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Atomic Resolution Holography Using Thermal Neutrons

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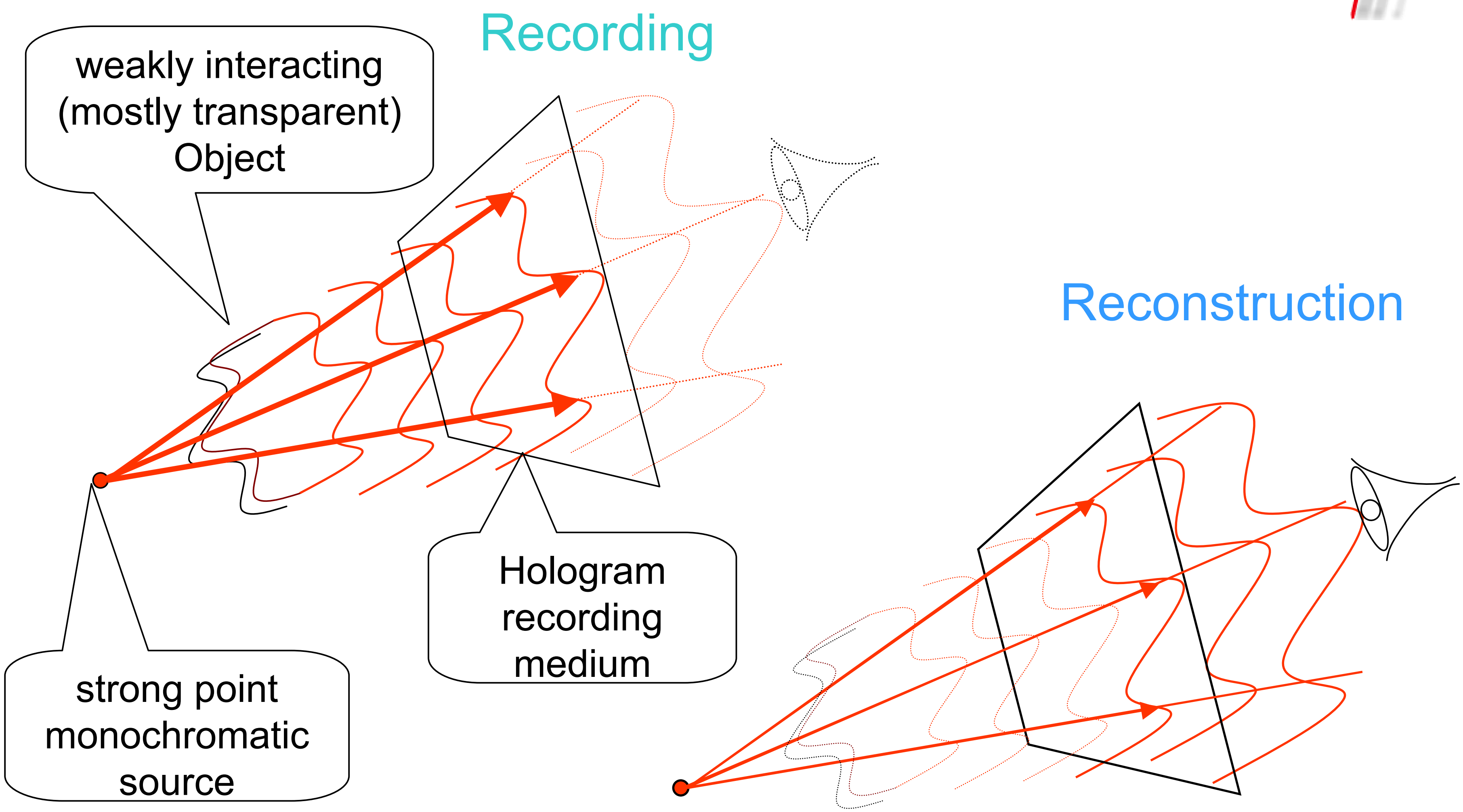
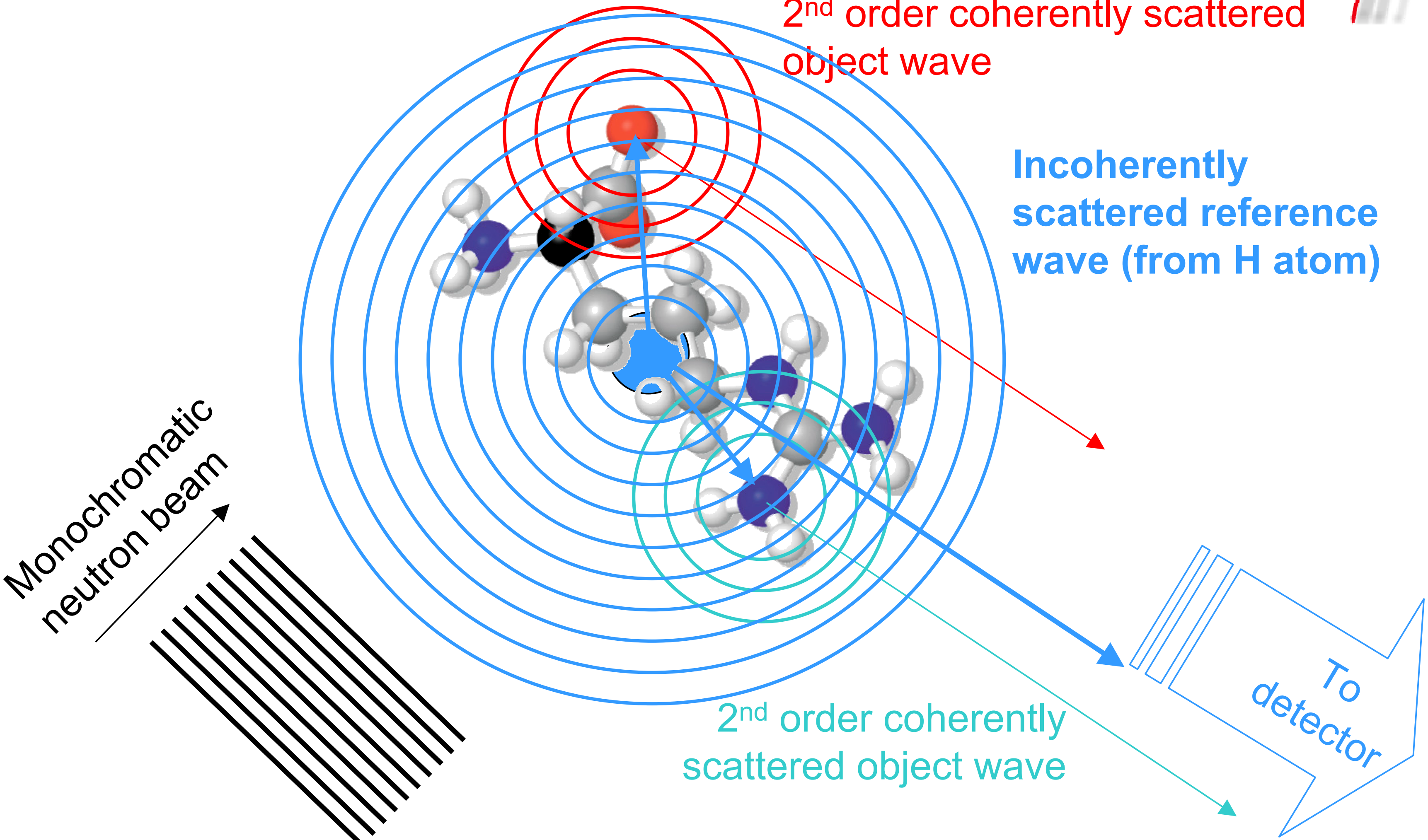
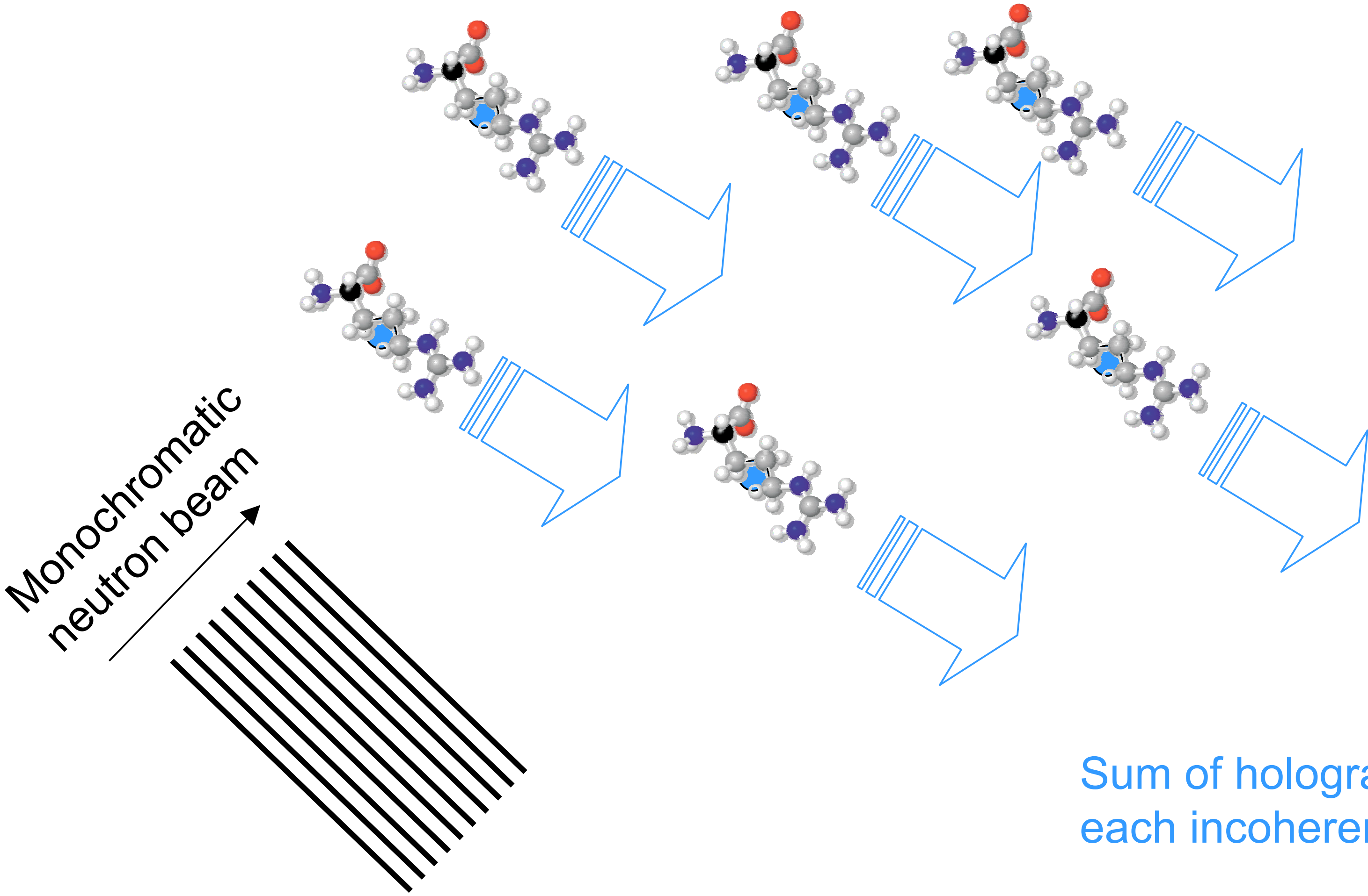
Gabor Holography



