

# Science with X-rays and Neutrons

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UCSD

PAC-11 NYC 2011

# What are particle accelerators good for?

- Solving the structure of the atom and the nucleus
- Unraveling the secrets of the Universe
- Finding the Higgs Boson
- Transmuting radioactive materials
- Medical applications
- Providing Neutron and X-ray Beams for studying the properties of materials !!!

# Partnerships

- IPNS Bob Kustom, Yang Cho, Bob Martin, Charlie Potts, Frank Brumwell, Jack Carpenter, David Price
- NSLS Ken Green, Rena Chasman, Ari van Steenburgen, Martin Blume
- APS John Galayda, Rod Gerig, Mike Borland, Glenn Decker, David Moncton, Gopal Shenoy
- LCLS John Galayda, Keith Hodgson

# Yes I Do Smile on Occasion



$$S(Q, \omega) \sim [1 - \exp(-\beta \omega)]^{-1} \operatorname{Im} \chi(Q, \omega)$$

$$S(Q) = \int d\omega S(Q, \omega) = |\langle \rho(Q) \rangle|^2$$

# Brightness & Fluxes for Neutron & X-Ray Sources

	Brightness ( $s^{-1}m^{-2}ster^{-1}$ )	dE/E (%)	Divergence ( $mrad^2$ )	Flux ( $s^{-1}m^{-2}$ )
Neutrons	$10^{15}$	2	$10 \times 10$	$10^{11}$
Rotating Anode	$10^{20}$	0.02	$0.5 \times 10$	$5 \times 10^{14}$
Bending Magnet	$10^{27}$	0.1	$0.1 \times 5$	$5 \times 10^{20}$
Undulator (APS)	$10^{33}$	10	$0.01 \times 0.1$	$10^{24}$

# Neutron Advantages

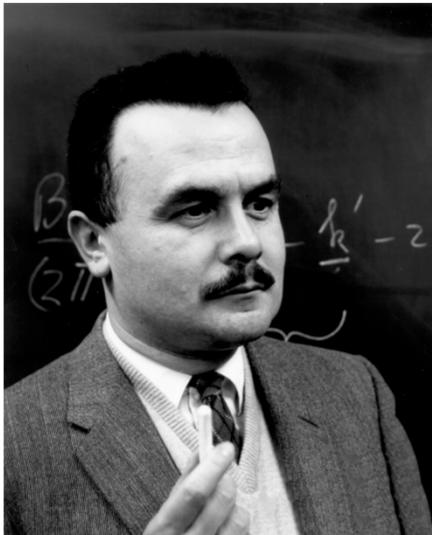
- Penetrating, but does no damage to sample
- H/D contrast matching can be used to study macromolecules in solution, polymers, etc.
- Strongly interacts with magnetic moments
- Energies match those of phonons, magnons, rotons, etc.

# Nobel Prize in Physics, 1994



Awarded for “pioneering contributions to the development of neutron scattering techniques for studies of condensed matter”

Bertram N. Brockhouse



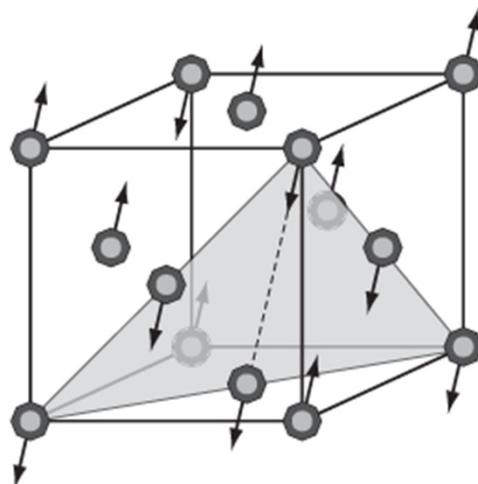
Development of  
neutron spectroscopy

Clifford G. Shull

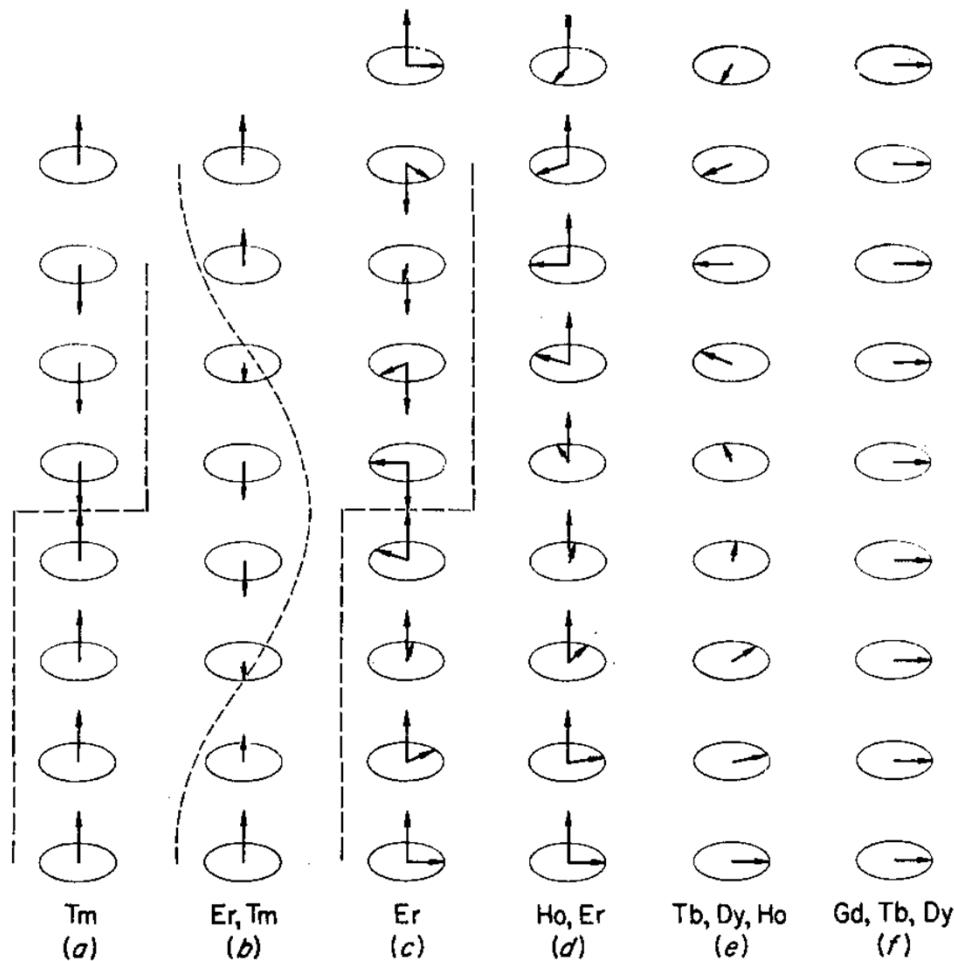


Development of the  
neutron diffraction technique

# First Study of an Antiferromagnetic Structure



Antiferromagnetic Structure of MnO  
(Shull and Wollan Phys. Rev. 83, 333 (1951))



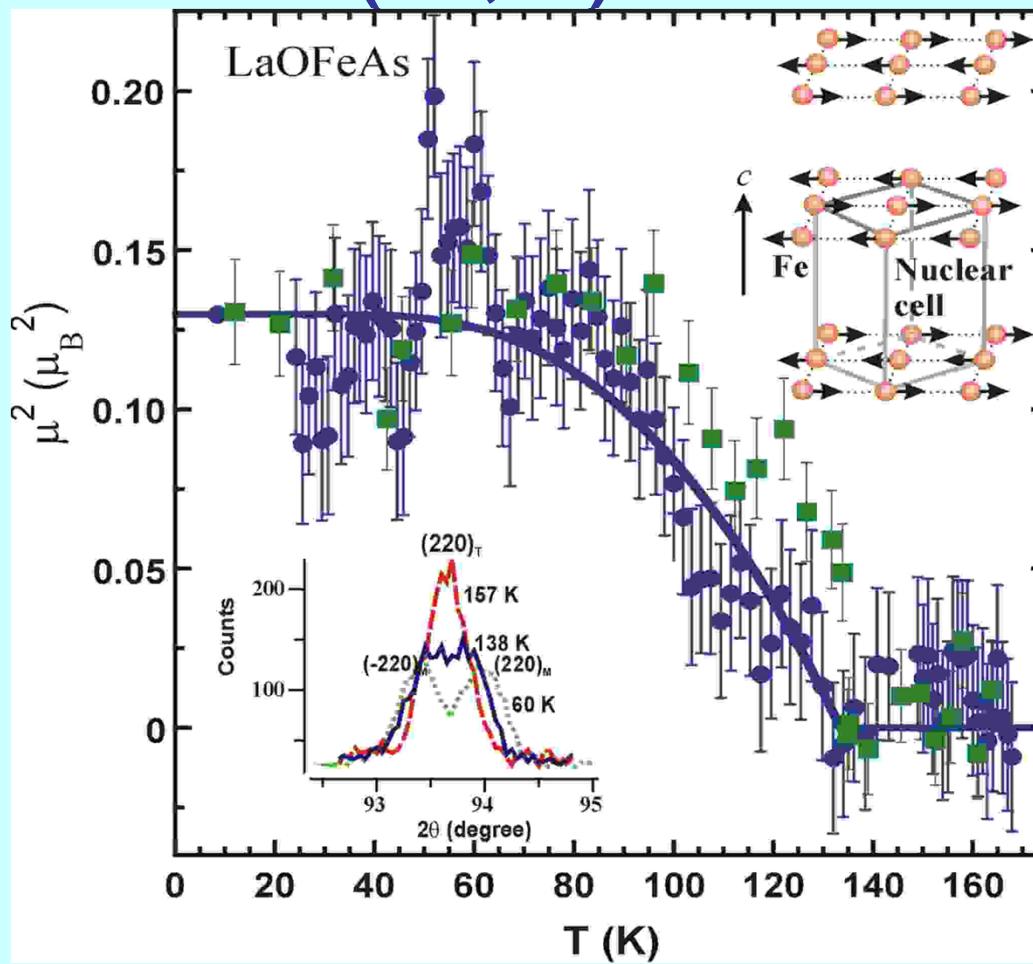
Magnetic Structure of the Rare Earth Metals  
(W.C. Koehler (1965))

QuickTime™ and a  
decompressor  
are needed to see this picture.

# Spin Structure of Cu ions in La<sub>2</sub>CuO<sub>4</sub>

Vaknin et al., PRL 1987

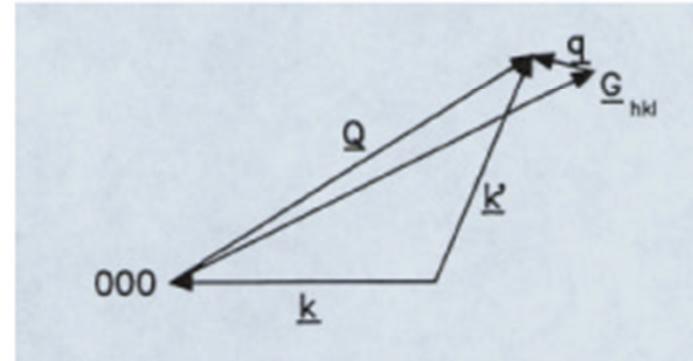
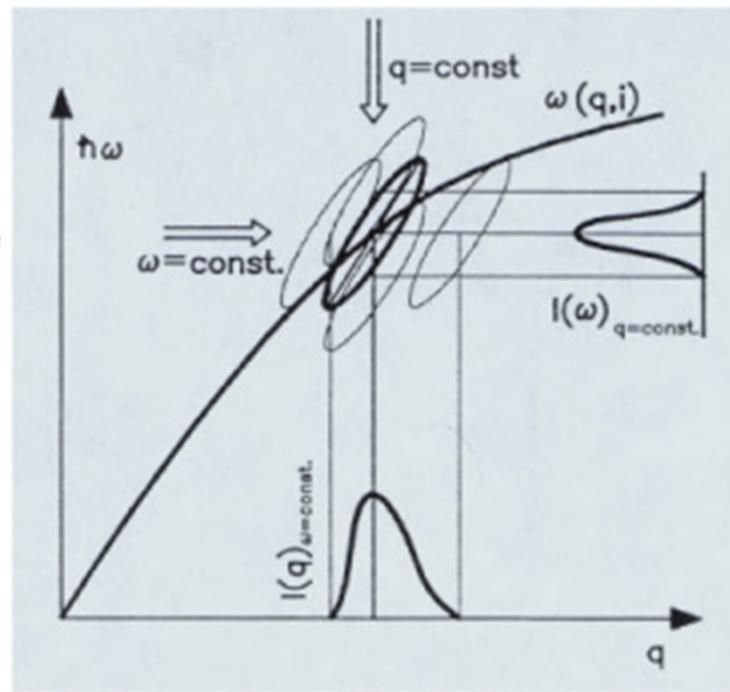
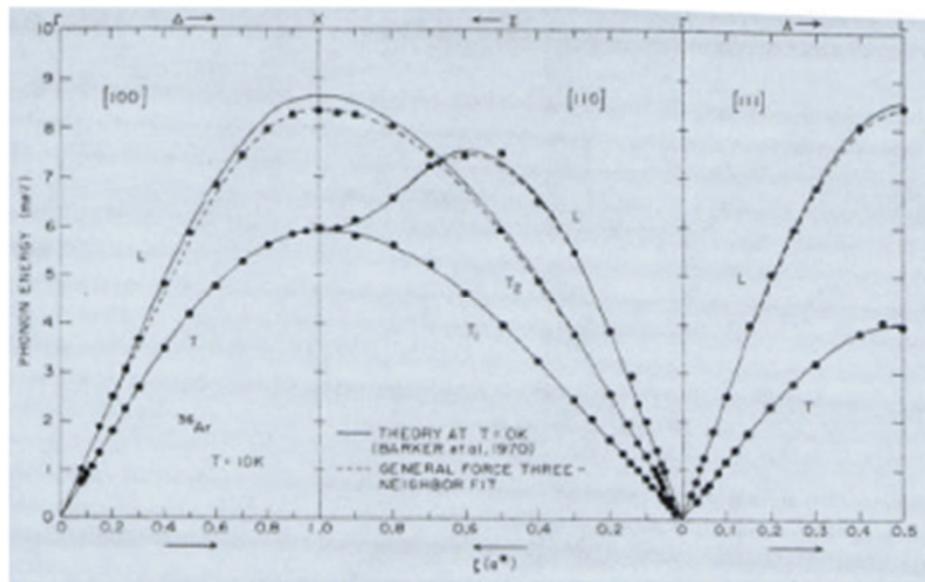
# Magnetic Structure La(O,F)FeAs



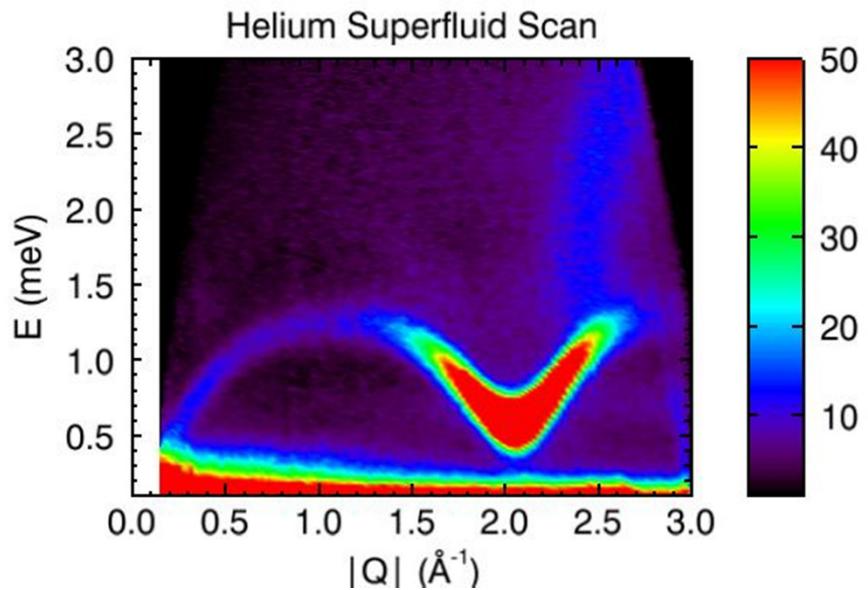
Magnetic Order Close to Superconductivity in the Iron-based Layered  $\text{La}(\text{O}_{1-x}\text{F}_x)\text{FeAs}$  systems, C. de la Cruz, Q. Huang, J. W. Lynn, J. Li, W. Ratcliff II, J. L. Zarestky, H. A. Mook, G. F. Chen, J. L. Luo, N. L. Wang, and P. Dai, P. Dai, *Nature* **453**, 899 (2008).

# Triple Axis Spectrometers Have Mapped Phonons Dispersion Relations in Many Materials

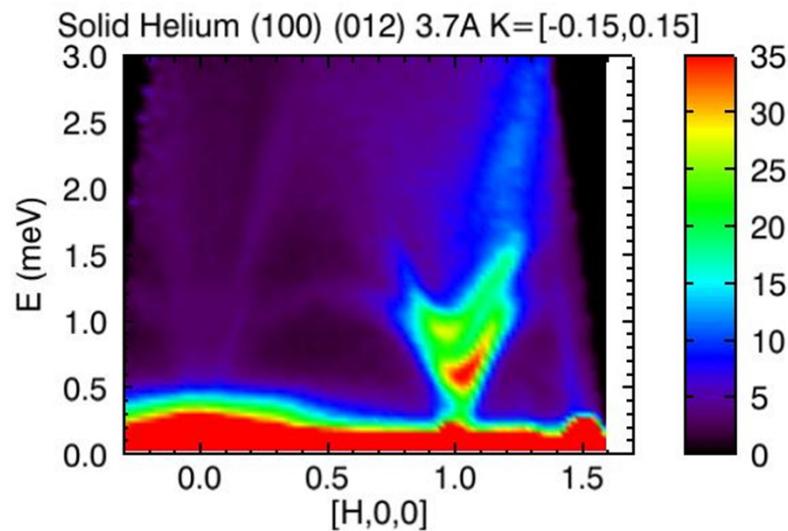
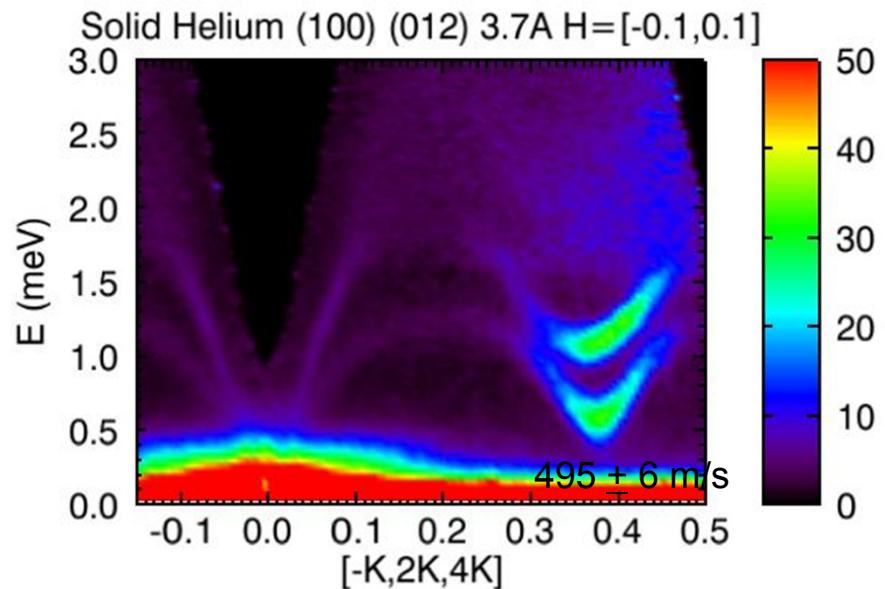
- Point by point measurement in  $(Q, E)$  space
- Usually keep either  $k_{\parallel}$  or  $k_F$  fixed
- Choose Brillouin zone (i.e.  $G$ ) to maximize scattering cross section for phonons
- Scan usually either at constant- $Q$  (Brockhouse invention) or constant- $E$



