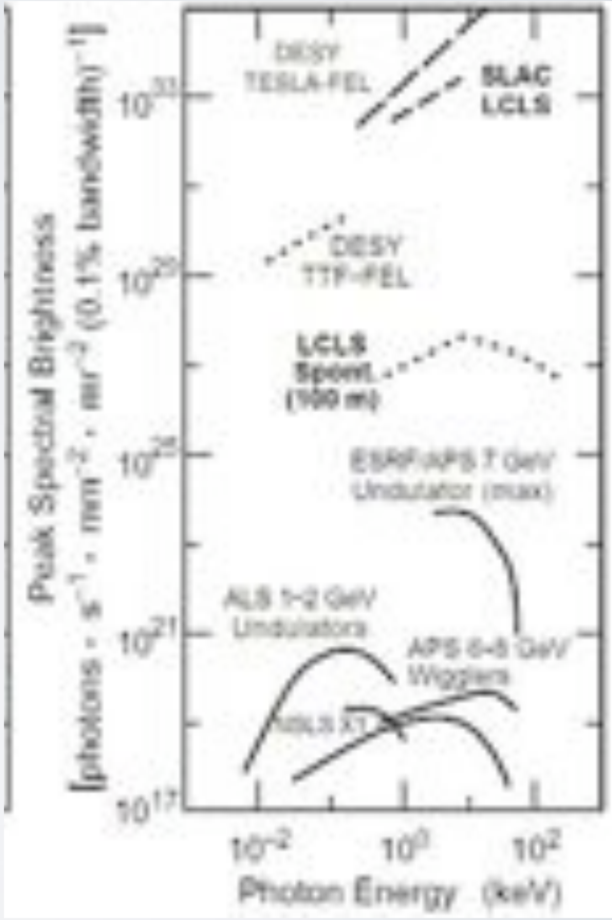
INVERSE PROBLEMS IN X-RAY SCIENCE

STEFANO MARCHESINI

- **overview of photon science**
- **inverse problems**
    - **linear (under-determined): tomography**
    - **non-linear: diffraction methods**
- **Inverse problems and the data deluge**



When new materials are discovered we want to know...

Structure (atomic resolution for crystals)

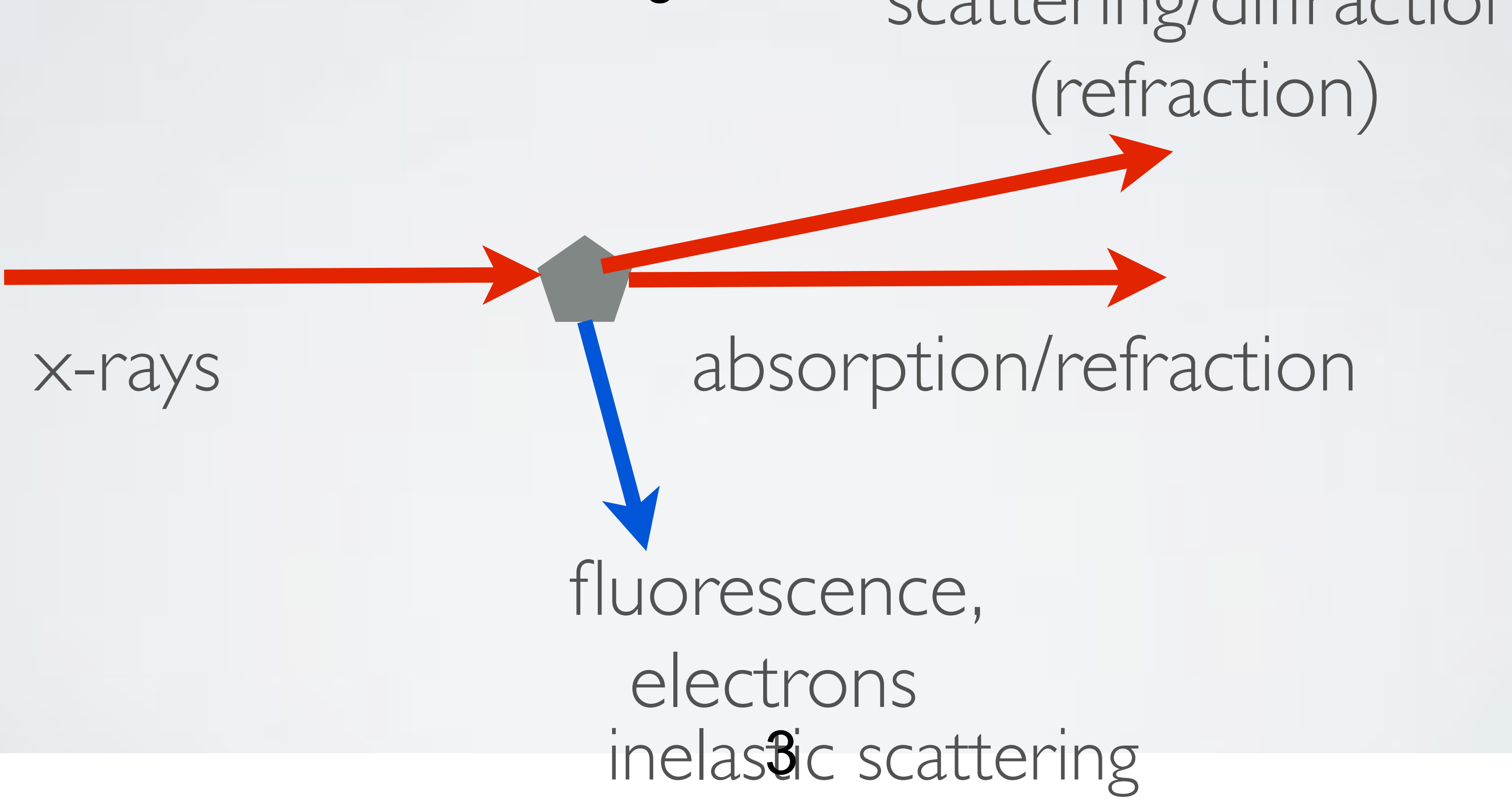
Electronic properties

Chemical structure

Dynamical evolution

X-ray interaction with matter

- wavelength ~atom size
- penetration ~microns-m
- Energy: ~valence bands and up
- time: femtoseconds and longer



APPLICATIONS OF X-RAY SCIENCE

- Solid state physics (spectroscopy: photoemission, inelastic scattering...)
- Biology (protein structures, cell imaging)
- Chemistry, Materials science (photovoltaics, batteries, catalysis, etc)
- Earth science
- Space science (stardust, metrology)
- Archeology
- Semiconductors (lithography, metrology)
- ...

ADVANCED LIGHT SOURCE SYNCHROTRON

FACILITY FACTS

~210	~600	$57.5M	5300	39 + beam test facility
Total ALS staff	Refereed publications per year	Average operating budget per year	Average number of operating hours per year	Number of beamlines

Experimental Techniques

Diffraction (~50%)

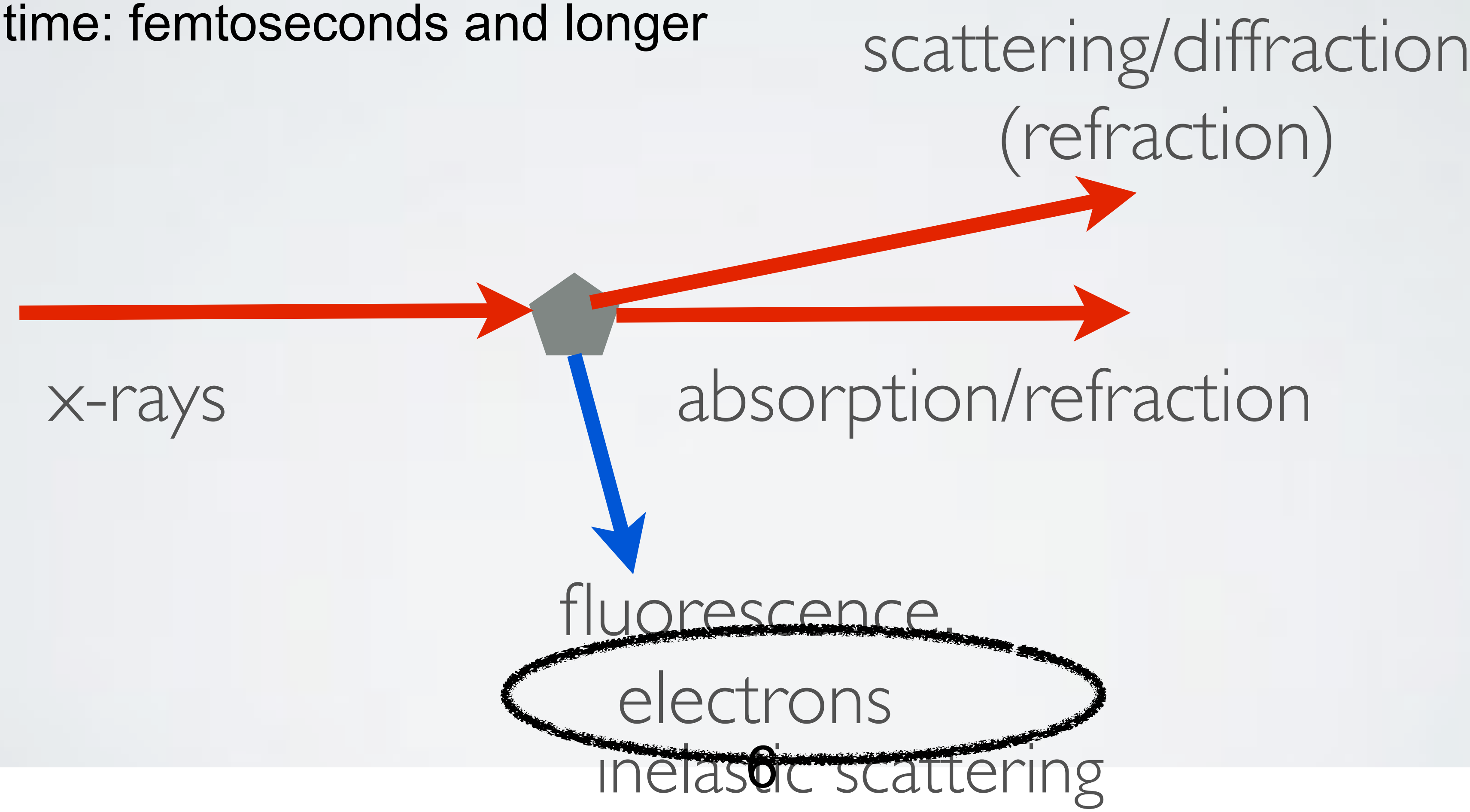
spectro-microscopy (~25%)

spectroscopy (~25%)

- 45 beamlines around the ALS
- covers a large range of science
  - structural biology
  - energy sciences
  - geo / environmental sciences
  - condensed matter physics
  - chemistry
  - materials science
  - EUV lithography

X-ray interaction with matter

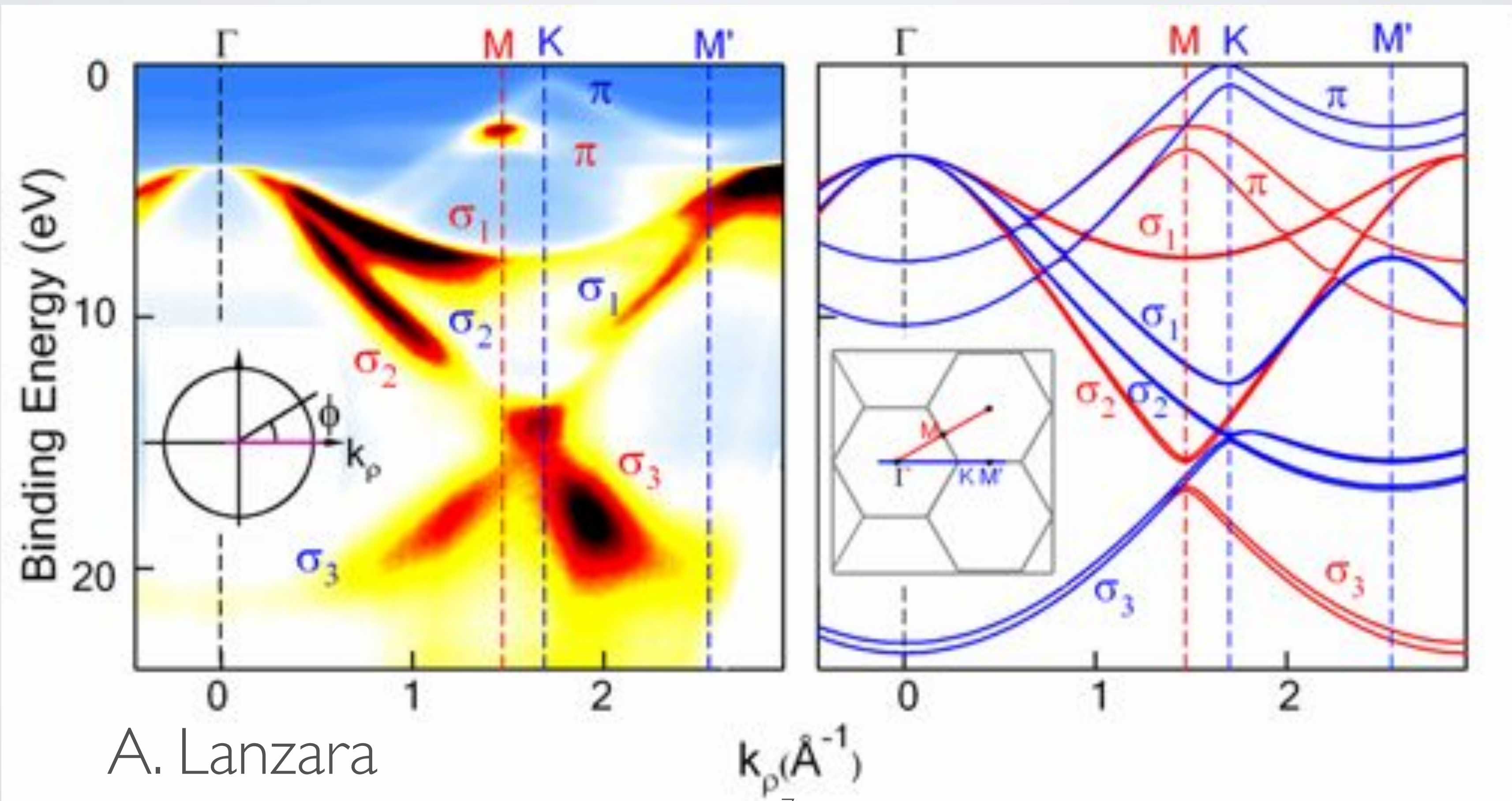
- wavelength ~atom size
- penetration ~microns-m
- Energy: ~valence bands and up
- time: femtoseconds and longer



