Reflectometry at the SNS

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Spallation Neutron Source Project
Oak Ridge National Laboratory

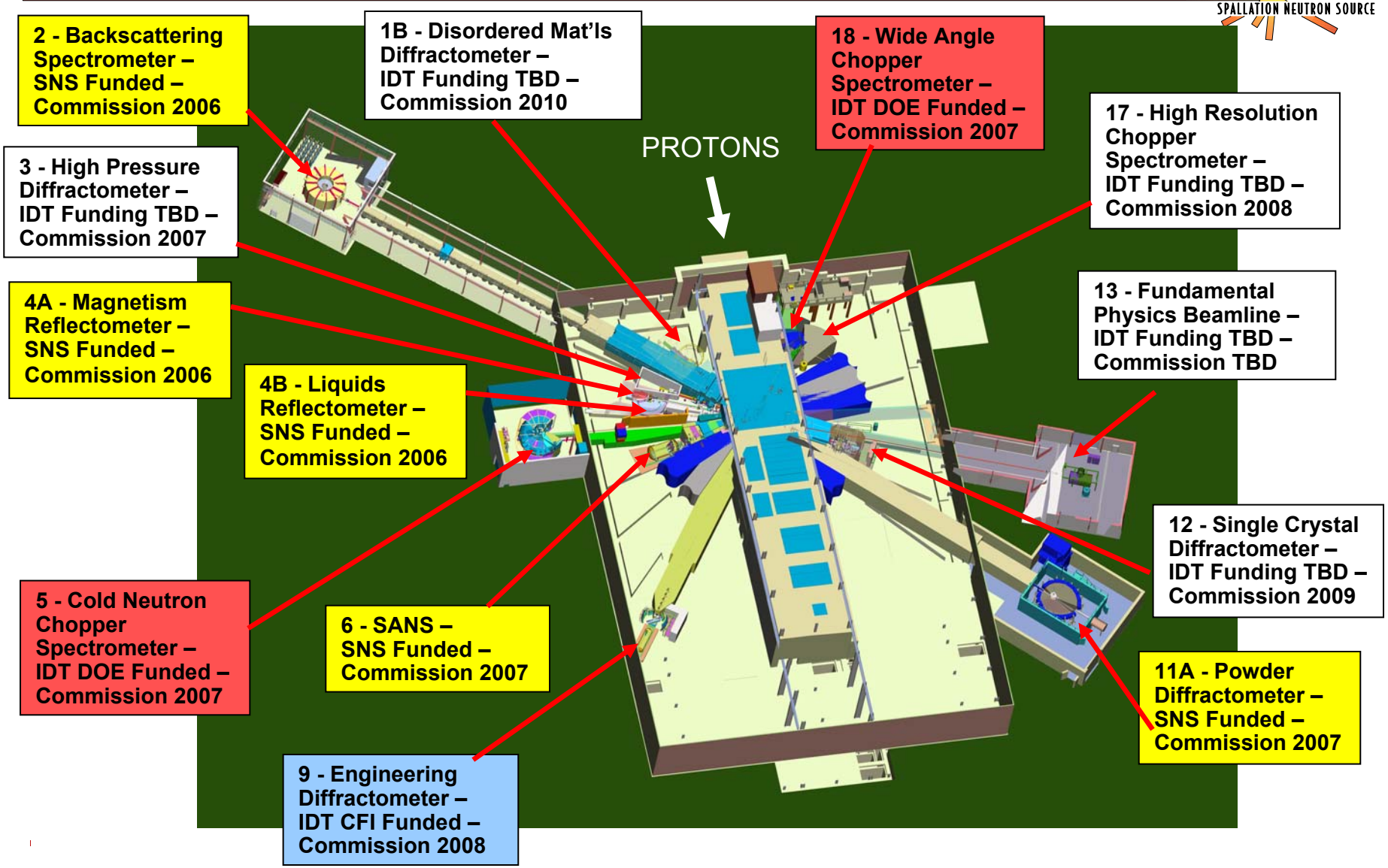
Structural Biology Task Force Meeting
Argonne, October 24, 2002

Outline

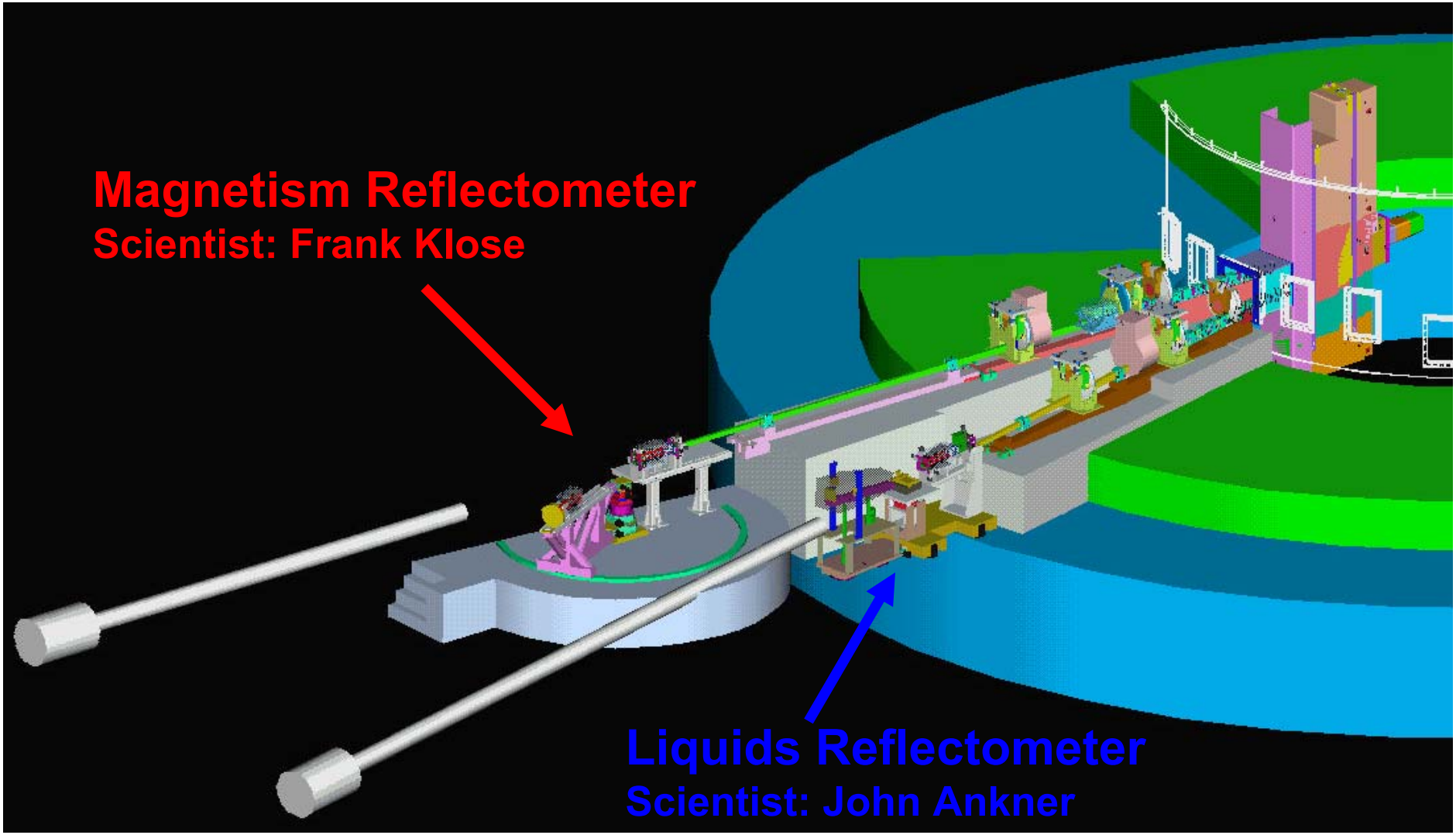
- Status of the SNS Reflectometers
- New Science:
  - Time dependent measurements
  - Measurements on lateral structures

Target Building with Neutron Scattering Instruments

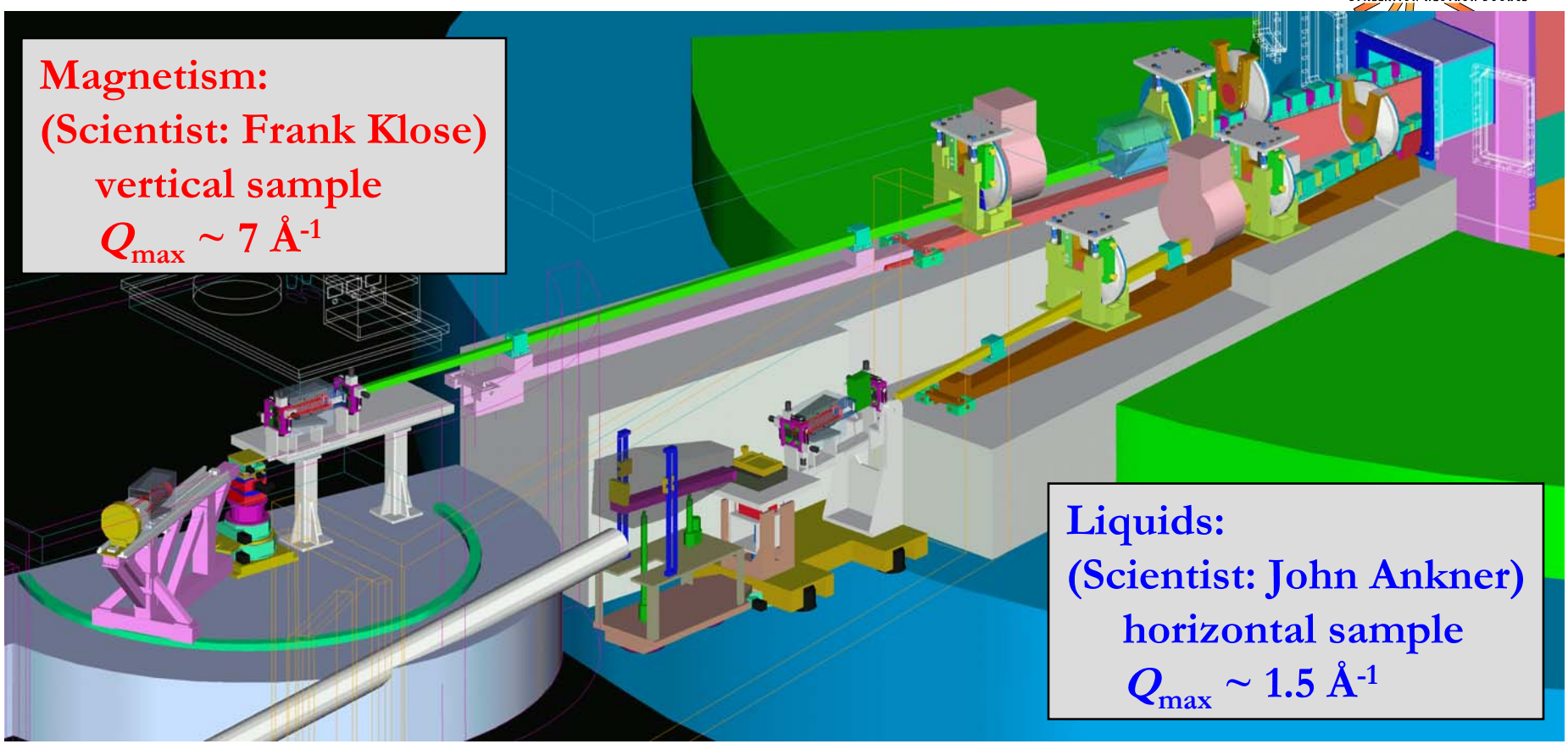
(8 instruments funded, 4 more in design funding phase)



