Diffraction

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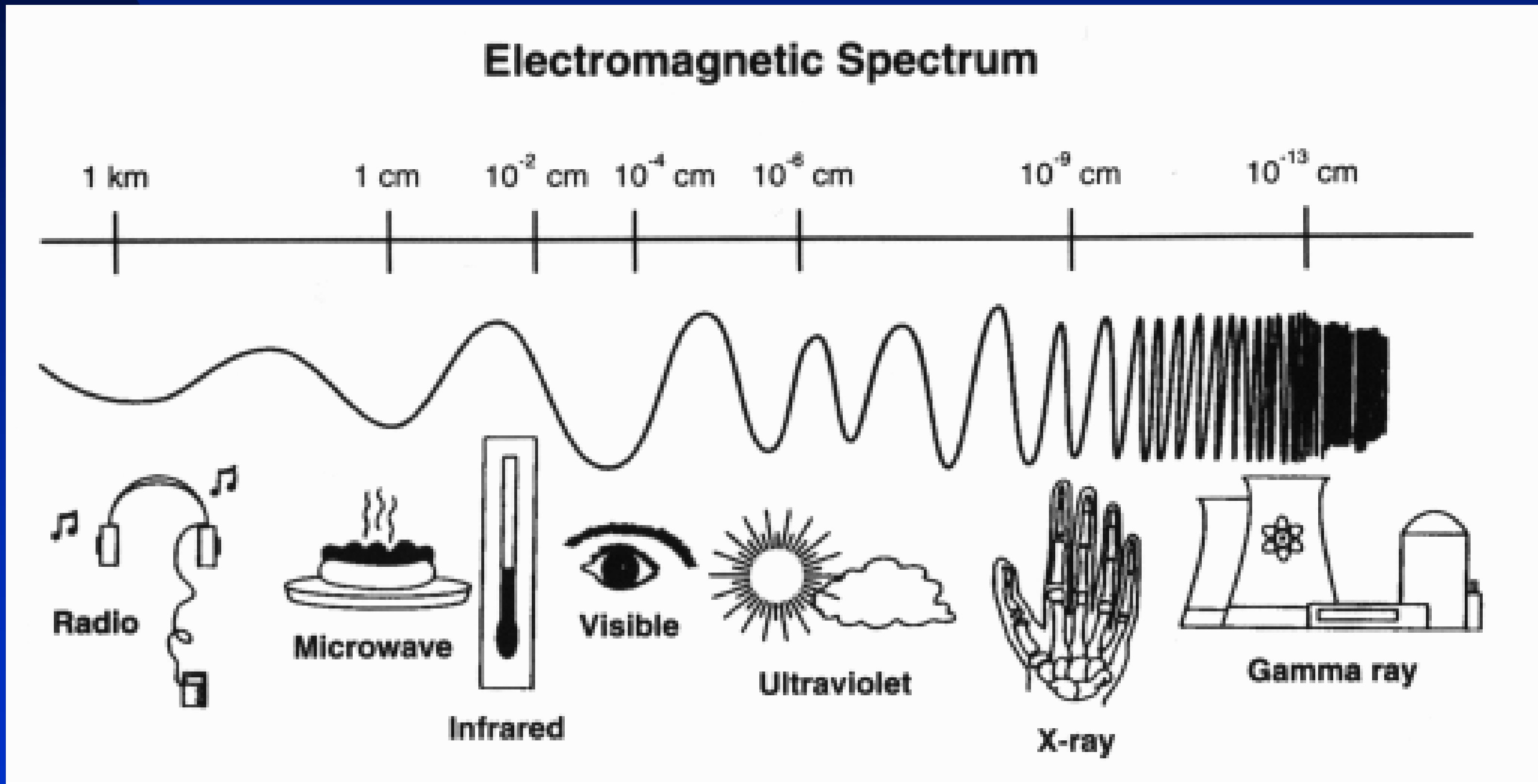
Part 1 Kinematical Theory

Introduction

- What I intend to give in this lecture:
  - Basic Concepts of "Diffraction"
    - Simpler Case: Kinematical Theory (Part 1)
    - Complicated Case: Dynamical Theory (Part 2)

- The speaker has been worked on experimental dynamical diffraction for 25 years. Now, he is in charge of X-ray optics for SPring-8, world's largest 3rd generation synchrotron facility in Japan.

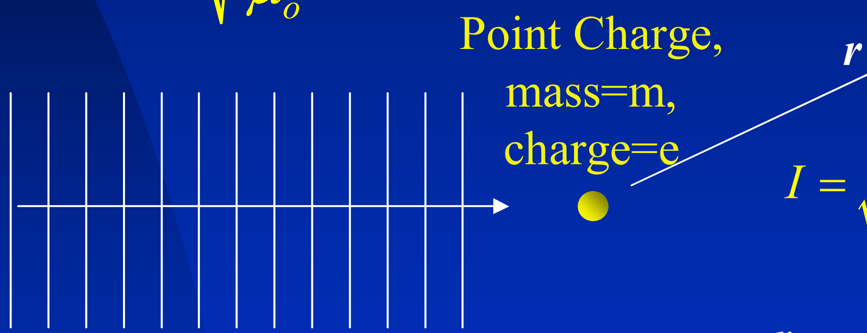
X-Rays as Shorter Wavelength Electromagnetic Wave



Scattering of x-rays by a point charge (Thomson Scattering)

$$E_{radiation} = \frac{1}{4\pi\varepsilon_0 c^2}\frac{e\left(\ddot{\boldsymbol{x}}\times\boldsymbol{r}\right)\times\boldsymbol{r}}{r^3}$$

$$I_0 = \sqrt{\frac{\varepsilon_o}{\mu_o}}E_0^2$$



$$I = \sqrt{\frac{\varepsilon_o}{\mu_o}}\left(\frac{e^2\sin\alpha}{4\pi mrc^2}E_0\right)^2 = r_0^2\sin^2\alpha\left(\frac{I_0}{r^2}\right)$$

$$r_0 = \frac{e^2}{4\pi\varepsilon_o mc^2} : Classical\ Electron\ Radius$$

$$= 2.81776\times10^{-15}\ m$$

Electromagnetic Plane Wave

$$\boldsymbol{E} = \boldsymbol{E}_0\exp\left[i\left(\boldsymbol{K}_o\cdot\boldsymbol{r}-\omega t\right)\right]$$

$$\boldsymbol{B} = \frac{\boldsymbol{k}}{\omega}\times\boldsymbol{E}_0\exp\left[i\left(\boldsymbol{K}_o\cdot\boldsymbol{r}-\omega t\right)\right] = \frac{\hat{\boldsymbol{k}}}{c}\times\boldsymbol{E}$$

