Nanostructure Evolution during Growth of Thin Films as seen by Coherent X-ray Dffraction

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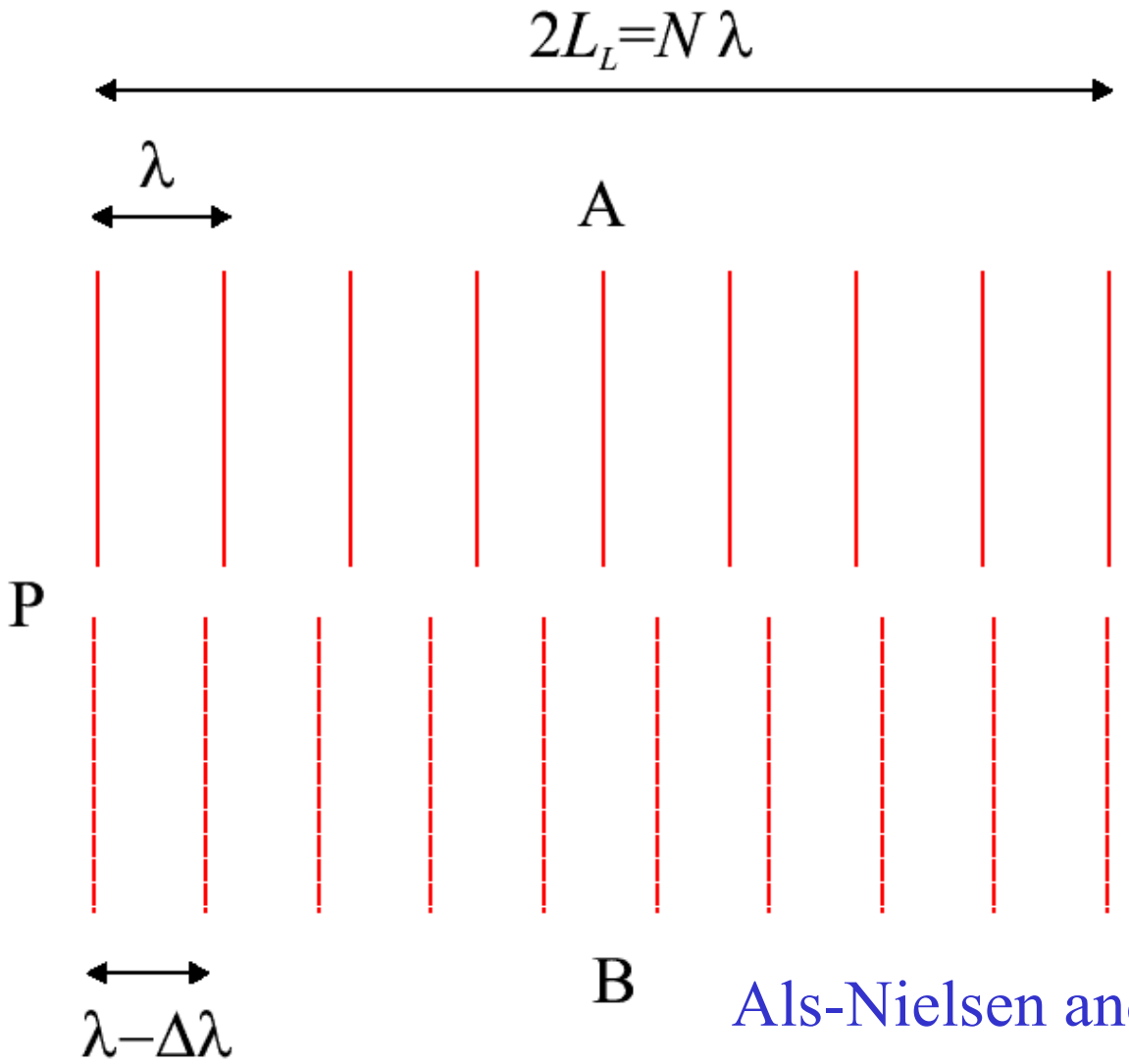
Outline

- Coherence in Diffraction
- Nanocrystal Images
- Coherent “Yoneda” geometry
- Au and Fe polycrystalline films
- Future prospects for CGISAXS

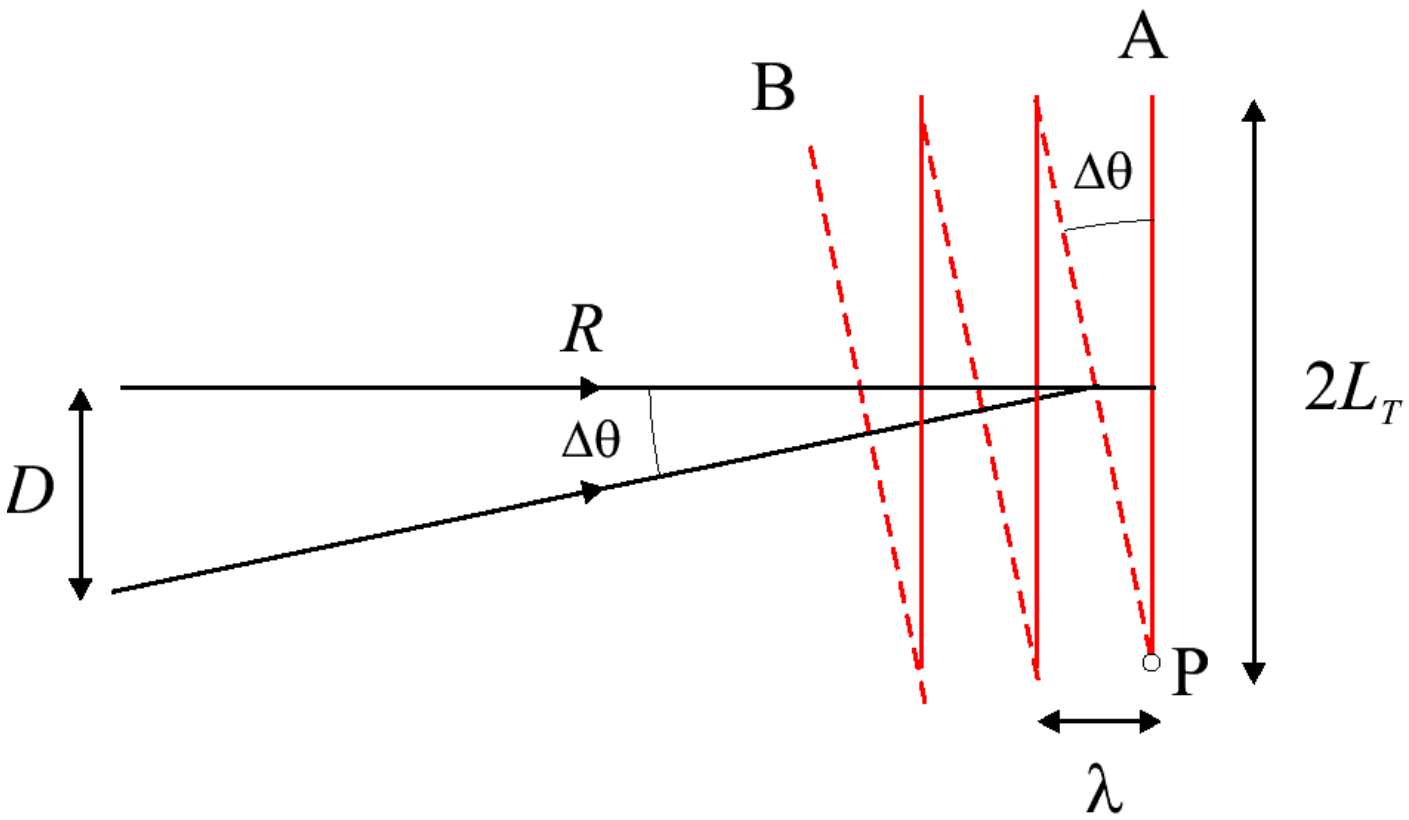
Longitudinal Coherence



$$L_L = \frac{1}{2}\frac{\lambda^2}{\Delta\lambda}$$

Als-Nielsen and McMorrow (2001)

Lateral (Transverse) Coherence



$$L_T = \frac{\lambda}{2}\frac{R}{D}$$

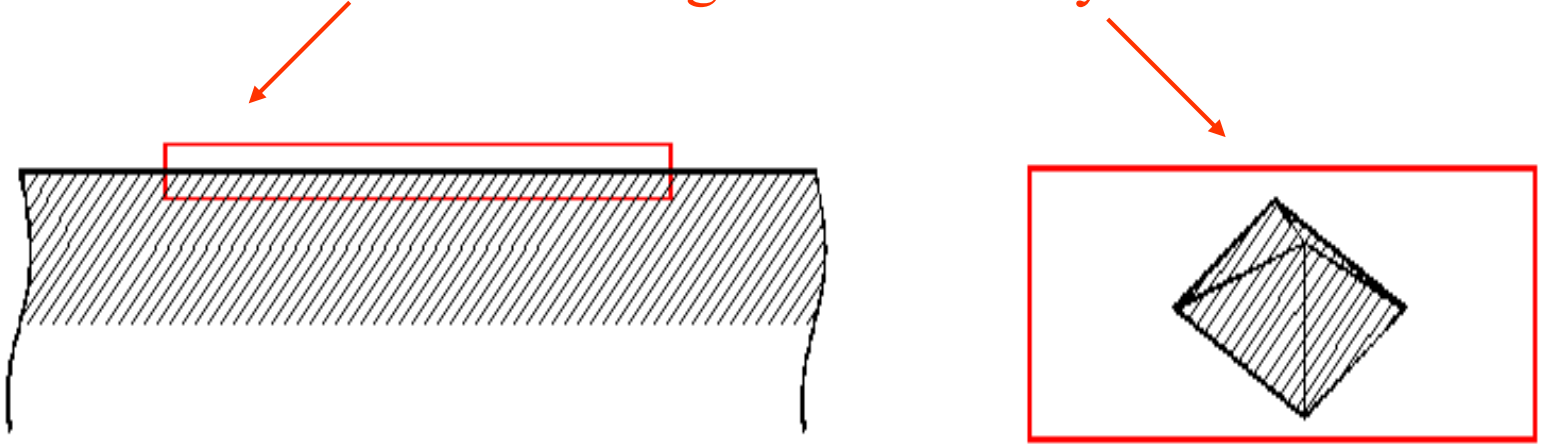
Als-Nielsen and McMorrow (2001)

Coherence at the APS or ESRF

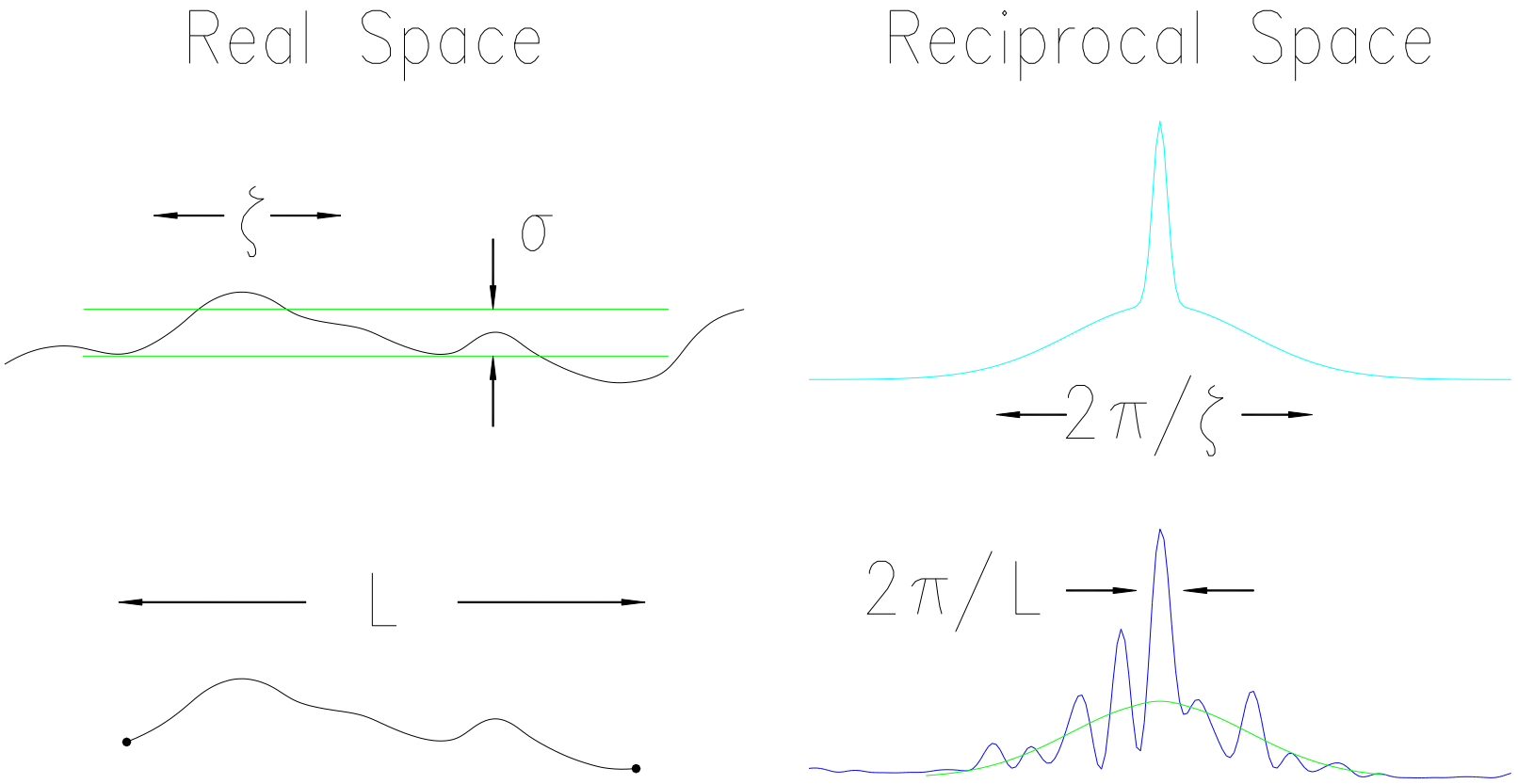
Typical of 3rd Generation (undulator) Synchrotron Source

