[redacted]

Applications of TW-class ultrashort X- ray pulses in scientific research

J. B. Hastings
SLAC National Accelerator Laboratory

Physics & Applications
Of High Efficiency
Free-Electron Lasers
Workshop

April 11-13, 2018 at the UCLA
California NanoSystem Institute

Thanks
First to the organizers for giving me the opportunity

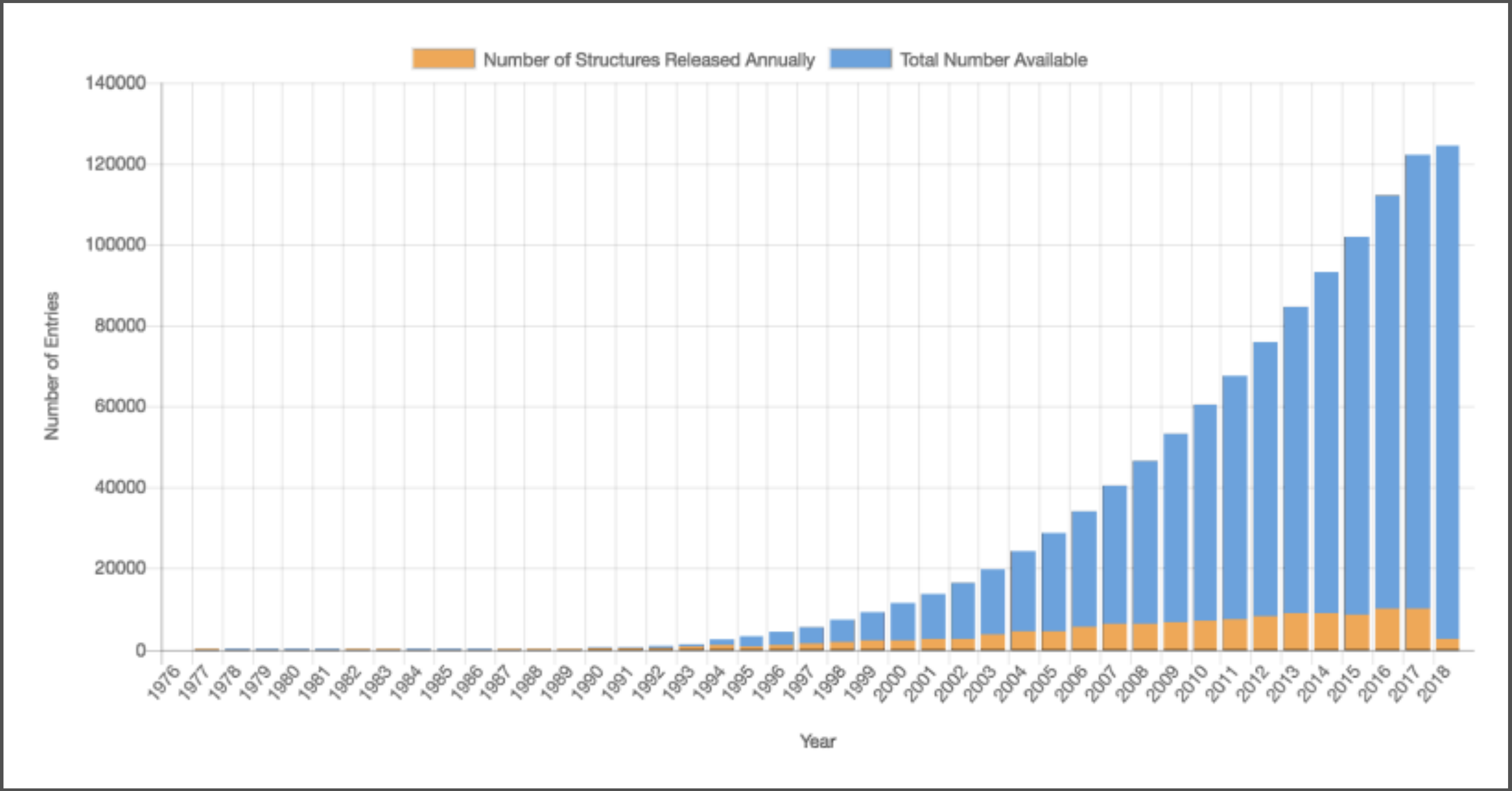
This talk is based on the work of the extraordinary team associated with LCLS from its concept to its future.

I particularly acknowledge my long standing collaboration with Paul Emma, Zhirong Huang, Claudio Pelligrini, David Reis and Giulio Monaco

X-rays are a unique analytical tool
What x-rays do best - imaging

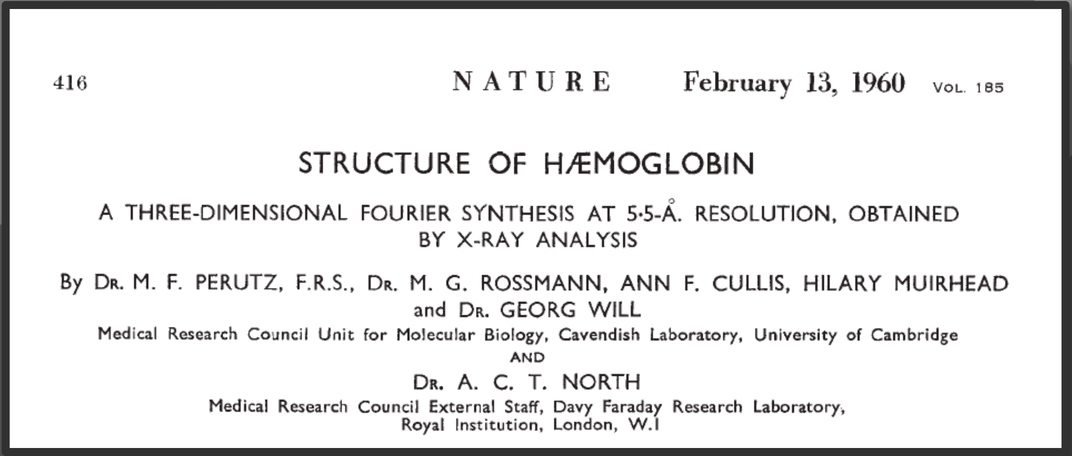
- Imaging in reciprocal space
  - Static structure
  - Dynamics-lattice vibrations
  - Resonant inelastic scattering
  - …….
- Imaging in real space
  - Scanning microscopies
  - Elemental mapping
  - Tomography
  - ……

Structures deposited in the Protein Data Bank



Structural biology:
Reciprocal space imaging of macro molecules

- Perutz et al. Hemoglobin
- MAD phasing
- *FEL*-Serial Femtosecond Crystallography
- *Storage Ring*-Serial crystallography
- Single particle imaging

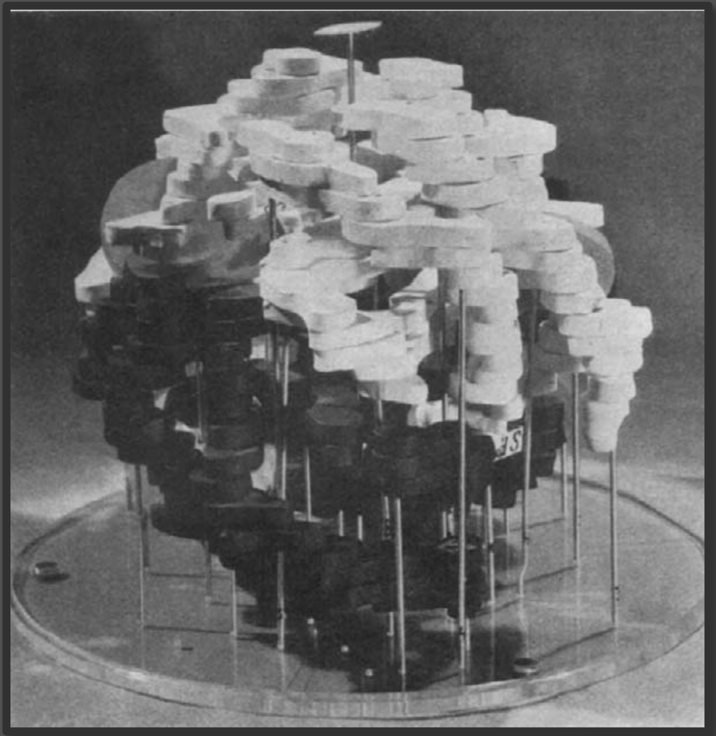


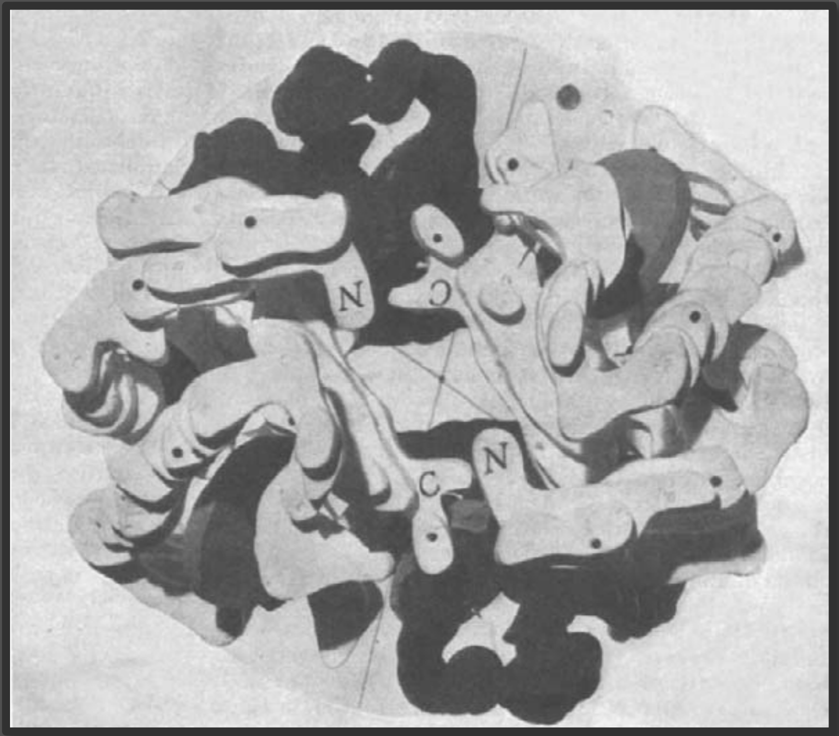
416 NATURE February 13, 1960 Vol. 185

STRUCTURE OF HÆMOGLOBIN

A THREE-DIMENSIONAL FOURIER SYNTHESIS AT 5·5-Å. RESOLUTION, OBTAINED
BY X-RAY ANALYSIS

By Dr. M. F. PERUTZ, F.R.S., Dr. M. G. ROSSMANN, ANN F. CULLIS, HILARY MUIRHEAD
and Dr. GEORG WILL
Medical Research Council Unit for Molecular Biology, Cavendish Laboratory, University of Cambridge
AND
Dr. A. C. T. NORTH
Medical Research Council External Staff, Davy Faraday Research Laboratory,
Royal Institution, London, W.1



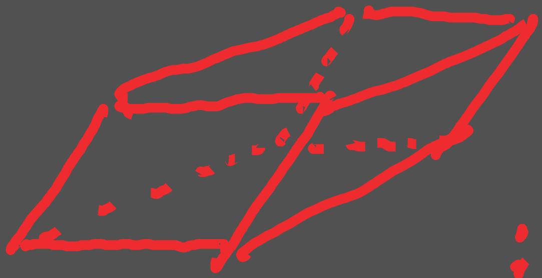


5.5 Å Resolution

Structural biology:
Reciprocal space imaging of macro molecules

- Perutz et al. Hemoglobin
- MAD phasing
- *FEL*-Serial Femtosecond Crystallography
- *Storage Ring*-Serial crystallography
- Single particle imaging

Intensity of diffraction from a small crystal (1)



$V_x = N_1\vec{a}_1 \cdot N_2\vec{a}_2 \times N_3\vec{a}_3$

$: N = N_1 N_2 N_3$ unit cells

$$E(\vec{Q}) = \sum_{uvw} \sum_{s} f_s e^{i\vec{Q}\cdot(\vec{R}_{uvw}+\vec{x}_s)}$$

define $F(\vec{Q}) = \sum_{s} f_s e^{i\vec{Q}\cdot\vec{x}_s}$

Intensity of diffraction from a small crystal (2)

$$E(\vec{Q}) = F(\vec{Q}) \sum_{uvw} e^{i\vec{Q}\cdot\vec{R}_{uvw}}$$

$$Q = \eta \vec{b}_1 + \kappa \vec{b}_2 + \mu \vec{b}_3$$

$$\Rightarrow E(\vec{Q}) = F(\vec{Q}) \sum_{uvw} e^{2\pi i \underbrace{(\eta u + \kappa v + \mu w)}_{\text{constructive interference} \equiv \text{integers}}}$$

u, v, w are integers

$\Rightarrow \eta, \kappa, \mu \equiv h\,k\,l$: are integers

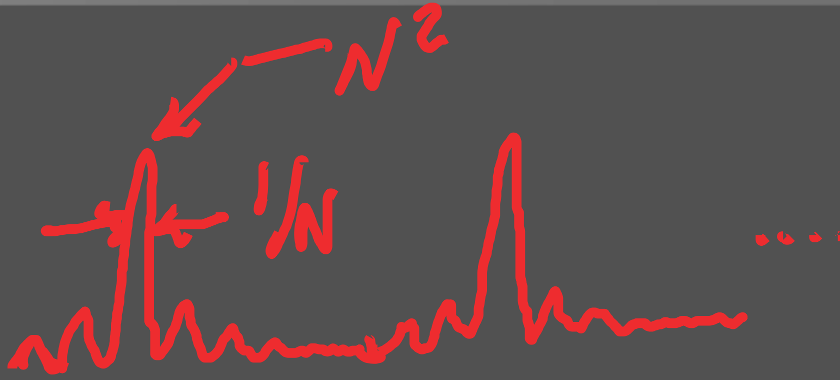
Intensity of diffraction from a small crystal (3)