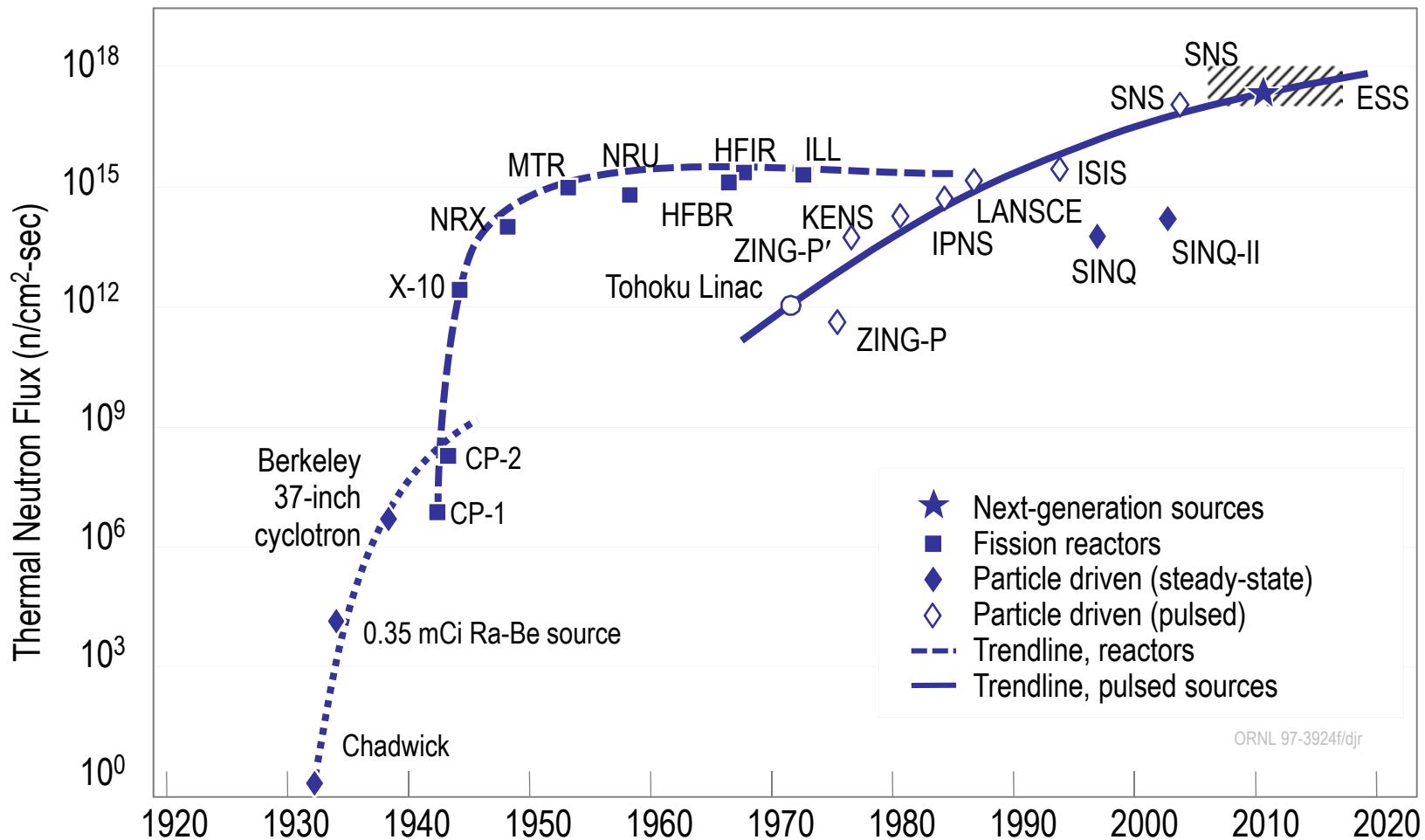
## Superfluid at melting Point



## Solid and liquid at melting



# We have come a long way since 1932



(Updated from *Neutron Scattering*, K. Skold and D. L. Price: eds., Academic Press, 1986)

# SNS Accelerator Complex

Front end:  
Produce  
1 ms long,  
chopped  
 $H^-$  beam

1 GeV linac

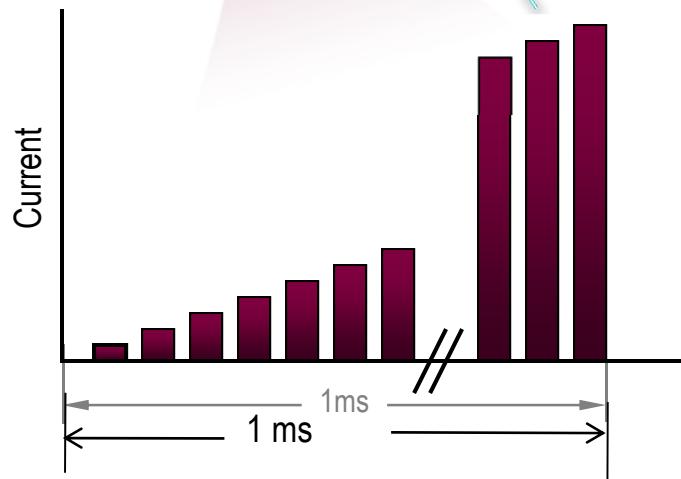
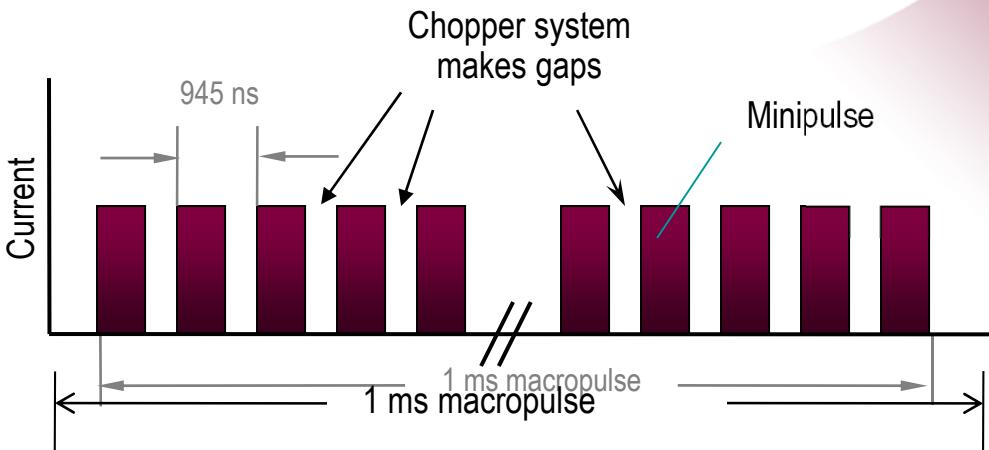
2.5 MeV

Front end

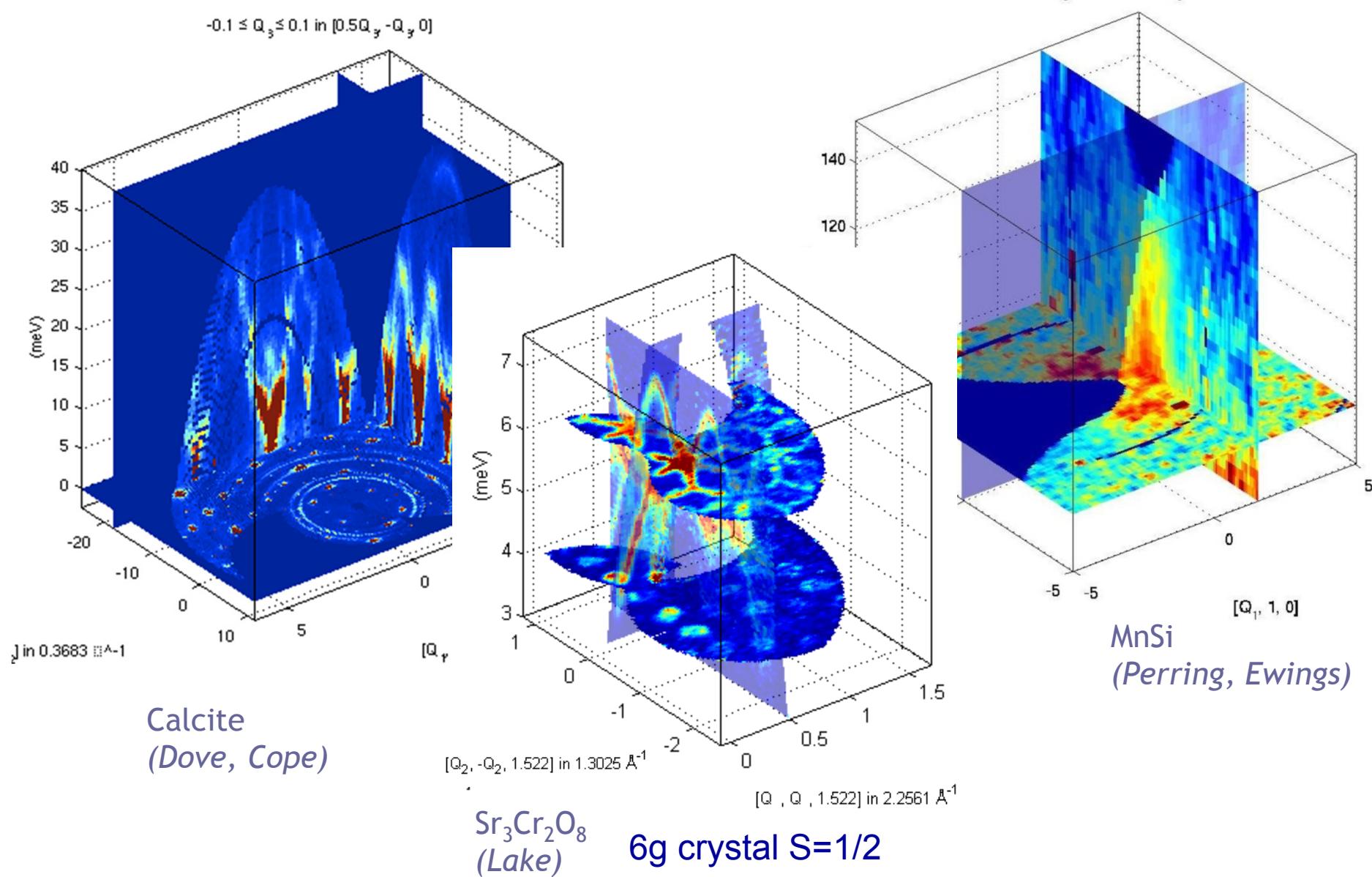
Linac

1000 MeV

Accumulator ring:  
Compress  
1 ms pulse  
to 700 ns



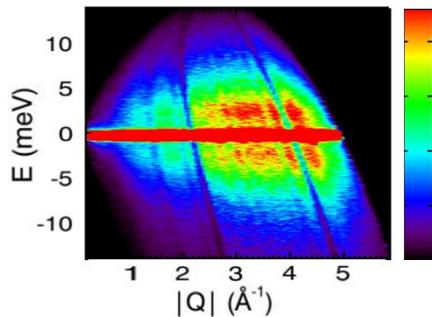
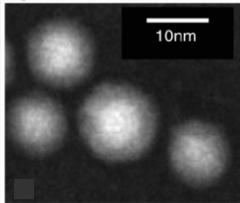
# Complete measurements of $S(Q, \omega)$ measurements on MERLIN spectrometer at ISIS



# Impacting a broad range of science

## Nanoscience

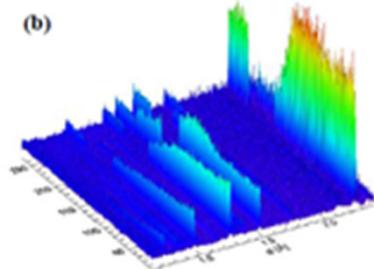
First observations  
of conventional spin  
waves  
in nanoparticles



Feygenson et al.,  
*Phys. Rev. Lett.* (submitted)

## In situ imaging for industry

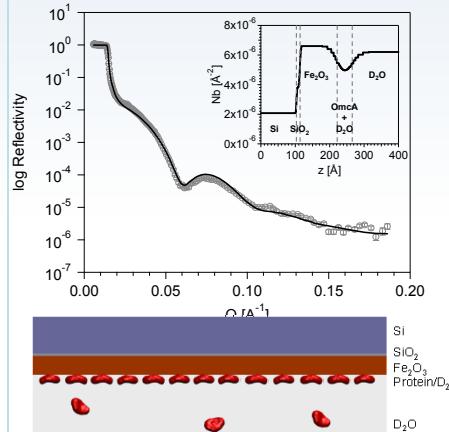
Providing industry  
with a look inside real  
materials



Ma et al.,  
*Scripta Mat.* (submitted)

## Bioremediation

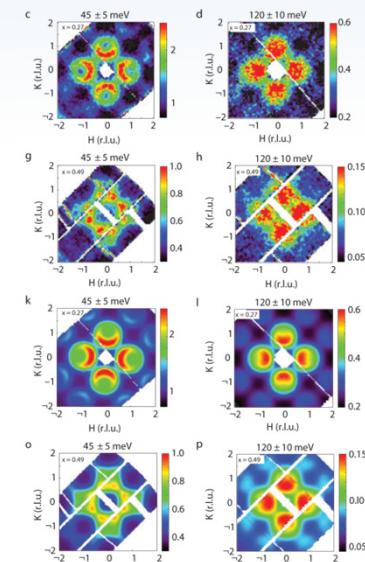
Probing molecular  
interactions between  
microbial-cell protein  
and mineral surfaces



Johs et al., *Biophys. J.*  
98, 3035 (2010)

## Superconductivity

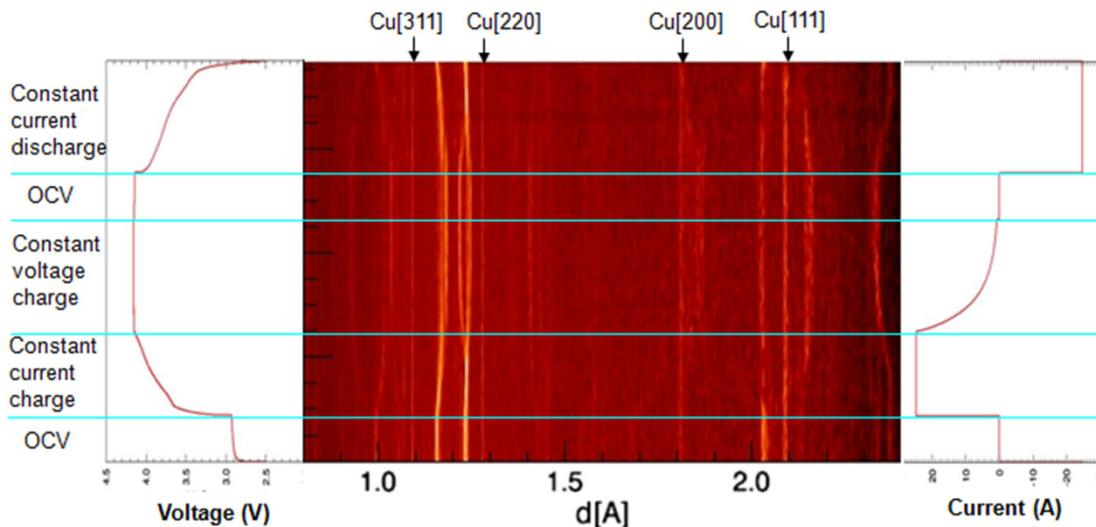
Evolution of spin  
excitations  
in superconductors:  
Strong evidence that  
superconductivity is  
related  
to magnetic interactions



Lumsden et al, *Nature Phys.*  
6, 182 (2010)

# Pushing the limits: Single pulse diffraction

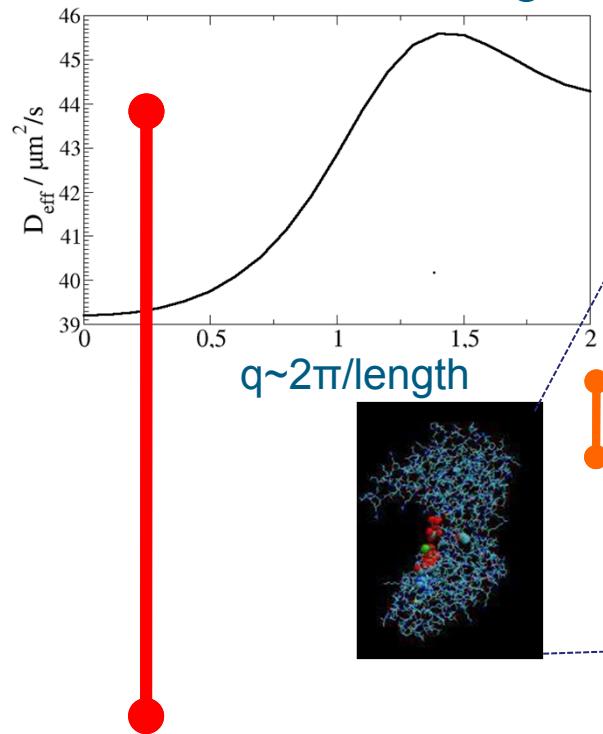
- In-situ study of structure evolution of Li-ion battery
  - Structure changes are clearly established
  - Time resolution for the present data  $\sim 2$  min, but this could easily be reduced to seconds
  - A new phase forms and dissolves during charge-discharge; more detailed analysis is under way
  - Time-resolved spatial mapping is possible