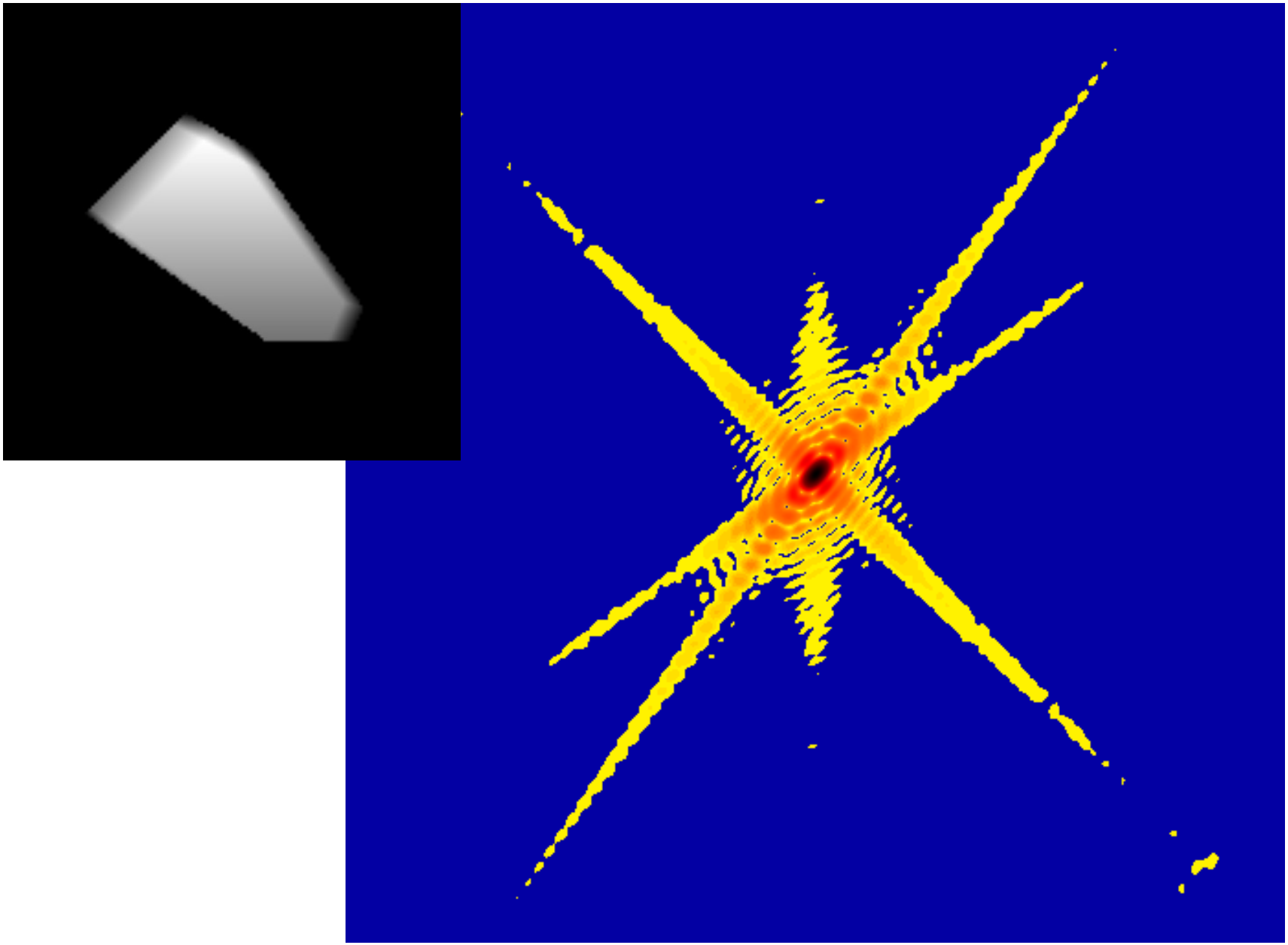
Coherence of	ξ_{VER}	ξ_{HORIZ}	ξ_{LONG}	Flux
Raw Undulator	35μm	9μm	0.004μm	2×10^{12}
Si(111) Monochromator	35μm	9μm	1μm	1×10^{10}
C(111) Monochromator	35μm	9μm	3μm	3×10^{9}

Coherent region defined by slits



Diffuse Scattering acquires Structure using CXD

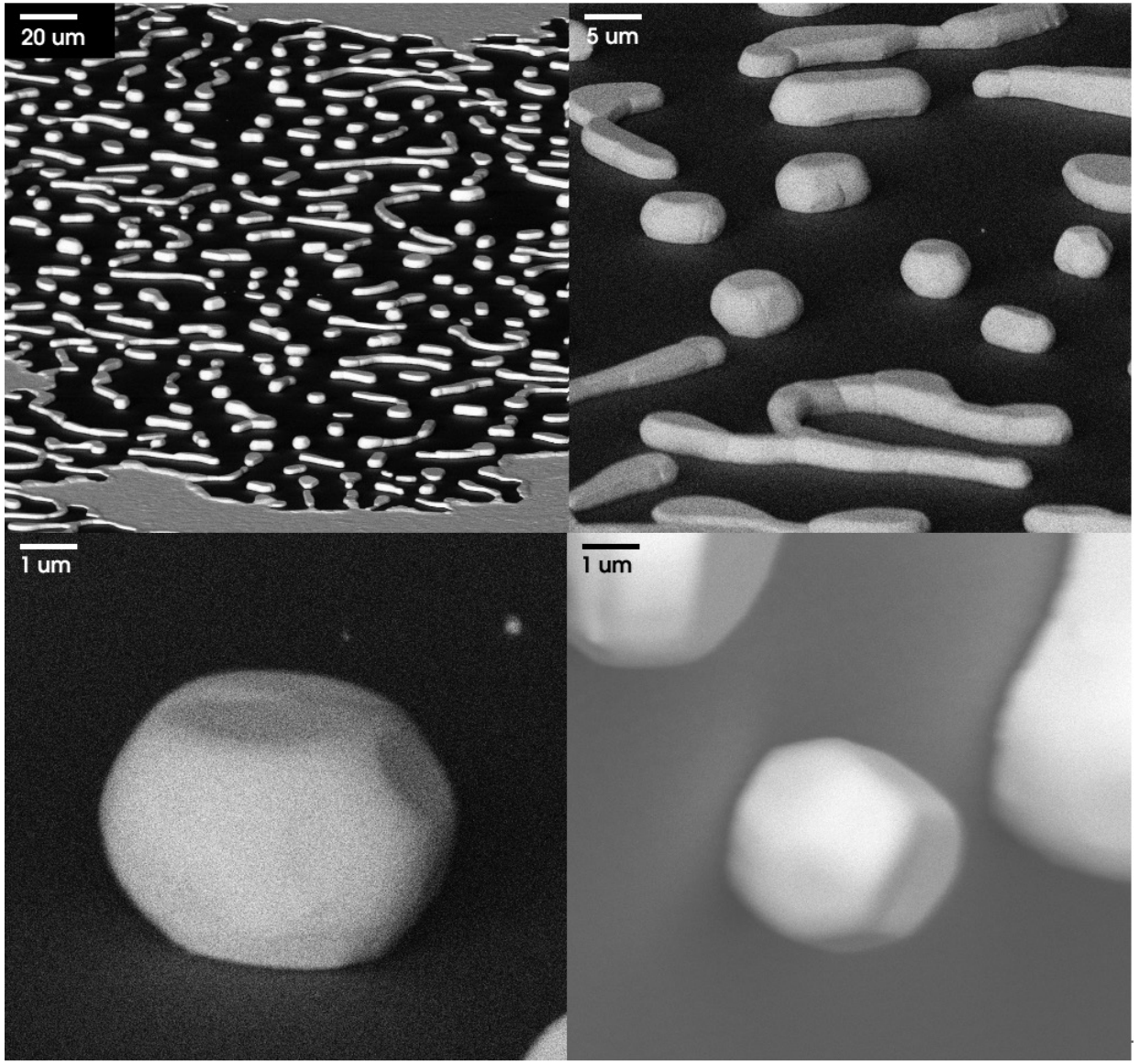




I. K. Robinson SRMS-4 Aug 2004

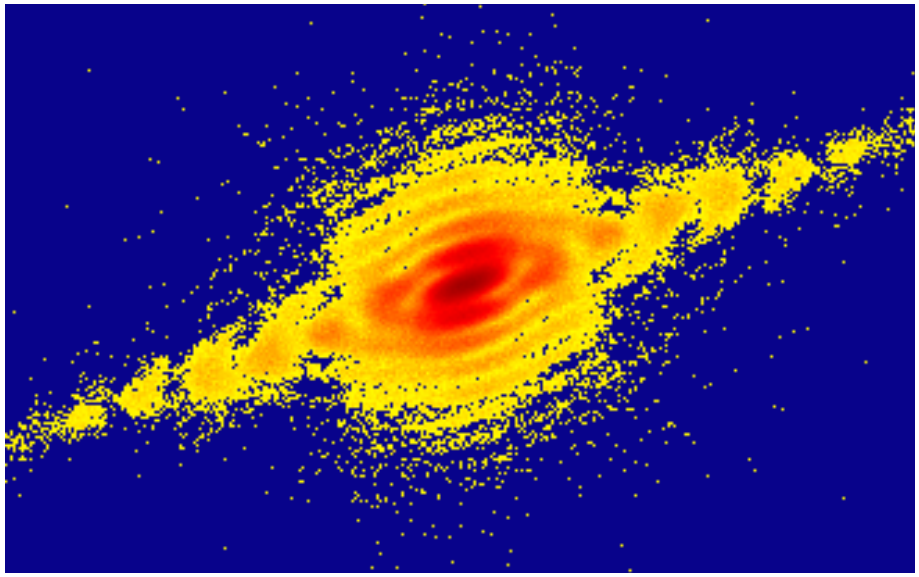
SEMS

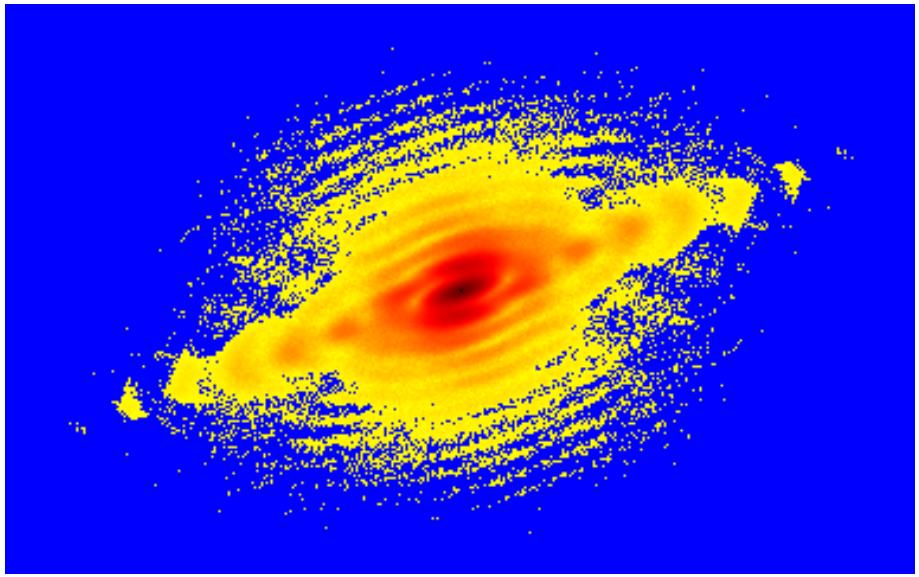
- Au blanket film
- Quartz substrate
- Annealed at 950°C for 70 hrs.