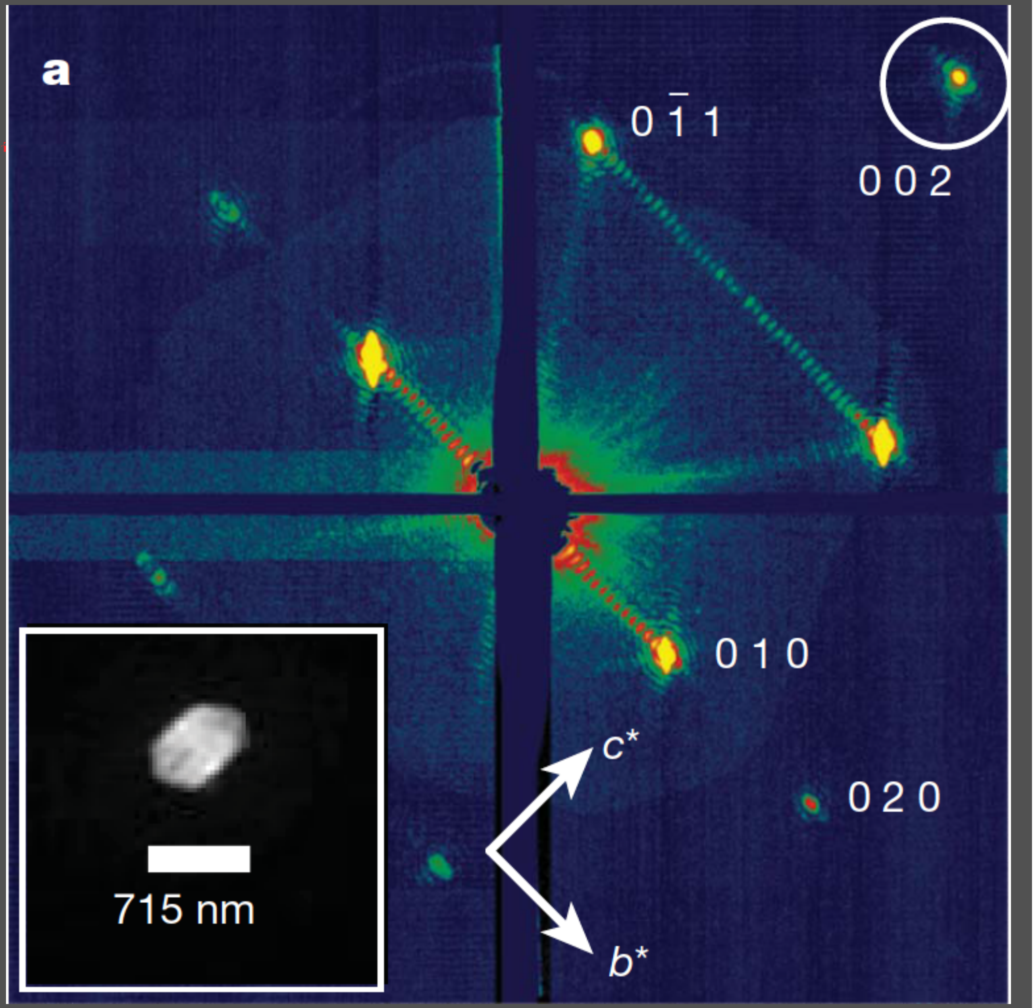
$\Rightarrow E(Q) = NF$; Bragg peak $\vec{Q} = \vec{G}$

$$I(G) \propto EE^* \propto N^2 |F_G|^2$$

$$I \propto |F|^2 \, \frac{\sin^2 \pi N_1 \eta/2}{\sin^2 \pi \eta/2} \cdot \frac{\sin^2 \pi N_2 K/2}{\sin^2 \pi K/2} \cdot \frac{\sin^2 \pi N_3 \mu/2}{\sin^2 \pi \mu/2}$$

Intensity of diffraction from a small crystal (4)





Chapman et al. Nature **470,** 73 (2011)

Integrated intensity more correctly

more correctly:

$$I(G) = I_0 r_e^2 \frac{V_{xtal}}{V_{cell}^2} \cdot \frac{\lambda^3}{\omega} LPA\, |F(G)|^2$$

$$|F_{hkl}|^2 \approx \text{contents of unit cell}$$

$$\Rightarrow I(G) \sim I_0 \cdot \underbrace{\frac{V_{xtal}}{V_{cell}}}_{N}$$

TWs ⇒

nanocrystals

Lattice dynamics

- Diffuse scattering
- Inelastic X-ray Scattering
- FELS open the opportunity to make measurements in the time domain

Determination of phonon dispersion in Al: A diffuse x-ray scattering study

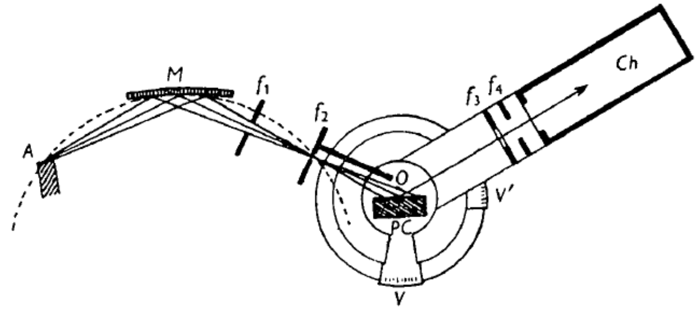
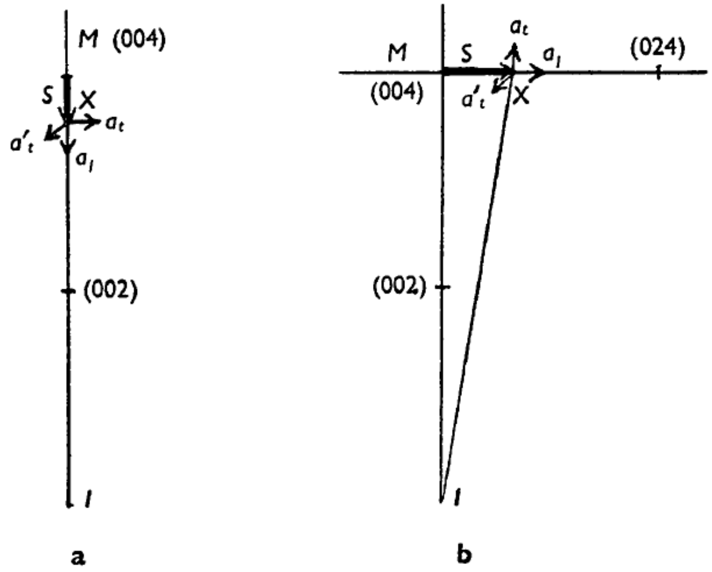
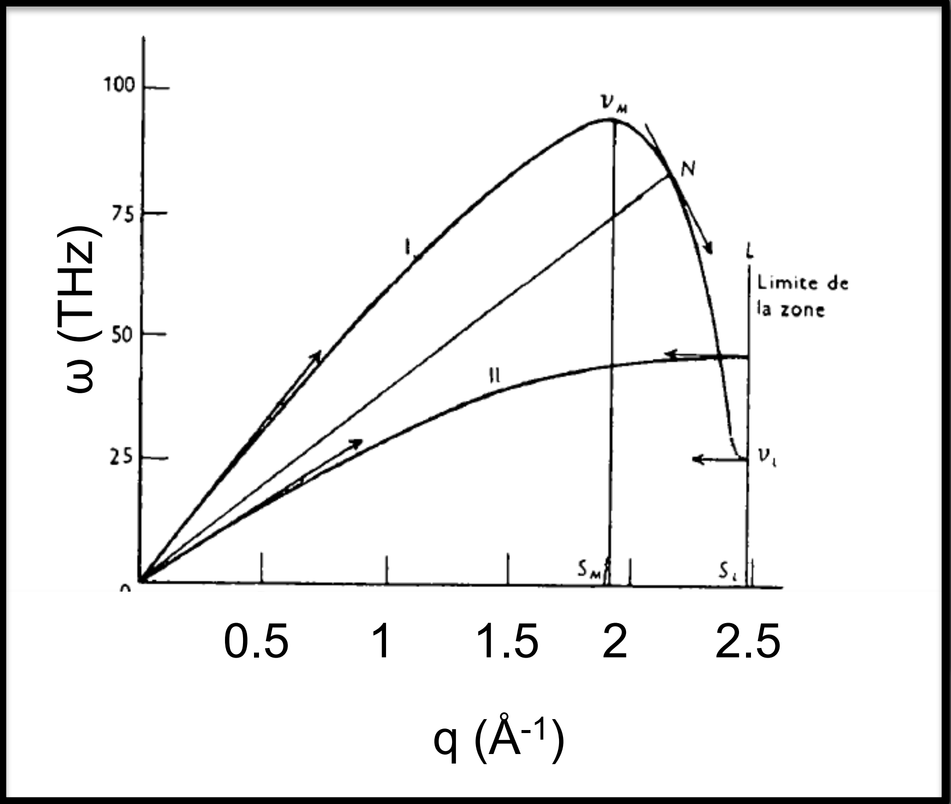


Fig. 1. Schéma général de l'appareillage: *A*, anticathode; *M*, monochromateur; f_1, f_2, f_3, f_4, fentes; *PC*, porte-cristal; *O*, axe du spectromètre; *Ch*, chambre d'ionisation; *V*, *V'*, verniers de lecture.



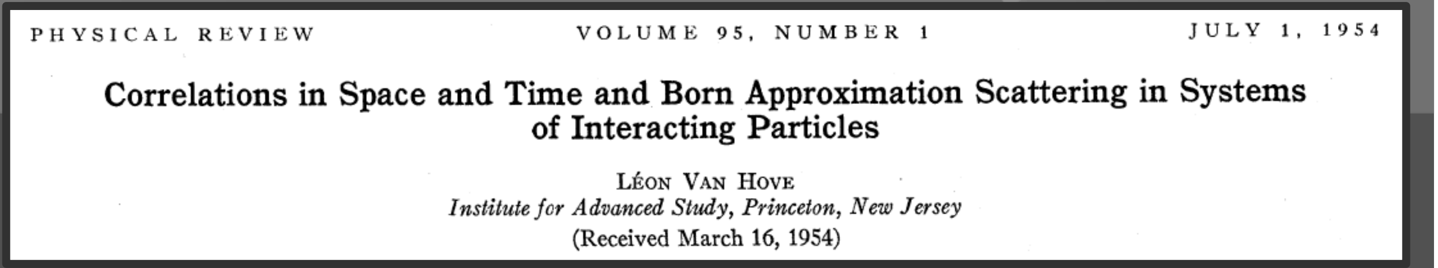


Dispersion des Vitesses des Ondes Acoustiques dans l"Aluminium, P. Olner, Acta Cryst**1**, 57 (1948)

Lattice dynamics

- Diffuse scattering
- Inelastic X-ray Scattering
- FELS open the opportunity to make measurements in the time domain

Inelastic X-ray Scattering (1)



PHYSICAL REVIEW VOLUME 95, NUMBER 1 JULY 1, 1954

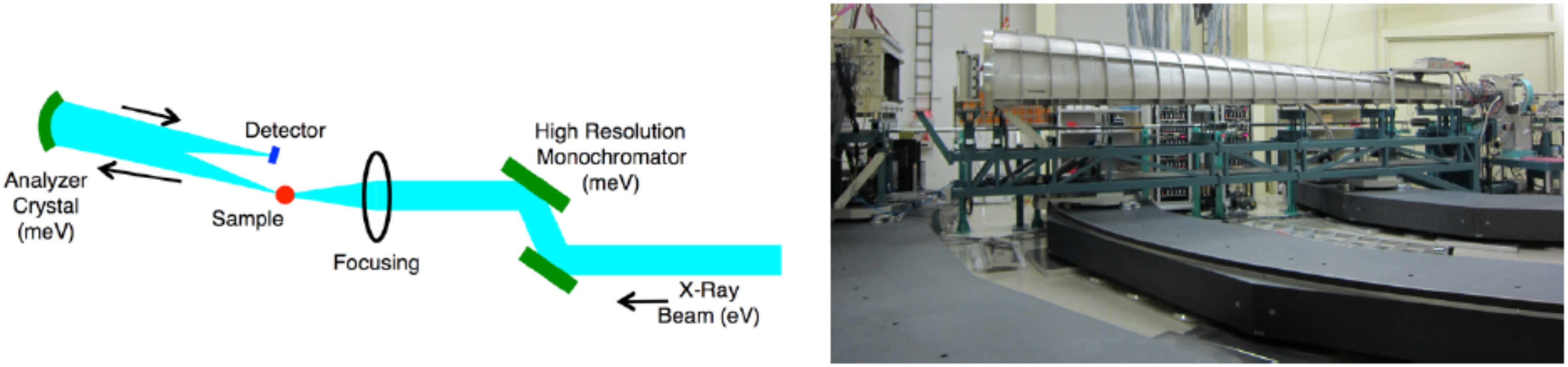
Correlations in Space and Time and Born Approximation Scattering in Systems of Interacting Particles

Léon Van Hove

Institute for Advanced Study, Princeton, New Jersey

(Received March 16, 1954)

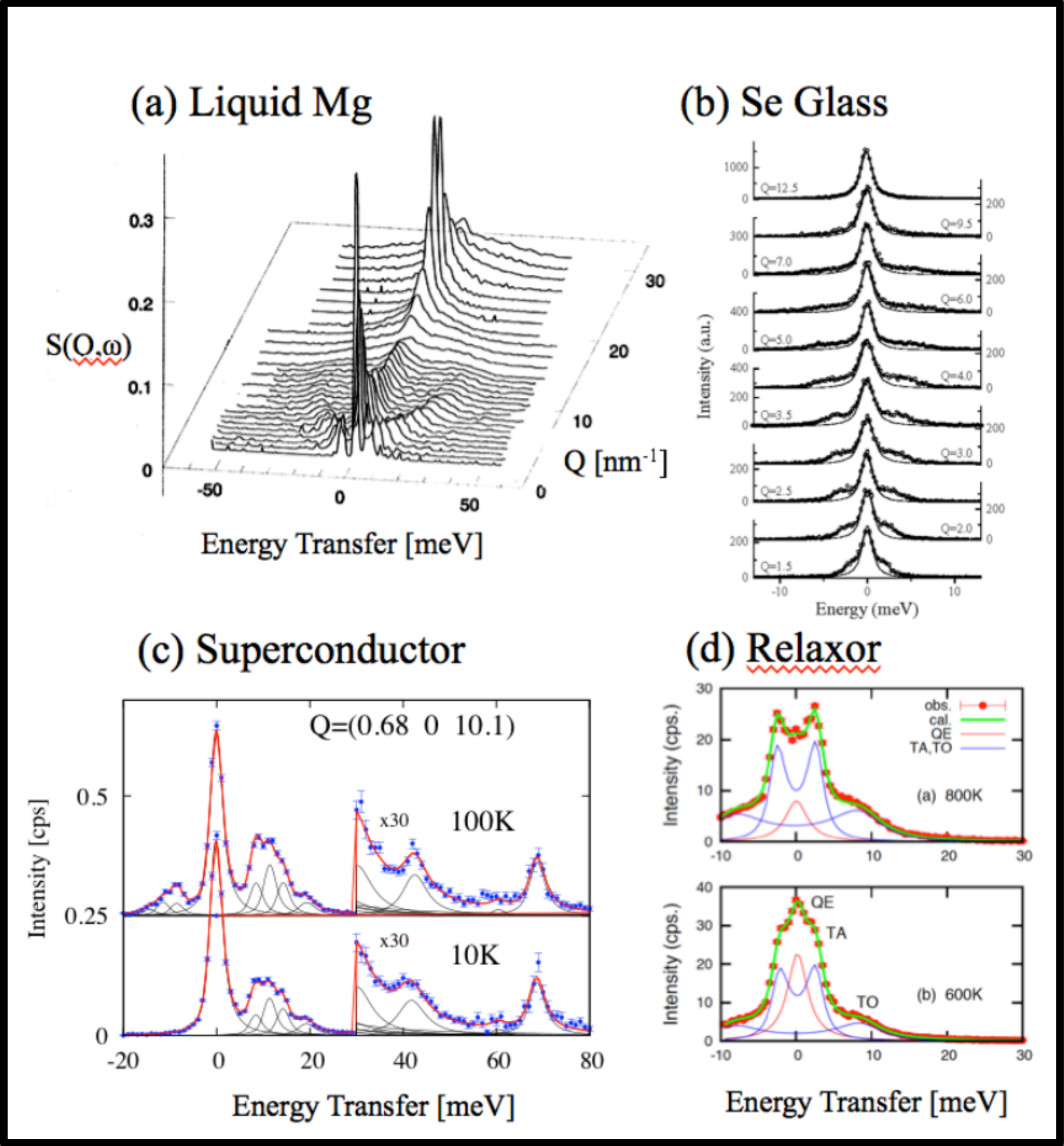
$$S(\vec{Q},\omega) \propto \sum_j \int \mathrm{d}t e^{i\omega t} \langle u_{j,\vec{Q}}(0) u_{j,-\vec{Q}}(t) \rangle$$



