Local Ordering of Fluids and Other Nanobeam Applications & Waveguides

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- Confining a fluid; why, how?
- Diffraction from fluid-filled nanocavity arrays
- Phase retrieval ⟹ fluid‘s density profile
- Nanofocus for diffraction from *single* cavity
- Outlook

ERL X-Ray Science Workshop #6, Cornell, USA, 23-24 June 2006

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Confined fluids

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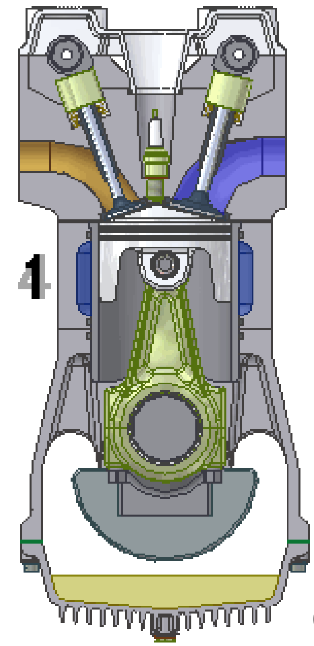
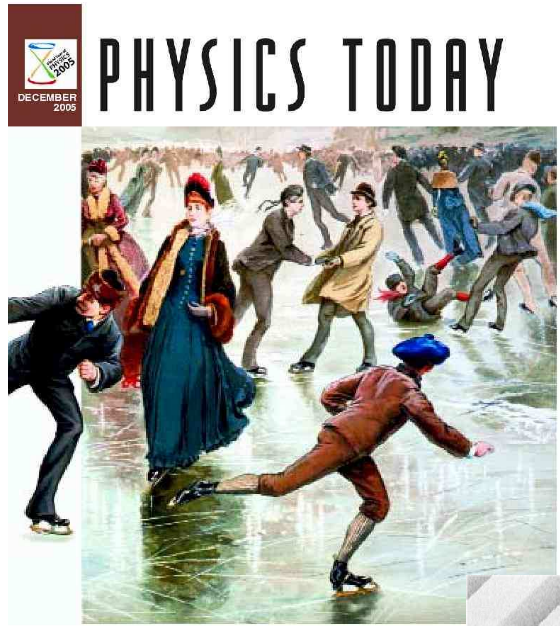
Focusing properties of waveguides and FZPs

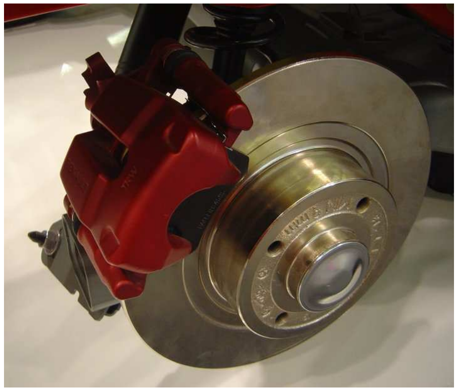
Christoph Bergemann, *Univ. Cambrige*
Franz Pfeiffer

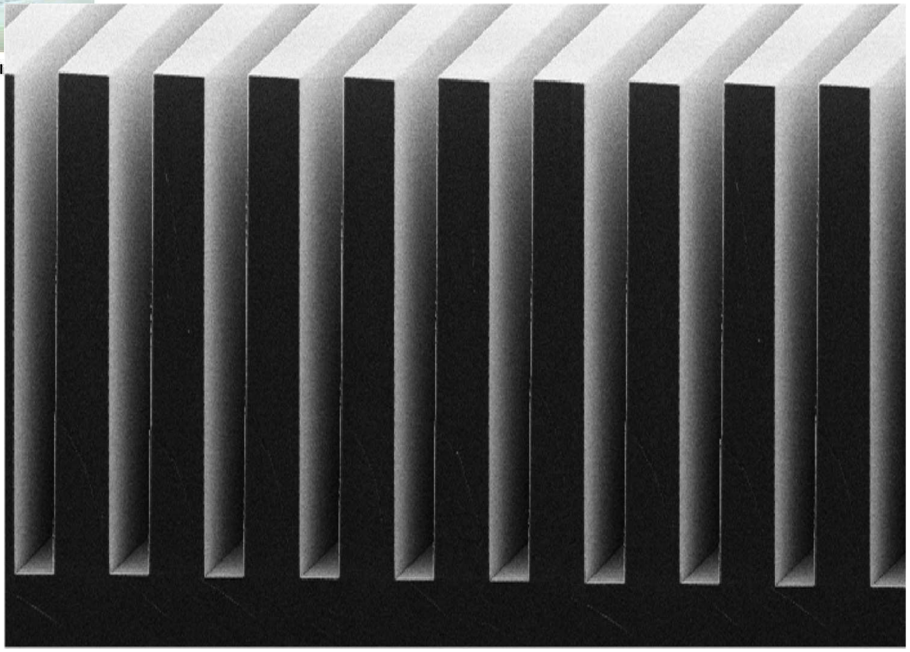
Confined fluids: Motivation



(from Wikipedia)