DISCOVERY OF MATERIALS ELECTRONIC BAND STRUCTURE

Solid state physics

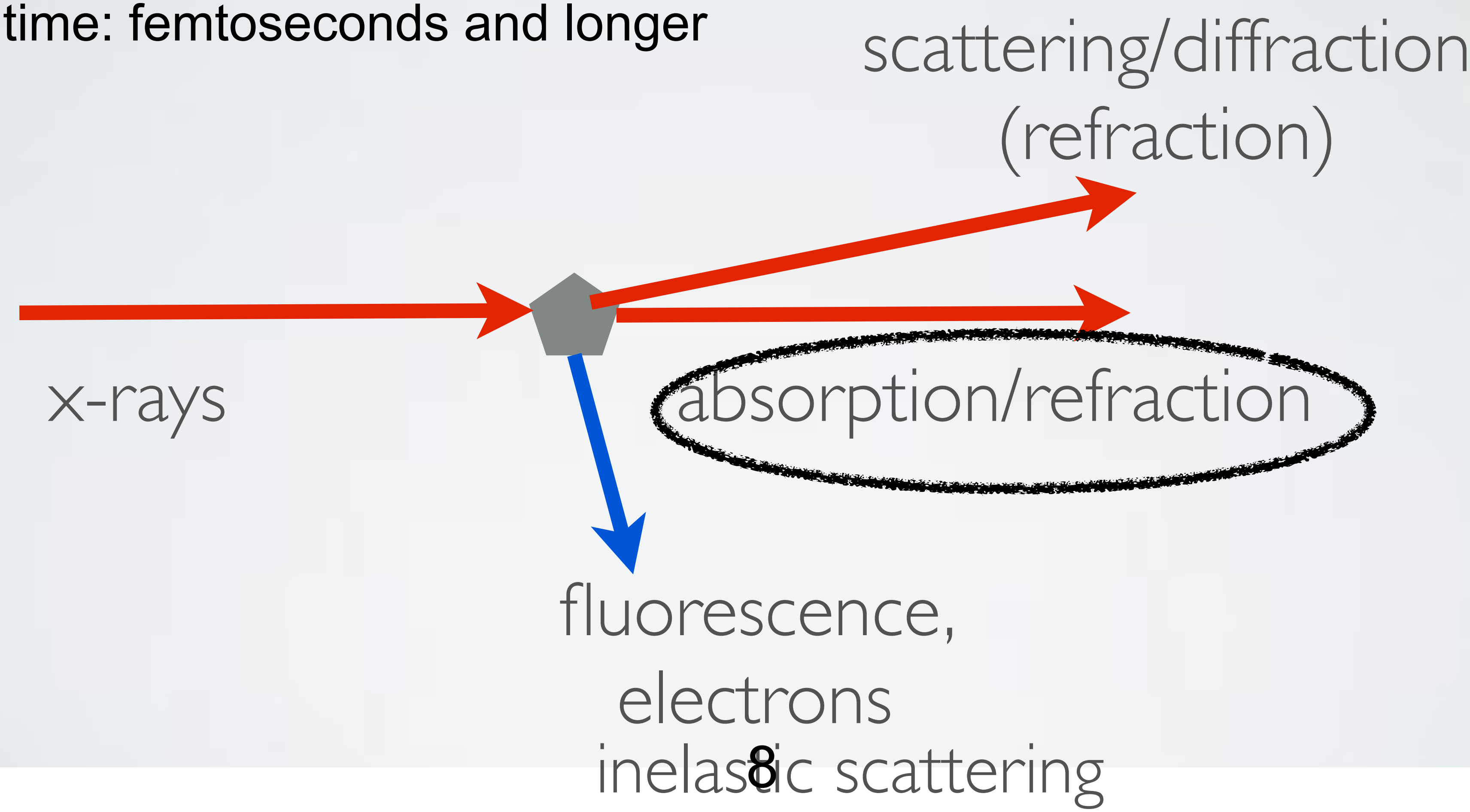
Angle resolved photoemission   Model



A. Lanzara

X-ray interaction with matter

- wavelength ~atom size
- penetration ~microns-m
- Energy: ~valence bands and up
- time: femtoseconds and longer



absorption

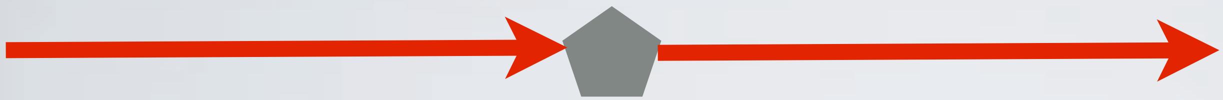
Contrast mechanism

$$I(x) = I_0(x)T(x)$$

absorption coefficient

$$e^{-\alpha(x,z_0)\Delta z}$$

absorption



Ray optics

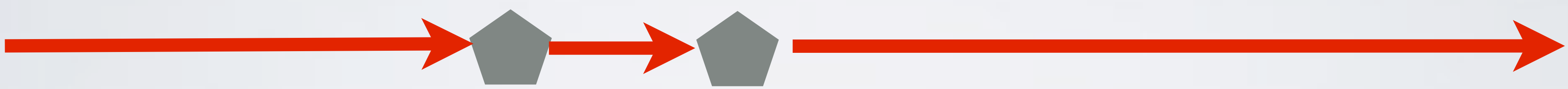
$$I' = I_0 e^{-\alpha(z)\Delta z}$$

refraction

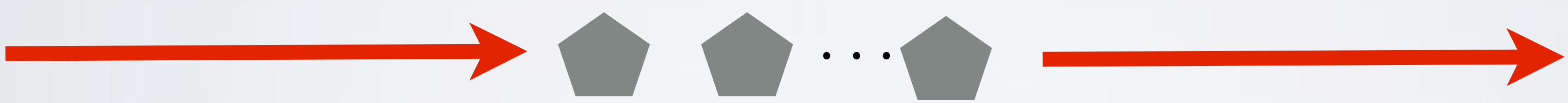
$$E' = E_0 e^{n(z)\Delta z} \qquad n = 1 - \delta + i\beta$$

wavefield

Ray optics, thick sample



$$e^{-\alpha(x,z_0)\Delta z} \quad e^{-\alpha(x,z_1)\Delta z}$$



$$e^{-\sum_i \alpha(x,z_i)\Delta z}$$

"Rythov approximation"

$$\log\left(\frac{I_z}{I_0}\right) = -\sum_i \alpha(x, z_i)\Delta z$$

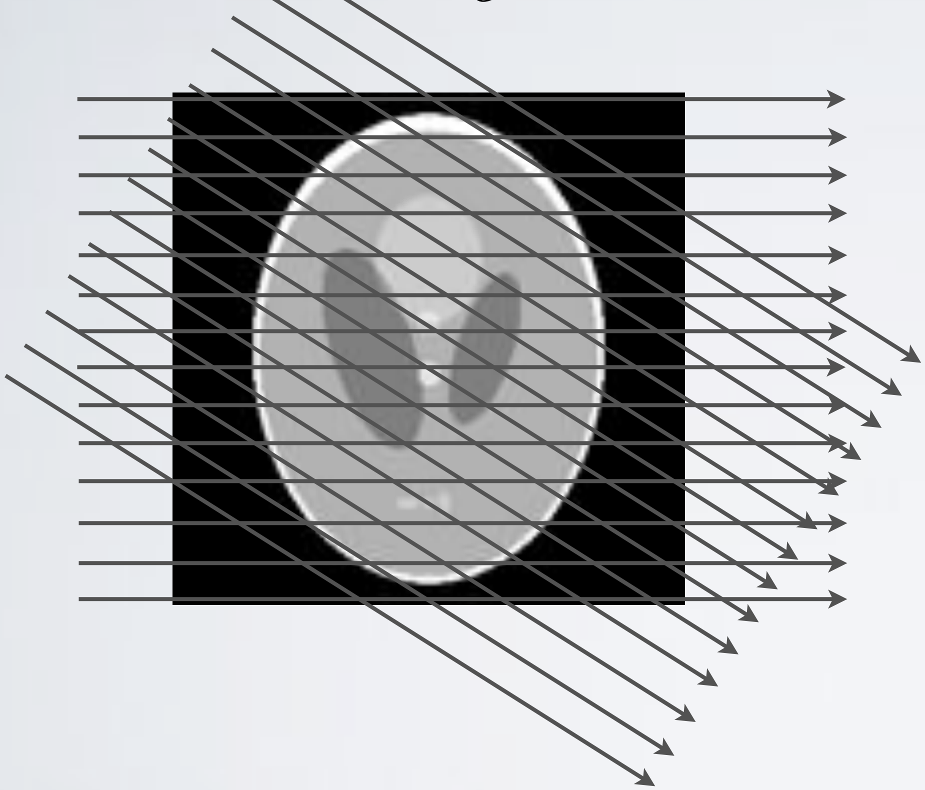
Born approximation

$$\frac{I_z}{I_0} = 1 - \sum_i \alpha(x, z_i)\Delta z$$

ray optics

absorption coefficient

$$e^{-\alpha(x,z_0)\Delta z}$$



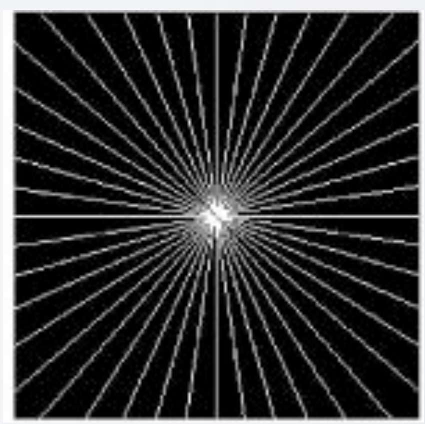
measure $\int \rho(x,y)d\boldsymbol{\tau}$

Tomography

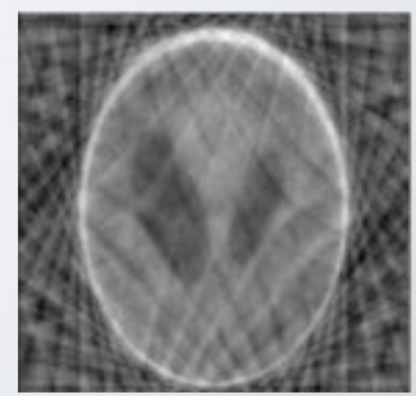
projection slice

$$\mathcal{F}_{1D}\int \rho(x,y)d\boldsymbol{\tau} = F_{2D}\rho(q\hat{\tau})$$

Fill out Fourier Slices

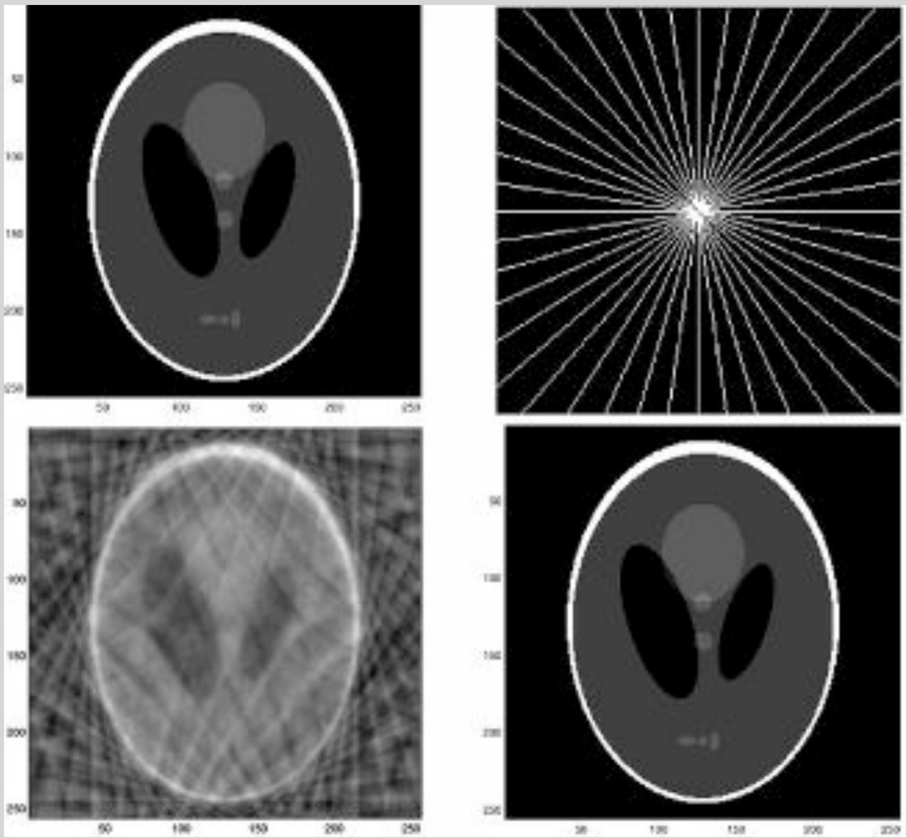


IFT



SHANNON WAS A PESSIMIST

Sparsifying algorithms
allow exact reconstruction
from undersampled data



Exact reconstruction using Total Variation norm (sparse gradient)

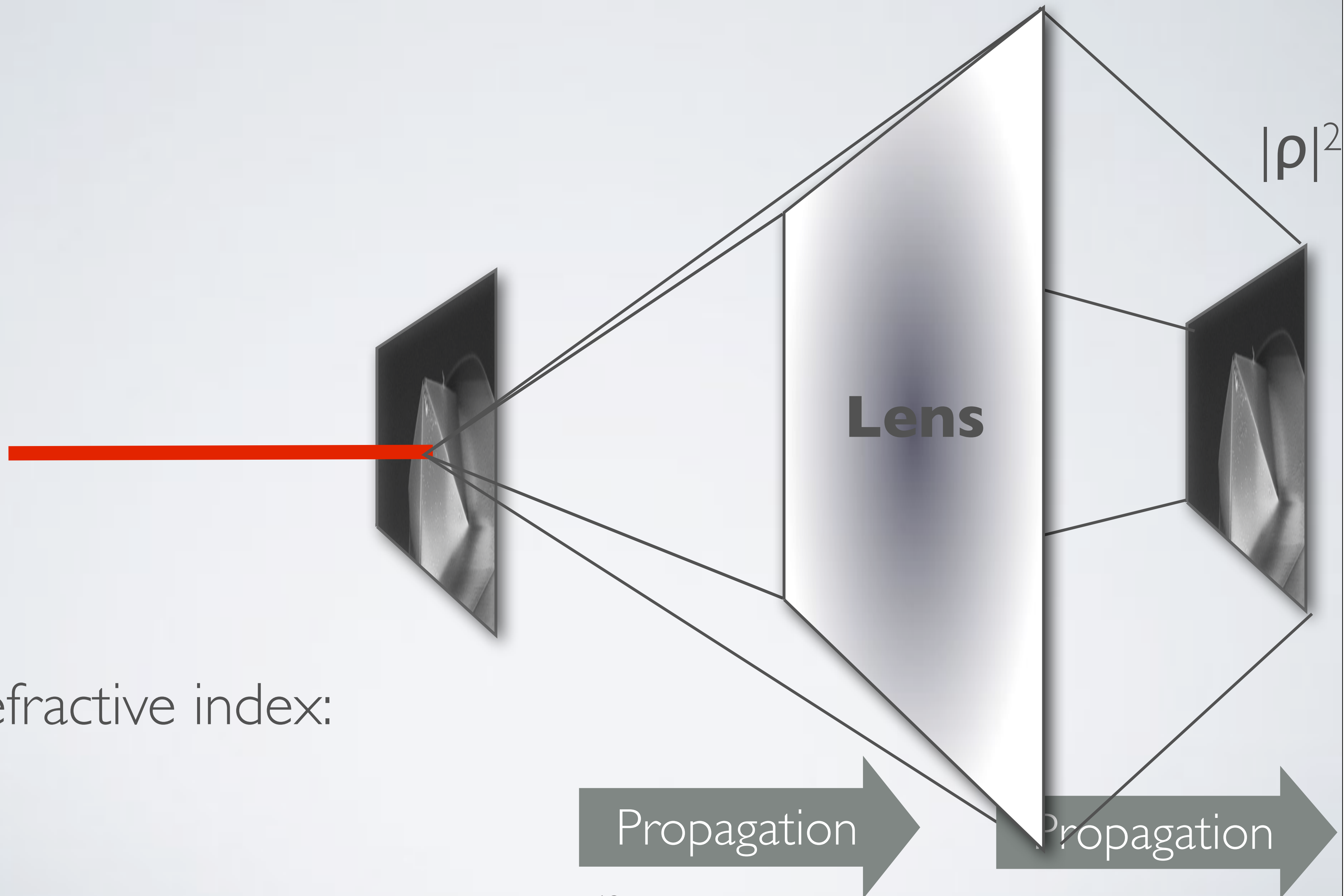
Candes, Romber, Tao ('06)