# Functional domain dynamics in proteins

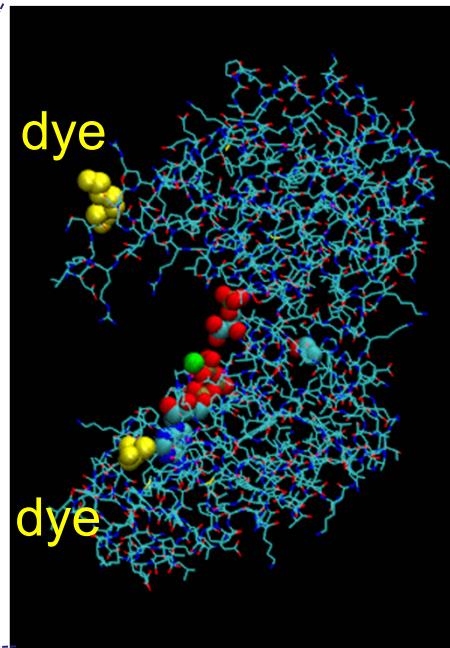
## NSE

0.5-50 nm length scale  
ps -  $\mu$ s time scale  
orientational average



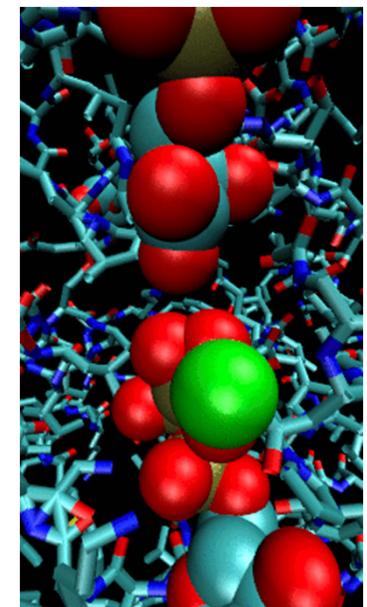
## FRET

fixed defined position  
 $> \mu$ s timescale



## NMR

ps - ms timescale  
small proteins



phosphoglycerate kinase

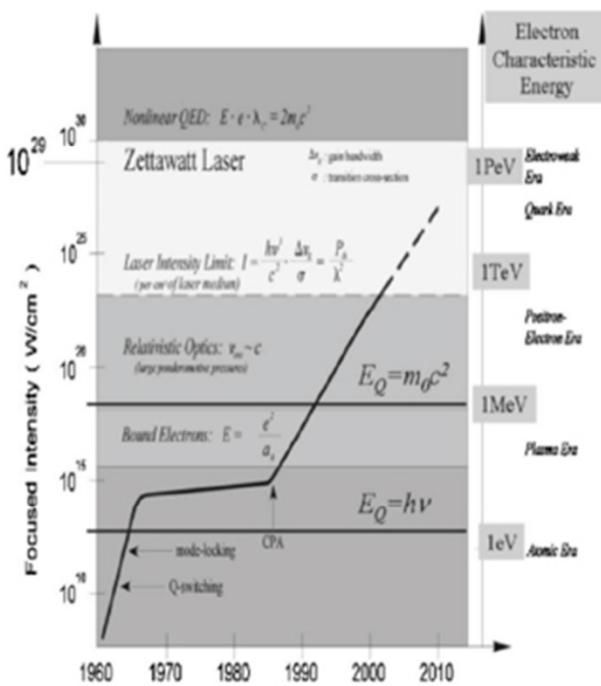
# Science with X-Rays

- Diffraction and crystal structures
- Structure Factors of liquids and glasses
- Structures of Thin Films
- ARPES
- EXAFS, XANES
- Studies of Magnetism with resonant XMS
- Inelastic X-ray scattering: phonons, electronic excitations
- X-ray Photon Correlation Spectroscopy
- Microscopy
- Imaging/Tomography

# Compare the evolution of high intensity optical and x-ray sources

## High-intensity at optical wavelengths

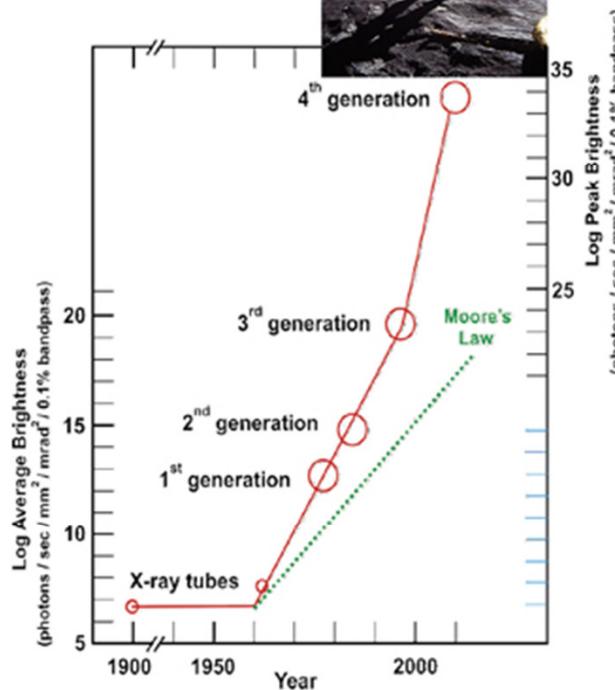
- high harmonic generation
- tabletop coherent x-ray radiation
- attosecond pulses



G. Mourou RMP 2006

## High-intensity at x-ray wavelengths

- ?
- ?
- ?



D. Moncton, George Brown

# Example 1: X-Ray Diffraction & structural biology

- D.C. Phillips presents the 3-D structure of lysozyme to the Royal Society in **1965**
- Linear polypeptide chain
- Folded model of the same amino acid sequence
- July 2009:  
58,588 structures in  
Protein Data Bank



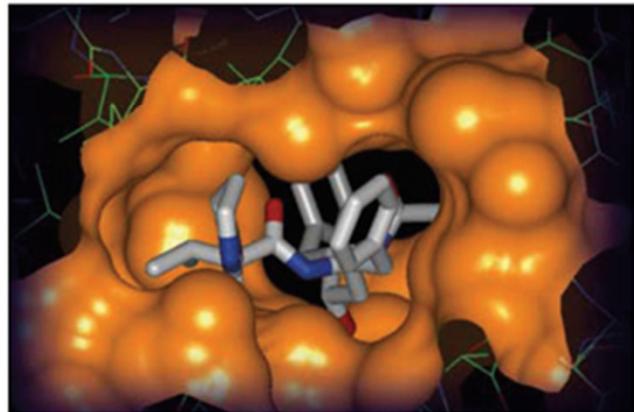
A single protein structure used to be the project of a scientific lifetime

Synchrotron Radiation - 8301 structures solved in 2009

# Synchrotron research on proteins has led to major advances in drugs to battle infection, HIV, cancer



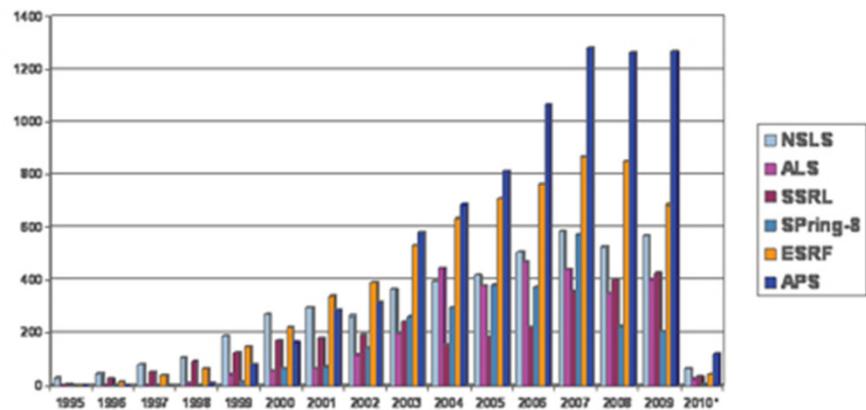
Renal cancer drug  
pazopanib™ developed in  
part based on APS  
research  
(GlaxoSmithKline)



Close-up view of the drug binding site  
within HIV protease ([Kaletra®](#), Abbott).



Ramakrishnan, Steitz and Yonath  
2009 Chemistry Nobel Laureates



APS protein structure  
output is almost twice  
that of any other light  
source



# X-rays dominant in protein structure determinations



## Snapshot: July 1, 2009

58,588 released atomic coordinate entries

### Molecule Type

- 54,141 proteins, peptides, and viruses
- 2,033 nucleic acids
- 2,381 protein/nucleic acid complexes
- 33 other

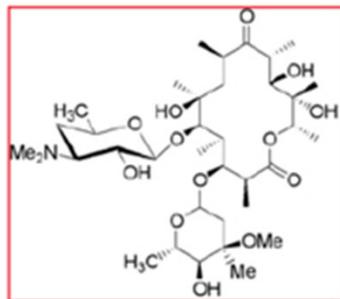
### Experimental Technique

- |        |                     |
|--------|---------------------|
| 50,284 | X-ray               |
| 7,914  | NMR                 |
| 243    | electron microscopy |
| 17     | hybrid              |
| 130    | other               |

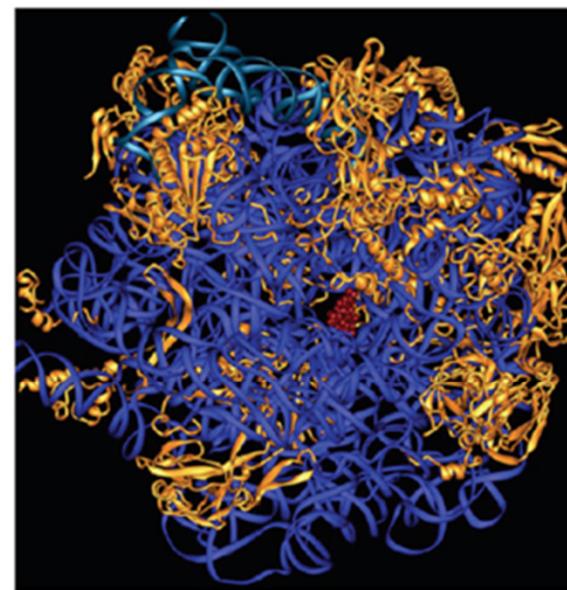
Year	Total Depositions
2000	2983
2001	3286
2002	3563
2003	4830
2004	5508
2005	6678
2006	7282
2007	8130
2008	7073
2009	8301
2010	1952
<b>TOTAL</b>	<b>59586</b>

# Designing antibiotics -

difference between bacterial and eukaryotic ribosomes is  
one amine group in the 2.5MD ribosome

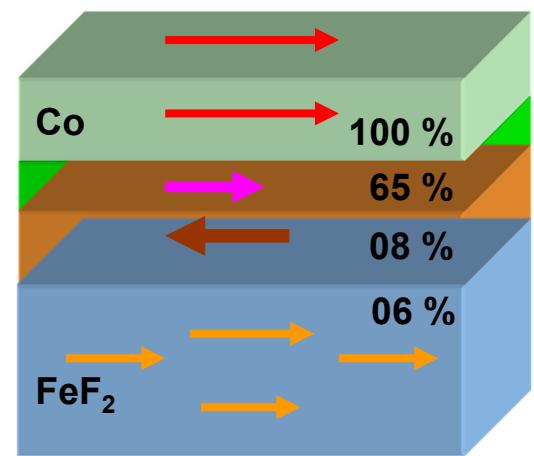
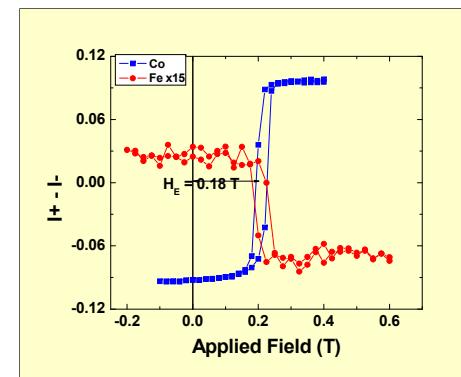
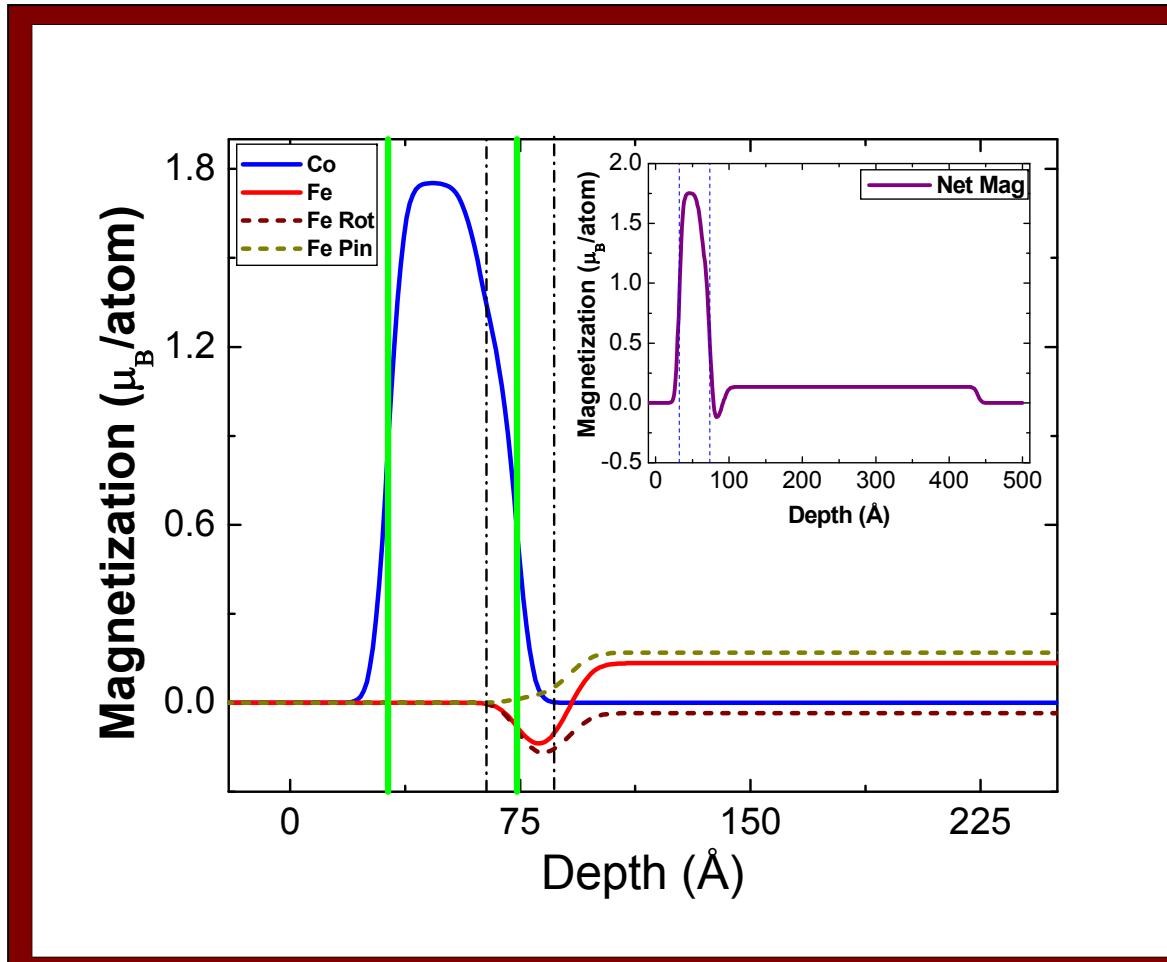


Erythromycin – a macrolide antibiotic that blocks protein synthesis by binding to bacterial ribosomes but not to eukaryotic ribosomes



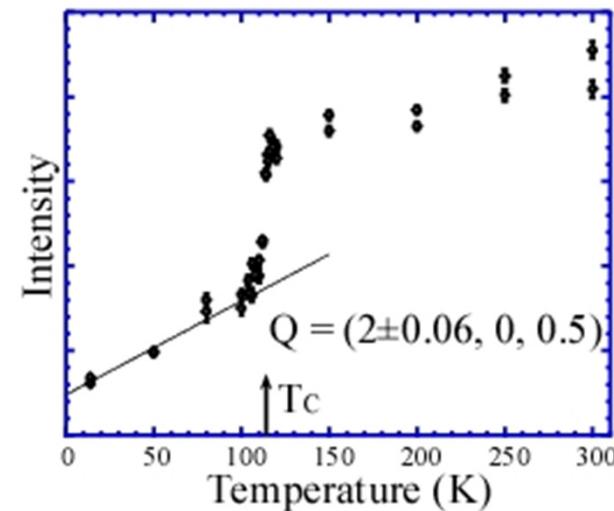
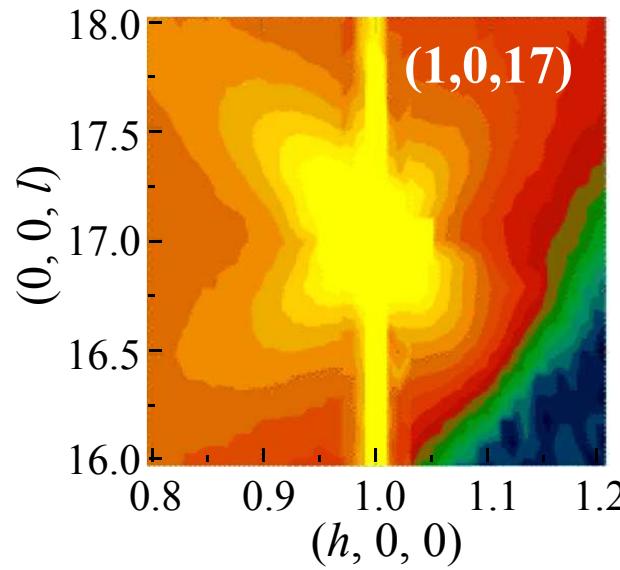
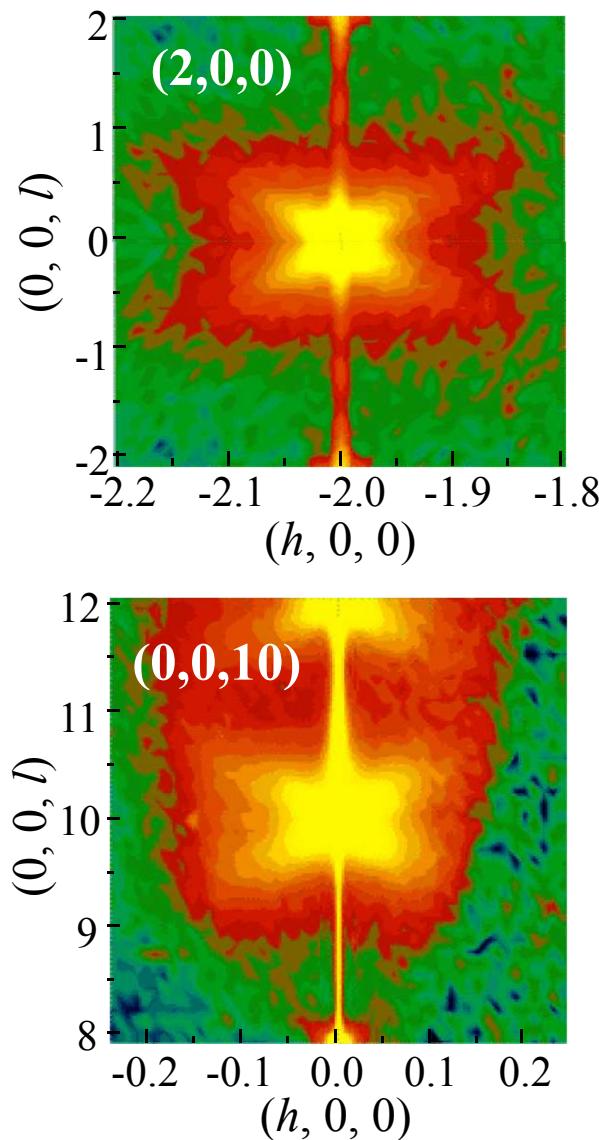
[www.molgen.mpg.de](http://www.molgen.mpg.de)

