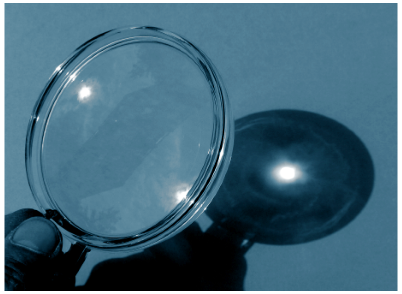
PAUL SCHERRER INSTITUT

TU Clausthal
Clausthal University of Technology

Jochen Stahn Laboratory for Neutron Scattering

Erwin Hüger
Florian Strauß
Harald Schmidt Institute of Metallurgy / Microkinetics Group



Focusing Reflectometry for In-situ Studies

Current trends and future perspectives in neutron reflectometry
08. - 09. 06. 2015, Lillestrøm, Norway

- Lithium Transport Through Thin Silicon Films
    - high-intensity specular reflectometry
        - the ***Selene*** guide
            - Amor & ***Selene***
                - reduction of the Li transport data
                    - ***Estia***

- Lithium Transport Through Thin Silicon Films
    - high-intensity specular reflectometry
        - the ***Selene*** guide
            - Amor & ***Selene***
                - reduction of the Li transport data
                    - ***Estia***

example for an *in-situ* study using n reflectometry

Lithium Transport Through Thin Silicon Films

in cooperation with E. Hüger, F. Strauß and H. Schmidt, TU Clausthal

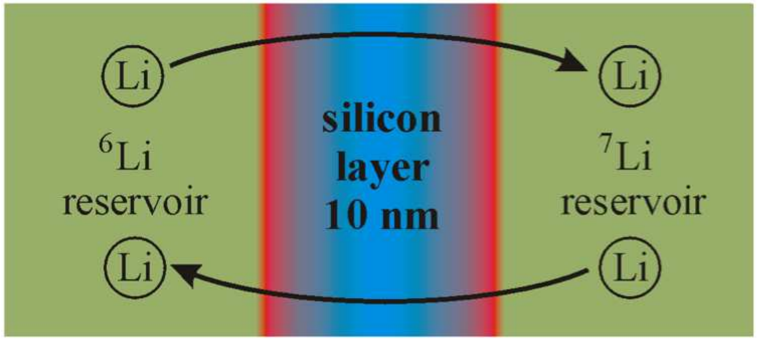
technological motivation:

- Si layers can be used in Li batteries to prevent oxidation of the electrodes
- Si films can be used as elecrodes in Li batteries

⇒ How fast does Li difuse through thin amorphos Si films?

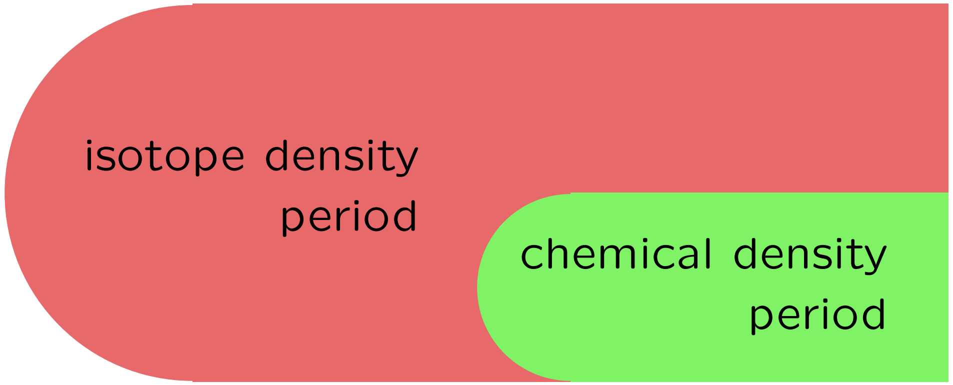
⇒ What is the solubility of Li in Si?