SNS Reflectometers



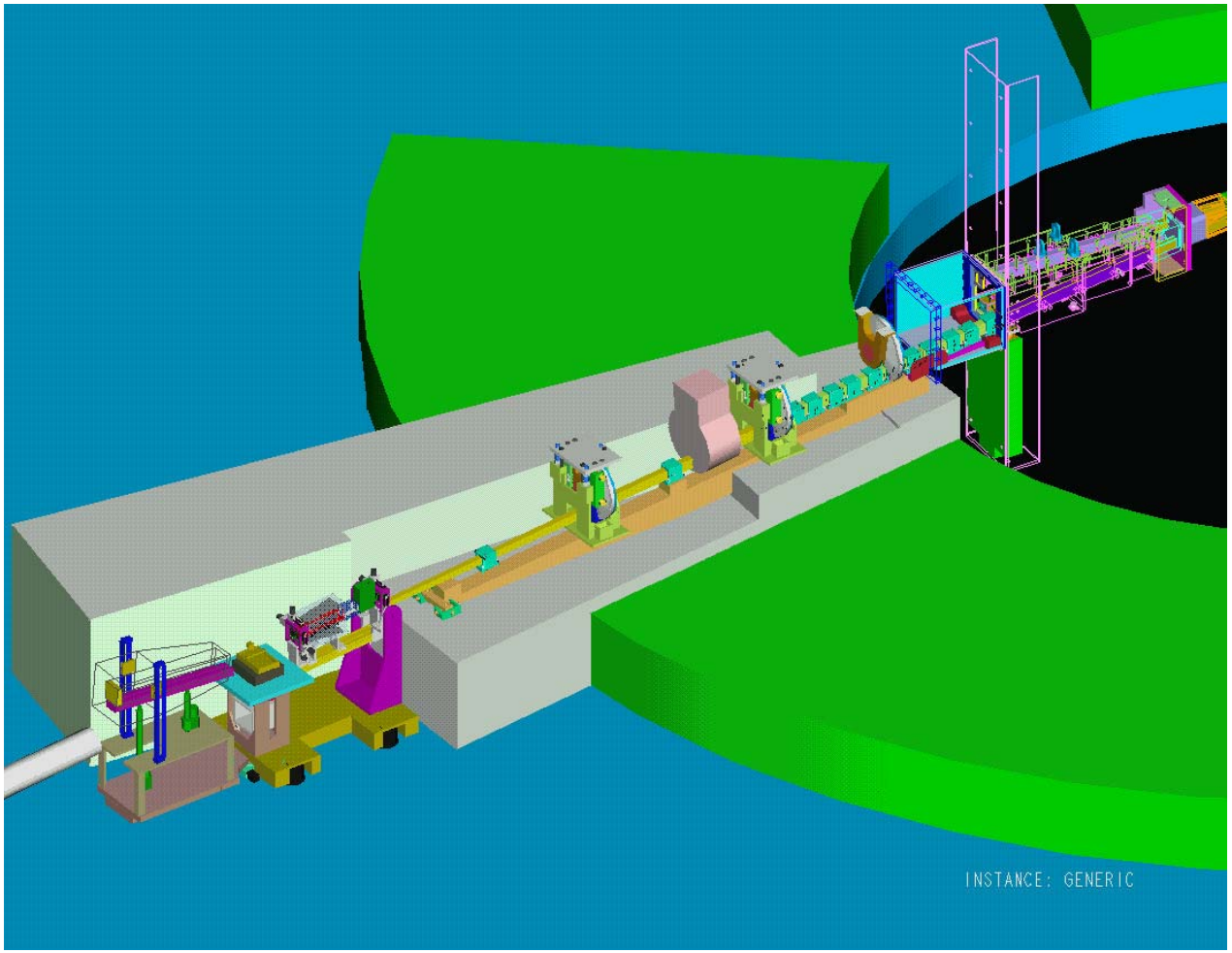
SNS Reflectometers



- 2 reflectometers sharing a single beamport
- Allows for both vertical sample (magnetism) and horizontal sample (liquids) studies