# Depth Dependent Magnetic Density Profile

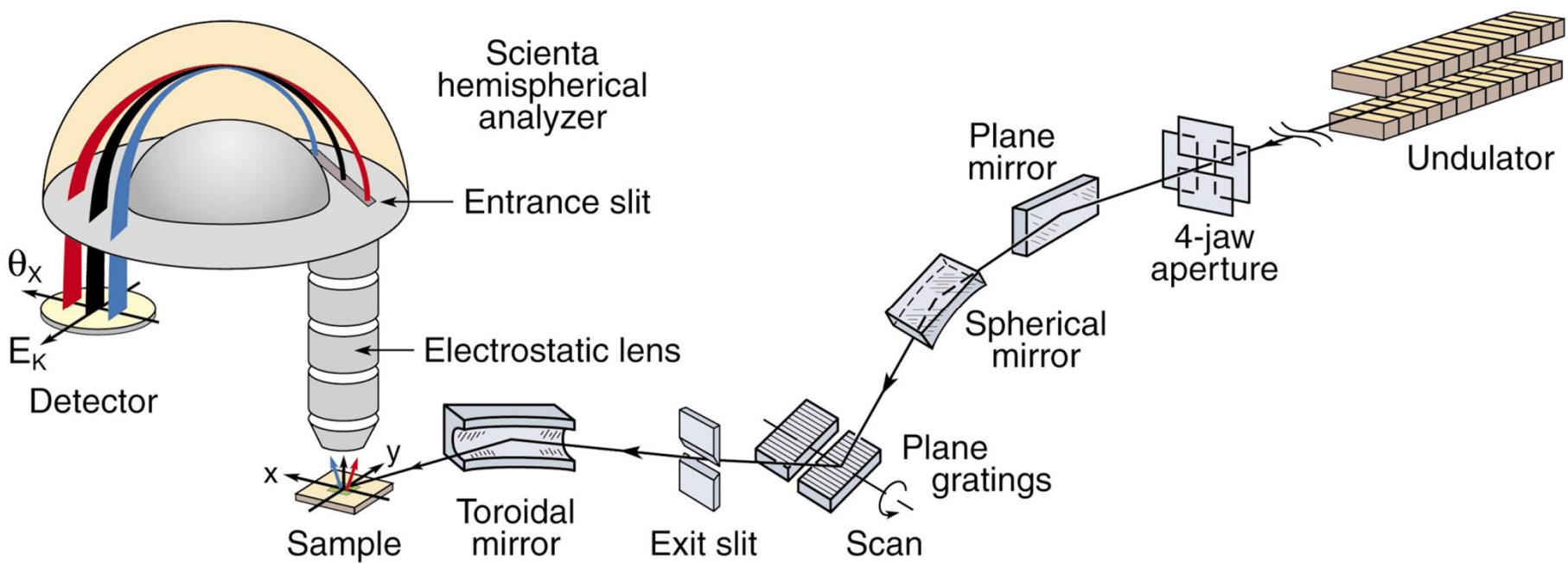


Exchange bias due to exchange interaction between Fe pinned and Fe rotating moments which is then mitigated across the Co/ $\text{FeF}_2$  interface  
Decreasing field is favorable to reversal and hence positive EB effect

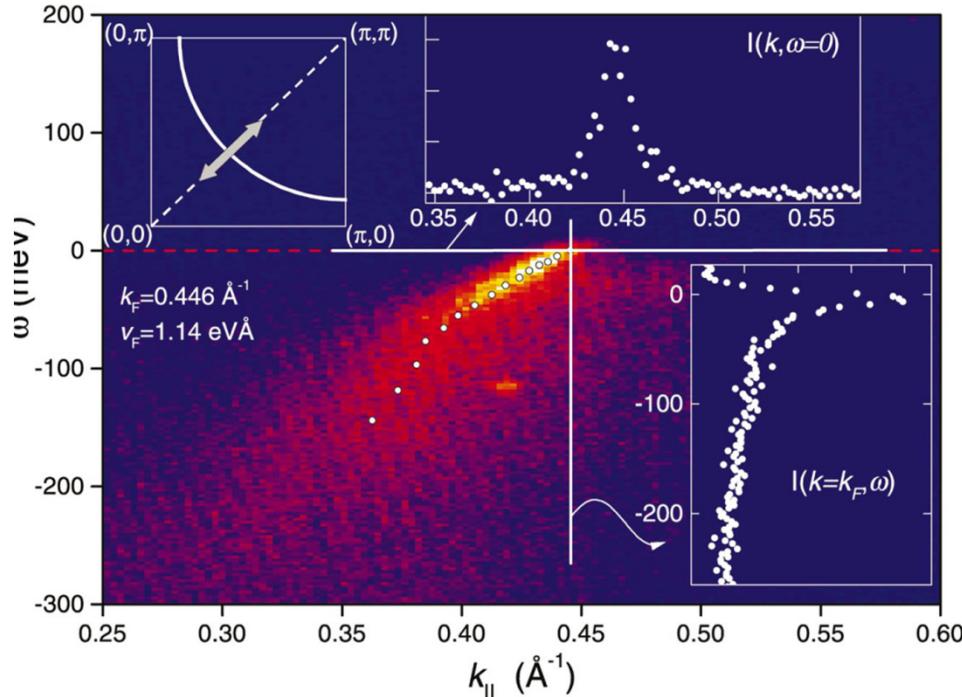
# Polarons via single-crystal diffuse scattering



L. Vasiliu-Doloc *et al.*, PRL 83, 4393 (1999).



IG. 6. Generic beamline equipped with a plane grating monochromator and a Scienta electron spectrometer (Color).



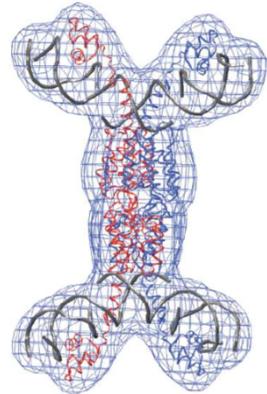
IG. 7. Energy ( $\omega$ ) vs momentum ( $k_{\parallel}$ ) image plot of the photoemission intensity from  $\text{Bi}_2\text{Sr}_2\text{Ca}\text{Cu}_2\text{O}_{8+\delta}$  along  $(0,0)$ - $(\pi,\pi)$ . This  $k$ -space cut was taken across the Fermi surface (see sketch of the 2D Brillouin zone upper left) and allows a direct visualization of the photohole spectral function  $A(\mathbf{k}, \omega)$  (although weighted by Fermi distribution and matrix elements). The quasiparticle dispersion can be clearly followed up to  $E_F$ , as emphasized by the white circles. Energy scans at constant momentum (right) and momentum scans at constant energy (upper right) define *energy distribution curves* (EDC's) and *momentum distribution curves* (MDC's), respectively. Afteralla, Fedorov, Johnson, Wells, *et al.*, 1999 (Color).

People want pretty pictures  
(in Space and Time!)

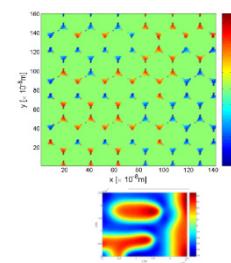
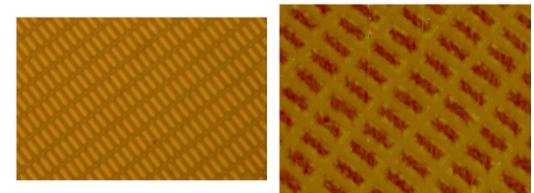
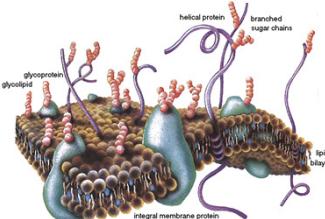
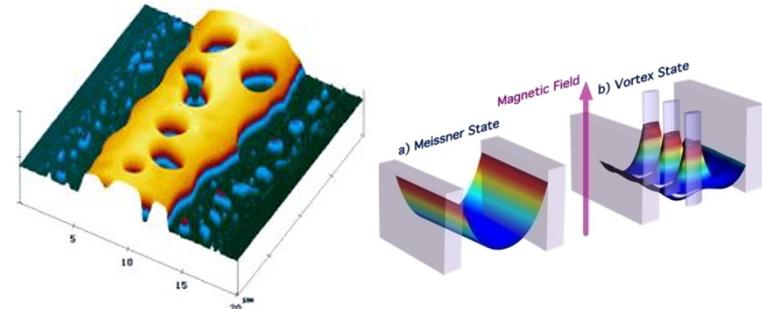
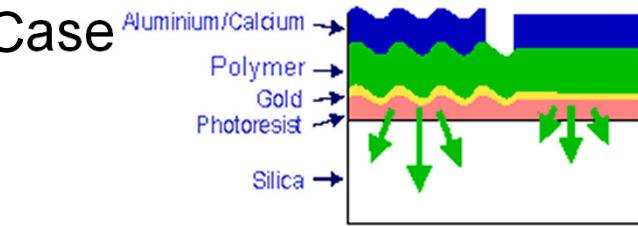
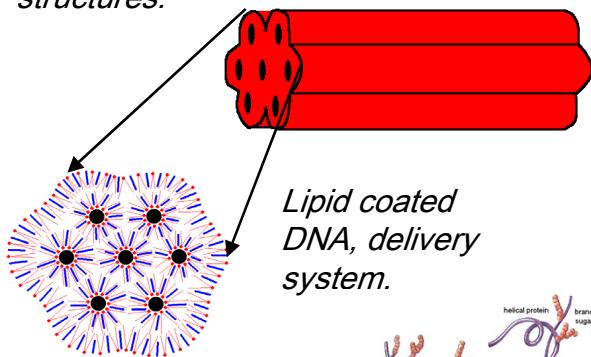
## ■ Advanced Materials

- Bio-Science

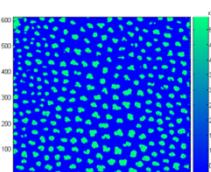
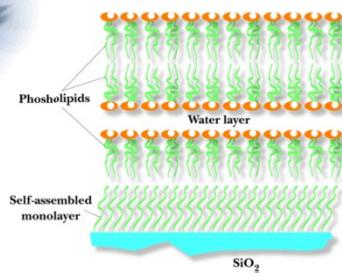
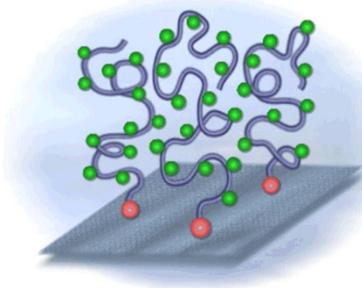
### The TS2 Science Case



*Overall size & shape of  
Tn3 resolvase (protein) – DNA  
complex, using neutron & X-ray  
contrasts plus known crystal  
structures.*



## ■ Soft Matter



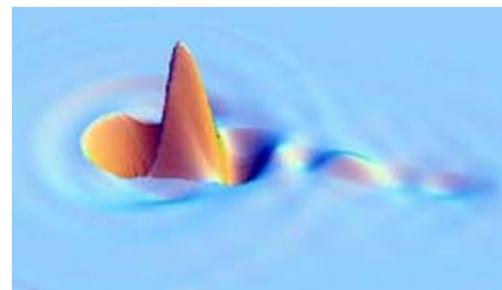
P. Abbamonte/Brookhaven National Lab [Phys. Rev. Lett. 92, 237401](#)

**Ripple effect.** Researchers used x rays to visualize the sloshing of electrons in water molecules.

They then calculated the wake of electron motion that would surround a gold ion moving through the fluid (above, click to see larger view).

See [animation](#) below. A research team has produced the fastest movies ever made of electron motion. Created by scattering x rays off of water, the movies show electrons sloshing in water molecules, and each frame lasts just 4 attoseconds

(quintillionths of a second). The results, published in the 11 June *PRL*, could let researchers "watch" chemical reactions even faster than those viewable with today's "ultrafast" pulsed lasers.



Phase Contrast Imaging

PEEM

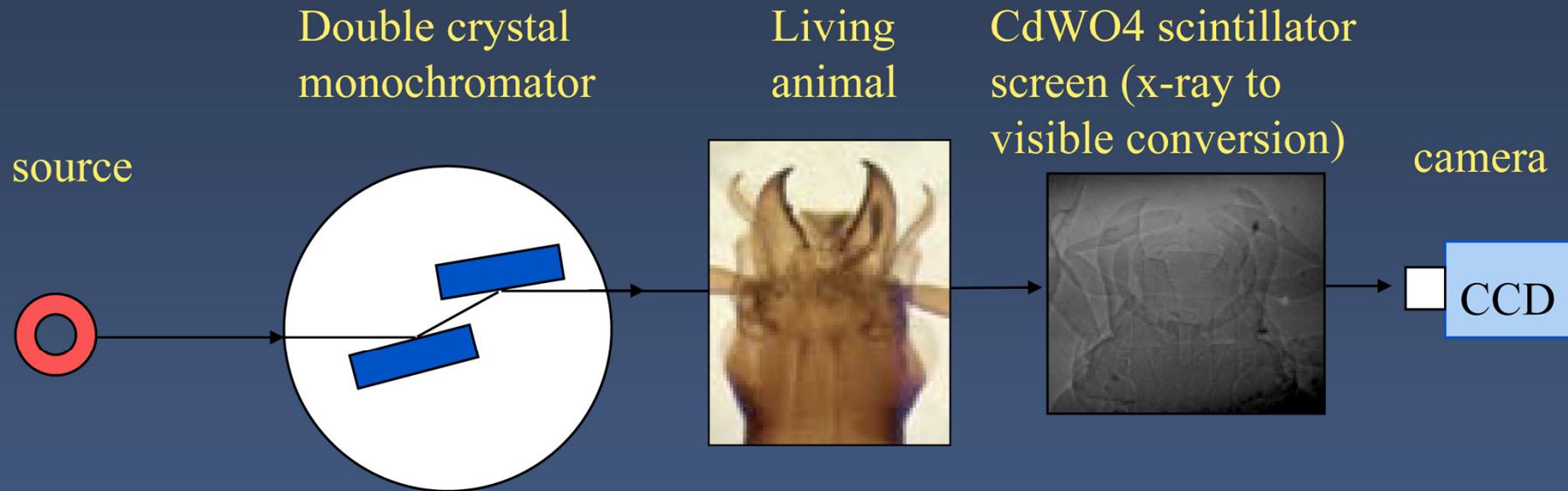
X-ray tomography

X-ray Microscopy/Fluorescence microscopy

X-Ray Holography

Phase retrieval imaging

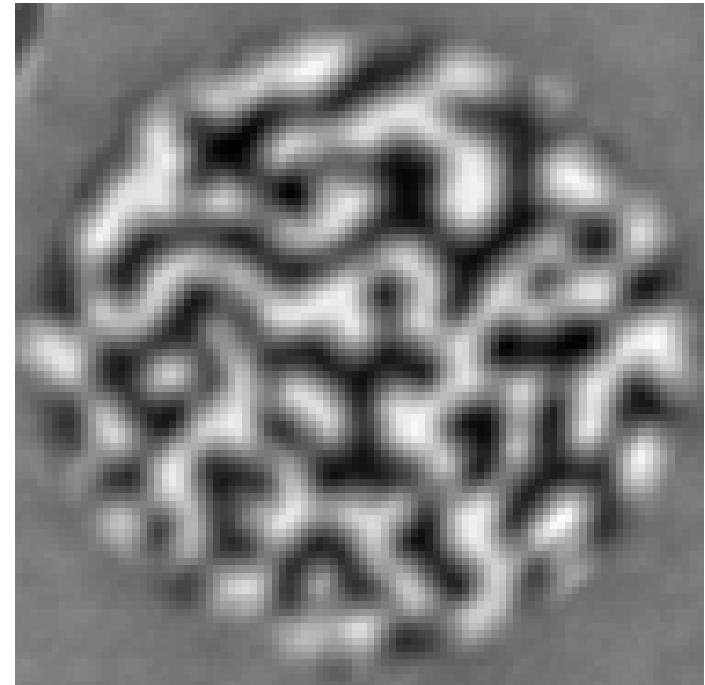
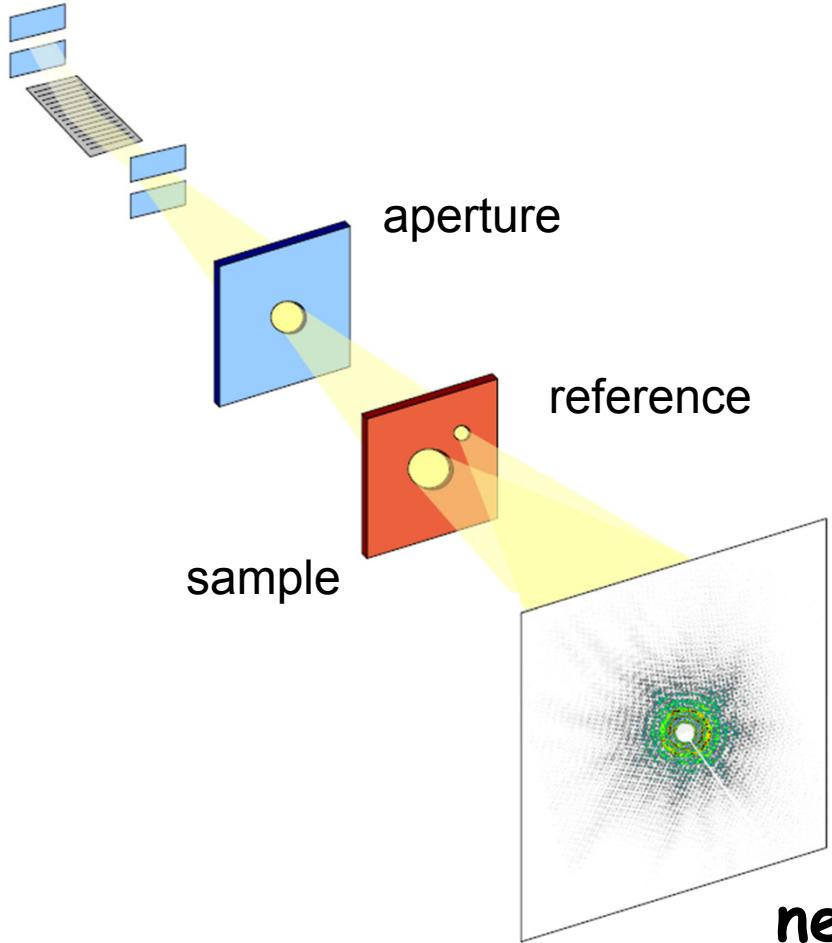
# Phase enhanced x-ray imaging:



X-ray images depend on differences in density within the sample.

Also: Beam coherence produces interference fringes (edge enhancement effects).

# Imaging via Soft X-ray Fourier Transform Holography



**needs circular polarized x-rays**

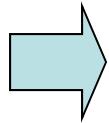
Method : S. Eisebitt et al. Nature 432, 885 (2004)

Resolution: L. M. Stadler et al. PRL, 100, 245503 (2008)

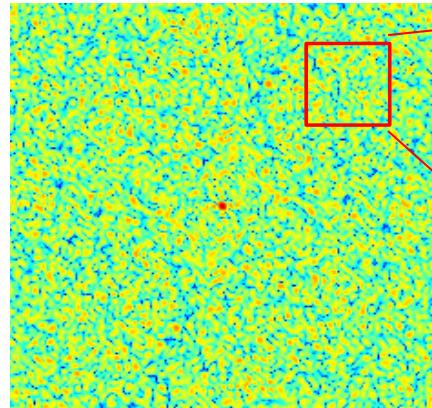
# Photon Correlation Spectroscopy+

Brownian Motion of 100 particles

QuickTime™ and a Video decompressor are needed to see this picture.



Diffraction Pattern

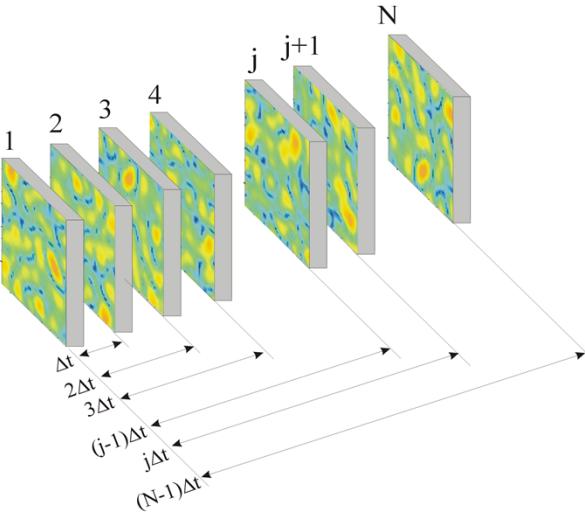
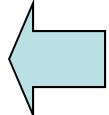
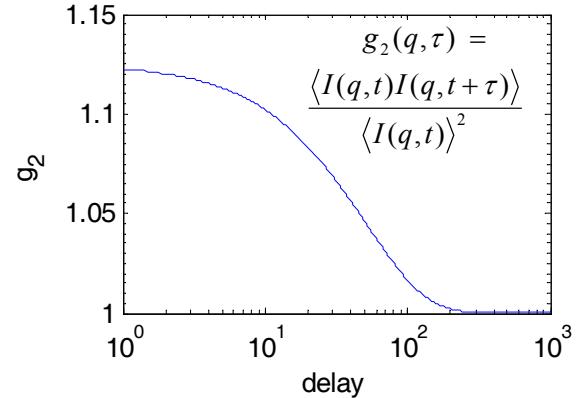


Speckles

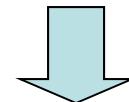
QuickTime™ and a Video decompressor are needed to see this picture.