Schematic of IXS spectrometer layout (left) and photograph of the 10m arm of the spectrometer of BL43LXU of SPring-8

Alfred Q. R. Baron, arXiv:1504.01098 [cond-mat.mtrl-sci]

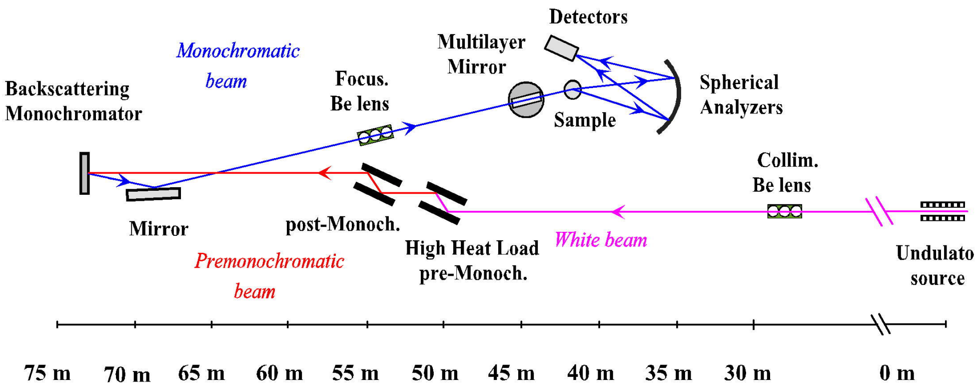
Inelastic X-ray Scattering (2)
(Rogue's gallery)



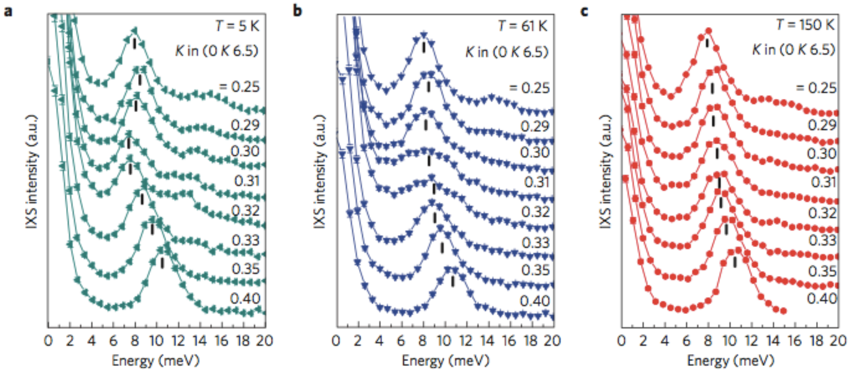
Alfred Q. R. Baron, arXiv:1504.01098 [cond-mat.mtrl-sci]

Inelastic X-ray Scattering:

$$S(\vec{Q},\omega) \propto \sum_j \int \mathrm{d}t\, e^{i\omega t}\langle u_{j,\vec{Q}}(0)\,u_{j,-\vec{Q}}(t)\rangle$$

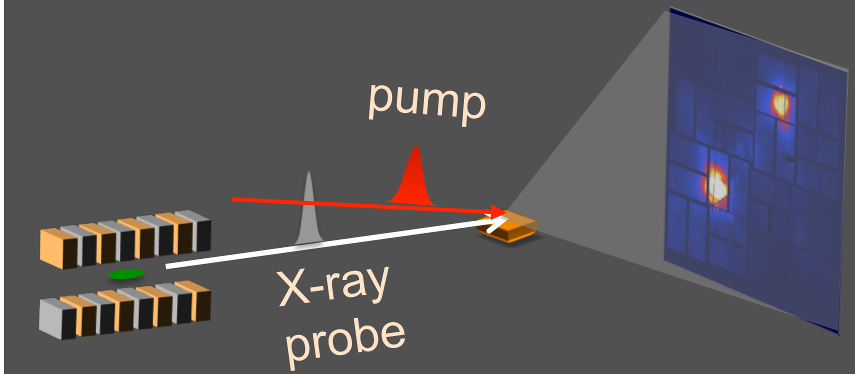


Underdoped YBCO



M. Le Tacon et. al, Nat. Phys. 10,52 (2014)

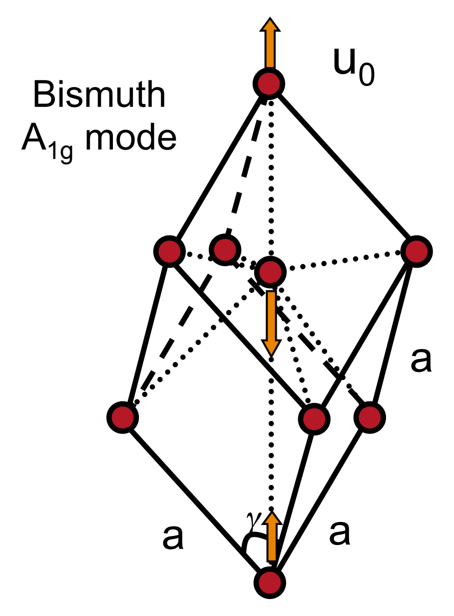
Time and momentum-domain x-ray scattering:

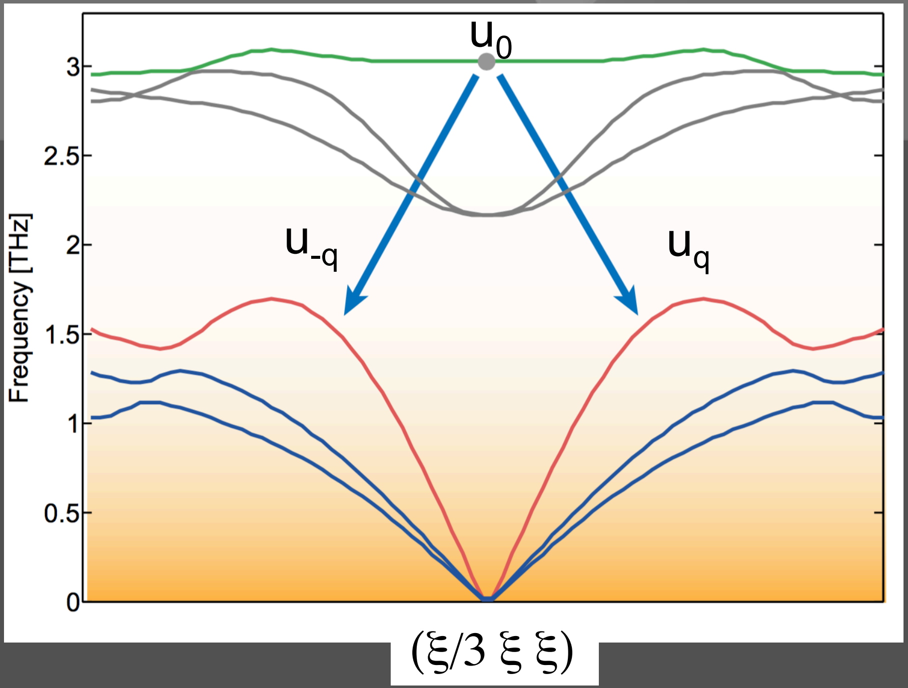


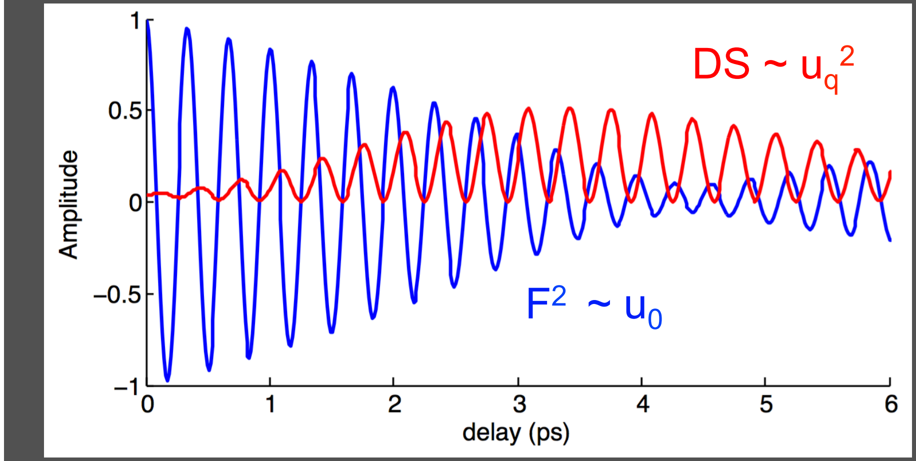
$$S(\vec{Q};\tau) \propto \sum_{j,j'}\langle u_{j,\vec{Q}}(\tau)\,u_{j',-\vec{Q}}(\tau)\rangle$$

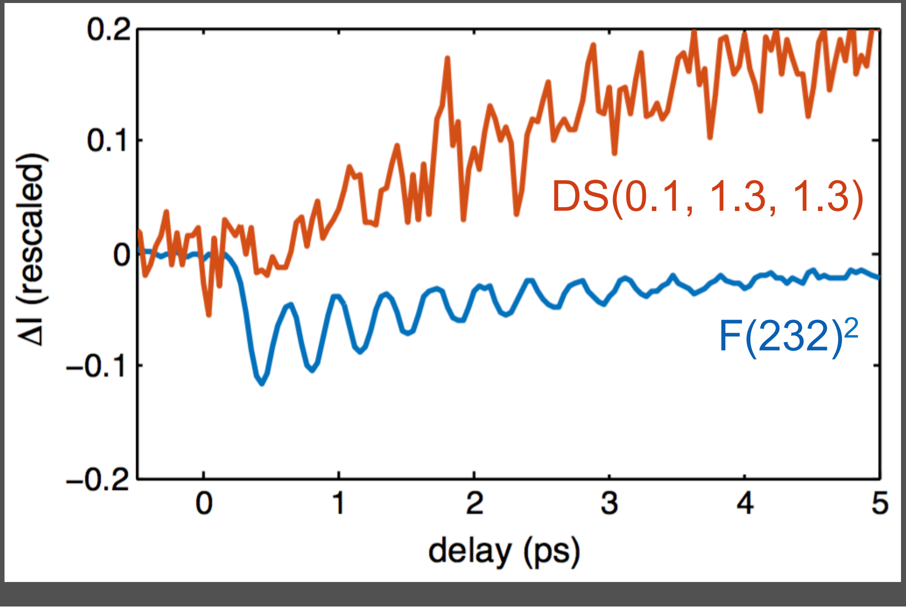
Trigo et al. Nature Physics. 9, 790, 2013