Lorentz Force

$$m\ddot{\boldsymbol{x}} = e(\boldsymbol{E}+\dot{\boldsymbol{x}}\times\boldsymbol{B})$$

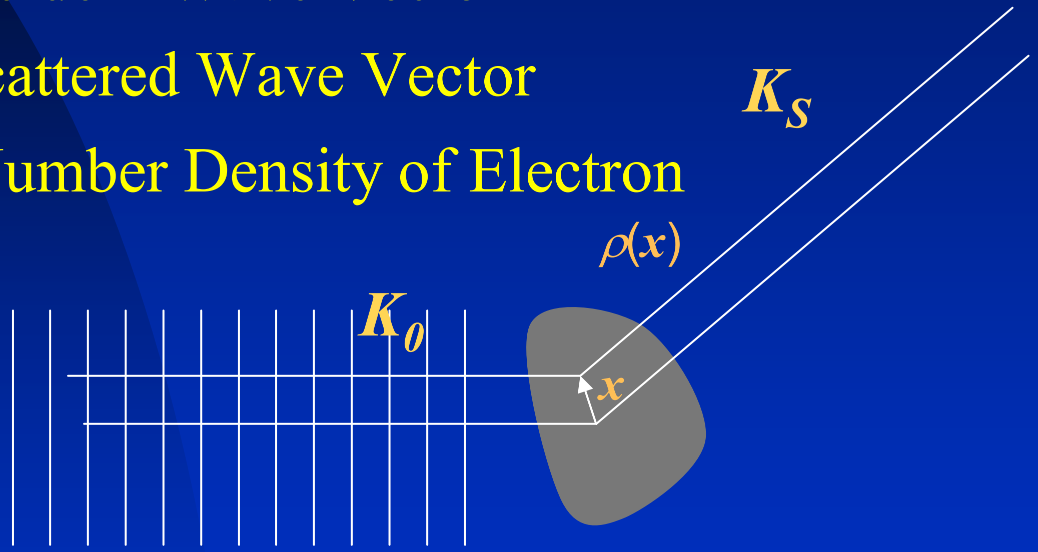
$$\left|\dot{\boldsymbol{x}}\right| << c \Rightarrow \ddot{\boldsymbol{x}} = \frac{e}{m}\boldsymbol{E}$$

Scattering of x-rays by distributed charge (1/2)

K_0 : Incident Wave Vector

K_S : Scattered Wave Vector

$\rho(x)$: Number Density of Electron



Contribution from a volume element d^3x at x

$$dE^{distribution}_{radiation} = E^{point\ charge}_{radiation}\,\rho(x)\exp\left[-i(K_s - K_o)\cdot x\right]d^3x$$

Scattering of x-rays by distributed charge (2/2)

$$\boldsymbol{E}_{radiation}^{distribution} = \boldsymbol{E}_{radiation}^{point\ charge} \iiint \rho(\boldsymbol{x}) \exp\left[-i\left(\boldsymbol{K}_s - \boldsymbol{K}_o\right)\cdot \boldsymbol{x}\right] d^3\boldsymbol{x}$$

3D Fourier Transform of Charge Density

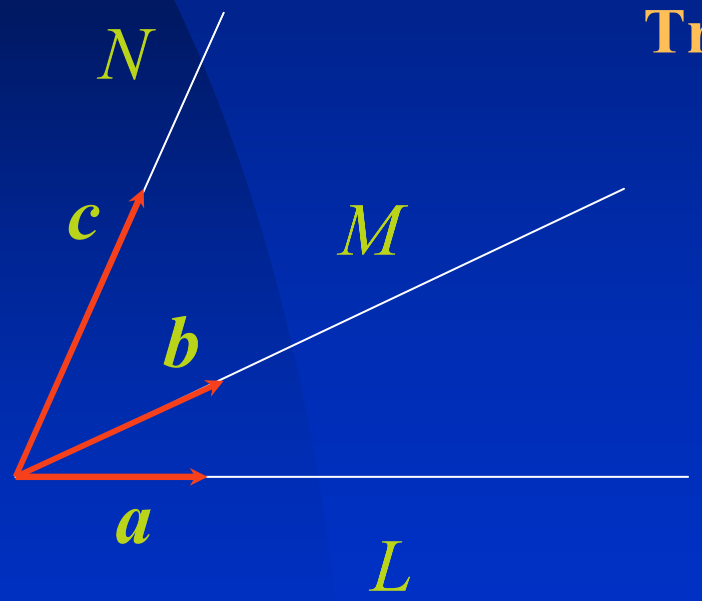
Scattered Intensity

$$I = I^{single} \left| \iiint \rho(\boldsymbol{x}) \exp\left[-i\left(\boldsymbol{K}_s - \boldsymbol{K}_o\right)\cdot \boldsymbol{x}\right] d^3\boldsymbol{x} \right|^2$$

$$= \frac{r_o^2 \sin^2 \alpha}{r^2} I_o \left| \iiint \rho(\boldsymbol{x}) \exp\left[-i\left(\boldsymbol{K}_s - \boldsymbol{K}_o\right)\cdot \boldsymbol{x}\right] d^3\boldsymbol{x} \right|^2$$