Intensity-intensity auto correlation

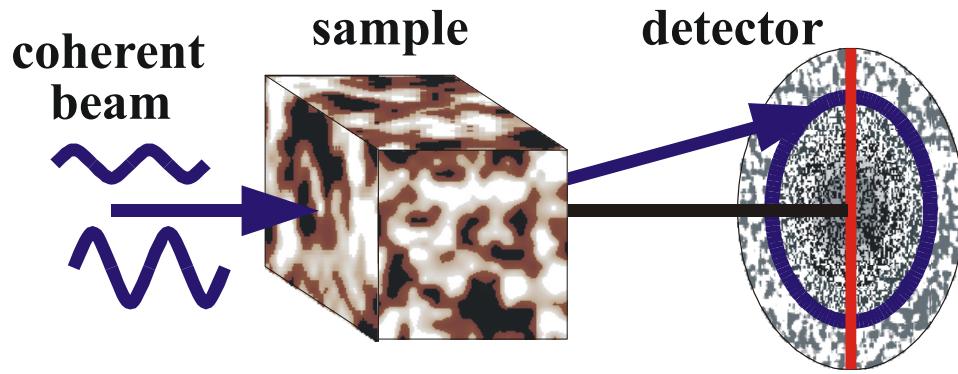
$$g_2(q, \tau) = \frac{\langle I(q, t)I(q, t + \tau) \rangle}{\langle I(q, t) \rangle^2}$$



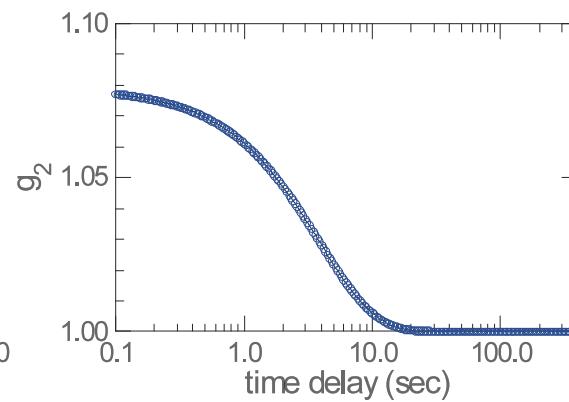
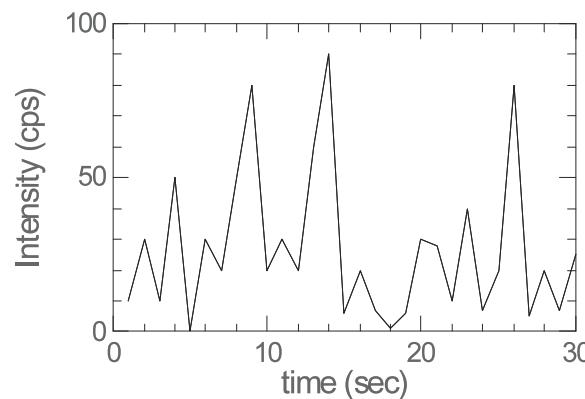
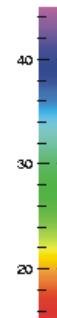
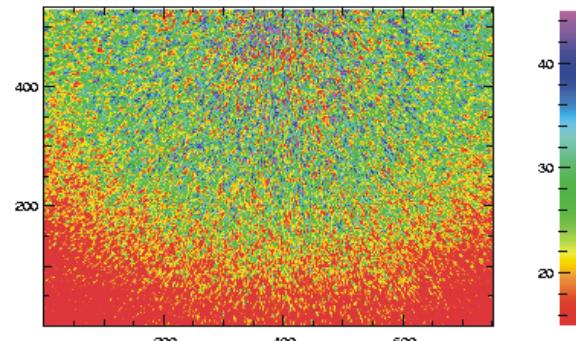
QuickTime™ and a Video decompressor are needed to see this picture.



# Photon Correlation Spectroscopy



X-ray speckle pattern from a static silica aerogel



$$g_2(\mathbf{q}, t) = \frac{\langle I(\mathbf{q}, t') I(\mathbf{q}, t' + t) \rangle}{\langle I(\mathbf{q}, t') \rangle^2}$$

$$\begin{aligned} g_2(t) &= 1 + \beta \exp(-2\Gamma t) \\ &= 1 + \beta \exp(-2t/\tau) \end{aligned}$$

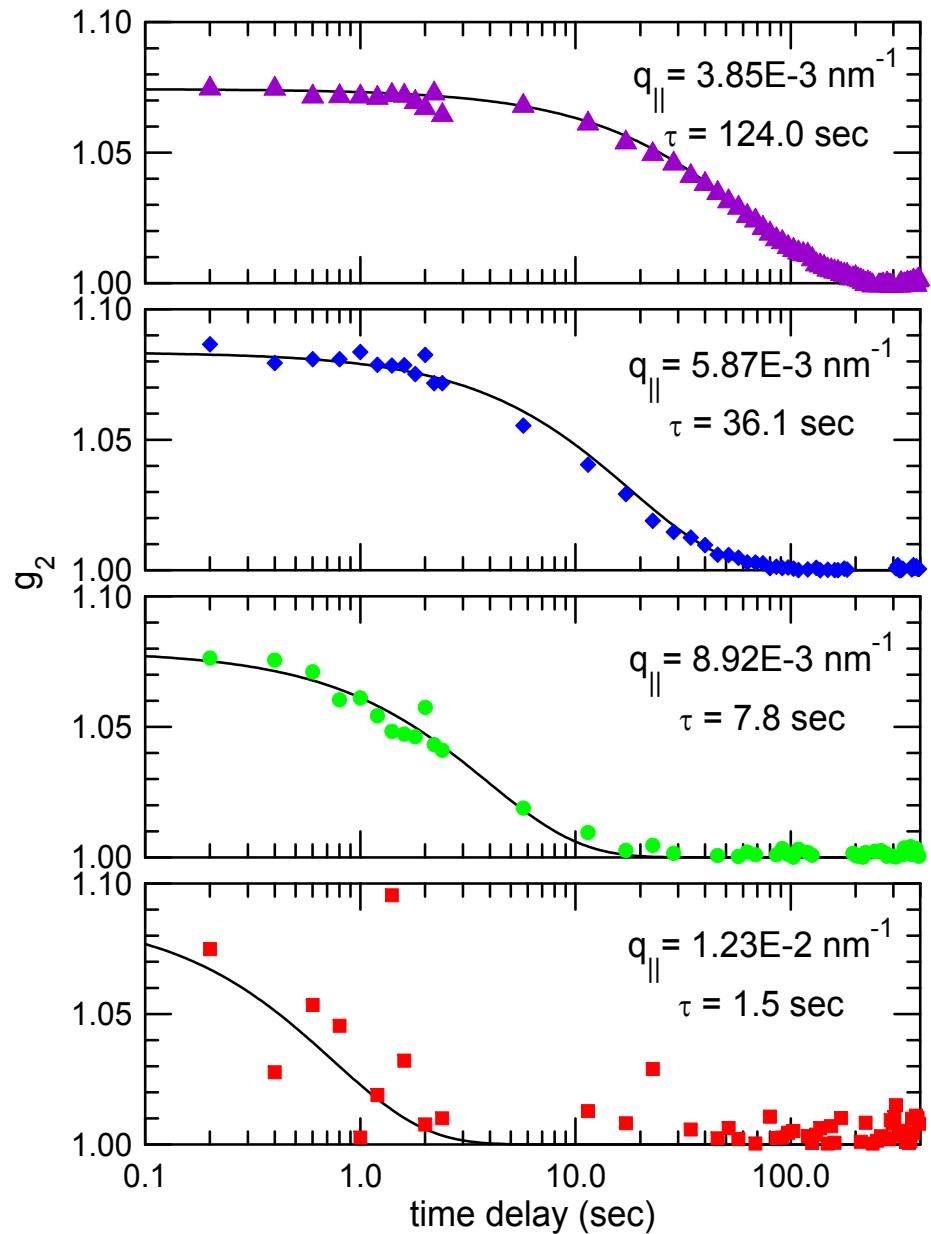
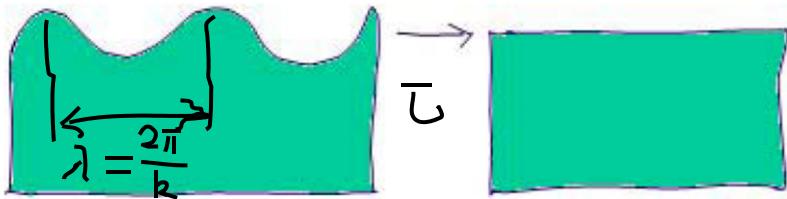
$\beta$ : speckle contrast

Siegert Reln.  
 $g_2(\mathbf{q}, t) = |g_1(\mathbf{q}, t)|^2$

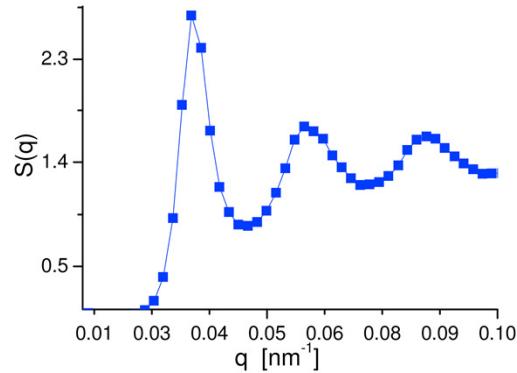
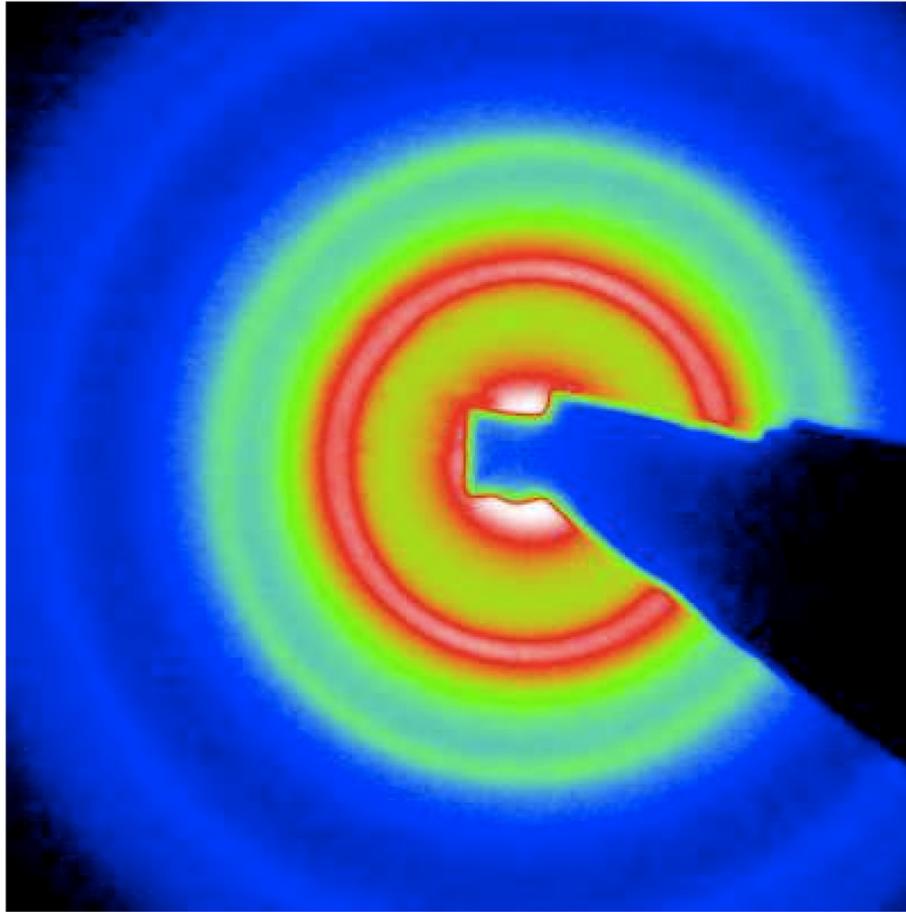
# Intensity Autocorrelation

$$g_2(q, \tau) = \frac{\langle I(q, t)I(q, t + \tau) \rangle}{\langle I(q, t) \rangle^2}$$

$$g_2(q, \tau) = 1 + \beta e^{-2t/\tau}$$



# Incoherent SAXS



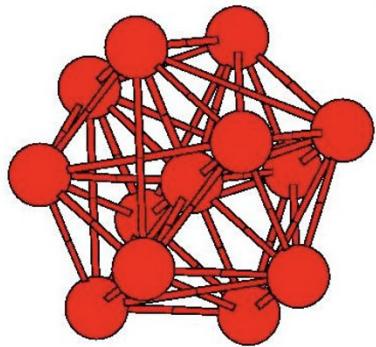
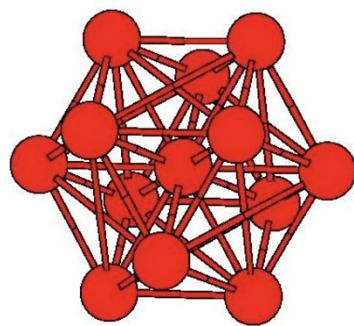
ensemble averaged structure factor

$$\langle S(Q) \rangle = 1 + n_0 \int (g(r) - 1) e^{iQr} dr$$

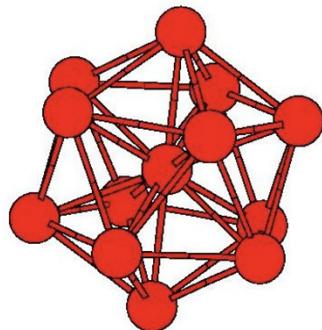
radial distribution function

$$g(r) = 4\pi r^2 n_0^{-2} \langle \rho(0) \rho(r) \rangle$$

information on local symmetries is lost



fcc and hcp structures can fill up space  
and form crystals



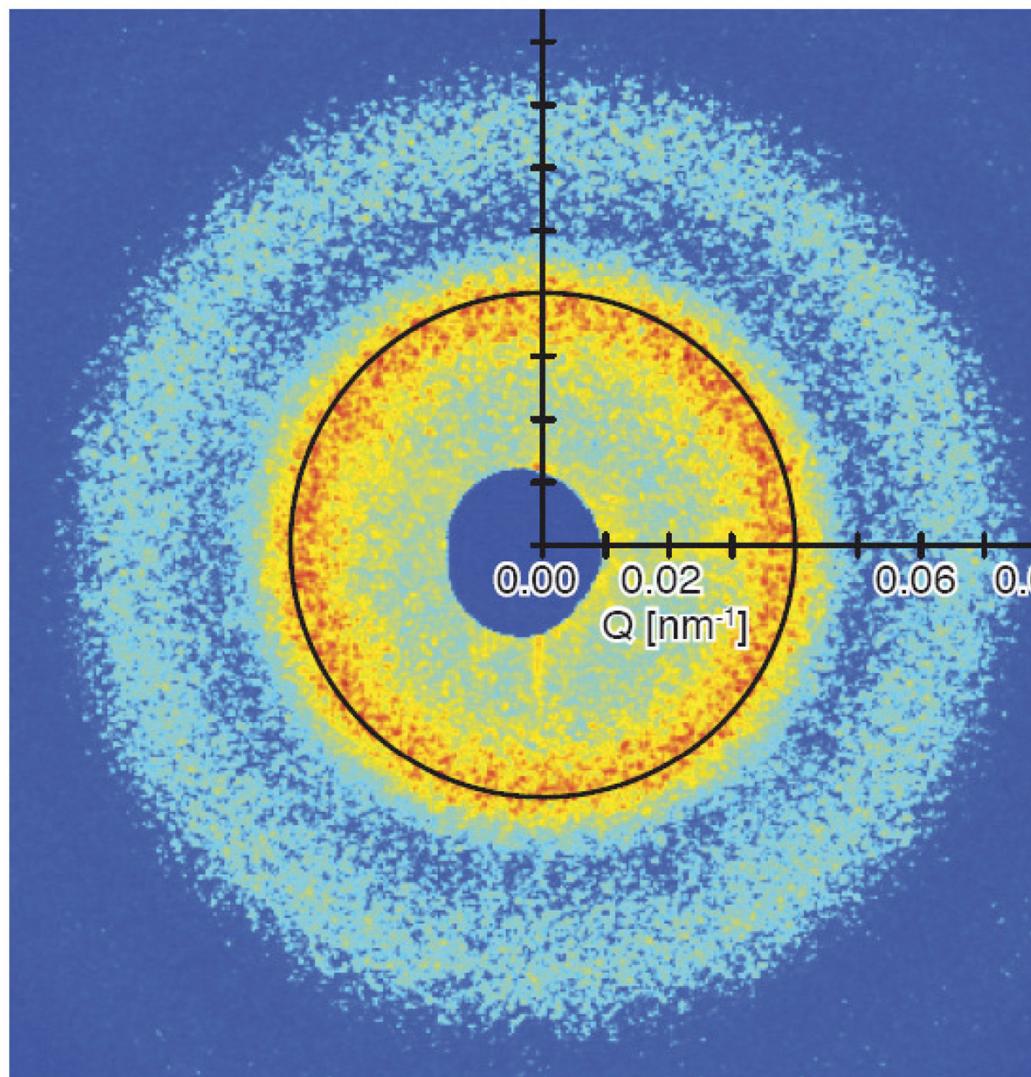
icosahedral structures can not fill space  
but may be energetically favored in liquids  
“locally favored structures (lsf)”