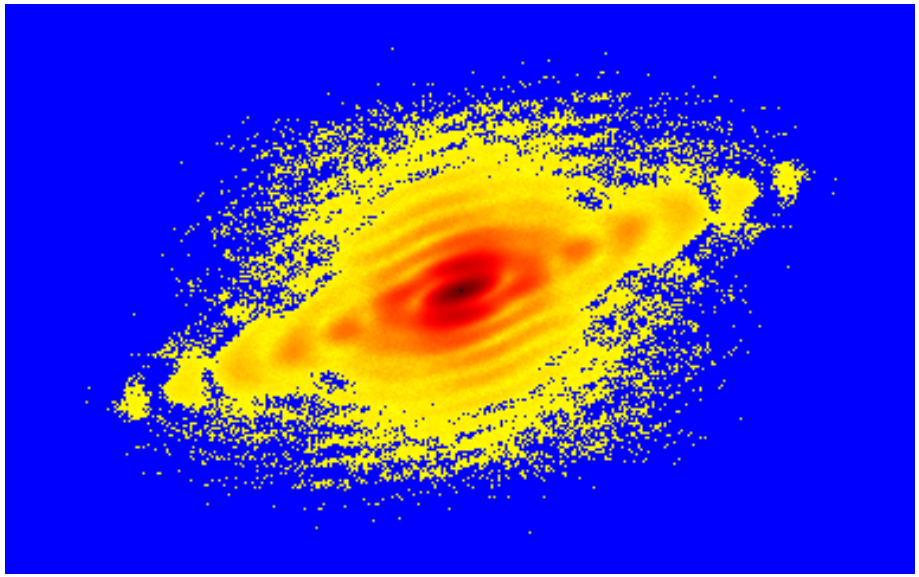


Symmetrized Data and two best fits

Chisq=0.0005

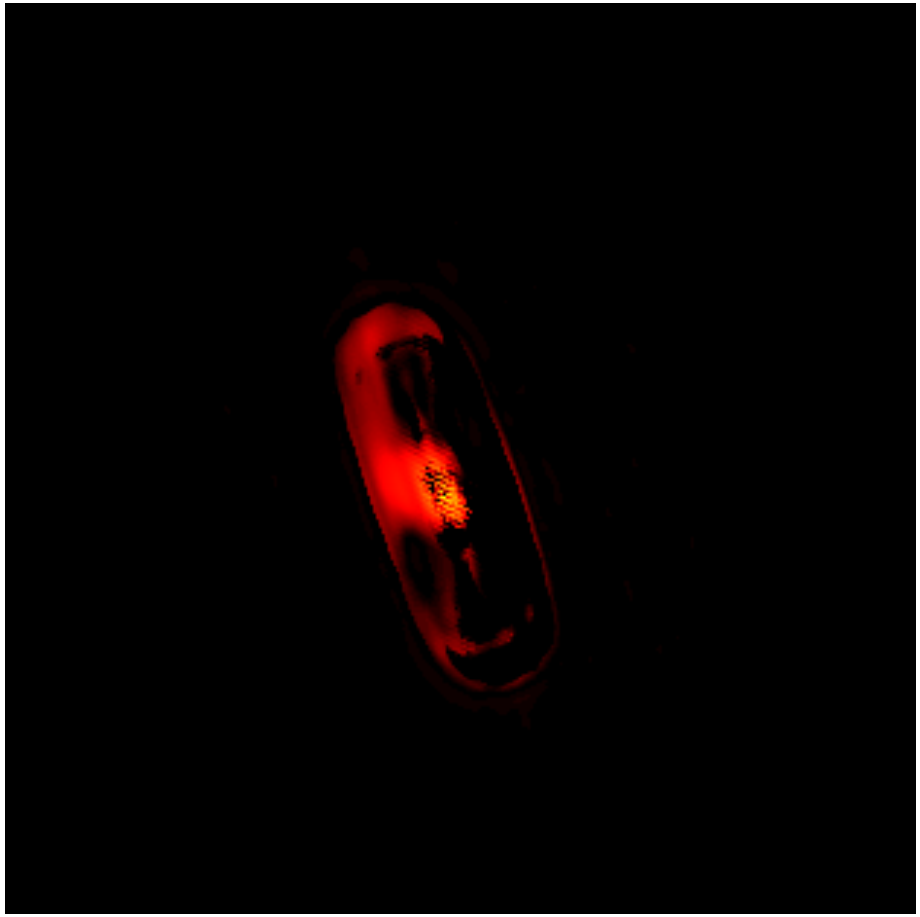


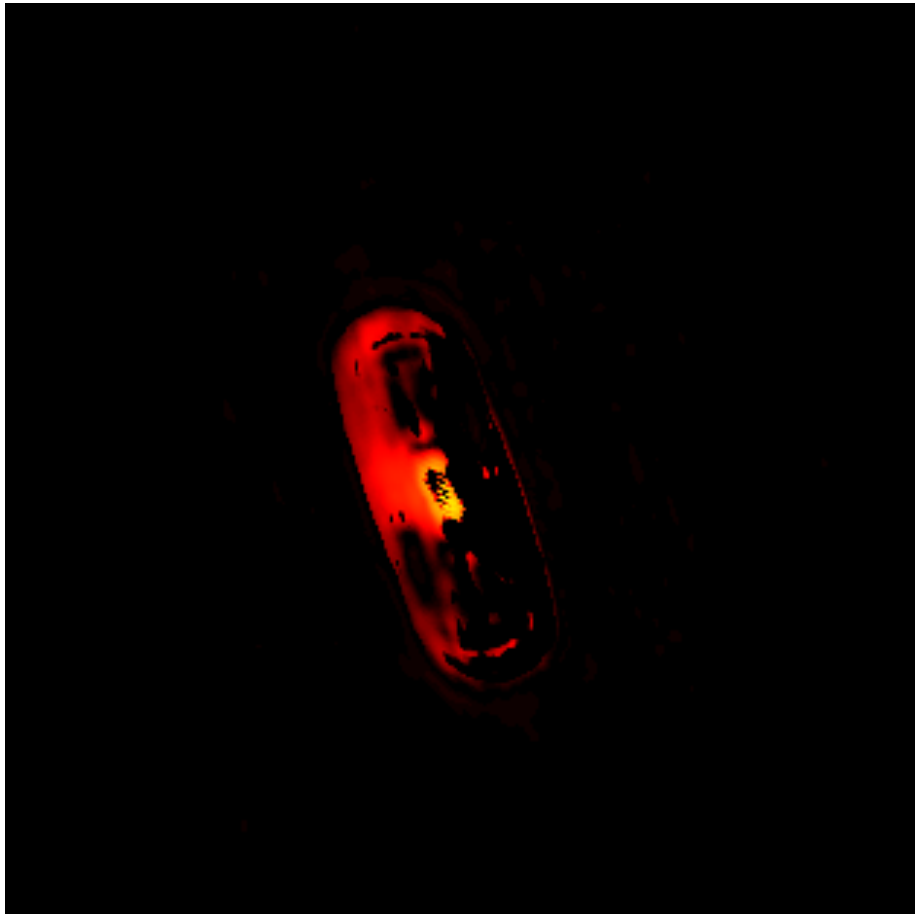


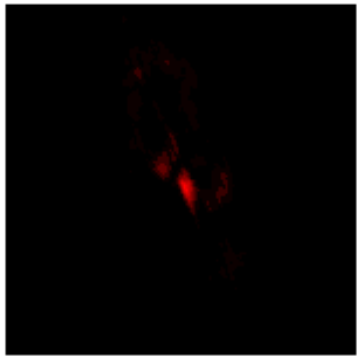
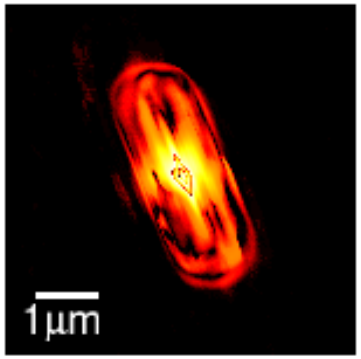


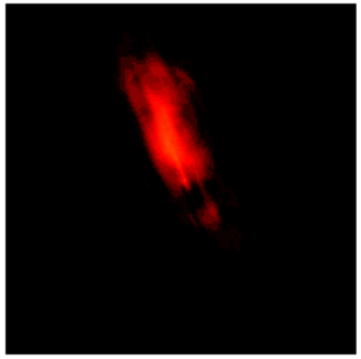
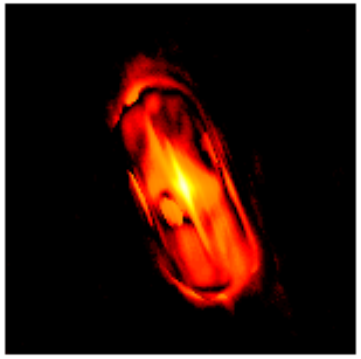
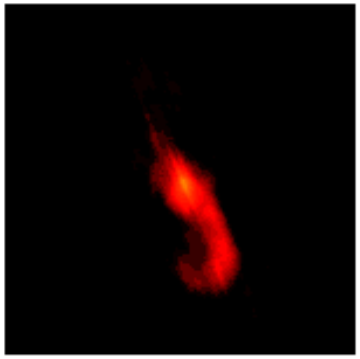
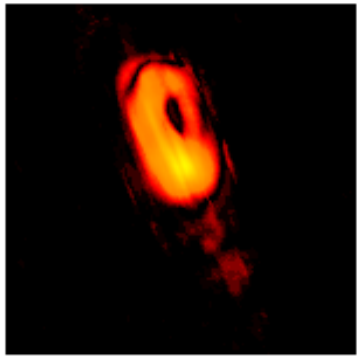
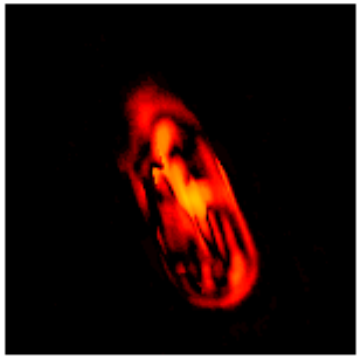
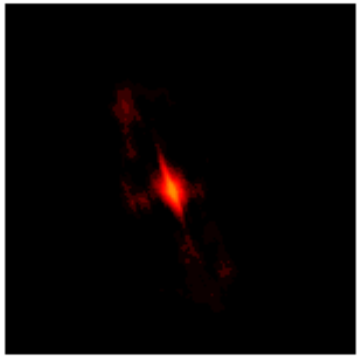
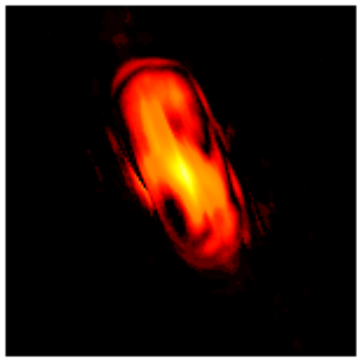
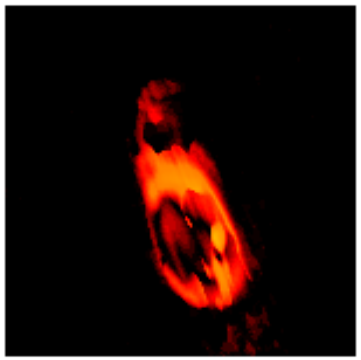
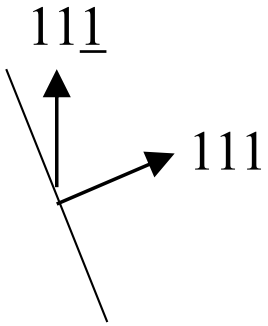
2D Reconstructions

chisquare = 0.0005

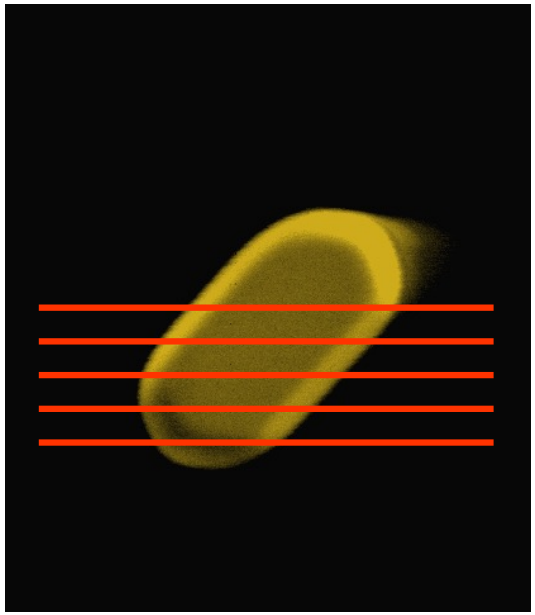




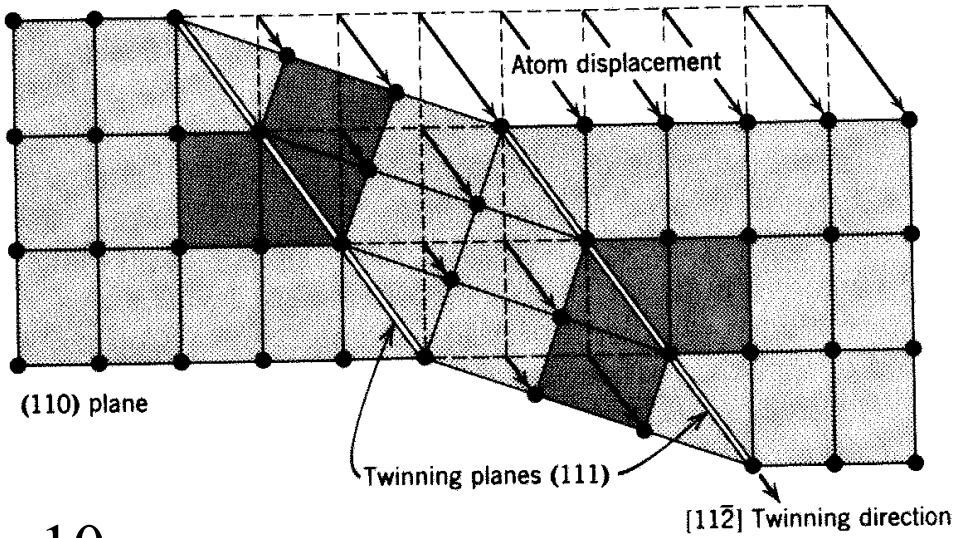










Slices through plan view SEM:



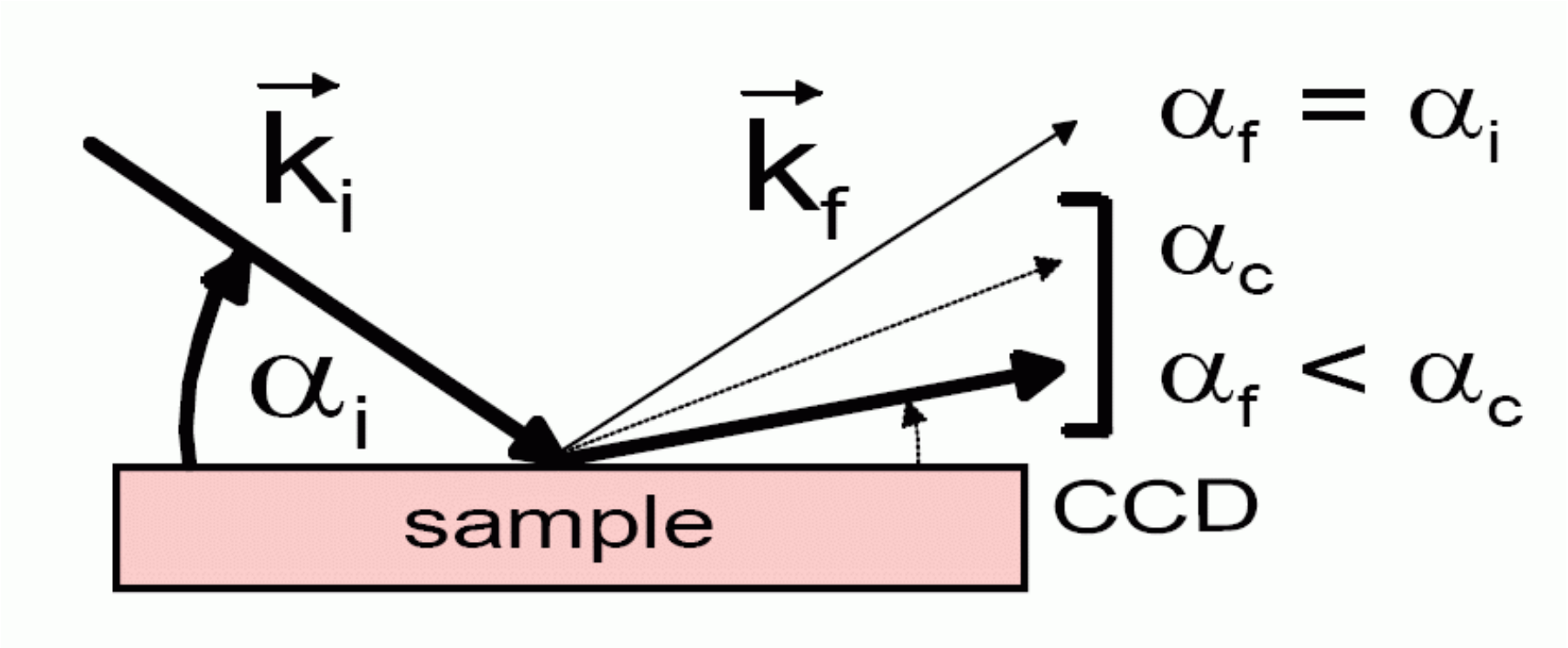
Twinning in deformed FCC metals

J. Wulff, "Structure and Property of Materials III" (1965)



~10μm
Cu

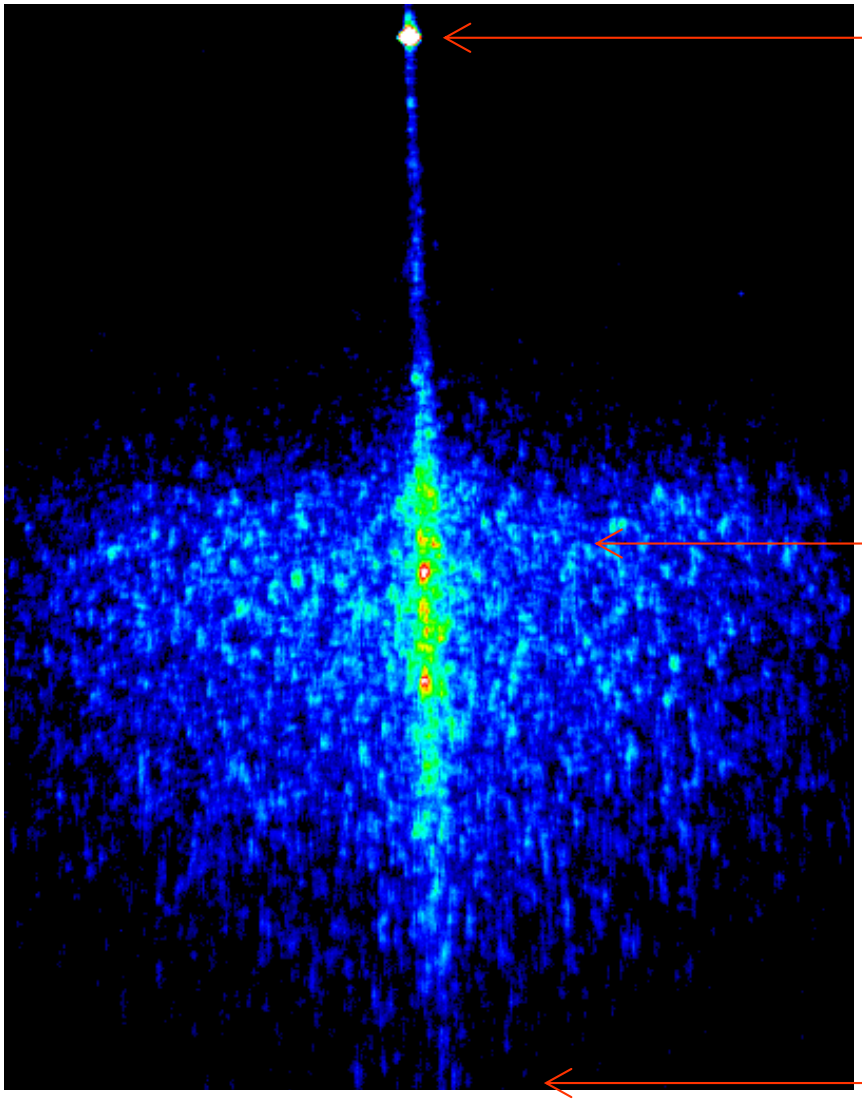
Coherent "Yoneda" sample geometry



F. Pfeiffer, W. Zhang and I. K. Robinson,
Applied Physics Letters 84 1847 (2004)

Structure in "Yoneda" Peak

Grazing-exit diffraction from a 1000A Au polycrystalline film

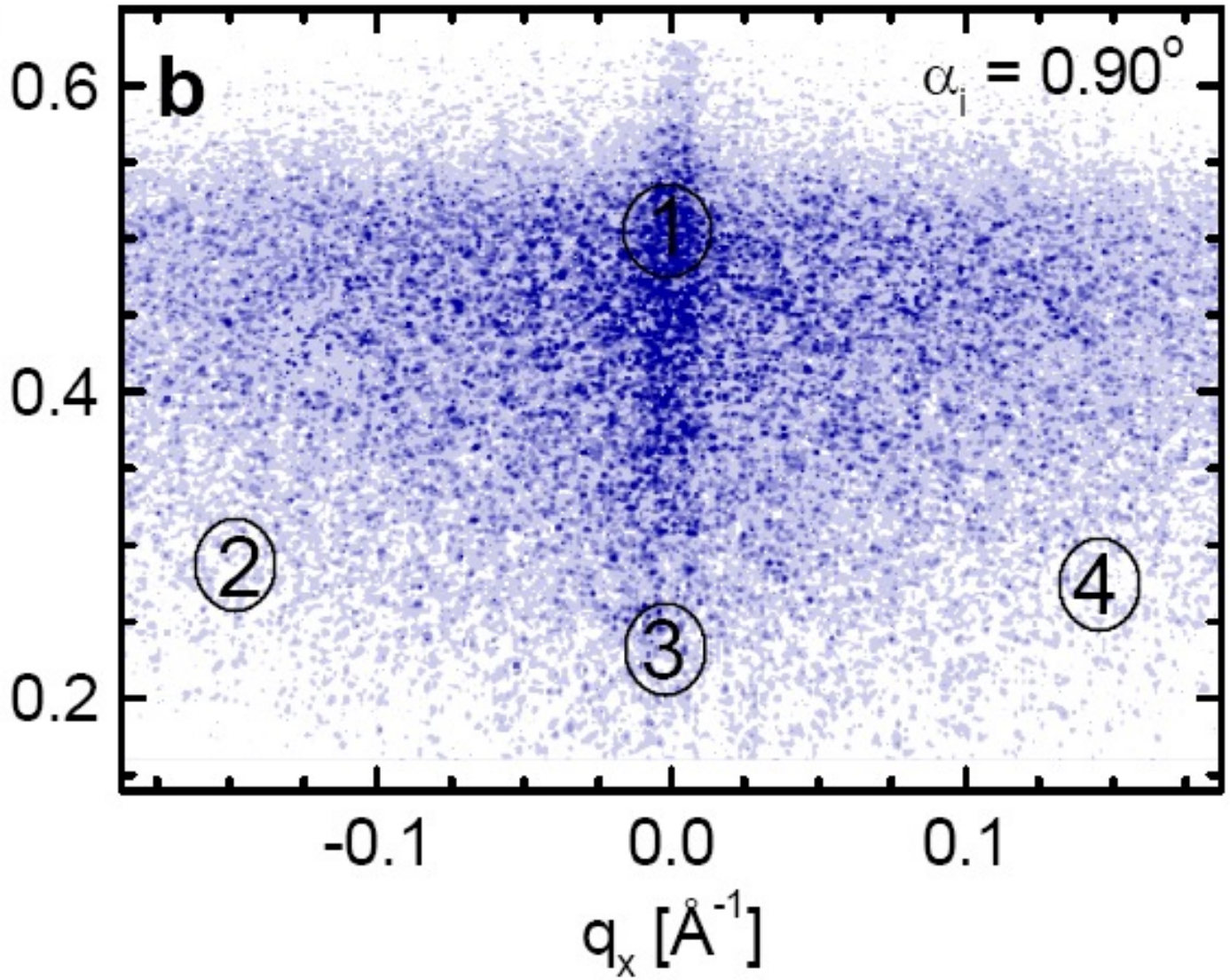


Specular ($\alpha_f \sim \alpha_i$)