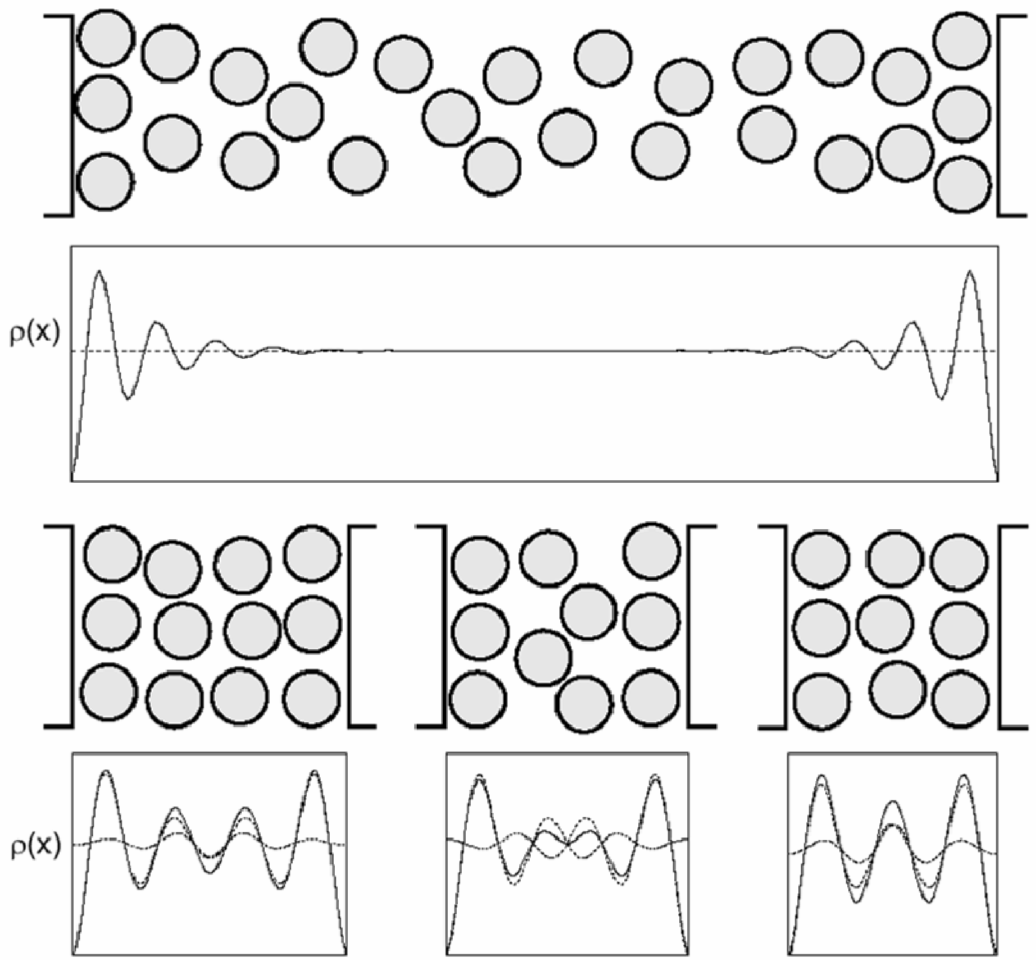






Microfluidics

Density profiles reveal confinement effects



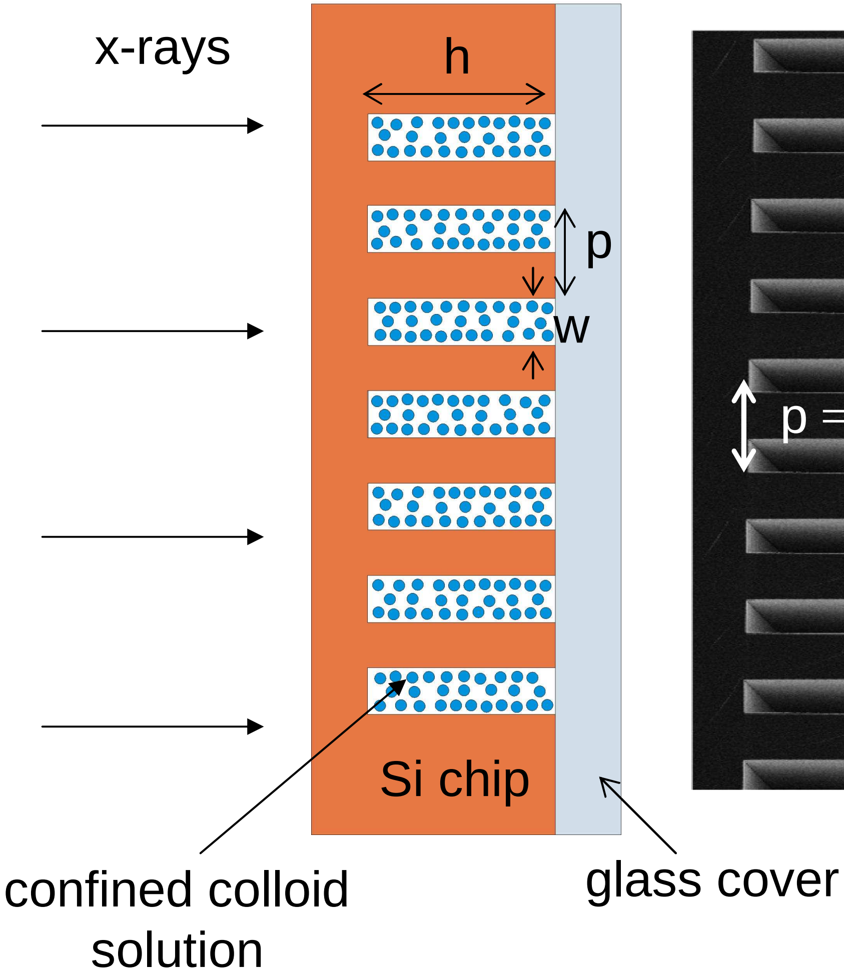
Ordering close to surfaces

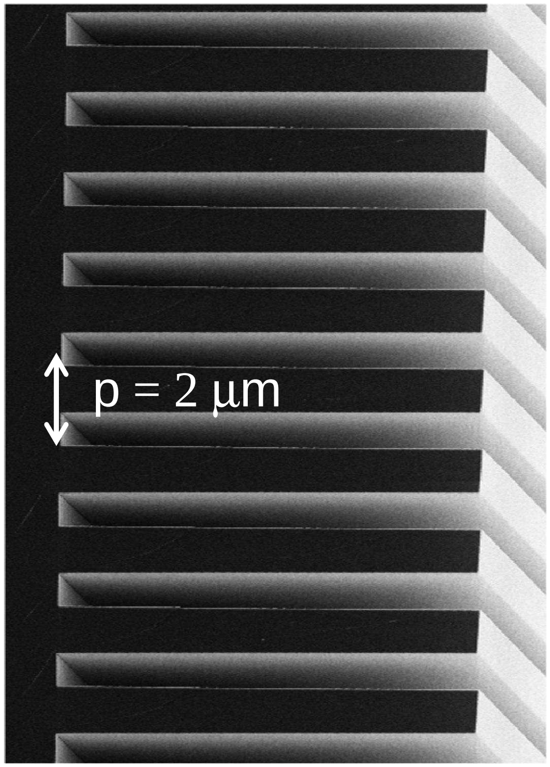
Integer/fractional effects

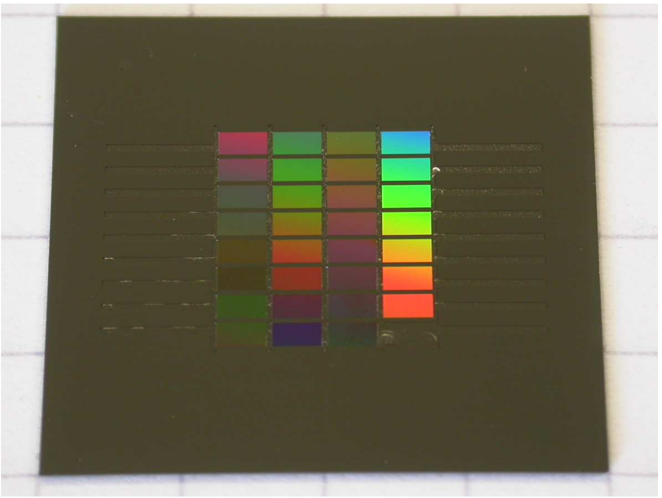
Do they exist?

Arrays of sample containers

O. Bunk et al., Phys. Rev. E75 (2007) 021501







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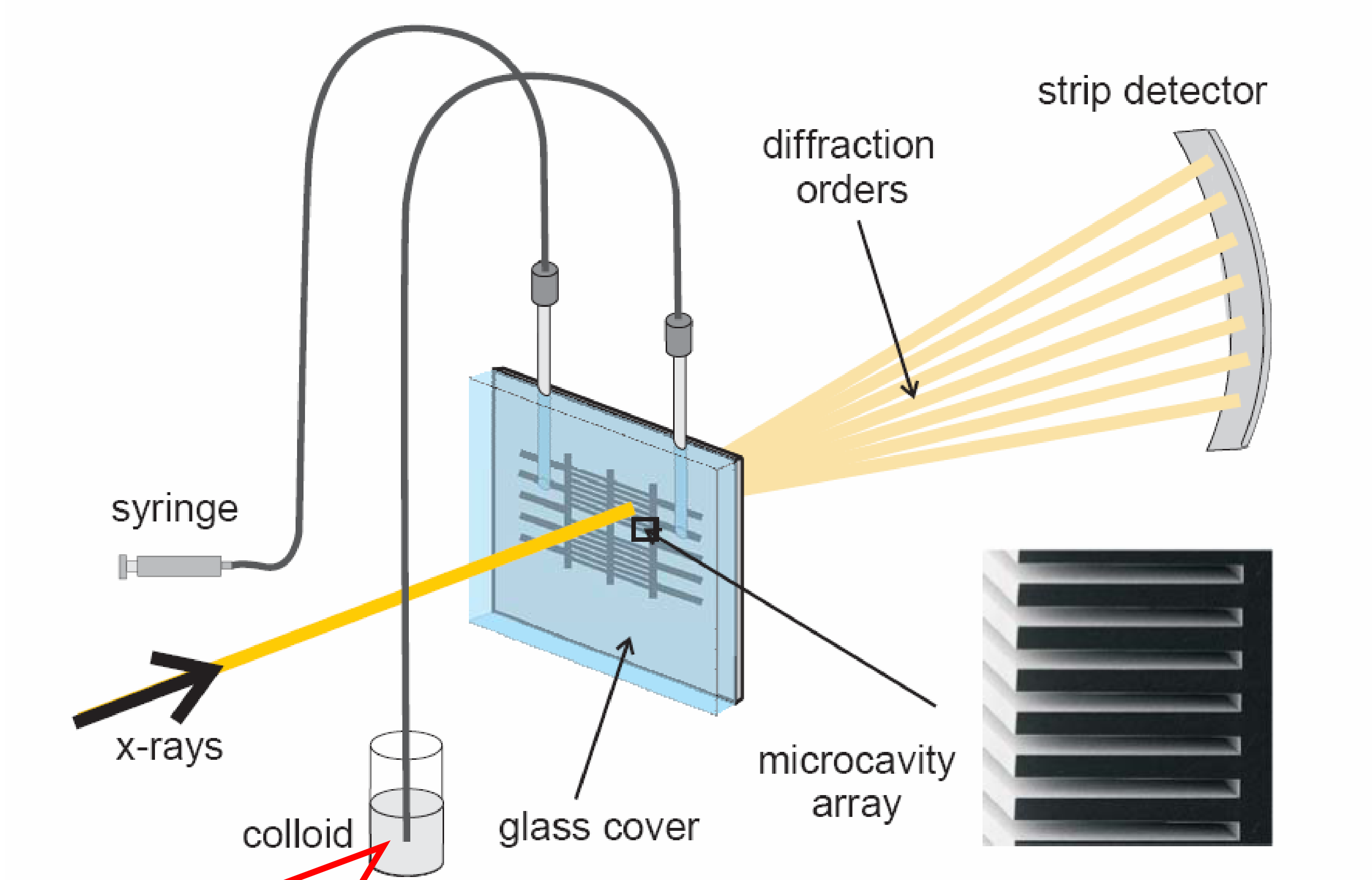
Scattering from filled microcavity array