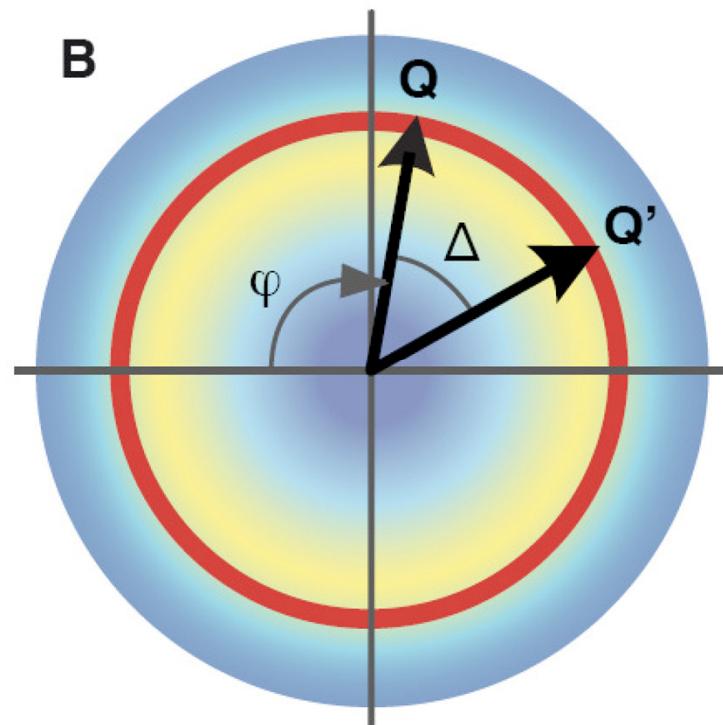
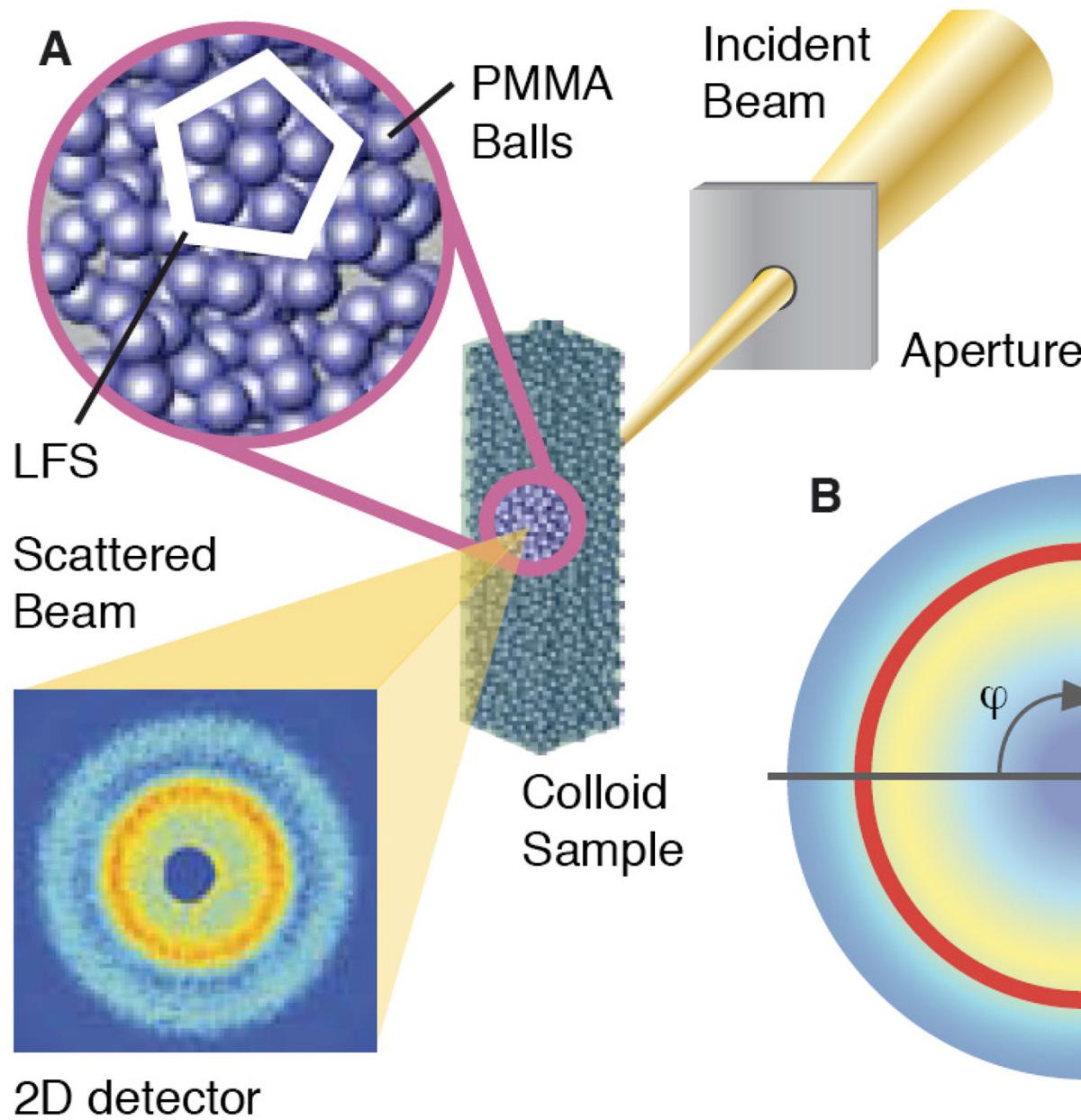
F. C. Frank, Proc. R. Soc. London A 215, 43 (1952).  
P. J. Steinhardt, D. R. Nelson, and M. Ronchetti, Phys.  
Rev. B 28, 784 (1983)

# Hard sphere systems – speckle pattern

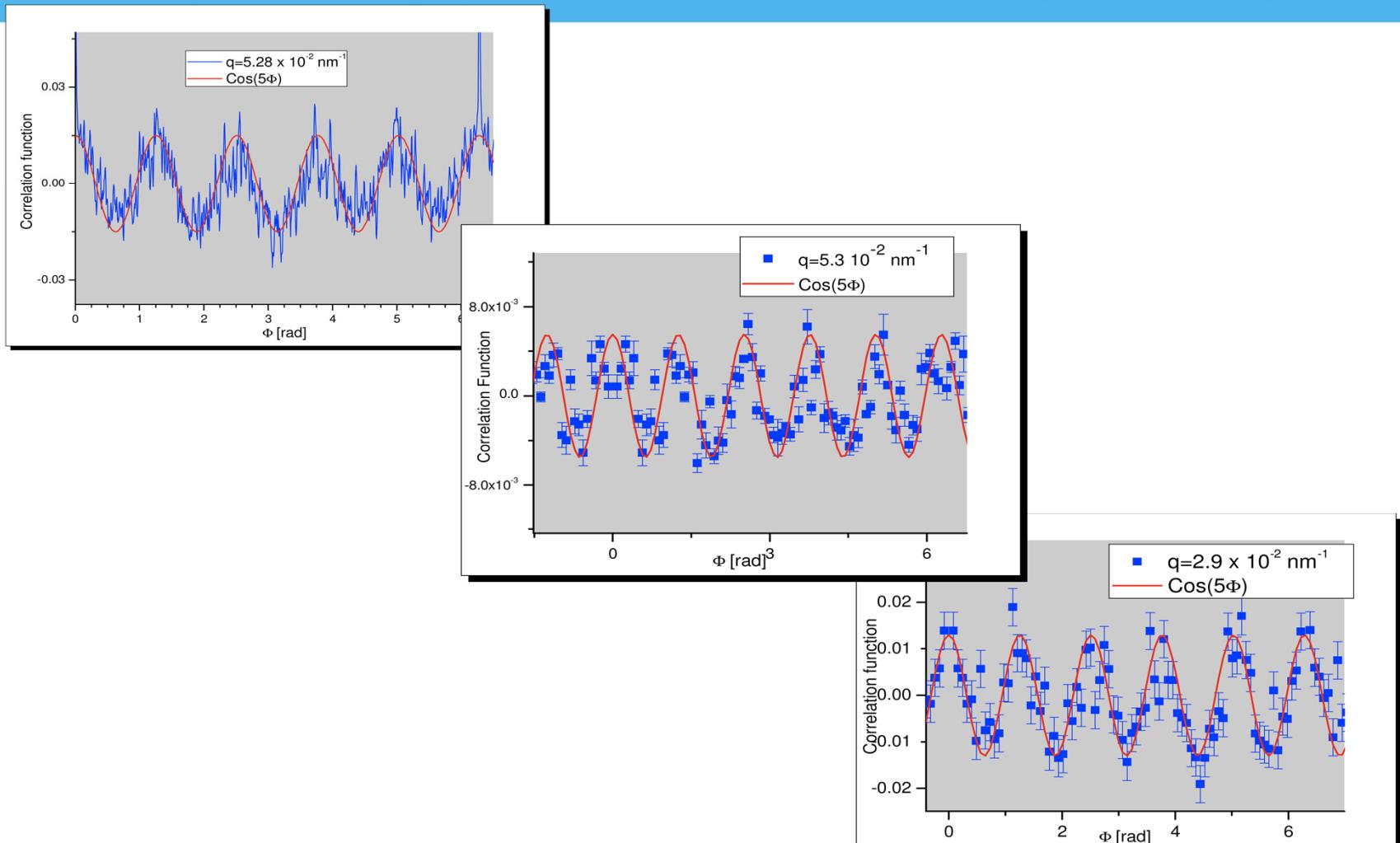
P.Wochner et al.

PNAS 2009



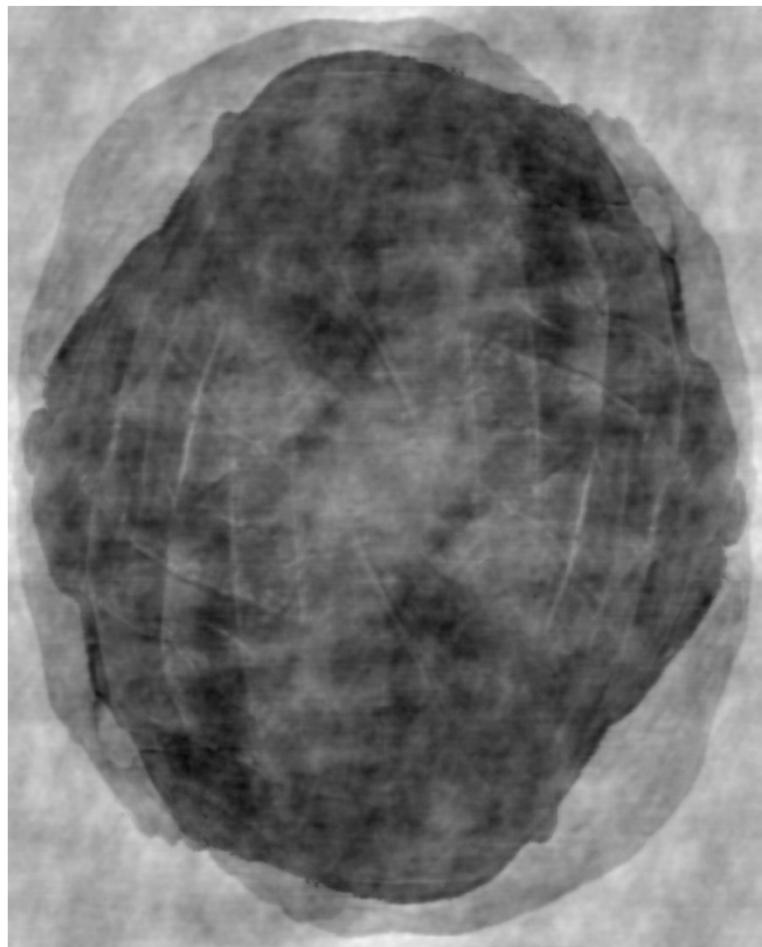


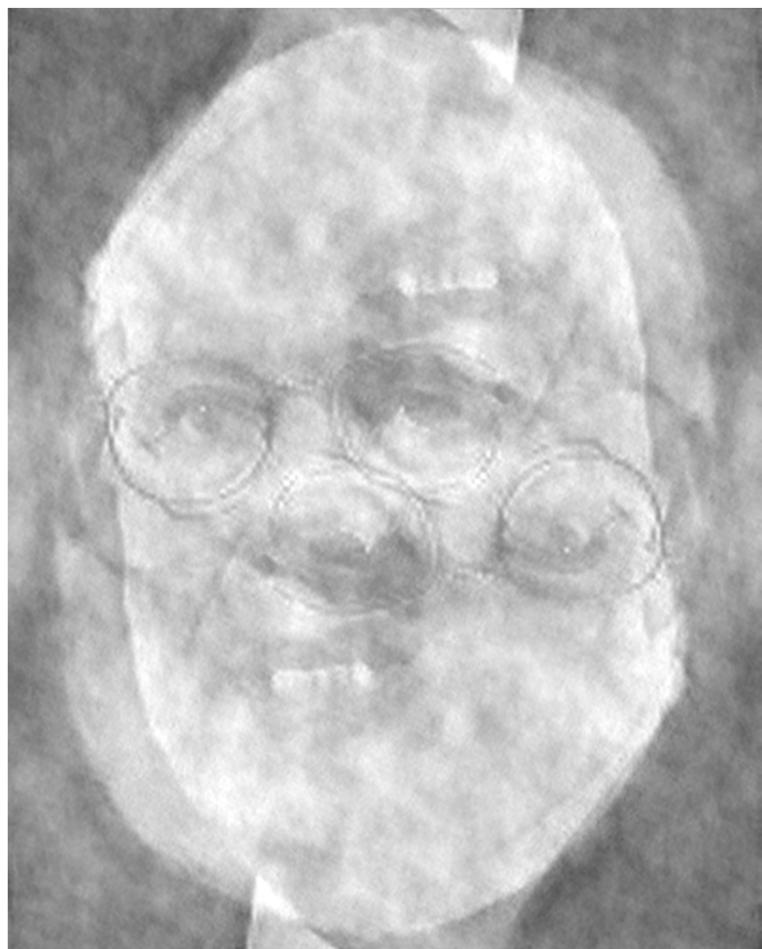
# 5-fold symmetry present in all hard sphere systems





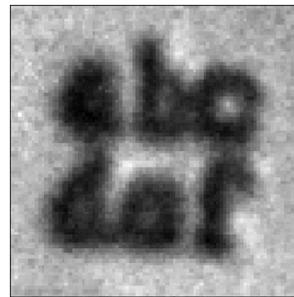
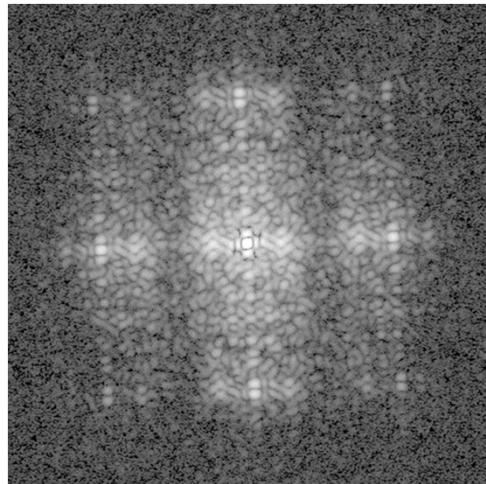
## The “Phase Problem”





Miao, Charalambous, Kirz, Sayre, *Nature* **400**, 342 (1999).

$\lambda=1.8$  nm  
soft x-ray  
diffraction  
pattern



Low angle data  
From optical  
micrograph

Scanning  
electron  
micrograph  
of object

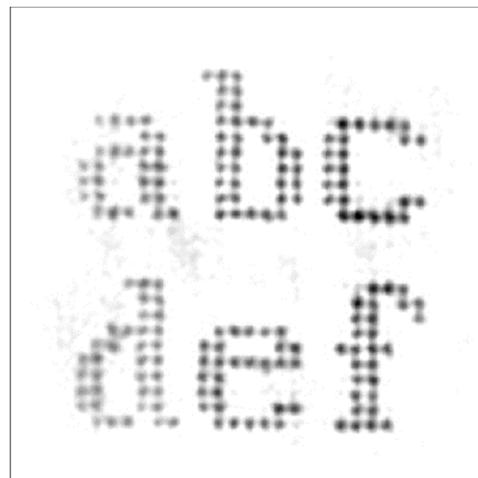
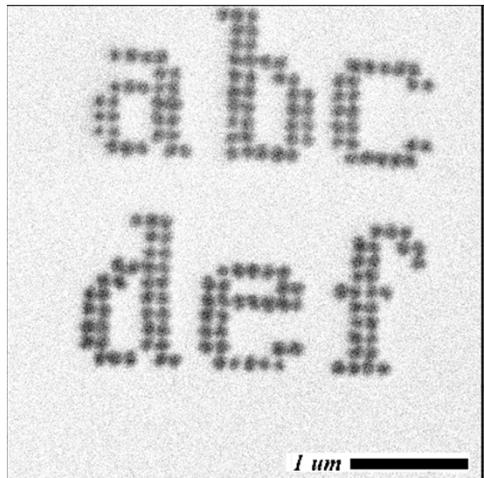


Image reconstructed  
from diffraction  
pattern ( $\theta_{\max}$   
corresponds to 80  
nm). Assumed  
positivity

# Reconstruction

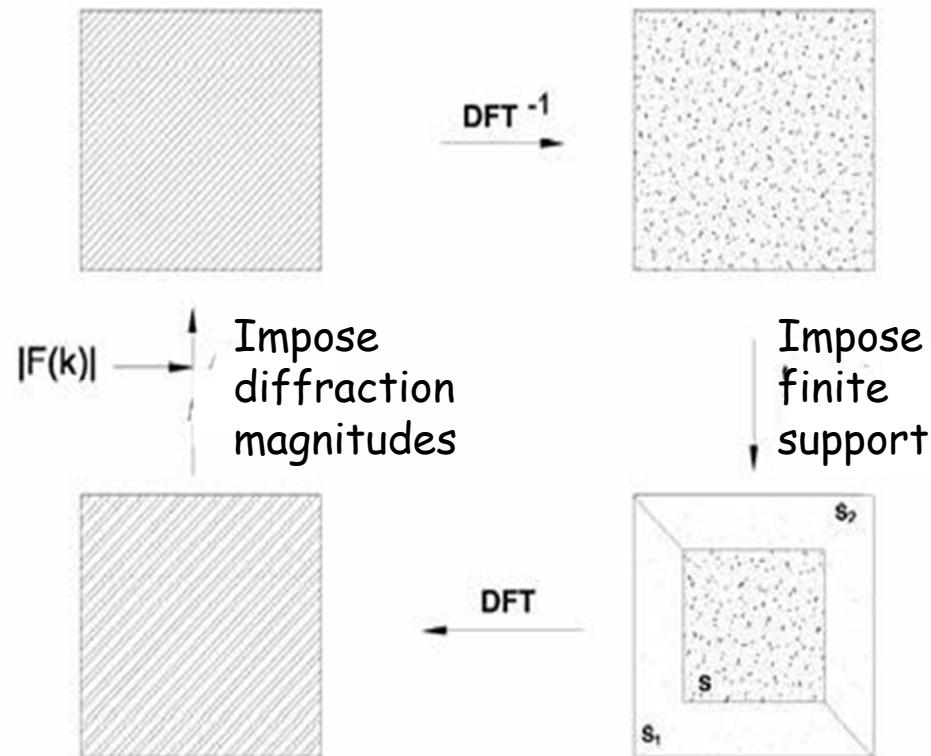
Equations can still not be solved analytically

Fienup iterative algorithm

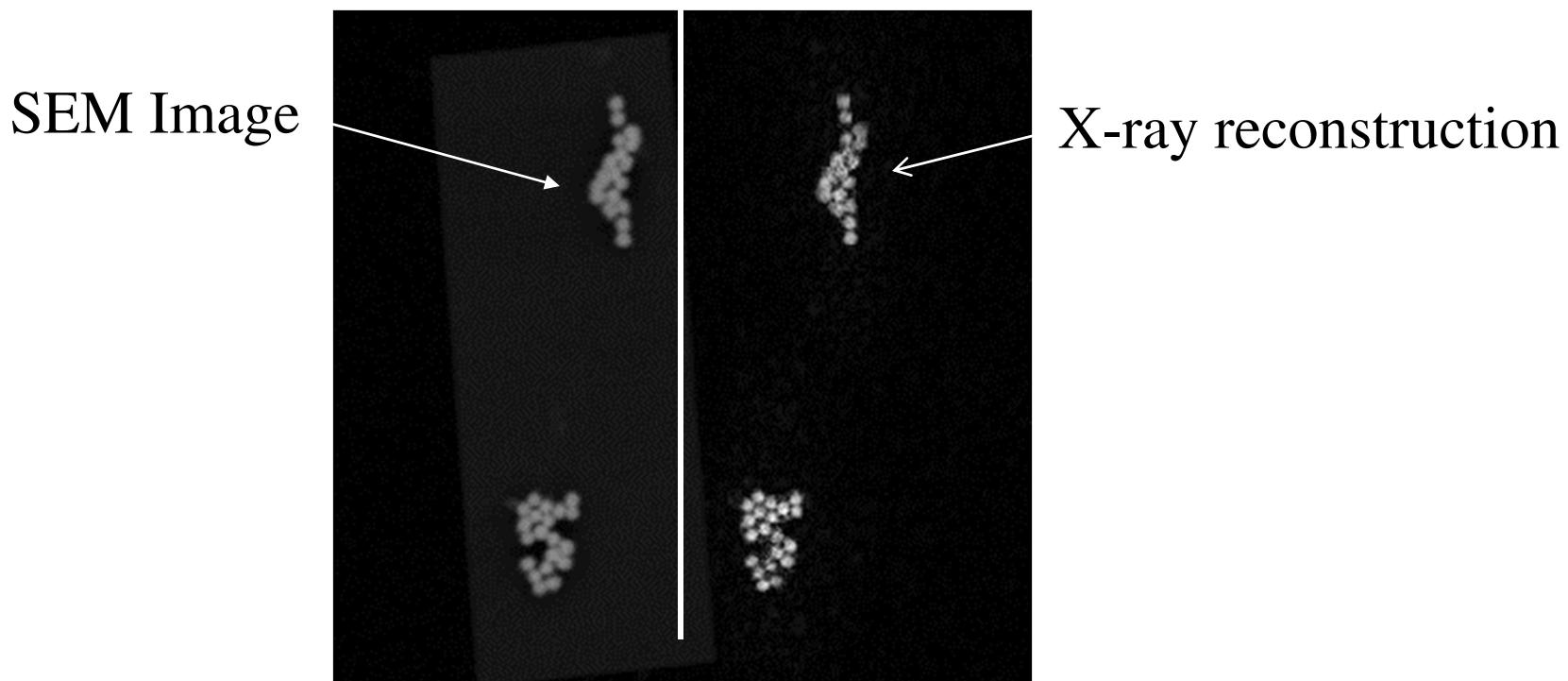
Reciprocal space

Real space

- Positivity of electron density helps!