- $R_{min} < 5\times10^{-10}$
- Sensitivity: Monolayers
- 50 -100 × NIST NG-1

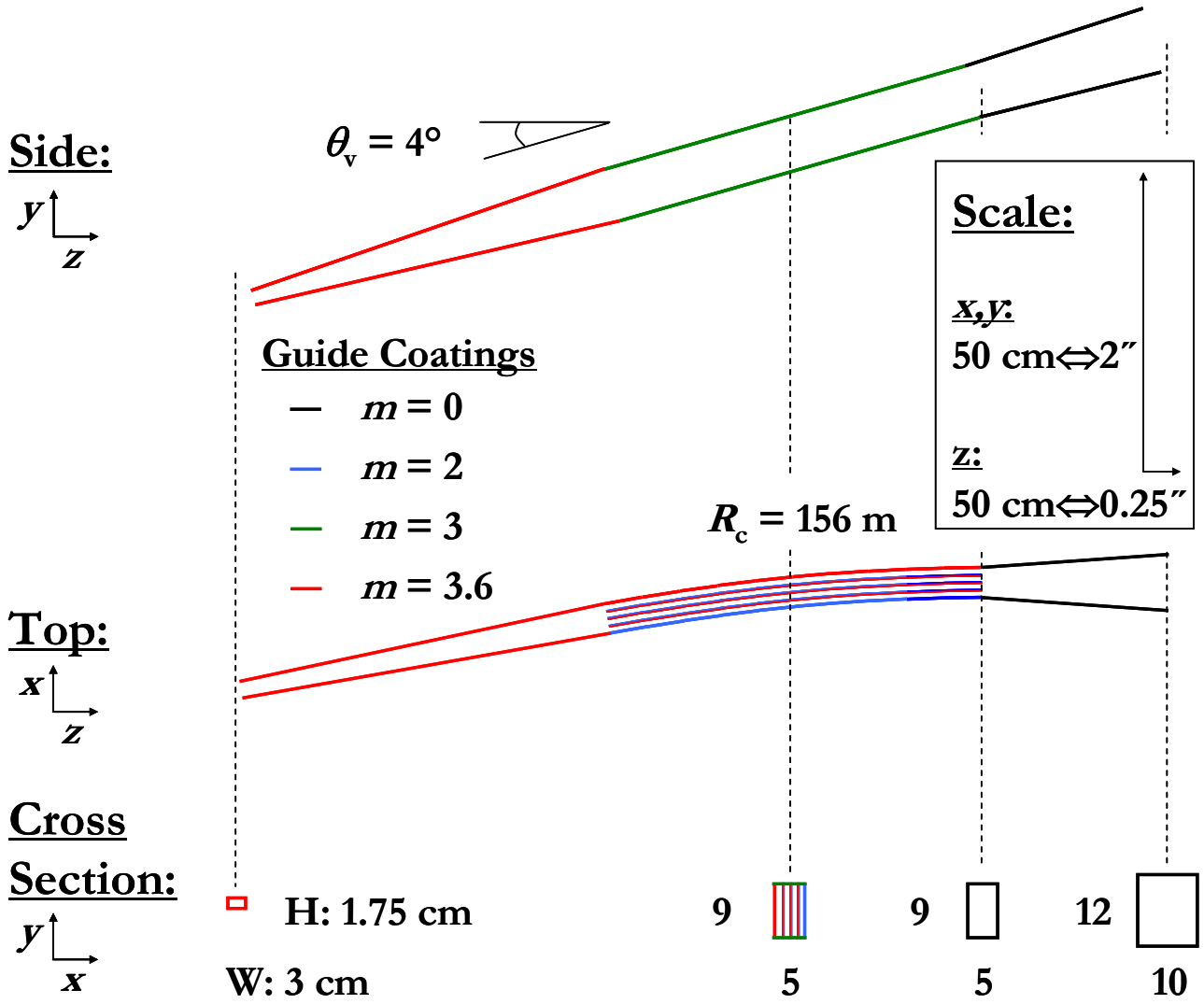
Liquids Reflectometer Concept



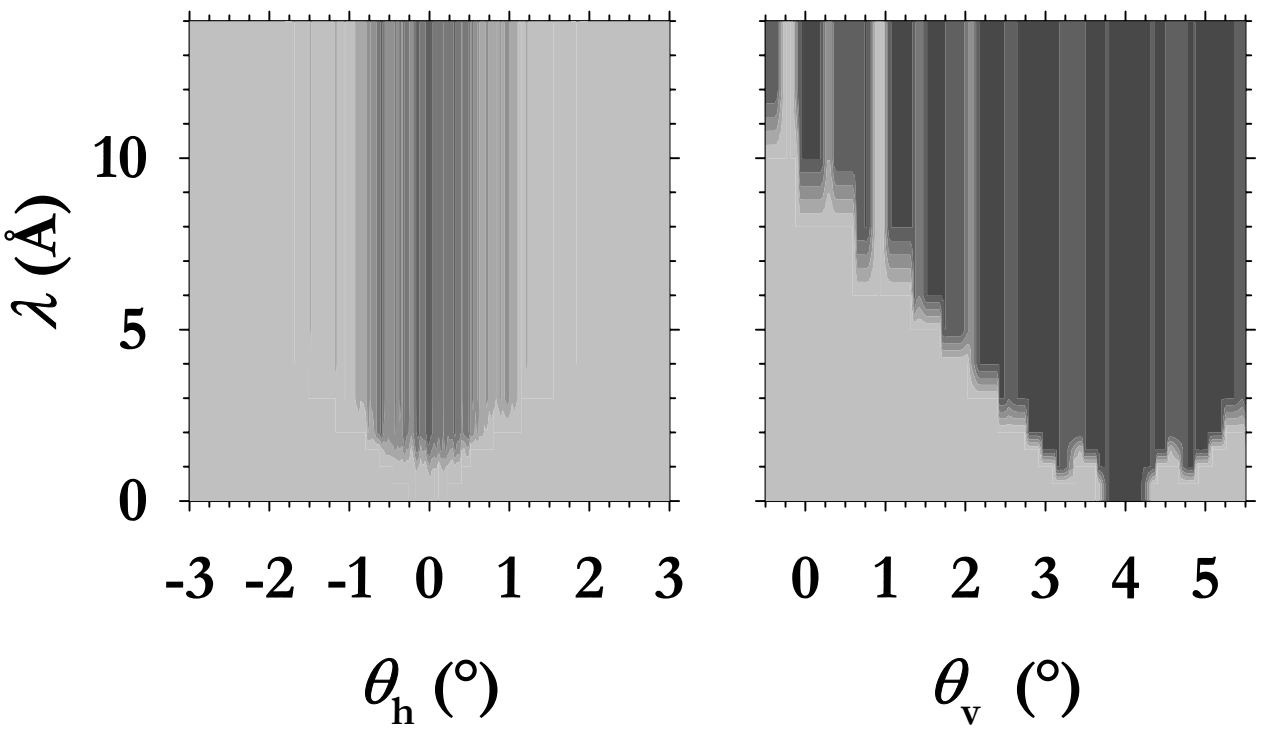
Optics Highlights

- 4° downward angle
- Multi-channel bender
- Tapered funnel

Incident Optics of the Liquids Reflectometer

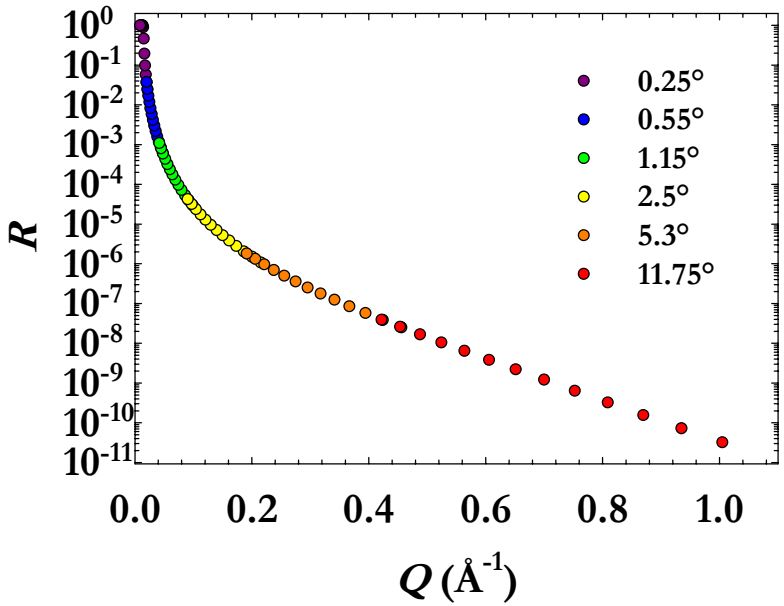


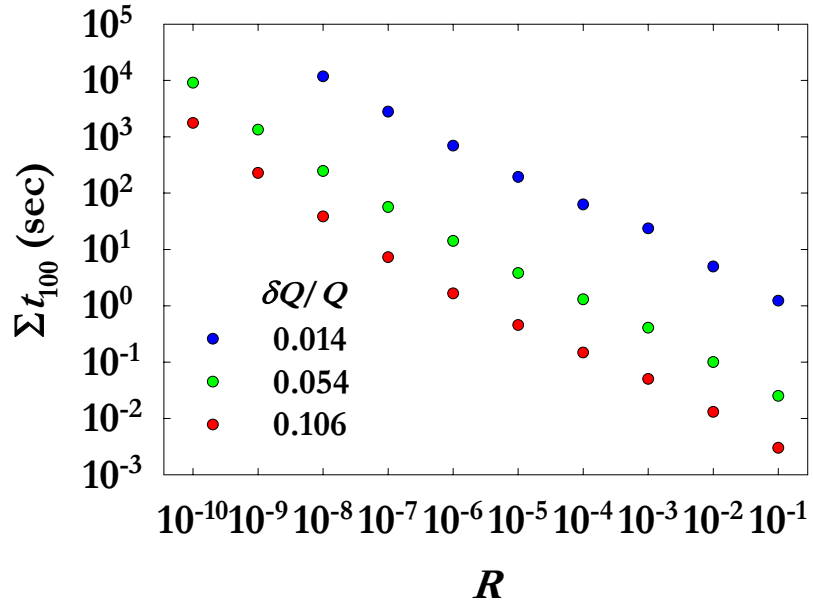
Optical Performance of the Liquids Reflectometer



- Wavelength cutoff: $\lambda < 1$ Å
- Horizontal divergence: $\pm 0.75°$
- Vertical divergence: $0 < \theta_v \leq 5°$ available

SiO_2 Measured as a Solid



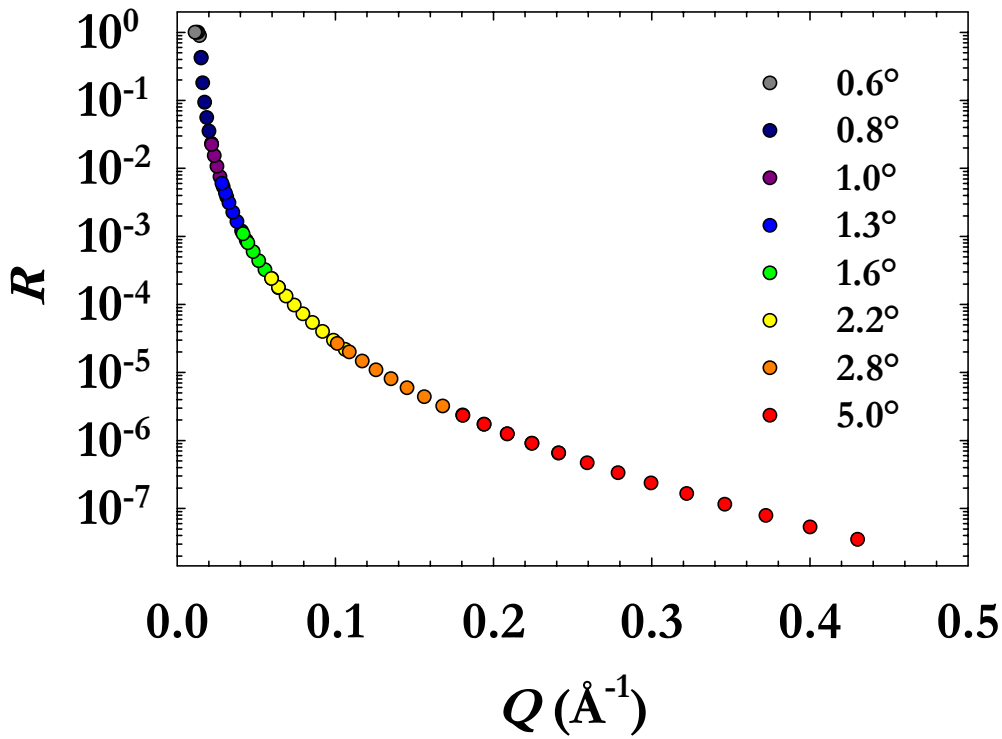


- Tilt sample and use $\theta_v = 4°$
- Bandwidth: 2.5 Å $\leq \lambda \leq$ 6.25 Å
- Use 6 incident angles θ_i

- Counting time Σt_{100}: cumulative time required to collect 100 counts for least-intense λ at each θ_i

$R_{min} \sim 10^{-10}$, $Q_{max} > 1$ Å^{-1} in several hours on ideal solid samples

SiO_2 Measured as a Liquid

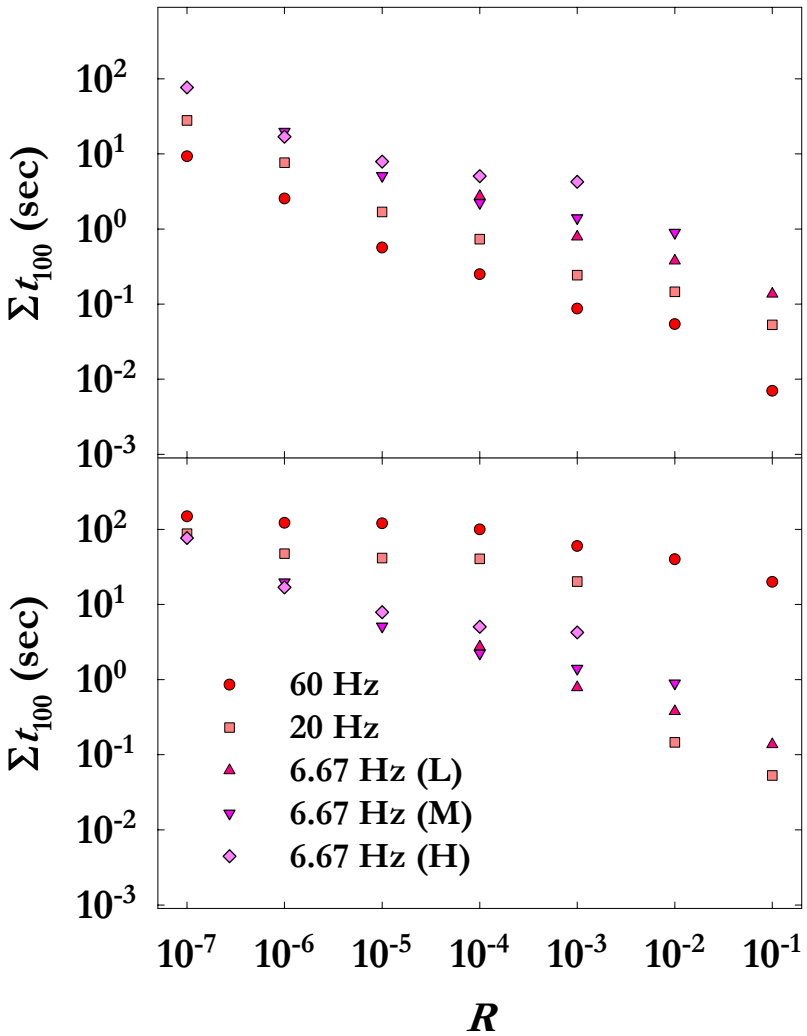


- Sample surface horizontal: slits select from $0 < \theta_v \leq 5°$ to define different θ_i
- Each θ_i uses different λ range – user must re-phase choppers
- Smaller dynamic range than solid
- More θ_i required (8)

$R_{min} \sim 10^{-7}$, $Q_{max} \sim 0.5$ Å^{-1} in 10 minutes for D_2O
(background limit of deuterated water sample)

Liquid Measurement Counting Times

- Resolution: $\delta Q/Q = 0.104$
- Raw counting times favor high frequencies (top)
- Assessing 20-sec penalty per each additional θ_i favors lower frequencies (bottom)
- User can choose mode

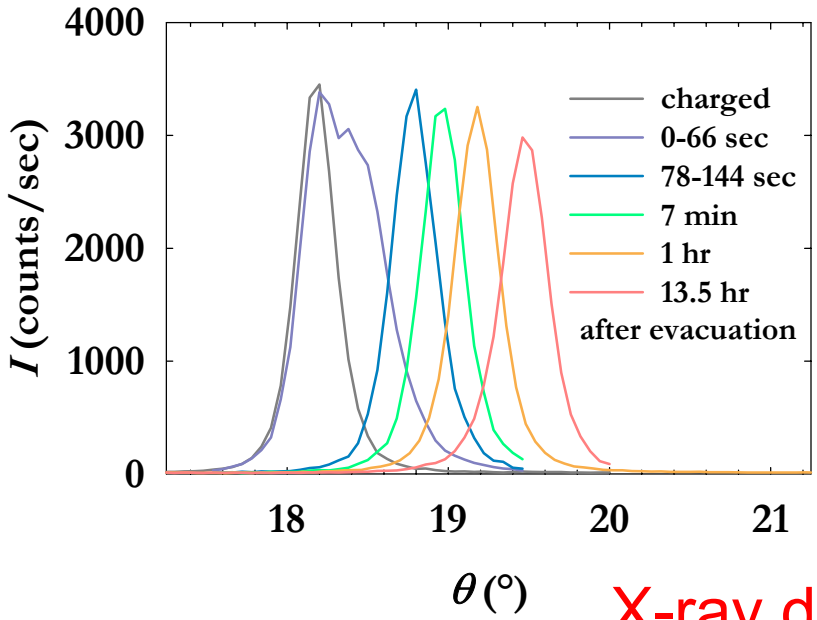


Scientific Possibilities

Example I:
Kinetics of Hydrogen Diffusion in Fe/Nb Multilayers
(Christine Rehm & John Ankner)

Time dependence: Fe/Nb-H outgassing

- [Fe 26 Å / Nb 1000 Å]$_5$ multilayer
- Loaded with H_2 at 900 mbar
- Time evolution of Nb Bragg peak ($Cu_{K\alpha}$) upon removal of H_2 atmosphere
- Earliest times (<100 sec) not well captured