O. Bunk et al., Phys. Rev. E75 (2007) 021501
A. Diaz and J.F. van der Veen, Thin Solid Films 515 (2007) 5645

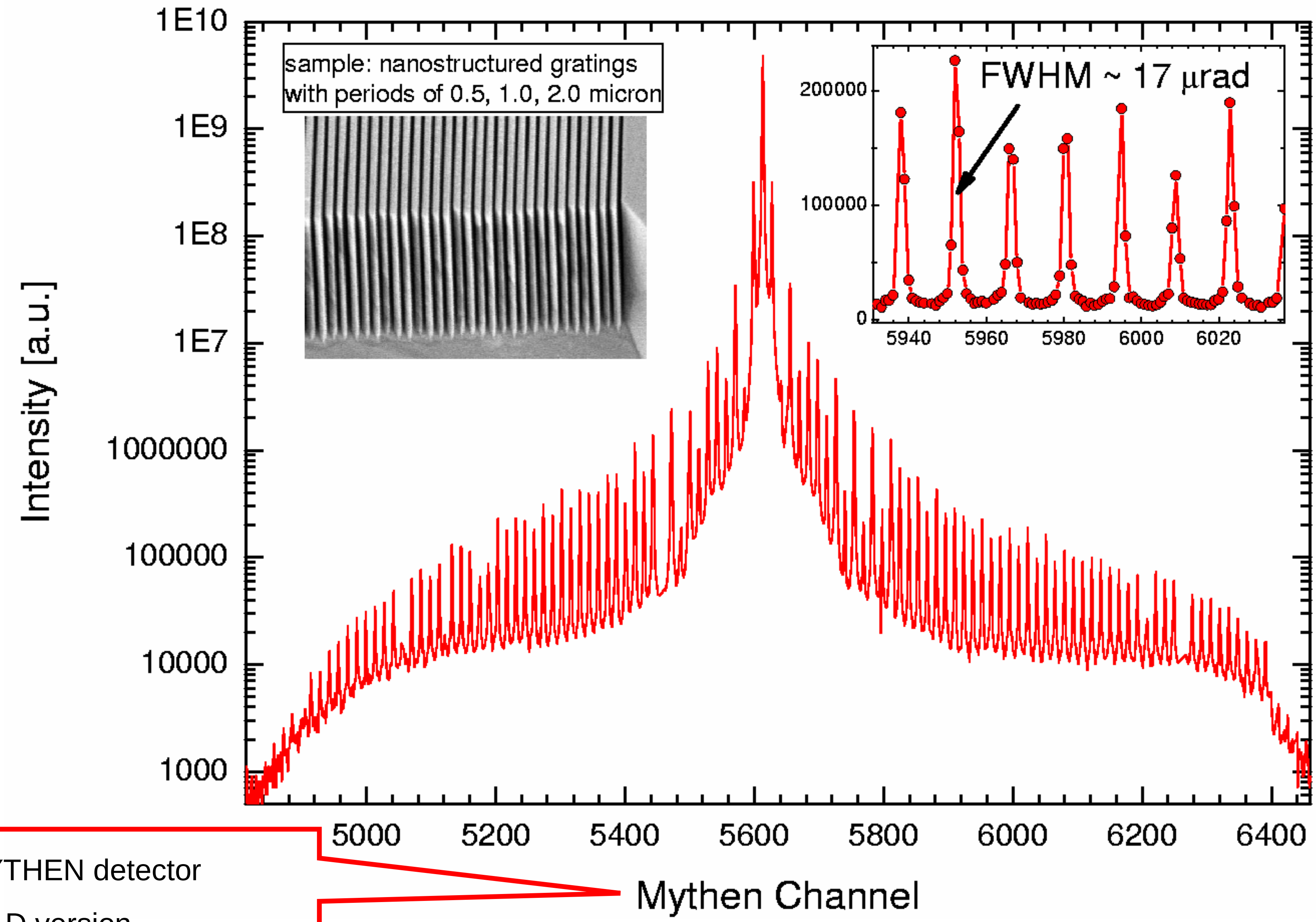


11 vol % silica spheres of 109 nm Ø

Polydispersity 3 %

solvent: 55% benzyl alcohol
and 45% ethanol

Diffraction pattern from cavity array

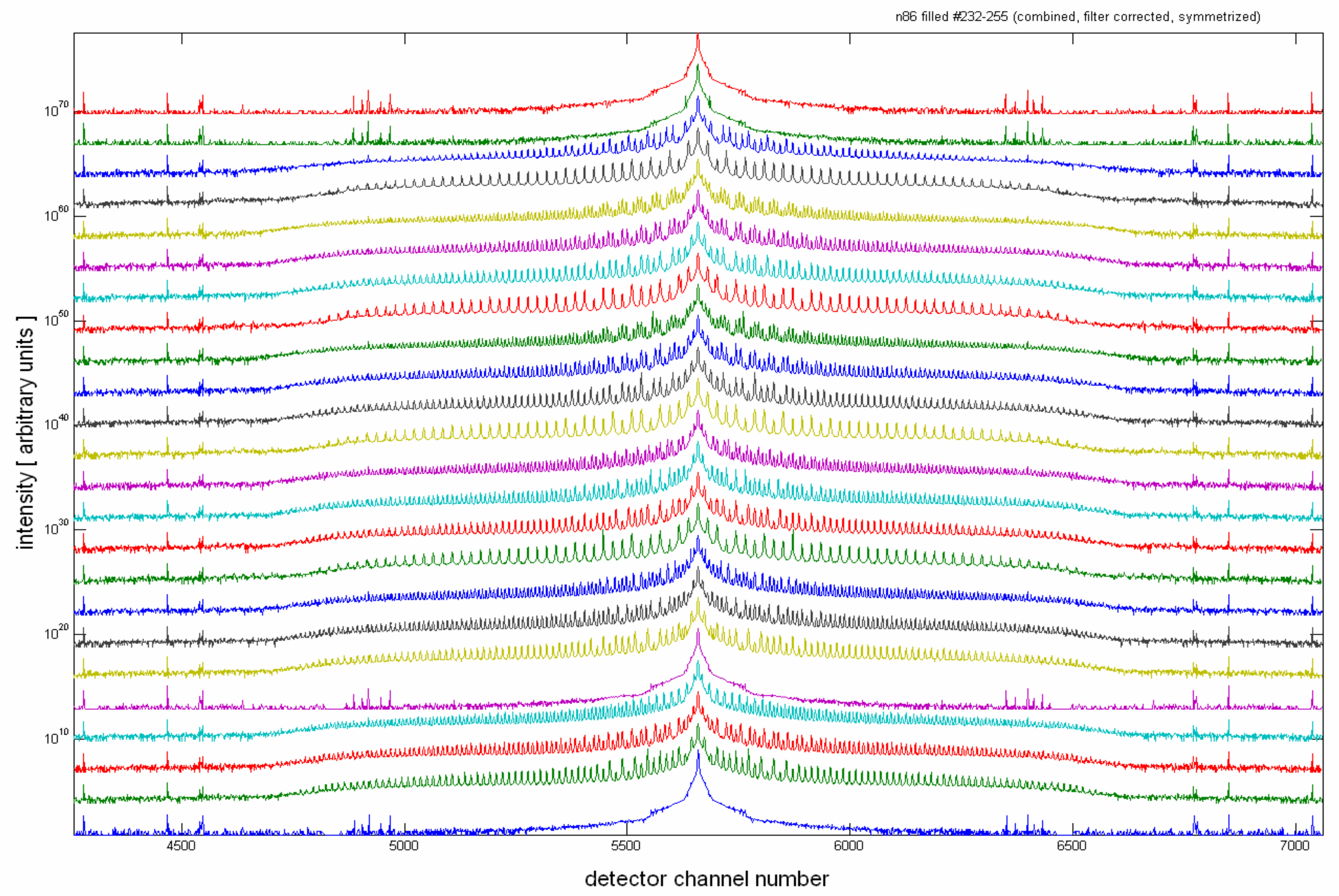


MYTHEN detector

is 1D version

of PILATUS pixel detector

One data set for each grating



Data analysis using iterative phase retrieval

O. Bunk et al., Acta Cryst. A63 (2007) 306 -314

Combining algorithms:

- Gerchberg Saxton's error reduction
- Hybrid Input Output (Fienup)

Constraints:

- constant phase and amplitude in Si walls
- amplitude and phase coupled in the gap
- ±0.1 rad phase range in the gap
- no restriction in the regions near the confining walls

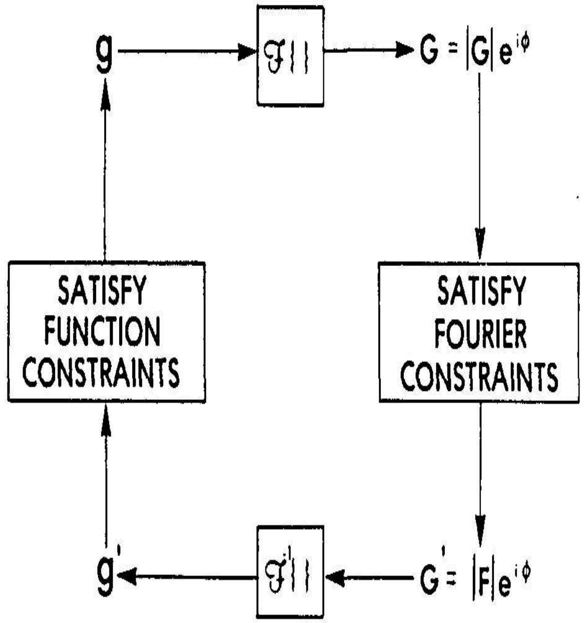
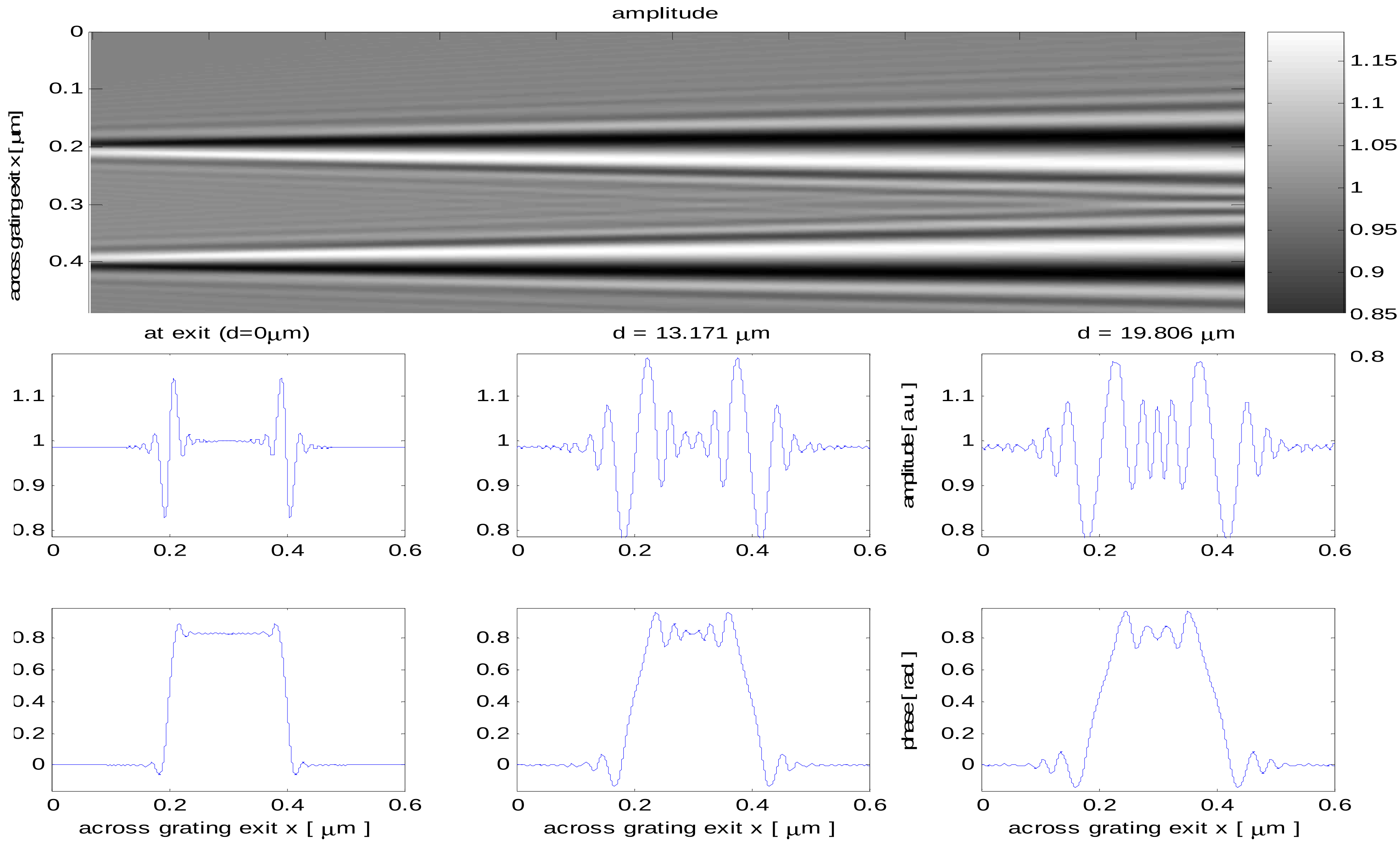


Fig. 1. Block diagram of the error-reduction (Gerchberg-Saxton) algorithm.