## Successful reconstruction of image from soft X-ray speckle alone.



50 nm diameter Gold Balls on transparent SiN membrane.

No “secondary image” was used

Approximate object boundary obtained from autocorrelation fn.

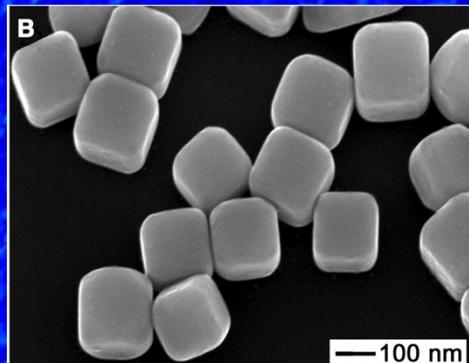
\*How to make an isolated object ? Use AFM to remove unwanted balls.

# *Imaging of individual nanoparticles at the APS*

Ross Harder, University of Illinois, Champaign

*Coherent diffraction pattern  
from 170 nm Ag particle*

*170 nm silver cubes*



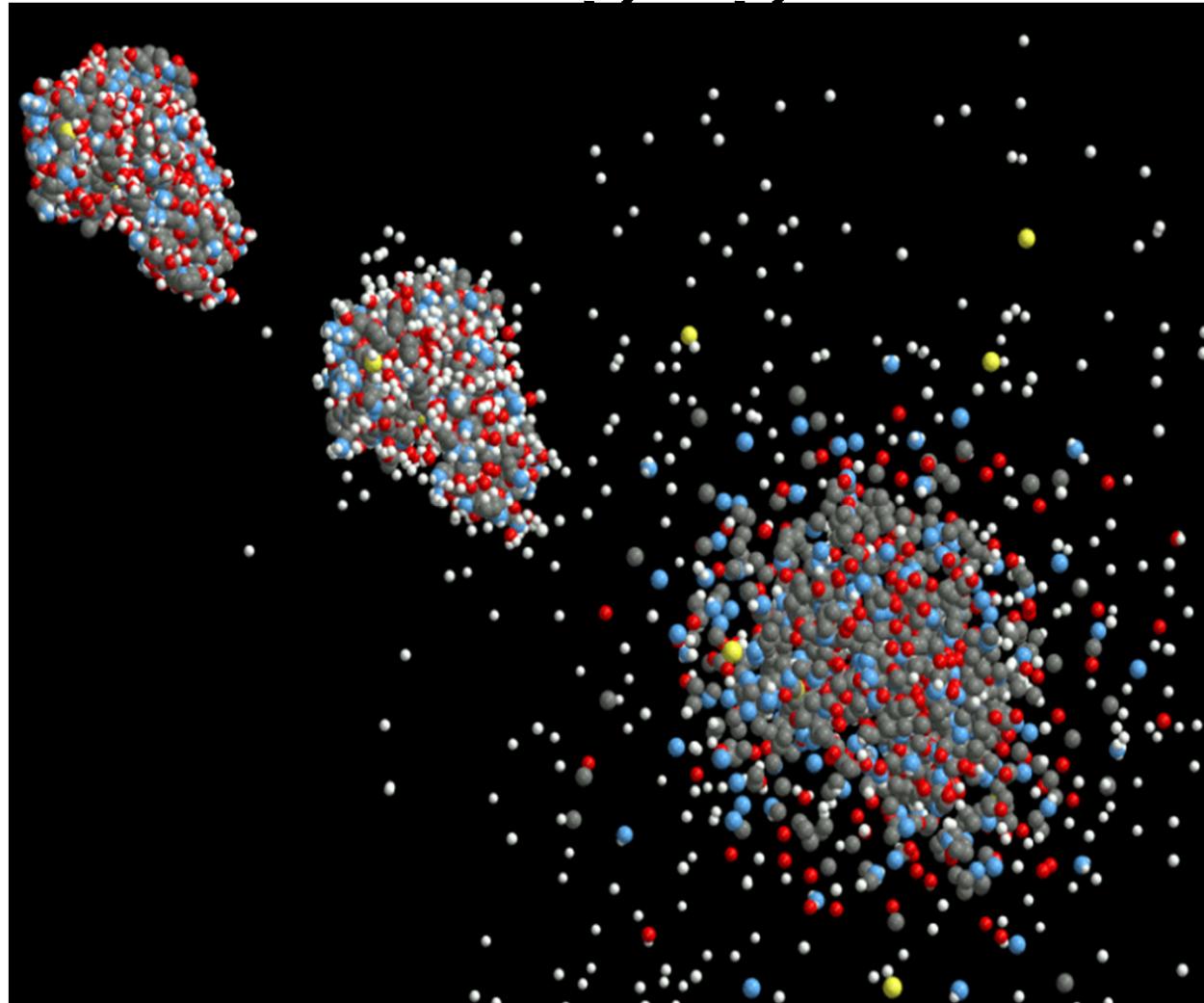
$5 \times 10^{-2} \text{ nm}^{-1}$

*inversion of  
diffraction pattern  
'lensless imaging'*

J.K. Robinson, et al., Science 298 2177 (2003)

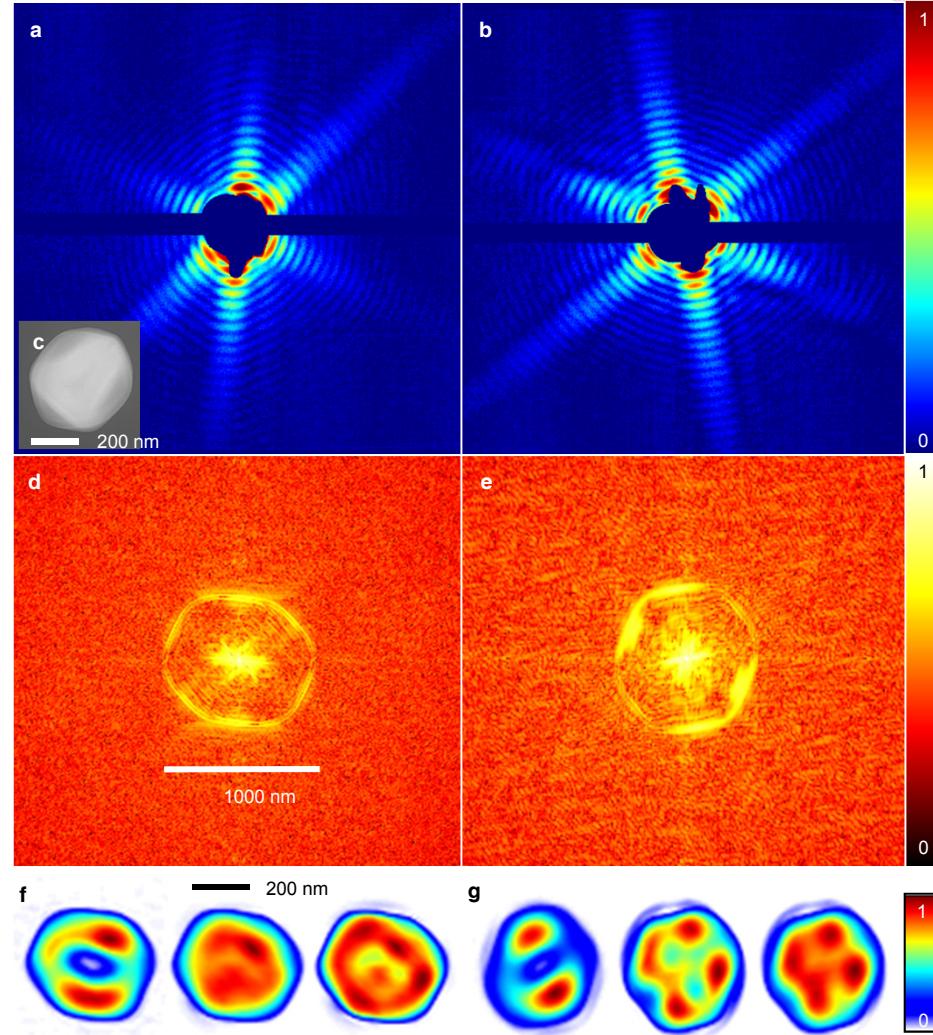
# Single molecule imaging?

- Atomic resolution structures known for *few* mammalian membrane proteins!
- Collect many single molecule diffraction patterns from fast x-ray pulses, and reconstruct?
- Lysozyme explodes in ~50 fsec
- R. Neutze *et al.*, *Nature* **406**, 752 (2000)



# Single mimivirus particles intercepted and imaged with an X-ray laser

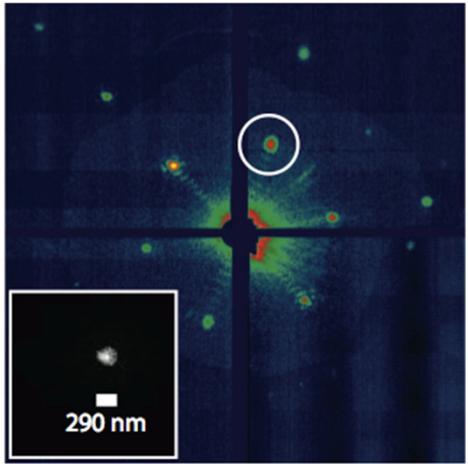
*Beyond crystallography: A new world in structural sciences*



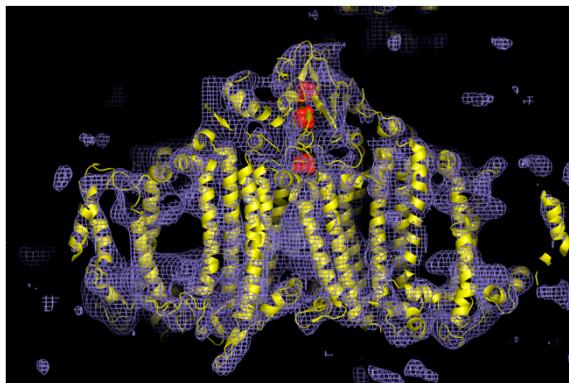
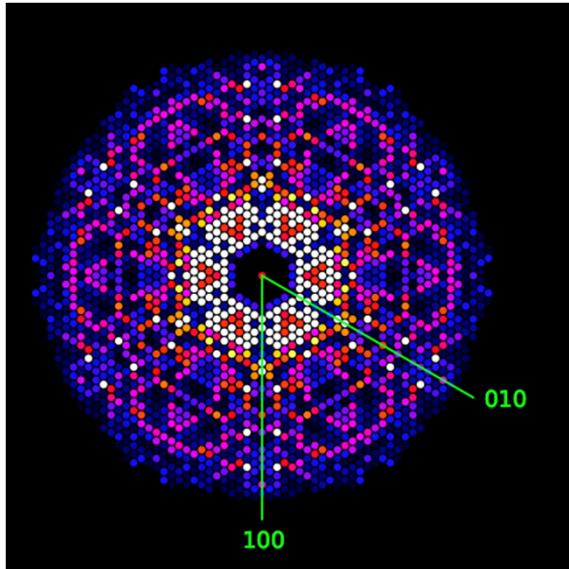
- A very short and extremely bright coherent X-ray pulse can be used to outrun key damage processes and obtain a single diffraction pattern from a large macromolecule, a virus, or a cell *without the need for crystalline periodicity*.
- Mimivirus is the largest known virus, *comparable in size to a small living cell*. It is too big for structure determination by electron microscopy and it cannot be crystallised.
- The structure of the intact virus was recovered from the flash diffraction pattern alone.
- There was no measurable sample deterioration.
- Death-rays: We expect high-resolution structures in such experiments with shorter and brighter photon pulses focused to a smaller area.
- Resolution can be further extended by averaging for samples available in multiple identical copies.

# Femtosecond x-ray nanocrystallography overcomes limitations of radiation damage

*A new paradigm opens up macromolecular structure determination to systems too small or radiation sensitive for synchrotron studies, and may save years of effort in crystallization trials*

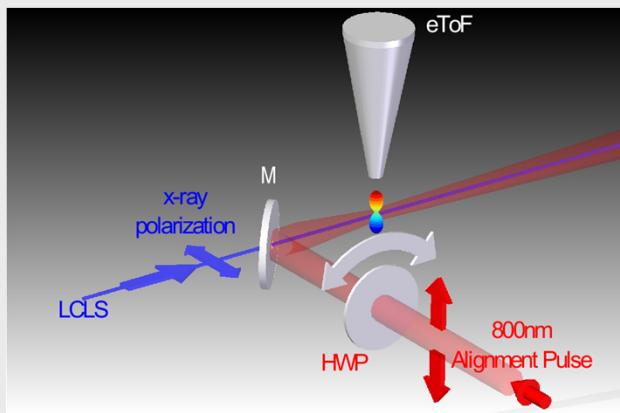


Single-shot diffraction patterns are recorded with 70 fs pulses. Coherent diffraction shows the crystal size is sub-micron (top left) and that the crystal has a perfect lattice. Individual shots are oriented in 3D and combined to build up the full information content of the underlying macromolecule (top right). This first demonstration was carried out at 2 keV photon energy, limiting the resolution to about 9 Å. (This will be improved with the dedicated CXI instrument.) The quality of the data are demonstrated by carrying out molecular replacement refinement (right). Structural details such as helices can be observed.

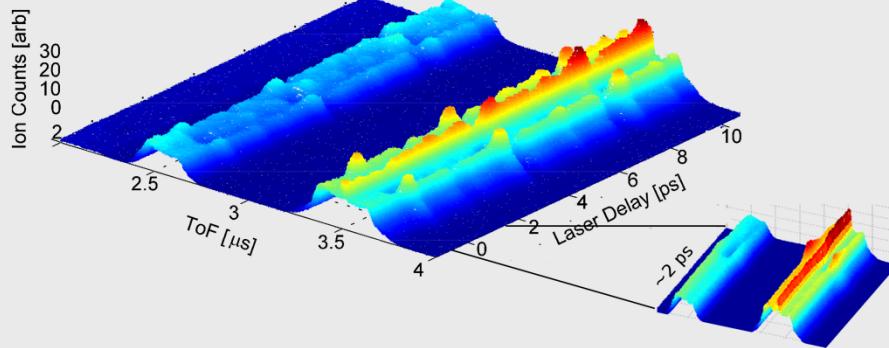


- The ultrafast LCLS x-ray pulses allow us to record “diffraction before destruction” where information is obtained before the onset of structural damage.
- Diffraction can be measured from sub-micron crystals containing less than a thousand molecules.
- Demonstrated using Photosystem I, a membrane protein, key to photosynthesis, that is extremely difficult to grow into large crystals.
- 30 single-crystal patterns per second were recorded from a liquid stream carrying a suspension of nanocrystals. 15,000 of these were indexed and combined into a full diffraction pattern which was analyzed with standard tools.
- Data are collected at room temperature. No cryogenic cooling or stabilization required.