Electronic Charge Distribution in Crystal

Crystal = 3D Regular Stacking of Molecules



Translation Symmetry

$$\rho(\boldsymbol{x}) = \rho(\boldsymbol{x} + l\boldsymbol{a} + m\boldsymbol{b} + n\boldsymbol{c})$$

l, m, n: integers

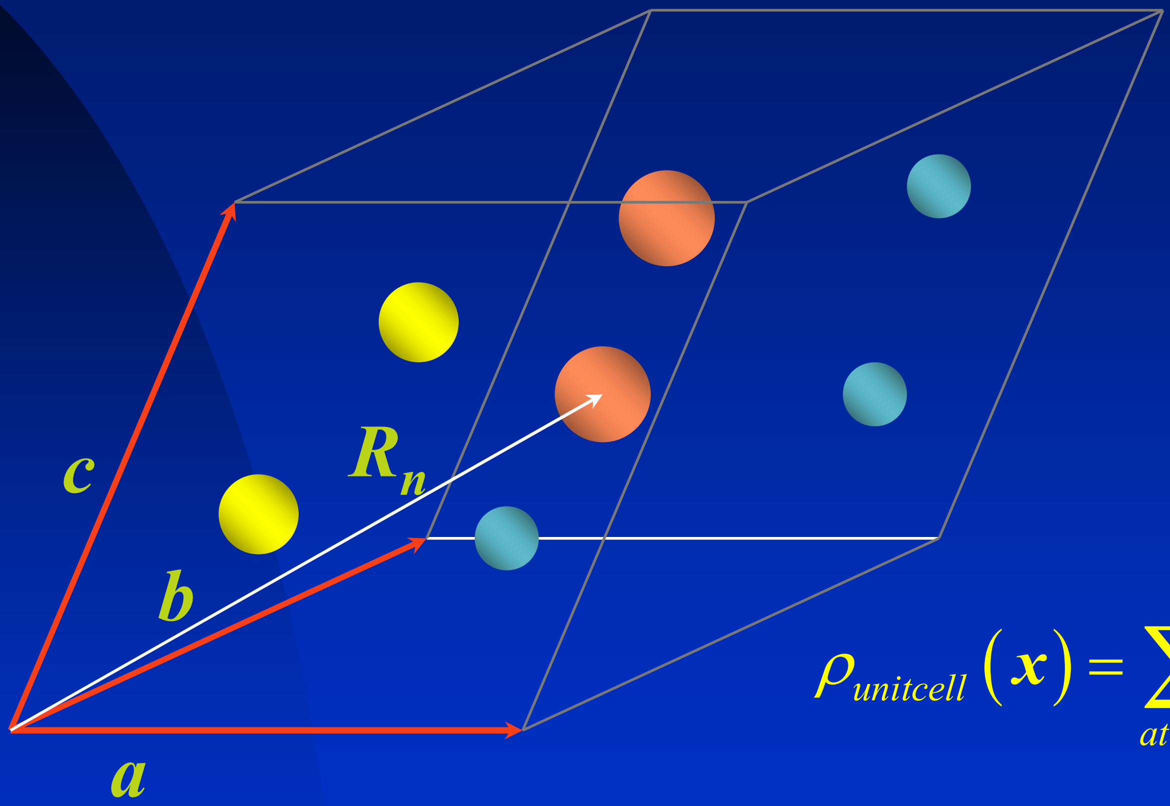
Fourier Transform of the Electronic Charge Distribution in Crystal

$$\int_{crystal} \rho(\boldsymbol{x}) e^{-i\boldsymbol{K}\cdot\boldsymbol{x}} d^3\boldsymbol{x} = \sum_{l=0}^{L-1}\sum_{m=0}^{M-1}\sum_{n=0}^{N-1} \int_{unitcell} \rho(\boldsymbol{x}+l\boldsymbol{a}+m\boldsymbol{b}+n\boldsymbol{c}) e^{-i\boldsymbol{K}\cdot(\boldsymbol{x}+l\boldsymbol{a}+m\boldsymbol{b}+n\boldsymbol{c})} d^3\boldsymbol{x}$$

$$= \sum_{l=0}^{L-1}\sum_{m=0}^{M-1}\sum_{n=0}^{N-1} e^{-i\boldsymbol{K}\cdot(\boldsymbol{x}+l\boldsymbol{a}+m\boldsymbol{b}+n\boldsymbol{c})} \int_{unitcell} \rho(\boldsymbol{x}) e^{-i\boldsymbol{K}\cdot\boldsymbol{x}} d^3\boldsymbol{x}$$

$$= \left(\frac{1-e^{-iL\boldsymbol{K}\cdot\boldsymbol{a}}}{1-e^{-i\boldsymbol{K}\cdot\boldsymbol{a}}}\right)\left(\frac{1-e^{-iM\boldsymbol{K}\cdot\boldsymbol{b}}}{1-e^{-i\boldsymbol{K}\cdot\boldsymbol{b}}}\right)\left(\frac{1-e^{-iN\boldsymbol{K}\cdot\boldsymbol{c}}}{1-e^{-i\boldsymbol{K}\cdot\boldsymbol{c}}}\right) \int_{unitcell} \rho(\boldsymbol{x}) e^{-i\boldsymbol{K}\cdot\boldsymbol{x}} d^3\boldsymbol{x}$$

Charge Density in Unit Cell



$$\rho_{unitcell}\left(\boldsymbol{x}\right)=\sum_{atom}\rho_{atom}^{(a)}\left(\boldsymbol{x}-\boldsymbol{R}_n\right)$$

$$\int_{unitcell}\rho\left(\boldsymbol{x}\right)e^{-i\boldsymbol{K}\cdot\boldsymbol{x}}d^3\boldsymbol{x}=\sum_{atom}\int_{atom}\rho_{atom}^{(a)}\left(\boldsymbol{x}-\boldsymbol{R}_n\right)e^{-i\boldsymbol{K}\cdot\boldsymbol{x}}d^3\boldsymbol{x}=\sum_{atom}e^{-i\boldsymbol{K}\cdot\boldsymbol{R}_n}\int_{atom}\rho_{atom}^{(a)}\left(\boldsymbol{x}\right)e^{-i\boldsymbol{K}\cdot\boldsymbol{x}}d^3\boldsymbol{x}$$

Atomic Scattering Factor, Structure Factor

Atomic Scattering Factor

$$f^{(a)}(\boldsymbol{K}) = \int_{atom} \rho_{atom}^{(a)}(\boldsymbol{x}) e^{-i\boldsymbol{K}\cdot\boldsymbol{x}} d^3\boldsymbol{x}$$