Fienup, Appl. Opt. (1982)

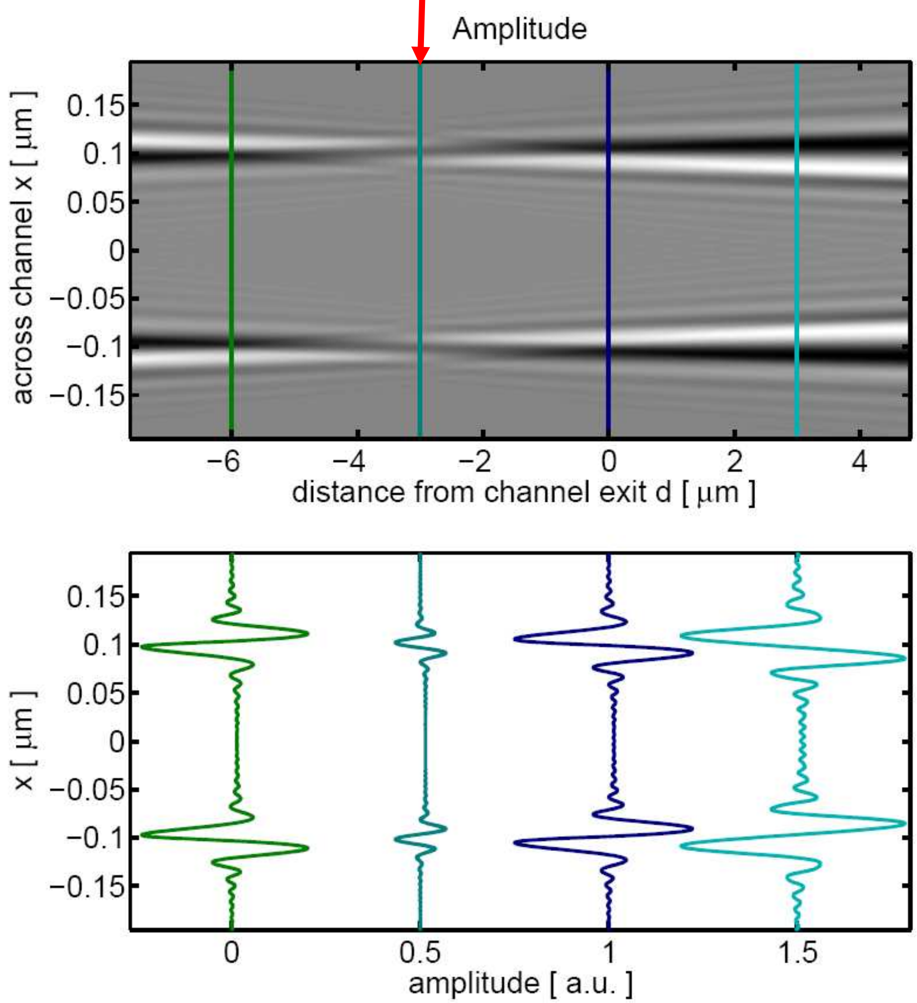
....but many valid solutions!



Free space *back* propagation in each iteration minimizes propagation effects

Halfway back

(thin object approach)

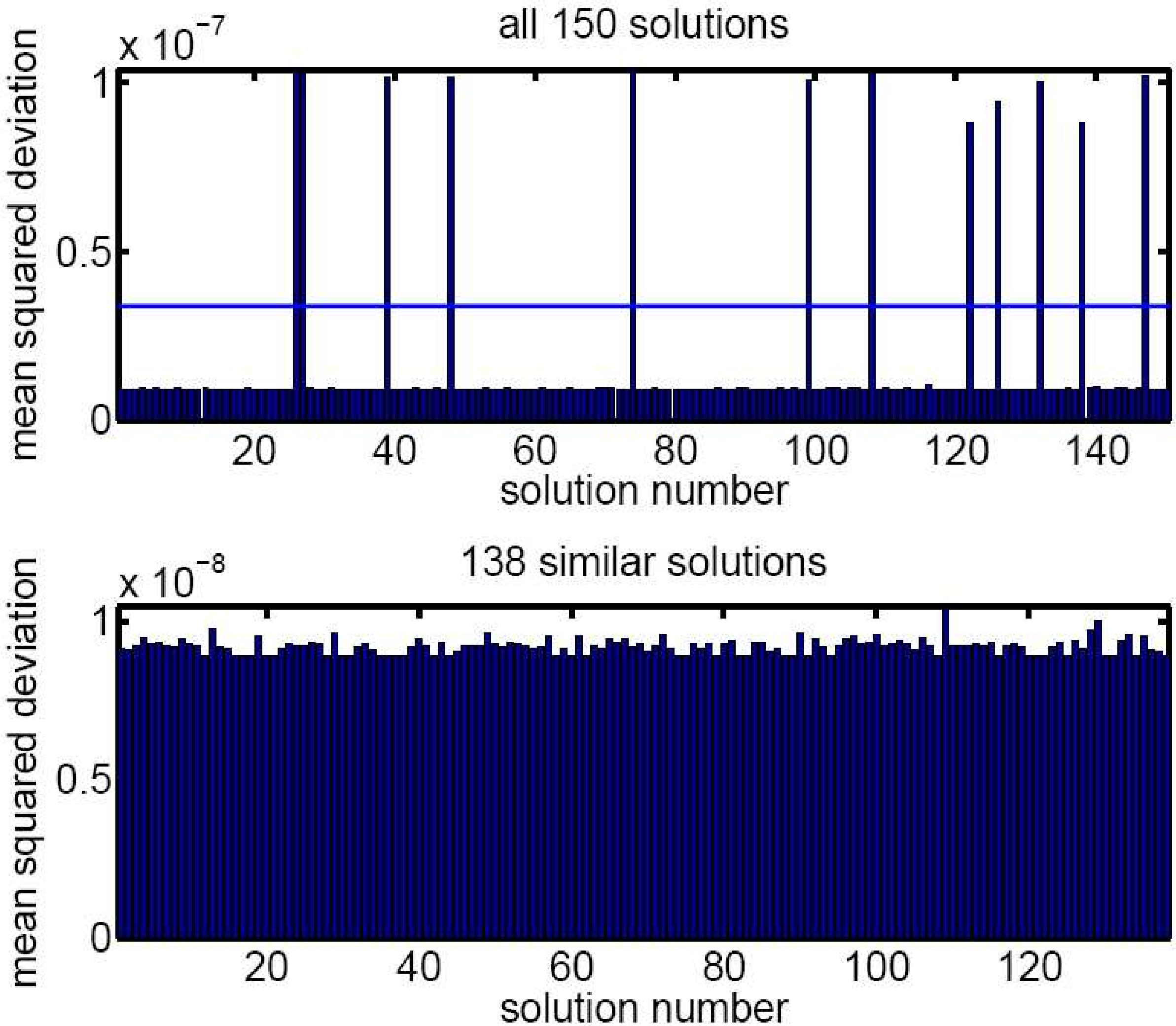


Fresnel propagator:

$$P(z) = e^{-izq^2/(2k)}$$

....allows for tighter constraints.