Atomic Structure Holography with thermal neutron waves

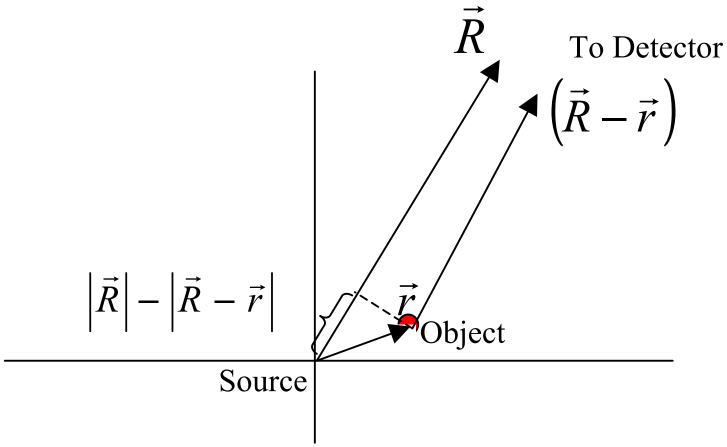


Atomic structure holography of macroscopic samples



Sum of holograms from each incoherent scatterer

S-Wave Diffraction for a Single Point Source



$$\psi_{ref}(\mathbf{R}) = a\frac{e^{ikR}}{R}$$

Time-independent (stationary), scalar, spherical waves centered at **R - r**

$$\psi_{obj}(\mathbf{R}) = a\sum_{j=1}^{j=n}\frac{e^{ikr_j}}{r_j} b_j \frac{e^{ik|\mathbf{R}-\mathbf{r}_j|}}{|\mathbf{R}-\mathbf{r}_j|}$$

$$\psi_{tot}(\mathbf{R}) = a\frac{e^{ikR}}{R}\left(1+\sum_{j=1}^{j=n}\left\{\frac{R}{|\mathbf{R}-\mathbf{r}_j|}\right\}\frac{b_j}{r_j}e^{i(kr_j - k\{R-|\mathbf{R}-\mathbf{r}_j|\})}\right)$$

Convert detector coordinates to direction

for R >> r :

$$k(|\mathbf{R}| - |\mathbf{R}-\mathbf{r}_o|) = k(\mathbf{r}_o \cdot \hat{\mathbf{R}}) = \mathbf{k}\cdot\mathbf{r}_o$$

$$\chi(\mathbf{k}) = \sum_{j=1}^{j=n}\frac{b_j}{r_j}e^{i(kr_j - \mathbf{k}\cdot\mathbf{r}_j)}$$

complex modulation function

detected intensity

$$I(\mathbf{k}) = \psi_{tot}^*(\mathbf{R})\psi_{tot}(\mathbf{R}) \approx \frac{a^*a}{R^2}\left(1 + 2\mathrm{Real}\{\chi(\mathbf{k})\} + |\chi(\mathbf{k})|^2\right)$$

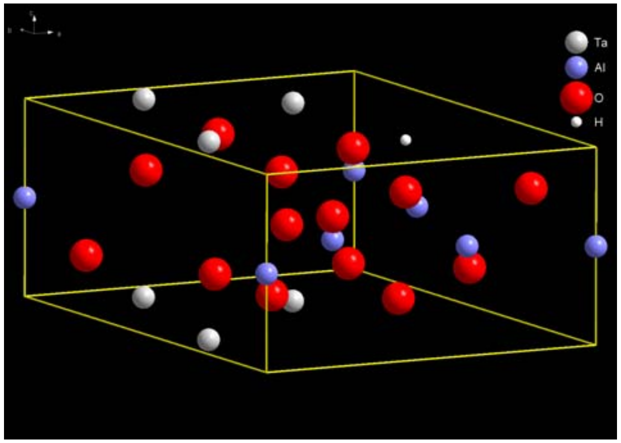
constant | small | much smaller

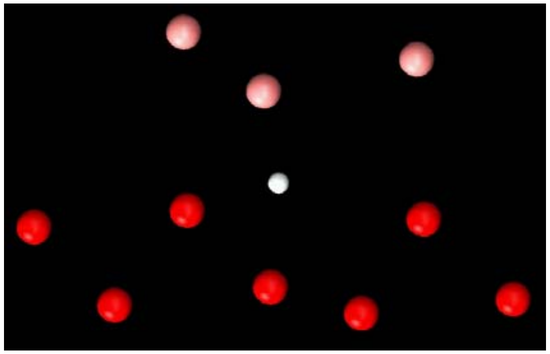
observed intensity modulation

An "Easy" Demonstration Sample

Simpsonite*

- No connection to Homer or Bart
- Mineral first discovered in Australia
- Simple structure
- 1 H atom