Structure Factor

$$F(\boldsymbol{K}) = \int_{unitcell} \rho(\boldsymbol{x}) e^{-i\boldsymbol{K}\cdot\boldsymbol{x}} d^3\boldsymbol{x} = \sum_{atom} e^{-i\boldsymbol{K}\cdot\boldsymbol{R}_n} f^{(a)}(\boldsymbol{K})$$

$$\int_{crystal} \rho(\boldsymbol{x}) e^{-i\boldsymbol{K}\cdot\boldsymbol{x}} d^3\boldsymbol{x} = \left(\frac{1-e^{-iL\boldsymbol{K}\cdot\boldsymbol{a}}}{1-e^{-i\boldsymbol{K}\cdot\boldsymbol{a}}}\right)\left(\frac{1-e^{-iM\boldsymbol{K}\cdot\boldsymbol{b}}}{1-e^{-i\boldsymbol{K}\cdot\boldsymbol{b}}}\right)\left(\frac{1-e^{-iN\boldsymbol{K}\cdot\boldsymbol{c}}}{1-e^{-i\boldsymbol{K}\cdot\boldsymbol{c}}}\right)\int_{unitcell} \rho(\boldsymbol{x}) e^{-i\boldsymbol{K}\cdot\boldsymbol{x}} d^3\boldsymbol{x}$$

$$= \left(\frac{1-e^{-iL\boldsymbol{K}\cdot\boldsymbol{a}}}{1-e^{-i\boldsymbol{K}\cdot\boldsymbol{a}}}\right)\left(\frac{1-e^{-iM\boldsymbol{K}\cdot\boldsymbol{b}}}{1-e^{-i\boldsymbol{K}\cdot\boldsymbol{b}}}\right)\left(\frac{1-e^{-iN\boldsymbol{K}\cdot\boldsymbol{c}}}{1-e^{-i\boldsymbol{K}\cdot\boldsymbol{c}}}\right) F(\boldsymbol{K})$$

Laue Function (1/3)

$$\frac{1-e^{-iL\boldsymbol{K}\cdot\boldsymbol{a}}}{1-e^{-i\boldsymbol{K}\cdot\boldsymbol{a}}}=\frac{e^{-i\frac{L\boldsymbol{K}\cdot\boldsymbol{a}}{2}}\left(e^{+i\frac{L\boldsymbol{K}\cdot\boldsymbol{a}}{2}}-e^{-i\frac{L\boldsymbol{K}\cdot\boldsymbol{a}}{2}}\right)}{e^{-i\frac{\boldsymbol{K}\cdot\boldsymbol{a}}{2}}\left(e^{+i\frac{\boldsymbol{K}\cdot\boldsymbol{a}}{2}}-e^{-i\frac{\boldsymbol{K}\cdot\boldsymbol{a}}{2}}\right)}=\frac{e^{-i\frac{L\boldsymbol{K}\cdot\boldsymbol{a}}{2}}\sin\frac{L\boldsymbol{K}\cdot\boldsymbol{a}}{2}}{e^{-i\frac{\boldsymbol{K}\cdot\boldsymbol{a}}{2}}\sin\frac{\boldsymbol{K}\cdot\boldsymbol{a}}{2}}$$

$$I_{crystal}=\frac{r_o^2\sin^2\alpha}{r^2}I_o\left|F\left(\boldsymbol{K}\right)\right|^2\left(\frac{\sin\frac{L\boldsymbol{K}\cdot\boldsymbol{a}}{2}}{\sin\frac{\boldsymbol{K}\cdot\boldsymbol{a}}{2}}\right)^2\left(\frac{\sin\frac{M\boldsymbol{K}\cdot\boldsymbol{b}}{2}}{\sin\frac{\boldsymbol{K}\cdot\boldsymbol{b}}{2}}\right)^2\left(\frac{\sin\frac{N\boldsymbol{K}\cdot\boldsymbol{c}}{2}}{\sin\frac{\boldsymbol{K}\cdot\boldsymbol{c}}{2}}\right)^2$$

Laue Function (2/3)

$$\boldsymbol{K}\cdot\boldsymbol{a}\to 0 \Rightarrow \left(\frac{\sin\frac{L\boldsymbol{K}\cdot\boldsymbol{a}}{2}}{\sin\frac{\boldsymbol{K}\cdot\boldsymbol{a}}{2}}\right)^2 \to L^2$$

$$\left(\frac{\sin\frac{L\boldsymbol{K}\cdot\boldsymbol{a}}{2}}{\sin\frac{\boldsymbol{K}\cdot\boldsymbol{a}}{2}}\right)^2 : \text{periodic function}$$

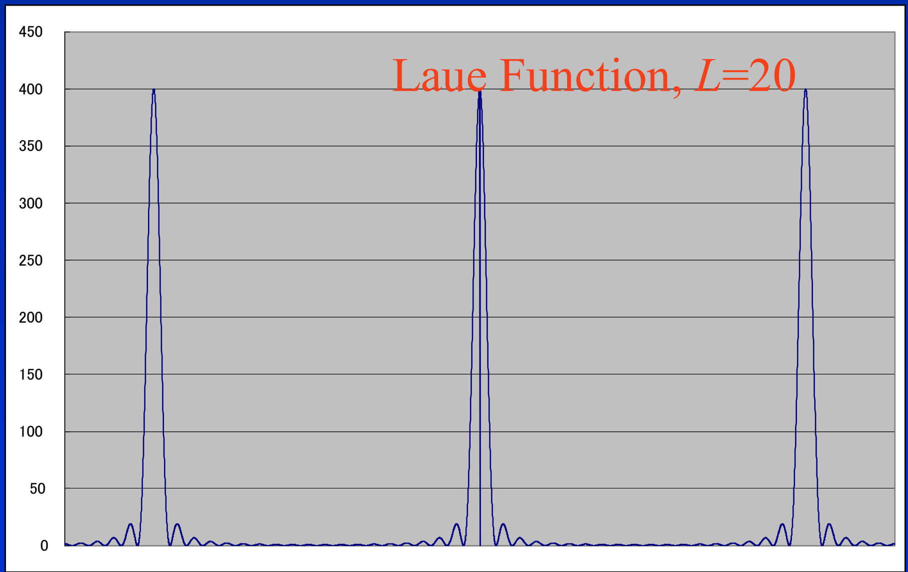
$$\boldsymbol{K}\cdot\boldsymbol{a}\to 2h\pi \Rightarrow \left(\frac{\sin\frac{L\boldsymbol{K}\cdot\boldsymbol{a}}{2}}{\sin\frac{\boldsymbol{K}\cdot\boldsymbol{a}}{2}}\right)^2 \to L^2\ ; h \text{ integer}$$