Phase retrieval: statistics



Refinement of initially random solutions

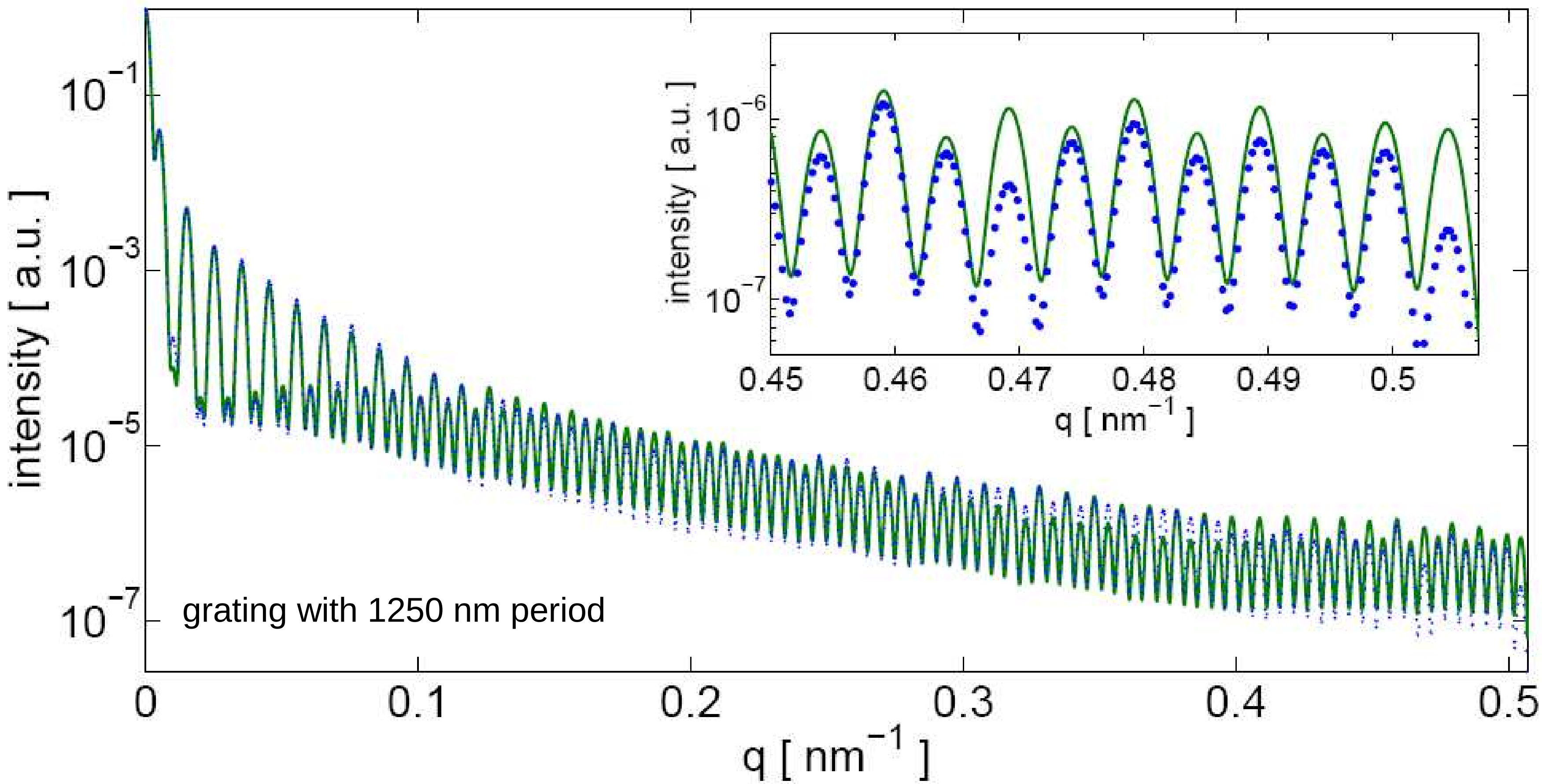
Rejecting dissimilar solutions

→ Averaging

Phase retrieval: how to…

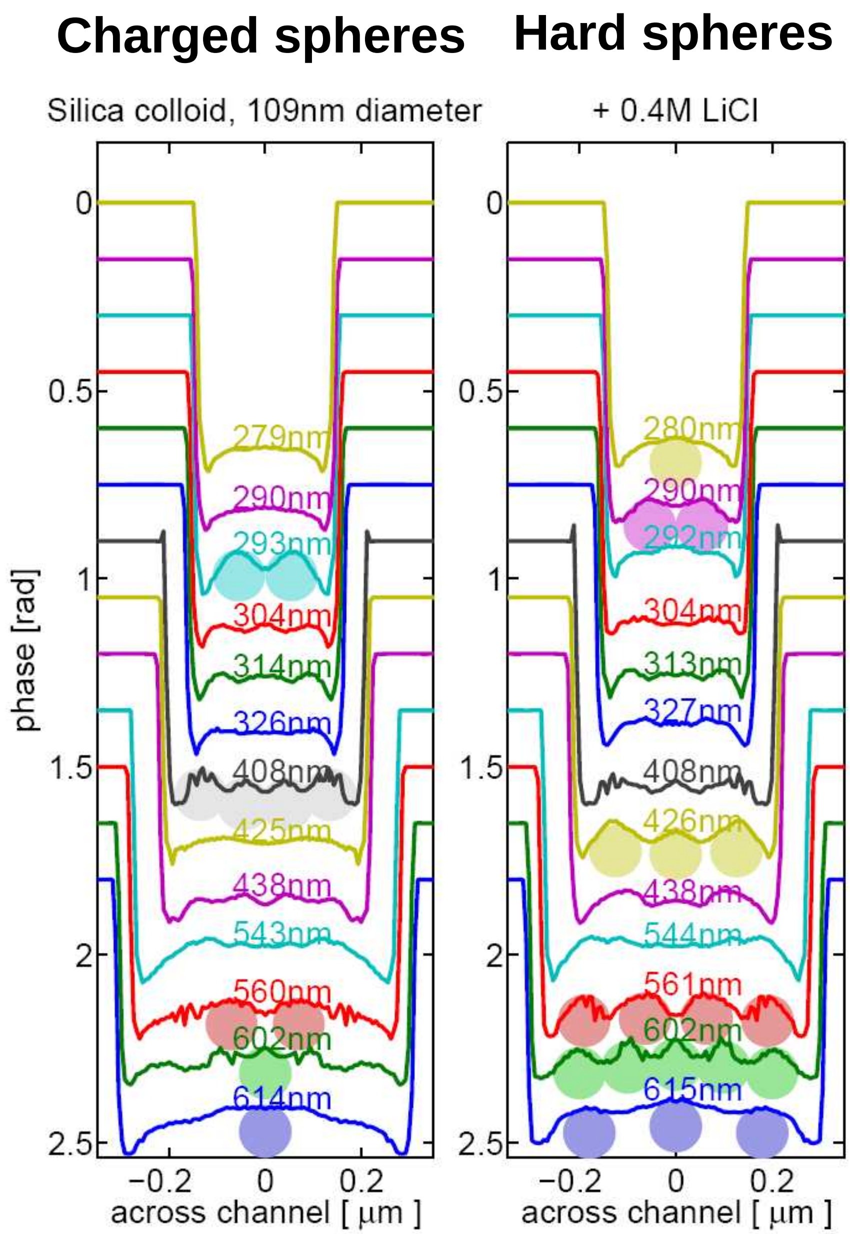
- The constraints determine the position of the reconstructed plane.
- Even thin objects like few microns deep gratings exhibit in their exit field propagation effects.
- Free space back propagation can be used to reduce propagation effects.
- Refinement of several initially random solutions
- Rejecting very dissimilar solutions
- Several solutions should be averaged to estimate the uncertainties.

Comparison measured/calculated intensities



O. Bunk et al., Acta Cryst. A63 (2007) 306 -314

Phase profiles of confined colloids

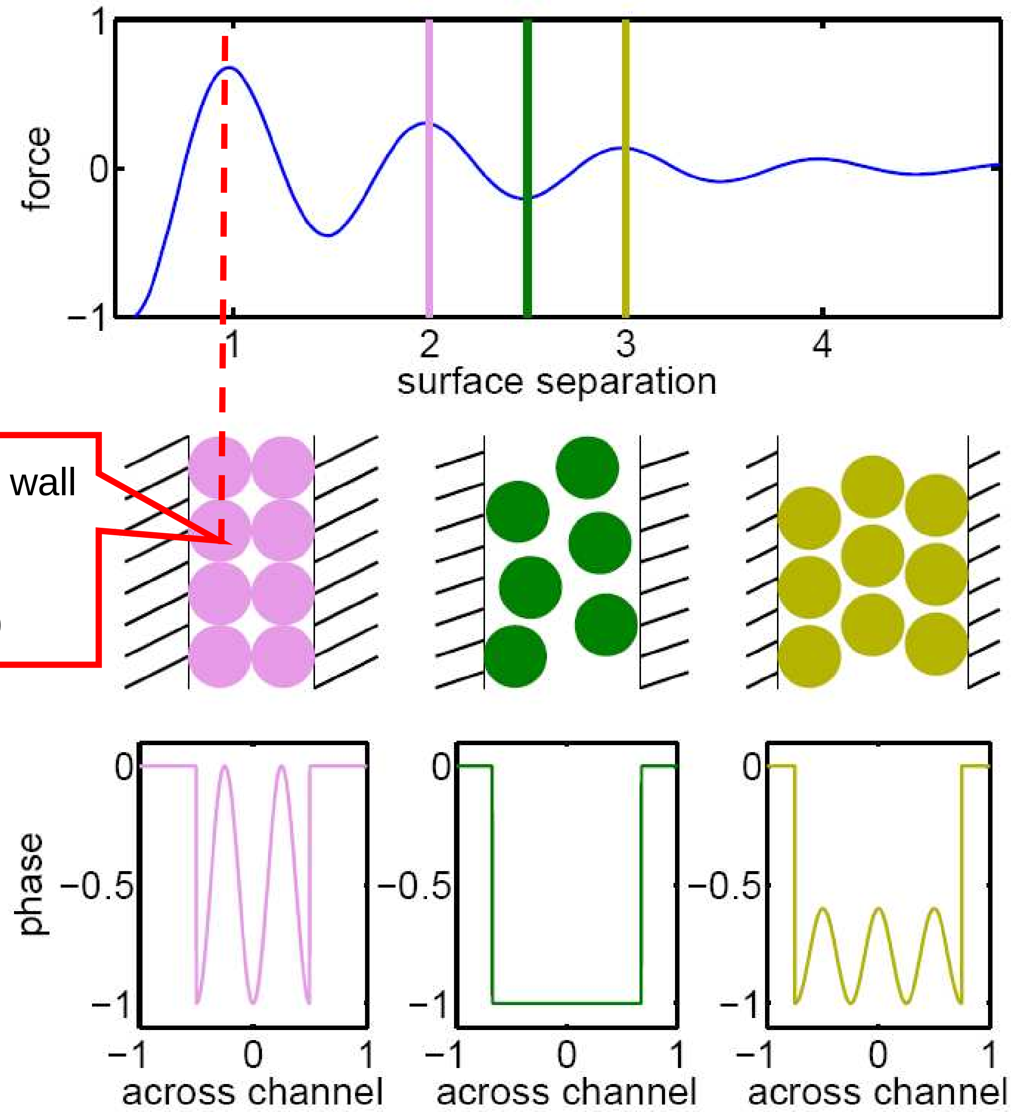


Ordering, if integer number of layers fit within gap.

Structural forces, as predicted!

10 vol% silica spheres in 55% benzyl alcohol 45% ethanol

Oscillatory ordering



SFA

J. Israelachvili et al.

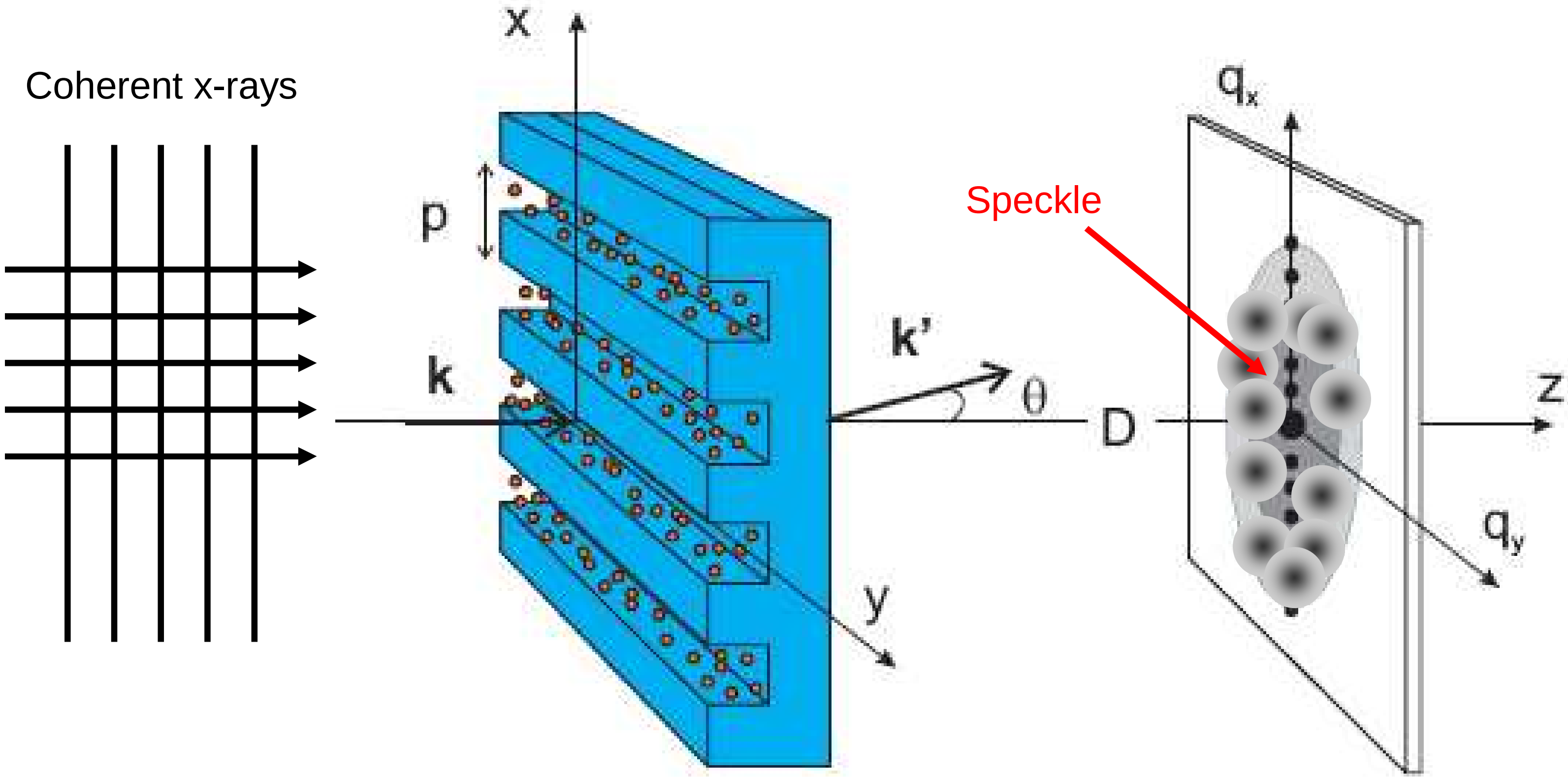
Higher pressure against wall

due to higher density

(Contact value theorem)