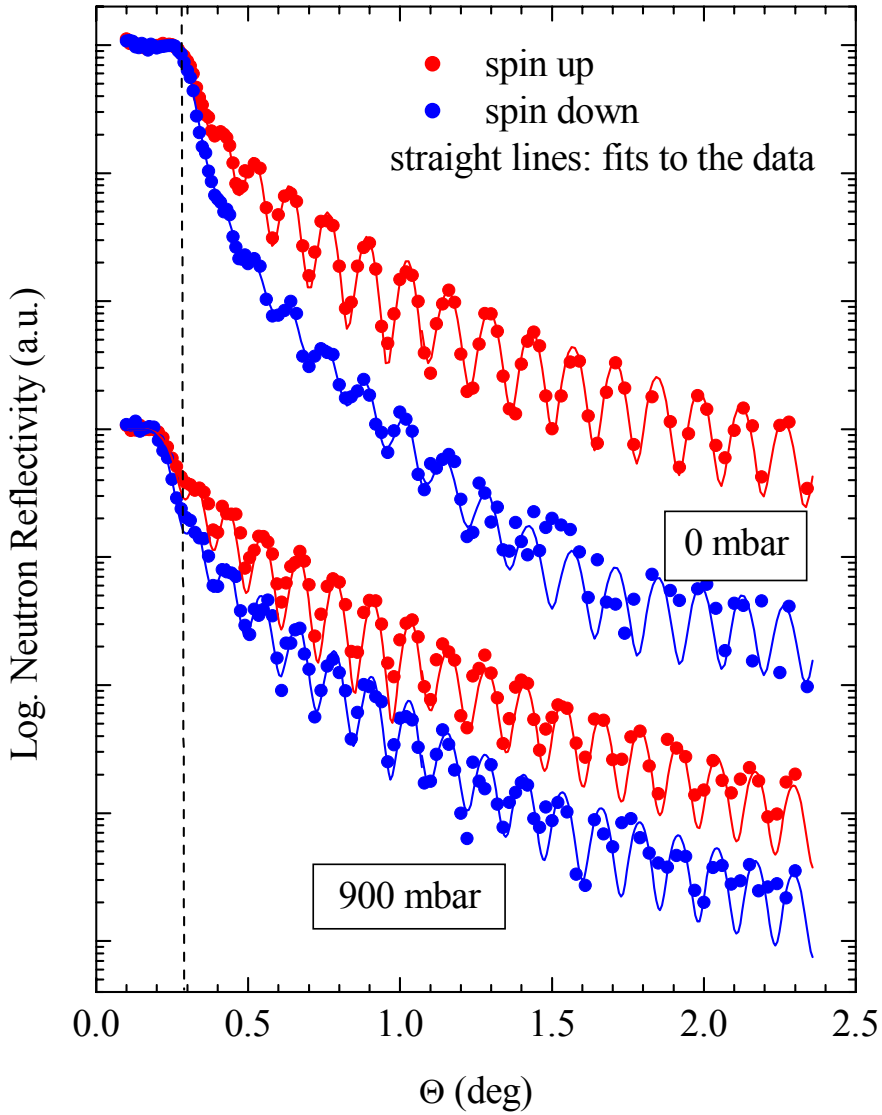


X-ray data

[Rehm et al., PRB 59, 3142 (1999)]

X-rays measure Nb lattice expansion only!
How can we measure hydrogen concentration?

Hydrogen Concentration Measurements in Nb Thin Films Using Neutron Reflectometry

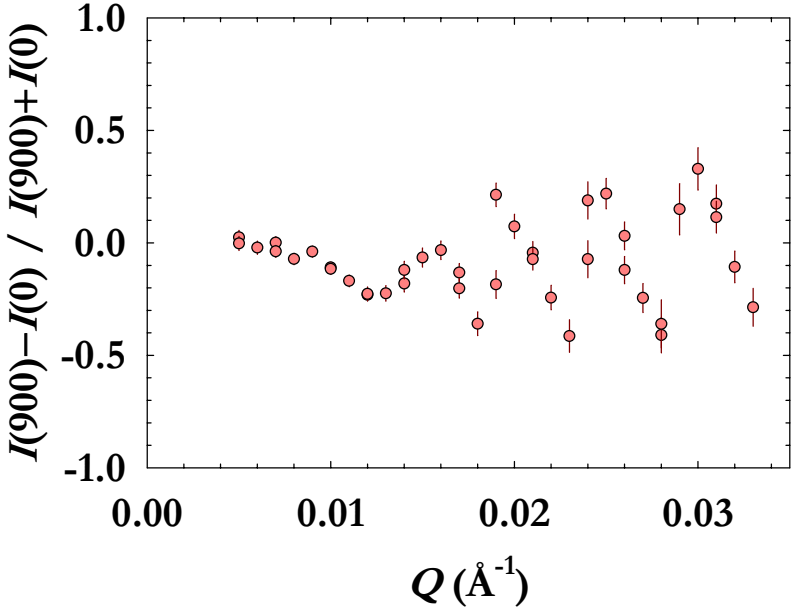


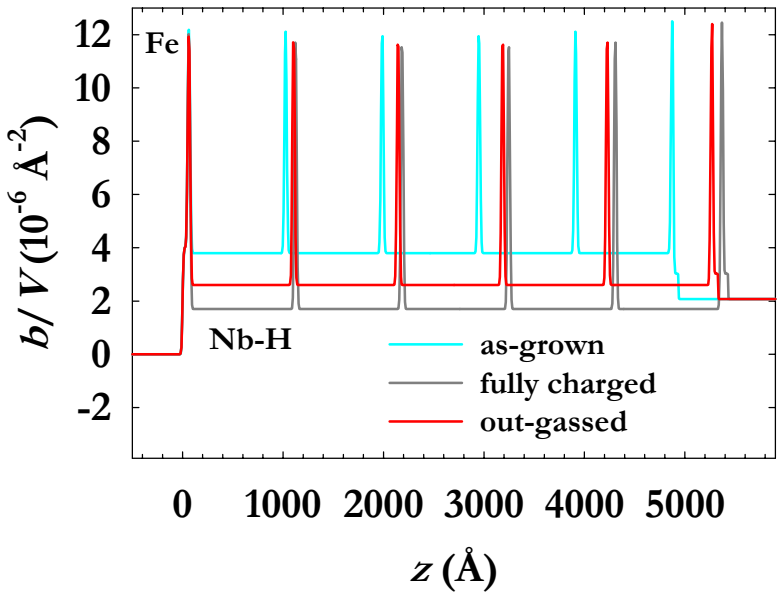
no hydrogen

95 [H]/[Nb] at.%.

[Rehm et al., PRB 59, 3142 (1999)]

Conventional Neutron Reflectivity Measurement

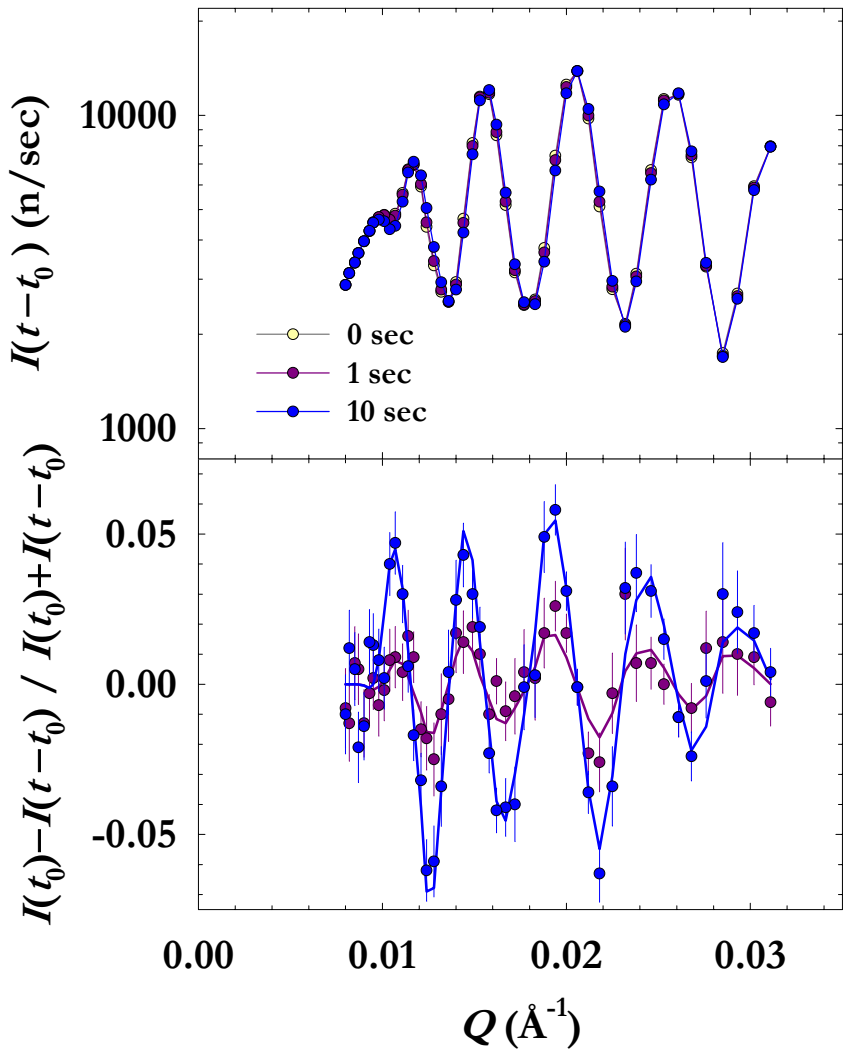




- Difference plot of charged vs. outgassed reflectivities
- Resolution: $\delta Q \approx 0.0015$ Å^{-1}
- Each (+) reflectivity required ~ 1 hr at HMI V6