Shannon theorem is worst case scenario

if we know that the signal is sparse (and other conditions)

see talk by Rebecca Willett

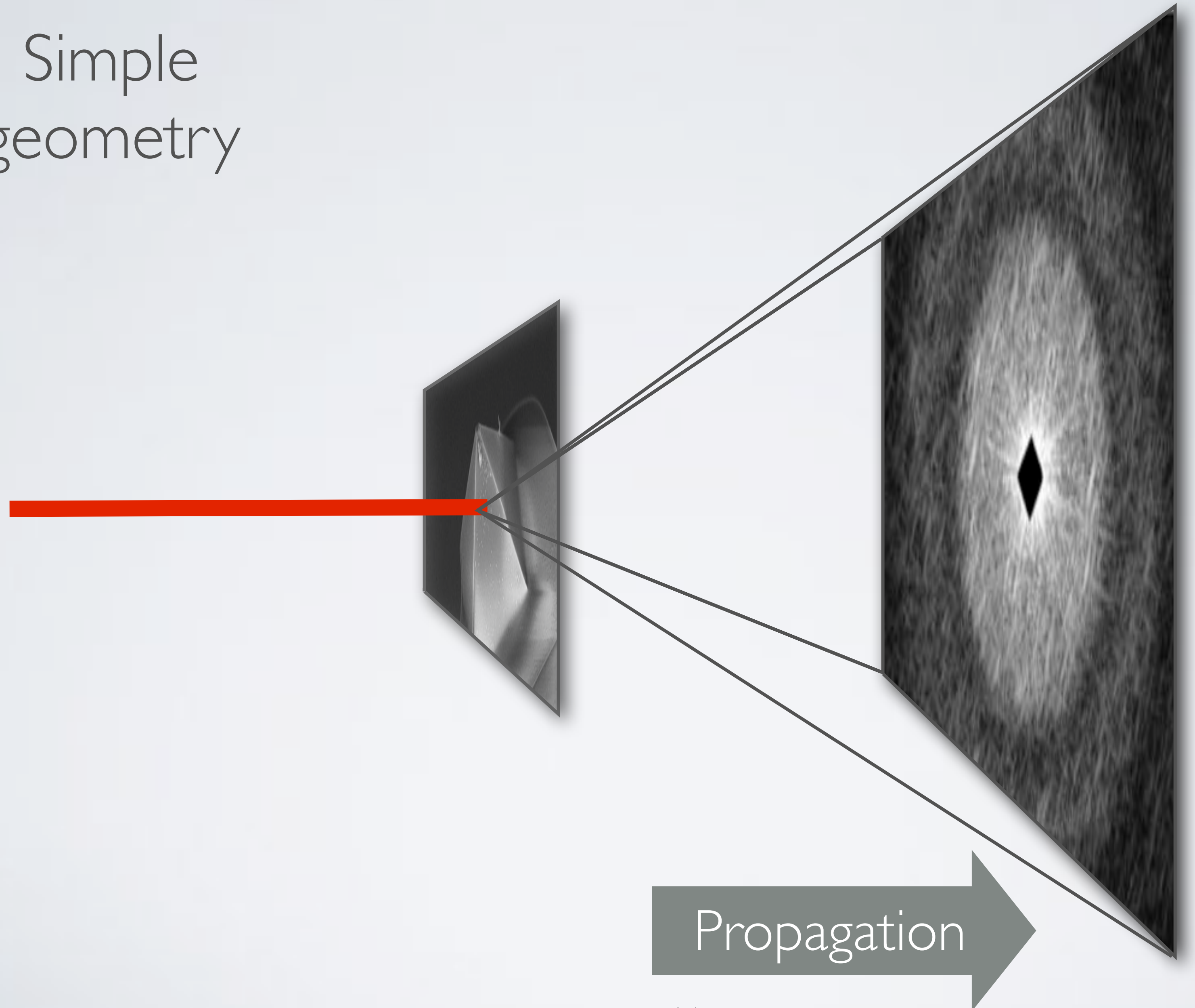
X-ray Microscopy the experiment



refractive index:

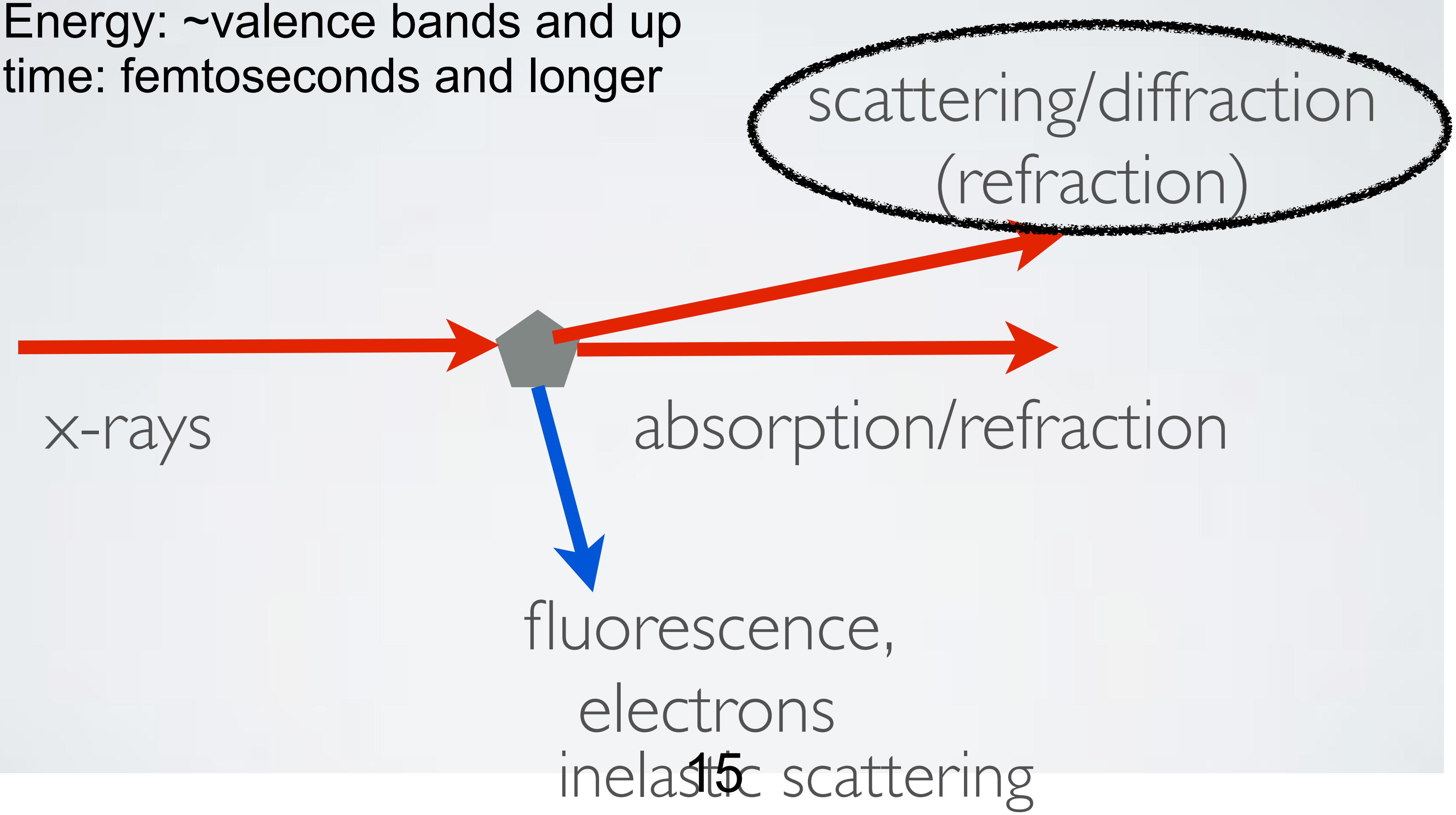
X-ray Diffraction: optical Fourier transform

Simple geometry



X-ray interaction with matter

- wavelength ~atom size
- penetration ~microns-m
- Energy: ~valence bands and up
- time: femtoseconds and longer



NONLINEAR DATA ANALYSIS

Diffraction & phase retrieval

TIMELINE

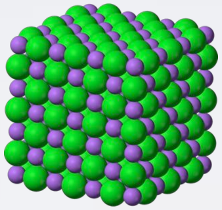
1905 Rontgen: x-ray imaging

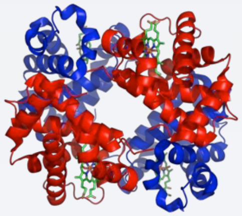
1912 Bragg **NaCl**

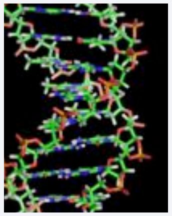
1951 Pauling et al **Alpha helix**
stereochemistry+diffraction:
1 spot (5.1 A period)

1953 Watson & Crick **DNA**
stereochemistry+diffraction
10 spots

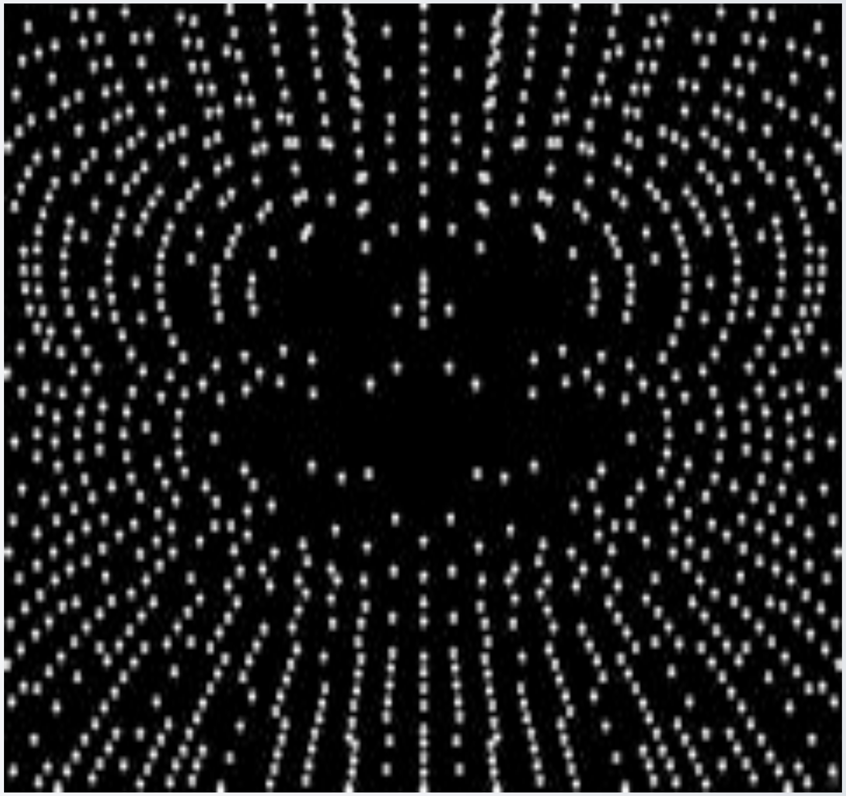
2012 ...