Similarly,

$$\boldsymbol{K}\cdot\boldsymbol{b}\to 2k\pi, k \text{ integer} \Rightarrow \left(\frac{\sin\frac{M\boldsymbol{K}\cdot\boldsymbol{b}}{2}}{\sin\frac{\boldsymbol{K}\cdot\boldsymbol{b}}{2}}\right)^2 \to M^2$$

and

$$\boldsymbol{K}\cdot\boldsymbol{c}\to 2l\pi, l \text{ integer} \Rightarrow \left(\frac{\sin\frac{N\boldsymbol{K}\cdot\boldsymbol{c}}{2}}{\sin\frac{\boldsymbol{K}\cdot\boldsymbol{c}}{2}}\right)^2 \to N^2$$



Laue Function (3/3)

- Scattering from a crystal is appreciable only when

$$\boldsymbol{K}\cdot\boldsymbol{a} = 2\pi h$$
$$\boldsymbol{K}\cdot\boldsymbol{b} = 2\pi k \qquad h,\ k,\ l\text{; integer}$$
$$\boldsymbol{K}\cdot\boldsymbol{c} = 2\pi l$$

- Then,

$$I_{crystal} = \frac{r_o^2 \sin^2\alpha}{r^2} I_o \left(LMN\right)^2 \left|F(\boldsymbol{K})\right|^2$$

Reciprocal Lattice (1/3)

Scattering from a crystal is appreciable only when

$$\boldsymbol{K}\cdot\boldsymbol{a} = 2\pi h$$
$$\boldsymbol{K}\cdot\boldsymbol{b} = 2\pi k$$
$$\boldsymbol{K}\cdot\boldsymbol{c} = 2\pi l$$

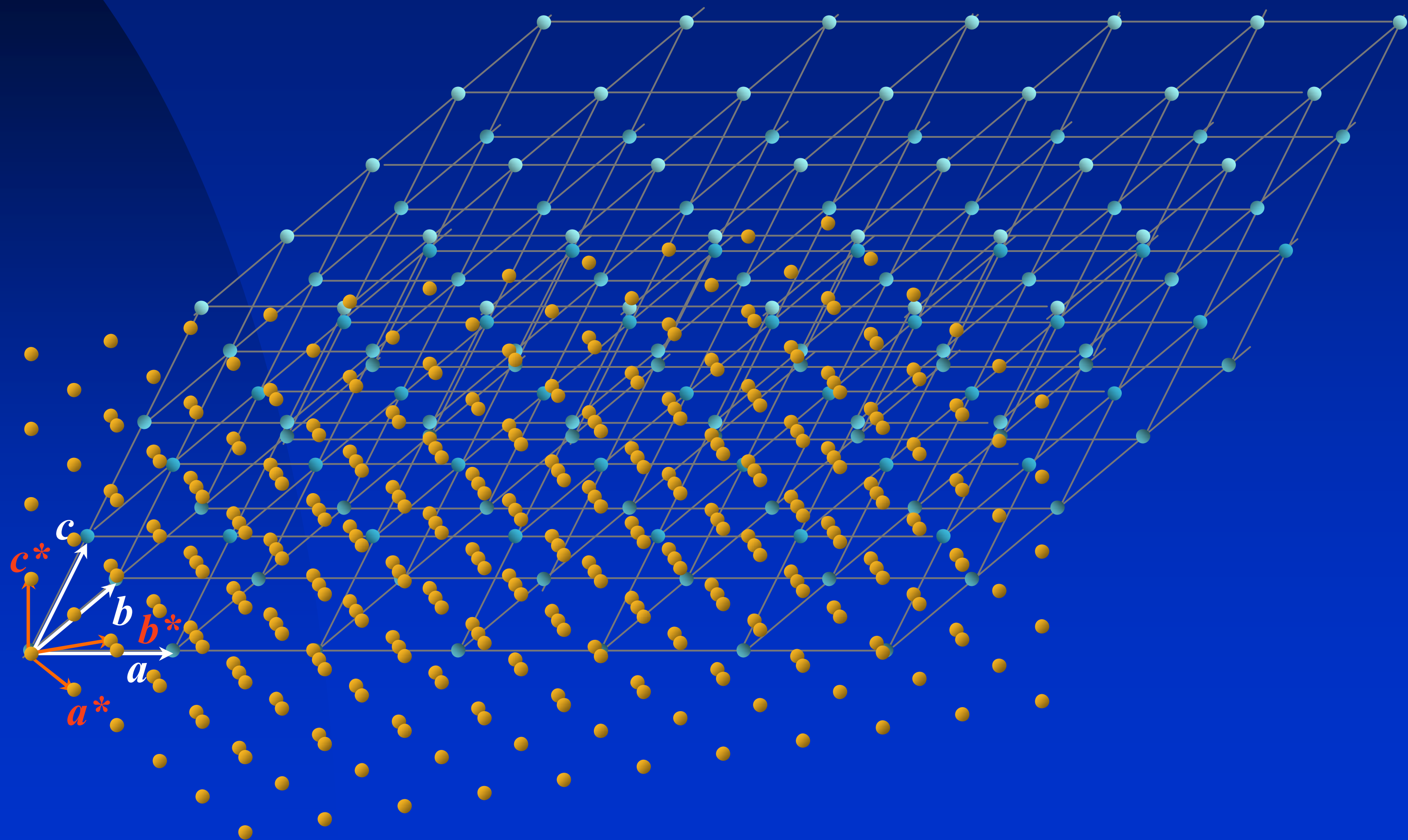
h, k, l : integer

Base Vectors of Reciprocal Lattice

$$\boldsymbol{a}^* = \frac{2\pi(\boldsymbol{b}\times\boldsymbol{c})}{\boldsymbol{a}\cdot(\boldsymbol{b}\times\boldsymbol{c})}, \boldsymbol{b}^* = \frac{2\pi(\boldsymbol{c}\times\boldsymbol{a})}{\boldsymbol{b}\cdot(\boldsymbol{c}\times\boldsymbol{a})}, \boldsymbol{c}^* = \frac{2\pi(\boldsymbol{a}\times\boldsymbol{b})}{\boldsymbol{c}\cdot(\boldsymbol{a}\times\boldsymbol{b})}$$

$$\boldsymbol{a}^*\cdot\boldsymbol{a} = 2\pi, \boldsymbol{a}^*\cdot\boldsymbol{b} = 0, \boldsymbol{a}^*\cdot\boldsymbol{c} = 0$$
$$\boldsymbol{b}^*\cdot\boldsymbol{b} = 2\pi, \boldsymbol{b}^*\cdot\boldsymbol{a} = 0, \boldsymbol{b}^*\cdot\boldsymbol{c} = 0$$
$$\boldsymbol{c}^*\cdot\boldsymbol{c} = 2\pi, \boldsymbol{c}^*\cdot\boldsymbol{a} = 0, \boldsymbol{c}^*\cdot\boldsymbol{b} = 0$$