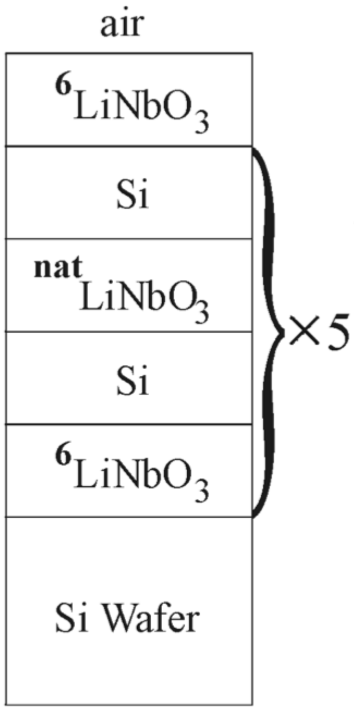
⇒ What is the influence of the Si:O:Li interface layer?

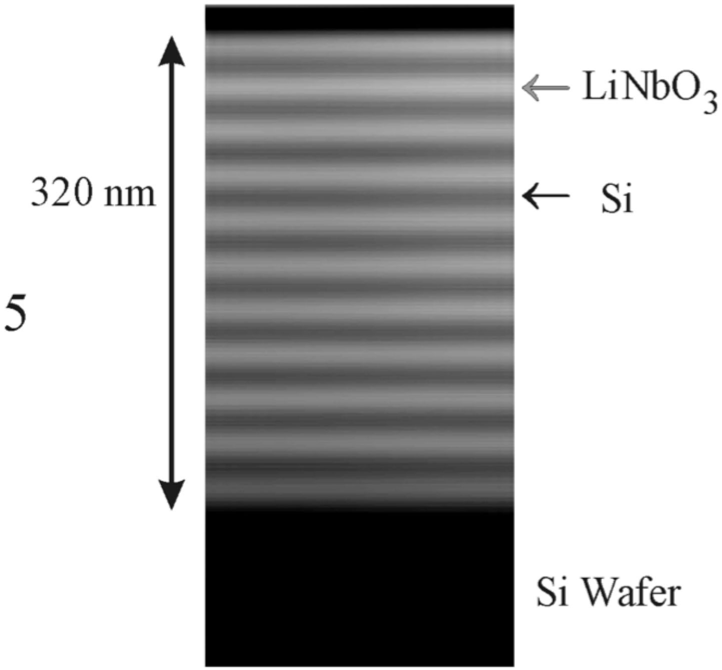


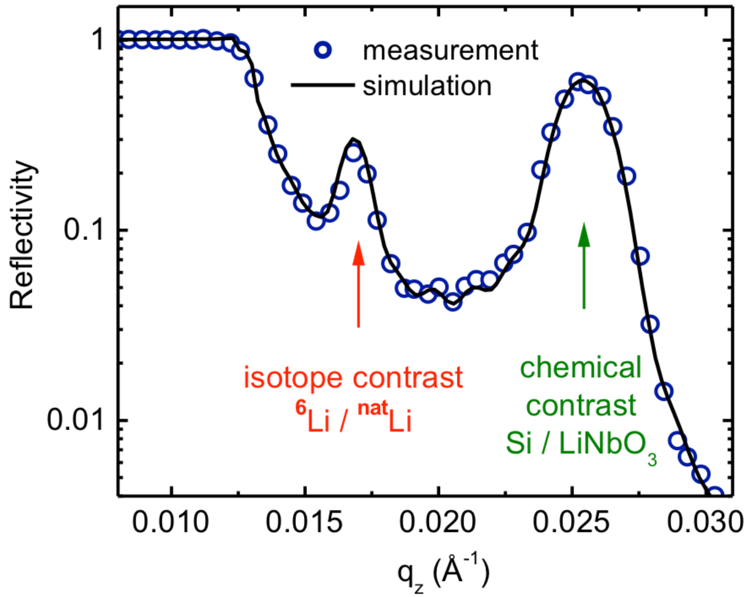
E. Hüger, *et al.*, Nano Letters 13 (2013) 1237.