X-ray diffraction
O. Bunk et al.,
Phys. Rev. E75
(2007) 021501

ERL: XPCS of confined fluids

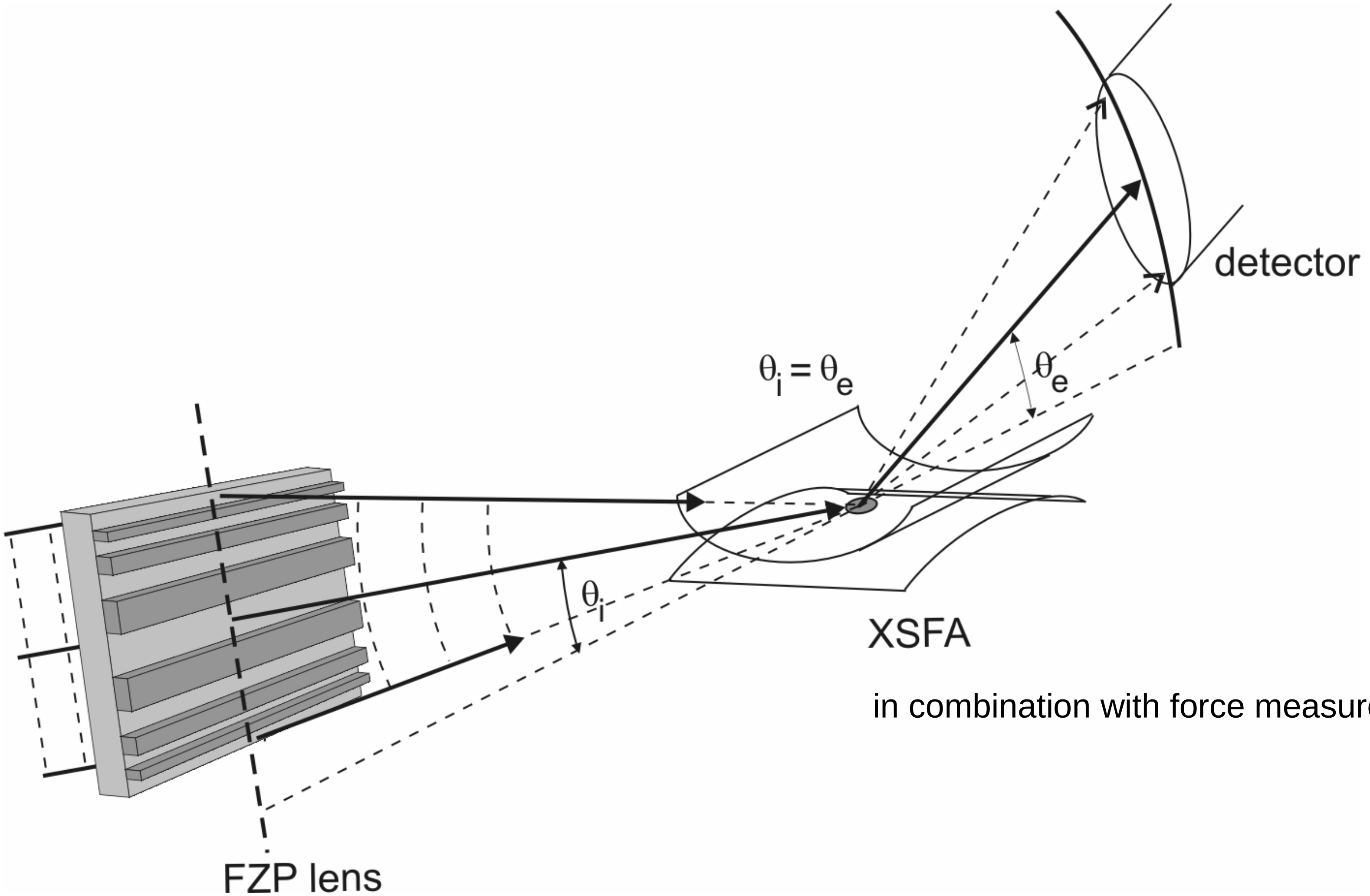


⇒ Correlation between stucture and dynamics

Confinement-induced slowing down?

Study also 2D confinement!

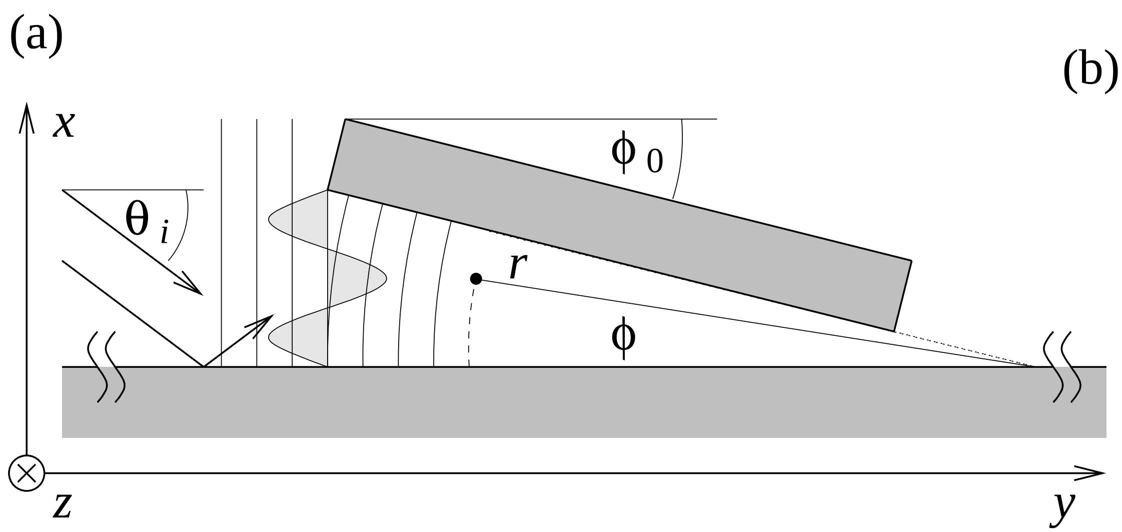
ERL: Nanofocus for studies of fluids confined in single nm-sized gap

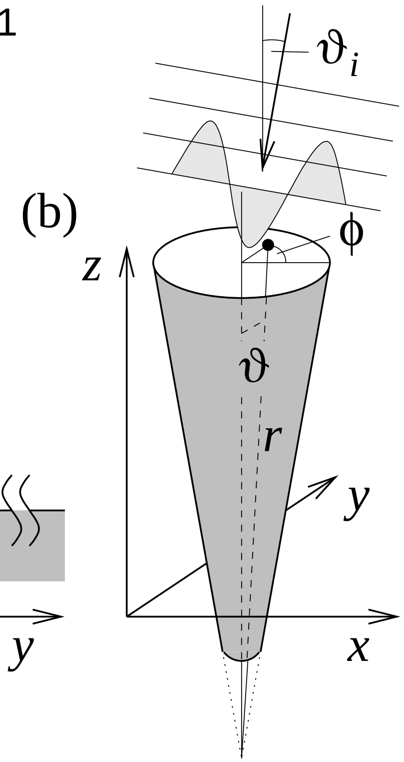


in combination with force measurements

Wedges and capillaries

C. Bergemann et al., PRL 91 (2003) 204801





M.J. Zwanenburg et al., Physica 283B (2000) 285

D. H. Bilderback et al., Science 263 (1994) 201

- Squeeze X-ray beams
- Become waveguides for small diameters
- Coupling into the waveguide critical for efficiency

Waveguide modes are found by solving Helmholtz equation: $\Delta u + n^2 k^2 u = 0$

Equivalence to particle-in-a-box problem

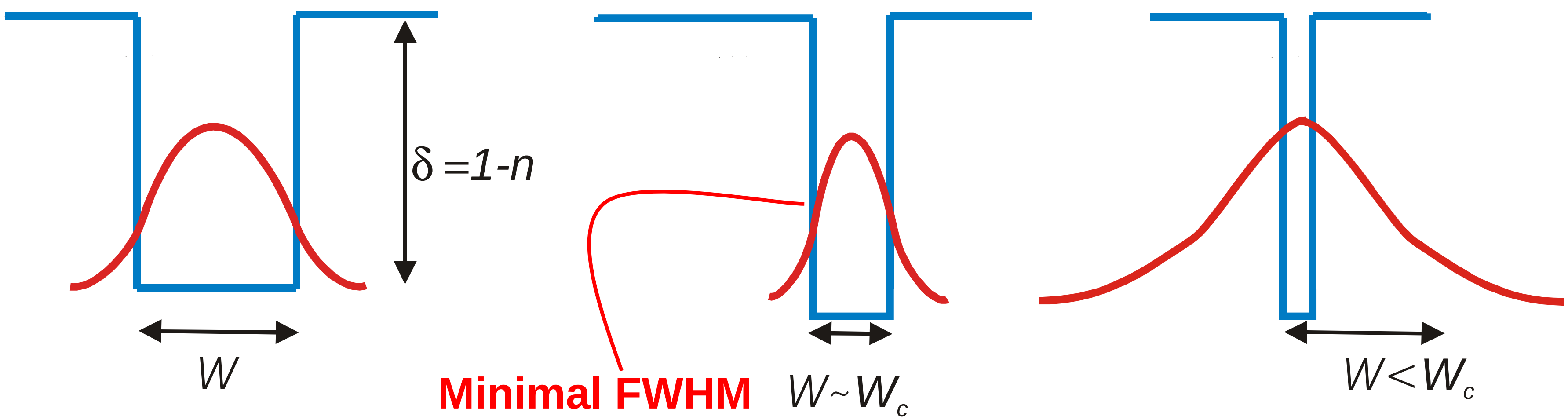
Write $u = \psi(x,y)e^{-iky}$, with ψ slowly varying envelope function.