Reciprocal Lattice (2/3)



Reciprocal Lattice (3/3)

- Reciprocal Lattice Vector

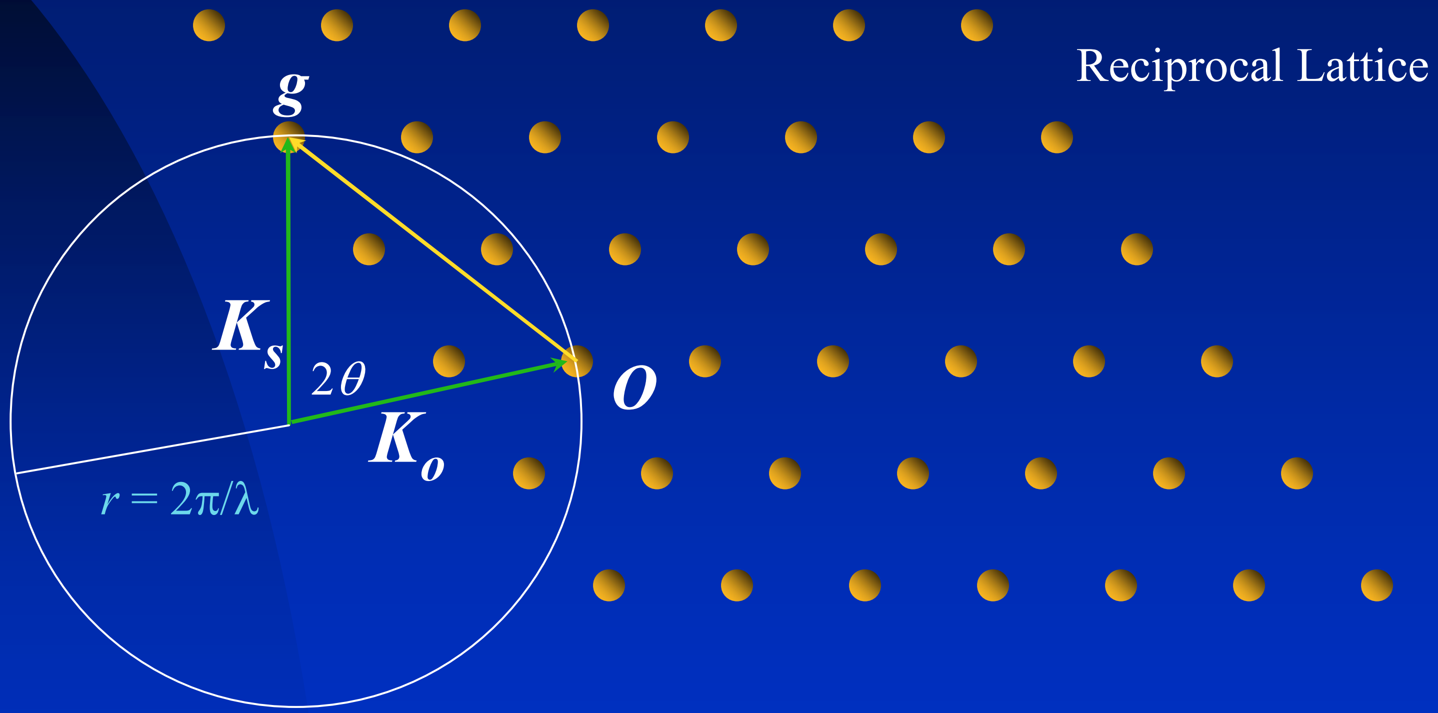
$$\boldsymbol{g} = h\boldsymbol{a}^* + k\boldsymbol{b}^* + l\boldsymbol{c}^*$$

h, k, l; integer

$$\boldsymbol{K} = \boldsymbol{g} \Rightarrow \boldsymbol{K}\cdot\boldsymbol{a} = 2\pi h, \boldsymbol{K}\cdot\boldsymbol{b} = 2\pi k, \boldsymbol{K}\cdot\boldsymbol{c} = 2\pi l$$

When the scattering vector, $\boldsymbol{K}=\boldsymbol{K}_s-\boldsymbol{K}_o$, corresponds to a reciprocal lattice vector, strong diffraction may be observed (necessary condition, but not a sufficient condition).

Ewald Sphere



$$|\boldsymbol{g}| = \frac{2\pi}{d_{hkl}}, |\boldsymbol{K}_o| = |\boldsymbol{K}_s| = \frac{2\pi}{\lambda} \Rightarrow \frac{2\pi}{\lambda}\sin\theta = \frac{\pi}{d_{hkl}}$$

Bragg Condition of Diffraction

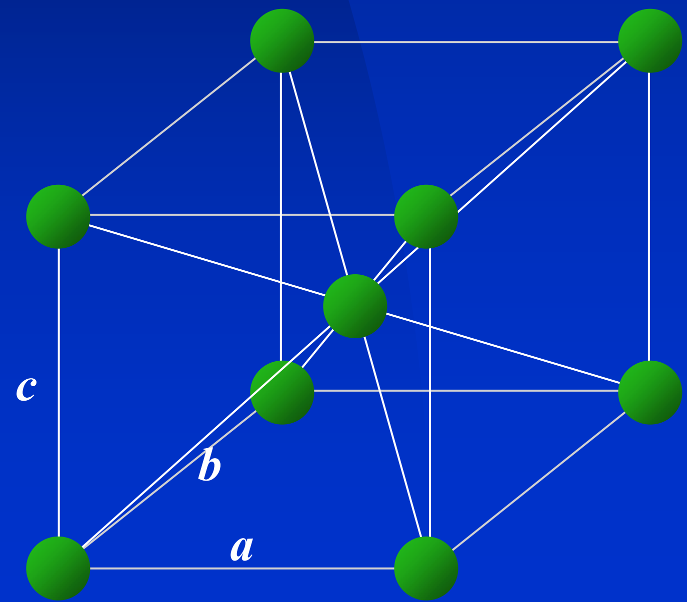
$$\Rightarrow \lambda = 2 d_{hkl} \sin\theta$$

Forbidden Reflection (1/2)

K=***g*** is a necessary condition for observing diffraction, but not a sufficient condition...