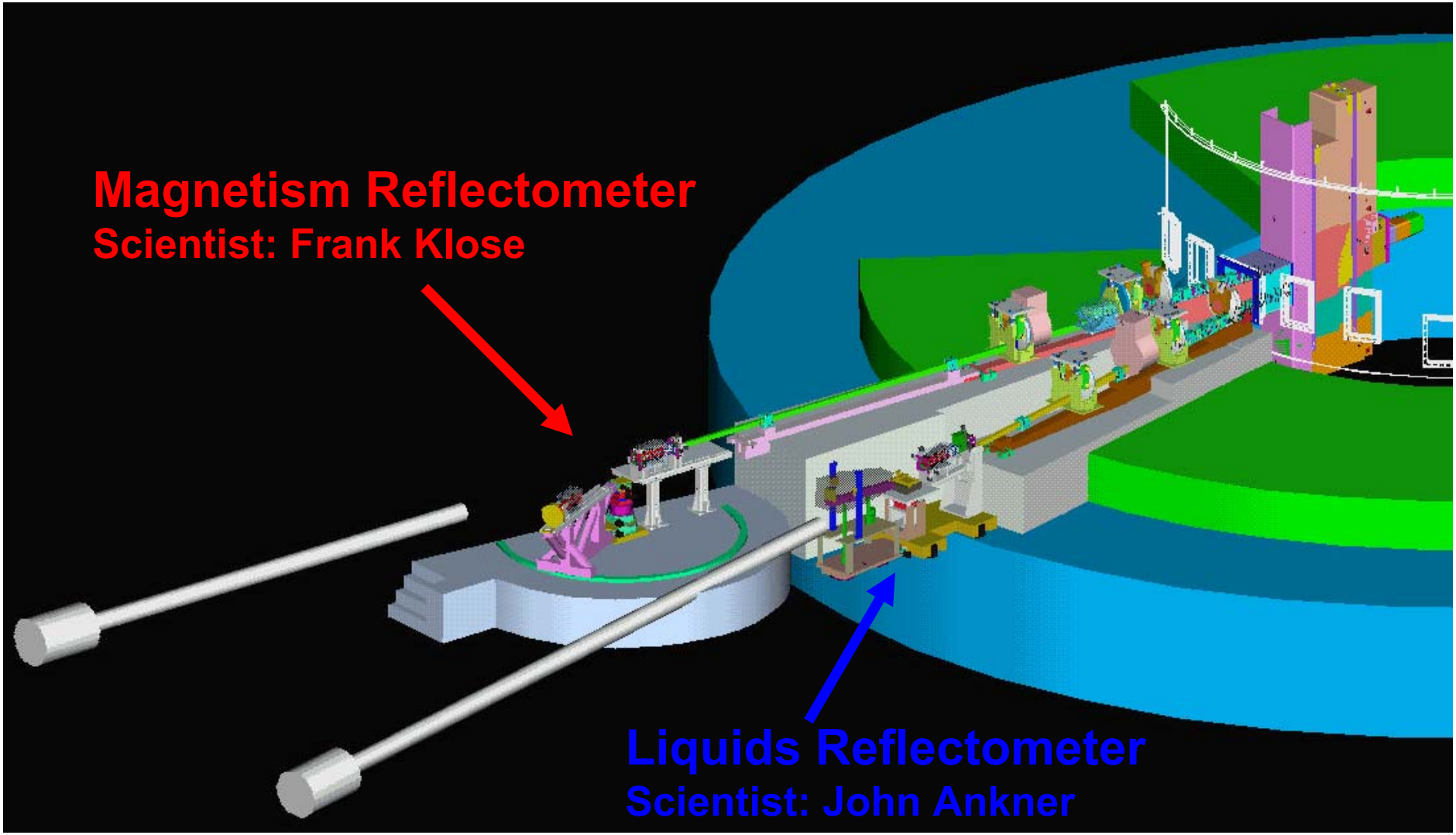
- Incomplete removal of H upon outgassing
- Fitted parameters used as basis for SNS calculation

Simulated measurement at SNS

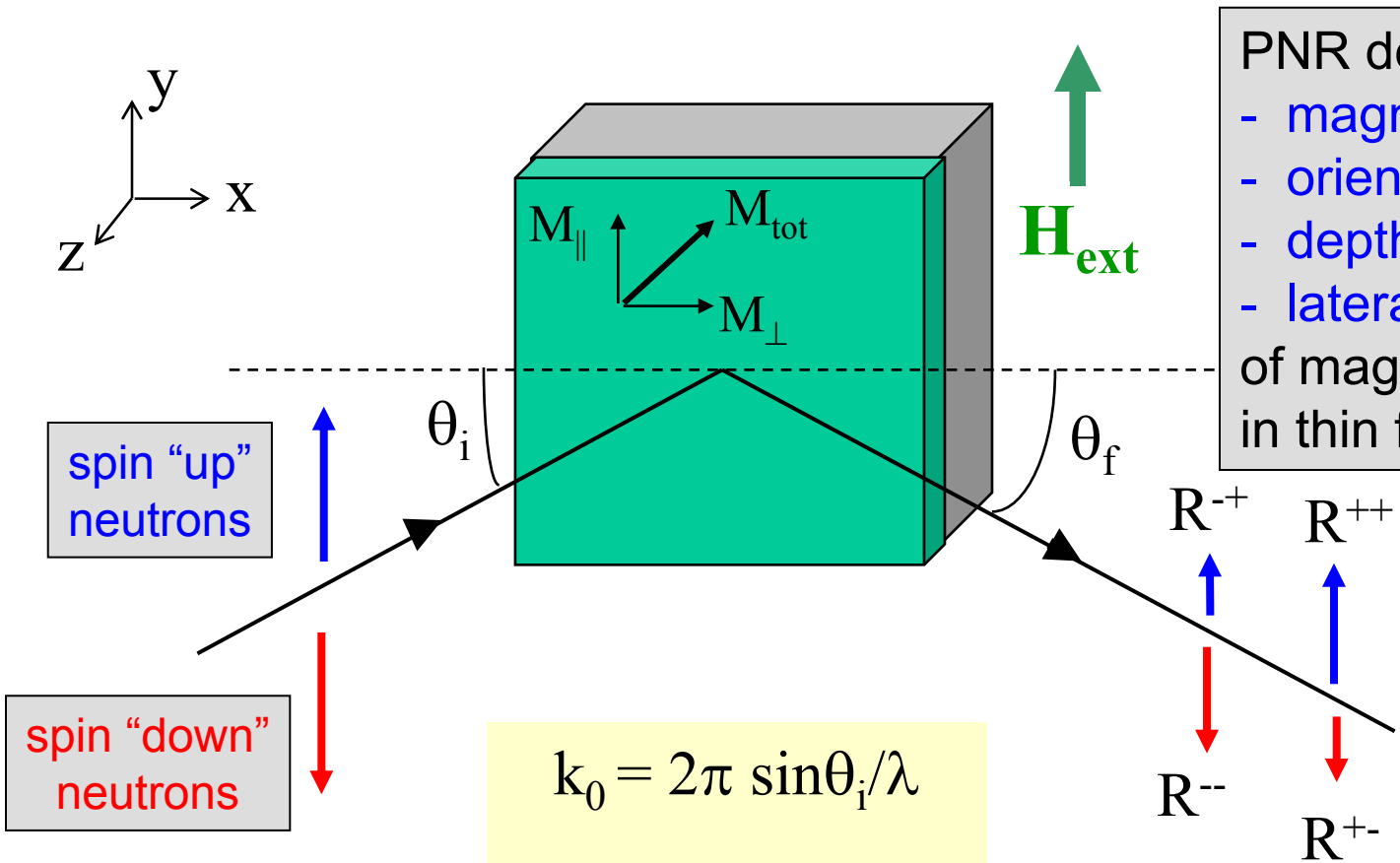


- 1-sec shots
- f = 30 Hz
- One angle: θ_i = 0.36°
- Film structure interpolated from x-ray and neutron data
- Scatter and error bars generated from calculated count rates

SNS Reflectometers



Polarized Neutron Reflectometry



PNR determines:
- magnitude
- orientation
- depth distribution
- lateral arrangements

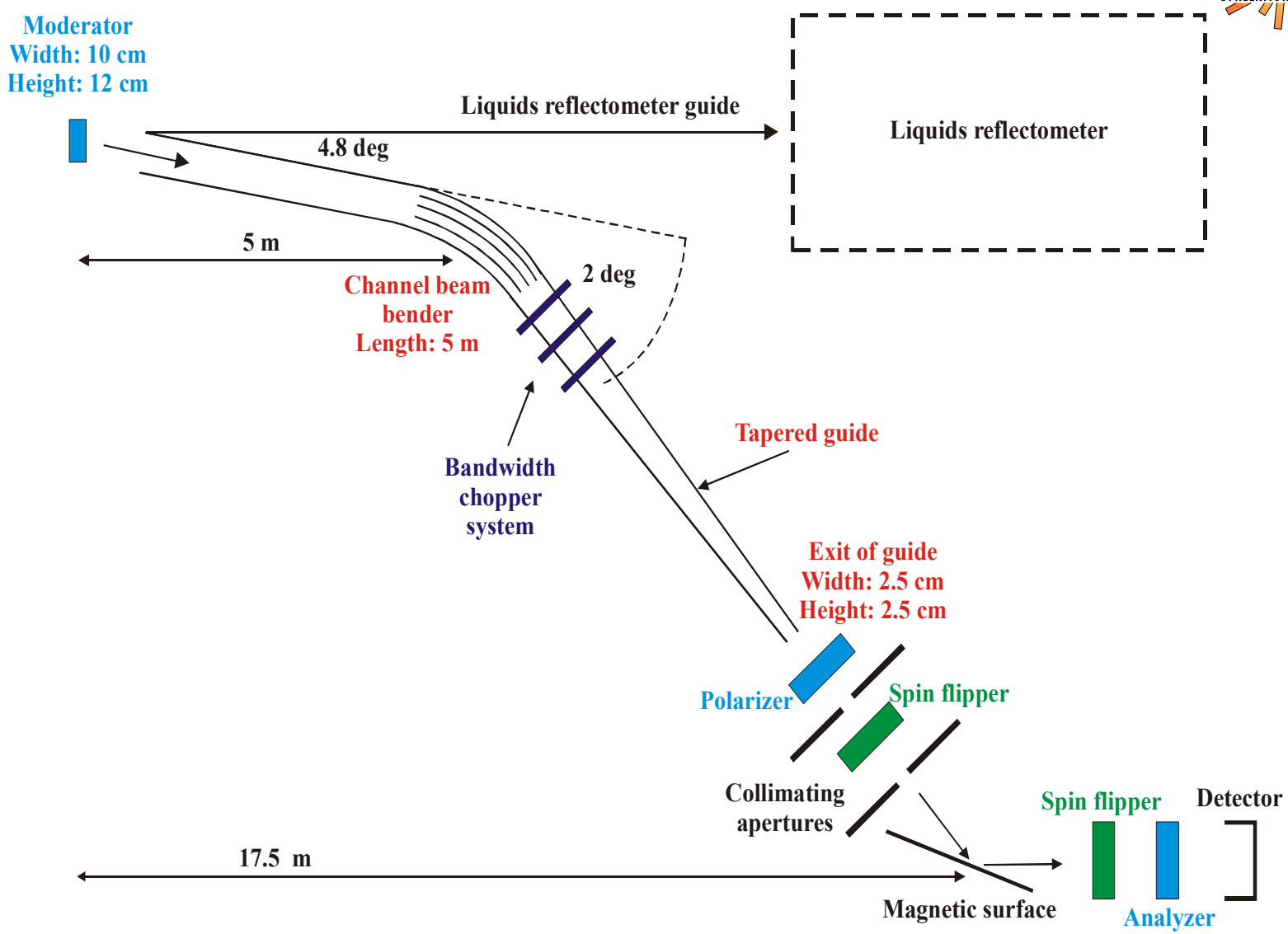
of magnetization vectors in thin film structures