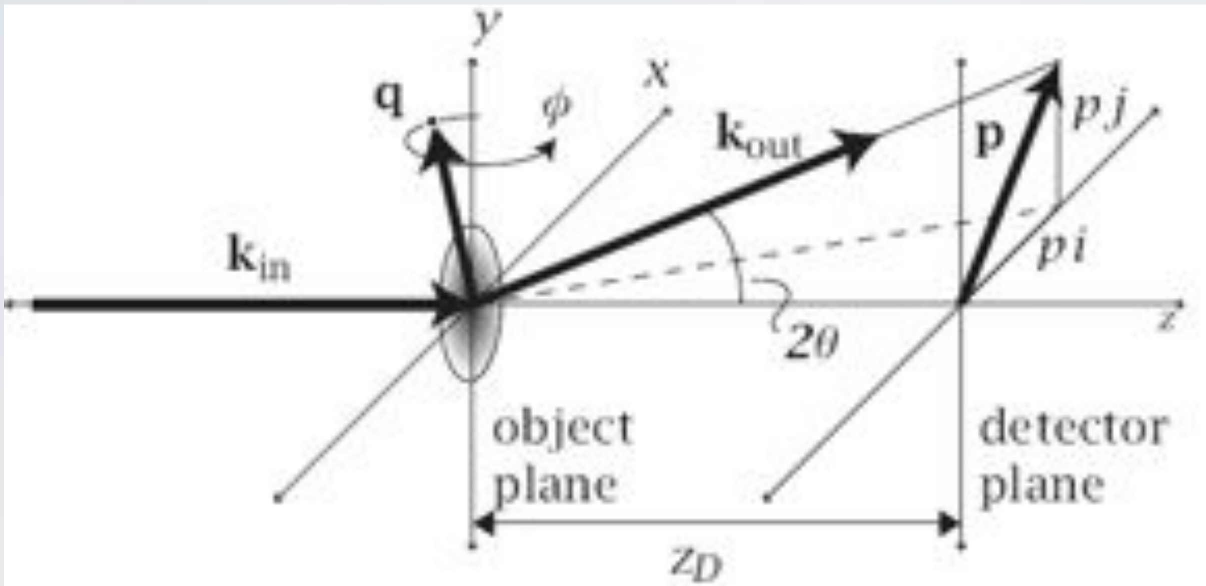


Diffraction pattern from a crystal

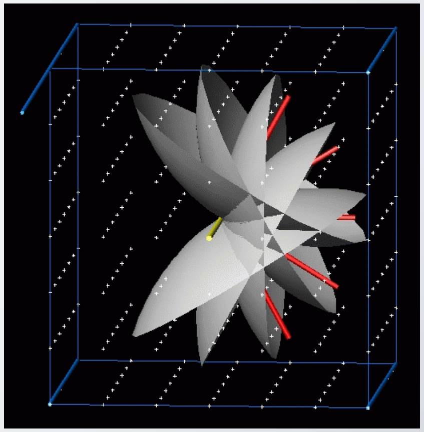


Elastic scattering and Fourier space

Each 2D measurement *I(p)* is a slice in Fourier space $I(\mathbf{p})=|F(\mathbf{q})|^2$



rotate for 3D

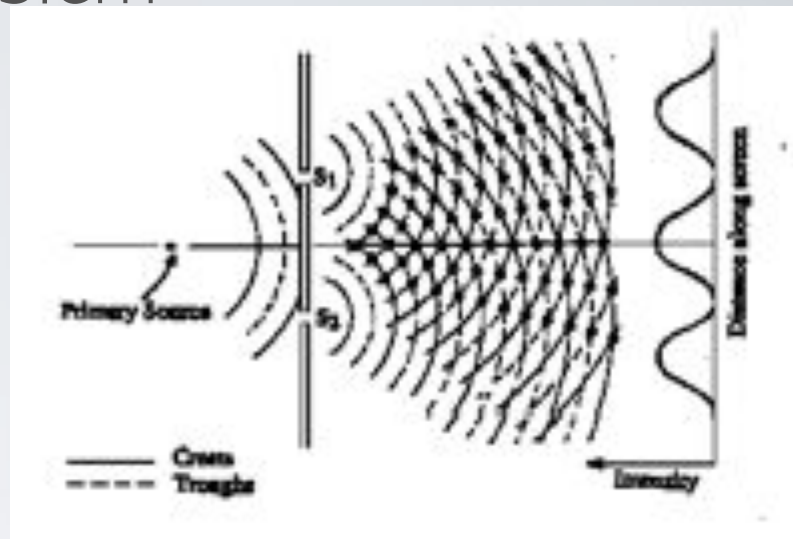


detector pixels

$$\begin{aligned}\mathbf{q}_{i,j} &= \mathbf{k}_{\text{out}} - \mathbf{k}_{\text{in}}\,, \\ &= \frac{1}{\lambda}\left(\frac{(p_i, p_j, z_D)}{\sqrt{p_i^2+p_j^2+z_D^2}} - (0,0,1)\right)\end{aligned}$$

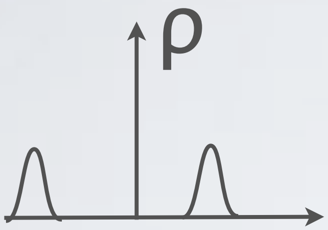
$$F(\boldsymbol{q}) = \mathcal{F}\rho(\boldsymbol{x})$$

X-ray Diffraction: the phase problem



$$F(\boldsymbol{q}) = \mathcal{F}\rho(\boldsymbol{x})$$

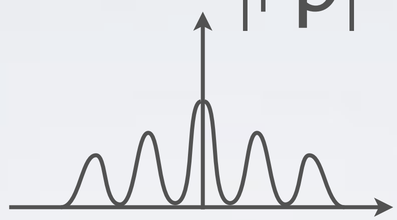
sample



position
(real space)

FT

$|F\rho|^2$

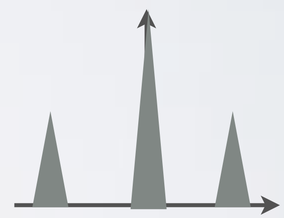


momentum
(Fourier space)

Find ρ

Measure $|F\rho|^2$

$\psi(-x)^* \, \psi(x)$



Autocorrelation
(real space)

IFT

$\rho(x)^* \, \rho(-x)$

SOLUTION IS NOT UNIQUE

Find ρ — Measure $|F\rho|^2$

Trivial

$\rho(x)$, $\rho(-x)$, $\rho(x+\Delta x)$, $e^{i\Phi}$

identity flip translation phase

---

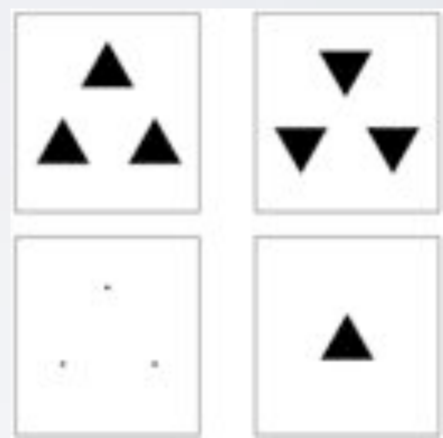
homometric structures

$\rho(x)=\rho_1(x)*\rho_2(x)$,

$\rho(x)=\rho_1(x)*\rho_2(-x)$

$\rho(x)$

$\rho_1(x)$



$\rho_2(x)$

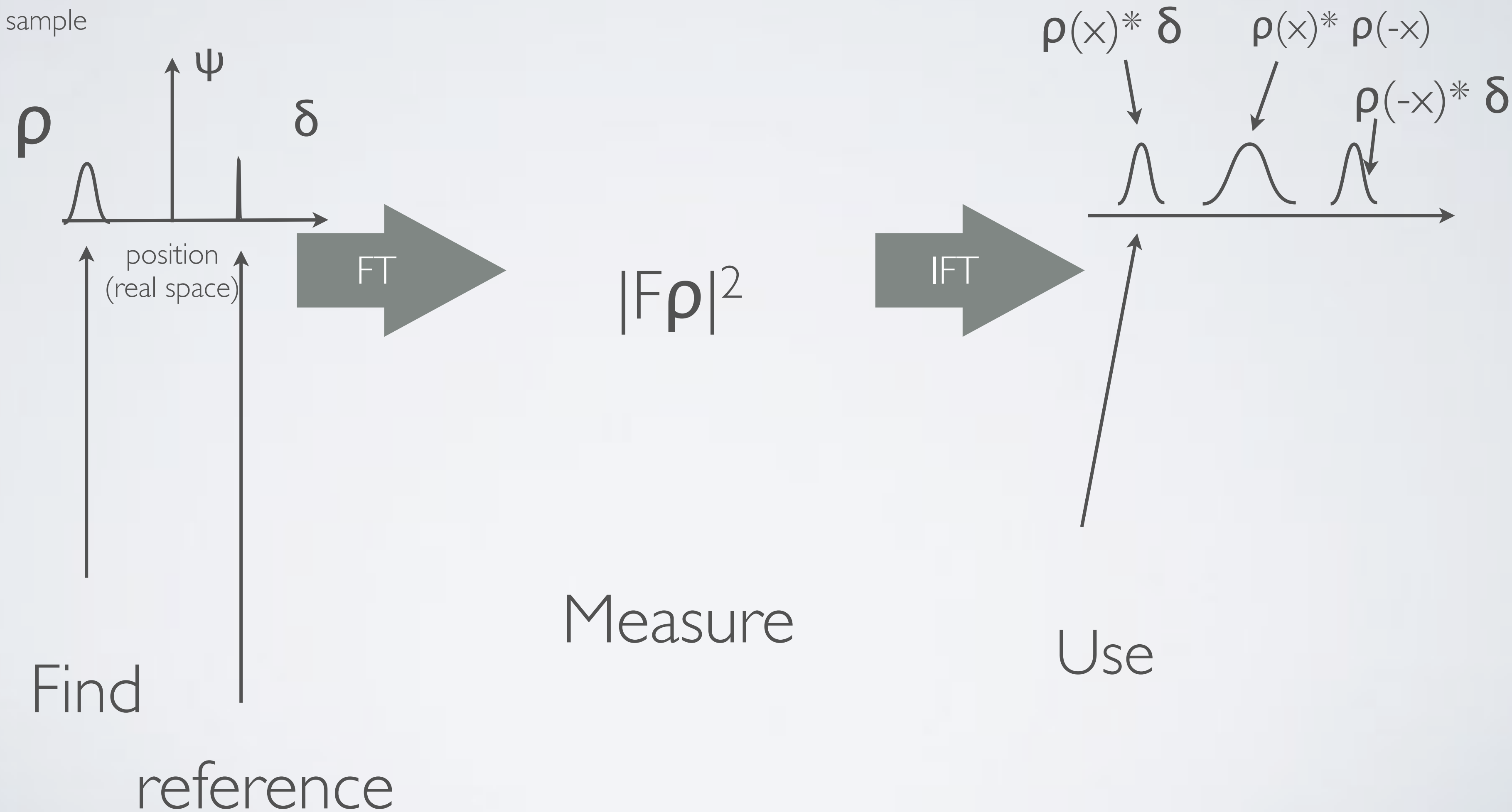
Bruck, Sodin (1979) Bates (1982) Hayes (1982)

homometric structures are rare

(factorable polynomials are rare in 2D and 3D)

X-ray Diffraction: the phase problem
solution (I)

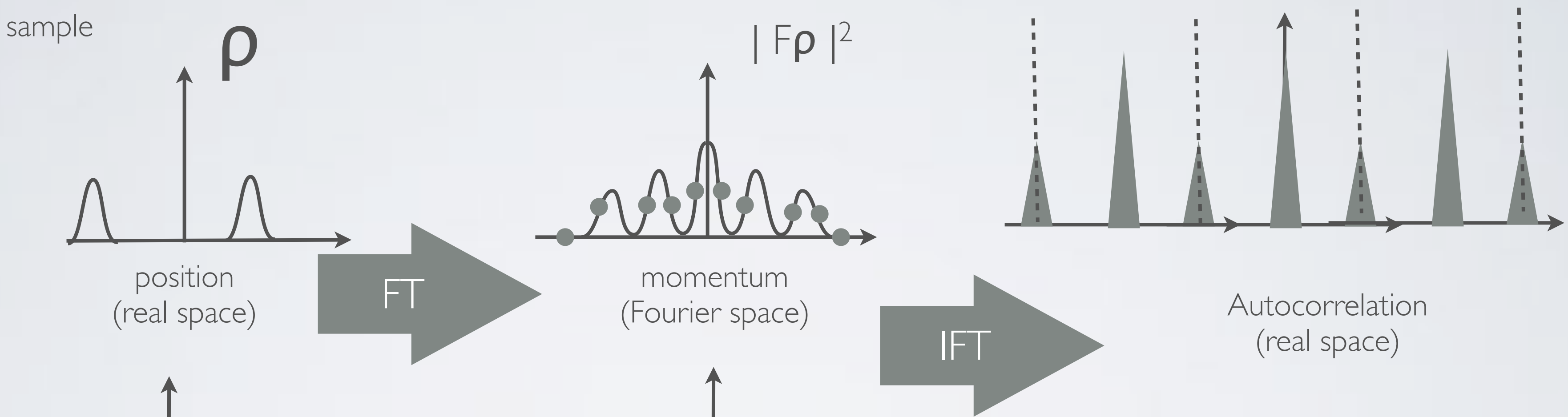
1946-51 (Gabor) Holography: introduce reference



X-ray Diffraction: the sampling problem

$$F(\boldsymbol{q}) = \mathcal{F}\rho(\boldsymbol{x})$$

ρ(-x)* ρ(x)



Find

ρ

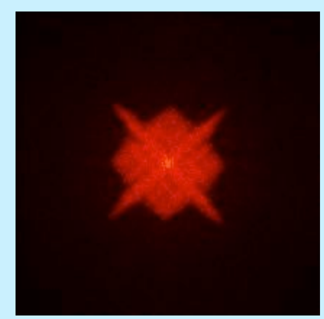
Measure

|Fρ|²

Sampling & aliasing

ρ

ρ(-x)* ρ(x)



OPTIMIZATION PROBLEM

Measure $I(\boldsymbol{q}) = c \left\langle |F(\boldsymbol{q})|^2 \right\rangle_{\Omega(\boldsymbol{q})}$

Find $F(\boldsymbol{q}) = \mathcal{F}\rho(\boldsymbol{x})$ Subject to: $\rho(\boldsymbol{x})$ finite, sparse or other prior

$\langle \cdot \rangle_{\Omega(\boldsymbol{q})}$ Average over e.g. wavelength, rotation, crystallite

$\boldsymbol{q}$ sampling, e.g. Bragg condition, beamstop

ITERATIVE METHODS

Start with some guess

F^{-1} transform

Real space constraint

$$\rho(\boldsymbol{q}) \to \mathcal{O}\rho(\boldsymbol{q})$$

F-transform

Fit the data

projection onto a non-convex set

BRANCHES OF DIFFRACTION

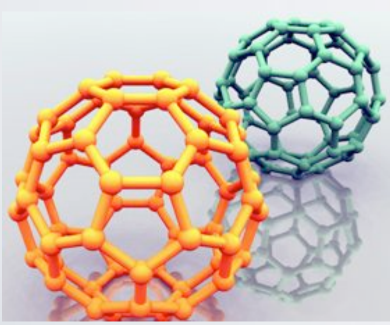
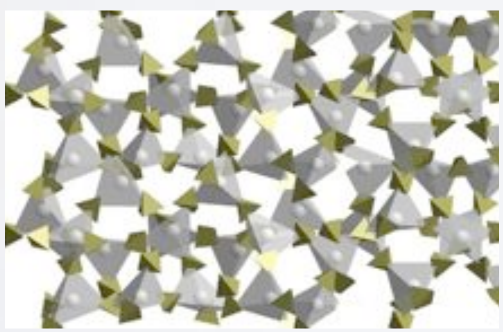
Diffraction

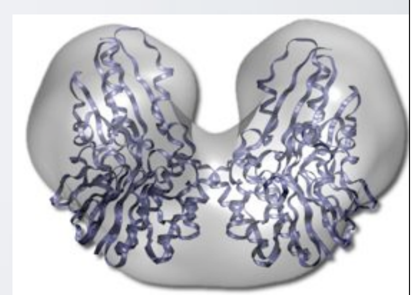
Small molecule crystal → Direct methods seeking sparsity

Small powder crystal → Direct methods, trial/error

protein crystal → +strong contrast atom → Direct methods, +maximum likelihood +model refinment