Parametric phonon resonance



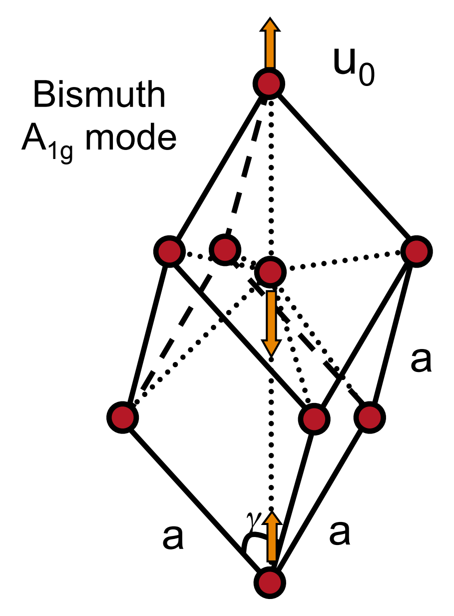


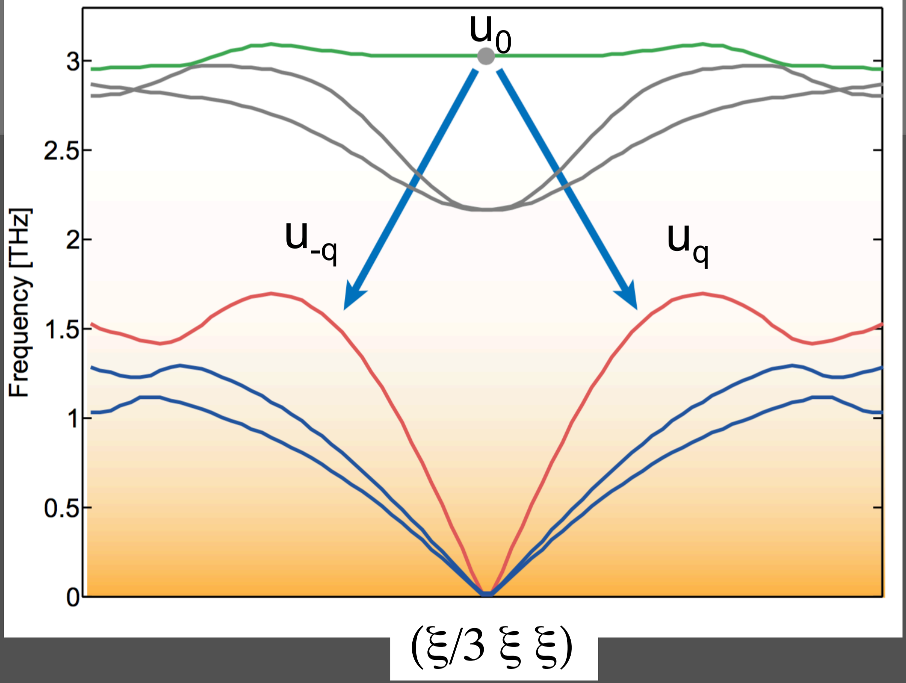


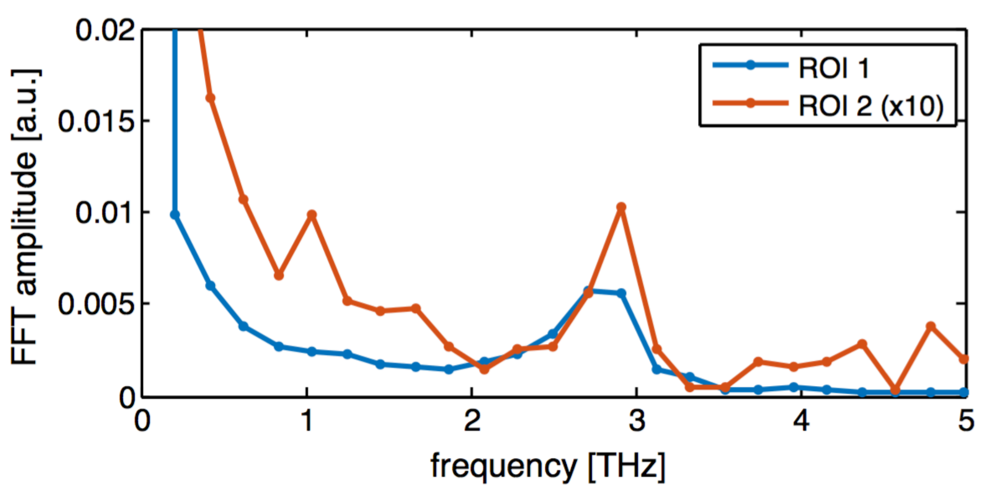


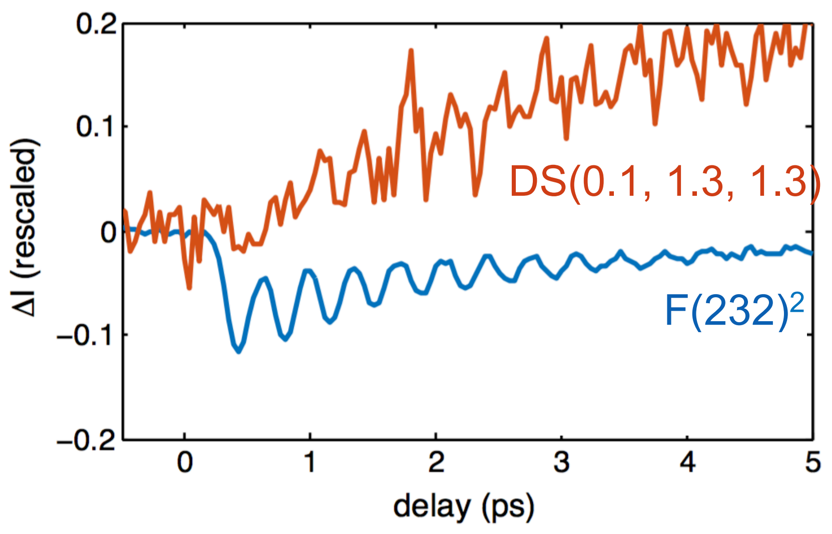
M. Trigo et al., unpublished

Parametric phonon resonance







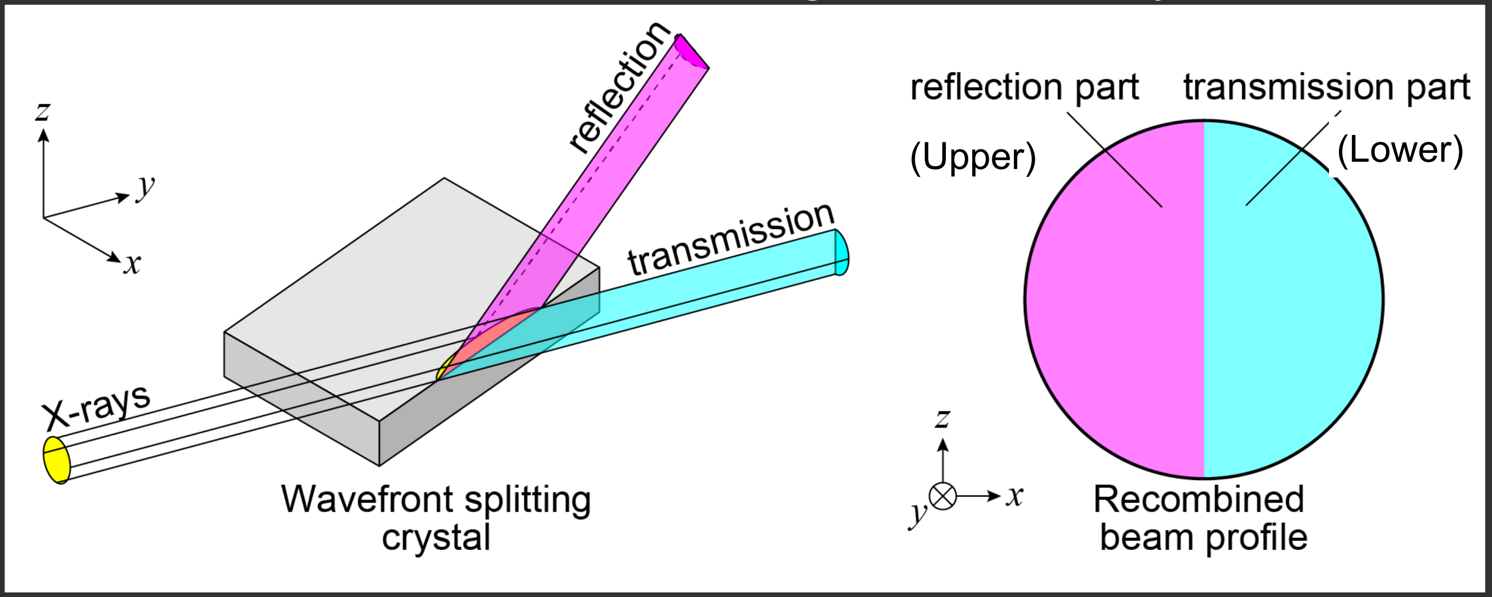


M. Trigo et al., unpublished

X-ray Optics

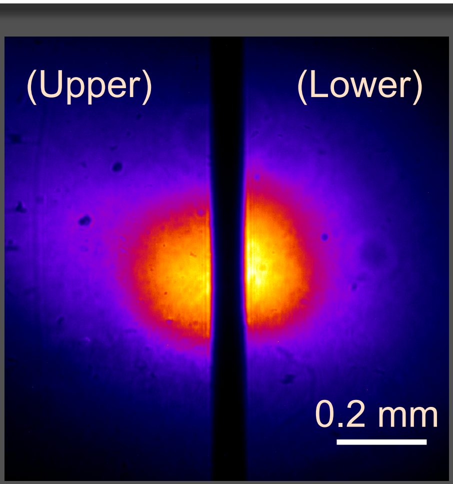
Split/Delay: Wavefront division *Osaka et al., SACLA*

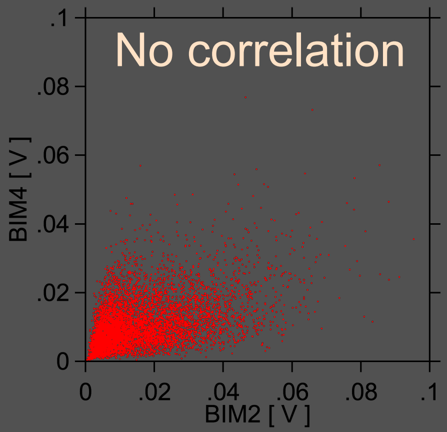
Wavefront division with edge-polished crystals



Advantages:
Reduced lattice strain
Control of intensity ratio
Spectral overlap

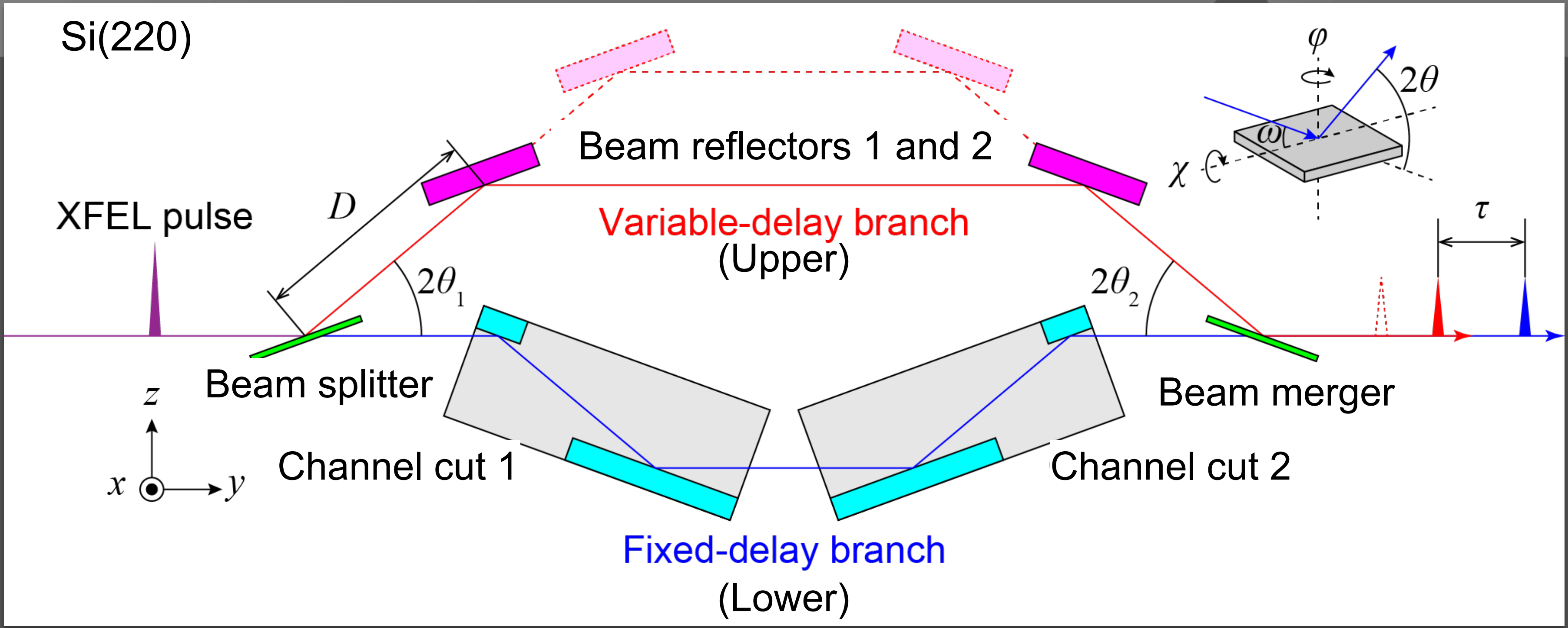
Issues:
Scattering from the edges.
Dead area due to imperfection of the edge.
Influence of pointing and profile fluctuations.





Averaged profile @CCD1 (0.4 m downstream)
Intensity correlation between the split pulses

Crystal arrangement of SDO system at SACLA



- **Crystal diffraction:**
- Large time delays (>ps)
- High energy resolutions ($\Delta E/E < 1\times10^{-4}$)

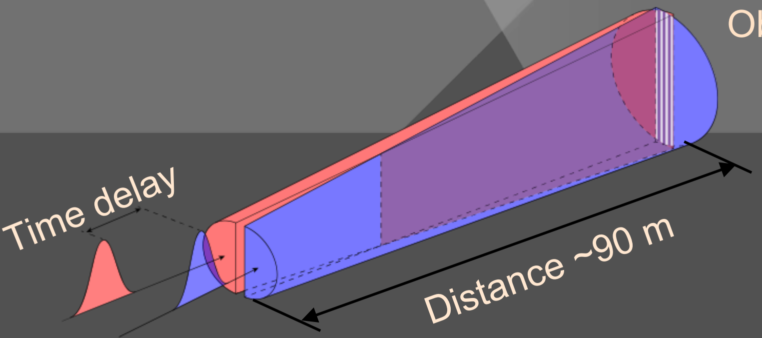
- **Two independent delay branches:**
- Enables access to time zero

- **Use of channel cuts:**
- Much stabilized operation

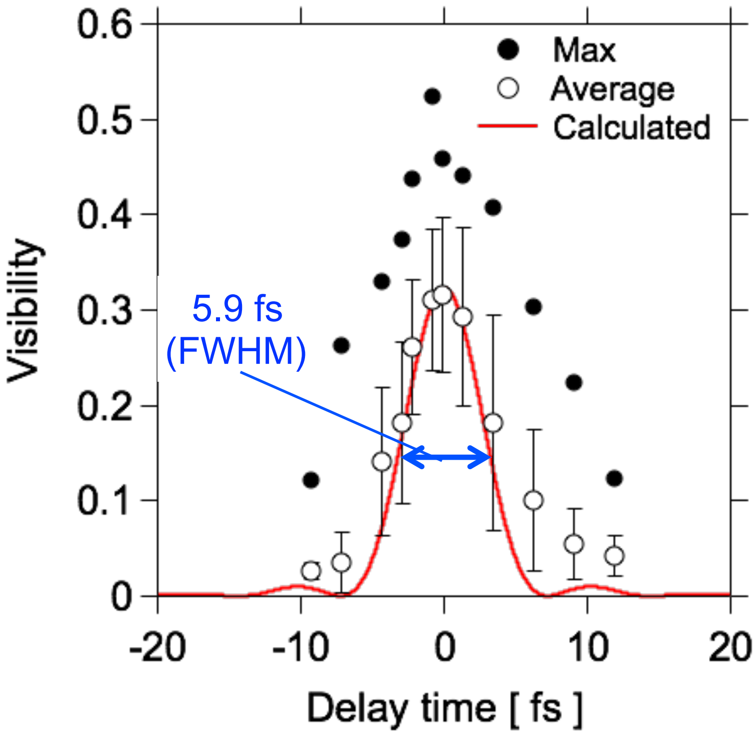
Fine temporal overlap: Interference observation

Observation of **interference fringes** produced between overlapping split pulses in spectrum, space, and **time**.

Visibility curves as a function of the time delay correspond to the coherence characteristic.