Substitute in Helmholtz eq. and neglect $\partial^2\psi/\partial y^2$.

$$\Longrightarrow \quad -\frac{1}{2k^2}\frac{\partial^2\psi}{\partial x^2} + \delta\cdot\psi = \frac{i}{k}\frac{\partial\psi}{\partial y}$$ Schrödinger equation

Wedge ⟺ Box of shrinking size

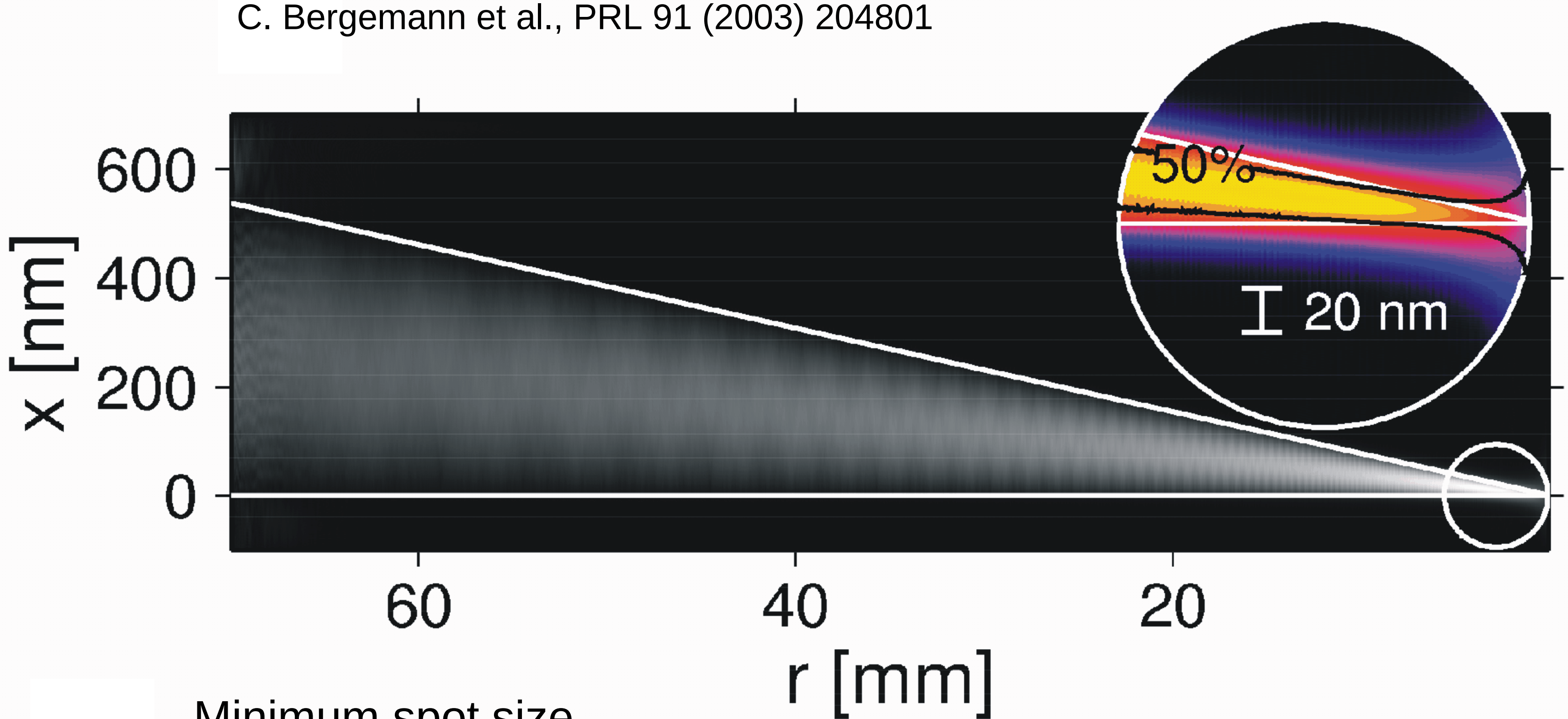
Lowest state remains bound:



C. Bergemann et al., PRL 91 (2003) 204801

What is the smallest possible spot size ?

C. Bergemann et al., PRL 91 (2003) 204801



Minimum spot size (FWHM): Δx_{min}=0.64 W_c, with

$$W_c = \frac{\lambda}{2\theta_c} = \frac{1}{2} \cdot \sqrt{\frac{\pi}{r_0 n_e}}$$

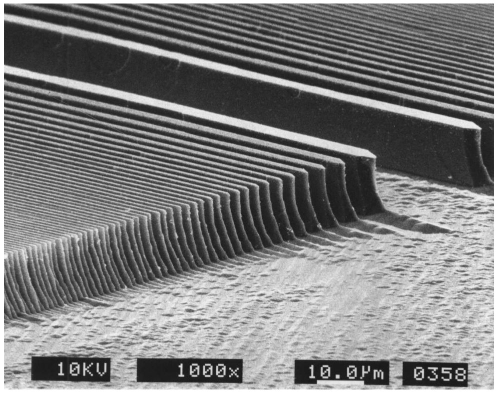
SiO_2: Δx_{min} = 13 nm
Au: Δx_{min} = 5 nm

Does such a limit on the spot size also exist for other X-ray focusing devices?

Ultimate resolution of Fresnel zone plates (FZPs) (I)

F. Pfeiffer et al., Phys. Rev. B73 (2006) 245331

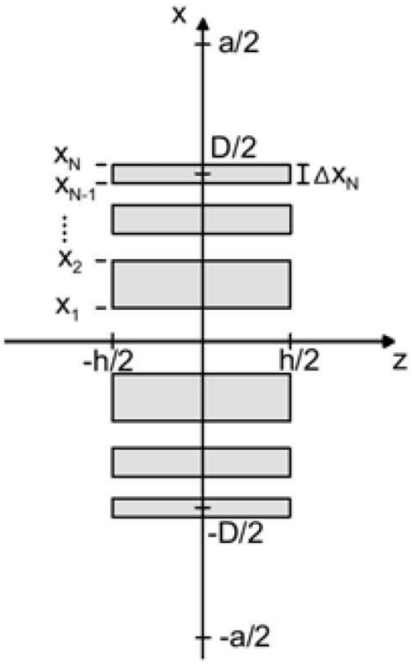
Theoretical minimum spot size:
$\Delta x_{min} \sim d_{min}$,
with d_{min} outermost zone width.



C. David et al.
Spectrochimica Acta B59 (2004) 1505

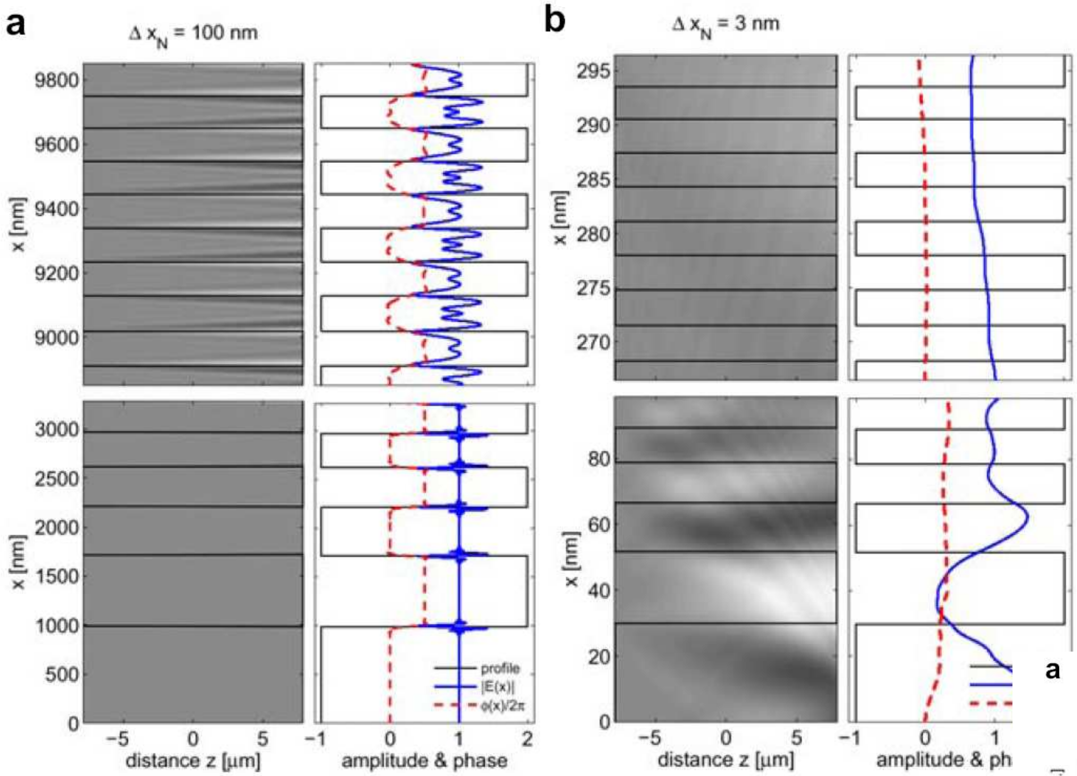
Solve for eigenfunctions of Schrödinger equation for the array of box potentials.

