Effect of Partial Coherence on the Reconstruction of Coherent X-ray Diffraction Patterns

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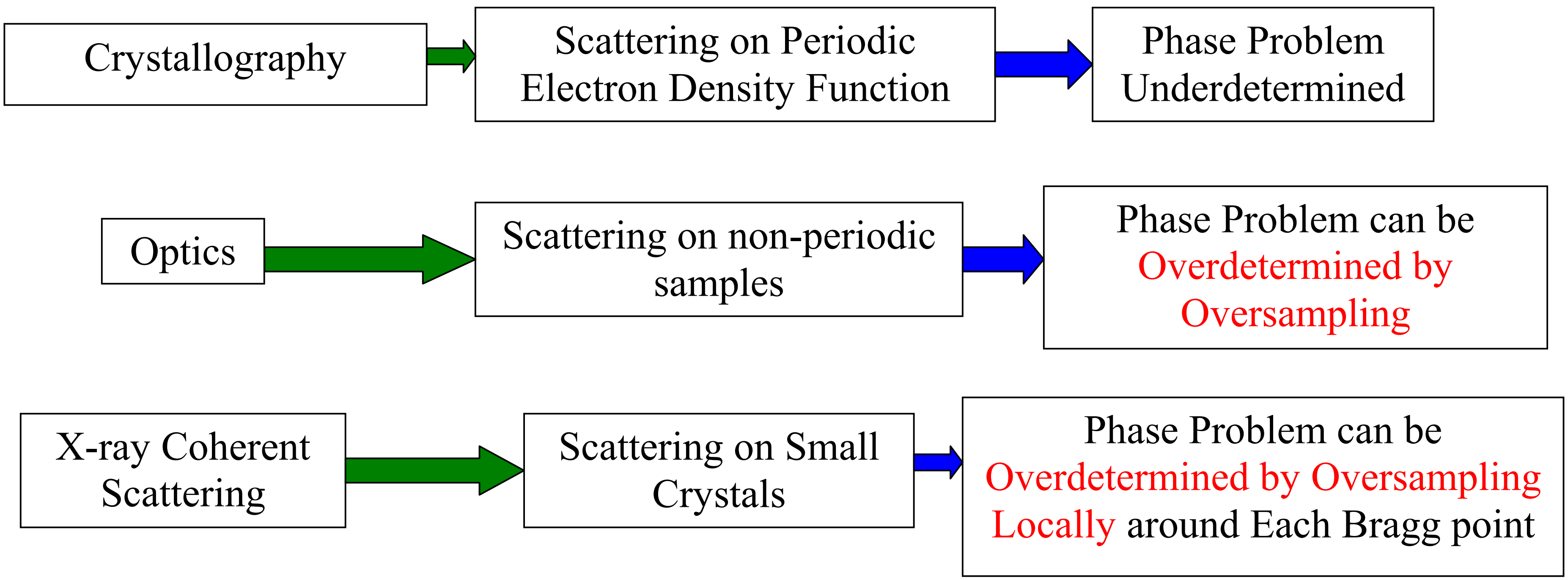
Phase Problem in Optics and Crystallography



Theory of Partially Coherent Radiation

Books:

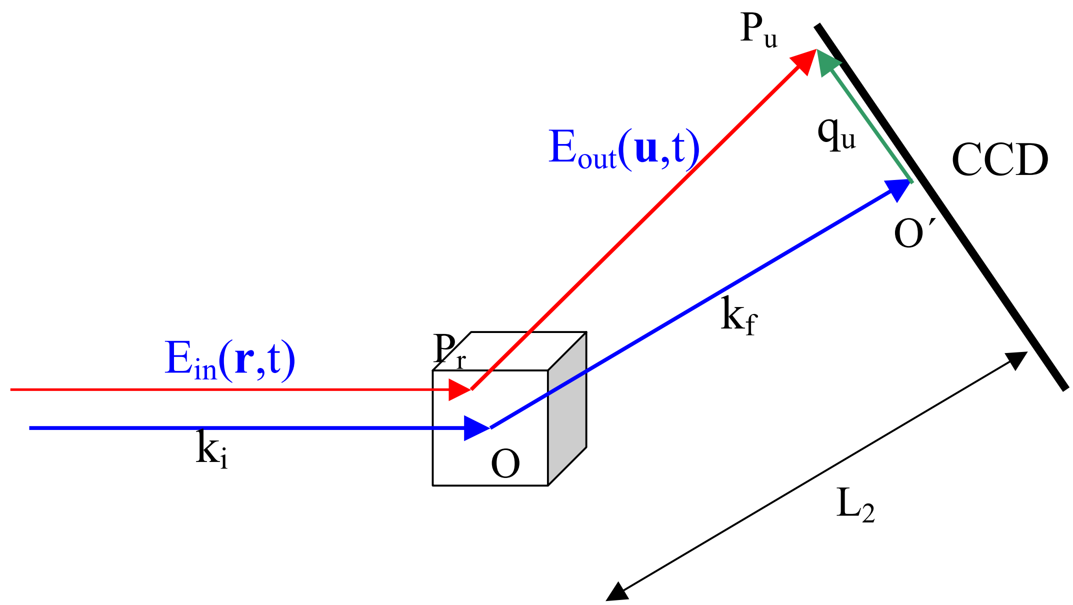
M. Born and E. Wolf, *Principles of Optics*;

J.W. Goodman, *Statistical Optics;*

L. Mandel and E. Wolf, *Optical Coherence and Quantum Optics*

We applied this general theory to a special problem of scattering partially coherent x-ray radiation on small crystal particles

Scattering of Partially Coherent Radiation



The incoming beam:

$$E_{in}(\mathbf{r},t)=A_{in}(\mathbf{r},t)\ \exp[ik_i r-i\omega t]$$

The scattered beam (Huygence-Fresnel principle):

$$E_{out}(\mathbf{u},t)=\int d\mathbf{r}\ \rho(\mathbf{r})\ A_{in}(\mathbf{r},t-\tau_r)\ (1/P_rP_u)\ \exp[i\mathbf{k_i r}-i\omega(t-\tau_r)]$$