Solution scattering → Monte carlo



BRANCHES OF DIFFRACTION

Diffraction

Single particle diffraction

Non convex methods (seeking compact object)

nanocrystallography

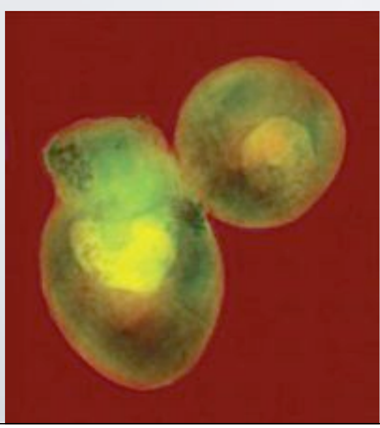
orientation determination (10^5 patterns) model fitting

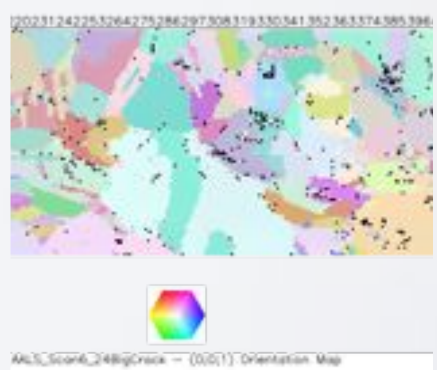
microcrystal

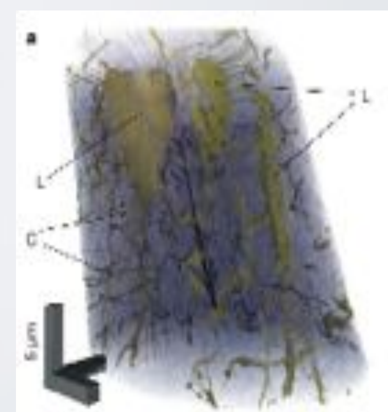
orientation determination model fitting

scanning diffraction

L_2 minimization 10^{11} unknowns

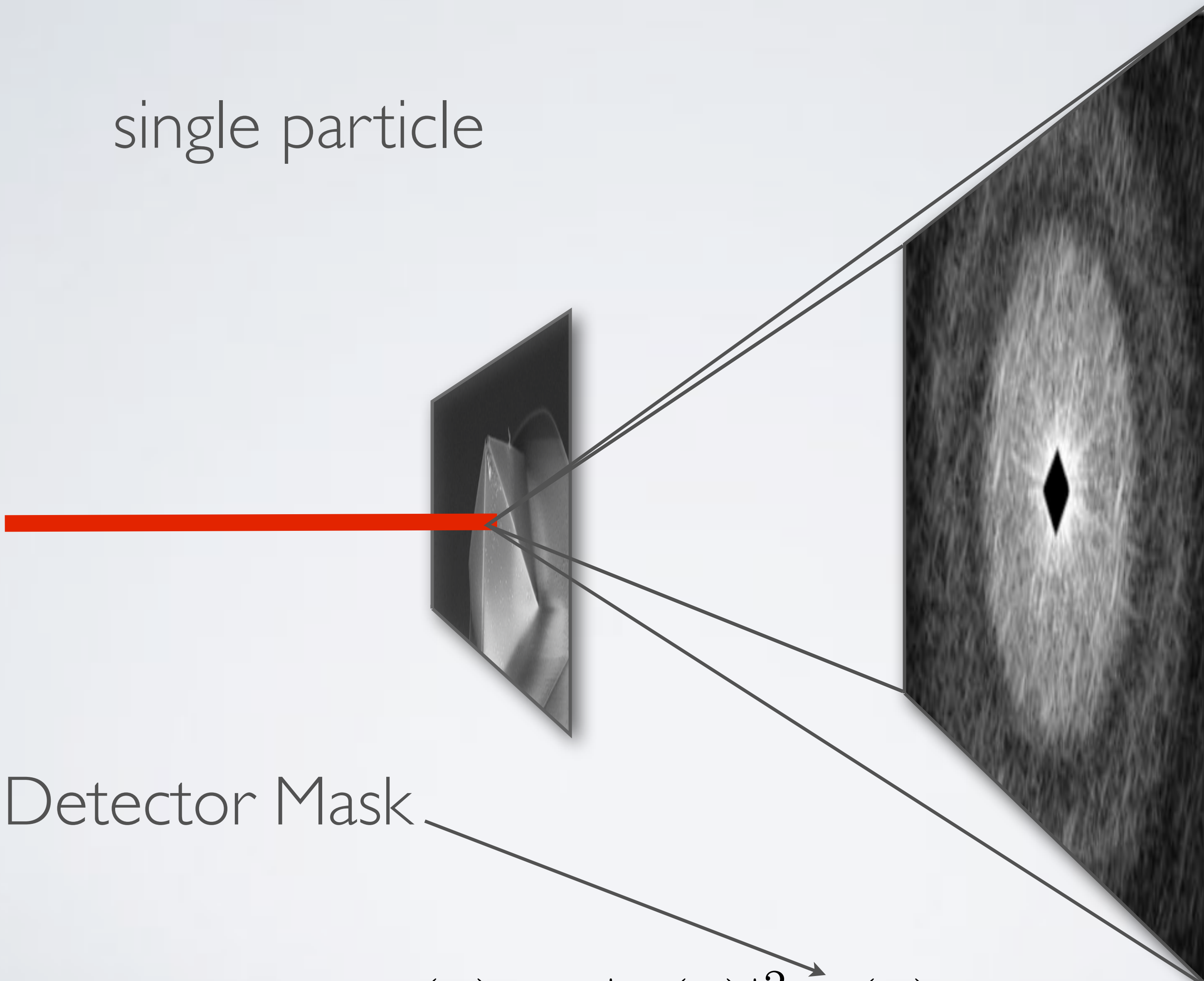






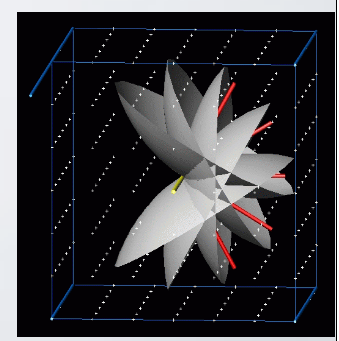
Sampling Schemes

single particle



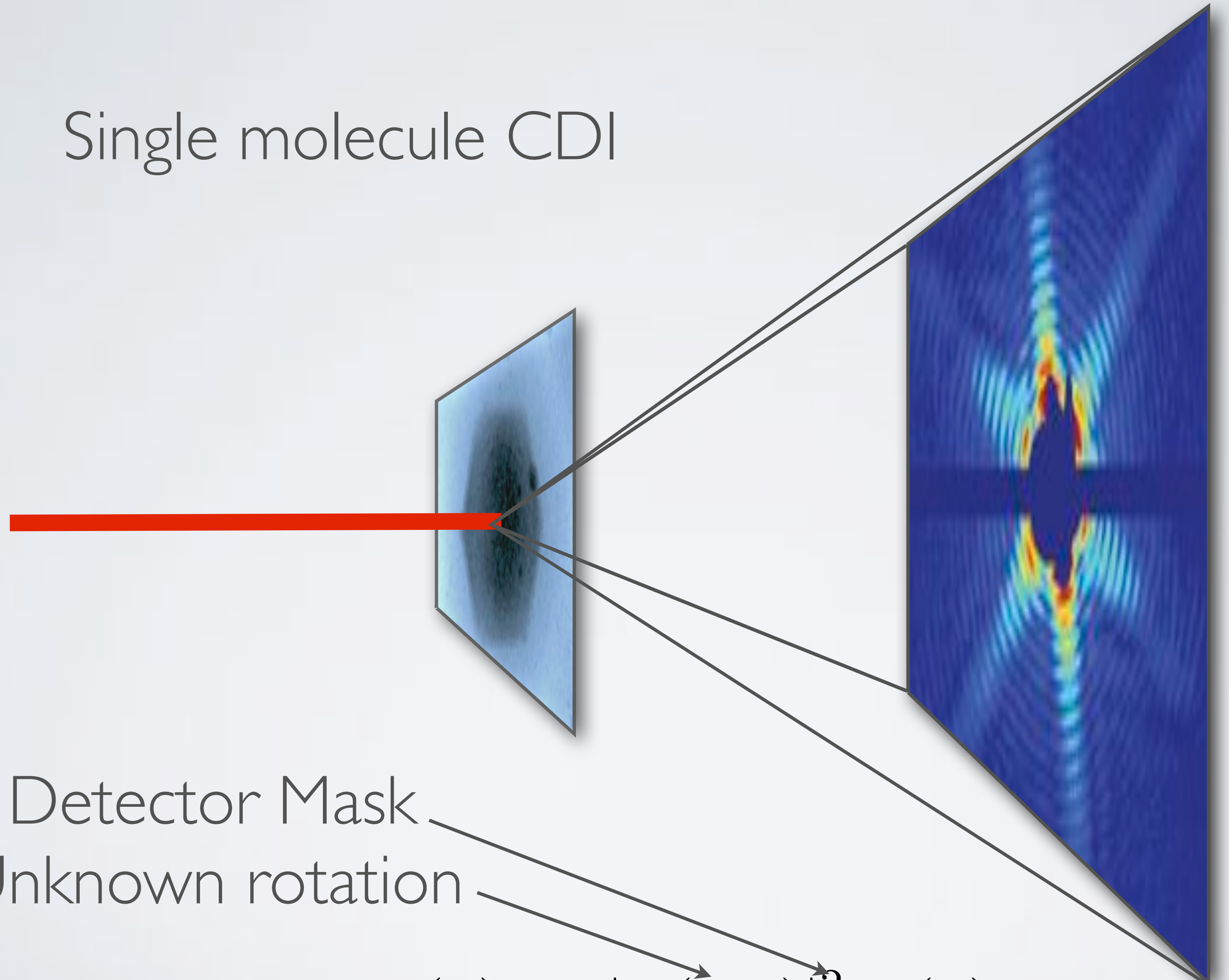
Detector Mask

$$I(\boldsymbol{q}) = c|F(\boldsymbol{q})|^2 M(\boldsymbol{q})$$



Sampling Schemes

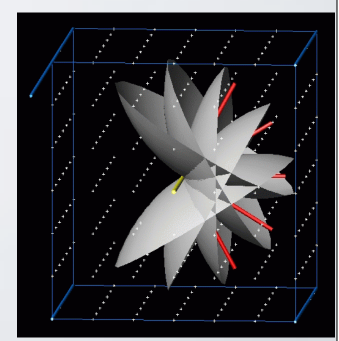
Single molecule CDI



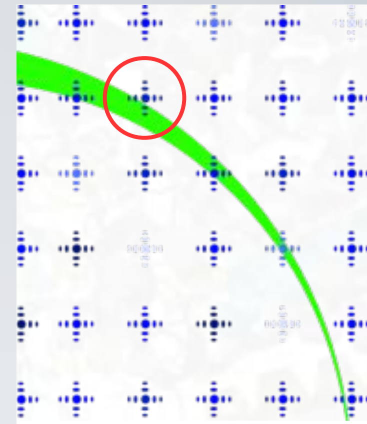
Detector Mask

Unknown rotation

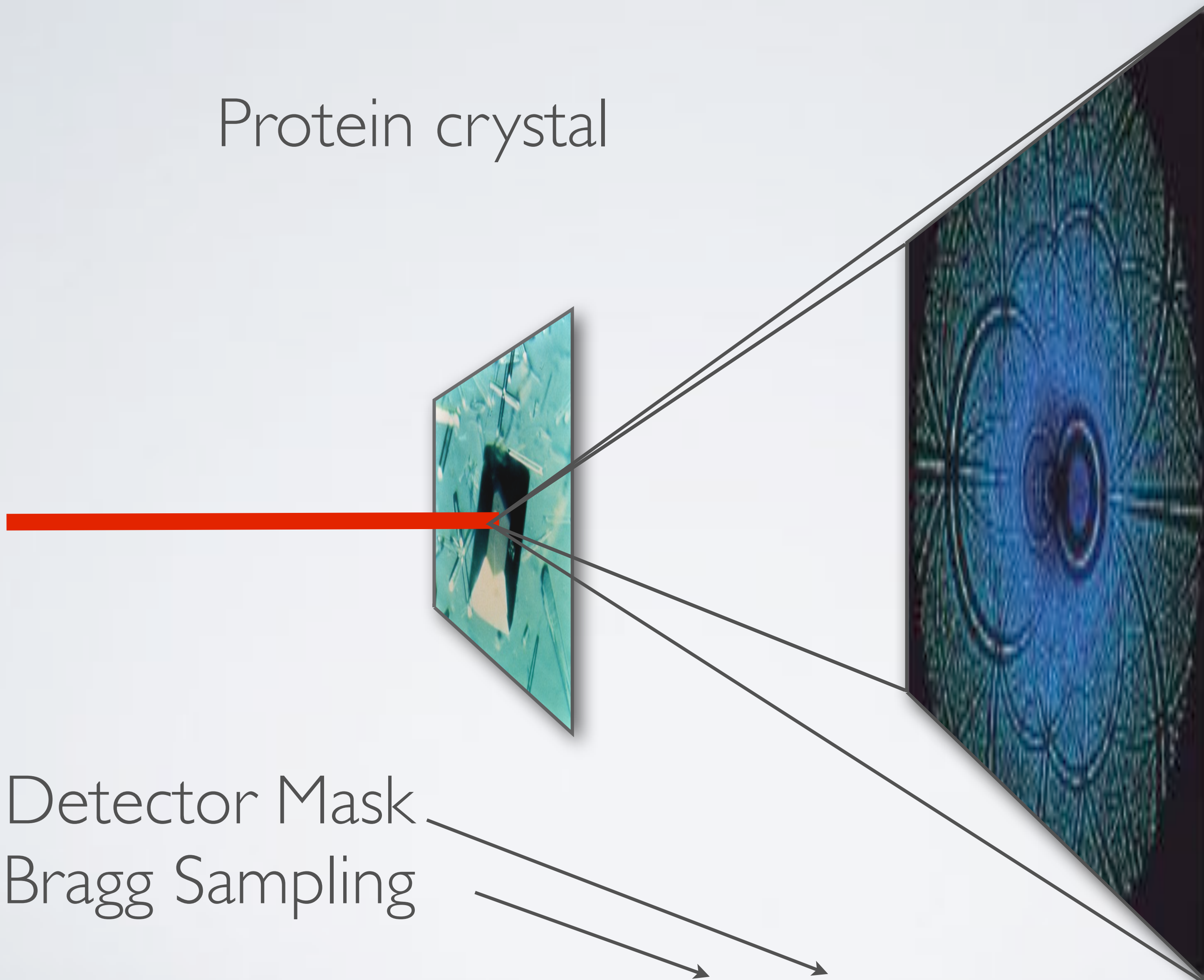
$$I(\boldsymbol{q}) = c|F(\boldsymbol{Rq})|^2 M(\boldsymbol{q})$$