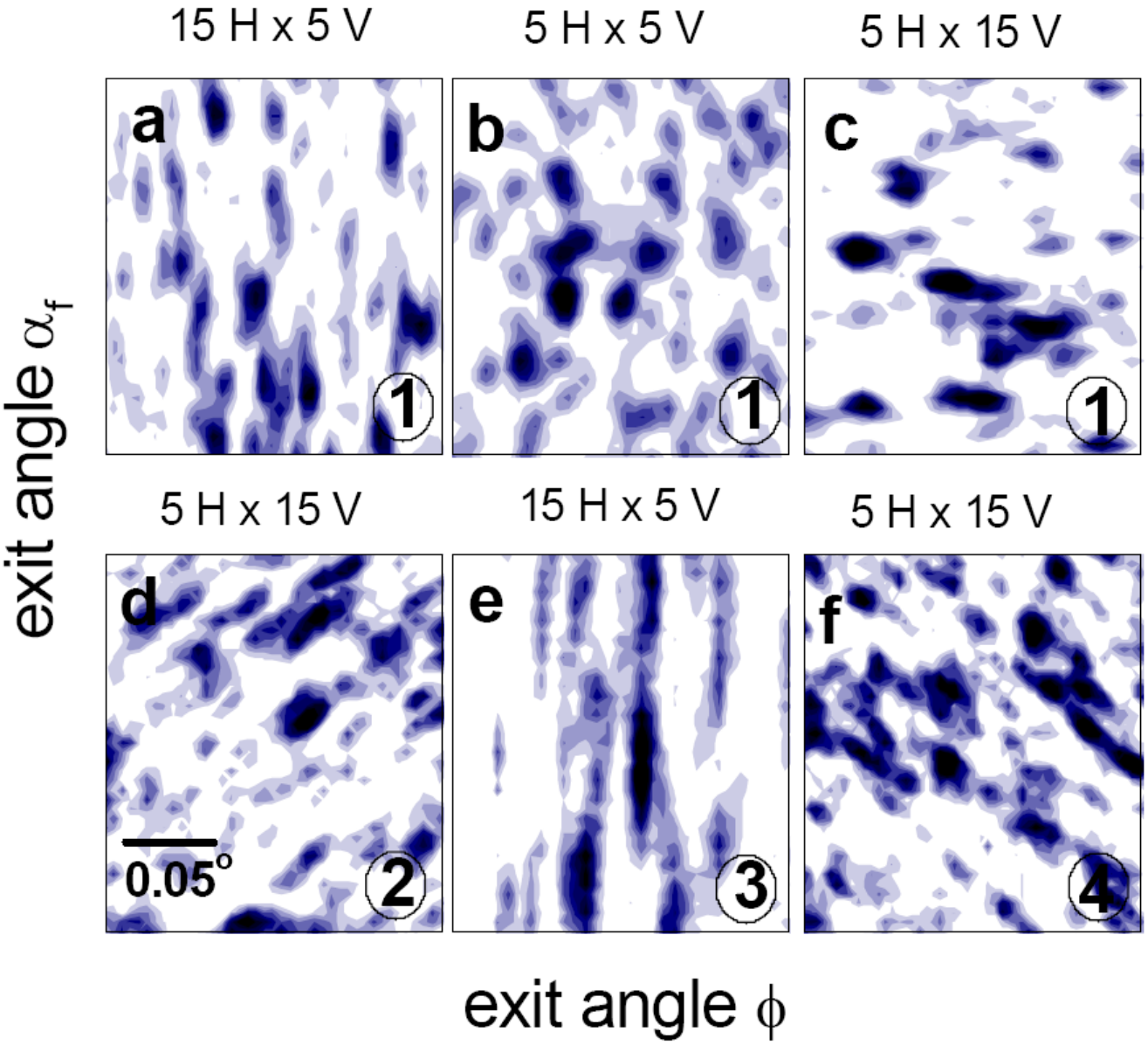
$\alpha_f \sim \alpha_c$

$\alpha_f \sim 0$

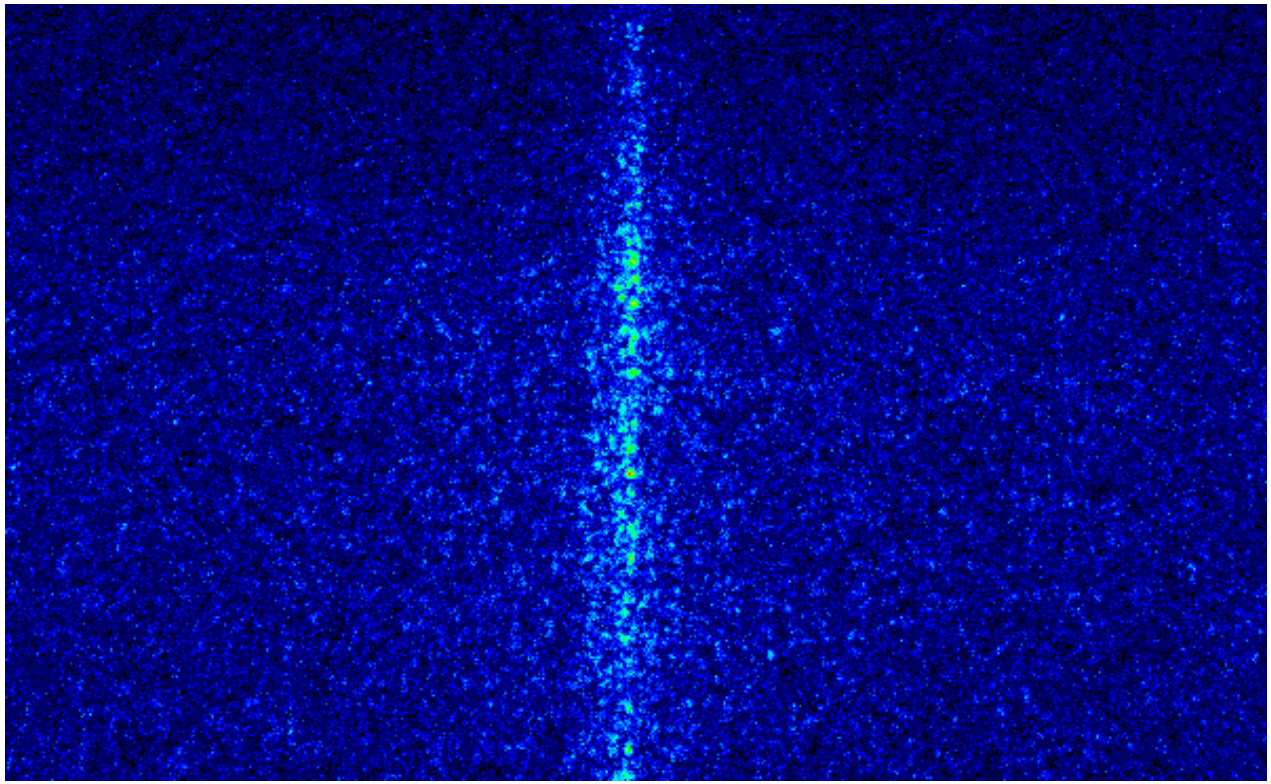
Anisotropy of speckles



Variation with entrance slit (microns)

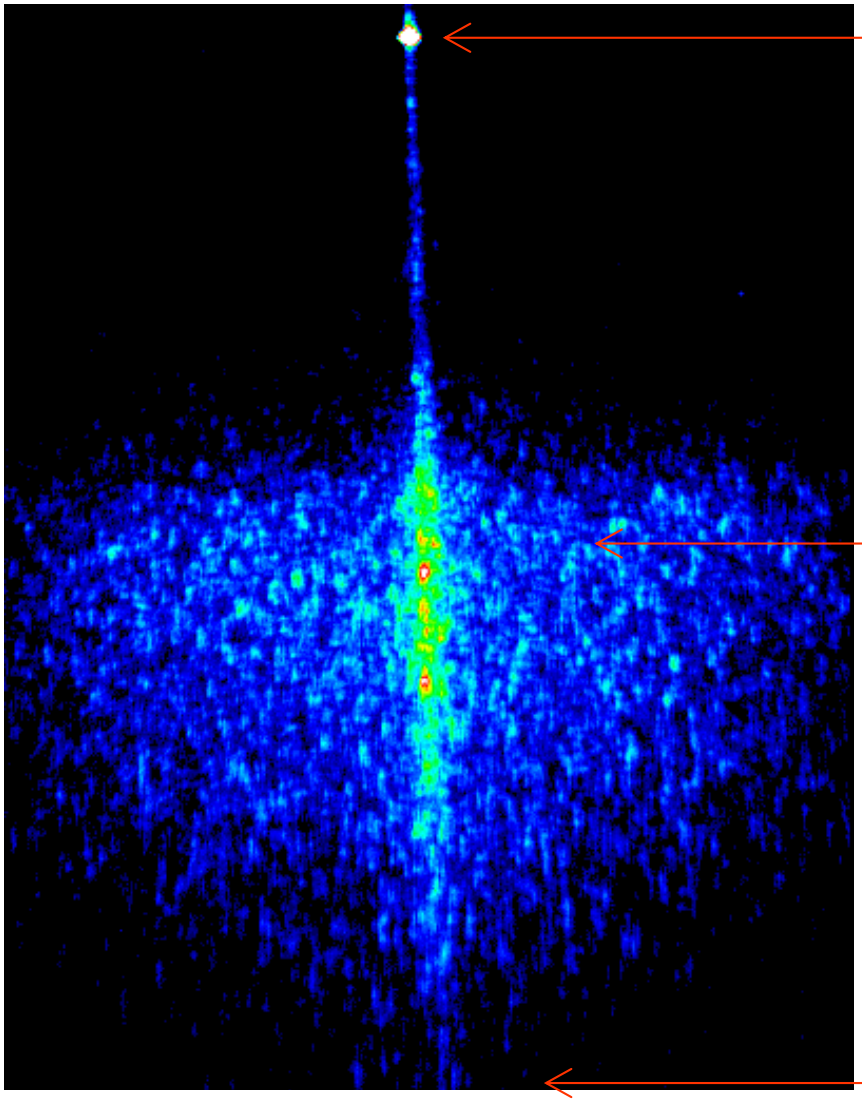


Angle series, 0.01° steps



Structure in "Yoneda" Peak

Grazing-exit diffraction from a 1000A Au polycrystalline film



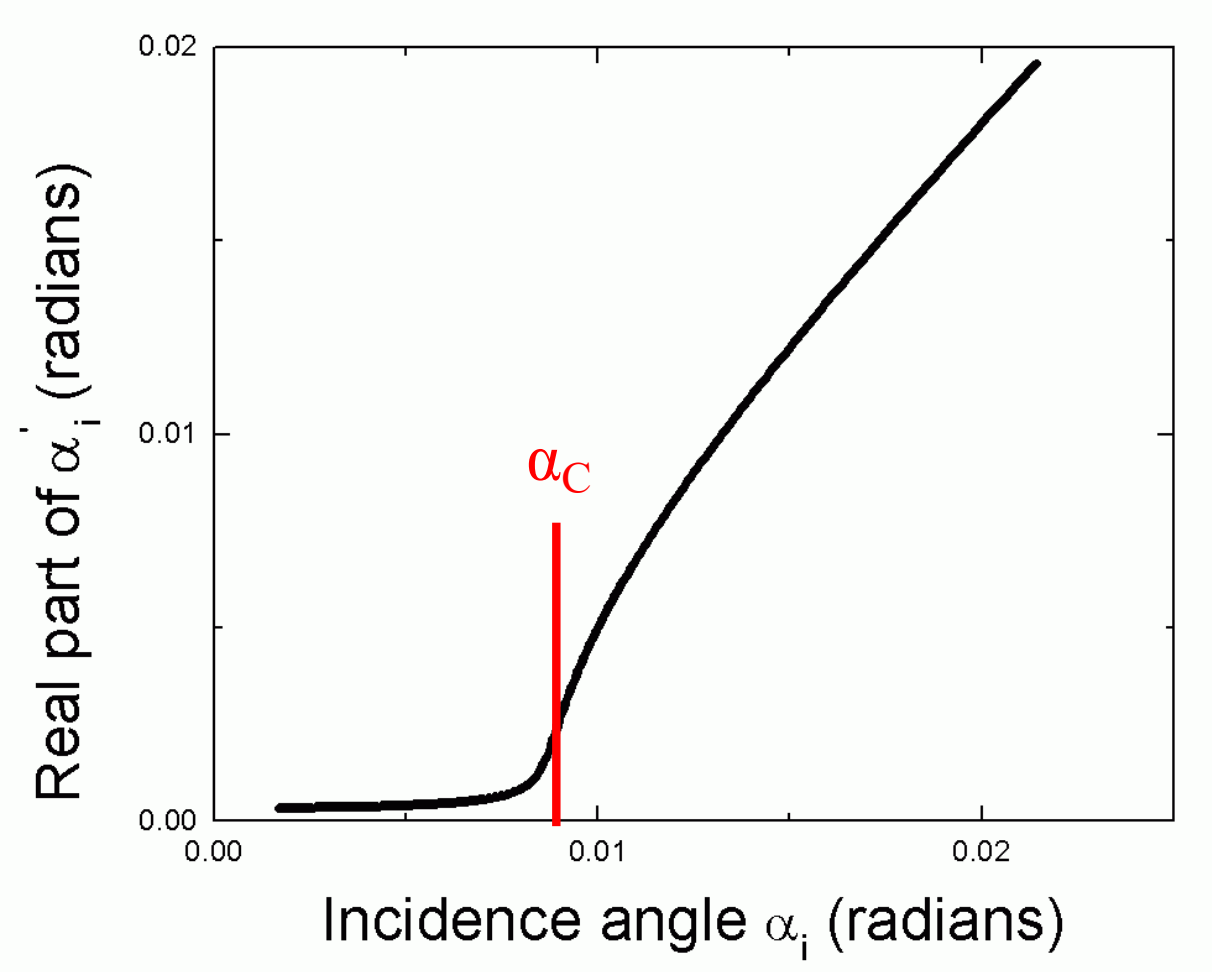
Specular ($\alpha_f \sim \alpha_i$)