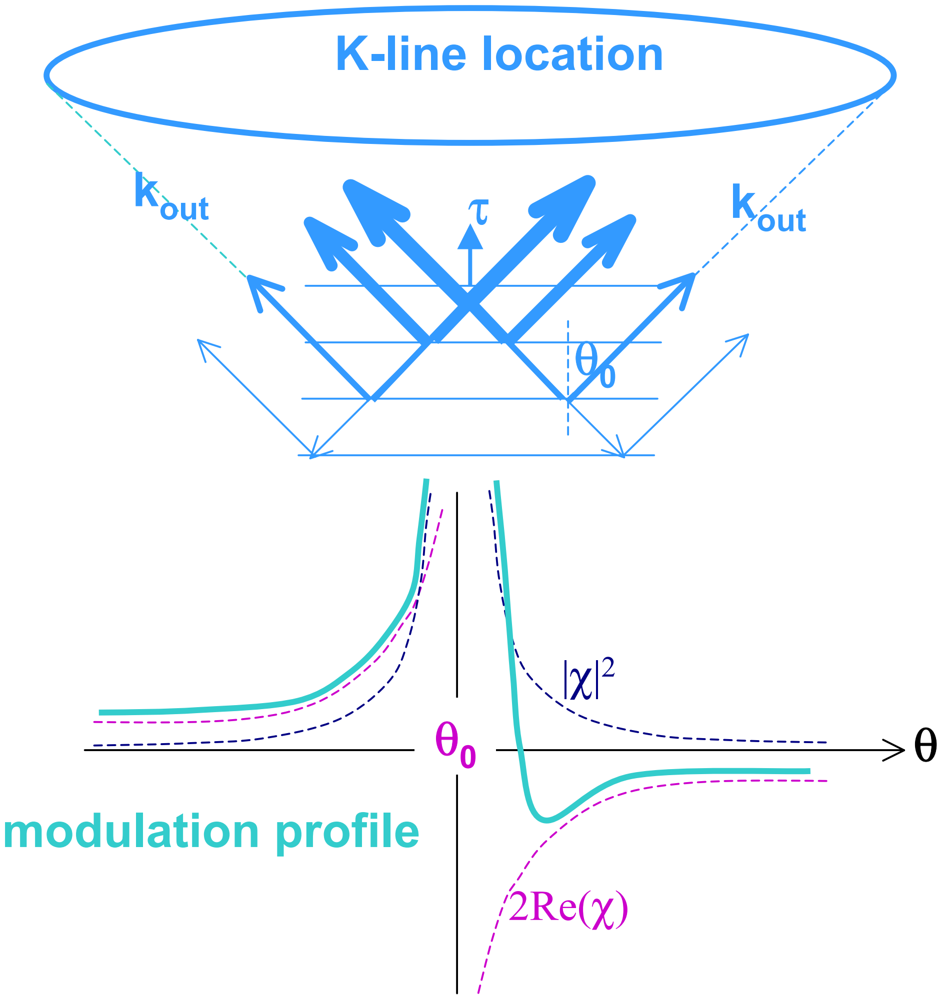




* Thanks to Ian Swainson (NRC) and Scott Ecrit (Canadian Museum of Nature)

Illustrations courtesy of Rob Hammond

s-Wave diffraction from planes: "K-Lines"



$$\chi(\mathbf{k}_{out}) = \int_{\mathbf{r}_o} \frac{b(\mathbf{r}_o)e^{i(kr_o - \mathbf{k}_{out}\cdot\mathbf{r}_o)}}{r_o}\,\mathrm{d}\mathbf{r}_o$$

$$B(\mathbf{q}) = \frac{1}{\sqrt{(2\pi)^3}}\int b(\mathbf{r})e^{-i\mathbf{q}\cdot\mathbf{r}}\mathrm{d}\mathbf{r}$$

$$\chi(\mathbf{k}_{out}) = \frac{4\pi}{\sqrt{(2\pi)^3}}\int \frac{B(\mathbf{q})}{|\mathbf{q}|^2 - 2\mathbf{k}_{out}\cdot\mathbf{q}}\mathrm{d}\mathbf{q}$$

$$B(\mathbf{q}) = \sum_{h,k,l} F_{hkl}\,\delta(\mathbf{q} - h\boldsymbol{\tau}_a - k\boldsymbol{\tau}_b - l\boldsymbol{\tau}_c)$$