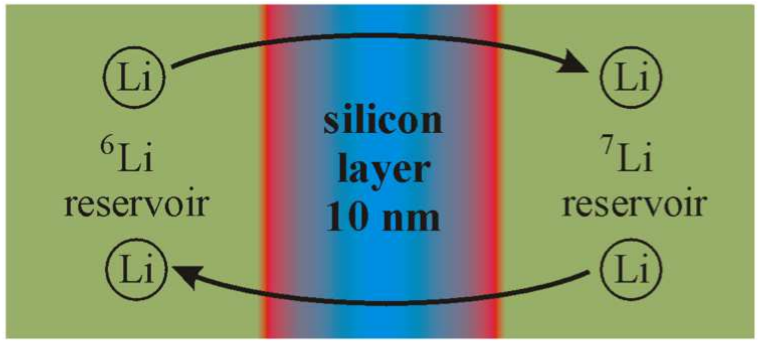
multilayer structure using the different densities of ^{6}Li and ^{7}Li



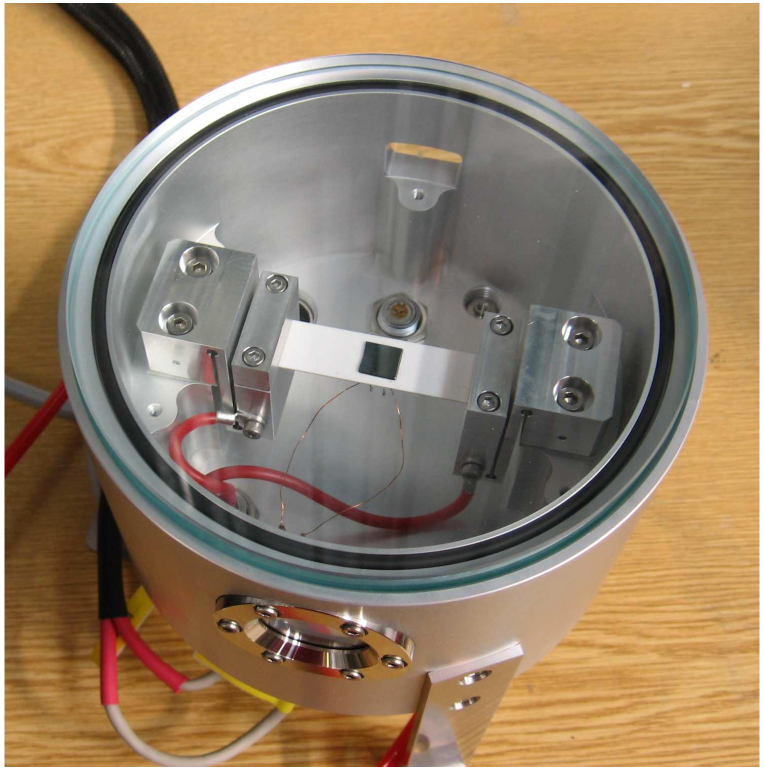








in-situ furnace



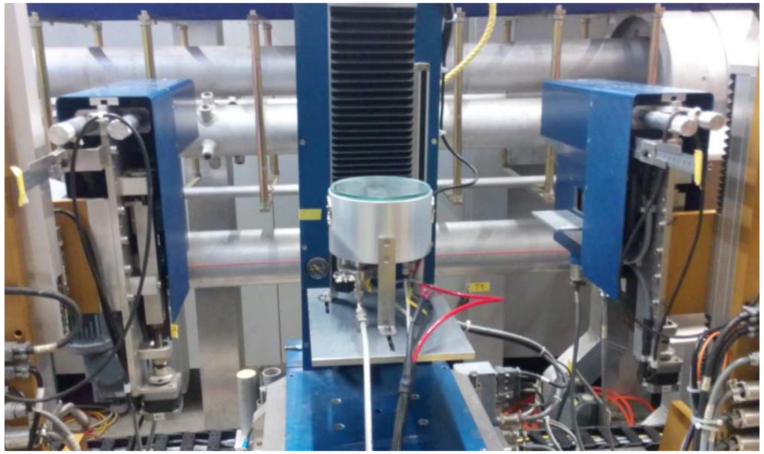
- $T \in [25^\circ\mathrm{C}, 500^\circ\mathrm{C}]$ here: $T = 240^\circ\mathrm{C}$
- $\dot{T} = 50\,\mathrm{Ks}^{-1}$ for heating
- $\dot{T} = 12\,\mathrm{Ks}^{-1}$ for cooling