If F(***K***)=0, then $I_{crystal} = 0$ even when ***K***=***g***.

Example 1, Body Center Cubic Lattice (bcc)



$$F(\boldsymbol{g}) = f^a(\boldsymbol{g})\left\{\exp(i\boldsymbol{g}\cdot\boldsymbol{0}) + \exp\left(i\boldsymbol{g}\cdot\left(\frac{1}{2}\boldsymbol{a}+\frac{1}{2}\boldsymbol{b}+\frac{1}{2}\boldsymbol{c}\right)\right)\right\}$$

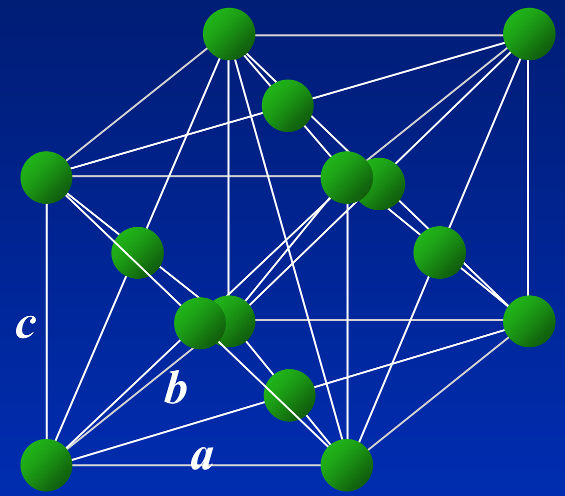
$$= f^a(\boldsymbol{g})\left\{1+\exp\left(i\pi(h+k+l)\right)\right\}$$

$$h+k+l = odd \Rightarrow F(\boldsymbol{g}) = 0 \text{ (Forbidden Reflection)}$$

$$h+k+l = even \Rightarrow F(\boldsymbol{g}) = 2f^a(\boldsymbol{g})$$

Forbidden Reflection (2/2)

Example 2, Face Center Cubic Lattice (fcc)



$$F(\mathbf{g}) = f^a(\mathbf{g})\left\{1 + \exp\left(i\pi(h+k)\right) + \exp\left(i\pi(k+l)\right) + \exp\left(i\pi(l+h)\right)\right\}$$

$$h,k,l \ \text{ all odd or all even} \Rightarrow F(\mathbf{g}) = 4f^a(\mathbf{g})$$

$$\text{Otherwise} \Rightarrow F(\mathbf{g}) = 0 \ (\text{Forbidden Reflection})$$

Special Topics

What we can measure in diffraction/scattering experiment
= Intensity

All phase information is lost!

Non-Crystalline Charge Distribution:

$$I = \frac{r_o^2 \sin^2 \alpha}{r^2} I_o \left| \iiint \rho(\boldsymbol{x}) \exp[-i\boldsymbol{K} \cdot \boldsymbol{x}] d^3\boldsymbol{x} \right|^2 = \sqrt{\frac{\varepsilon_o}{\mu_o}} \boldsymbol{E} \cdot \boldsymbol{E}^*$$

$\boldsymbol{E}^*$: complex conjugate of $\boldsymbol{E}$

If

$$E = \frac{r_o \sin \alpha}{r} \sqrt{I_o \sqrt{\frac{\mu_o}{\varepsilon_o}}} \iiint \rho(\boldsymbol{x}) \exp[-i\boldsymbol{K} \cdot \boldsymbol{x}] d^3\boldsymbol{x}$$

$$= \sqrt{I \sqrt{\frac{\mu_o}{\varepsilon_o}}} \exp(i\varphi)$$

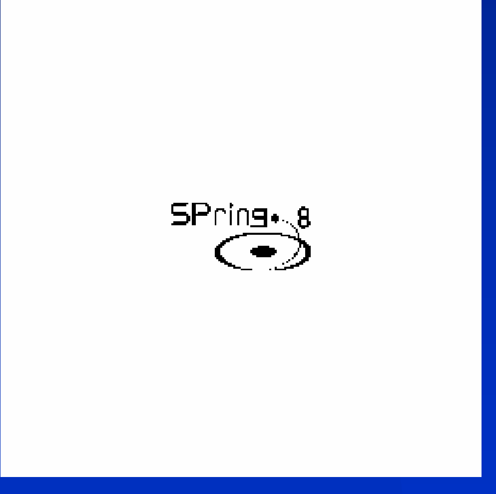
is obtained, we can calculate $\rho(\boldsymbol{x})$ by Fourier inversion.

Iterative Phase Retrieval
(Jianwei Miao & David Sayre)

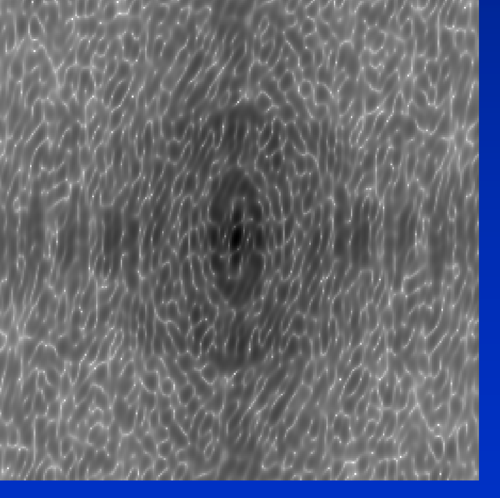
X-ray intensity data: Phase Information is Lost!

Scattered pattern in Far Field with Coherent Illumination, Phase can be retrieved.

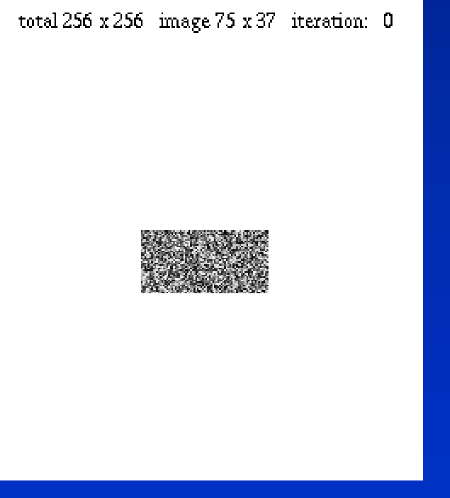
Phase Retrieval → Iterative Algorism developed by Gerchberg & Saxon, followed by the improvement by Fienup (Opt. Lett. 3 (1978) 27.)