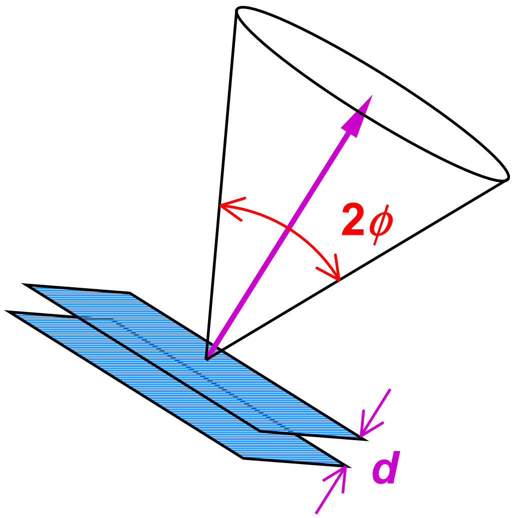
$$\chi(\mathbf{k}_{out}) = \frac{4\pi}{\sqrt{(2\pi)^3}}\sum_{h,k,l}\frac{F_{hkl}}{|\boldsymbol{\tau}_{hkl}|^2 - 2\mathbf{k}_{out}\cdot\boldsymbol{\tau}_{hkl}}$$

$$I_{1s} \cong \frac{a^*a}{R^2}\left[1 + 2\,\mathrm{Re}(\chi(\mathbf{k}_{out})) + |\chi(\mathbf{k}_{out})|^2\right]$$

What We Expect to See

Interference leads to cones of scattering with:

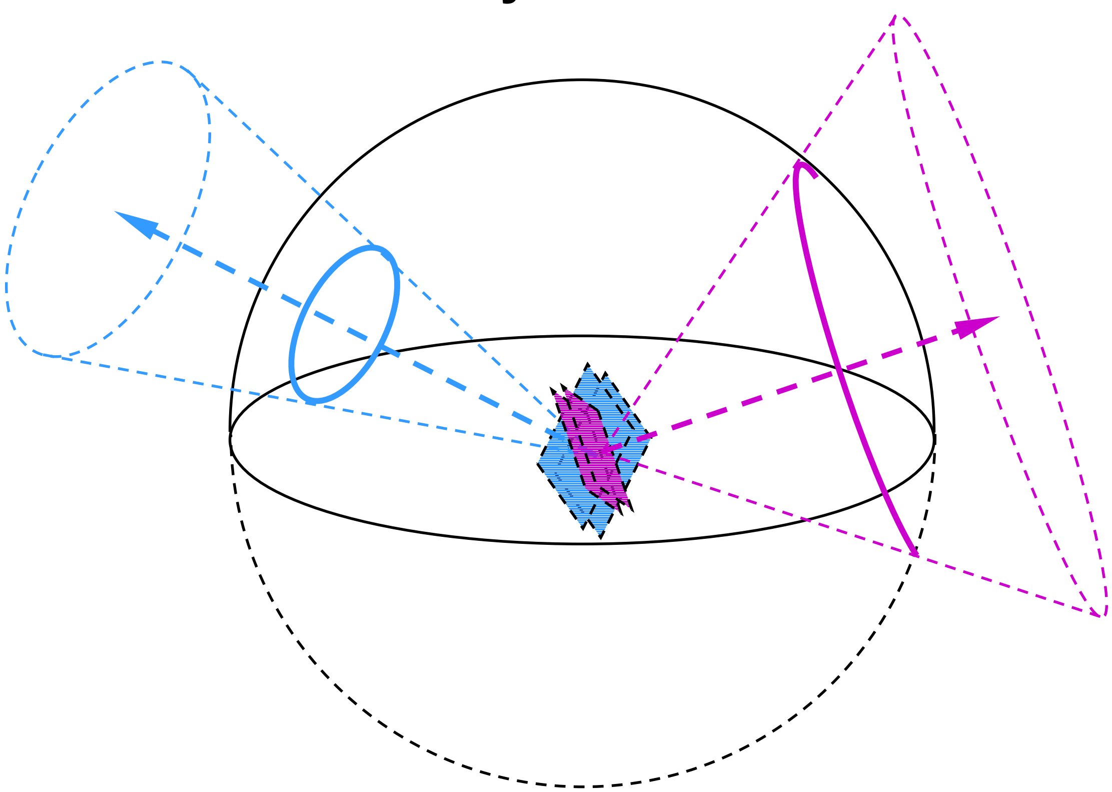
- cone axis parallel to the plane normal
- characteristic angle relates wavelength and atomic spacing



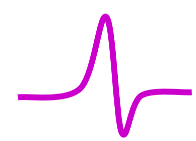
$$\lambda = 2d \cos \phi$$

Expect to See “K-Lines”

Looking over a sphere of scattering, these cones intersect the sphere forming rings, known as Kossel Lines, Kikuchi Lines or collectively as “K-Lines”