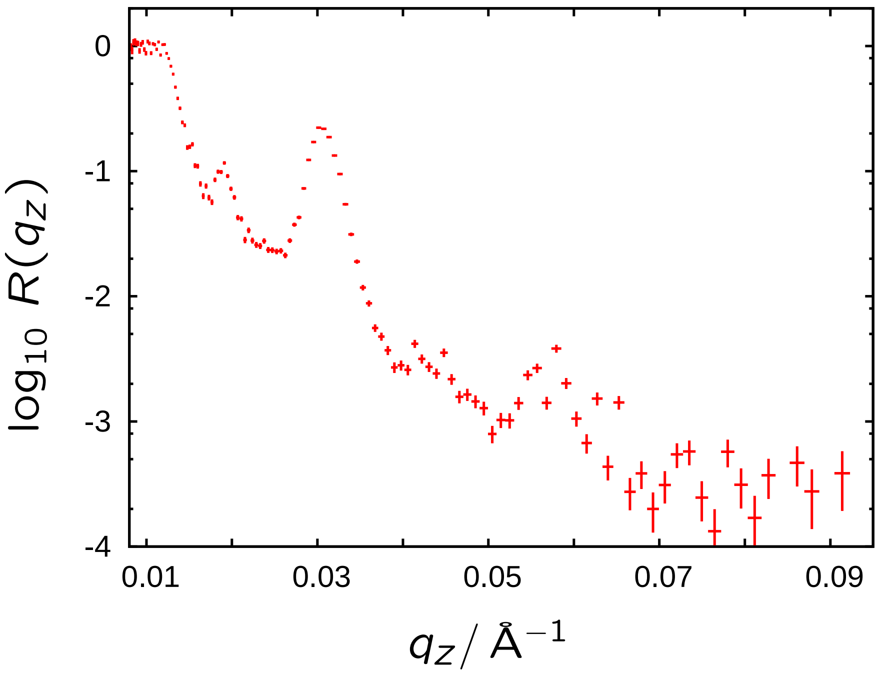
time-structure

- interval
  (measurements at RT in between annealing periods)
- continous measurement

instrument:

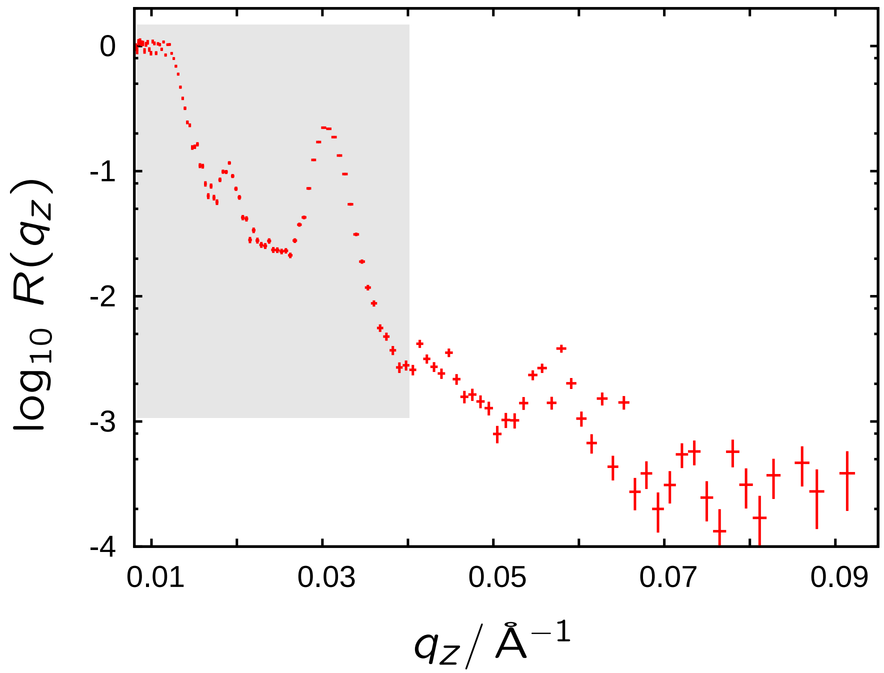
Amor — a TOF reflectometer at PSI, Switzerland