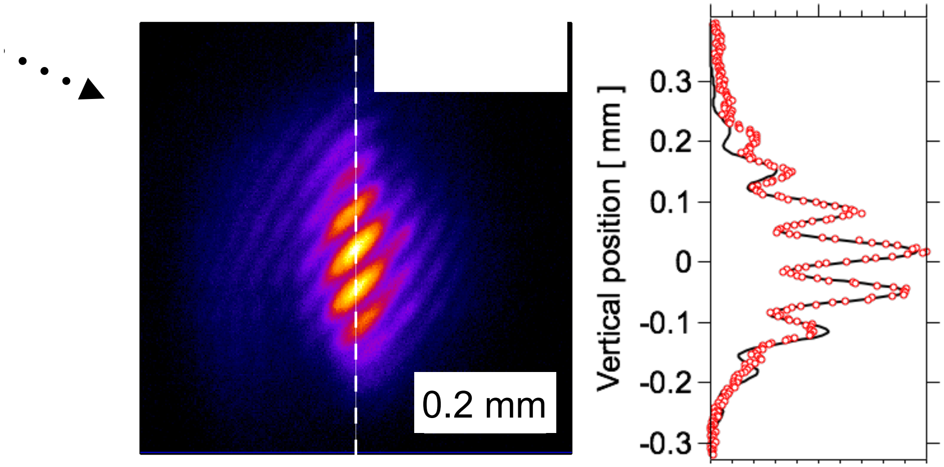


Rough sketch of experiment setup



Fitting function $f(z)$ with a **visibility** V:

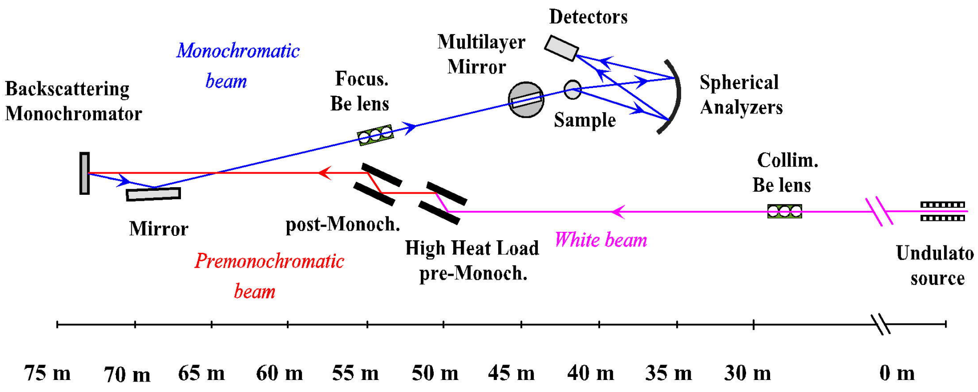
$$f(z) = A\exp\left[-\frac{(z-\mu)^2}{2\sigma^2}\right]\left[1+\cos\left(2\pi\frac{z-\mu}{\delta}+\phi\right)V\right]$$



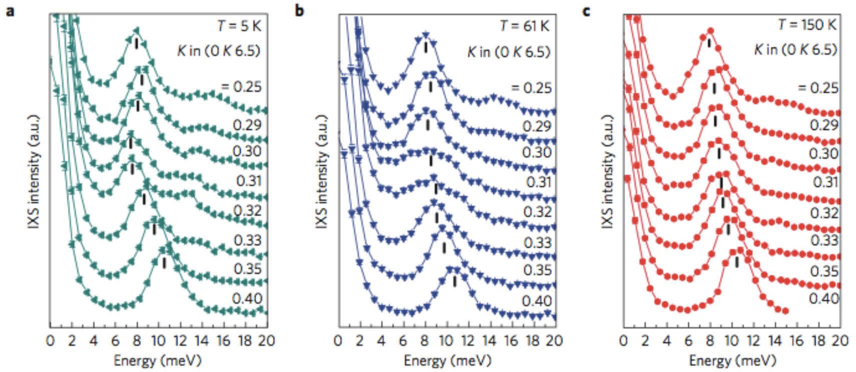
Determination of time zero within <5 fs accuracy

Inelastic X-ray Scattering:

$$S(\vec{Q},\omega) \propto \sum_j \int \mathrm{d}t e^{i\omega t}\langle u_{j,\vec{Q}}(0)u_{j,-\vec{Q}}(t)\rangle$$

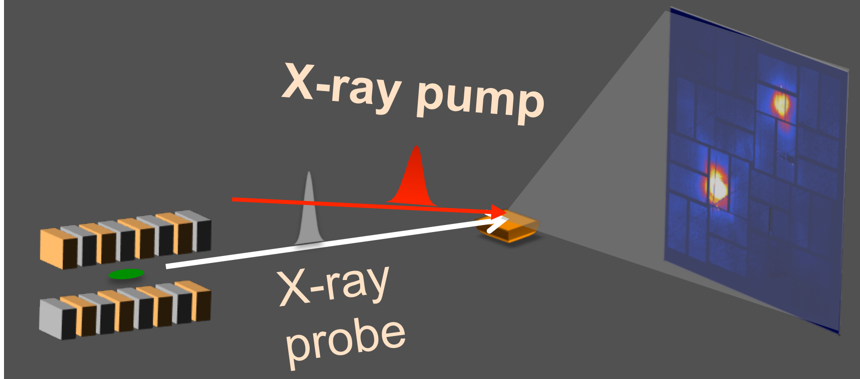


Underdoped YBCO



M. Le Tacon et. al, Nat. Phys. 10,52 (2014)

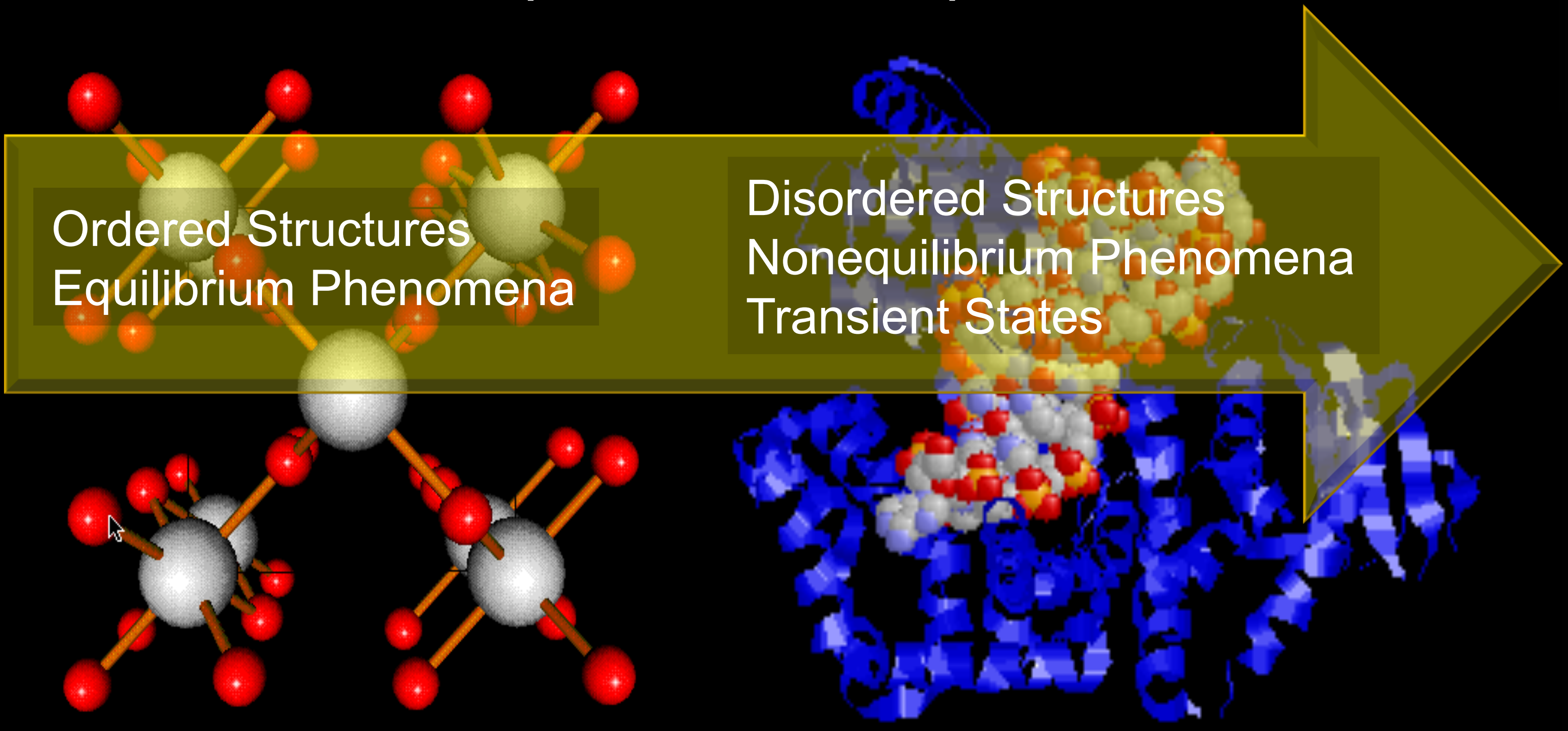
Time and momentum-domain x-ray scattering: X-ray pump-X-ray probe



$$S(\vec{Q};\tau) \propto \sum_{j,j'}\langle u_{j,\vec{Q}}(\tau)u_{j',-\vec{Q}}(\tau)\rangle$$

Trigo et al. Nature Physics. 9, 790, 2013

Future Role of FELs (After H. Dosch)



Ordered Structures
Equilibrium Phenomena

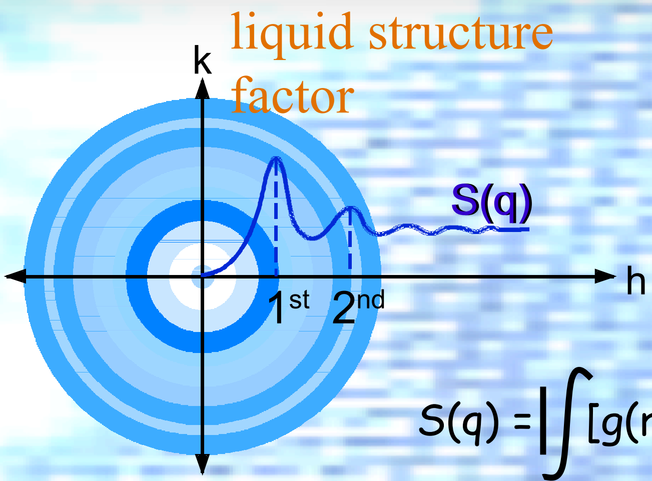
Disordered Structures
Nonequilibrium Phenomena
Transient States

Era of Crystalline Matter
Conventional X-ray Probes

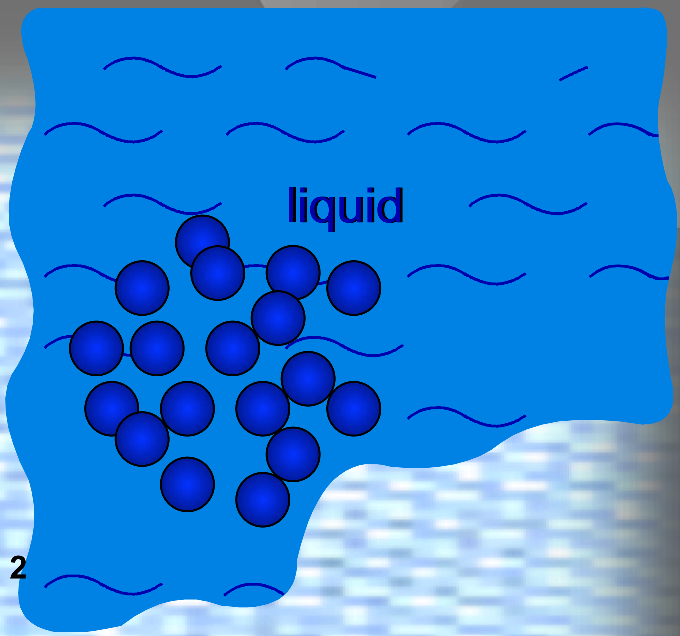
Era of Disordered Matter
Coherent X-ray Probes

1900 2000 future

Local Order in Liquids



$$S(q) = \left| \int [g(r)-1]\, e^{iqr}\, dr \right|^2$$



pair correlation function

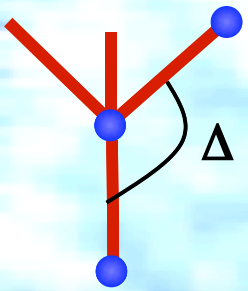
$g(r) = \langle \rho(r)\rho(0) \rangle$

(radial information)