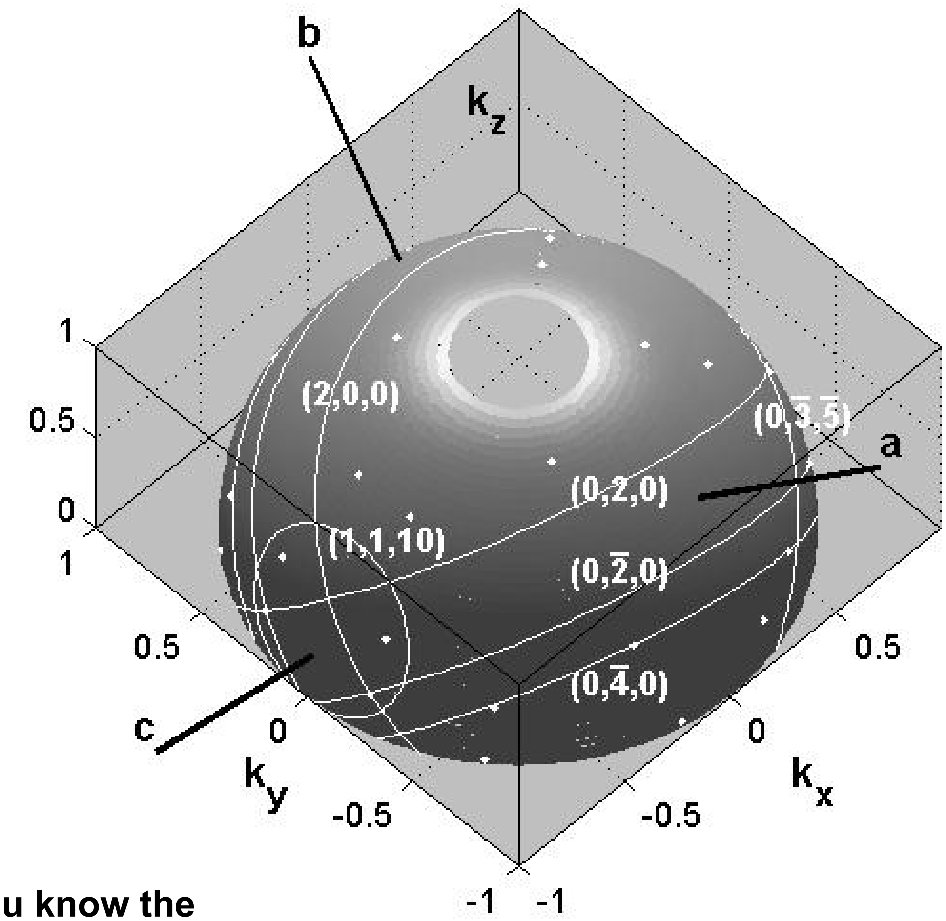


Intensity variations across *the K-lines*



Due to symmetry, a hemisphere is sufficient

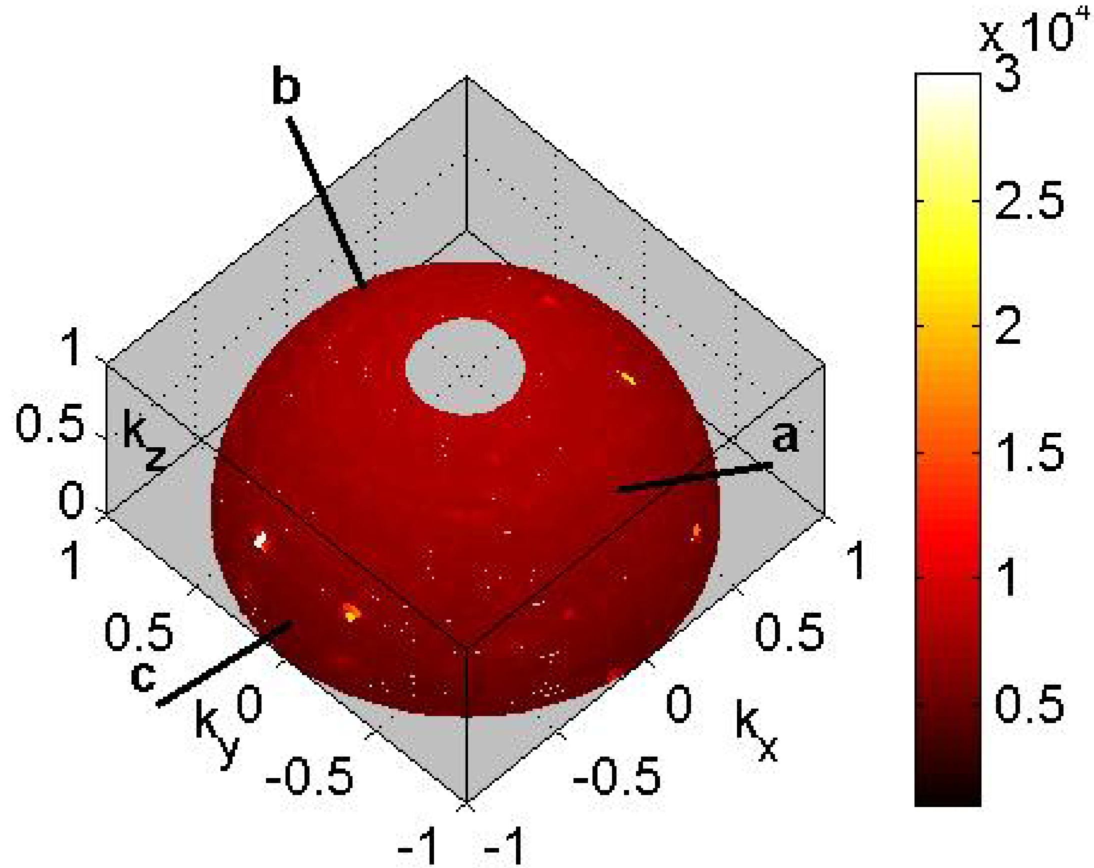
Expect to See “K-Lines”



Can easily calculate once you know the crystal orientation and radiation wavelength.

