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## *Opportunities for X-ray Microscopy with the APS Upgrade*

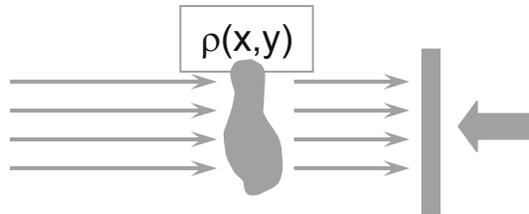
*Stefan Vogt, Barry Lai, Jörg Maser and Qun Shen*

*X-ray Science Division - Advanced Photon Source*

## *APS Upgrade Planning Meeting for Microscopy, July 17th - Agenda*

- Welcome - Gabrielle Long, XSD
- Anticipated Capabilities of the Upgraded Storage Ring - Michael Borland, ASD
- APS upgrade and future prospects of nanobeam - Barry Lai, XSD
- Some Challenges in Micro-beam X-ray Strain Analysis - Cevdet Noyan, Columbia University
- Impact of improved spatial resolution and spectroscopy capabilities for the biogeochemistry and environmental science - Kenneth Kemner, Bio-ANL
- Subcellular Transition Metal mapping reveals new pathways in Infectious Disease - Thomas O'Halloran, Northwestern University
- Impact of Proposed Upgrades on Microbeam Studies in the Earth and Planetary Sciences - Steve Sutton, University of Chicago
- High Resolution Element-Selective Microscopy Using X-ray Enhanced Scanning Tunneling Microscopy - Volker Rose, CNM
- Achromatic optics for new frontier in XRF and XRD - Jon Tischler, Oak Ridge Nat'l Lab
- Discussion
  
- Plus contributions & comments by email, phone, etc:  
Gayle Woloschak, Tanja Paunesku, Steve Heald, Paul Evans, James Penner-Hahn, Juergen Thieme, Peter Ingram, Zhounghou Cai, ...

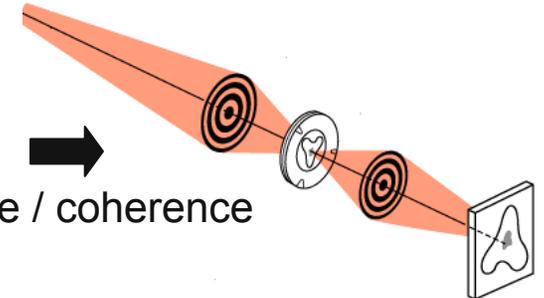
# X-ray Microscopy



*Direct imaging (radiography), with magnification in visible light \**

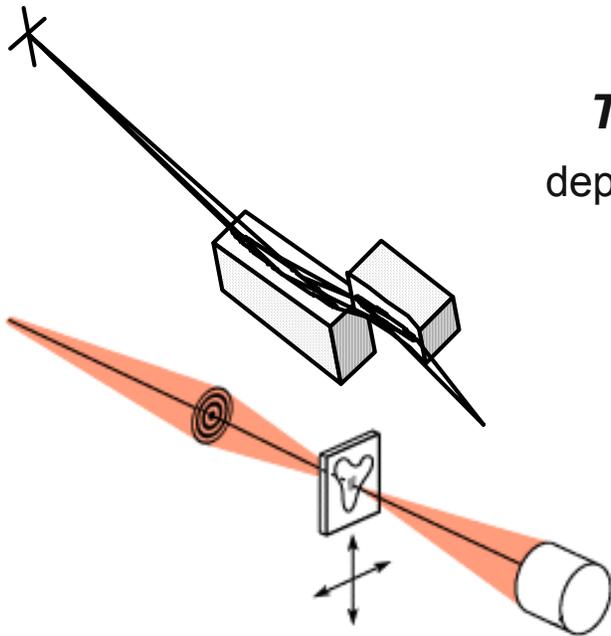
## **Transmission microscope (TXM)**

depends only on total flux, but not brilliance / coherence

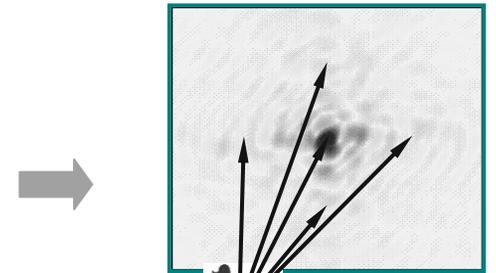


## **Scanning microscope (SXM)**

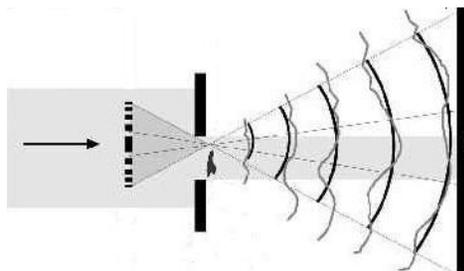
depends only on coherent flux, directly benefits from reduced emittance