$\alpha_f \sim \alpha_c$

$\alpha_f \sim 0$

Refraction by Gold Interface

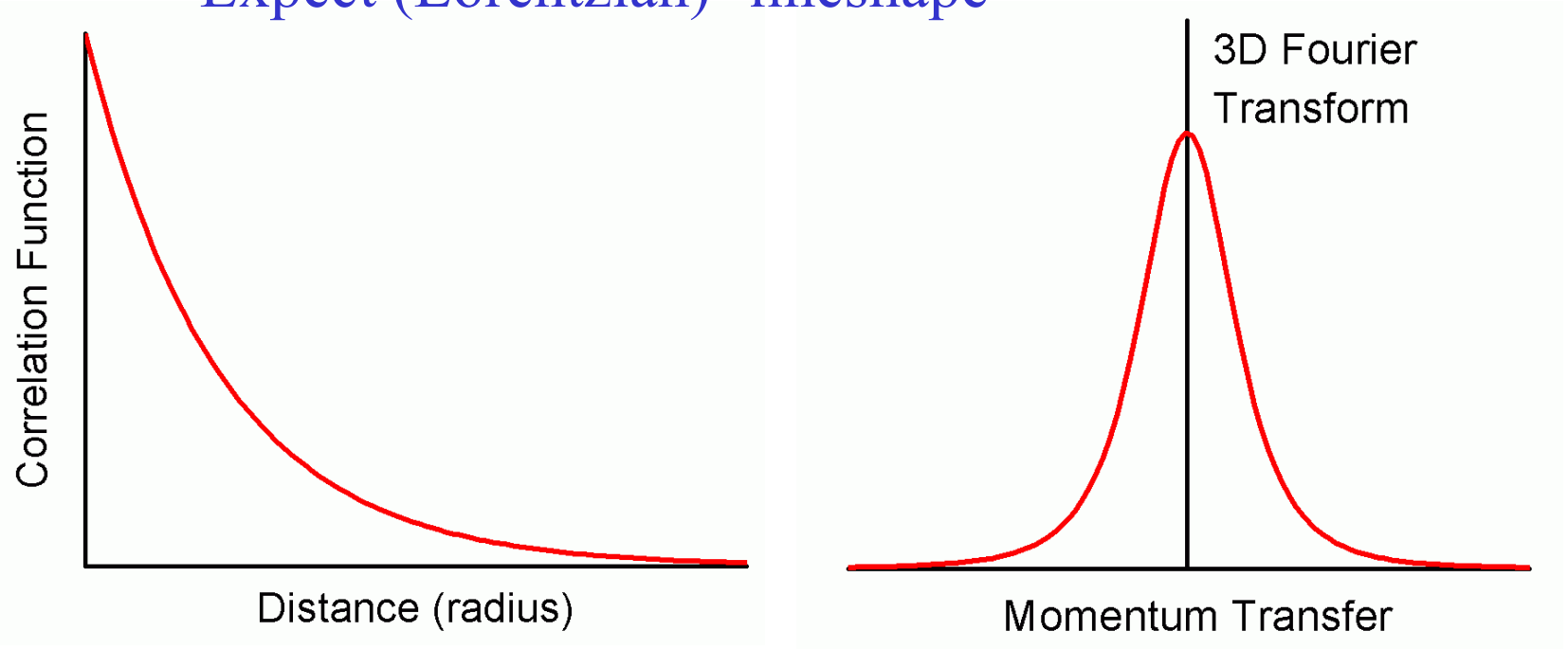
$$\alpha_i' = \sqrt{\alpha_i^2 - 2\delta + 2i\beta}$$



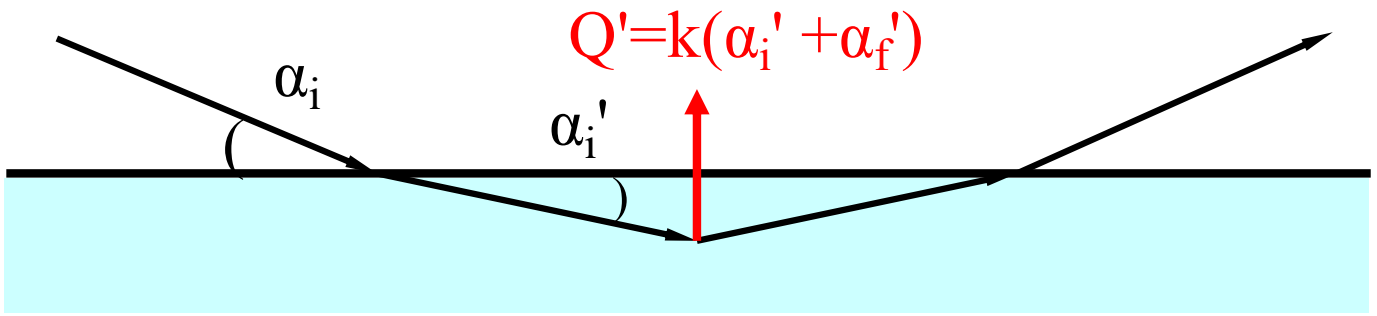
E=8.92keV
d=3.76×10^{-5}
β=2.9×10^{-6}

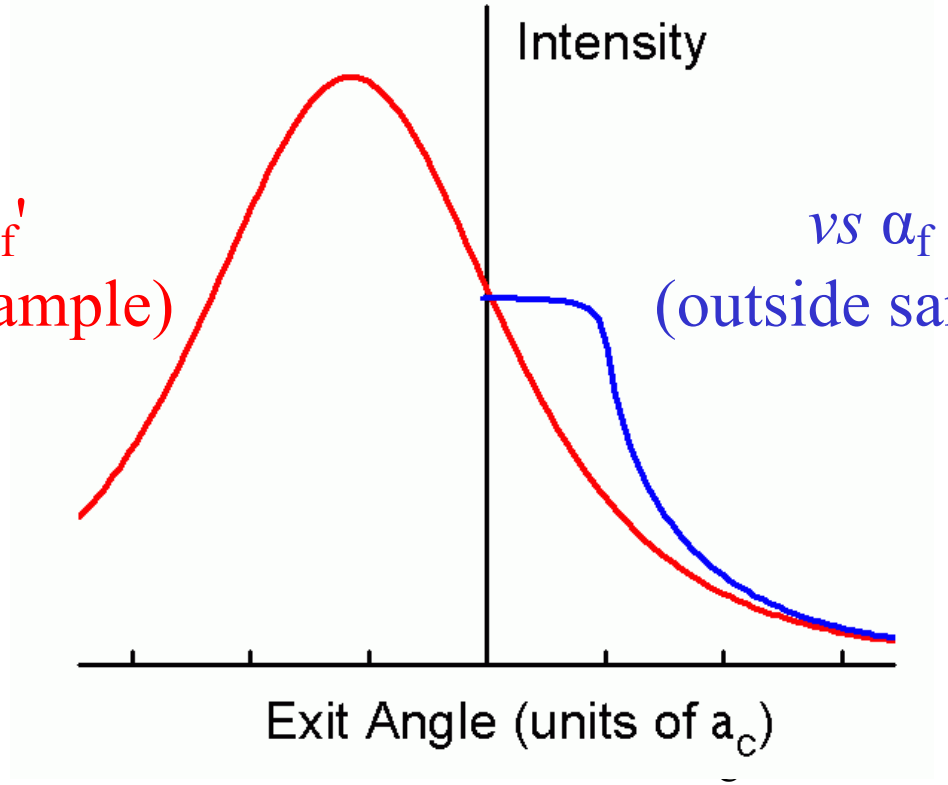
Model of grain correlations

- Density of film assumed uniform, except at location of grain boundaries
- Assume exponential size distribution in 3D
- Expect (Lorentzian)^2 lineshape



Effect of refraction on lineshape





Transmission Function

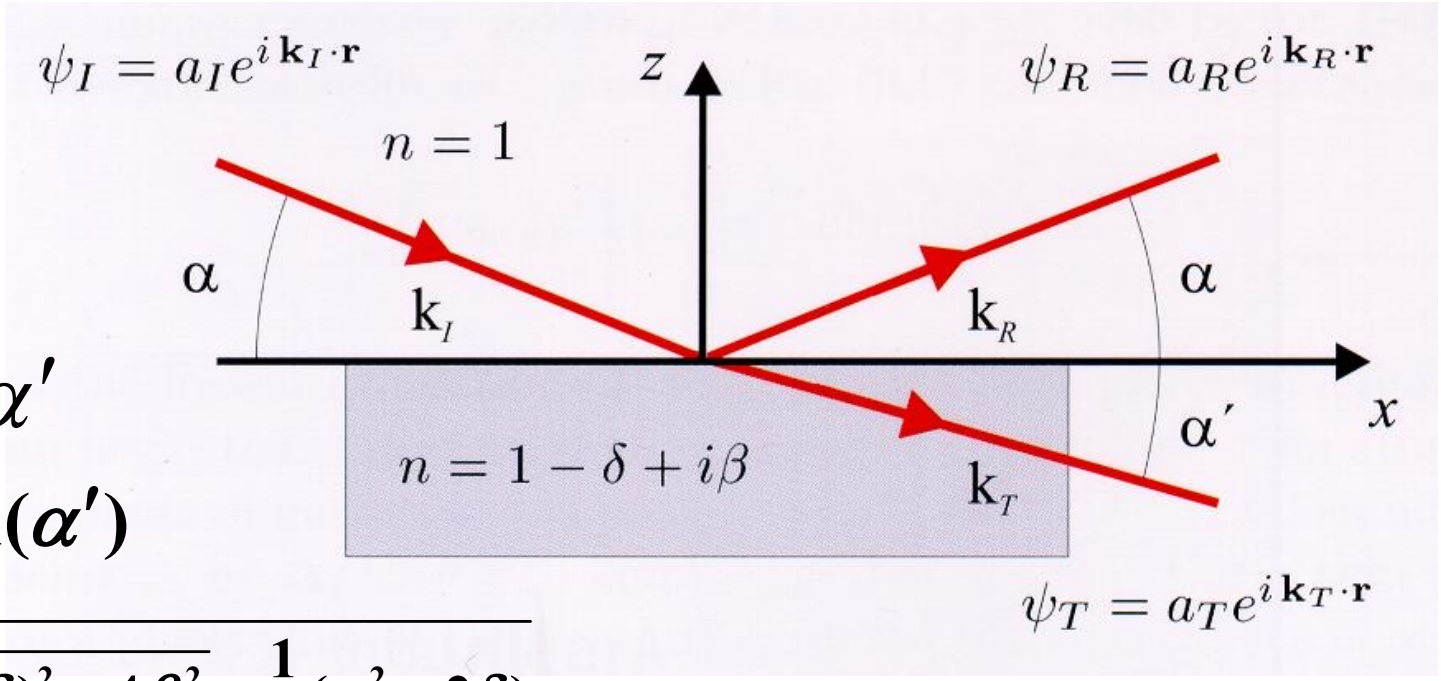
- Refractive index

$$n = 1 - \delta + i\beta$$

- Snell's law

$$\cos\alpha = n\cos\alpha'$$

$$\alpha' = \mathrm{Re}(\alpha') + \mathrm{Im}(\alpha')$$



$$\mathrm{Re}(\alpha') = \sqrt{\frac{1}{2}\sqrt{(\alpha^2 - 2\delta)^2 + 4\beta^2} + \frac{1}{2}(\alpha^2 - 2\delta)}$$

$$\mathrm{Im}(\alpha') = \sqrt{\frac{1}{2}\sqrt{(\alpha^2 - 2\delta)^2 + 4\beta^2} - \frac{1}{2}(\alpha^2 - 2\delta)}$$

- Amplitude Transmittivity

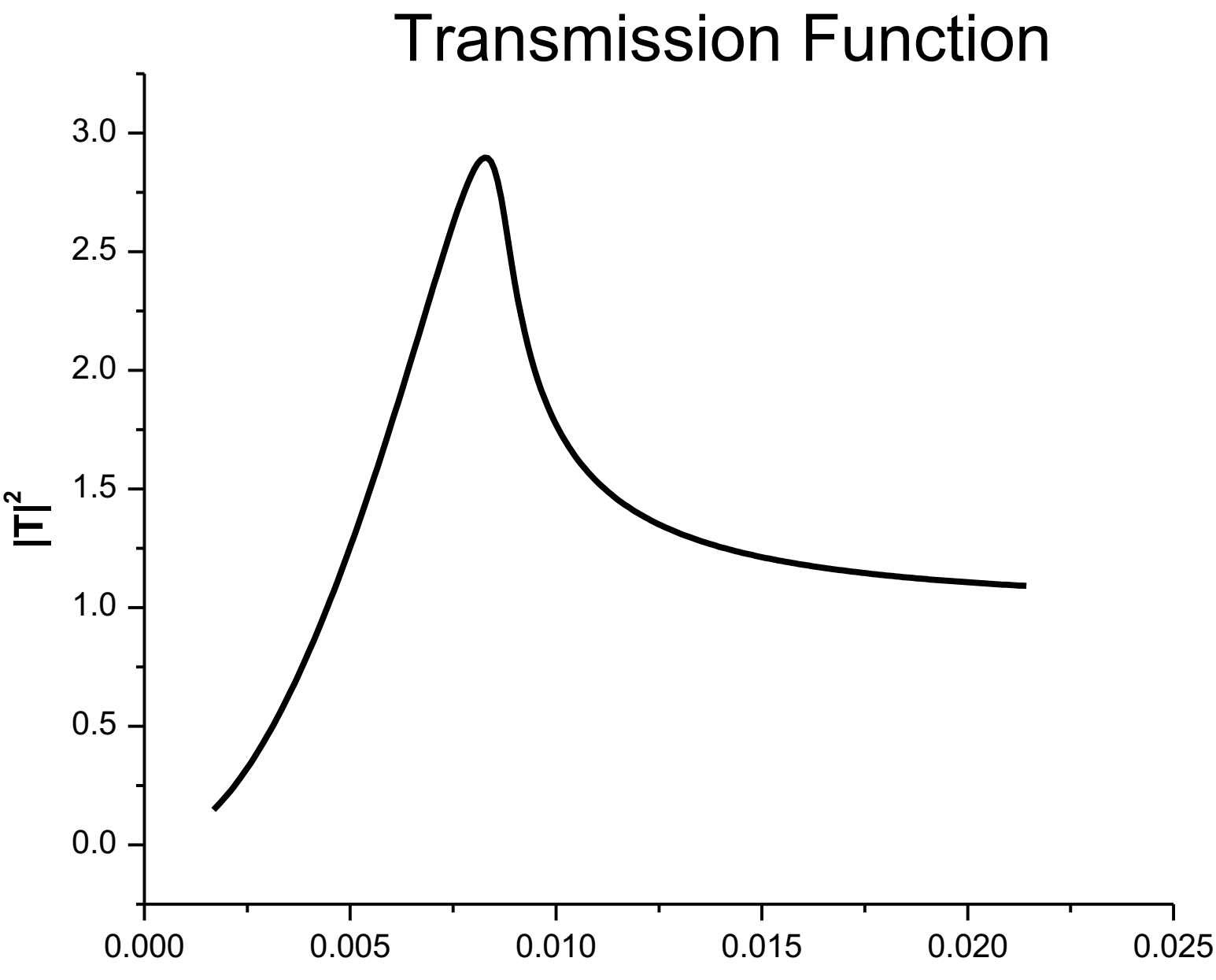
$$T = \frac{\alpha_T}{\alpha_I} = \frac{2\alpha}{\alpha + \alpha'}$$