$\tau_r = P_rP_u/c$ – time delay for radiation propagation between P_r and P_u

For $L_2 \gg D$ (paraxial approximation): $P_rP_u \approx L_2 - \mathbf{n_f r} + (\mathbf{u}-\mathbf{r})^2/(2\,L_2)$

$$E_{out}(\mathbf{u},t)=A(u,t)\ [\exp(ikL_2)/L_2]\ \exp(-i\omega t)$$

Scattering amplitude

$$A(\mathbf{u},t) = \int d\mathbf{r}\rho(\mathbf{r})A_{in}(\mathbf{r},t-\tau_r)P_{L_2}(\mathbf{u}-\mathbf{r})e^{-i\mathbf{q}\cdot\mathbf{r}},\ \mathbf{q} = \mathbf{k}_f-\mathbf{k}_i$$

Here

$$P_{L_2}(\mathbf{u}-\mathbf{r}) = \frac{1}{i\lambda L_2}e^{i(k/2L_2)(\mathbf{u}-\mathbf{r})^2}.$$

is the propagator (Green's function).

In the **far–field** (Fraunhofer) limit

$$(E_\gamma \simeq 8 \text{ keV},\ L_2 \simeq 3 \text{ m})$$

$$kD^2/(2L_2) << 1 \Longrightarrow D << 10\mu m.$$

neglecting r^2 in $P_{L_2}(\mathbf{u}-\mathbf{r})$

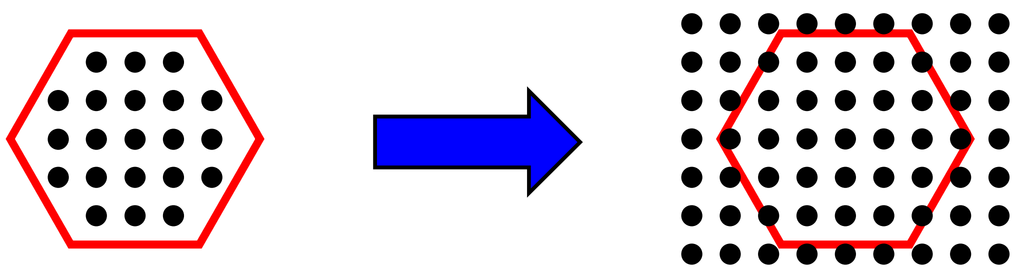
Scattering amplitude

$$A(\mathbf{q}',t) = \int d\mathbf{r}\rho(\mathbf{r})A_{in}(\mathbf{r},t-\tau_r)e^{-i\mathbf{q}'\cdot\mathbf{r}},\ \mathbf{q}' = \mathbf{q}+\mathbf{q}_u$$

where $\mathbf{q}_u = (k/L_2)\mathbf{u}$.

Considerations:

1. The scattering particle is a crystalline sample with a periodic electron density function
2. The amplitude $A_{in}(\mathbf{r},t-\tau_r)$ is a slow varying function on the size of the unit cell



$$\rho(\mathbf{r}) = \rho_\infty(\mathbf{r})\cdot s(\mathbf{r})$$

From theory of Fourier transformation (convolution theorem)

$$A(\mathbf{q}',t) = [F(q)/v] \sum_n A_n(\mathbf{q}'-\mathbf{h}_n,t),$$

where $\mathbf{h}_n$ - reciprocal lattice vector and

$$A_n(\mathbf{q},t) = \int d\mathbf{r}\ s(\mathbf{r})\ A_{in}(\mathbf{r}, t-\tau_r)\ e^{-i\mathbf{qr}}$$

$s(\mathbf{r})$ - shape function of crystal

Coherent X-ray beam

$$A_{in}(\mathbf{r}, \mathbf{t} - \tau_{\mathbf{r}}) \longrightarrow const$$

Scattering amplitude

$$A(\mathbf{q}) = \frac{\mathbf{F(q)}}{\mathbf{v}} \sum_{\mathbf{n}} \mathbf{S}(\mathbf{q} - \mathbf{h_n}),$$

where $S(\mathbf{q})$ Fourier integral of shape function:

$$S(\mathbf{q}) = \int \mathbf{s(r)e^{-iq \cdot r}dr}$$

Scattering intensity by crystal of finite dimensions $(D >> a)$ – periodic function

$$I(\mathbf{q}) = |\mathbf{A(q)}|^{\mathbf{2}} = \frac{|\mathbf{F(q)}|^{\mathbf{2}}}{\mathbf{v^2}} \sum_{\mathbf{n}} |\mathbf{S}(\mathbf{q} - \mathbf{h_n})|^{\mathbf{2}}.$$

In the vicinity of the reciprocal point

$$\mathbf{h_n} = \mathbf{h},\ \mathbf{q} \simeq \mathbf{h}$$

$$I(\mathbf{Q}) = \frac{|\mathbf{F(h)}|^{\mathbf{2}}}{\mathbf{v^2}} |\mathbf{S(Q)}|^{\mathbf{2}},\ \ \mathbf{Q} = \mathbf{q} - \mathbf{h}$$

General properties:

1. For arbitrary form of crystal shape intensity distribution is periodic function of **q**

2. For unstrained crystal:

- Maximum value of intensity distribution:

$$I_{max} = |F(\mathbf{h}_n)|^2\, V^2 / v^2.$$

- Intensity distribution is locally centrosymmetric around every $\mathbf{h}_n$

$$s(-\mathbf{q}) = s^*(\mathbf{q}) \rightarrow I(-\mathbf{Q}) = I(\mathbf{Q})$$

- It has the same shape for every reciprocal lattice point $\mathbf{h}_n$

Partially coherent x-ray beam

Intensity at the position $\mathbf{u}$ of the detector plane at Bragg point $\mathbf{h_n} = \mathbf{h}$

$$I(\mathbf{Q}) = \langle \mathbf{A}(\mathbf{Q},\mathbf{t})\mathbf{A}^*(\mathbf{Q},\mathbf{t})\rangle_{\mathbf{T}} = \frac{|\mathbf{F}(\mathbf{h})|^{\mathbf{2}}}{\mathbf{v}^{\mathbf{2}}}\left|\mathbf{A_h}(\mathbf{Q},\mathbf{t})\right|^{\mathbf{2}}$$