*Coherent diffraction microscopy\**



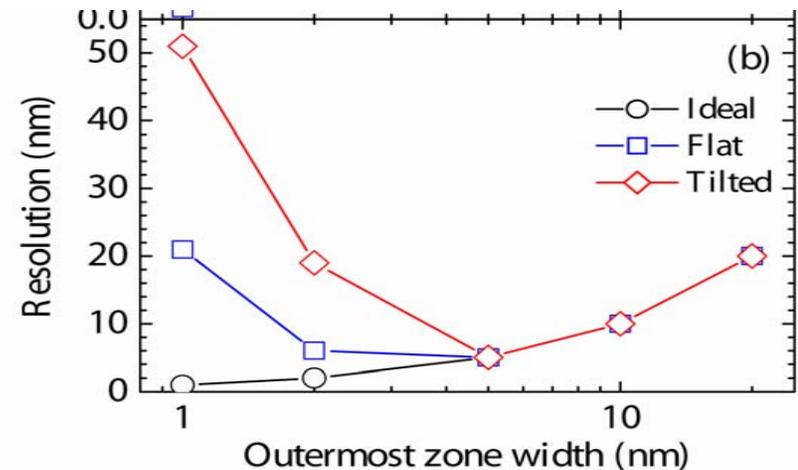
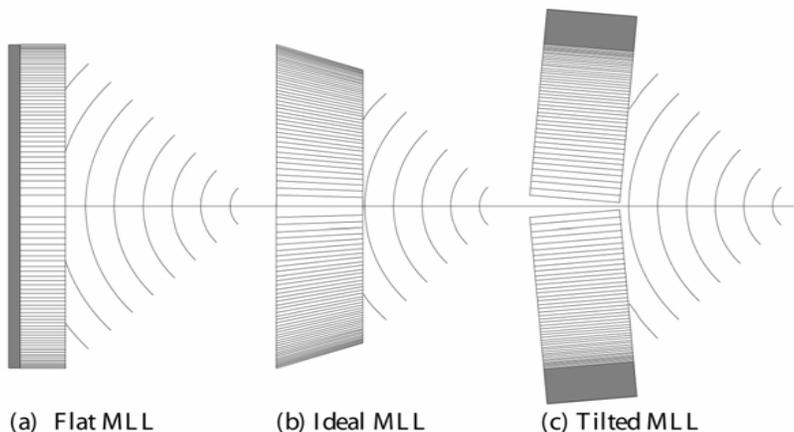
*Holography\**



\*: see following talk by Qun Shen

# Scanning X-ray Microscopy

- To achieve diffraction limited spot size, must illuminate X-ray optics coherently
- => any increase in brilliance directly increases focused flux accordingly
- Currently, high-res X-ray optics 'routinely' achieve  $\leq 200\text{nm}$ ;  
on Japanese 1km beamline:  $40\text{nm} \times 30\text{nm}$  with Kirkpatrick-Baez mirrors
- ANL demonstrated with new Multilayer Laue Lens  $< 20\text{ nm}$  in 1D (Maser *et al*)



Ideal structure: resolution approaching  $\delta = 1\text{ nm}$  feasible,

Tilted MLL:  $\delta = 5\text{ nm}$  feasible

Kang *et. al*, PRL, 2006

## Upgrade benefits for Scanning Microscopies

- Increased current (2x more flux)
- Longer straights (up to 5x more flux due to longer IDs)
- AND reduced emittance (3nm -> 1nm)
  - 3x more focussed flux or
  - 3x smaller spot size
- **VERY exciting**: possibility of further reduced emittance (ERL ?)
  - DIRECTLY translates into smaller spot / more focussed flux for harder X-rays
  - Ideal: ‘round’ source (e.g., 25 um diameter)
  - 3 nm emittance -> 0.03 nm **100x more focussed flux**/ smaller spot)
  - Additionally:
    - *no need for beam defining aperture*
    - *remove astigmatism*
    - *don’t ‘throw away’ incoherent photons*

## Implications for Scanning Microscopy

10 keV	Brilliance [ph/s/mm <sup>2</sup> /mrad <sup>2</sup> /0.1%]	Flux density * [ph/s/0.01%/μm <sup>2</sup> ]	
		(200 nm) <sup>2</sup> spot	(5 nm) <sup>2</sup> spot
<b>Today</b> 100 mA, 3.0 nm UA, L=2.4 m	4.0x10 <sup>19</sup>	9.5x10 <sup>10</sup>	1.5x10 <sup>14</sup>
<b>Upgrade</b> 200 mA, 1.0 nm UA, L=8.0 m	1.4x10 <sup>21</sup>	3.4x10 <sup>12</sup>	5.4x10 <sup>15</sup>
<b>Upgrade improved brilliance</b> 100 mA, 0.03 nm UA, L=8.0 m	2.3x10 <sup>22</sup>	5x10 <sup>13</sup>	9.0x10 <sup>16</sup>

Optics improvement: 1600

Source improvement

600x

35x

17x

Detector improvement: 6 x

⇒ Signal gain = 336kx – 5.5Mx

## Sensitivity & spatial resolution:

Today:

- (100 mA, 3.0 nm, UA, L=2.4 m)
- XRF detector collects 6% of  $4\pi$ SR

	Spot size		
sample thickness [um]	200 [nm]	20 [nm]	5 [nm] (0.1s)
0.1 [um]	3500	35	15
10 [um]	26000	260	60

Future:

- Upgraded (200 mA, 1.0 nm, UA, L=8.0 m), = 40x more coherent flux
- plus assume XRF detector collects 30% of  $4\pi$ SR

	Spot size		
sample thickness [um]	200 [nm]	20 [nm] (0.03s)	5 [nm] (0.002s)
0.1 [um]	180	6	4
10 [um]	1800	50	25

10 keV incident beam energy, biological sample in water (frozen hydrated)  
 minimum detectable Zn [#atoms], limited by rad damage:

For materials sciences samples, radiation damage less of an issue, expect higher sensitivity

## APS upgrade vs detectors & optics ?

- Most of the improvement in sensitivity comes from detector and optics improvement. What role does APS (ring & ID upgrade) play ?