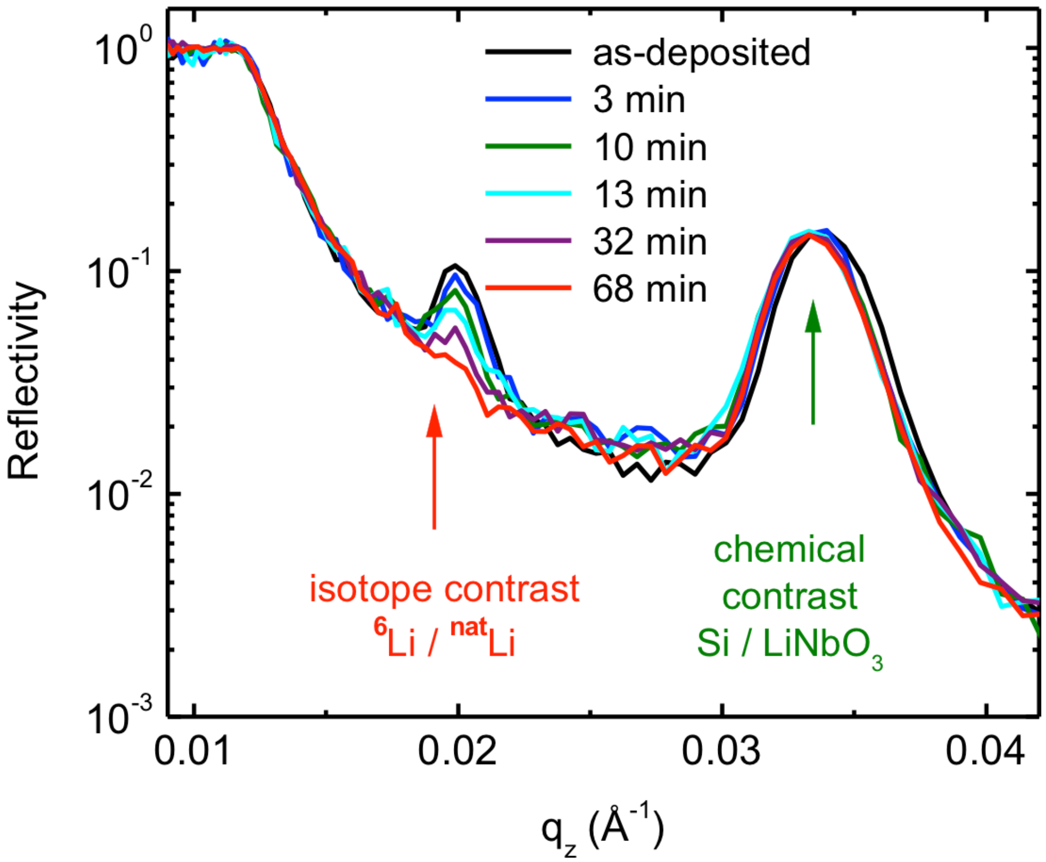




reflectivity during annealing at 240°C
measurement time 1.5 min

reflectivity during annealing
measurement time 1.5 min
after various times



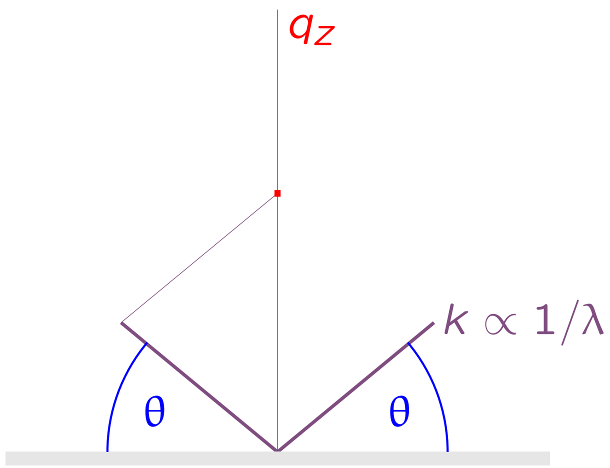


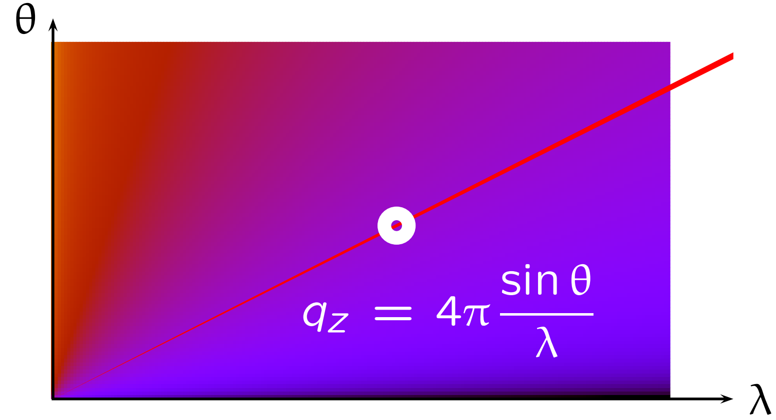
ml is chemically stable

Li contrast is vanishing

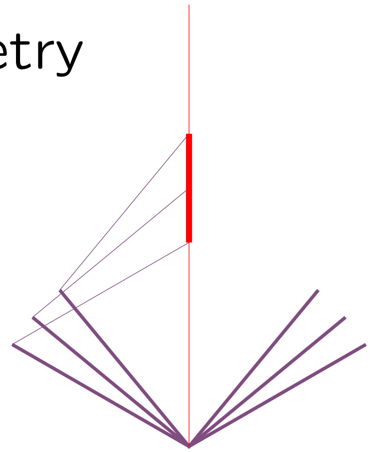
- Lithium Transport Through Thin Silicon Films
  - high-intensity specular reflectometry
    - the ***Selene*** guide
      - Amor & ***Selene***
        - reduction of the Li transport data
          - ***Estia***