$$I(\mathbf{Q}) = \frac{|\mathbf{F}(\mathbf{h})|^{\mathbf{2}}}{\mathbf{v}^{\mathbf{2}}}\mathbf{drdr's}(\mathbf{r})\mathbf{s}(\mathbf{r}')\boldsymbol{\Gamma}_{\mathbf{in}}(\mathbf{r},\mathbf{r}',\boldsymbol{\Delta}\tau)\mathbf{e}^{-\mathbf{iQ}\cdot(\mathbf{r}-\mathbf{r}')}$$

$$\mathbf{Q} = \mathbf{q}' - \mathbf{h} = \mathbf{q_u} + \mathbf{q} - \mathbf{h},$$

where $\Delta\tau = (P_r P_u - P_{r'} P_u)/c$ is a time delay.

Mutual coherence function

$$\Gamma_{in}(\mathbf{r},\mathbf{r}',\tau) = \langle \mathbf{A_{in}}(\mathbf{r},\mathbf{t})\mathbf{A^*_{in}}(\mathbf{r}',\mathbf{t}+\tau)\rangle_{\mathbf{T}}\,.$$

For the **cross-spectral pure** light

$$\Gamma_{in}(\mathbf{r},\mathbf{r}',\tau) = \sqrt{\mathbf{I}(\mathbf{r})}\sqrt{\mathbf{I}(\mathbf{r}')}\gamma_{\mathbf{in}}(\mathbf{r},\mathbf{r}')\mathbf{F}(\tau).$$

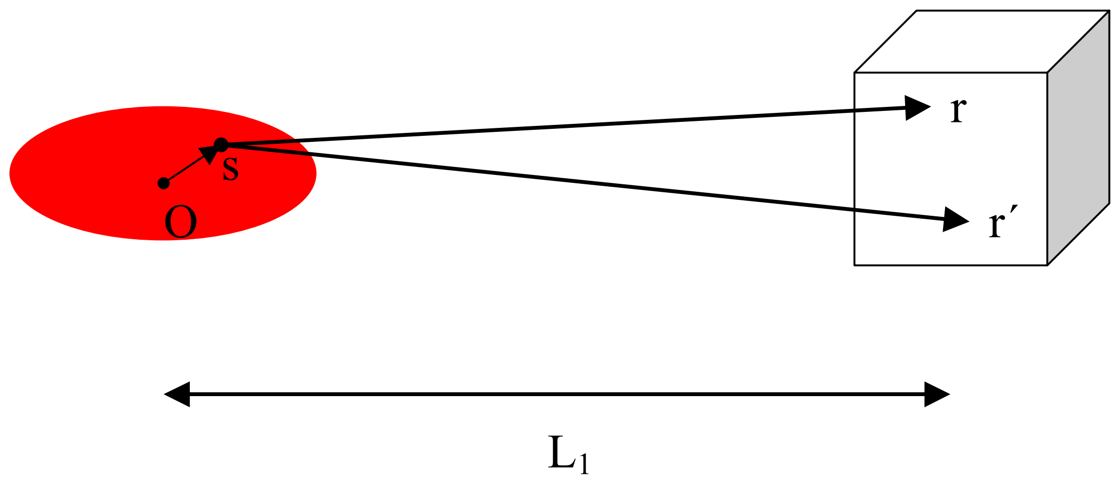
Here

$$I(\mathbf{r}) =< |\mathbf{A_{in}}(\mathbf{r},\mathbf{t})|^{\mathbf{2}} >_{\mathbf{T}};\ \mathbf{I}(\mathbf{r}') =< |\mathbf{A_{in}}(\mathbf{r}',\mathbf{t})|^{\mathbf{2}} >_{\mathbf{T}}$$

$\gamma_{in}(\mathbf{r},\mathbf{r}')$ – complex degree of coherence, $F(\tau)$ – time autocorrelation function.

Considerations:

1. Incident radiation is coming from a planar incoherent source on the distance L_1 from the sample
2. Gaussian distribution of intensity of the source
3. $L_1 >> S$ and D; paraxial approximation



Complex degree of coherence (van Cittert-Zernike theorem):

$$\gamma_{in}(\mathbf{r}-\mathbf{r}') = e^{i\psi} \int d\mathbf{s}\, I(\mathbf{s}) \exp[-i\,(k/L_1)\,(\mathbf{r}-\mathbf{r}')\cdot\mathbf{s}] / \int d\mathbf{s}\, I(\mathbf{s}),$$

$$\psi = (k/2L_1)\,(r^2 - r'^2),$$

where $I(\mathbf{s})$ – intensity distribution of the incoherent source

Synchrotron source

Gaussian intensity distribution :

$$I(s_x, s_y) = \frac{I_0}{2\pi\sigma_x\sigma_y} e^{-\frac{1}{2}(s_x^2/\sigma_x^2 - s_y^2/\sigma_y^2)},$$

$\sigma_{x,y}$ – halfwidths of intensity distribution

Complex degree of coherence (far-field, $\psi \ll 1$)

$$\gamma_{in}(\mathbf{r}_\perp - \mathbf{r}'_\perp) = \exp\left(-\frac{(\mathbf{x}-\mathbf{x}')^2}{2\xi_x^2} - \frac{(\mathbf{y}-\mathbf{y}')^2}{2\xi_y^2}\right).$$

$\mathbf{r}_\perp$ and $\mathbf{r}'_\perp$ – projections of $\mathbf{r}$ and $\mathbf{r}'$ across the beam propagation direction.