**SPEED !!!** - only this will make most experiments feasible

- Example scan of a 5x5 micron area in x-ray fluorescence (e.g., cell nucleus, or part of a semiconductor structure)
- Today (100 mA, 3.0 nm, UA, L=2.4 m)
- XRF detector collects 6% of  $4\pi$ SR
- Upgraded (200 mA, 1.0 nm, UA, L=8.0 m)
- plus assume XRF detector collects 30% of  $4\pi$ SR

dwel time [s]	resolution [nm]	scan time [h]
1	200	0.2
1	20	17.4
0.1	5	27.8

dwel time [s]	resolution [nm]	scan time [h]
1	200	0.2
0.03	20	0.5
0.002	5	0.6

***New Science***

***Materials Science***

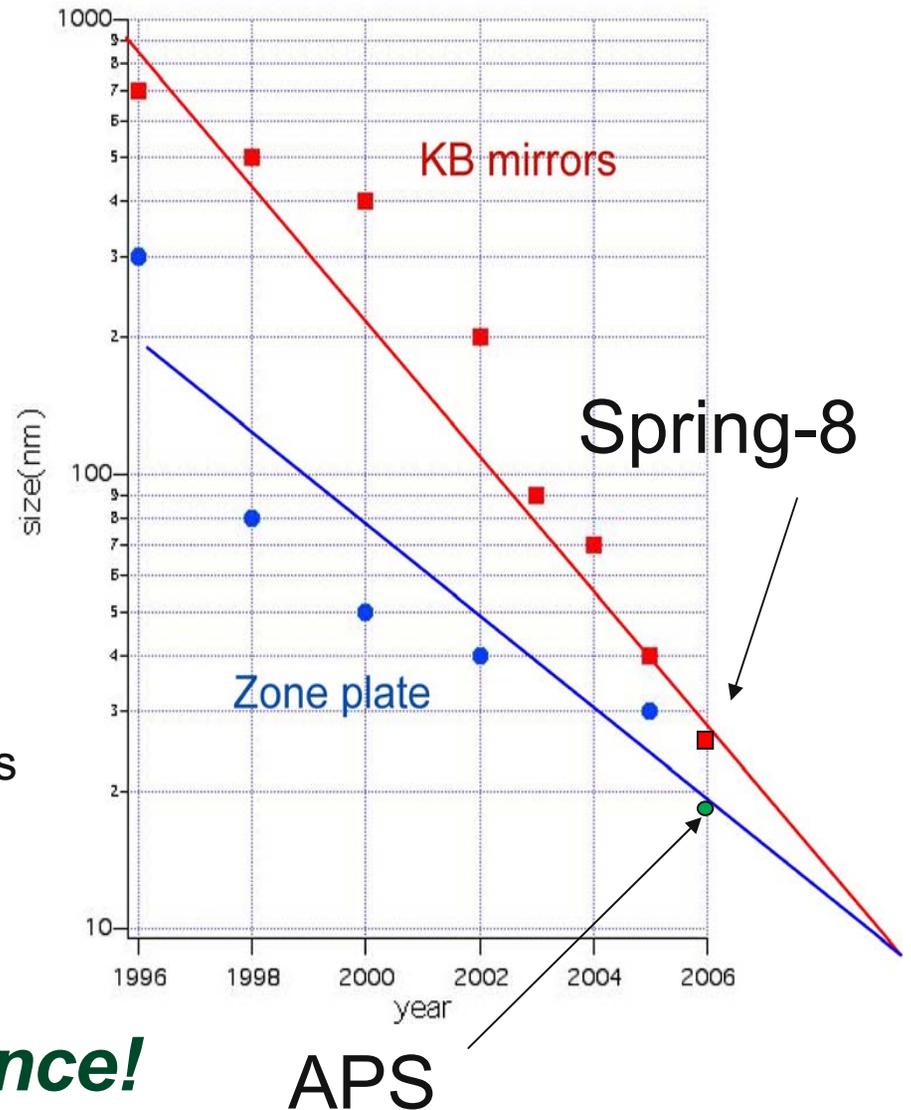
## Small beams change everything!

- Mapping!
  - Chemistry
  - Local structure
  - Heterogeneity
  - Defects/ correlations
- Combinatorial/ ease of sample preparation

3 motivations for 1-nm probe:

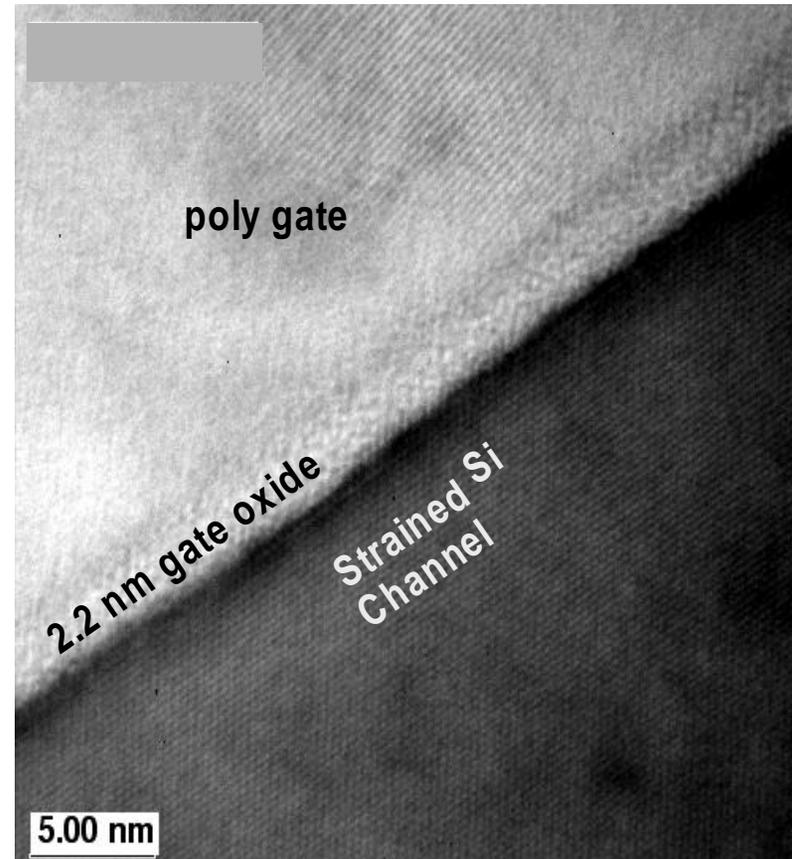
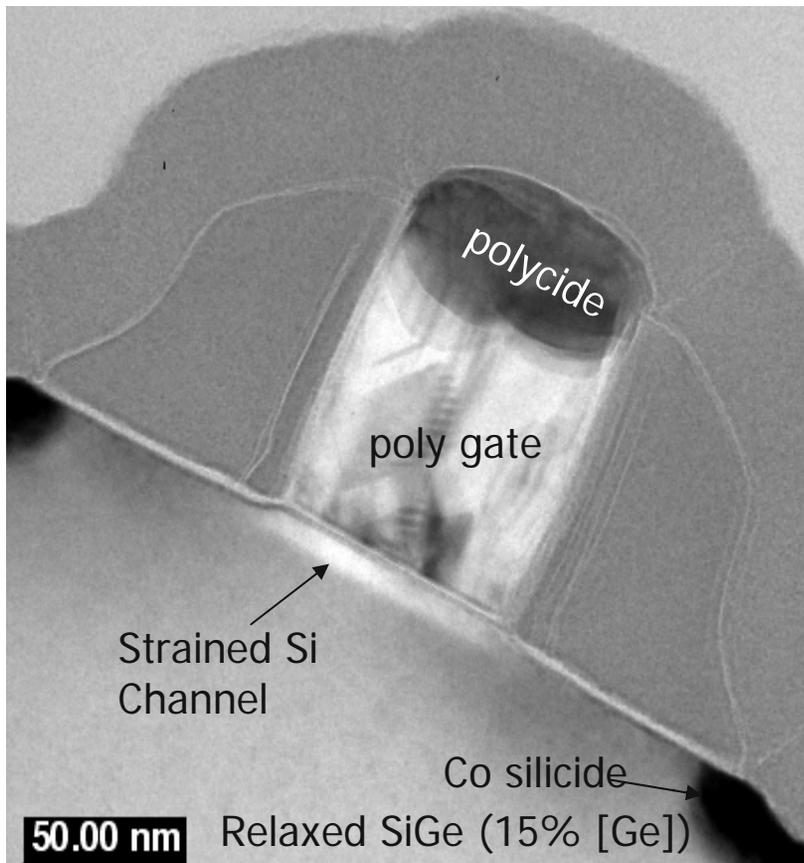
- Improve signal-to-background for individual nanoparticle
  - Isolate from matrix/ surrounding particles
- Resolve nano regions
- Look at small volumes below surface along boundaries etc.

**Probably essential for science!**



Slide adapted from Jon Tischler

## Cross-sectional TEM of Strained-Si NMOSFET



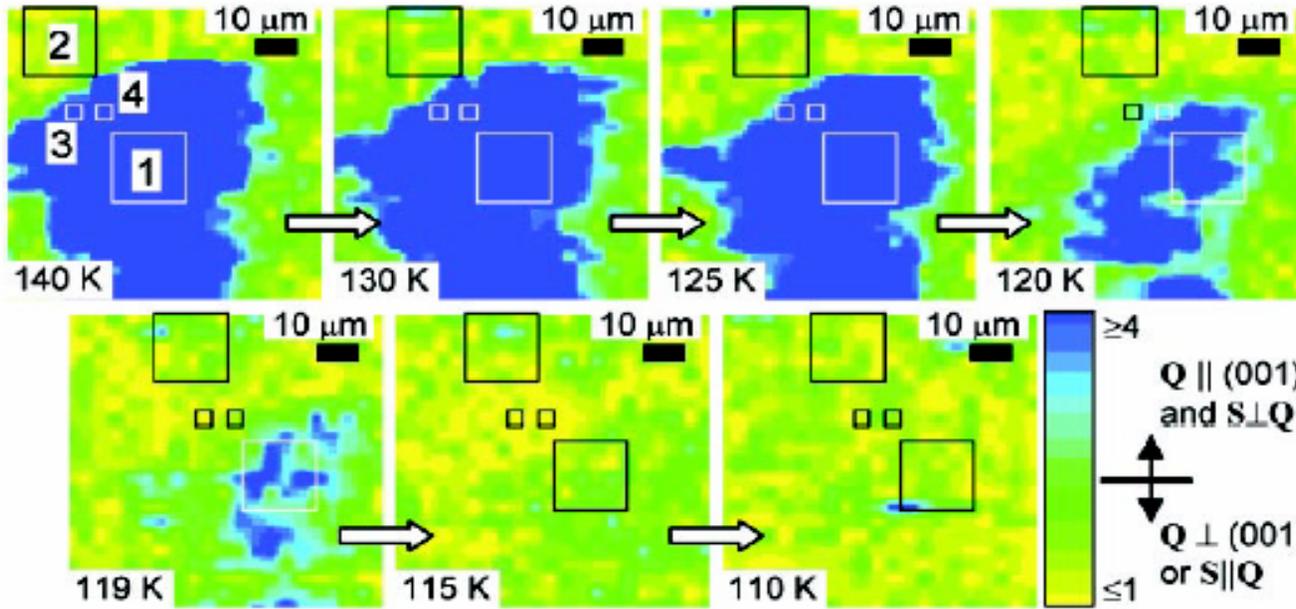
- Quality of epitaxial layers maintained during CMOS process steps
- Gate oxide with smooth interface formed by thermal oxidation