# Femtosecond time-resolved x-ray experiments study aligned “hollow” molecules

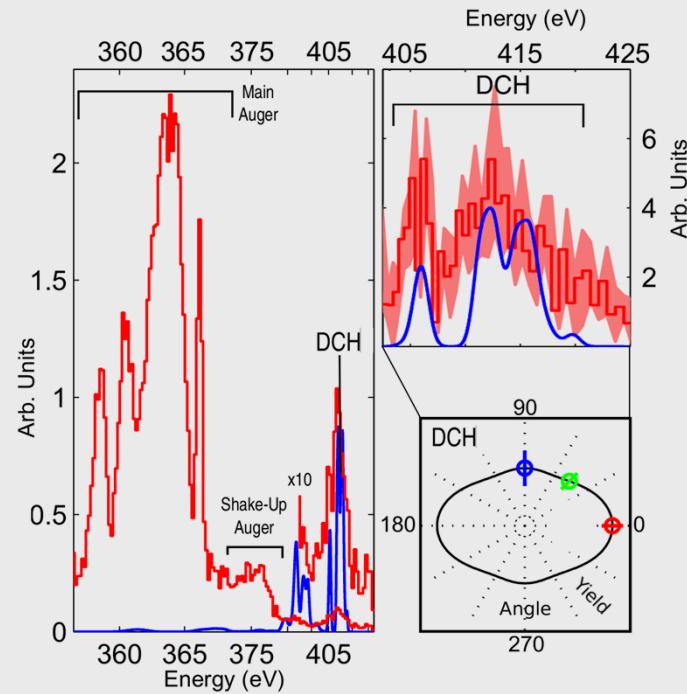


Laser pump/x-ray probe ion time of flight spectra



- 10 fsec x-ray pulse arrives within 150 fsec of peak in alignment
- X-ray pulse produces molecules with a hollow atom (double core hole)
- First spectra of intramolecular dynamics...
  - in strong x-ray fields in the molecular frame

“Hollow” molecule Auger spectrum in the molecular frame

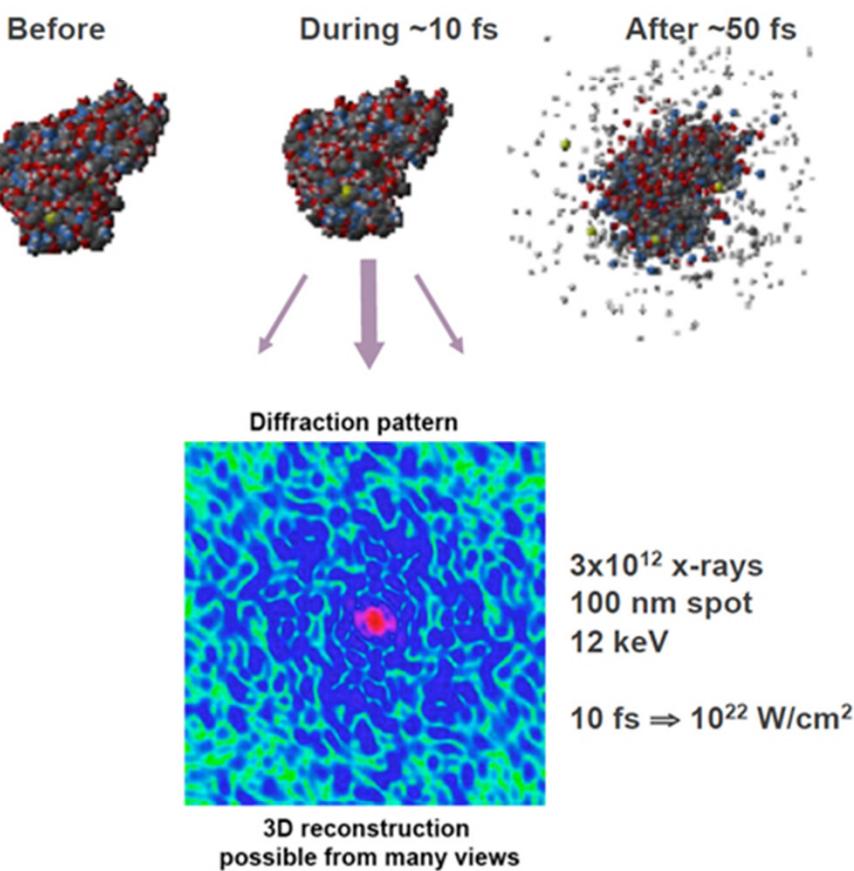


Measurement in red  
*Ab initio* calculation in blue

J. Cryan, J. Glownia, P. Bucksbaum, R. Coffee, et al., Phys. Rev. Lett., **105**, 083004 (2010); Opt. Exp. **18**, 17620 (2010)

# AMO questions at the ultraintense x-ray frontier

- fundamental nature of x-ray damage at high intensity
  - Coulomb explosion
  - electronic damage
  - behavior at  $10^{22} \text{ W/cm}^2$  -  $1\text{\AA}$
- nonlinear x-ray processes
  - role of coherence
- quantum control of inner-shell processes



Neutze, Wouts, van der Spoel, Weckert, Hajdu Nature 406, 752 (2000)

# LCLS Experiment 1 - Oct 1, 2009

Nature of the electronic response to

$10^5$  x-rays/ $\text{\AA}^2$

80 - 340 fs

800 - 2000 eV

$\sim 10^{18}$  W/cm $^2$

Original single molecule imaging parameters, Neutze et al. Nature (2000)

$3 \times 10^{12}$  x-rays/(100 nm) $^2$  =  $3 \times 10^6$  x-rays/ $\text{\AA}^2$

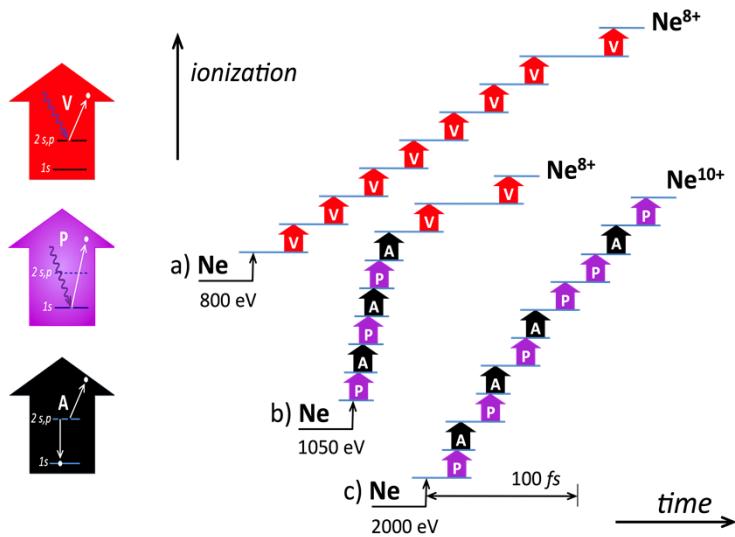
10 fs

$\sim 10^{22}$  W/cm $^2$



# Femtosecond electronic response of atoms to ultraintense x-rays

*Understanding the response of matter to ultraintense x-ray irradiation is vital for LCLS applications*

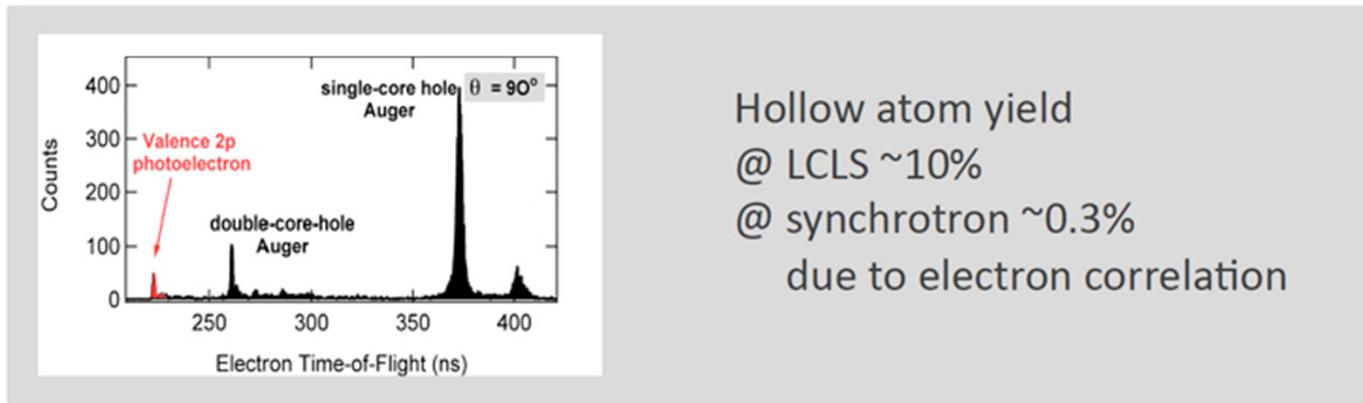


*Multiphoton ionization of atoms occurs within a single LCLS pulse of ~100 fs duration and fluence of ~ $10^{12}$  x-rays per square micron. The multiple ionization proceeds through a sequence of single electron ejections. Three types of electrons are ejected, valence (V), inner-shell (P) and Auger (A), as depicted on the left of the diagram. One can control the ionization mechanism.*

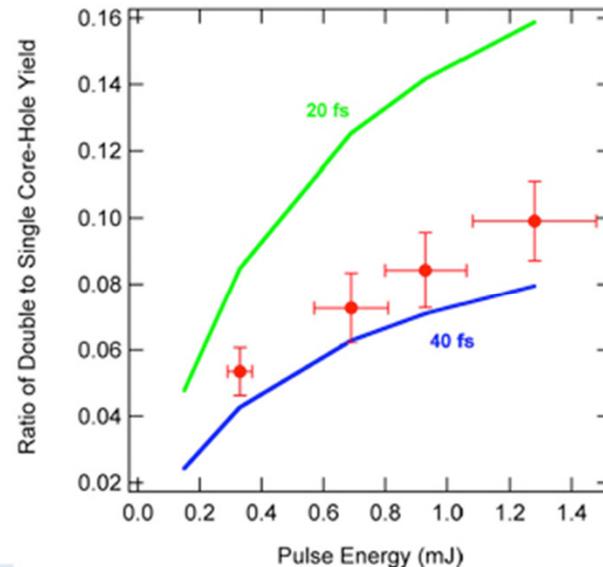
*Studying the prototypical neon atom: at photon energies below the K-edge (870 eV) the outer electrons are stripped (VVV... process), whereas at photon energies above the K-edge the inner electrons are initially ejected, followed by femtosecond Auger decay (PAPA... process). At very high intensity, both 1s core electrons can be removed prior to refilling by Auger decay to produce exotic hollow atoms.*

- LCLS provides x-ray pulses with peak intensities more than a billion times greater than those from any existing synchrotron source. Understanding the electronic response at extreme x-ray intensities is fundamental.
- By studying a simple target, neon, where the dominant interaction changes as a function of photon energy from outer- to inner-shell absorption, a general understanding can be achieved.
- Observe full stripping of neon via a six-photon, ten-electron process within a single ~100 fs x-ray pulse – the first observed multiphoton x-ray process.
- Observe x-ray induced transparency via hollow atom formation at high x-ray intensities – predict this phenomenon will be generally observed in molecules, solids – allowing one to control the penetration depth in matter with pulse duration.
- Straightforward rate equation model reproduces the observed trends – suggesting that more complex systems may be modeled.

# Hollow atom production: deliberate, huge and an indicator of x-ray pulse duration



1050 eV,  
nominal electron bunch  
duration ~80 fs

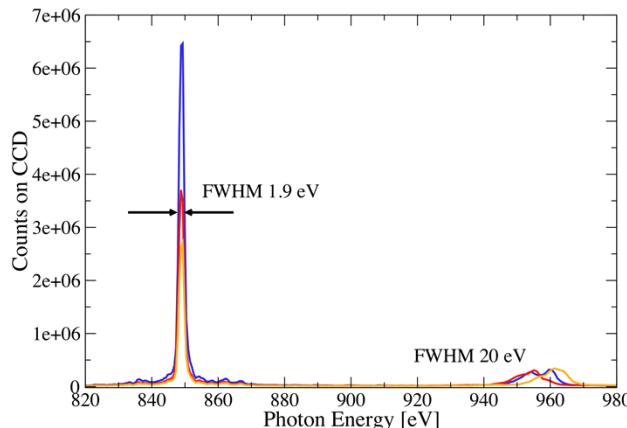


# Summary of ultra-intense x-ray interaction phenomena

- Target changes during a single 100 fs x-ray pulse at fluences similar to that for single molecule imaging
  - six-photon, ten-electron stripping of neon ( $\sim 10^{12}/\mu\text{m}^2$ )
  - multiphoton absorption probability high when fluence  $> 1/\sigma$
- Intensity-induced x-ray transparency – a general phenomena
  - transient x-ray transparency caused by formation of hollow atoms
  - hollow atoms  $\sigma_{\text{scatt}}/\sigma_{\text{abs}}$  is increased – advantageous for imaging
- Straightforward rate equation calculations capture essential physics
- Femtosecond time-scale atomic processes provide FEL diagnostics



# First realization of an atomic inner-shell x-ray laser at 850 eV, by ultrafast photoionization of Neon with the LCLS

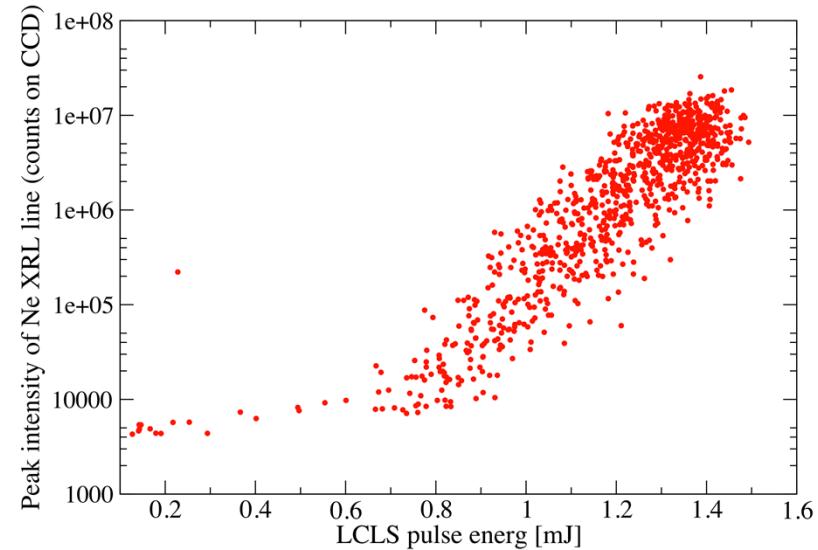


**Single shot spectra of the transmitted LCLS line at around 960 eV and the Ne K-a line at 850 eV (1.9 eV instrument-limited width). The brightest shot resulted in  $2 \times 10^9$  photons in the Ne K-a line, with a conversion efficiency of  $10^{-3}$ .**

- The photoionization-based atomic x-ray lasing scheme was first proposed in 1967 (Duguay and Renzepis)
- Due to requirement of an extremely fast and intense x-ray pump source, it could never be realized so far.
  - The experiment is a first step in the virtually unexplored field of non-linear quantum optics with x-rays and opens the pathway to new, non-linear spectroscopic methods in the x-ray regime

- Focusing LCLS pulses of 960 eV into a Neon gas sample to a spot size of  $\sim 1.5$  mm, a long narrow plasma column is produced on a femtosecond timescale by photoionization of the K-shell, resulting in a population inversion of the Ne K-a transition.
- Fluorescence photons of the front-end of the plasma column get amplified by stimulated emission, resulting in ultrabright, fs

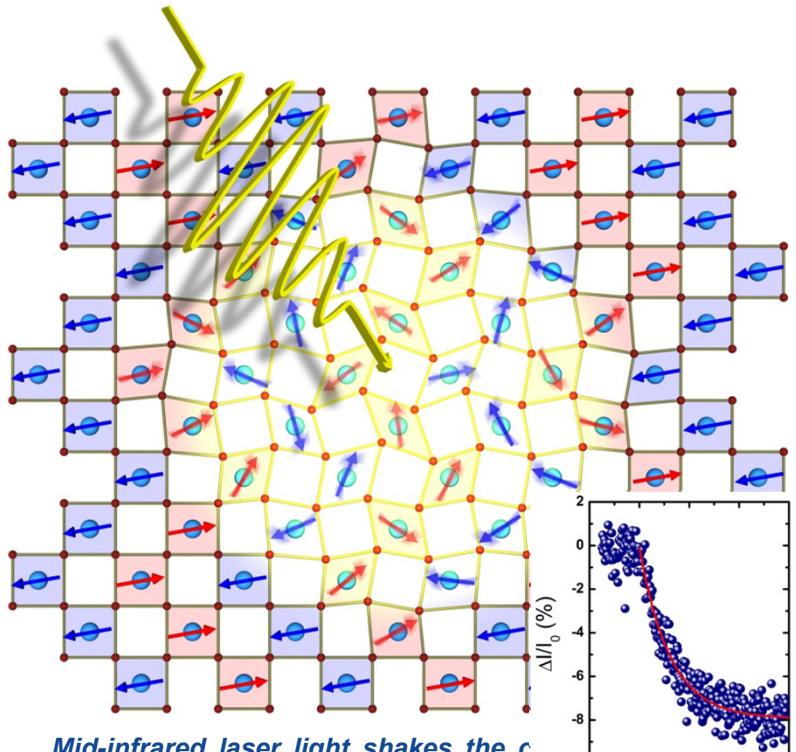
x-ray pulses at 850 eV  
**Peak intensity of the Ne K-a line as a function of the pumping power of the LCLS. Doubling the pumping intensity gives a rise in the peak power of four orders of magnitude.**



N. Rohringer et al., manuscript in preparation

# Controlling magnetism in complex oxide with lattice excitation

**New approach for magnetic switching disclosed at the LCLS Free Electron Laser**



Mid-infrared laser light shakes the c layered manganite  $\text{La}_{0.5}\text{Sr}_{1.5}\text{MnO}_4$  excitation of a vibrational mode. Ant...  
is in this way perturbed, as viewed by time-resolved resonant soft X-ray diffraction from the magnetic superlattice.

- To date, light control of magnetization has always been induced by near-infrared light. This type of stimulation is associated with large dissipation, as eV-photon energies are used to drive the rearrangement of the microscopic order on the meV energy scale.
- Electronic properties of complex oxides can be controlled by optical excitation of lattice vibrations in the mid infrared, providing an alternate path to steer condensed matter on ultrafast timescales.
- Recent experiments at the LCLS Free Electron Laser utilizing time-resolved resonant soft X-ray diffraction demonstrate that the magnetic order in complex oxides can be disordered through optical lattice manipulation.
- The magnetic response time is 12ps, likely dictated by spin-lattice relaxation rates.

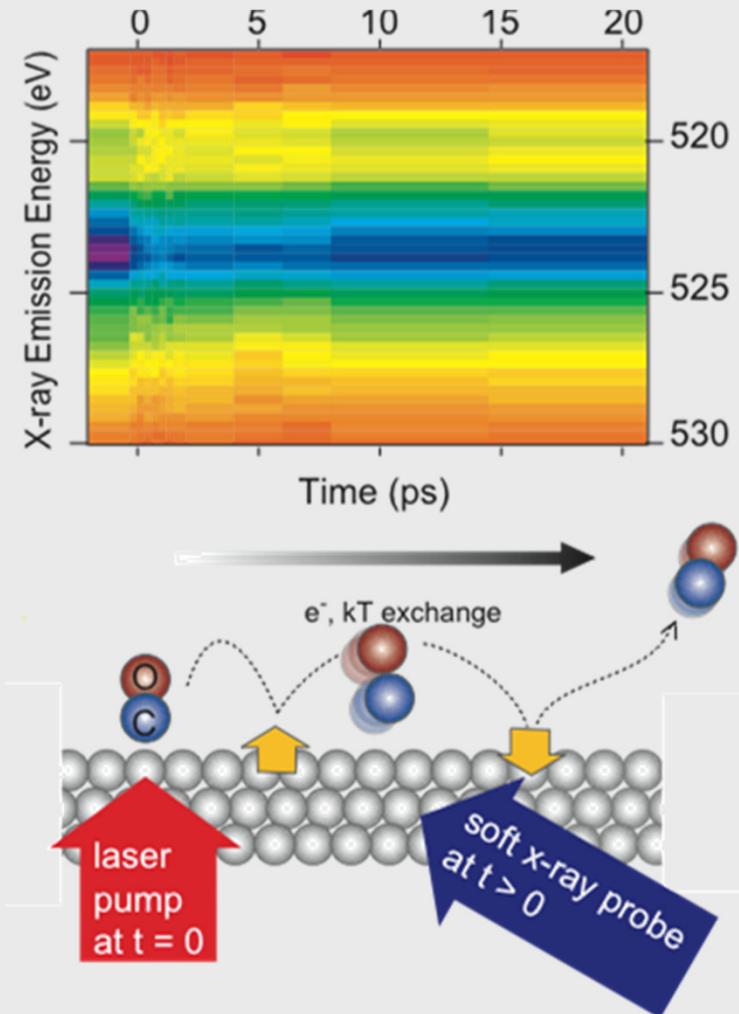


Center for  
Free-Electron Laser  
Science



# LCLS pump-probe experiments: CO on Ru(001)

- Nearly all chemical reactions of importance to society occur at interfaces
- Catalysis is a trillion dollar industry and is also at the core of chemical energy transformations
  - The first pump probe experiment at LCLS investigated a simple reaction step involving breaking of the CO-metal bond
  - The experiments use a 400 nm optical laser pump pulse, and a LCLS pulses to probe the CO electronic structure state though O K-emission spectroscopy as a function of time delay.
  - The results shows the time evolution of a transient CO state where the chemical bond strength to the surface is reduced



PIs: Anders Nilsson, Hirohito Ogasawara, Dennis Nordlund (SLAC), Wilfried Wurth (DESY), Alexander Föhlich (Helmholtz center), Henrik Öström (Stockholm U) and Martin Wolf (Fritz Haber Institute)

## Conclusion

Much of the most exciting recent science with Neutrons and X-rays has resulted from fruitful collaborations between accelerator physicists and CM and Materials Scientists and Crystallographers and Biologists

Hopefully, this will continue, leading to more exciting science over the next decade,  
So, this is not.....

THE END