Incidence

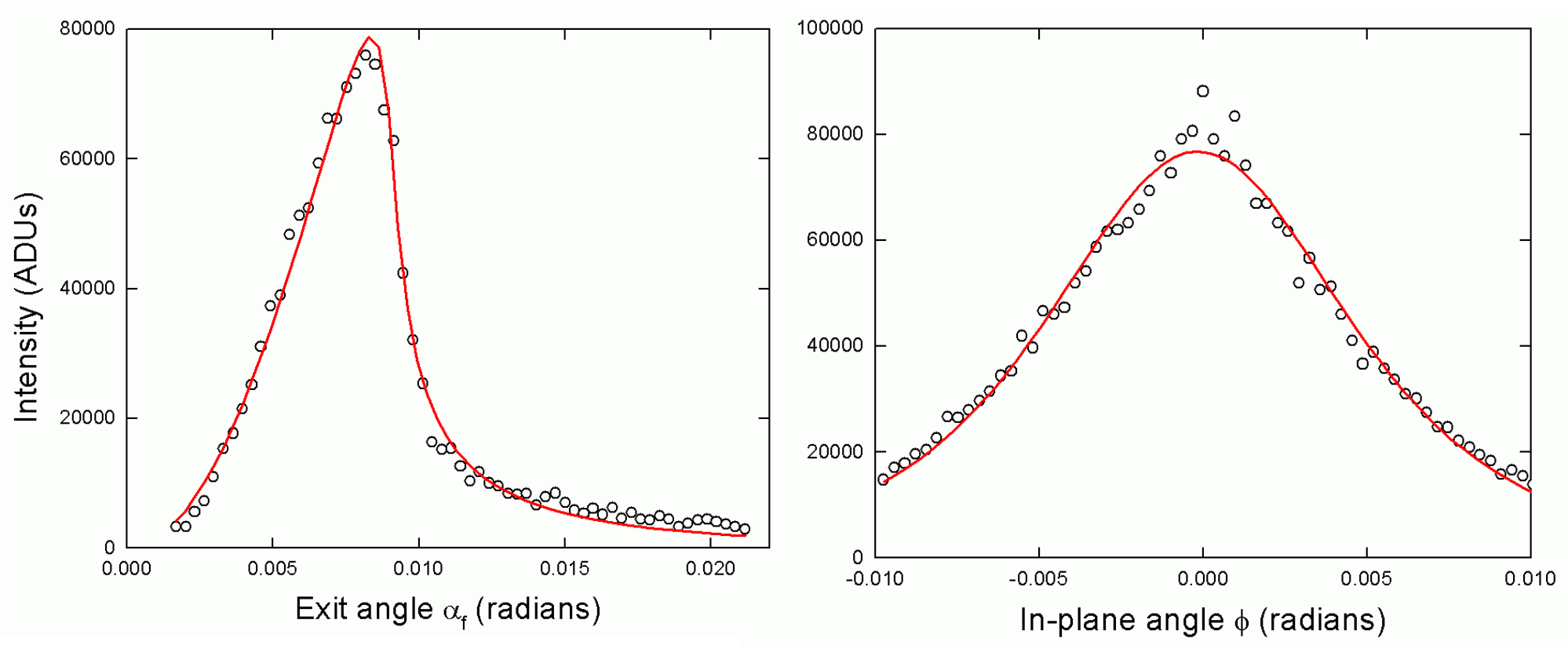
$$T(\vec{k}_1) = \frac{2\alpha_i}{\alpha_i + \alpha_i'} = const$$

Exit

$$T(\vec{k}_2) = \frac{2\alpha_f}{\alpha_f + \alpha_f'}$$

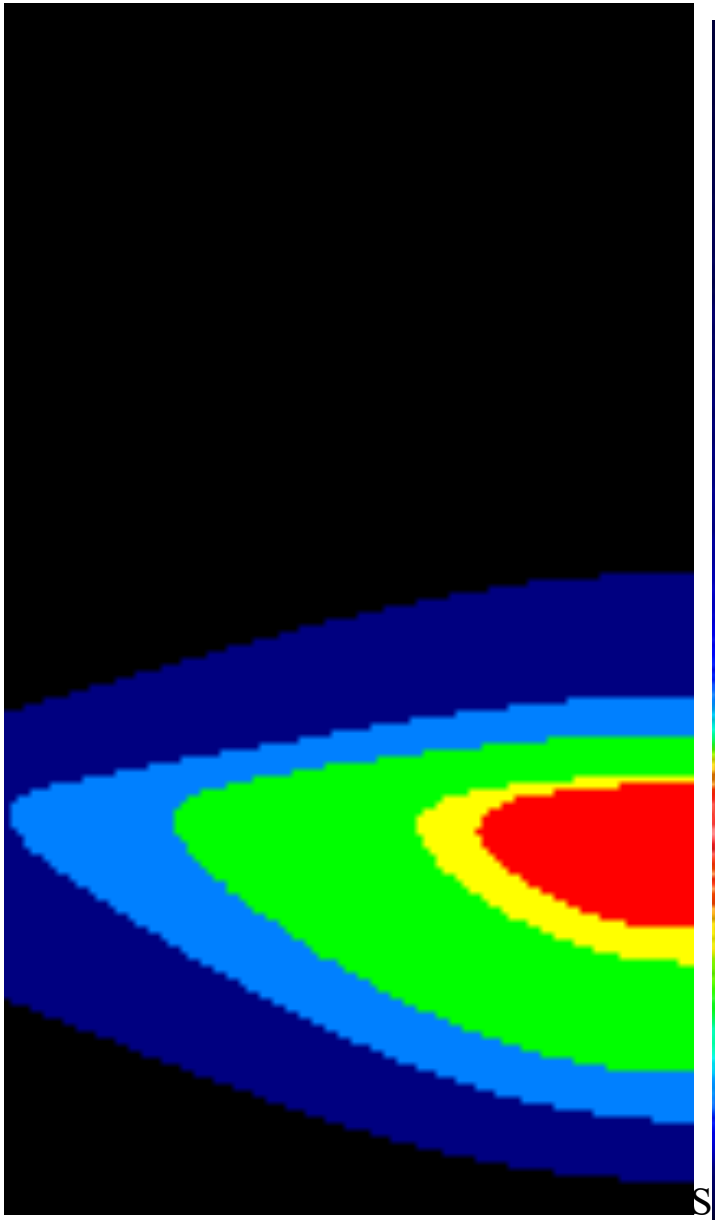

Transmission Function
3.0
2.5
2.0
1.5
1.0
0.5
0.0
|T|²
0.000
0.005
0.010
0.015
0.020
0.025
α