Ken Rim  
-IBM

- ⇒ today's manufacturer's are already able to produce nm scale structures.
- ⇒ X-rays are a method choice to probe such materials (nondestructive, buried layers, in situ...)
- ⇒ But: need x-ray beam of the same order of magnitude as structure size.

Slide courtesy Cev Noyan, with modification

# Antiferromagnetic Domain Evolution in Chromium

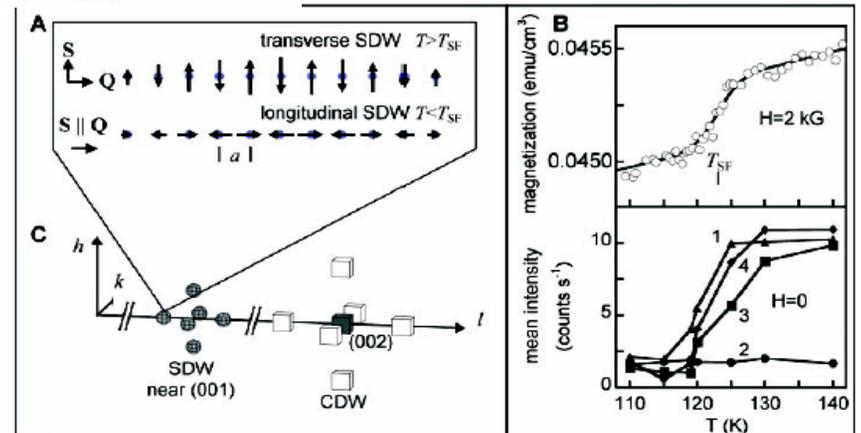


P.G. Evans, E.D. Isaacs, G. Aeppli, Z. Cai, B. Lai, *Science* 295, 1042 (2002).

Spin-flip transition  $T$  is non-uniform within domain

- Diffraction contrast allows imaging of regions with different magnetic order

To study and elucidate role of domain boundaries (few nm), need better spatial resolution!  
Or too look at smaller structures



Slide courtesy P. Evans

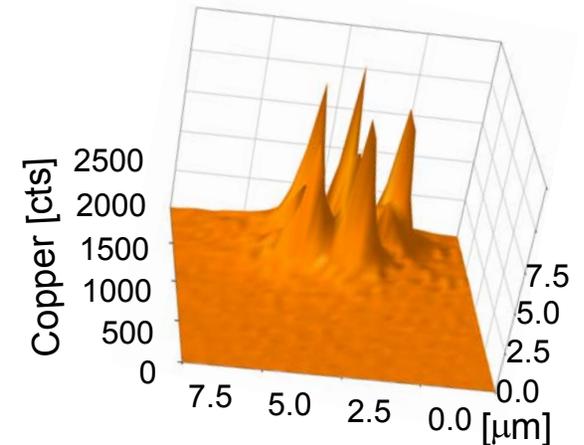
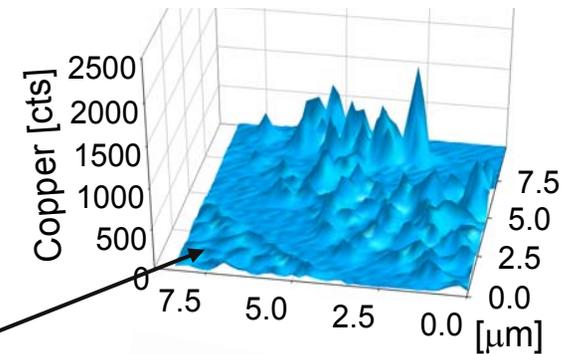
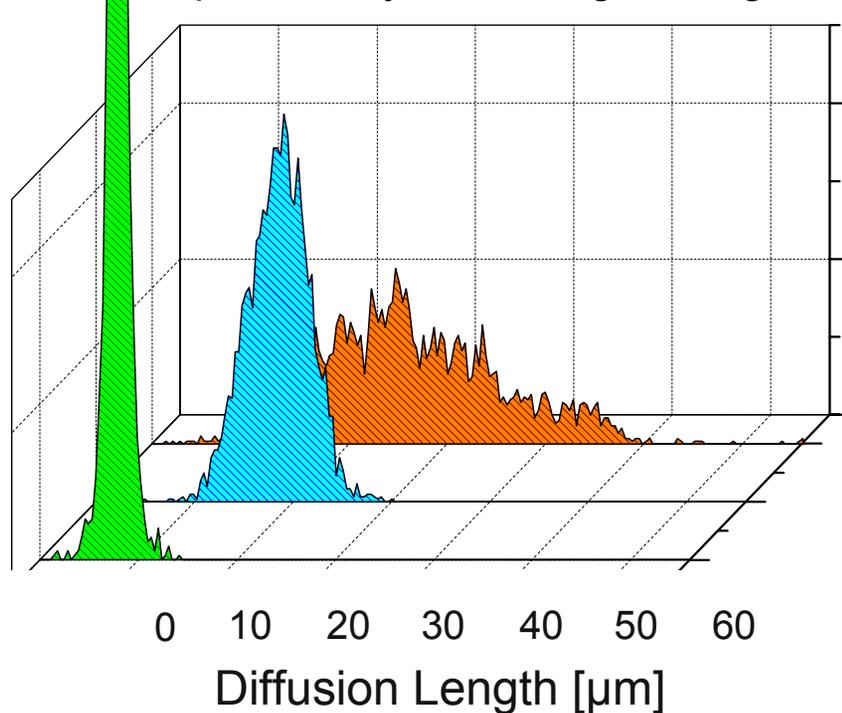
# Controlling metal-impurity nanodefects for low-cost solar cells (dirty silicon)