measurement of **four** cross-sections

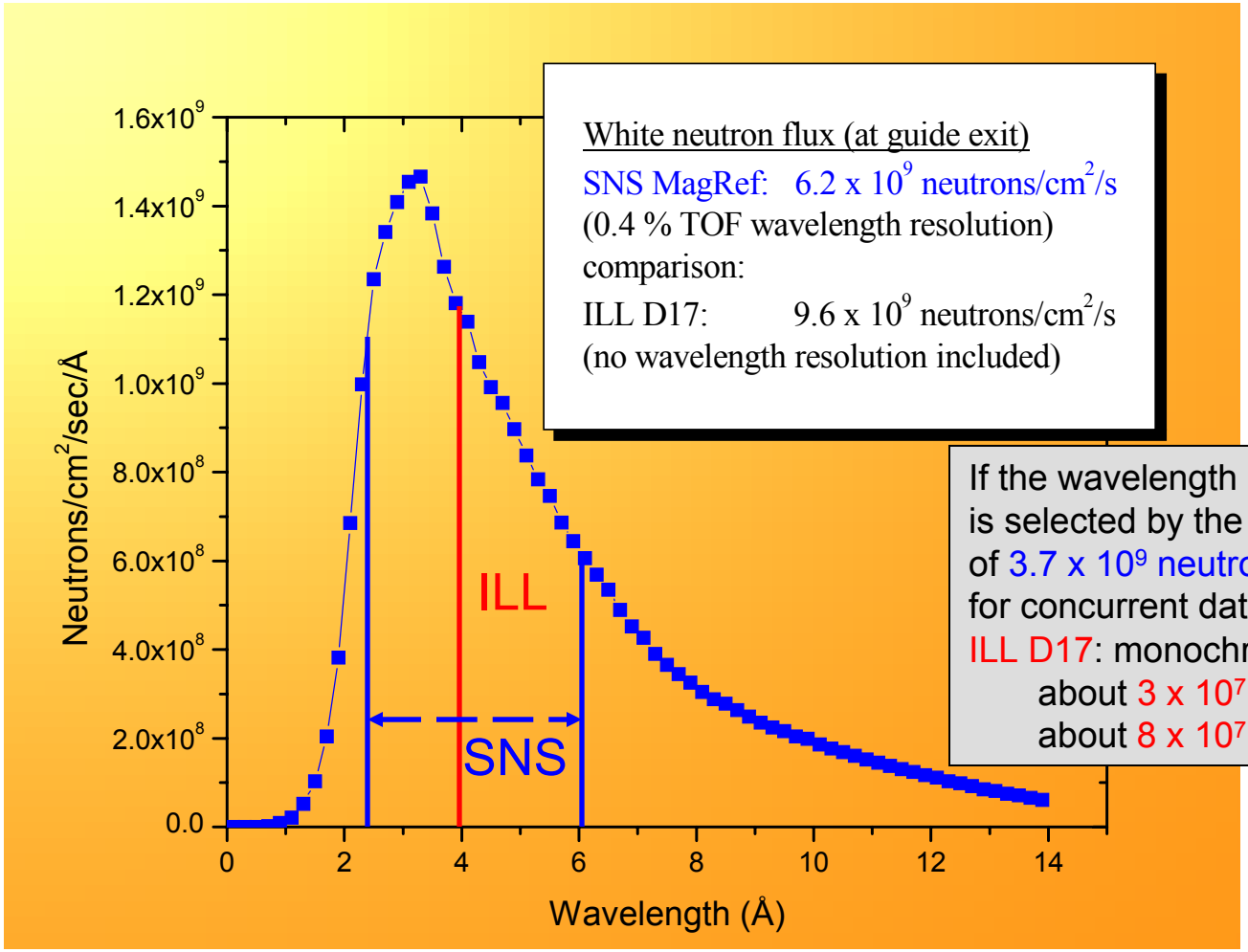
$$k_0 = 2\pi \sin\theta_i/\lambda$$

$$q = 4\pi \sin\theta_i/\lambda$$

Neutron Beam Optics of the Magnetism Reflectometer (schematic)



Flux comparison: SNS Reflectometer (2MW) vs. D17 at ILL



White neutron flux (at guide exit)

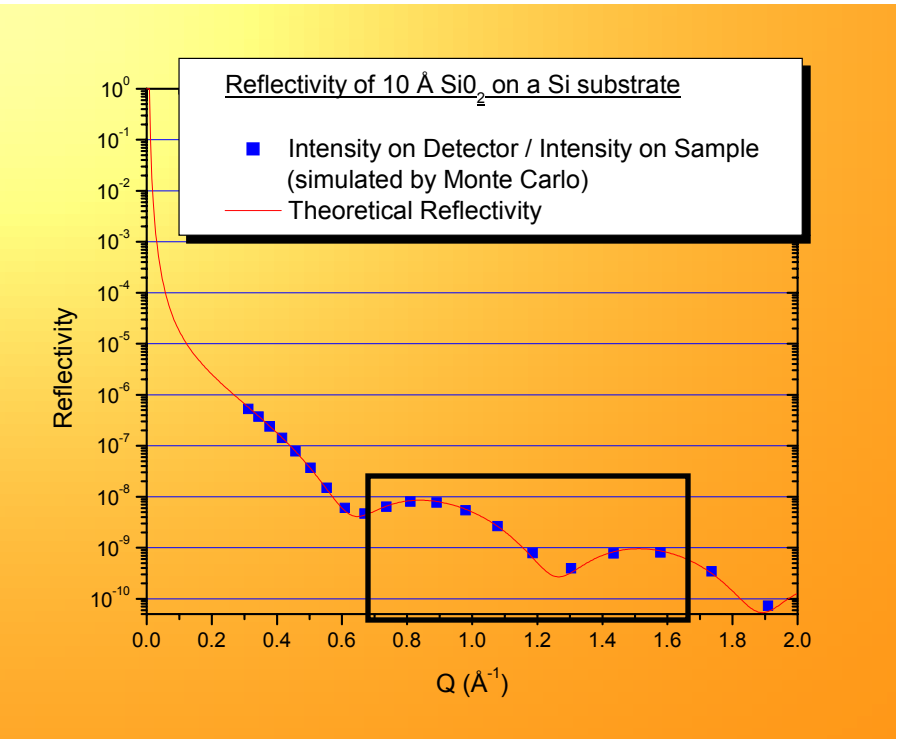
SNS MagRef: 6.2×10^9 neutrons/cm^2/s
(0.4 % TOF wavelength resolution)
comparison:
ILL D17: 9.6×10^9 neutrons/cm^2/s
(no wavelength resolution included)

If the wavelength band of 2.6 Å to 6.1 Å is selected by the choppers, a neutron flux of 3.7×10^9 neutrons/cm^2/s can be used for concurrent data collection.

ILL D17: monochromatic beam, λ =4 Å
about 3×10^7 at 0.4% λ resolution
about 8×10^7 at 1% λ resolution

F. Klose, W.T. Lee
Applied Physics A. Material Science & Processing (ICNS 2001 Proceedings, accepted for publication)
W.T. Lee and F. Klose, SPIE Proceedings, Volume 4509 (2001), p. 145 - 156

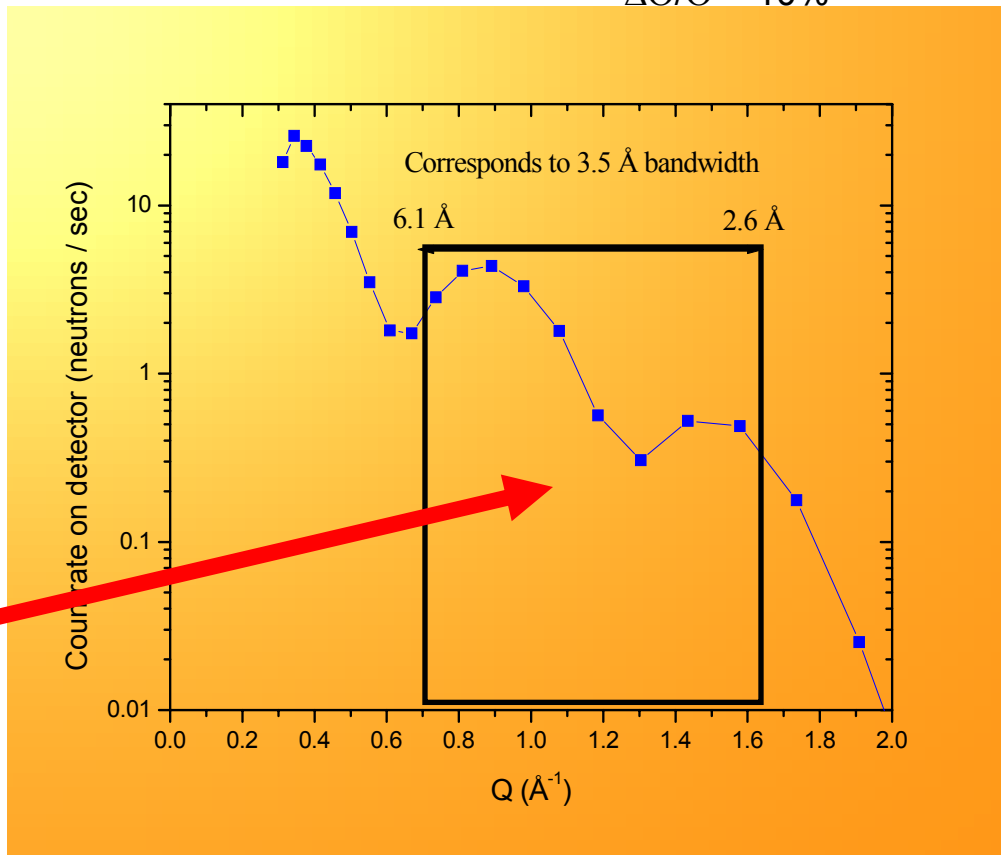
Simulated Detector Count Rates (10^{-8} – 10^{-10} reflectivity range)



10 Å SiO_2 on Si
Sample size:
25 mm x 25 mm

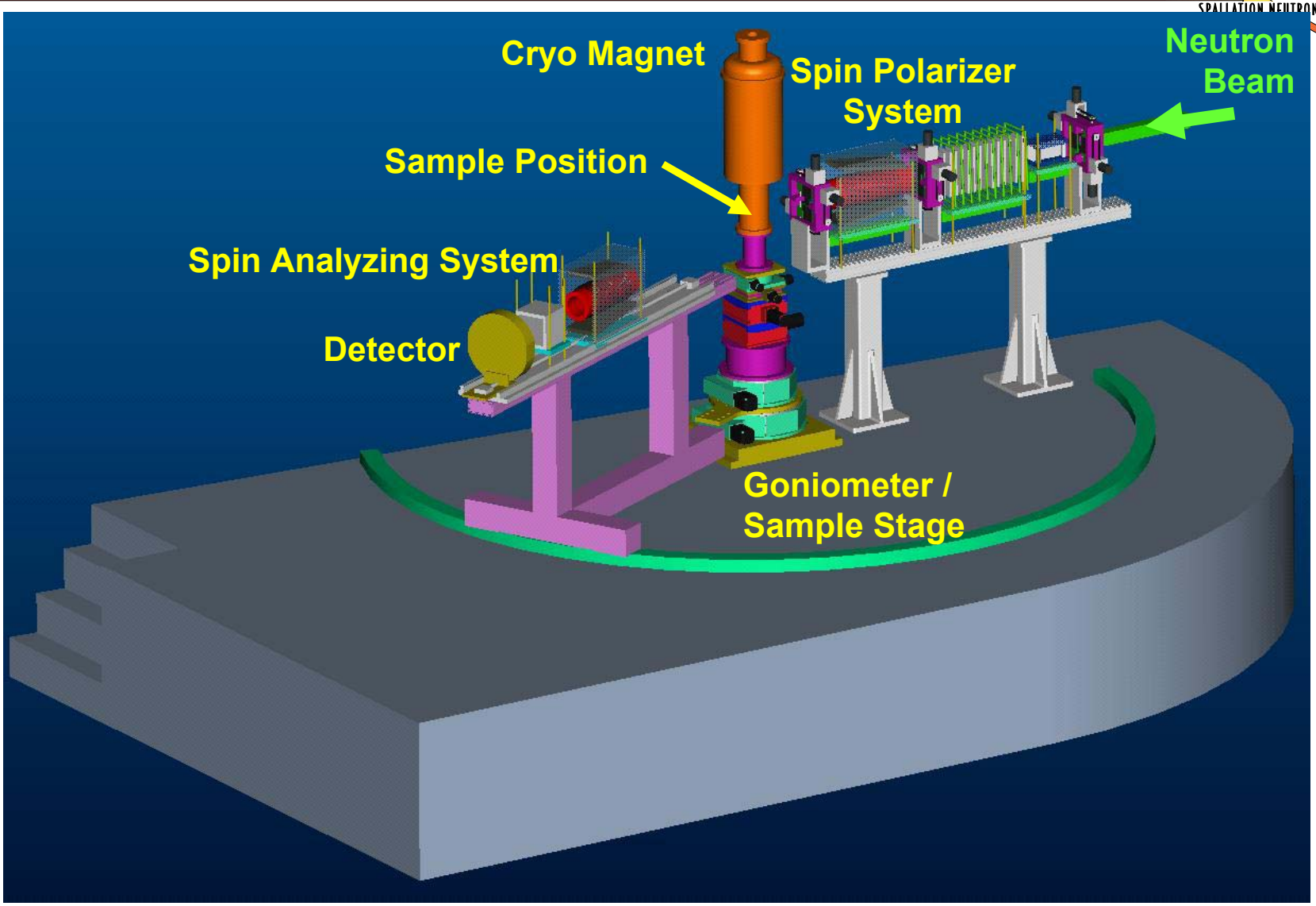
Scattering Angle:
Θ = 20 deg.