Sampling Schemes



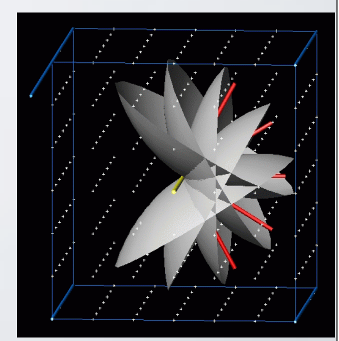
Protein crystal



Detector Mask

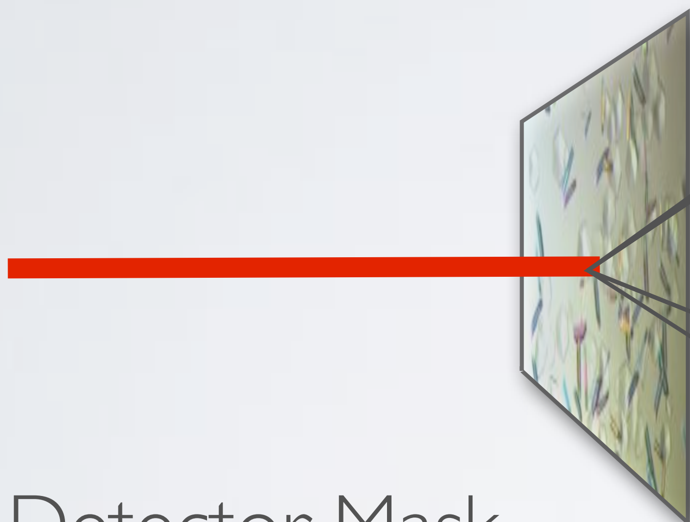
Bragg Sampling

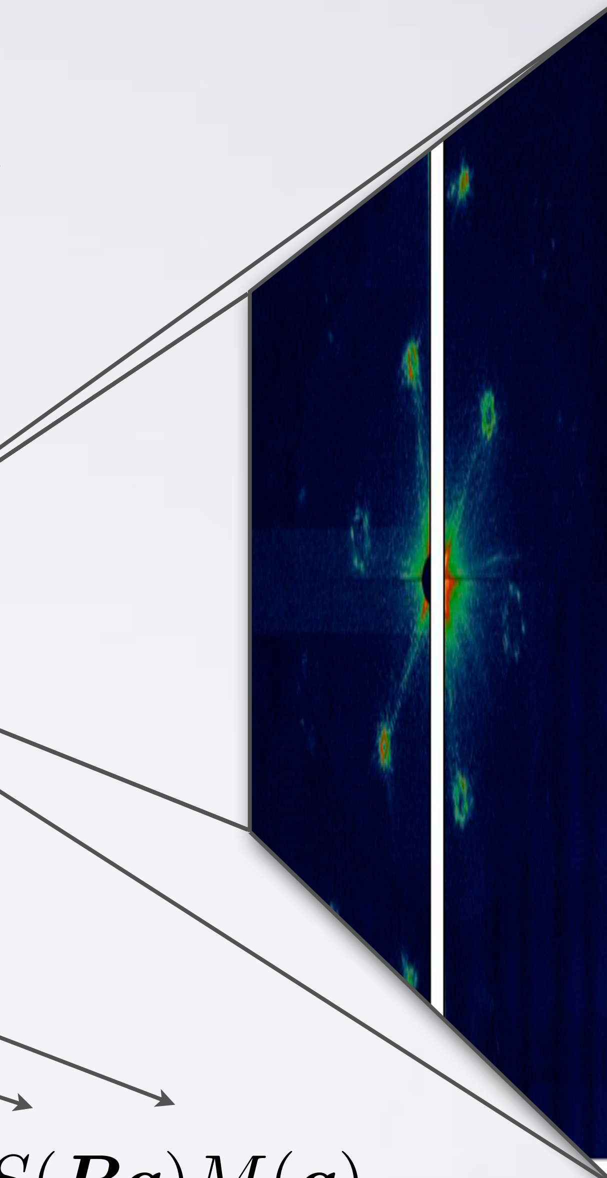
$$I(\boldsymbol{q}) = c|F(\boldsymbol{Rq})|^2 S(\boldsymbol{Rq}) M(\boldsymbol{q})$$



Sampling Schemes

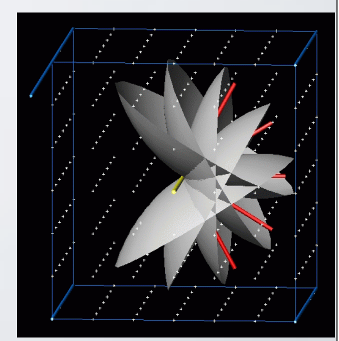
Protein nano-crystals





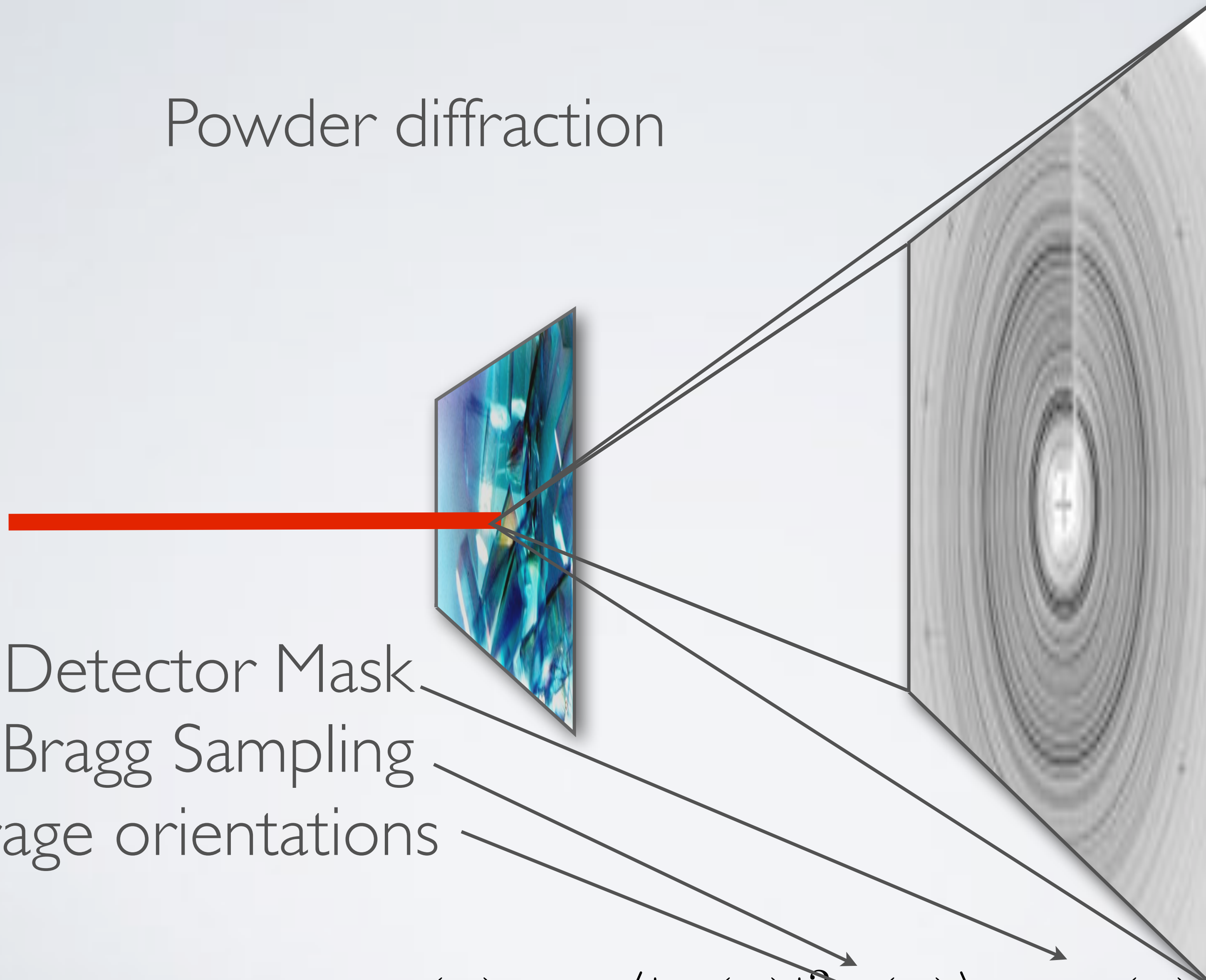
Detector Mask
Bragg Sampling
Unknown rotation

$$I(\boldsymbol{q}) = c|F(\boldsymbol{Rq})|^2 S(\boldsymbol{Rq}) M(\boldsymbol{q})$$



Sampling Schemes

Powder diffraction

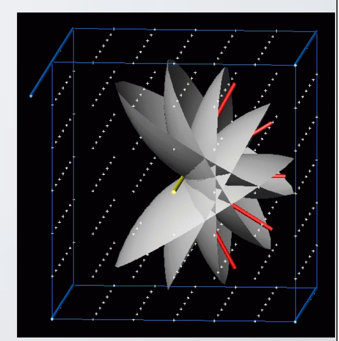


Detector Mask

Bragg Sampling

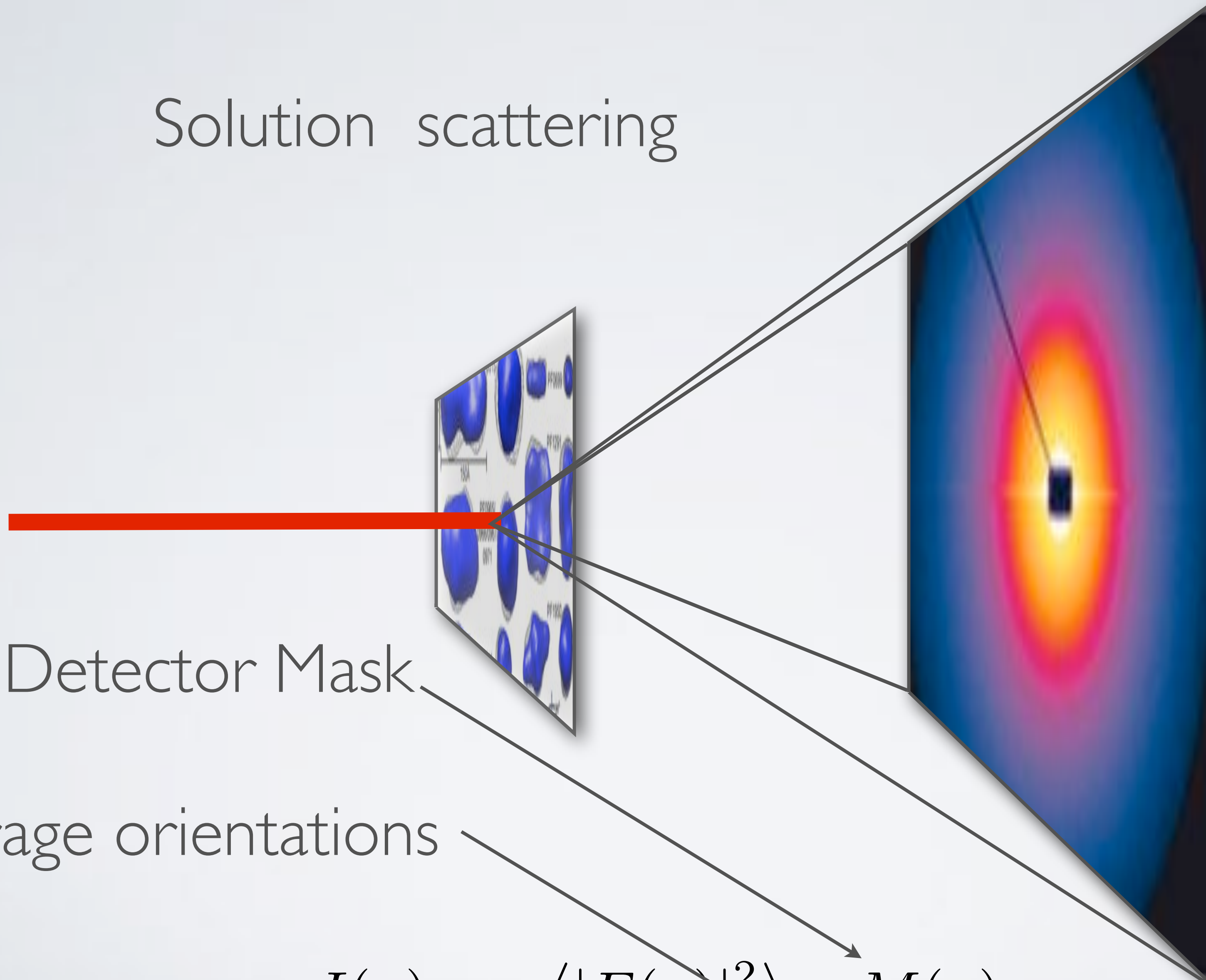
average orientations

$$I(\boldsymbol{q}) = c\left\langle |F(\boldsymbol{q})|^2 S(\boldsymbol{q}) \right\rangle_{|\boldsymbol{q}|} M(\boldsymbol{q})$$