T. Buonassisi, et al.,  
*Nature Materials* **4**, 676-679 (2005)

Currently, 90% of photovoltaic devices used crystalline silicon (semiconductor grade); problem: limited availability, expensive

Alternative: Low-cost Metallurgical-grade Silicon Feedstock, but high impurity leads to high recombination activity, and thus low efficiency

- Problem is caused by metal impurities, affecting minority carrier diffusion length
  - Nanoprecipitates (10s nm) very detrimental
  - inclusions (up to 25  $\mu\text{m}$ ) seem O.K.
- Address problem by defect engineering



- But: to fully characterise the system, need to be able to study single nanoprecipitates
- Currently, this is not possible because of lacking spatial resolution and focussed flux

***New Science***

***Environmental Science & Geoscience***



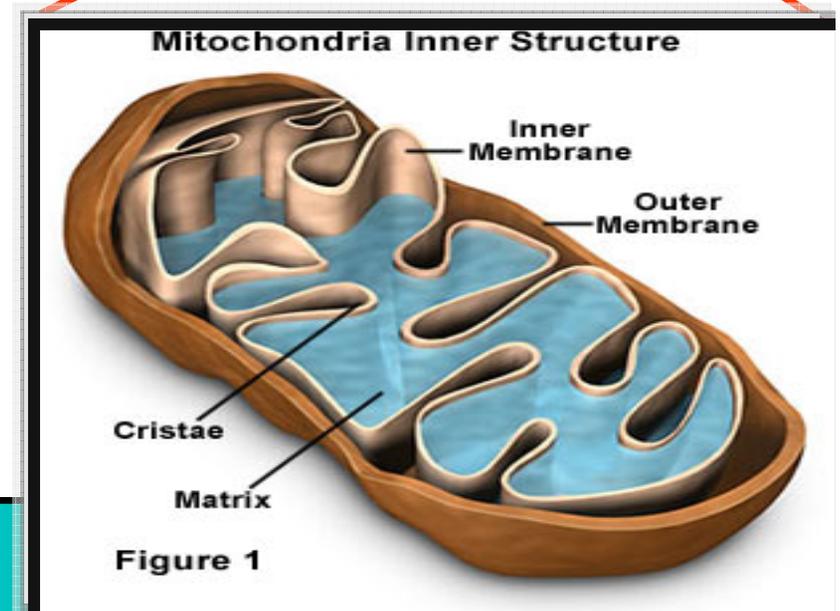
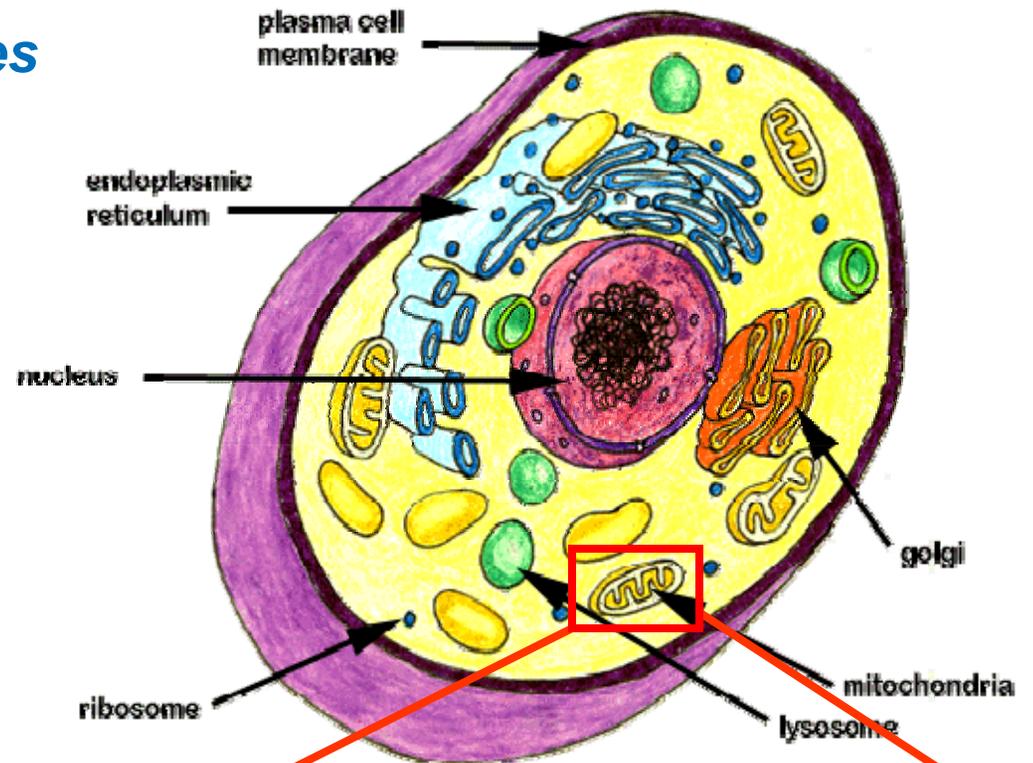
***New Science***