thermal average:
temporal
partially coherent beam

Bond-orientational order $g(\Delta)$

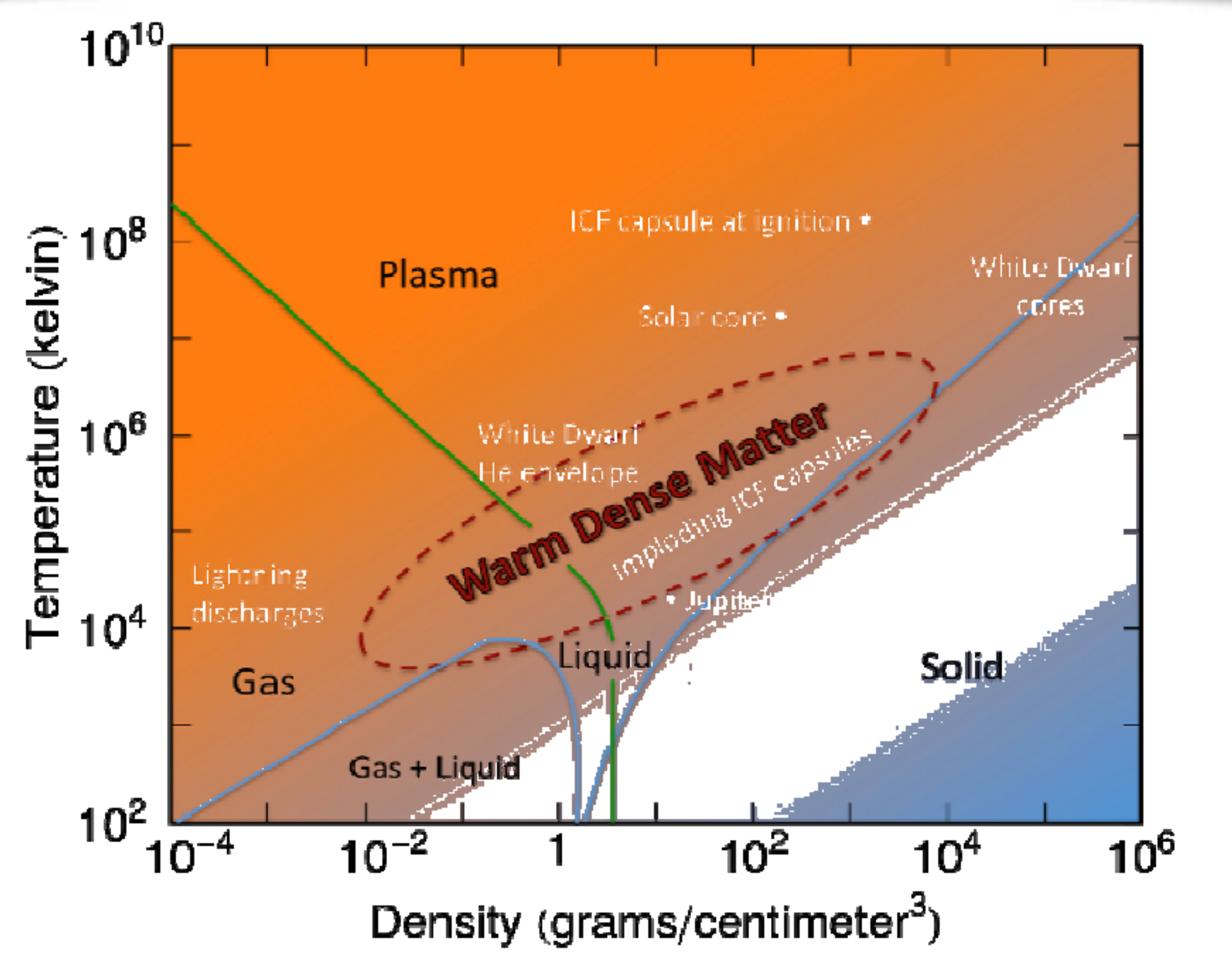
azimuthal information



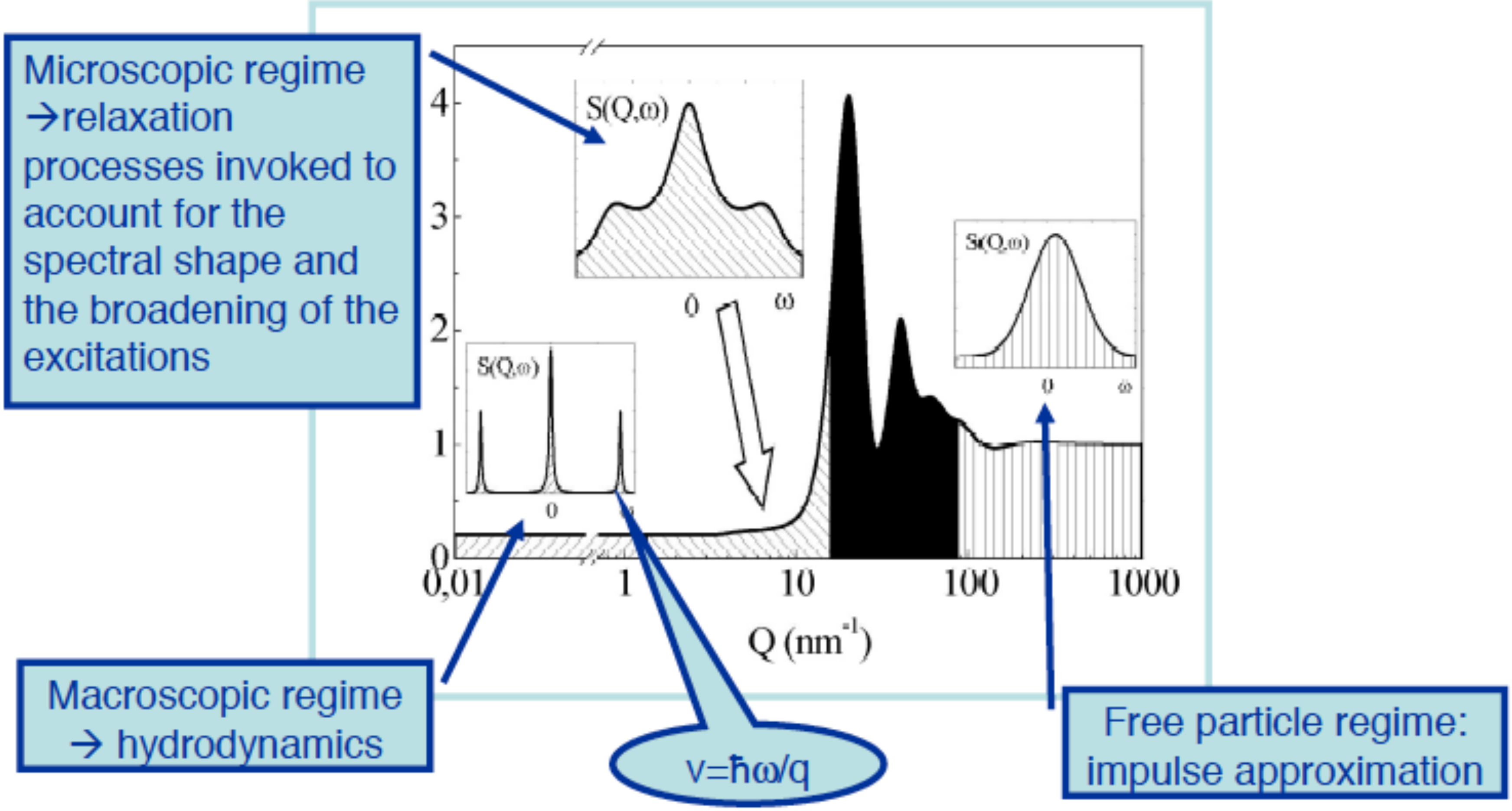
to measure angles in nanoworld:
beyond 2-body correlations !!

(After H. Dosch)

Warm dense matter



Dynamic – inelastic x-ray scattering in liquids (1)



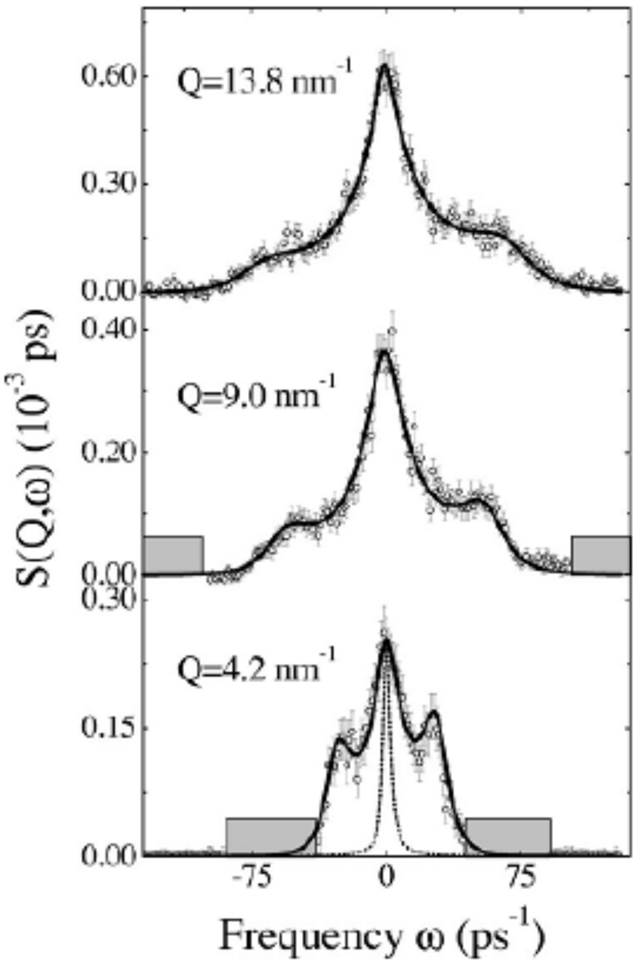
Microscopic regime
→relaxation processes invoked to account for the spectral shape and the broadening of the excitations

Macroscopic regime
→ hydrodynamics

v=ħω/q

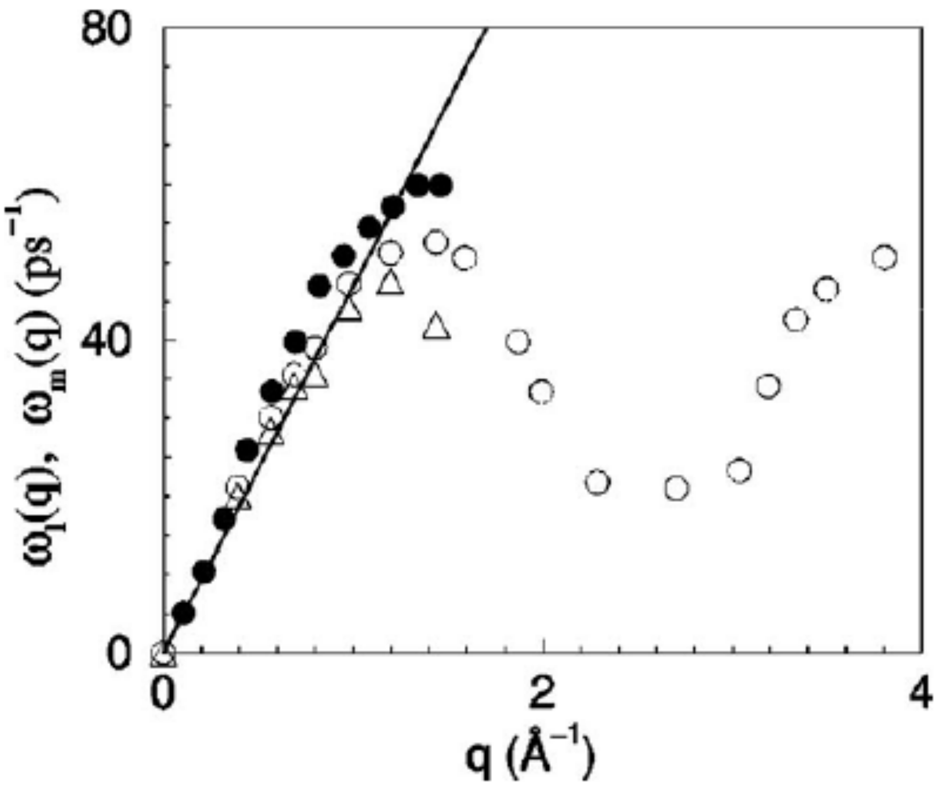
Free particle regime:
impulse approximation

Dynamic – inelastic x-ray scattering in liquids (2)



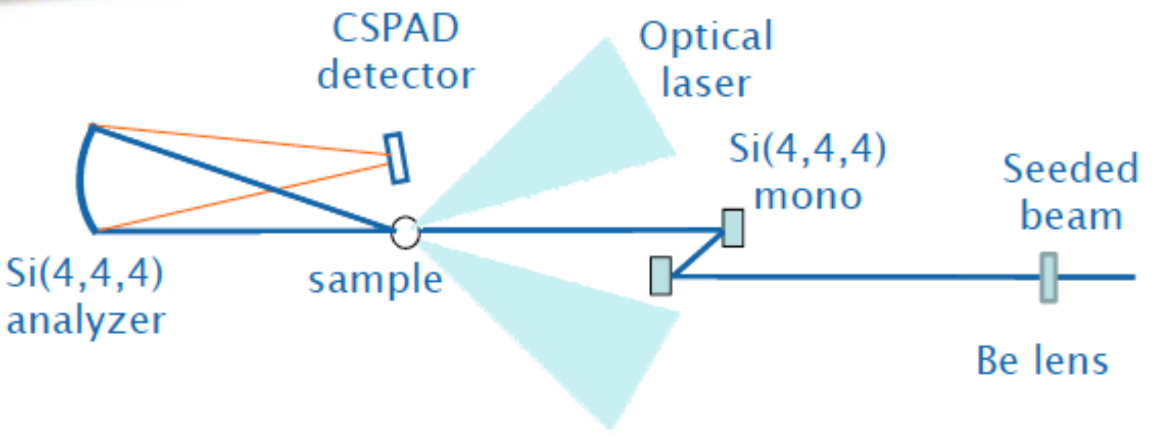
Scopigno et al., PRE 63, 011210 (00)

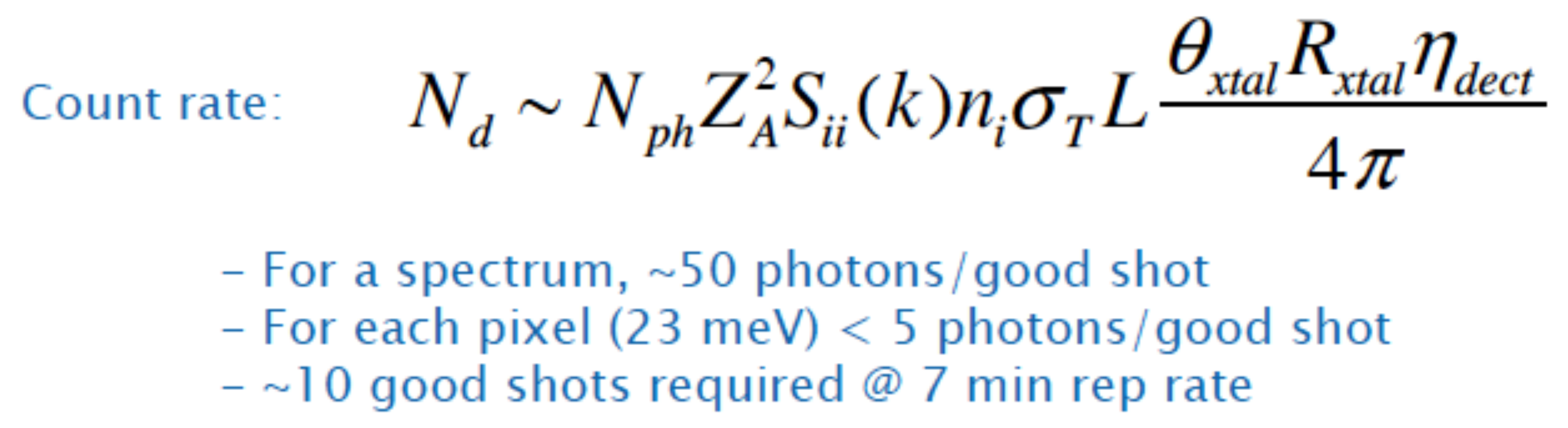
Dispersion curve:
solid-like behavior



Gonzalez et al., PRB 65, 084201 (02)

Inelastic scattering from Al – T=2 eV, $\rho=2\rho_0$




Count rate:

$$N_d \sim N_{ph} Z_A^2 S_{ii}(k) n_i \sigma_T L \frac{\theta_{xtal} R_{xtal} \eta_{dect}}{4\pi}$$

- For a spectrum, ~50 photons/good shot
- For each pixel (23 meV) < 5 photons/good shot
- ~10 good shots required @ 7 min rep rate

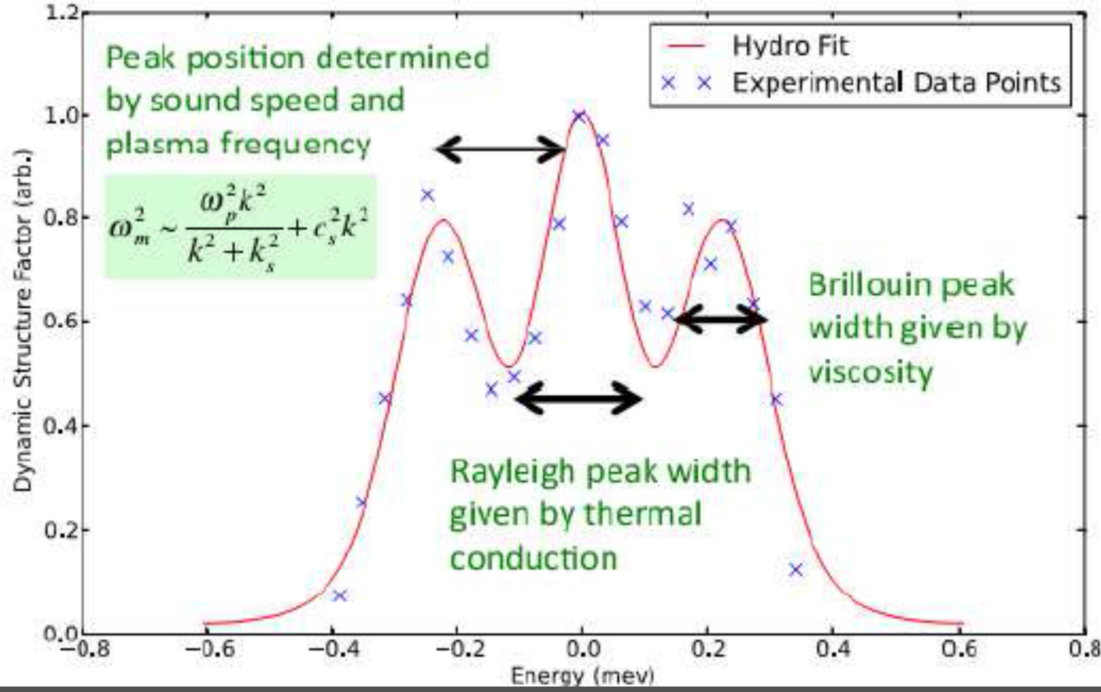
First results – S(k,ω) for one k value

Rayleigh peak

Brillouin peaks

$$\frac{S(k,\omega)}{S(k)/2\pi}=\frac{\gamma}{\gamma-1}\frac{c_s^2k^2}{\omega_m^2}\frac{2D_Tk^2}{\omega^2+(D_Tk^2)^2}+\frac{\omega_{m\gamma}}{\omega_m}\left[\frac{\sigma k^2}{(\omega+\omega_m)^2+(\sigma k^2)^2}+\frac{\sigma k^2}{(\omega-\omega_m)^2+(\sigma k^2)^2}\right]$$

Schmidt et al., PRE (2012)



Peak position determined by sound speed and plasma frequency

$$\omega_m^2 \sim \frac{\omega_p^2k^2}{k^2+k_s^2}+c_s^2k^2$$

Brillouin peak width given by viscosity

Rayleigh peak width given by thermal conduction

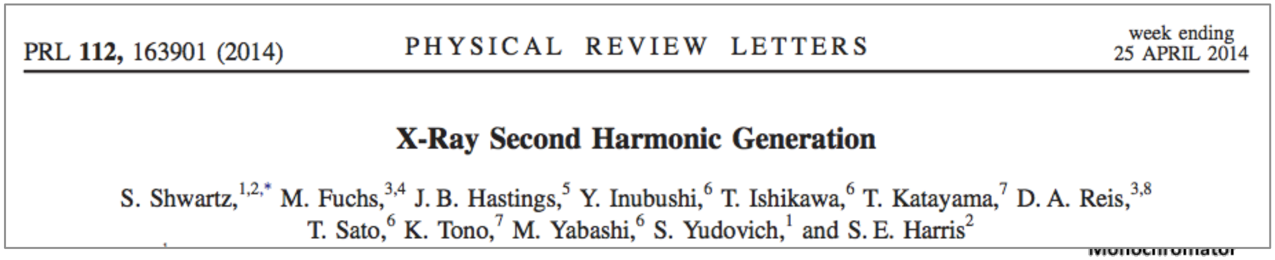
Conclusions

- Structural biology – real nanocrystals
- X-ray split and delay – x-ray pump x-ray probe – new vision for lattice dynamics
- Liquids and novel, transient states of matter
- non-linear x-ray scattering
- …….