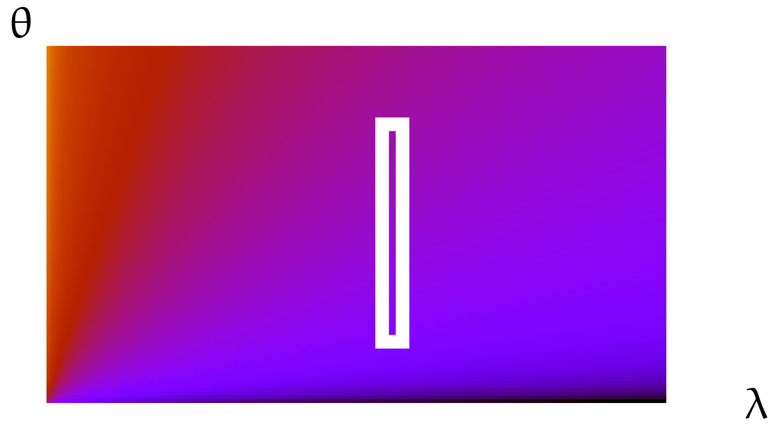
specular reflectometry



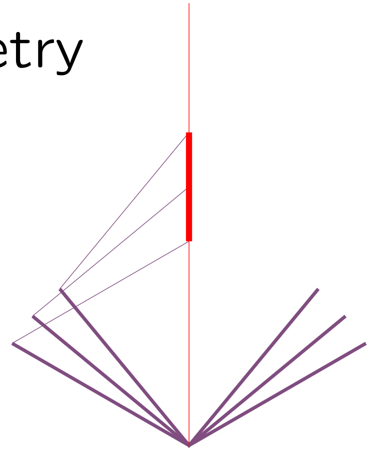


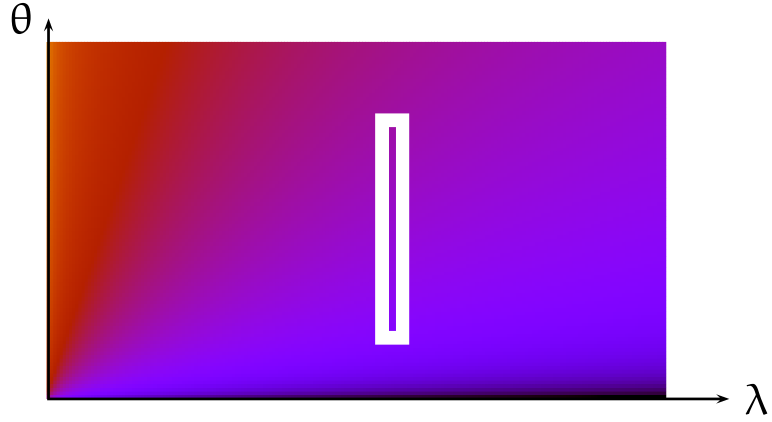
angle-dispersive reflectometry



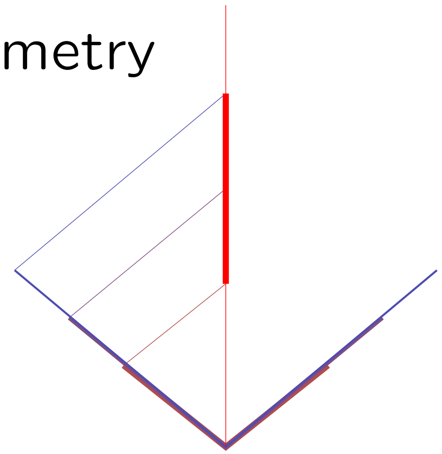


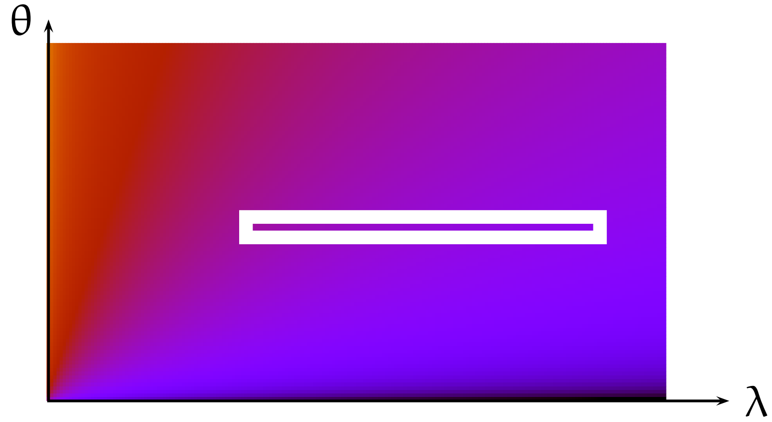
angle-dispersive reflectometry