***Biology & Life Sciences***

## a 'typical' cell, and its structures

Typical sizes of cell structures and organelles:

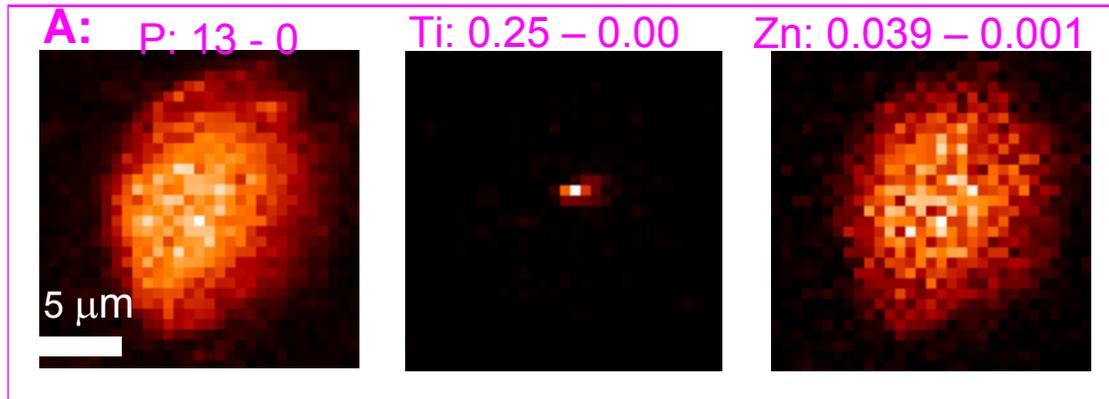
- nucleus: 2-5  $\mu\text{m}$
- **mitochondrion: 0.5x2  $\mu\text{m}$  (cellular respiration), w/ substructure !**
- ribosome: 25 nm (protein synthesis from mRNA)
- chromatin fiber: 20 nm diam. (DNA double helix on histones)
- microtubuli: 20 nm diam. (cytoskeleton)
- membrane thickness: 8 nm



## Visualizing single nanoparticles in cells

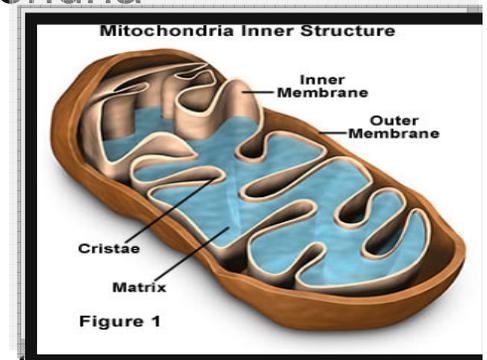
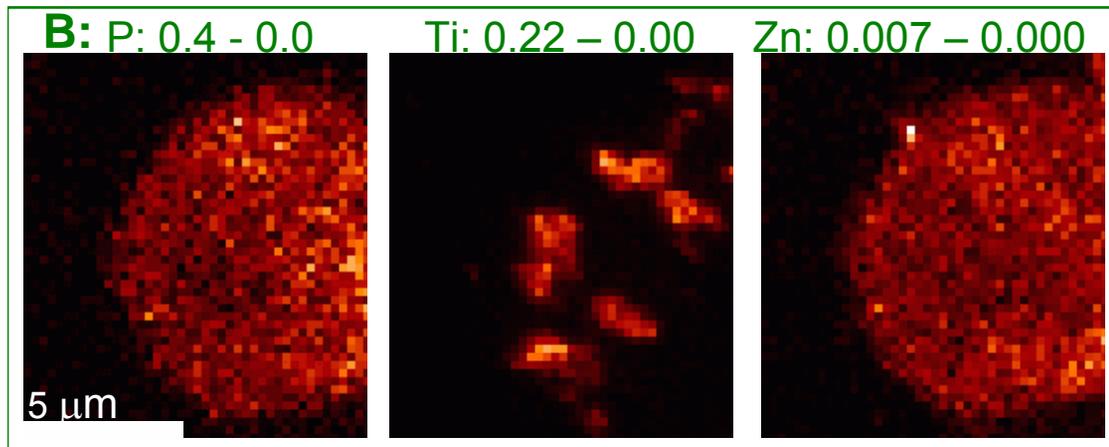
- Numerous developments to create functional nanocomposites that **combine** properties for
    - imaging (in its application to humans, e.g. Gd as a contrast agent for MRI)
    - therapy (e.g., TiO<sub>2</sub> with photo-induced cleavage of DNA)
    - targeting (e.g., sequence specific DNA, visualize via optical fluorescence)
  - But: before being able to test on subject, need to confirm in vitro:
    - Do the nanocomposites enter the cells ?
    - Do they 'find' the right target ?
    - Do they ONLY interact with the right target (e.g., toxicity) ?
    - Do the different components remain joined ?
- ⇒ Need to be able to find and localise a single nanocomposite with 'just a few' active metals.
- ⇒ For sufficient sensitivity, need small (<20nm) beam
- ⇒ To determine localisation precisely, need <10nm beam (8nm membrane double layer)
- ⇒ Need to image several whole cells (10x10 μm<sup>2</sup>), ideally tomographically

