] Ken Kemner,[1] Gary Mitchell,[3] Paolo Monteiro,[6] Conal Murray,[7] Tijana Rajh,[1] Volker Rose,[1] Zhonghou Cai,[1] Ian McNulty,[1] Lydia Finney,[1] Chris Jacobsen,[1] Stefan Vogt[1]
[1] ANL, [2]MIT, [3]Dow Chemical Comnany, [4]University of Connecticut, [5]Georgia Institute of Technology, [6]University of California, Berkeley, [7]IBM

Variable-Temperature Nano-Fluorescence Mapping

- Capability: Variable temperature stage from 90 K → > 800 °C with nanoprobe
  - Discovery: Thermodynamics and kinetics of defect generation and annihilation



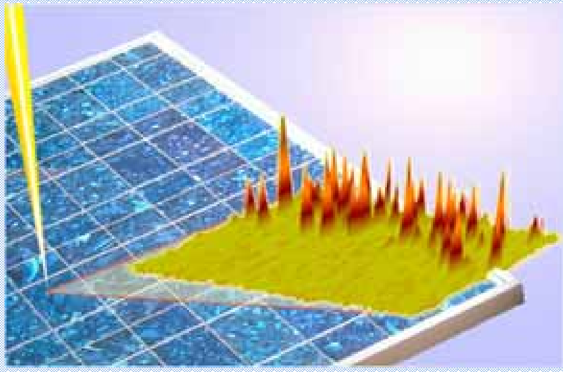
Courtesy T. Buonassisi



Hudelson *et al.*, *Adv. Mater*. **22**, 3948 (2010)

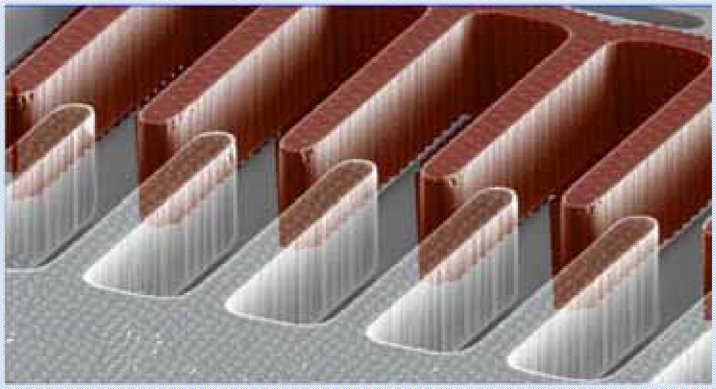
Science Thrust:

Photovoltaics



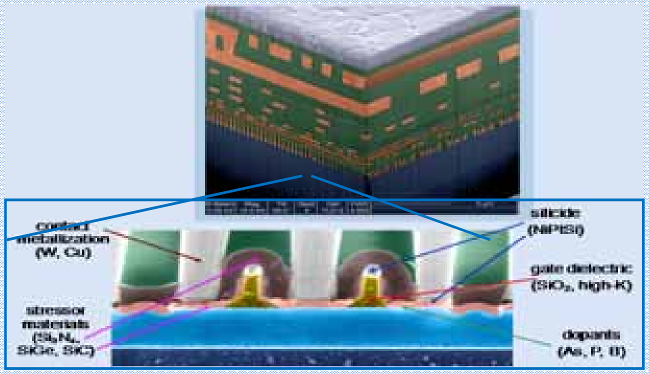
Courtesy T. Buonassisi, MIT

Energy Storage



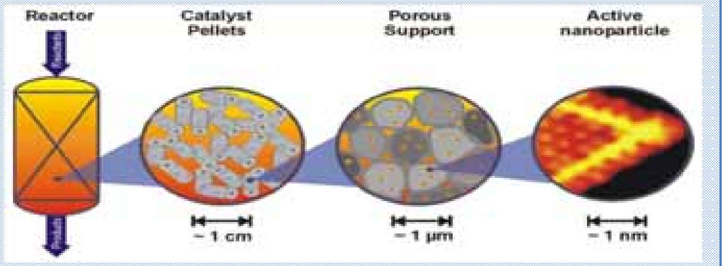
Courtesy T. Rajh, ANL

Nanoscale Electronics



Courtesy C. Murray, IBM

Catalysis



Courtesy G. Mitchell, Dow Chemical

Summary

- **Current Capabilities: CNM/APS Hard X-ray Nanoprobe**
  - Nanoscale Diffraction, fluorescence, tomography at 30 – 50 nm
    - Strain in materials and devices, study of structural phase transitions
    - Structure/composition of geopolymers, biological systems
    - Advanced building materials, energy storage systems

- **Towards Nanofocusing of Hard X-rays**
  - APS Goal: 5 nm focus at 25 keV
    - Nanoscale impurities and contaminants at defects (solar cells, ULK dielectrics)
    - Advanced energy materials (e.g. nanoparticle based battery electrodes, fuel cell materials)
  - Approach: compact MLL microscope/pocket nanoprobe

- **Future: In-Situ Nanoprobe Beamline**
  - 20 nm – 50 nm resolution at 10 – 1000x increased flux
  - In-situ fluorescence spectroscopy, tomography
  - Sensitivity to << 100 atoms
  - Coherent scattering to map defects
  - Applications: Photovoltaics, Energy Storage, Nanoscale Electronics, Catalysis, Biomedical applications