[redacted]

Also from two to one:
second harmonic generation

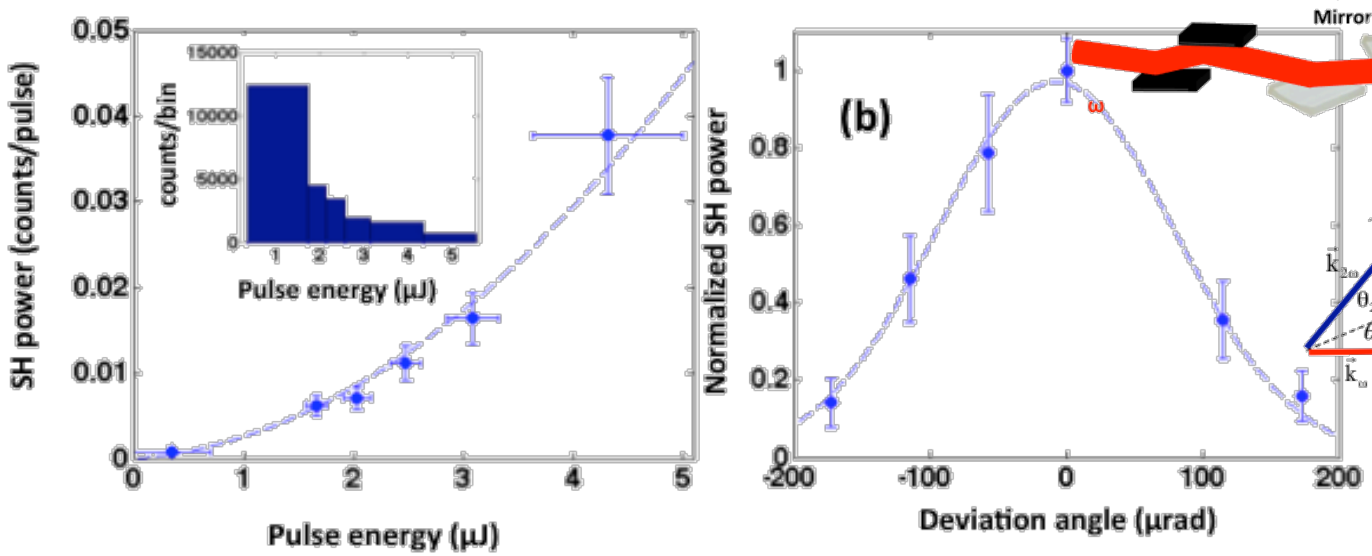
$$J_{2\omega} \propto \eta \rho_G J_\omega$$

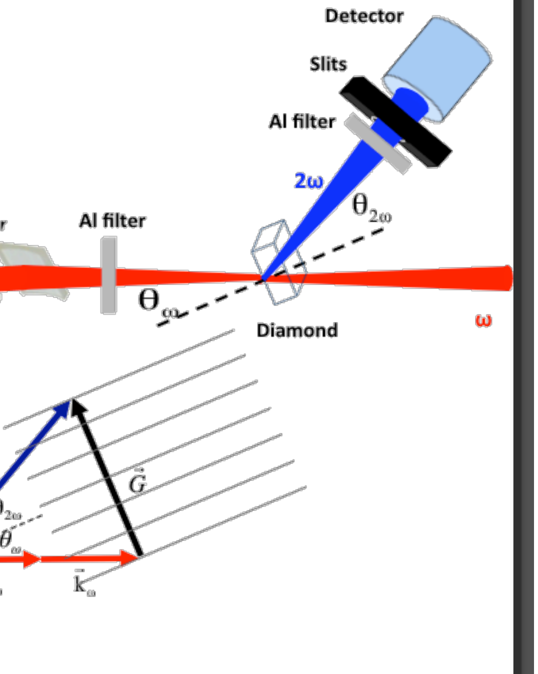


PRL **112,** 163901 (2014) PHYSICAL REVIEW LETTERS week ending 25 APRIL 2014

X-Ray Second Harmonic Generation

S. Shwartz,[1,2,*] M. Fuchs,[3,4] J. B. Hastings,[5] Y. Inubushi,[6] T. Ishikawa,[6] T. Katayama,[7] D. A. Reis,[3,8]
T. Sato,[6] K. Tono,[7] M. Yabashi,[6] S. Yudovich,[1] and S. E. Harris[2]





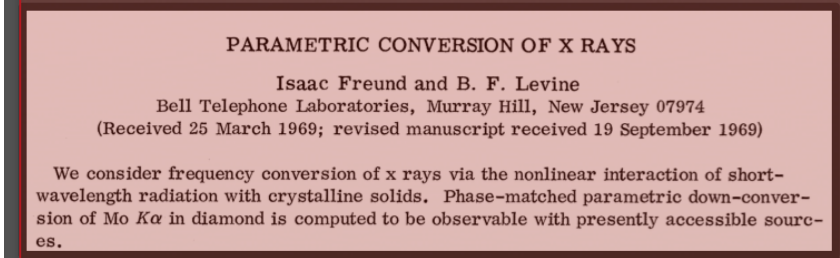
Peak Efficiency ~6X10^{-11} @10^{16} W/cm^2 (~3X10^9 V/cm)

Made possible by advent of hard x-ray FELs (2009 LCLS, 2011 SACLA)

Non-Linear X-ray Optics
(before FELs)

From one x ray to two…

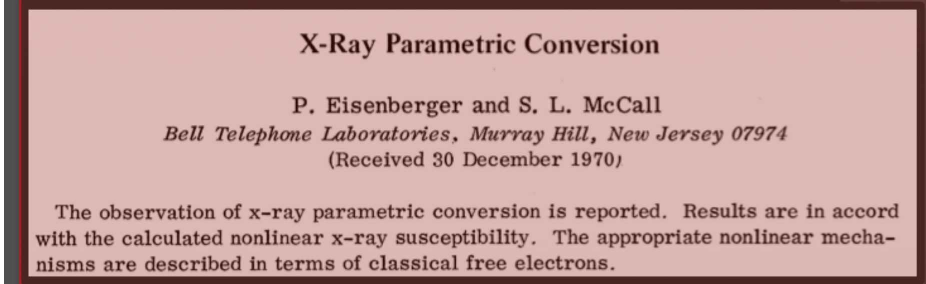
Where we were



PARAMETRIC CONVERSION OF X RAYS

Isaac Freund and B. F. Levine
Bell Telephone Laboratories, Murray Hill, New Jersey 07974
(Received 25 March 1969; revised manuscript received 19 September 1969)

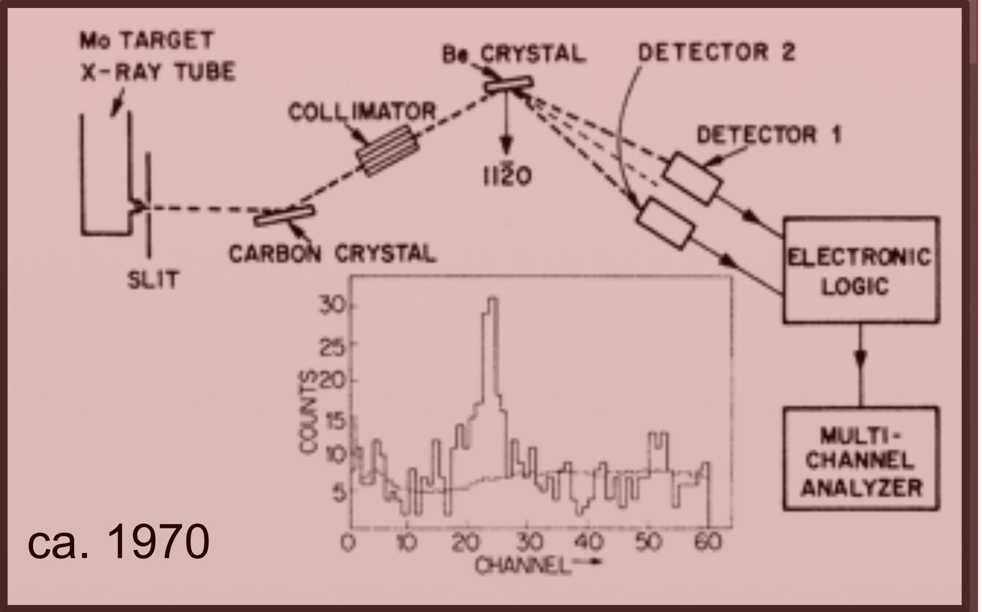
We consider frequency conversion of x rays via the nonlinear interaction of short-wavelength radiation with crystalline solids. Phase-matched parametric down-conversion of Mo *Kα* in diamond is computed to be observable with presently accessible sources.



X-Ray Parametric Conversion

P. Eisenberger and S. L. McCall
Bell Telephone Laboratories, Murray Hill, New Jersey 07974
(Received 30 December 1970)

The observation of x-ray parametric conversion is reported. Results are in accord with the calculated nonlinear x-ray susceptibility. The appropriate nonlinear mechanisms are described in terms of classical free electrons.



ca. 1970

~ 1 coincidence count/100 minute

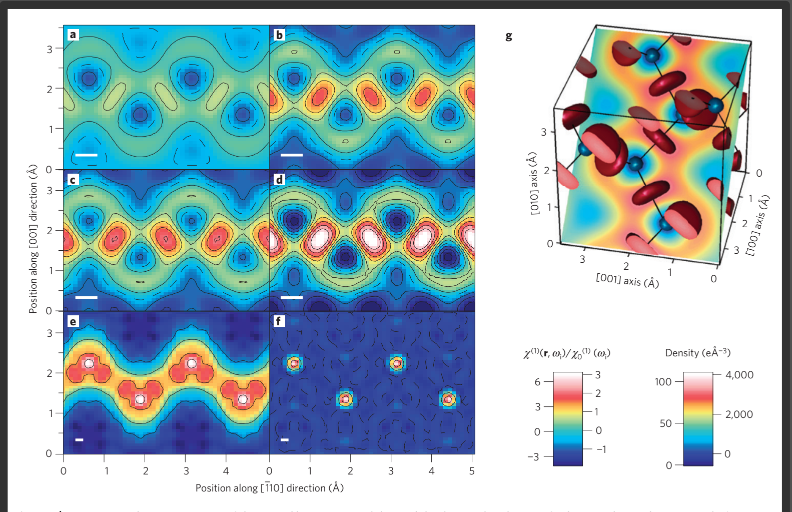
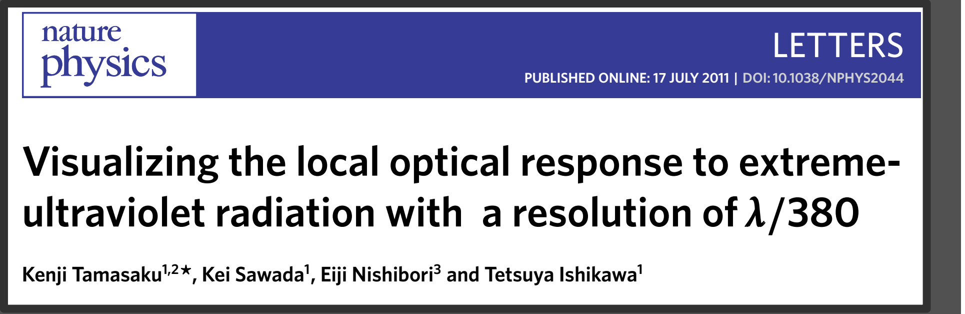