Sampling Schemes

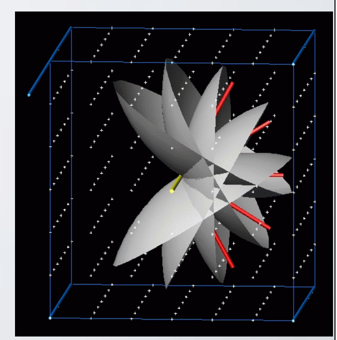
Solution scattering



Detector Mask

average orientations

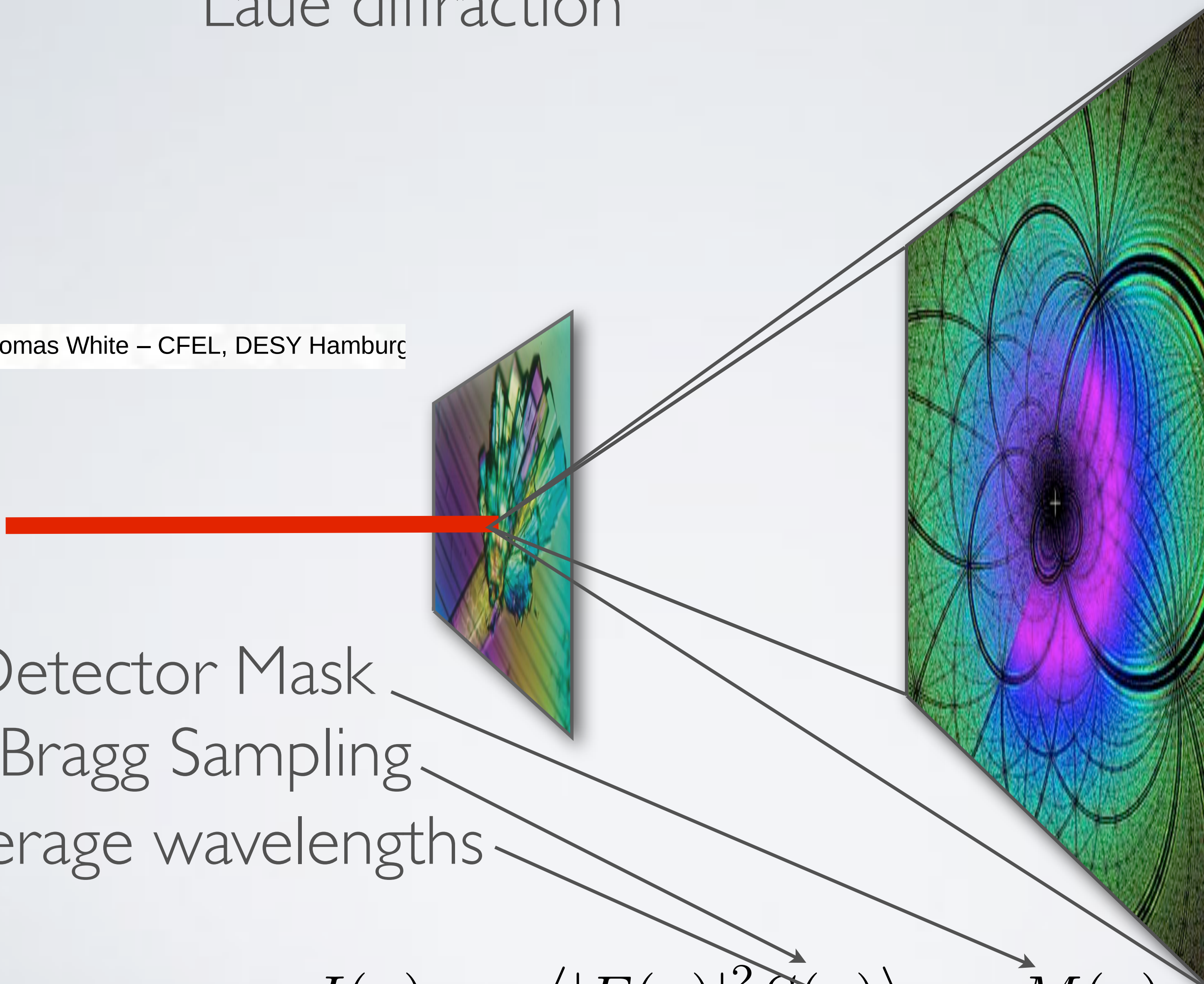
$$I(\boldsymbol{q}) = c\left\langle |F(\boldsymbol{q})|^2 \right\rangle_{|\boldsymbol{q}|} M(\boldsymbol{q})$$



Sampling Schemes

Laue diffraction

Thomas White – CFEL, DESY Hamburg

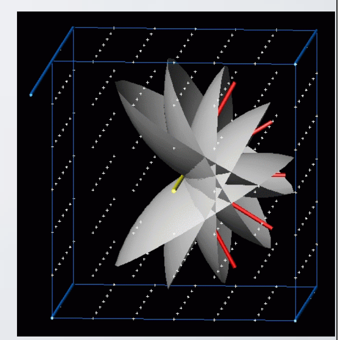


Detector Mask

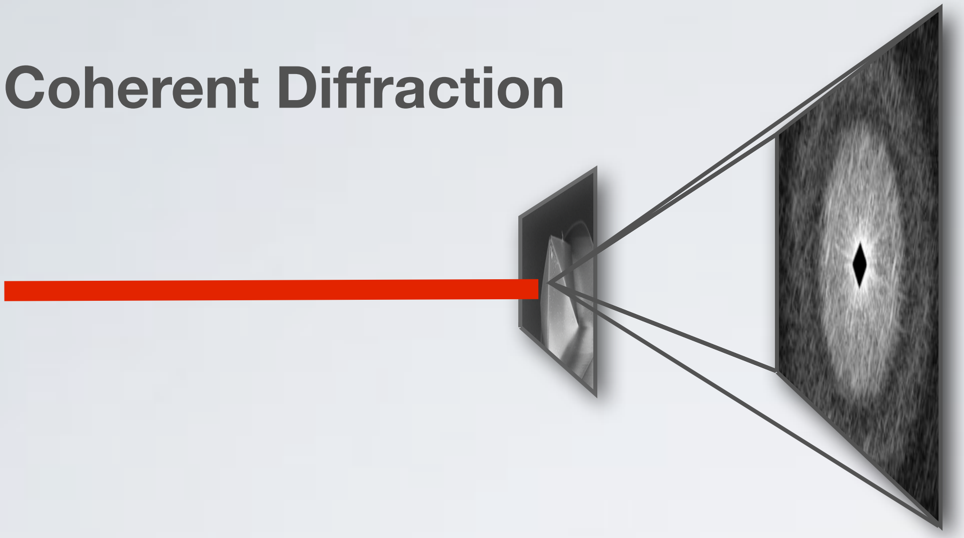
Bragg Sampling

average wavelengths

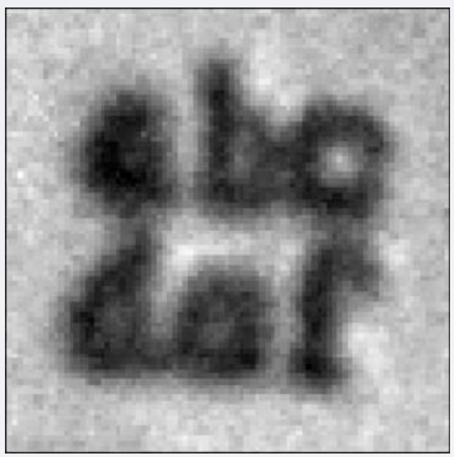
$$I(\boldsymbol{q}) = c\left\langle |F(\boldsymbol{q})|^2 S(\boldsymbol{q}) \right\rangle_{\Omega(\boldsymbol{q})} M(\boldsymbol{q})$$



Coherent diffractive imaging (1999)



+



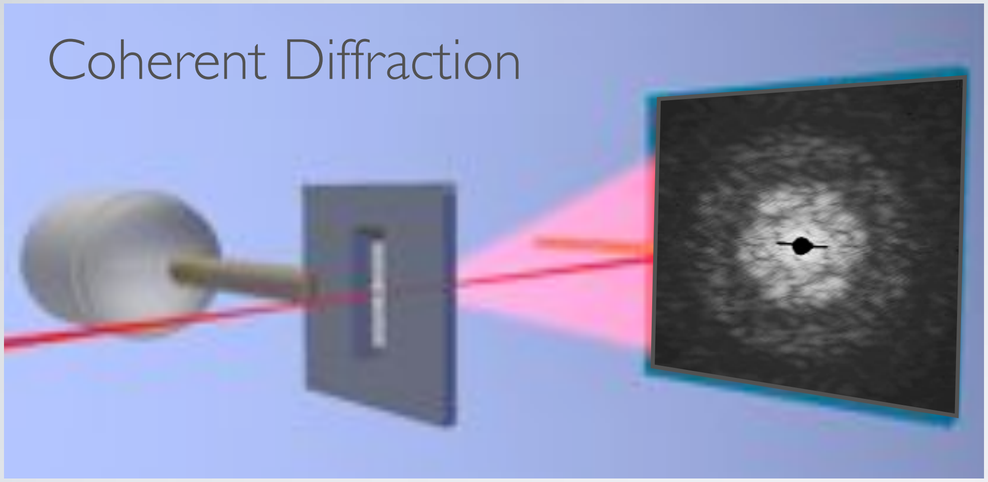
=

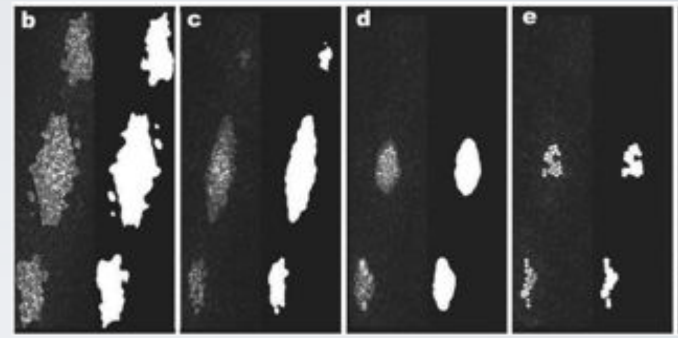
Resolution extended by phasing alogirhtms

nature International weekly journal of science

J. Miao, P. Charalambous, J. Kirz & D Sayre, Nature 400, (1999)

Ab-initio cohrent diffractive imaging

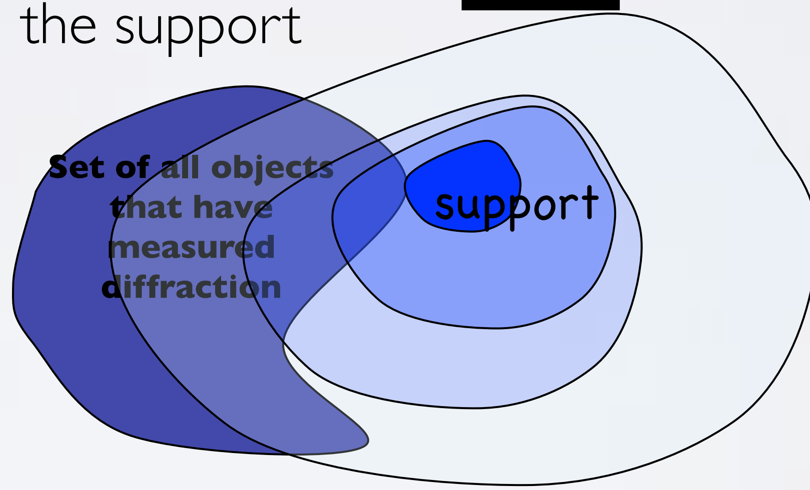




iteratively shrinking
the support

=

+

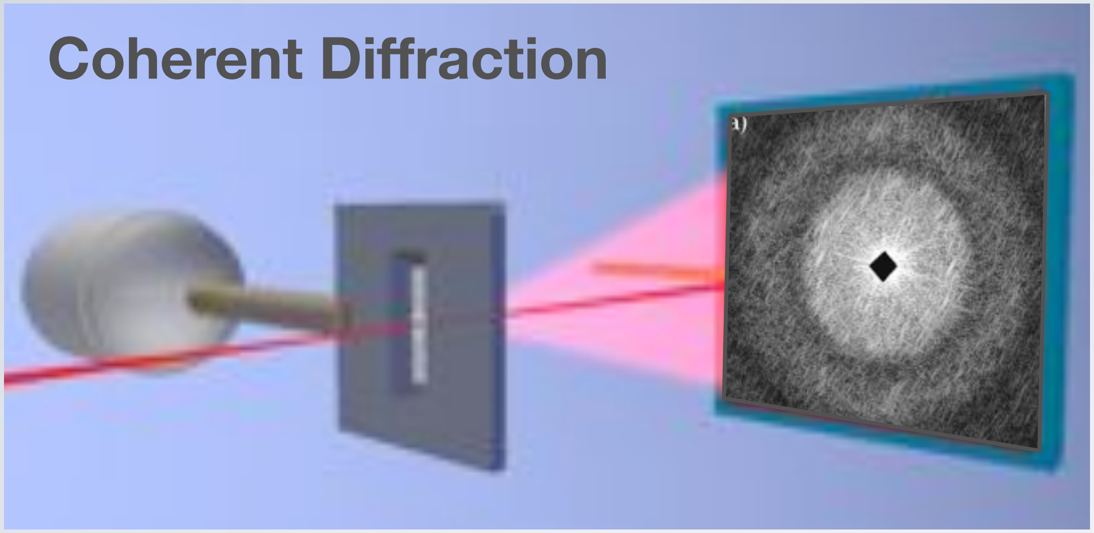


Find sparsest
solution
that fits the data

Physical Review B
condensed matter and materials physic

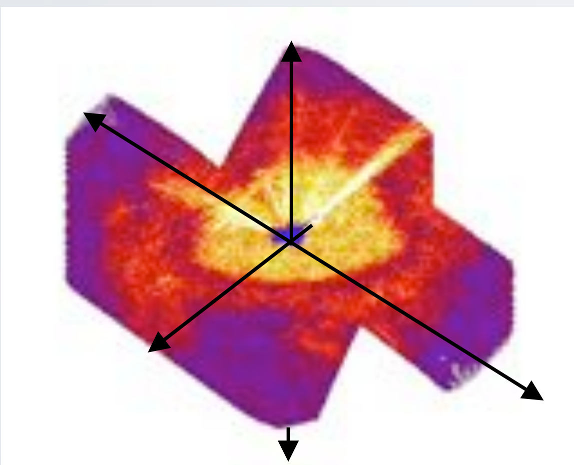
S. Marchesini, H. He, H. N. Chapman et al. PRB 68, 140101(R) (2003),

THREE DIMENSIONAL CDI



- Established billion-element phasing

Diffraction data



ab-initio Reconstruction

- 15 nm resolution

Journal of the Optical Society of America A

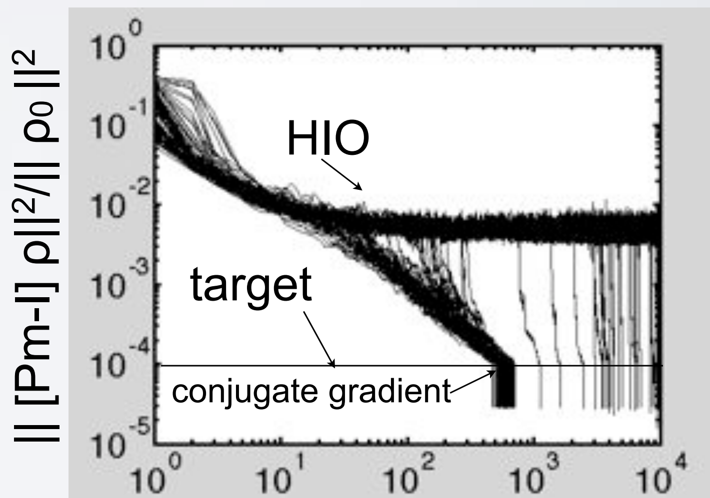
H. N. Chapman, A. Barty, S. Marchesini, et al. JOSAA 23, 1179-1200 (2006)

IT DOESN'T ALWAYS WORK

- Support determination algorithms (shrinkwrap) are not robust
- Even with simulated data, reconstruction is not guaranteed.
- Missing data is a main source of problem