Resolution:
$\Delta\lambda/\lambda$ = 10%
$\Delta\Theta/\Theta$ = 10%

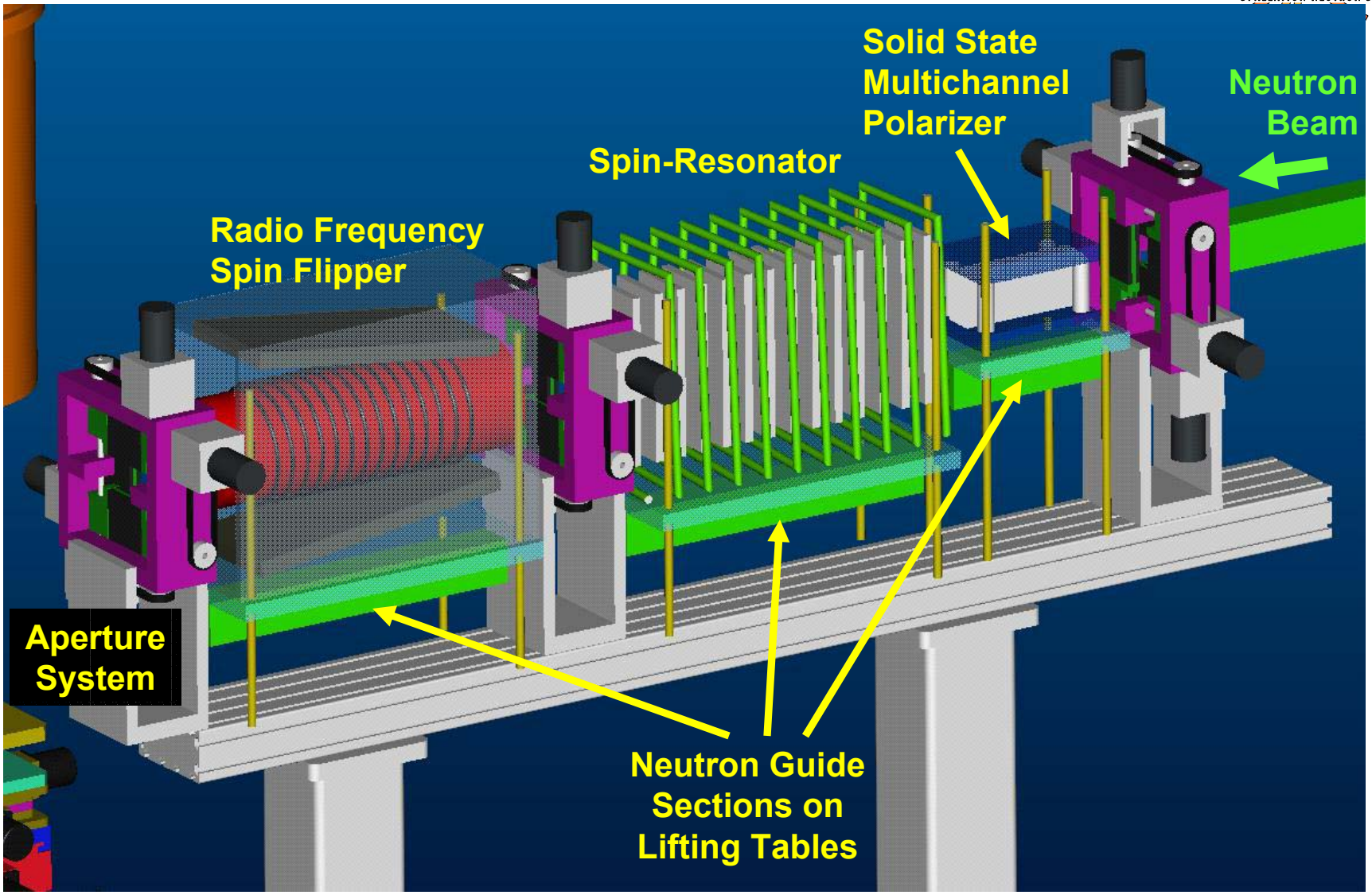


Marked region can be measured within 8 minutes ! (>100 counts per TOF channel)