Transverse coherence length

$$\xi_{x,y} = \frac{L_1}{k\sigma_{x,y}}$$

For the parameters of APS source

$$E_\gamma = 8 \text{ keV},\ L_1 = 40 \text{ m}$$

$$\sigma_x \simeq 350\mu m \Rightarrow \quad \xi_x \simeq 3\mu m$$
$$\sigma_y \simeq 50\mu m \Rightarrow \quad \xi_y \simeq 20\mu m$$

Time autocorrelation function

Exponential form (Lorentzian power spectral density)

$$F(\tau) = F_0 \exp(-\tau/\tau_{||})$$

Longitudinal correlation length

$$\xi_{||} = c\tau_{||},\ \xi_{||} = \frac{2}{\pi}\left(\frac{\lambda^2}{\Delta\lambda}\right)$$

For a Si (111) double-crystal monochromator

$$\Delta\lambda/\lambda \simeq 3 \times 10^{-4}\ ,\ \lambda \simeq 1.5\ \text{Å}$$

$$\xi_{||} \simeq 0.32\mu m.$$

In the **far-field limit**

$$F(\Delta\tau) = F(|\mathbf{r}_{||} - \mathbf{r}'_{||}|) = \mathbf{F_0} \exp(-|\mathbf{r}_{||} - \mathbf{r}'_{||}|/\xi_{||}),$$

$\mathbf{r}_{||}$ and $\mathbf{r}'_{||}$ – components of $\mathbf{r}$ and $\mathbf{r}'$ along the beam.

Scattered intensity

$$I(\mathbf{Q}) = \frac{|\mathbf{F(h)}|^2}{\mathbf{v^2}} \int \mathbf{dr}\varphi_{\mathbf{11}}(\mathbf{r})\gamma_{\mathbf{in}}(\mathbf{r}_\perp)\mathbf{F}(|\mathbf{r}_\parallel|)\mathbf{e}^{-\mathbf{iQ\cdot r}}$$

where

$$\varphi_{11}(\mathbf{r}) = \int \mathbf{dr's(r')s(r'+r)}$$

$\varphi_{11}(\mathbf{r})$ – **autocorrelation function** of the shape function $s(\mathbf{r})$.

Coherent limit

$$\xi_\perp, \xi_\parallel \to \infty \Longrightarrow \gamma_{in}(\mathbf{r}_\perp), \mathbf{F}(|\mathbf{r}_\parallel|) \to \mathbf{1}$$

$$I_{coh}(\mathbf{Q}) = \frac{|\mathbf{F(h)}|^2}{\mathbf{v^2}} \int \mathbf{dr}\varphi_{\mathbf{11}}(\mathbf{r})\mathbf{e}^{-\mathbf{iQ\cdot r}} = |\mathbf{A(Q)}|^{\mathbf{2}},$$

where $A(\mathbf{Q})$ is a kinematically scattered amplitude

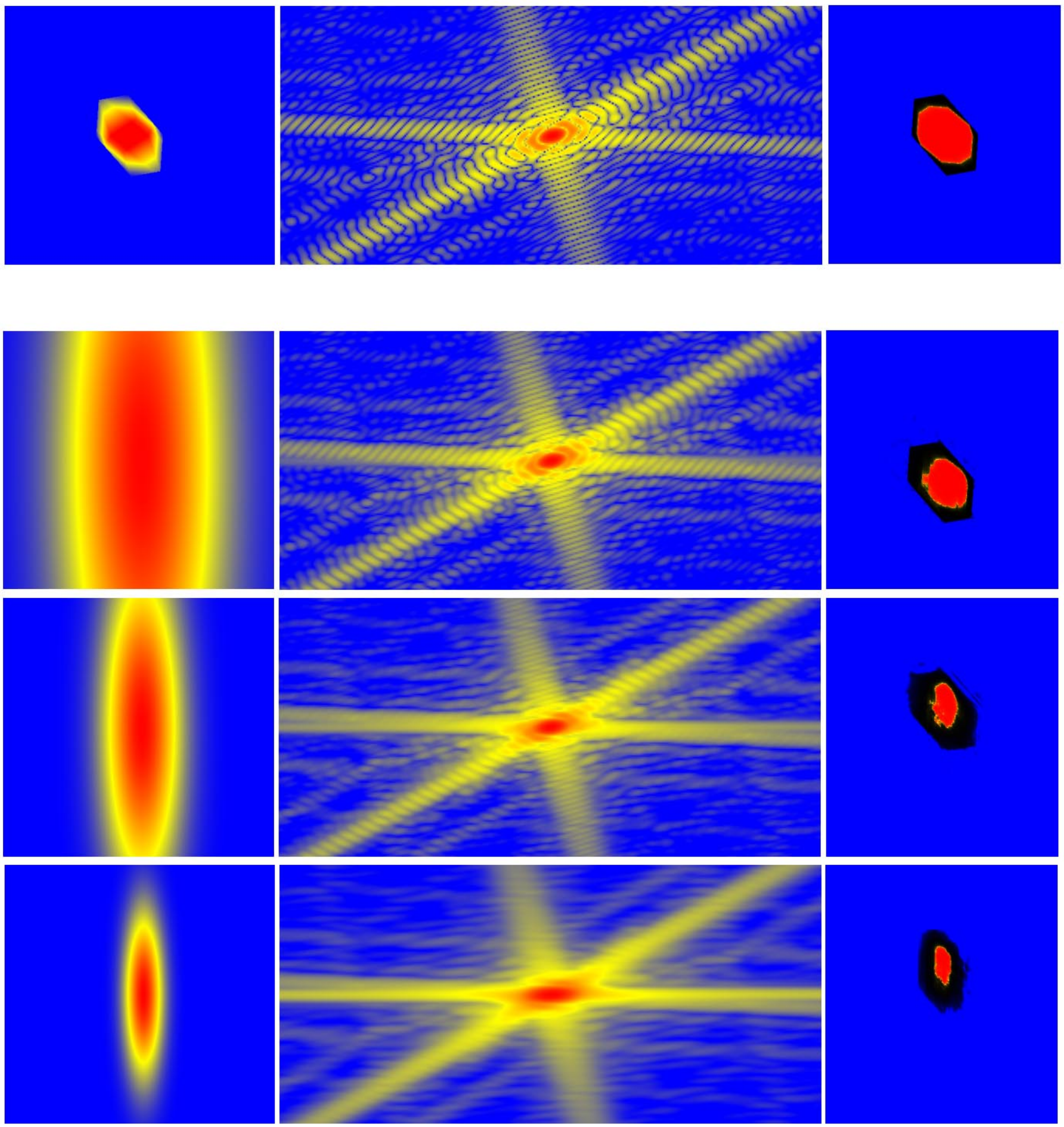
$$A(\mathbf{Q}) = (\mathbf{F(h)/v}) \int \mathbf{drs(r)e}^{-\mathbf{iQ\cdot r}}$$