Table 1. Summary of various algorithms

Algorithm	Iteration $\rho^{(n+1)} =$
ER	$\boldsymbol{P_s P_m} \rho^{(n)}$
SF	$\boldsymbol{R_s P_m} \rho^{(n)}$
HIO	$\begin{cases} \boldsymbol{P_m} \rho^{(n)}(\boldsymbol{r}) & \boldsymbol{r} \in S \\ (\boldsymbol{I} - \beta \boldsymbol{P_m})\rho^{(n)}(\boldsymbol{r}) & \boldsymbol{r} \notin S \end{cases}$
DM	$\{\boldsymbol{I} + \beta \boldsymbol{P_s} \; [(1+\gamma_s)\,\boldsymbol{P_m} - \gamma_s \boldsymbol{I}] - \beta \boldsymbol{P_m} \; [(1+\gamma_m)\,\boldsymbol{P_s} - \gamma_m \boldsymbol{I}]\}\rho^{(n)}$
ASR	$\frac{1}{2}[\boldsymbol{R_s R_m} + \boldsymbol{I}]\rho^{(n)}$
HPR	$\frac{1}{2}[\boldsymbol{R_s}\,(\boldsymbol{R_m} + (\beta - 1)\boldsymbol{P_m}) + \boldsymbol{I} + (1-\beta)\boldsymbol{P_m}]\rho^{(n)}$
RAAR	$\left[\frac{1}{2}\beta\,(\boldsymbol{R_s R_m} + \boldsymbol{I}) + (1-\beta)\boldsymbol{P_m}\right]\rho^{(n)}$



WHAT FIX-POINT ITERATIONS WORK BEST?

$$\rho(\boldsymbol{q}) \to \mathcal{O}\rho(\boldsymbol{q})$$

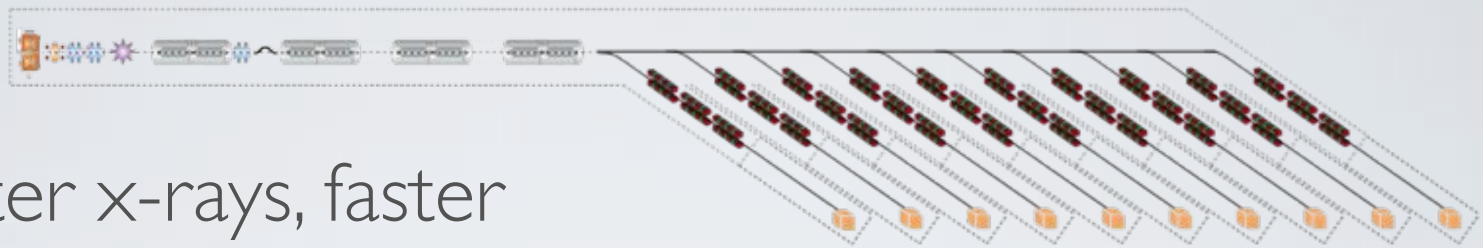
Table 1. Summary of various algorithms

Algorithm	Iteration $\rho^{(n+1)} =$
ER	$\boldsymbol{P_s}\boldsymbol{P_m}\rho^{(n)}$
SF	$\boldsymbol{R_s}\boldsymbol{P_m}\rho^{(n)}$
HIO	$\begin{cases} \boldsymbol{P_m}\rho^{(n)}(\boldsymbol{r}) & \boldsymbol{r} \in S \\ (\boldsymbol{I} - \beta\boldsymbol{P_m})\rho^{(n)}(\boldsymbol{r}) & \boldsymbol{r} \notin S \end{cases}$
DM	$\{\boldsymbol{I} + \beta\boldsymbol{P_s}\,[(1+\gamma_s)\,\boldsymbol{P_m} - \gamma_s\boldsymbol{I}] - \beta\boldsymbol{P_m}\,[(1+\gamma_m)\,\boldsymbol{P_s} - \gamma_m\boldsymbol{I}]\}\rho^{(n)}$
ASR	$\frac{1}{2}[\boldsymbol{R_s}\boldsymbol{R_m} + \boldsymbol{I}]\rho^{(n)}$
HPR	$\frac{1}{2}[\boldsymbol{R_s}\,(\boldsymbol{R_m} + (\beta-1)\boldsymbol{P_m}) + \boldsymbol{I} + (1-\beta)\boldsymbol{P_m}]\rho^{(n)}$
RAAR	$\left[\frac{1}{2}\beta\,(\boldsymbol{R_s}\boldsymbol{R_m} + \boldsymbol{I}) + (1-\beta)\boldsymbol{P_m}\right]\rho^{(n)}$

arXiv:physics/0603201

arXiv:0809.2006

DATA DELUGE OPPORTUNITIES



Brighter x-rays, faster detectors

Statistical inference

(Sparse modeling)

recovering a small number of most relevant variables in high-dimensional data

More of the same

more samples, more theory

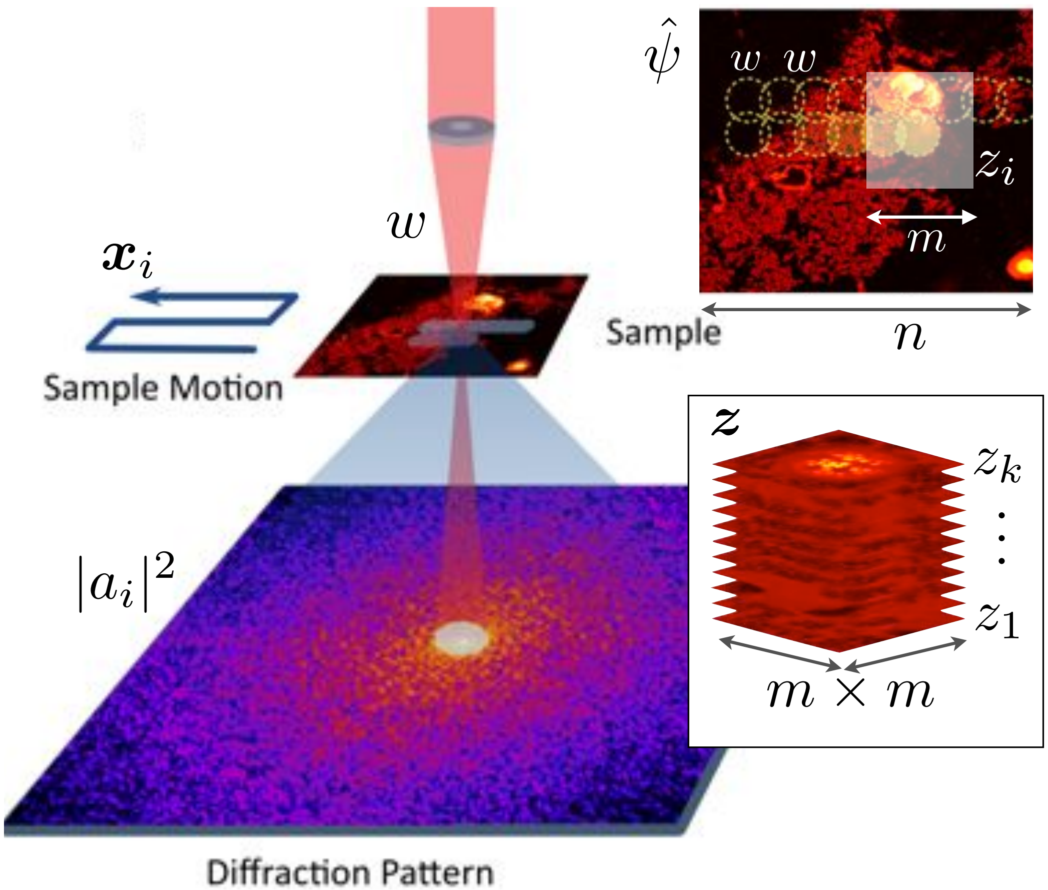
faster

Reduction

Reduce multi-frame data into higher resolution, higher contrast (SNR) images, volumes or movies.

Examples: ptychography, (nanocrystallography, single molecule, A. Barty talk)

ptychography:
combine scanning microscopy with diffraction



diffraction data

FFT

unknown

$$a_{\mathbf{x}}(\mathbf{q}) \quad = \quad \left|\mathcal{F} w(\mathbf{r})\hat{\psi}(\mathbf{r}+\mathbf{x})\right|$$

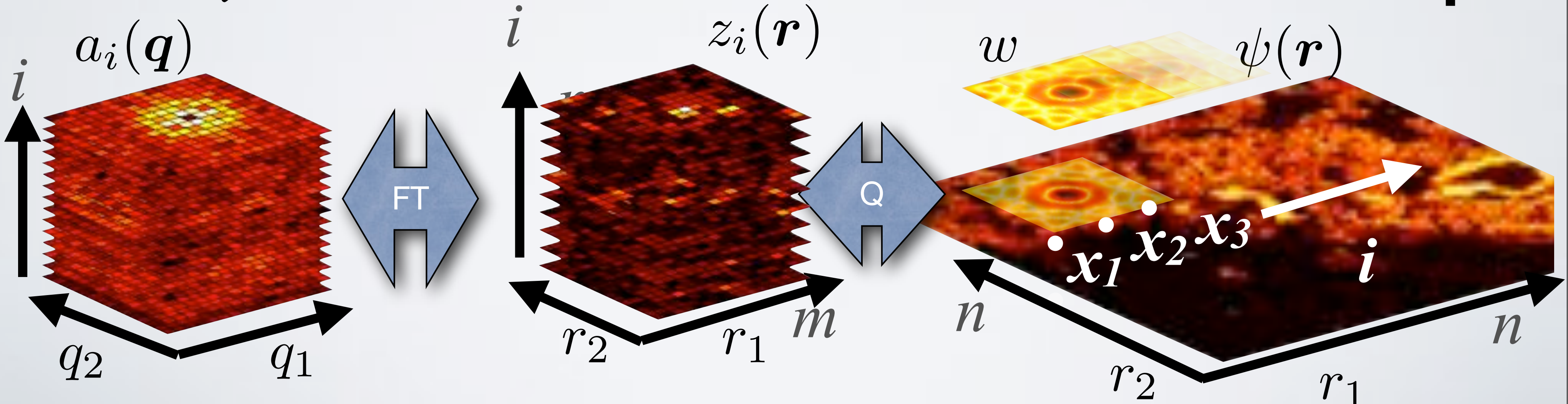
probe

Karle, Hoppe, ~1970
Rodemburg ~1980...
Chapman ~1990...
Pfeiffer ~2007
Thibault ~2008

....

amplitude $\boldsymbol{a}$ | Fourier transform $\boldsymbol{F}$ | scanning illumination $\boldsymbol{Q}$ | unknown ψ

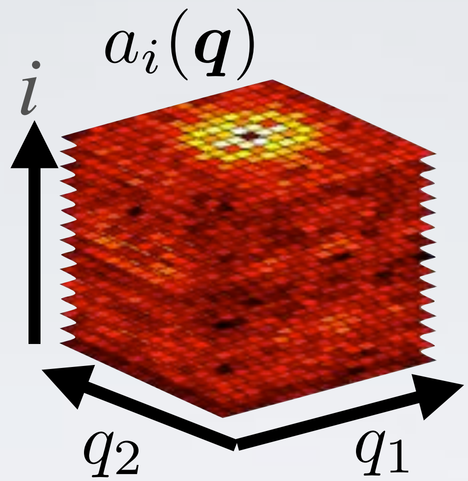
$$\begin{pmatrix} a_1 \\ a_2 \\ \vdots \end{pmatrix} = \left| \begin{pmatrix} \mathcal{F} &&&&& \\ & \mathcal{F} &&&& \\ && \mathcal{F} &&& \\ &&& \mathcal{F} && \\ &&&& \mathcal{F} & \\ &&&&& \mathcal{F} \end{pmatrix} \begin{pmatrix} \boxed{w \quad Q_1} \\ \boxed{w \quad Q_2} \\ w \\ w \\ w \\ w \end{pmatrix} \begin{pmatrix} \psi \end{pmatrix} \right|$$

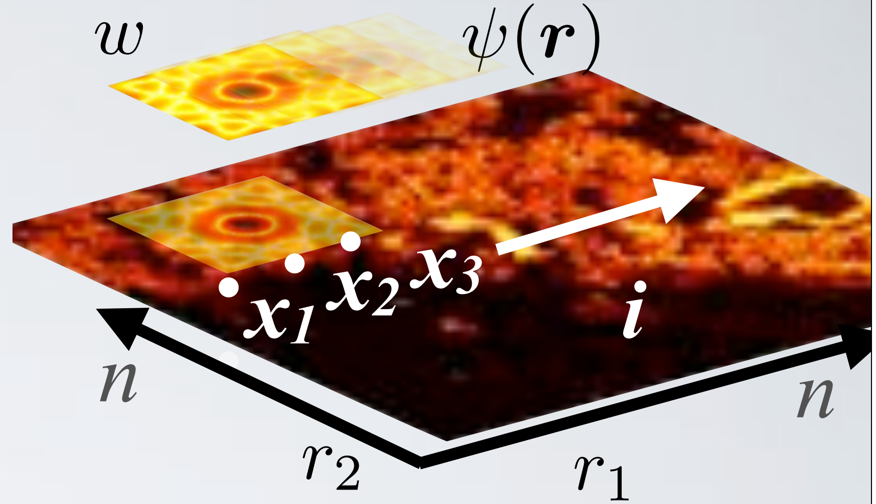


Scanning Diffractive imaging

data unknown

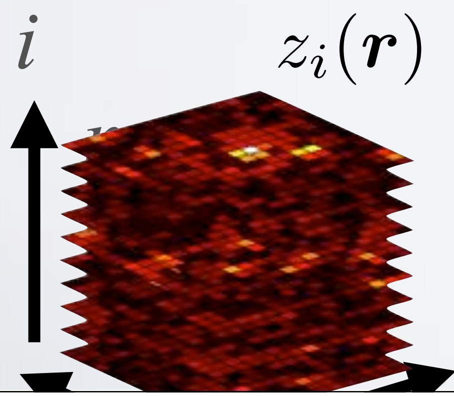
$$\boldsymbol{a} = |\boldsymbol{F}\mathbf{Q}\hat{\psi}|$$





introduce intermediate variable $\mathbf{z}$

$$\boxed{\begin{aligned} \boldsymbol{a} &= |\boldsymbol{F}\hat{\mathbf{z}}|, \\ \hat{\mathbf{z}} &= \mathbf{Q}\hat{\psi}, \end{aligned}}$$



projections

fit data $$P_F\mathbf{z} = \boldsymbol{F}^* \frac{\boldsymbol{F}\mathbf{z}}{|\boldsymbol{F}\mathbf{z}|} \cdot \boldsymbol{a}.$$

satisfy "overlap" $$P_Q = \mathbf{Q}(\mathbf{Q}^*\mathbf{Q})^{-1}\mathbf{Q}^*,$$

ptychographic data (5.3.2.1) ALS

Ptychography

super-resolution combining scanning with diffraction

(future) Combine

- Tomography
- Blind deconvolution
- Phase retrieval
- multiple scattering
- Vibrations
- denoising

ALS Beamline 5.3.2, 2012

Scanning microscopy

scanning diffraction

removed from web version

X 7 resolution enhancement

T Tyliszczak, R. Celestre, A D. Kilcoyne, , A. Schirotzek, T. Warwick(ALS),

CONCLUSIONS

- Experiments in photon science are very diverse
- Sparse modeling is a powerful method to extract information from noisy data
- High frame rate enables to achieve higher SNR or resolution: imaging of samples previously impossible
- Phase retrieval remains an open issue