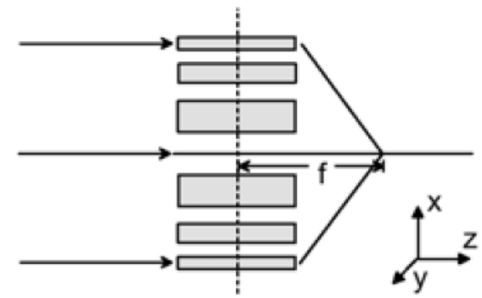
- Outermost zone width d_{min} =1.5 nm,
- Nr. of zones: 600
- $\lambda = 0.1$ nm
- Silicon $\delta = 3.17 \times 10^{-6}$
- Thickness 15.8 µm
- Standing waves up to order 6000 as basis functions

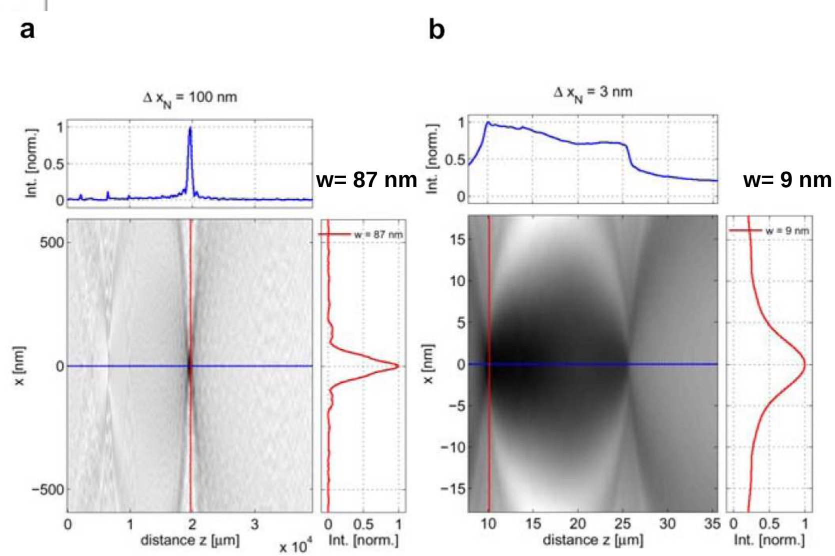


Diagonalize 6000×6000 matrix……..

Illumination by plane wave







F. Pfeiffer et al., Phys. Rev. B73 (2006) 245331

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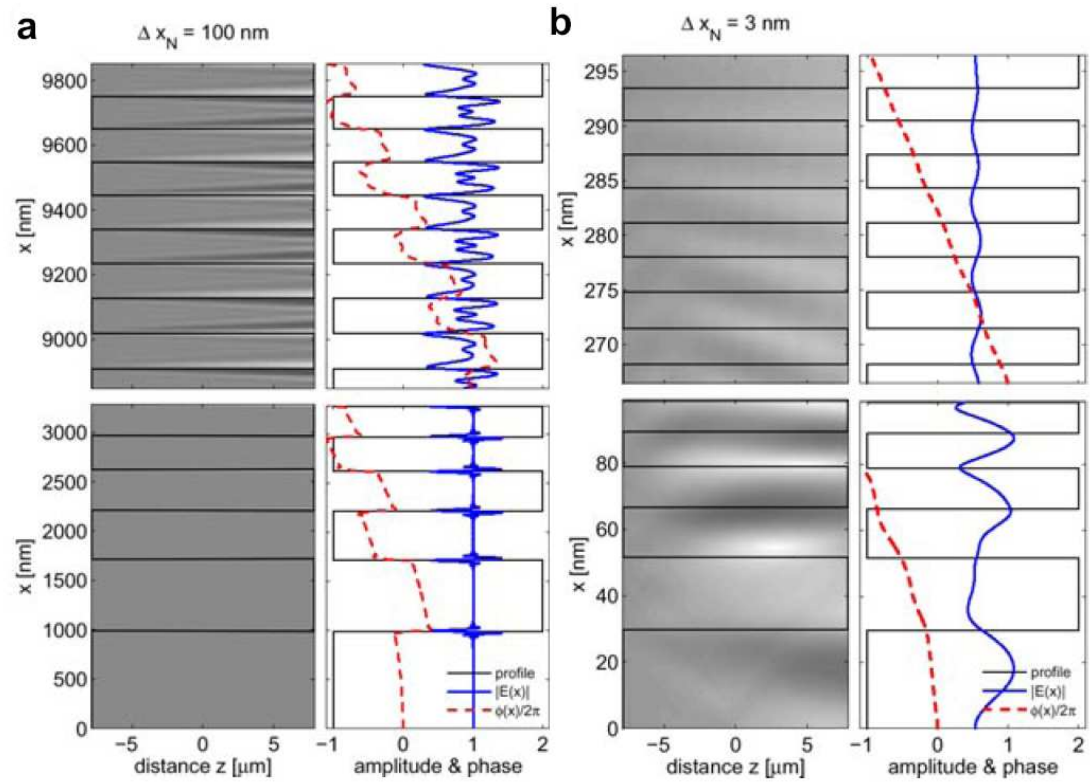
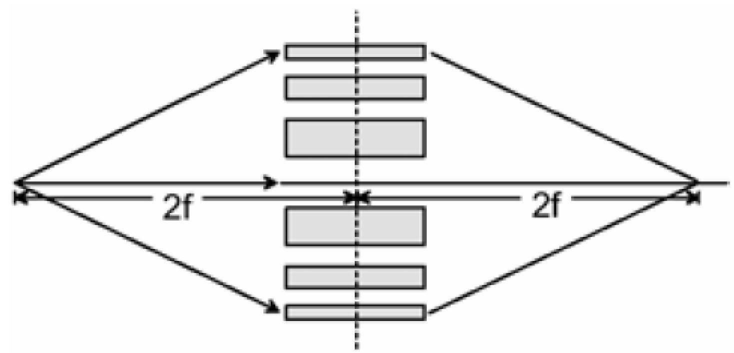
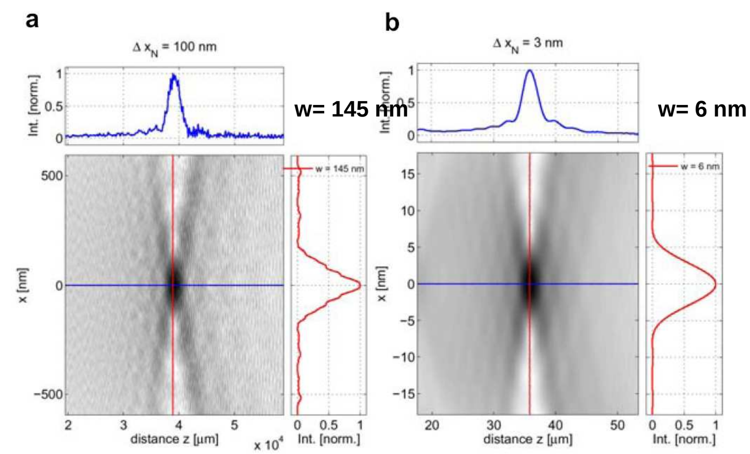
Illumination by cylindrical wave

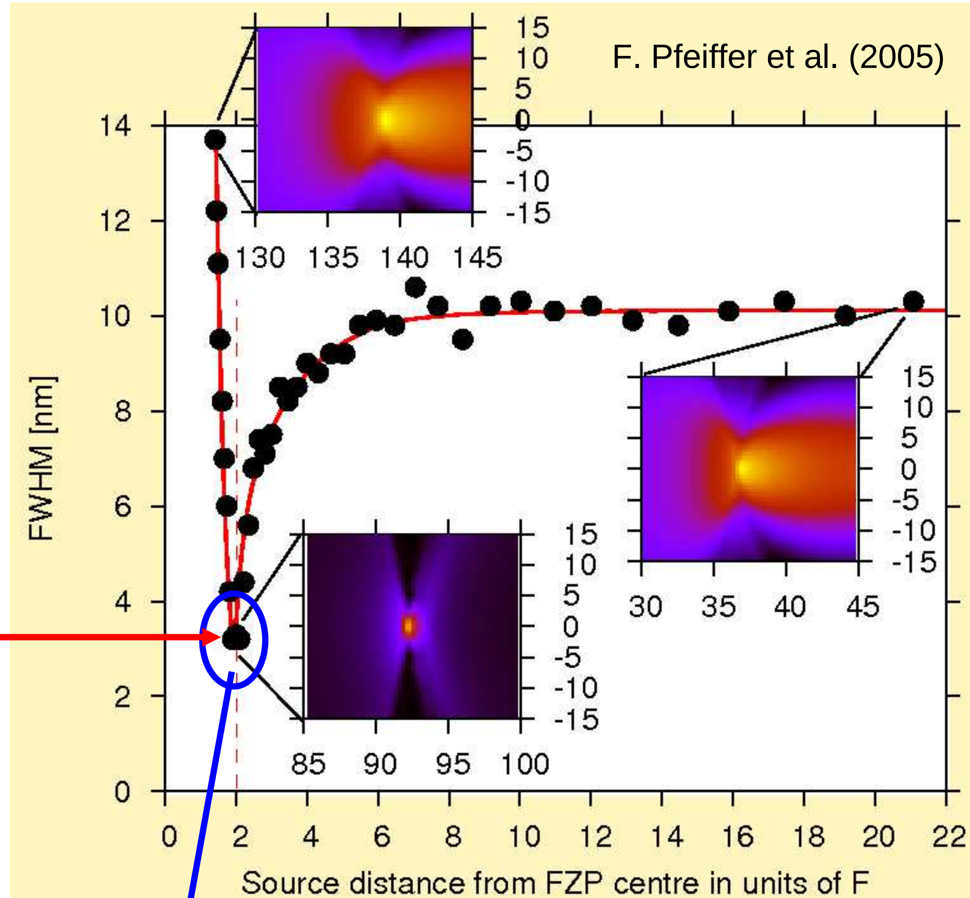




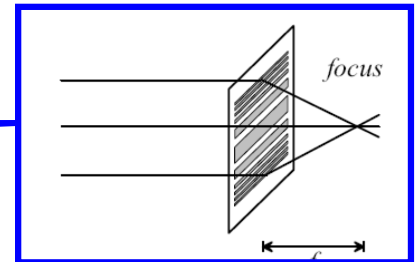
F. Pfeiffer et al., Phys. Rev. B73 (2006) 245331

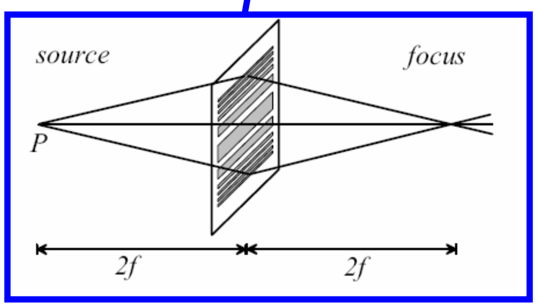


Ultimate resolution of FZPs



FWHM ~ 3 nm





- There is NO fundamental limit on the spot size for FZPs.
- Neither for adiabatic refractive lens arrays! (C.G. Schroer and B. Lengeler, PRL 94 (2005) 054802)

F. Pfeiffer et al., Phys. Rev. B73 (2006) 245331

Outlook

- Confined fluids correlate structure and dynamics
  Relevance: lubrication, wetting, transport though narrow pores (membranes), protein folding, etc.
- Nanofocusing required for studies of single pores
  FZPs: no fundamental limit on minimum spot size . But there are practical limits......