From convolution theorem

$$I(\mathbf{Q}) = \frac{\mathbf{1}}{(\mathbf{2}\pi)^{\mathbf{3}}} \int \mathbf{dQ'I_{coh}(Q')}\widetilde{\mathbf{\Gamma}}(\mathbf{Q}-\mathbf{Q'})$$

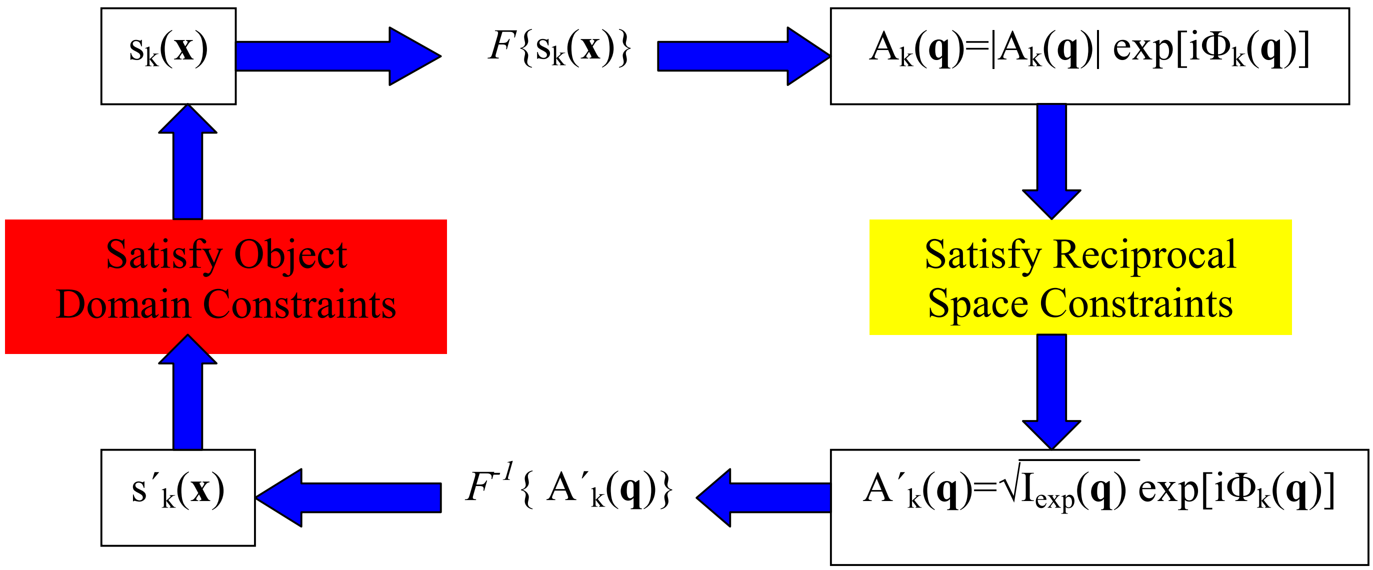
where

$$\tilde{\Gamma}(\mathbf{Q}) = \int \mathbf{dr}\gamma_{\mathbf{in}}(\mathbf{r}_\perp)\mathbf{F}(|\mathbf{r}_\parallel|)\mathbf{e}^{-\mathbf{iQr}}$$



Iterative Methods for Phase Retrieval

Gerchberg – Saxton – Fienup algorithm



Error-reduction algorithm (ER)

$$s_{k+1}(\mathbf{x})=\begin{cases} s'_k(\mathbf{x}), \ \mathbf{x} \text{ in } S_{ob} \\ 0, \ \mathbf{x} \text{ not in } S_{ob} \end{cases}$$

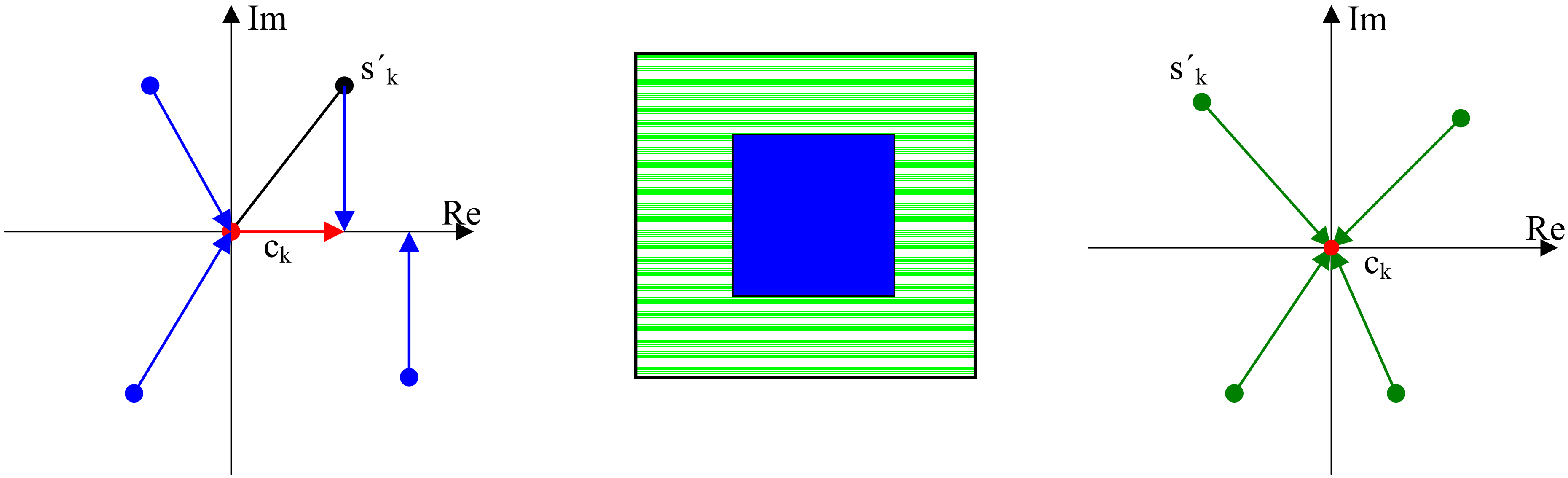
Hibrid input-output algorithm (HIO)