Real Space Image



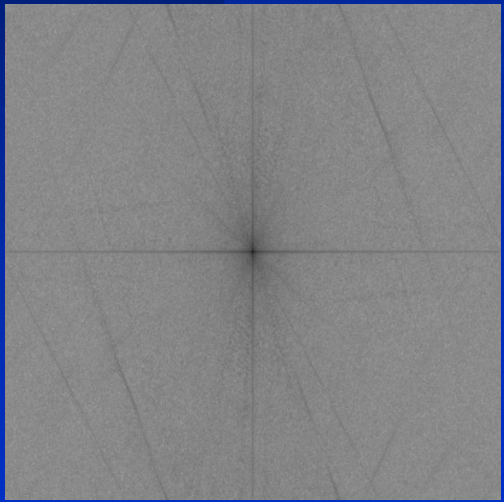
Scattered Intensity



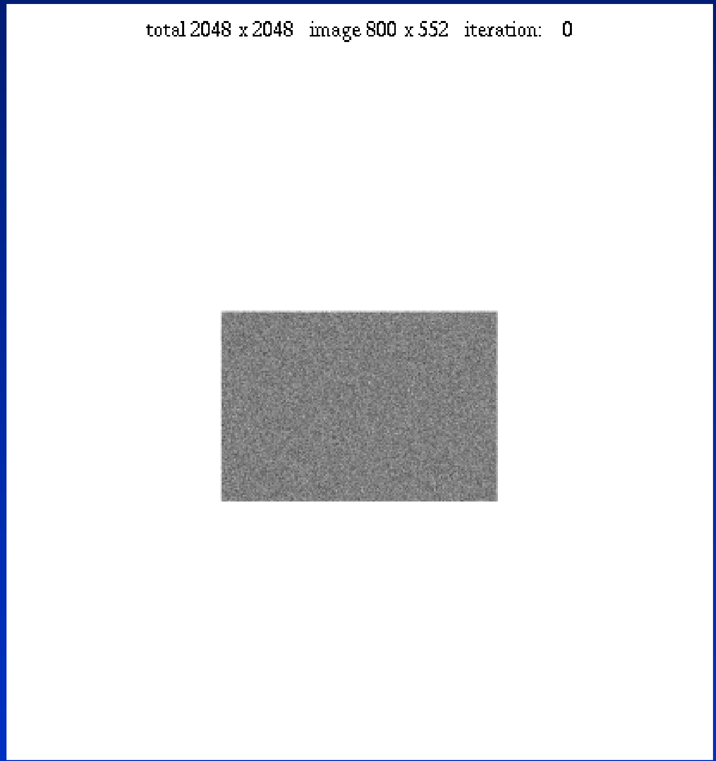
Phase Retrieval

Reconstruction of Complex Real Space Images

Real Space



Scattered Intensity



Phase Retrieval

Original Image

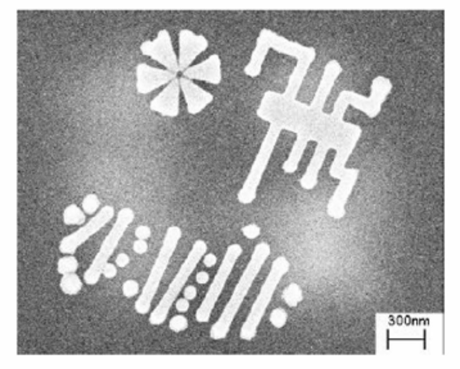
Reconstructed Image

After 5000 iteration

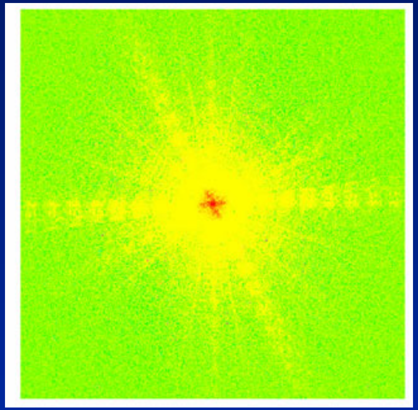
3D Diffraction Microscopy

Miao *et al. PRL* (2002)

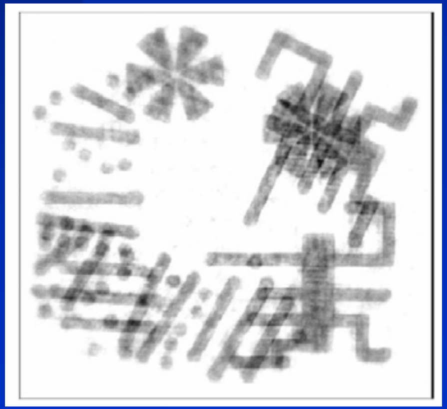
Two Layer Ni Pattern



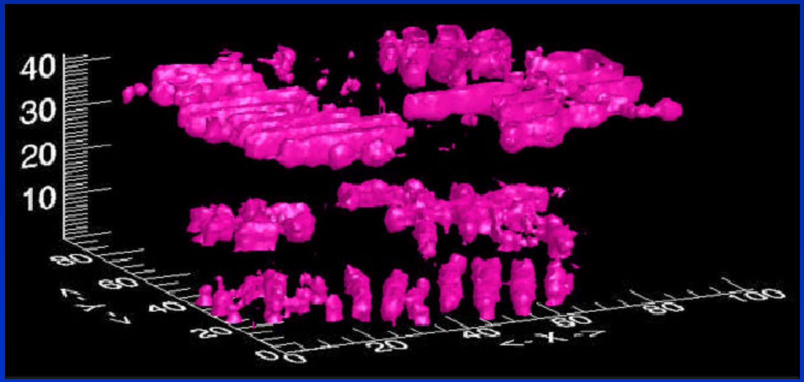
SEM image of Ni pattern on SiN



Coherent Scattering Pattern



2D Reconstructed Image
(<10 nm resolution)



3D Reconstructed Image (~50 nm resolution)