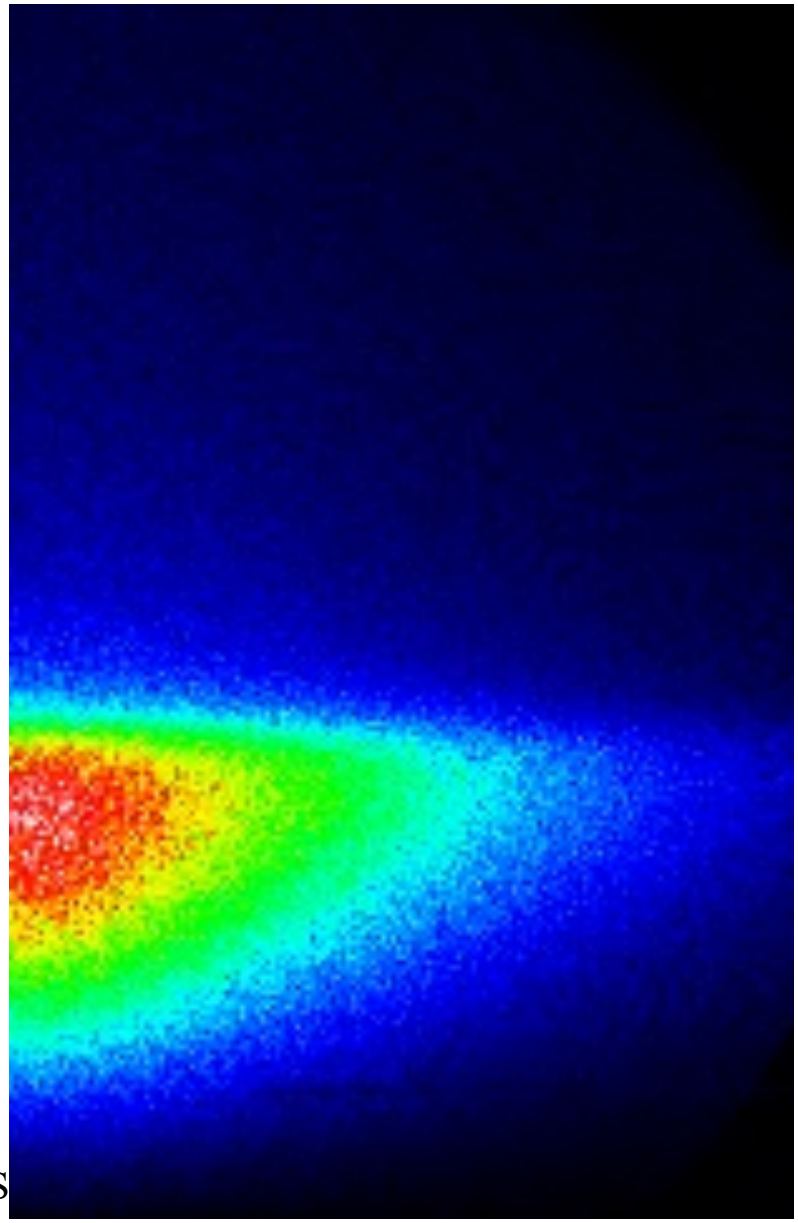
Fits to (Lorentzian)^2 Profiles

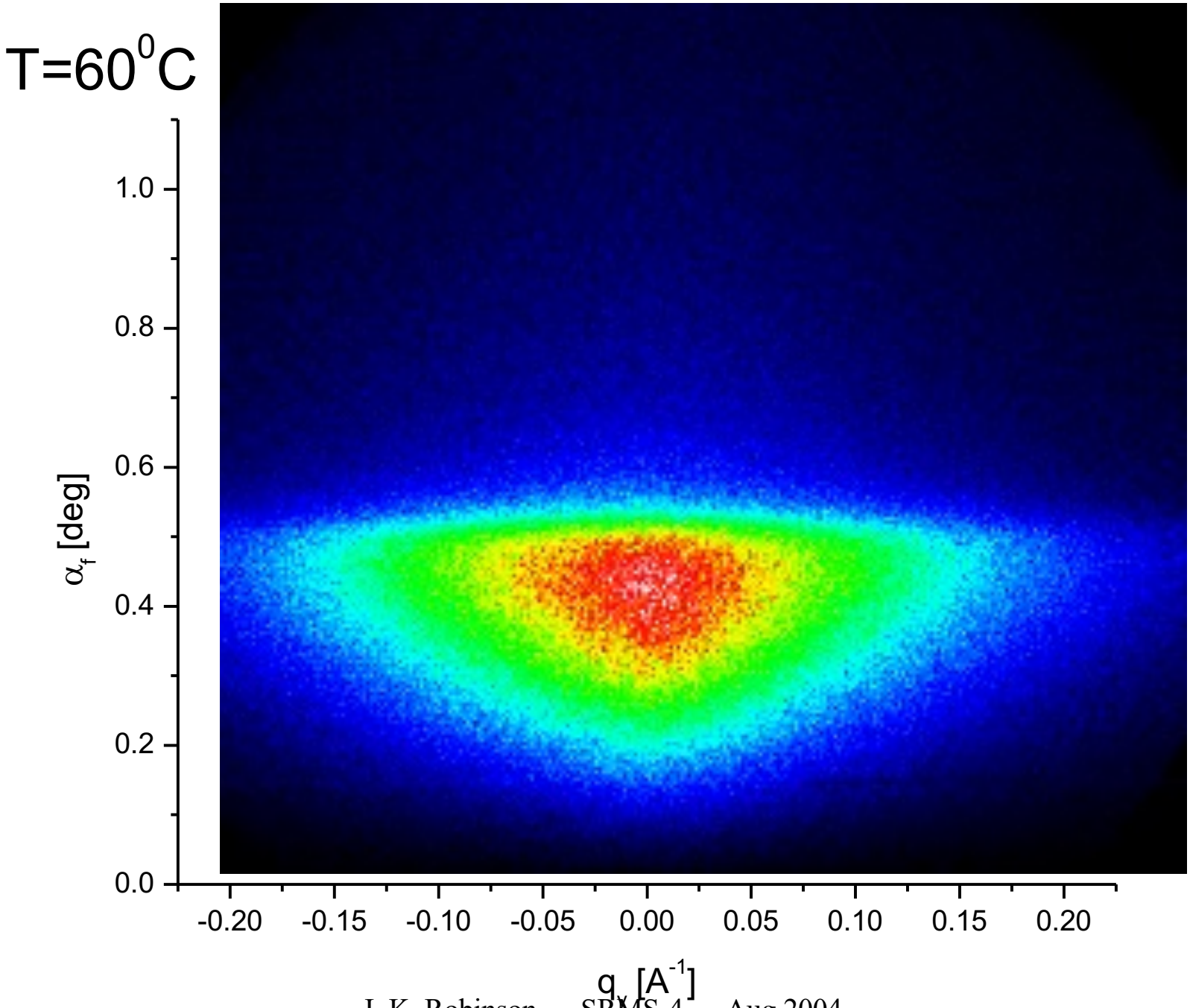


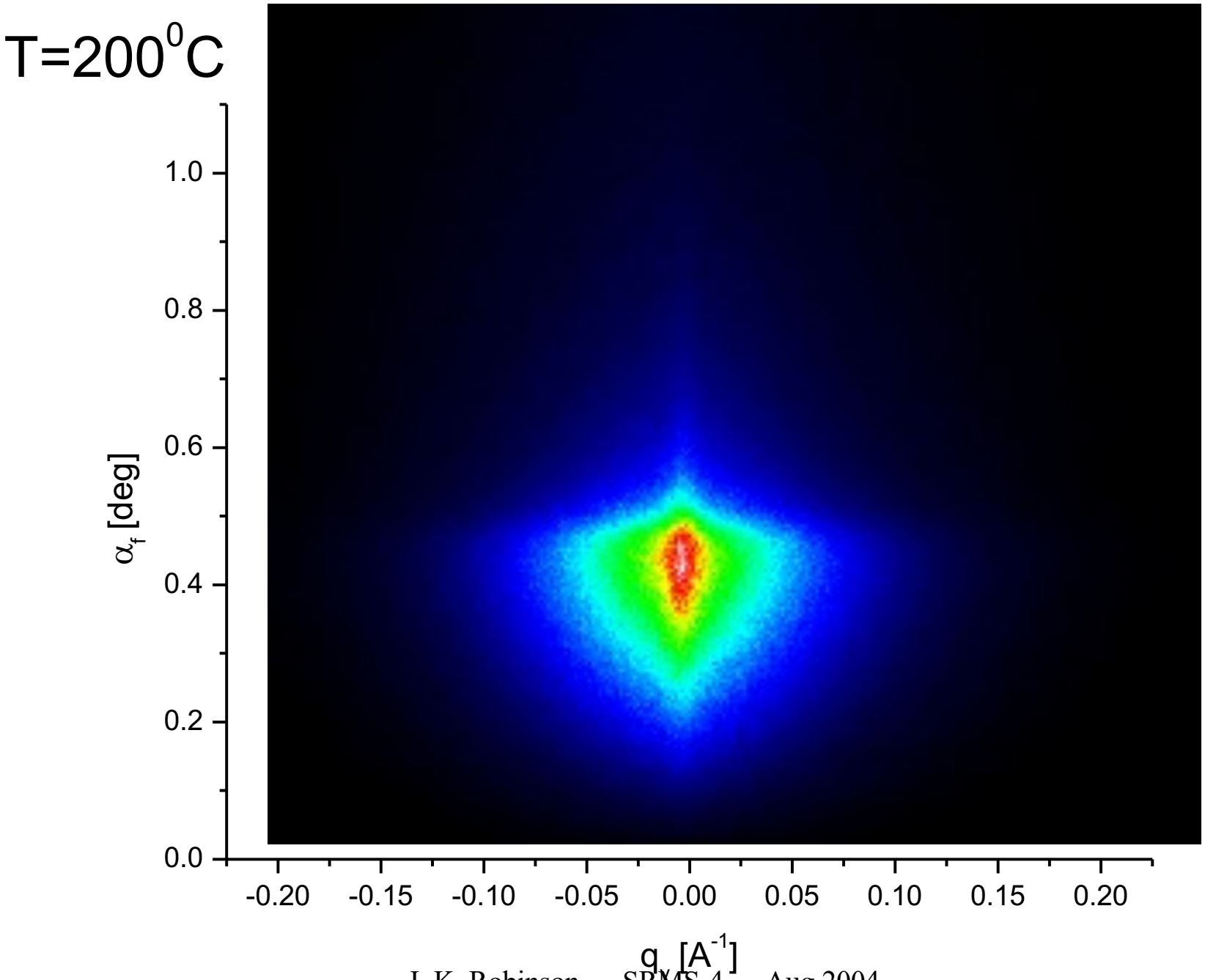
Fit params: center (x,y), amplitude, width=0.0084, α_i=0.011

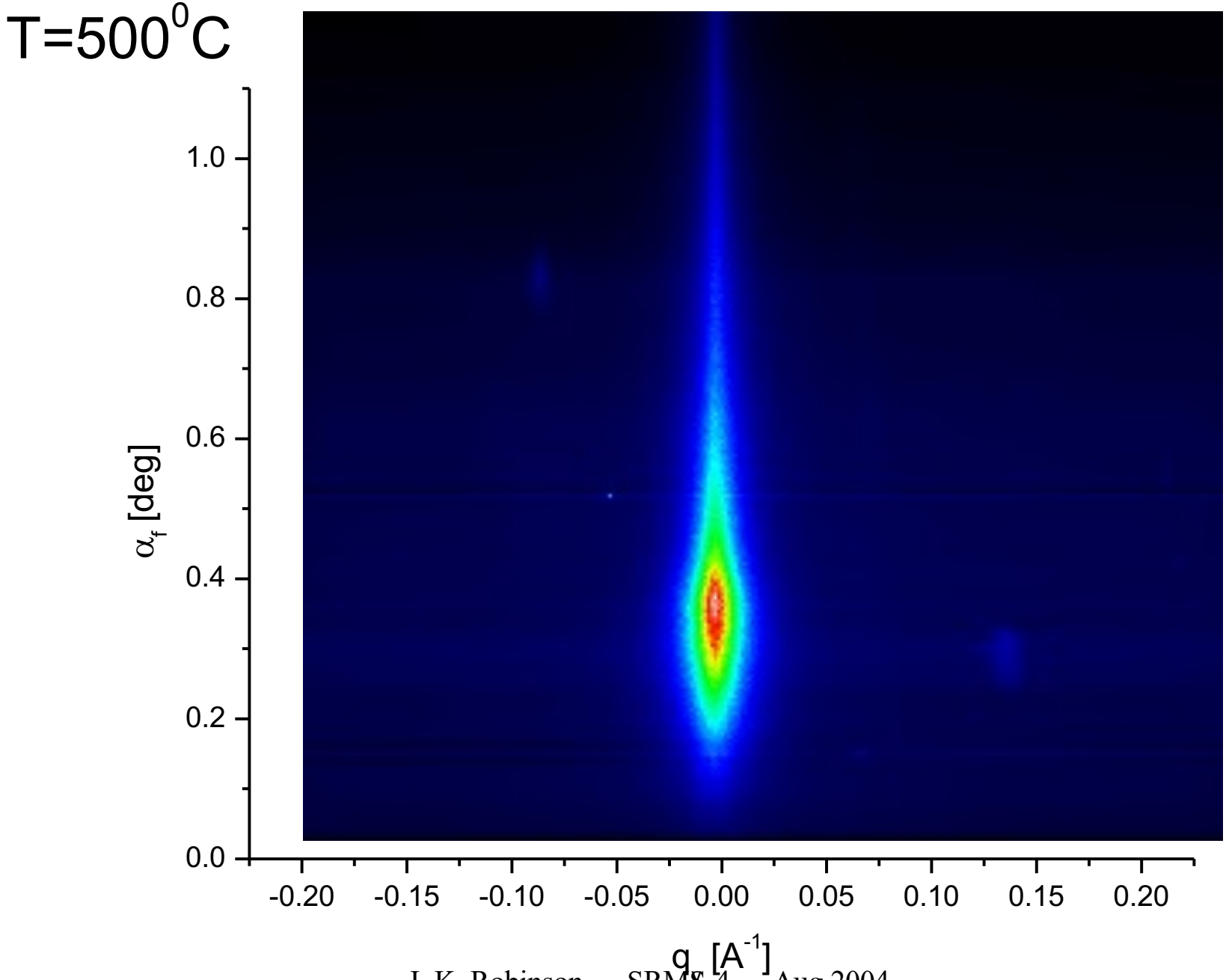
Simulations of CCD data



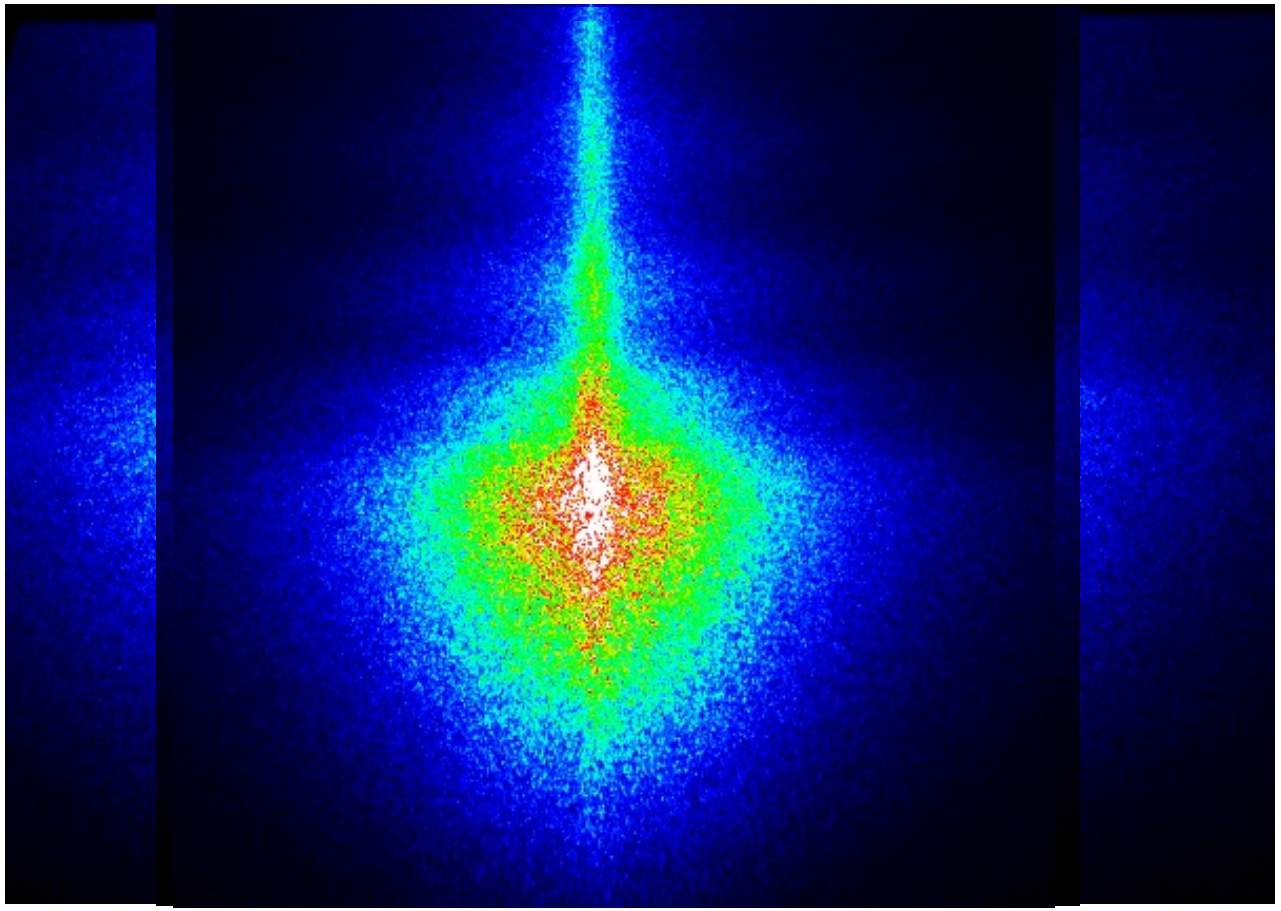



T=60^{0}C
1.0
0.8
0.6
0.4
0.2
0.0
α_f [deg]
-0.20
-0.15
-0.10
-0.05
0.00
0.05
0.10
0.15
0.20
q_x [A^{-1}]


T=200^{0}C
1.0
0.8
0.6
0.4
0.2
0.0
α_f [deg]
-0.20
-0.15
-0.10
-0.05
0.00
0.05
0.10
0.15
0.20
q_x [A^{-1}]


T=500^{0}C
1.0
0.8
0.6
0.4
0.2
0.0
α_f [deg]
-0.20
-0.15
-0.10
-0.05
0.00
0.05
0.10
0.15
0.20
q_z [A^{-1}]

Annealing Fe(110) film at 350C



Conclusions and Future Outlook

- GISAXS explained by refraction + $|T(\alpha)|$
- CGISAXS sees individual grain distribution
- Surface effects are relatively unimportant
- Invert diffraction pattern to an image
- Study evolution grain by grain
- Other metals, kinds of film
- Island to film transition