Figure 3 | Reconstructed microstructures of the optical linear susceptibility and the density distribution of valence and core electrons. a–d, The



nature physics

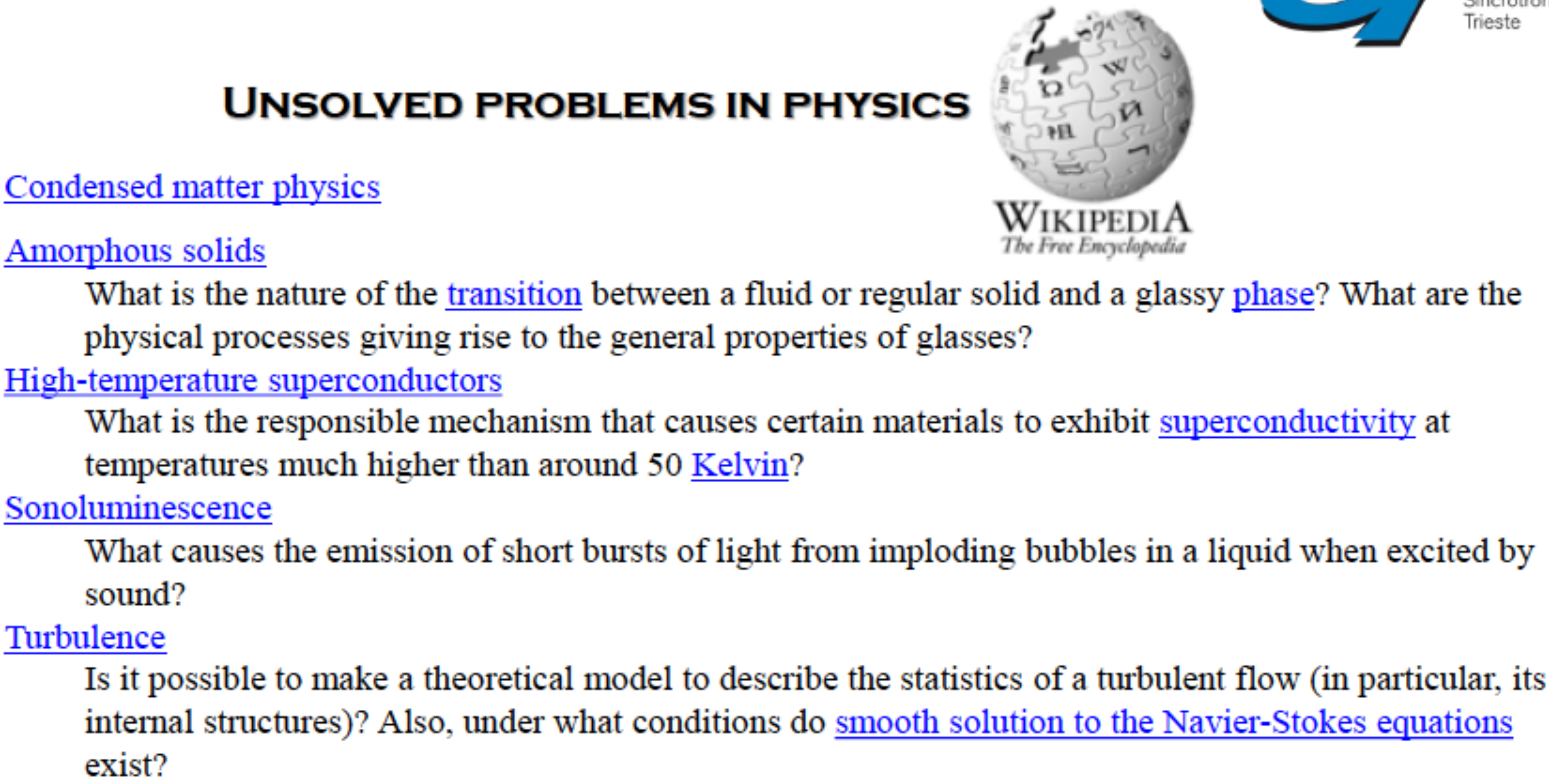
LETTERS

Visualizing the local optical response to extreme-ultraviolet radiation with a resolution of $\lambda/380$

Kenji Tamasaku[1,2]★, Kei Sawada[1], Eiji Nishibori[3] and Tetsuya Ishikawa[1]

ca. 2011

Why Disordered Systems ?



UNSOLVED PROBLEMS IN PHYSICS

WIKIPEDIA
The Free Encyclopedia

Condensed matter physics

Amorphous solids

What is the nature of the transition between a fluid or regular solid and a glassy phase? What are the physical processes giving rise to the general properties of glasses?

High-temperature superconductors

What is the responsible mechanism that causes certain materials to exhibit superconductivity at temperatures much higher than around 50 Kelvin?

Sonoluminescence

What causes the emission of short bursts of light from imploding bubbles in a liquid when excited by sound?

Turbulence

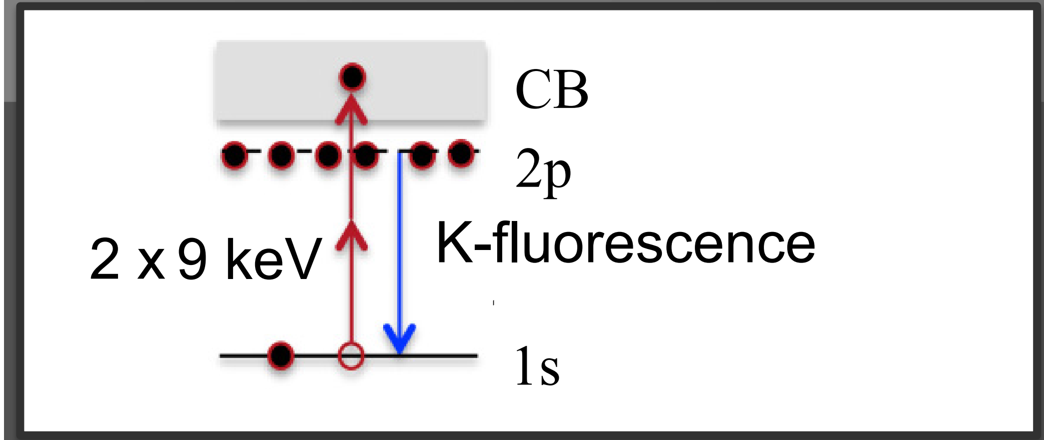
Is it possible to make a theoretical model to describe the statistics of a turbulent flow (in particular, its internal structures)? Also, under what conditions do smooth solution to the Navier-Stokes equations exist?

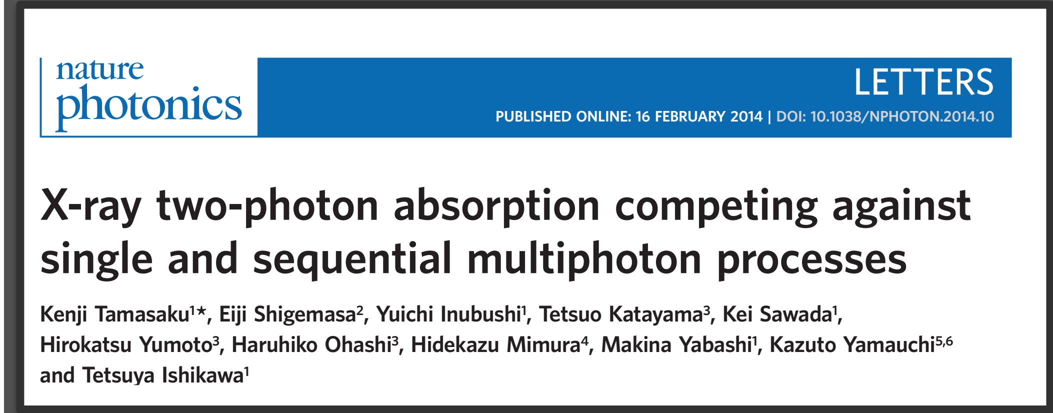
Glass is a very general state of condensed matter → a large variety of systems can be transformed from liquid to glass

The liquid-glass transition cannot be described in the framework of classical phase transitions since T_g depends on the quenching rate → one cannot define an order parameter showing a critical behaviour at T_g

Two-photon K-shell absorption

Non-relativistic Z^{-6} dipole scaling



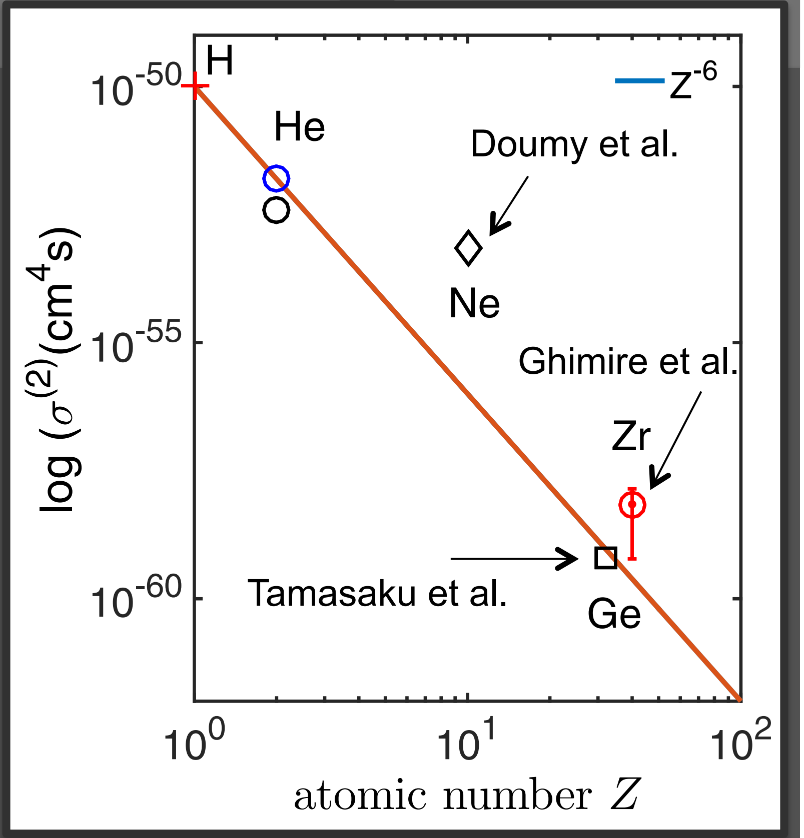


nature photonics

LETTERS

X-ray two-photon absorption competing against single and sequential multiphoton processes

Kenji Tamasaku[1]*, Eiji Shigemasa[2], Yuichi Inubushi[1], Tetsuo Katayama[3], Kei Sawada[1], Hirokatsu Yumoto[3], Haruhiko Ohashi[3], Hidekazu Mimura[4], Makina Yabashi[1], Kazuto Yamauchi[5,6] and Tetsuya Ishikawa[1]



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Nonsequential two-photon absorption from the *K* shell in solid zirconium

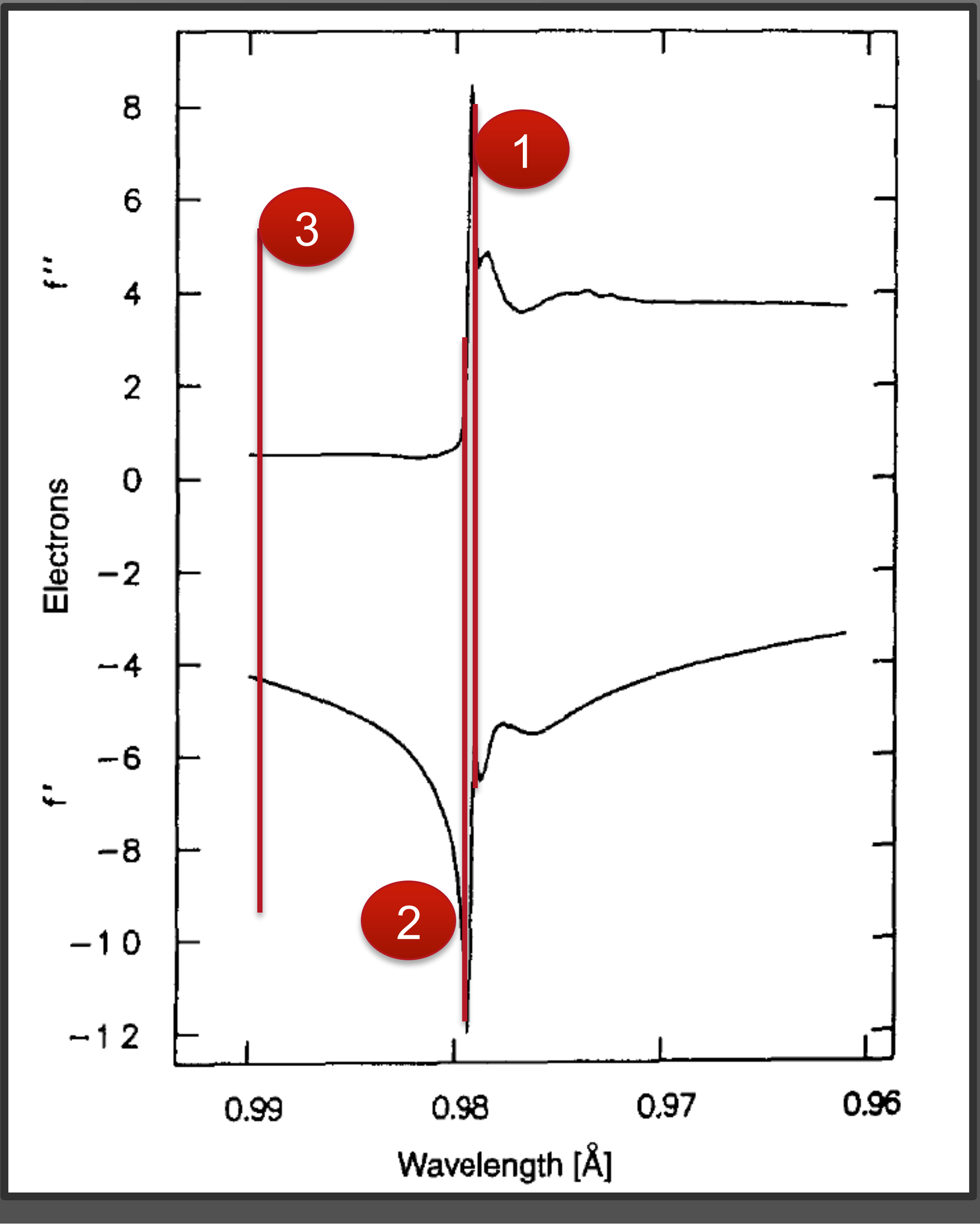
Shambhu Ghimire,[1,*] Matthias Fuchs,[2] Jerry Hastings,[3] Sven C. Herrmann,[4] Yuichi Inubushi,[5] Jack Pines,[4] Sharon Shwartz,[6] Makina Yabashi,[5] and David A. Reis[1,7,†]

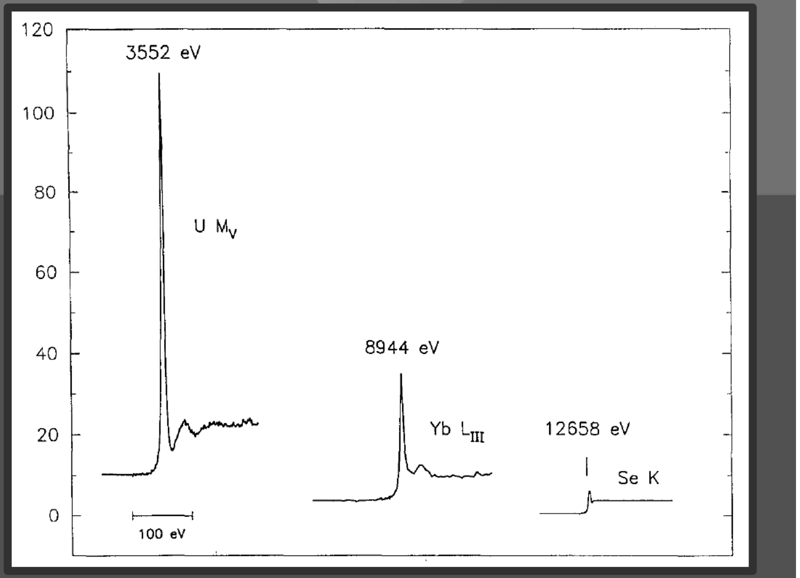
Non-Linear X-ray Optics
(again)

Structural biology:
Reciprocal space imaging of macro molecules

- Perutz et al. Hemoglobin
- MAD phasing
- *FEL*-Serial Femtosecond Crystallography
- *Storage Ring*-Serial crystallography
- Single particle imaging

MAD Phasing





Wayne A. Hendrickson and Craig M. Ogata, Methods in Enzymology Volume 276, 494 (1997)

Lattice dynamics

- Diffuse scattering
- Inelastic X-ray Scattering
- FELS open the opportunity to make measurements in the time domain