(Fienup-Millane)

$$s_{k+1}(\mathbf{x})=\begin{cases} s'_k(\mathbf{x}), \ |c_k(\mathbf{x})- s'_k(\mathbf{x})|<\varepsilon \\ s_k(\mathbf{x}) + \beta[c_k(\mathbf{x})- s'_k(\mathbf{x})], \ |c_k(\mathbf{x})- s'_k(\mathbf{x})|>\varepsilon \end{cases}$$

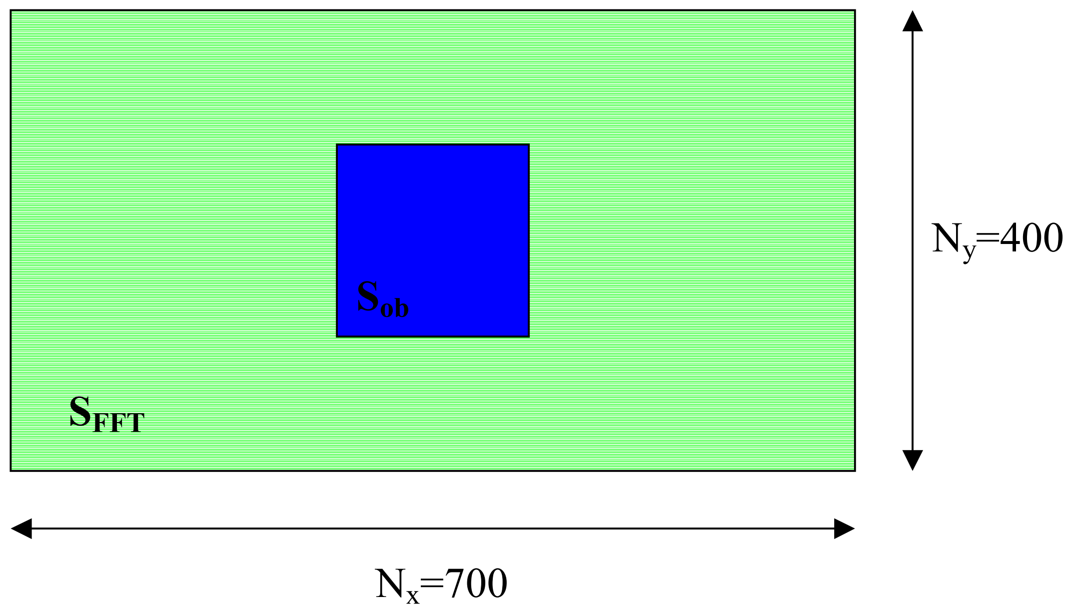
Best convergence with: $\beta = 0.8 \div 0.9$ and $\varepsilon = 0.01$

Constrain for Reconstruction of Small Crystal Shape



$$c_k(\mathbf{x}) = \begin{cases} \mathrm{Re}[c_k] = \mathrm{Re}[s'_k(\mathbf{x})];\ \mathrm{Im}[c_k] = 0, \text{ If } \mathbf{x} \text{ in } S_{ob}, \text{ and } \mathrm{Re}[s'_k(\mathbf{x})] > 0 \\ \mathrm{Re}[c_k] = 0;\ \mathrm{Im}[c_k] = 0, \text{ If } \mathbf{x} \text{ not in } S_{ob}, \text{ or } \mathrm{Re}[s'_k(\mathbf{x})] < 0 \end{cases}$$

Oversampling



Oversampling condition:

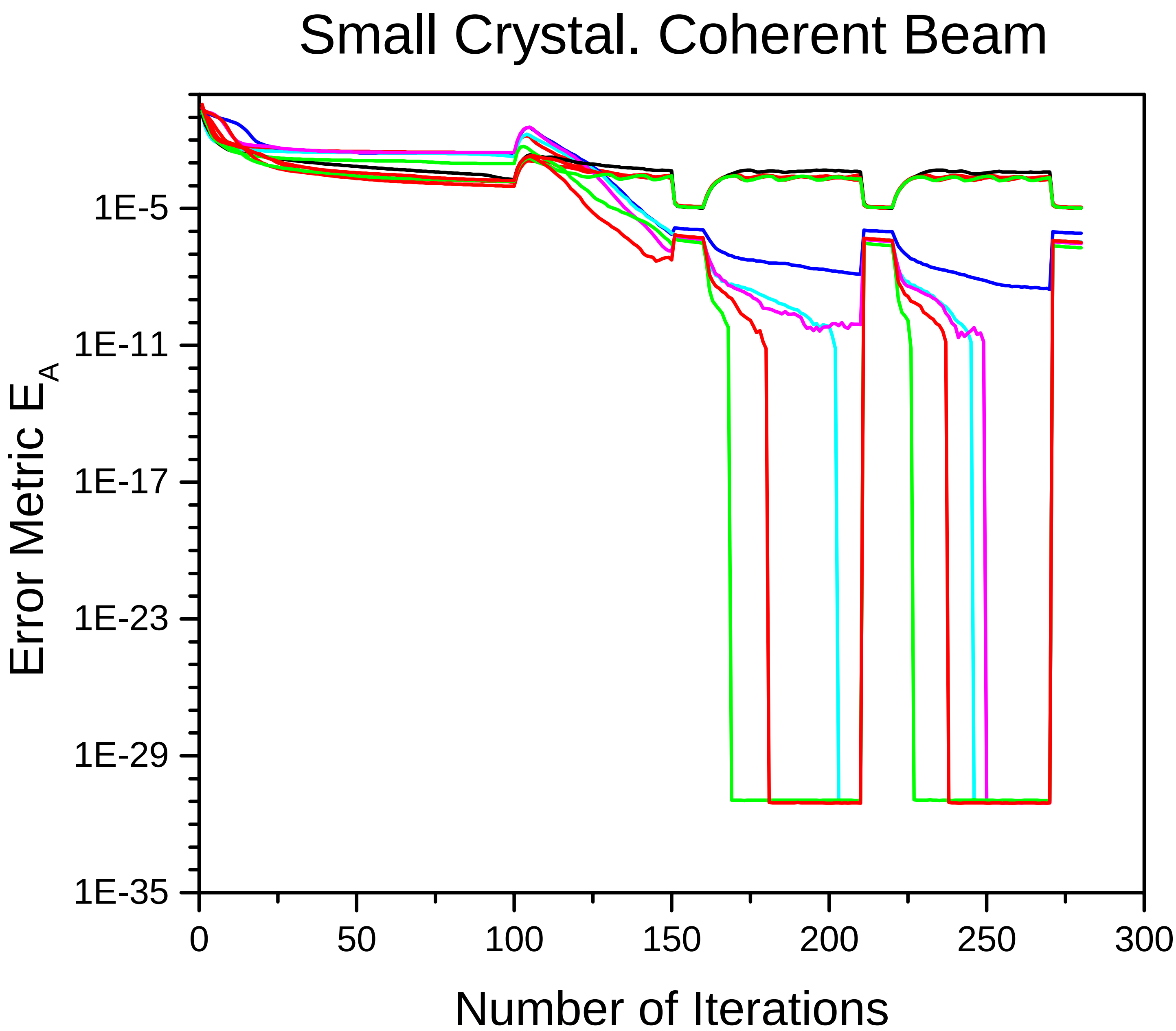
$$\sigma = S_{FFT}/S_{object} > 2$$

In our calculations:

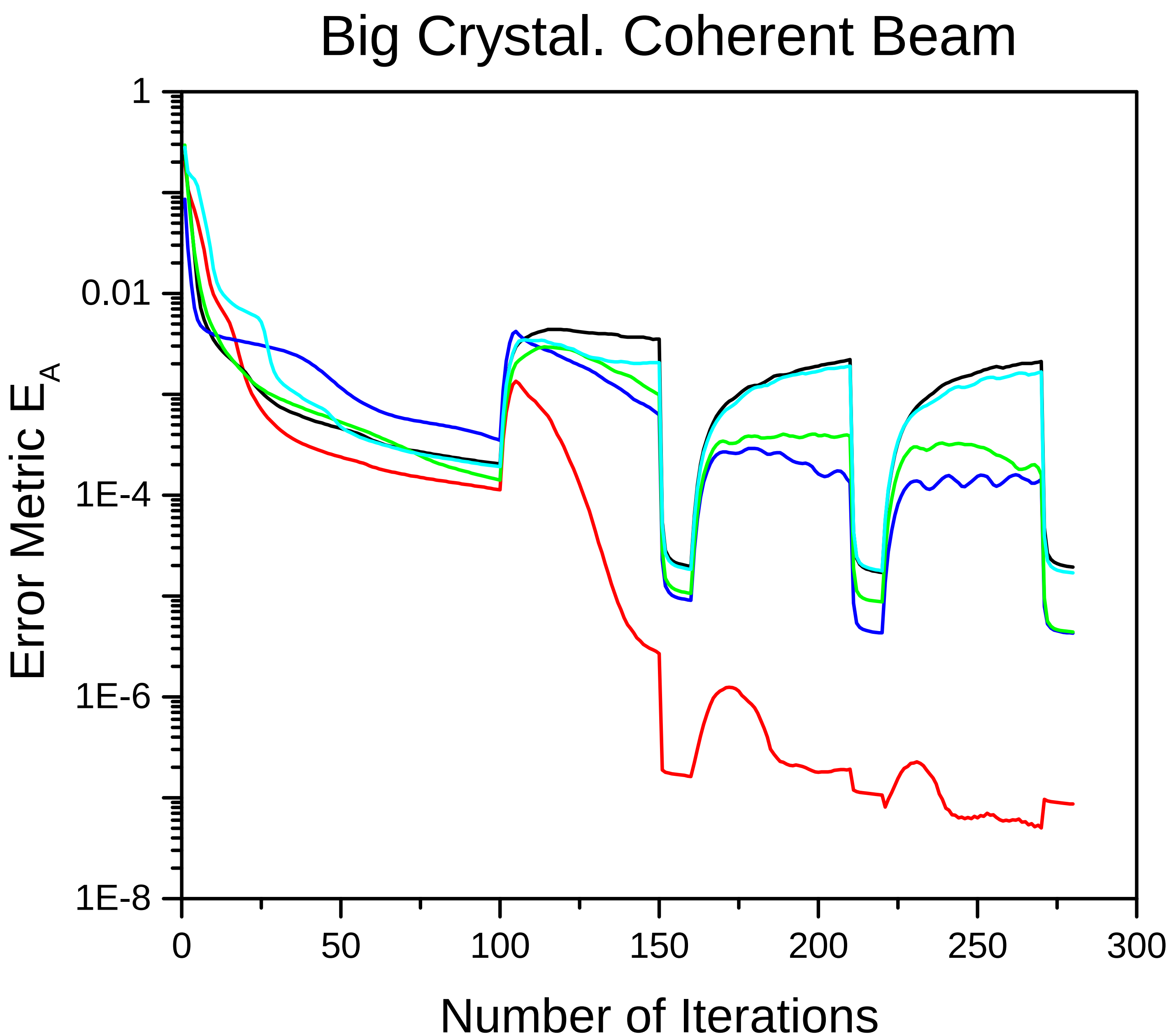
$$\sigma = S_{FFT}/S_{object} > 7$$

Error Metric

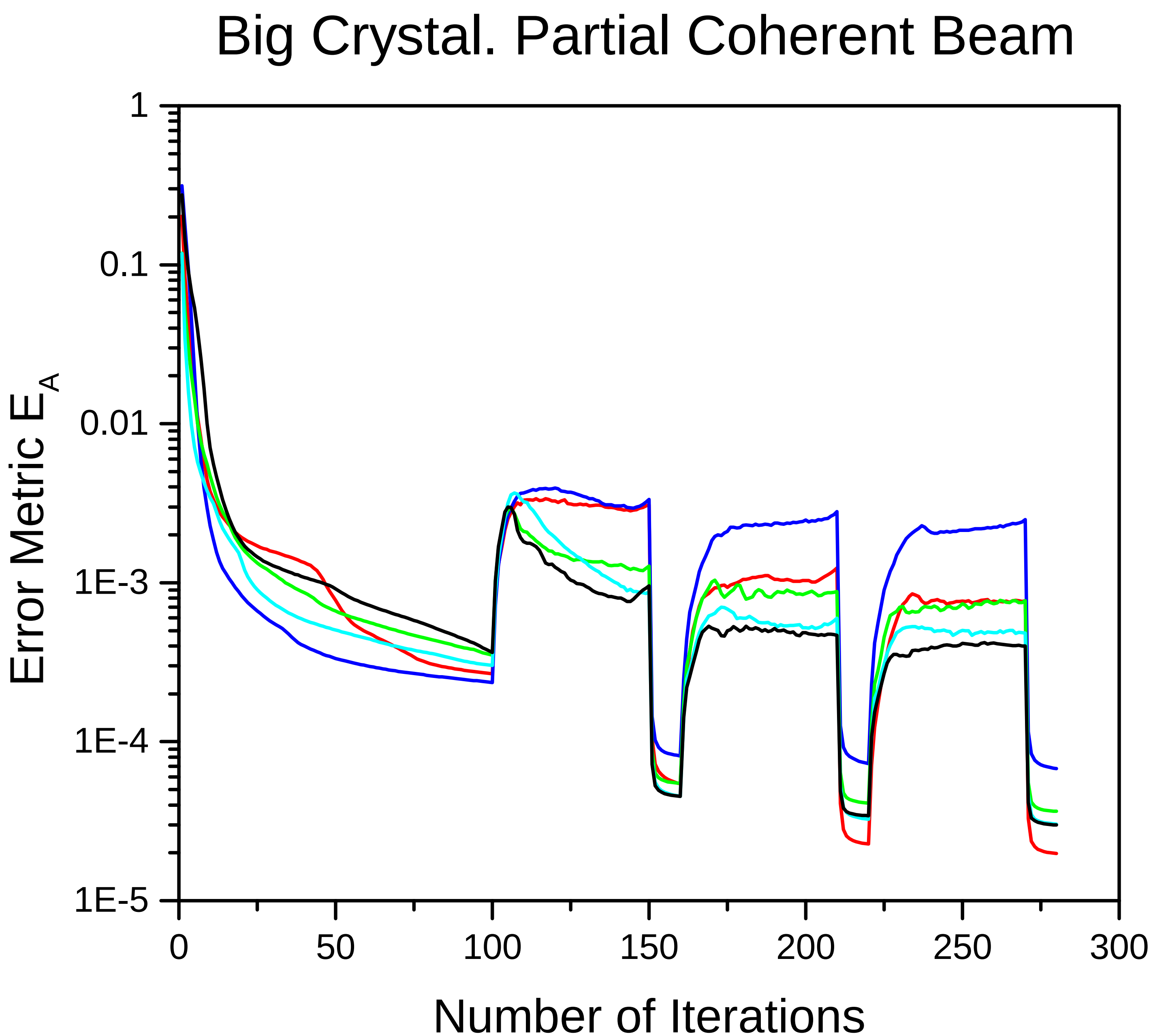
$$E_A = \frac{\Sigma_{qx,qy} \left[\ |A_k(q_x,q_y)| - \sqrt{I_{exp}(q_x,q_y)}\ \right]}{\Sigma_{qx,qy} \left[\ I_{exp}(q_x,q_y)\ \right]}$$



Small Crystal. Coherent Beam
Error Metric E_A
1E-5
1E-11
1E-17
1E-23
1E-29
1E-35
0
50
100
150
200
250
300
Number of Iterations



Big Crystal. Coherent Beam
Error Metric E_A
Number of Iterations
1
0.01
1E-4
1E-6
1E-8
0
50
100
150
200
250
300


Big Crystal. Partial Coherent Beam
Error Metric E_A
1
0.1
0.01
1E-3
1E-4
1E-5
0
50
100
150
200
250
300
Number of Iterations

Reconstruction of Big Crystal. Partial Coherent Beam.

ζ_x	ζ_y	Σ_x (BG)	Σ_y (BG)	Σ_x (BS)	Σ_y (BS)
∞	∞	107.5	127.5	90	92.5
91	367	107.5	127.5	72.5	75
45	183	112.5	127.5	42.5	72.5
22	91	75	125	25	60
11	45	45	115	15	42.5

$\zeta_{x,y}$ – values of transverse correlation length (pixels)

$\Sigma_{x,y}$ (BG) – size of the background level (pixels)

$\Sigma_{x,y}$ (BS) – size of the bright central spot (pixels)

How we can explain this effect

Intensity of scattered radiation

$$I(q_x, q_y) = \frac{|F(\mathbf{h})|^{\mathbf{2}}}{v^2} \int dxdy \varphi_{11}^z(x, y) \gamma_{in}(x, y) e^{-iq_x x - iq_y y}$$

where $\varphi_{11}^z(x, y)$ is projection of 3D autocorrelation function:

$$\varphi_{11}^z(x, y) = \int dz \varphi_{11}(x, y, z)$$

In the **limit of small correlation lengths**:

$$\xi_{x,y} << D$$

we obtain for intensity:

$$I(q_x, q_y) \sim \frac{|F(\mathbf{h})|^{\mathbf{2}}}{v^2} \varphi_{11}^z(0) \int dxdy \gamma_{in}(x, y) e^{-iq_x x - iq_y y}$$

Inversion of this expression gives **complex degree of coherence**:

$$\gamma_{in}(x, y)$$

with the **typical size of area** with maximum intensity

$$S \approx \xi_x \times \xi_y$$

Summary

- Theory of partial coherent radiation was applied to scattering on small crystals
- Phase retrieval of diffraction patterns from small crystals was discussed
- Effects introduced by partially coherent radiation on the reconstructed images were investigated