## TiO<sub>2</sub>-DNA nanocomposites as intracellular probes



Units: μg/cm<sup>2</sup>

- **A:** scan of a MCF7 cell transfected with nanocomposites targeted to nucleolus
- **B:** scan of a PC12 cell transfected with nanocomposites targeted to mitochondria



### Promising Future: Nanocomposites as tools for Gene therapy ?

- Correct defective genes responsible for disease development, e.g.,  
*destroying mutated and dominant genes (e.g., oncogenes)*

*But: need to be able to RESOLVE cellular targets of nanocomposites, to determine specific localisation, and ability to 'see' single nanocomposites*

T. Paunesku *et al*, Nature Materials 2003

## Probing inter-membrane spaces

- Membranes are interfaces between different environments
- In many infectious diseases, a parasite is engulfed by a vacuole, which becomes a battleground. The parasite needs to sequester nutrients from it, the cell may try to 'starve' the parasite, or poison it, by adjusting the local metal content.
- Currently we canNOT resolve this space, the ability to do so could improve our understanding of mechanisms of infectious diseases significantly (as well as our understanding of basic cell biology)

Invasion of red blood cells by *Plasmodium falciparum* (Malaria parasite)

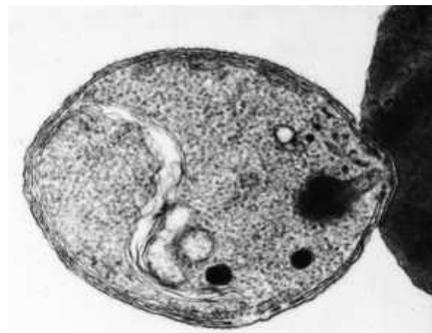
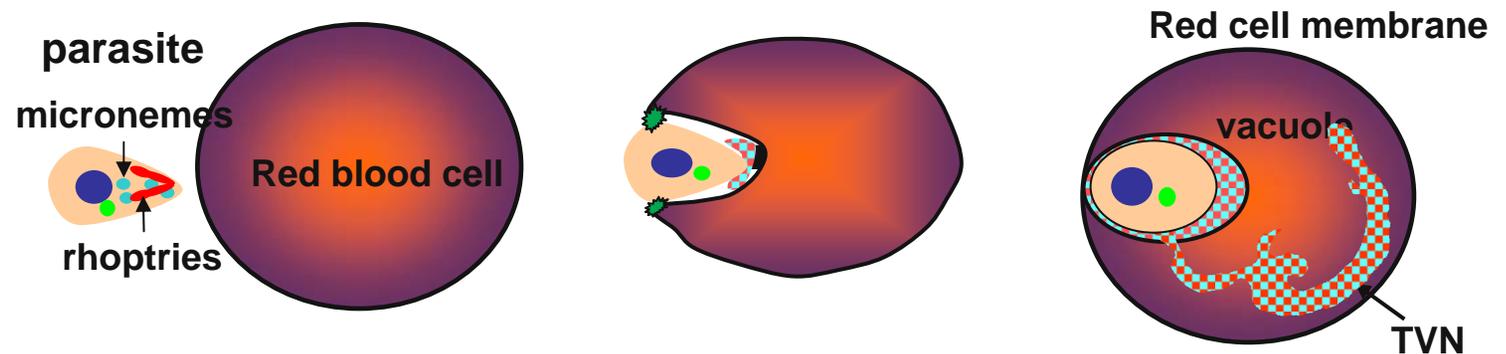


Illustration courtesy  
of Tom O'Halloran

## Summary: The APS upgrade & X-ray Microscopy

- Upgrade **is required**, to take scanning X-ray microscopy to the next level, and allow acquisition of high resolution data in realistic timeframes
- it **must be matched** by improvements at the beamlines
  - Optics (high spatial resolution, stability)
  - Detectors (large solid angle)
  - Data analysis (quantification, automation)
  - Correlative experiments with other techniques (IR, visible light, EM, ..)
  - (Specimen Preparation / Environment )
- But: shutdown time is problematic. A year is 'acceptable' but anything longer is highly problematic. (in particular for 'only' a factor 3 in coherence)
- Further reduction of emittance would be a HUGE boon for scanning microprobes
  - Directly further increase focussed flux density
  - Improve resolution, sensitivity

## ***New Science: mostly through higher resolution, sensitivity some examples are:***

- Material Sciences:
  - Investigate grain boundaries
  - Investigate nano-impurities
  - Characterise novel nanostructures, e.g., in applications for semiconductor industry
- Environmental and Geo-Sciences
  - Enable studies at high pressures and temperature that have been unattainable => new materials & phenomena
  - Allow study of metal-influenced process on and near bacterial surfaces => improve our understanding on how they interact and influence their environment
- Biology and Life Sciences
  - detect and map single nanovectors in cells and tissues (e.g., combine Gd base MRI contrast agent, with TiO<sub>2</sub>-DNA active component), correlate exactly to target (<=10nm) – currently impossible
  - Measure and compare cytoplasmatic (host cell), vesicular, as well as the elemental content of parasites, to significantly improve our understanding of infectious diseases
  - Potential to detect and localise individual metalloproteins in cells, providing an exciting new tool for cell/molecular biology

*Thank you for your attention !!*