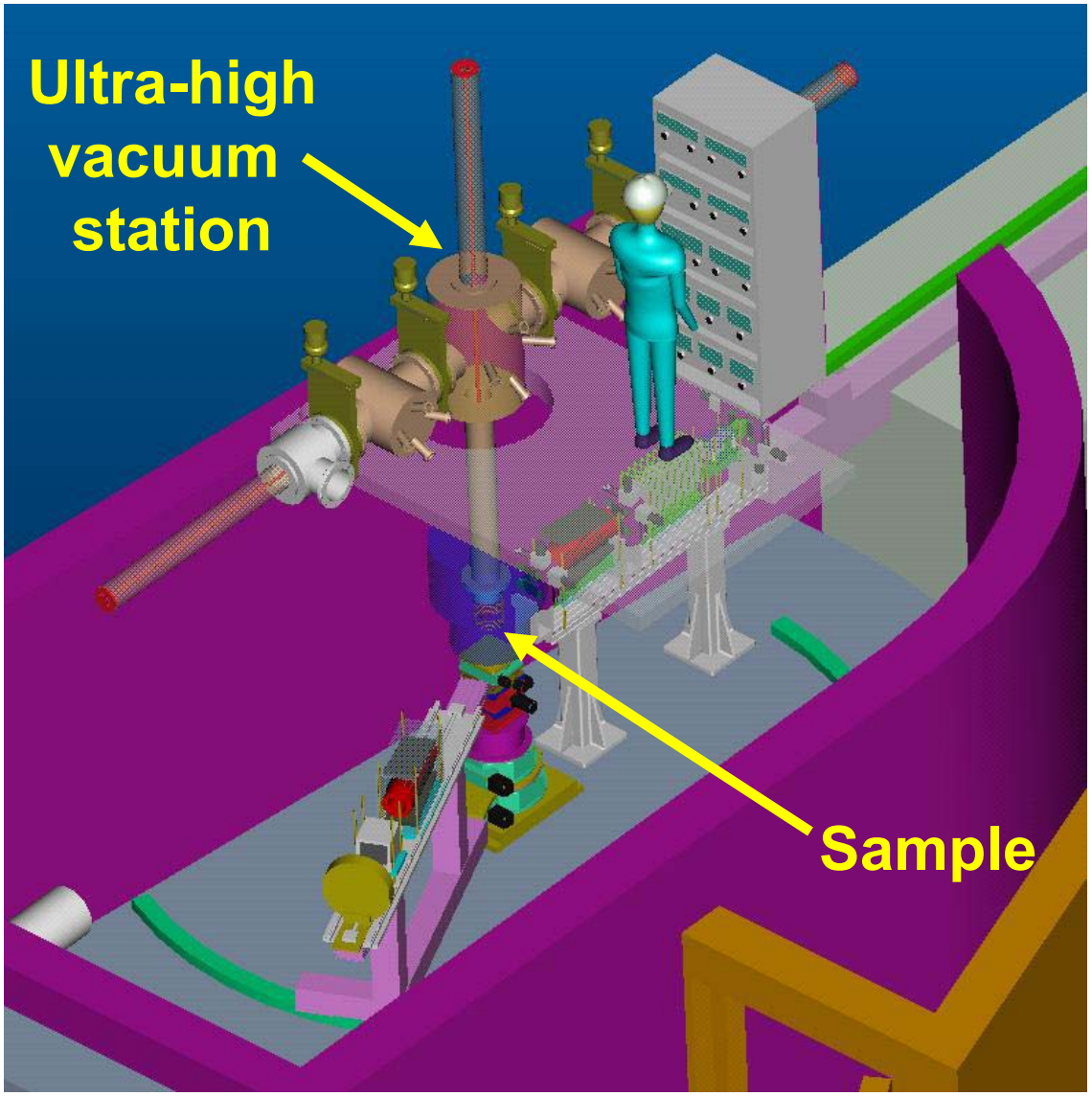
Sample Area With Cryo Magnet Installed



Spin Polarizer System



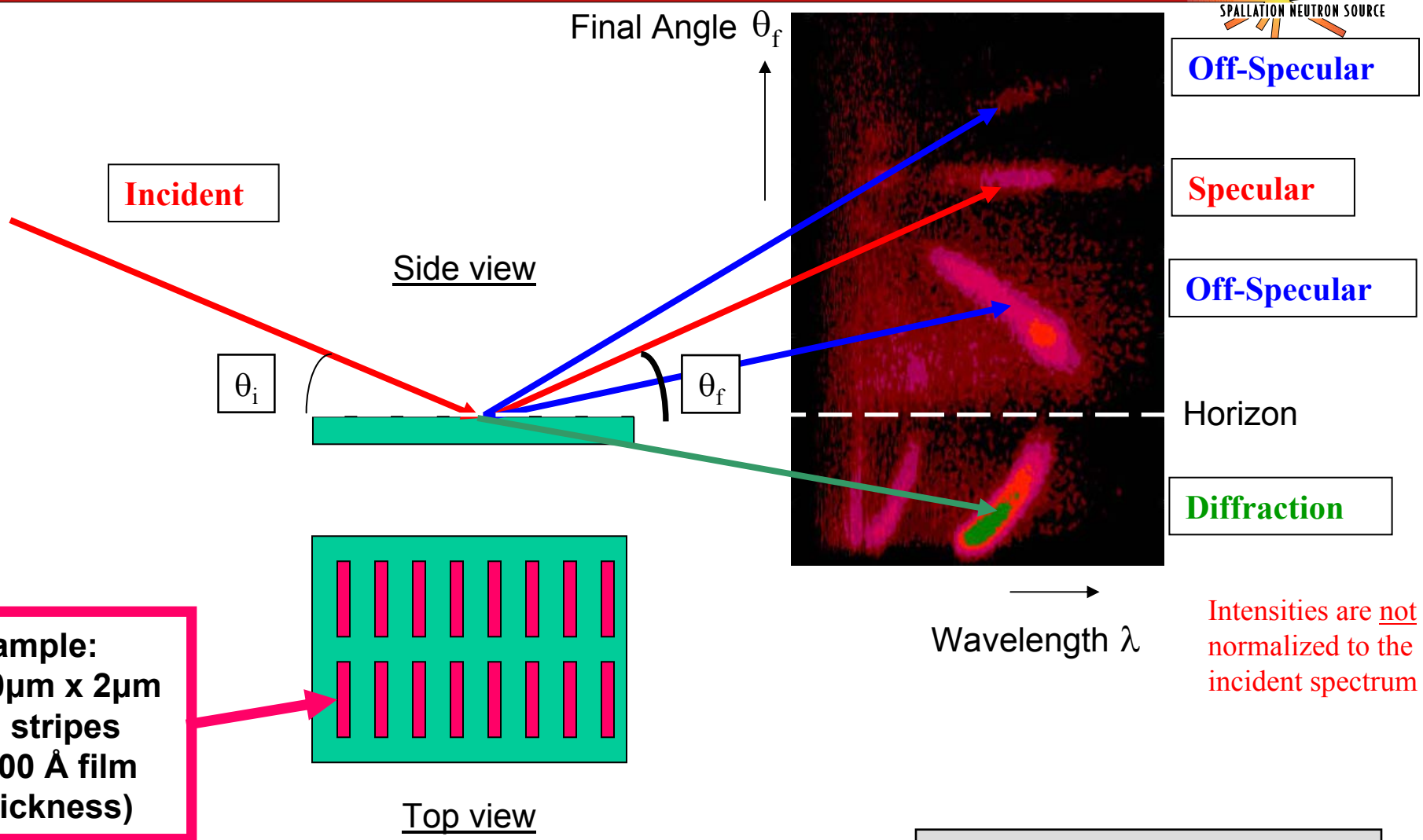
XYZ Magnet and UHV Station



Scientific Possibilities

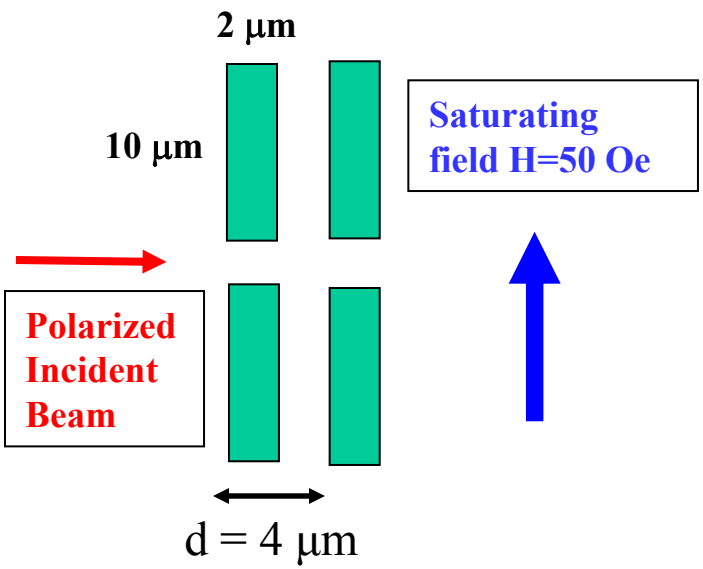
Example II:
Off-Specular Scattering from Laterally Structured Films
(Hal Lee & Frank Klose)

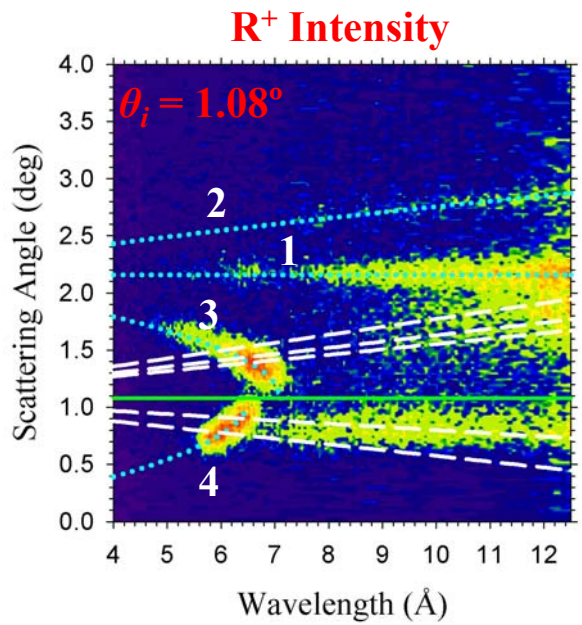
Off-Specular Scattering From Lateral Structures

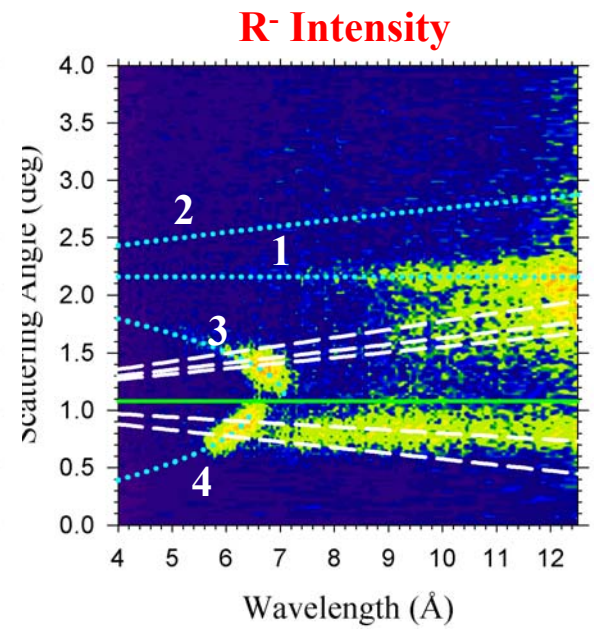


W.T. Lee & F. Klose, 2002

Polarized Neutron Time-Of-Flight Off-Specular Scattering







Intensities are normalized to the incident spectrum

1: specular reflection

2,3: off-specular reflections (above horizon)

$$\theta = \theta_i + \sqrt{\theta_i^{\,2} + 2n\lambda / d} \quad \text{with } n = +/- 1$$

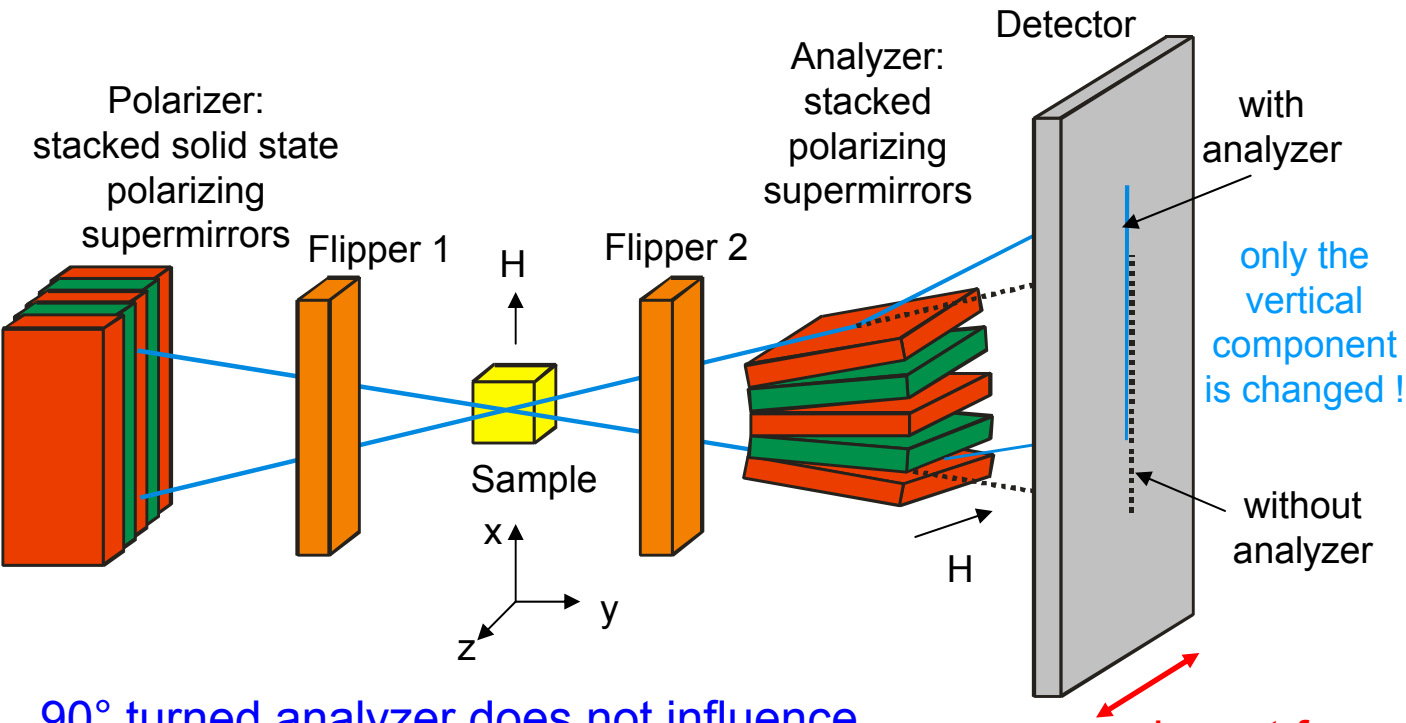
4: off-specular diffraction (below horizon)

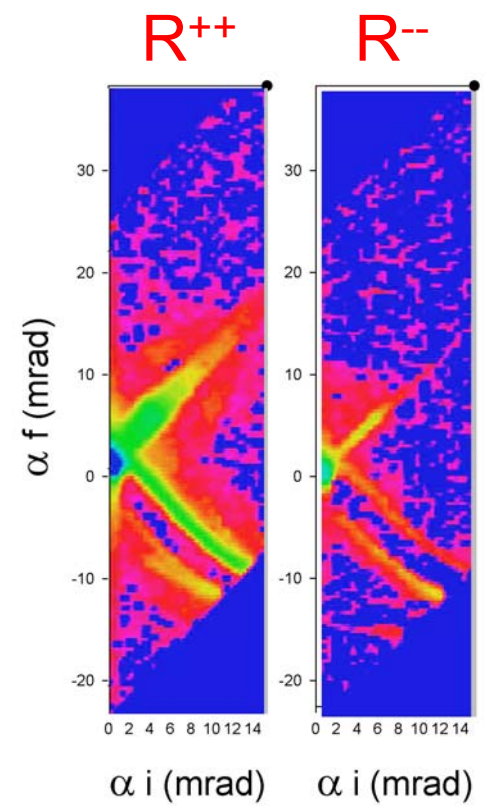
$$\theta = \theta_i + \sqrt{\theta_i^{\,2} + 2n\lambda / d - Nb\lambda^2 / \pi} \quad \text{with } n = -1$$

(dashed lines are the critical edges: from the horizon to higher/lower angles, the silicon edge -above the horizon only- and the permalloy edges)

W.T. Lee, F. Klose, B. Toperverg, PNCMI Conference Proceedings, 2002

Polarized Neutron Off-Specular Scattering with Polarization Analysis





90° turned analyzer does not influence the z-component of the neutron!

relevant for diffuse scattering

W.T. Lee, F. Klose, B. Toperverg, U. Ruecker
measured at HADAS reflectometer, FZ Juelich, 2002

Outlook for Reflectometry (and Other Instruments) at SNS

Elastic Scattering

- Higher Flux
  - ⇒ Smaller Samples
  - ⇒ Shorter Measurements
    - allow determination of phase diagrams
    - allow kinetics to be studied in "real time"
  - ⇒ Sensitivity to Weak Signals
    - detects small moments and spatial dependence with high q
- Specular + Diffuse Scattering
  - ⇒ Information on Domains and other Lateral Structures

Inelastic Scattering

- Combine Reflectometry with Inelastic Scattering