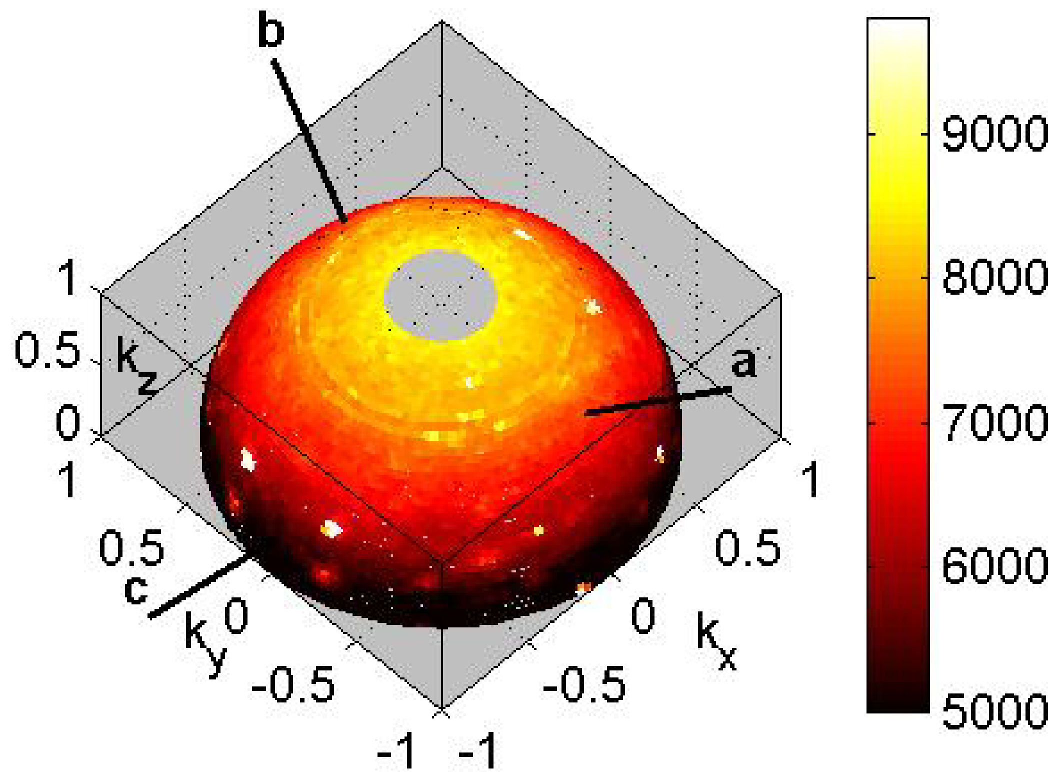
K-lines:
A Needle in a Hay Stack

- Bragg peaks very intense
- Hologram data is in the featureless “background”

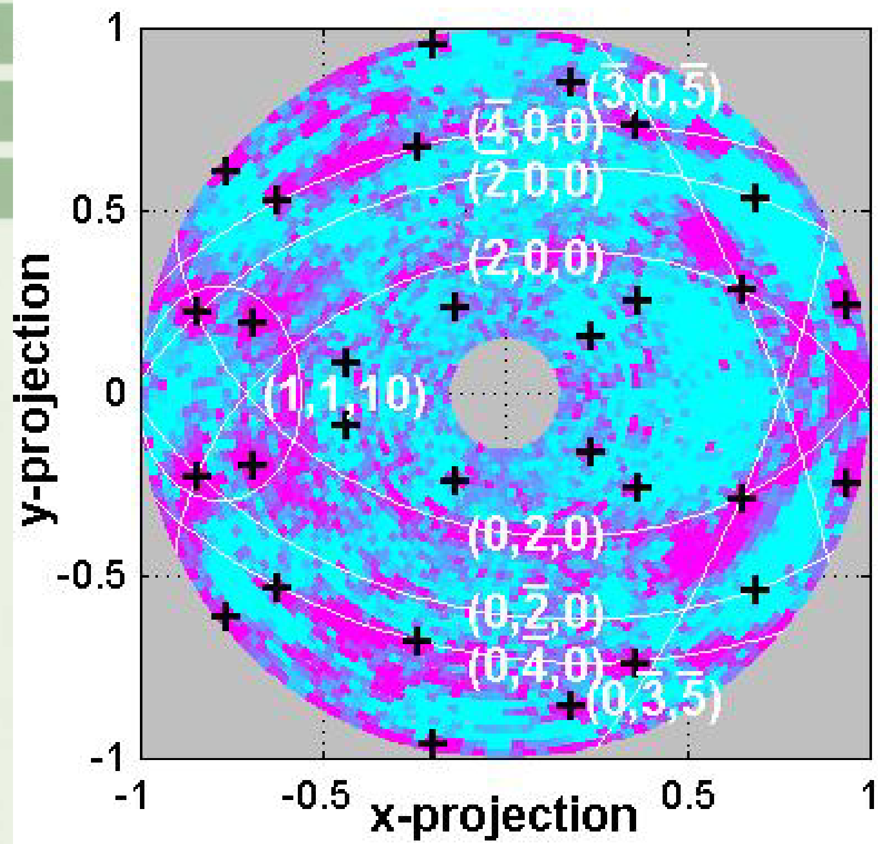


Looking for the Needle
Reduce Dynamic Range

- The “background” is not featureless, but still dominated by slowly varying intensity
  - Attenuation
  - Debye-Waller factor
- Remove by fitting to slowly varying functions in ϕ and θ



Found the Needle
K-Lines Appear



Observation of Kossel and Kikuchi Lines in Thermal Neutron Incoherent Scattering
B. Sur, R. B. Rogge, R. P. Hammond, V. N. P. Anghel, and J. Katsaras
PRL, **88** (6), 065505 (2002)

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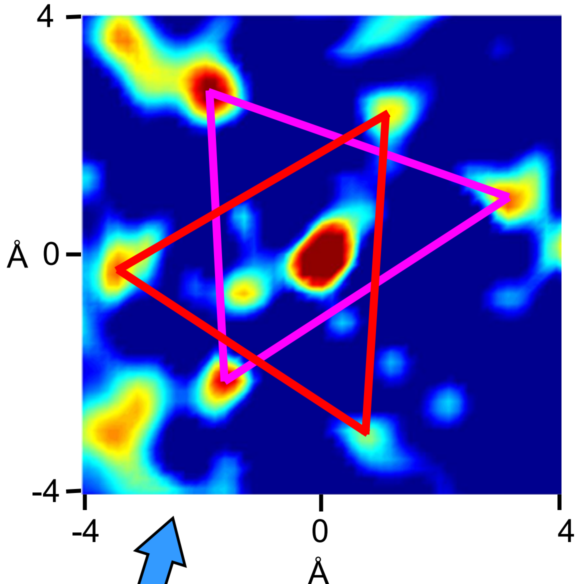
The Reconstruction

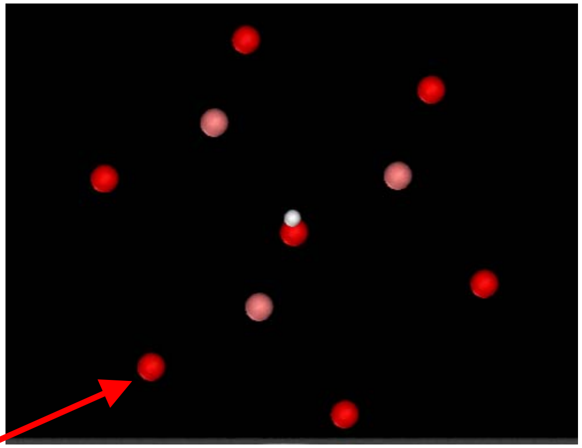


Slice of 3D data set taken at the known distance *below* the H atom to the plane of ***red*** O atoms

No model system required

Crystal structure falls out of reconstruction

Obtain a full 3D view (H atom view)



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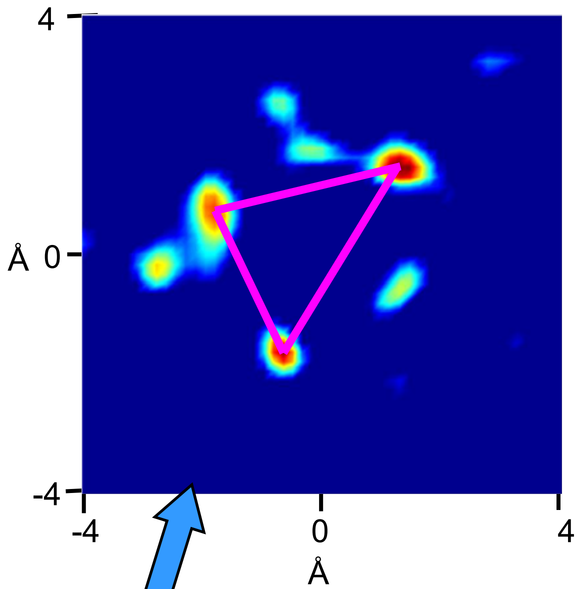
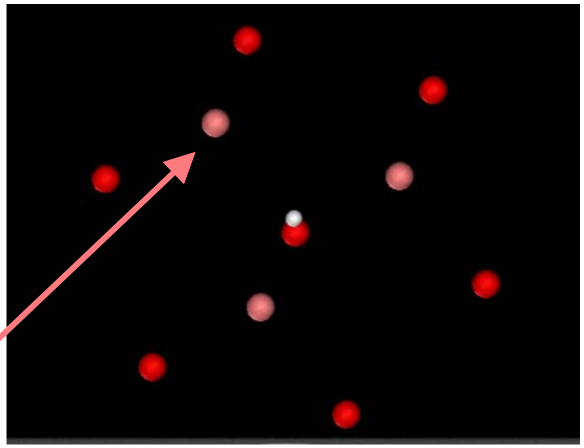
The Reconstruction





Slice from 3D data set taken at the known distance *above* the H atom to the plane of *light red* O atoms

Applications

- Hologram of atoms in general… *
  - Solves the phase problem
  - Permits study of systems that cannot be crystallized (e.g. biological macromolocules, catalyst particles, nanostructures);
  - Has been used to:
    - locate dopant atoms in semiconductors and alloys;
    - probe ordering in alloys, quasicrystals;
    - look at interfaces and films

* J. Spence, Nature (News and Views), **410,** 1037-1040 (2001)

Applications
Neutron Holography

- Neutrons are coherently scattered equally well by light and heavy atoms
- Neutrons can distinguish similar-Z atoms and can distinguish some isotopes
- The internal source H atom is abundant in biological materials and polymers
- H and D scatter very differently
- Neutrons are gentle, causing little or no damage to delicate systems

Take advantage of the complementarity of neutrons to tackle those systems that cannot be easily studied with electromagnetic radiation