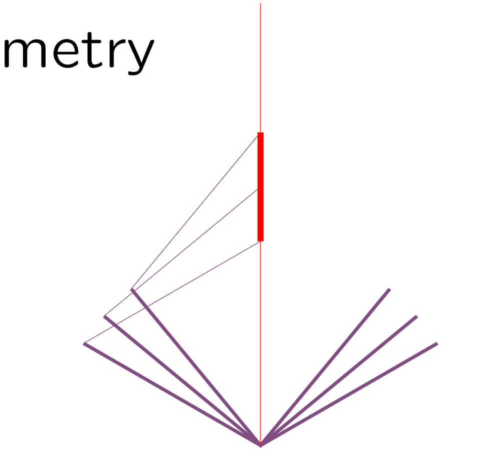


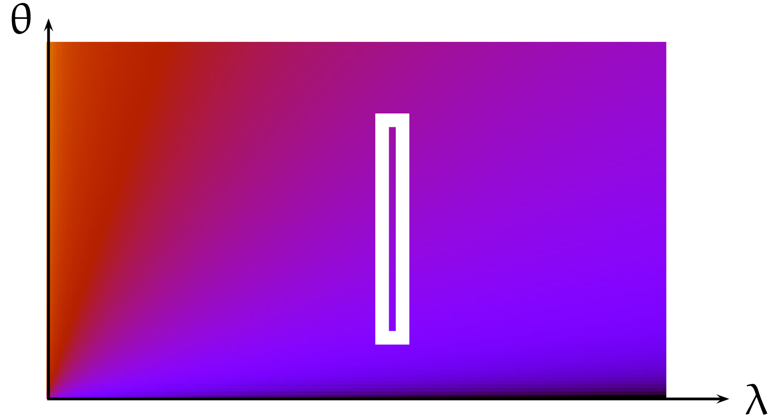
energy-dispersive reflectometry



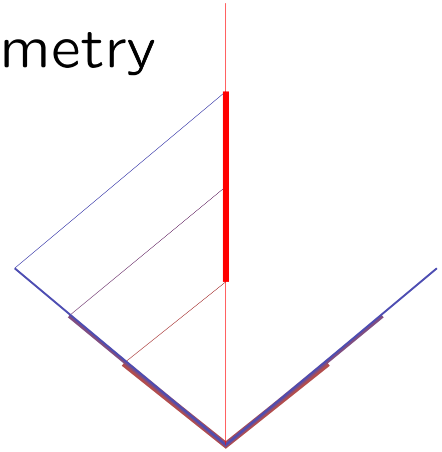


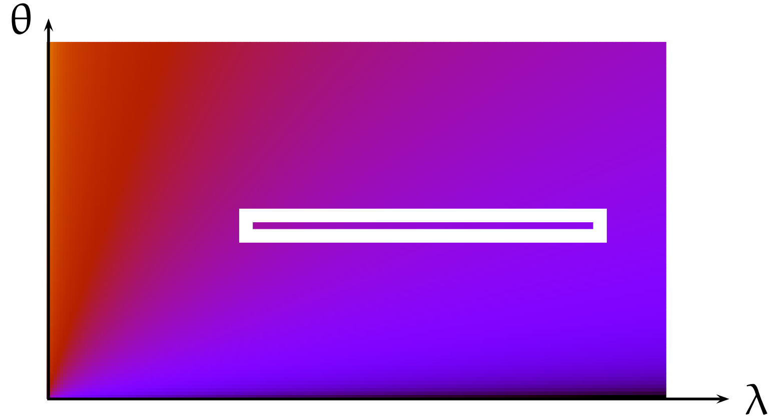
angle-dispersive reflectometry



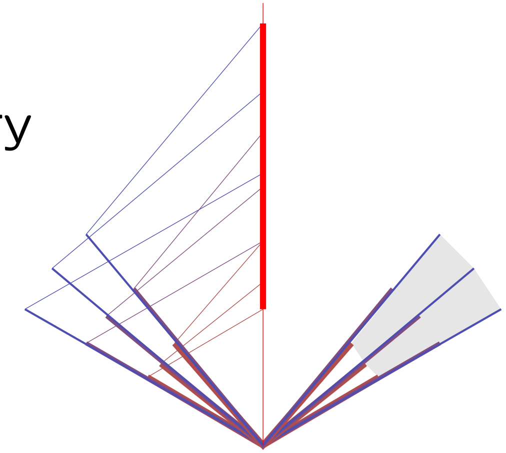


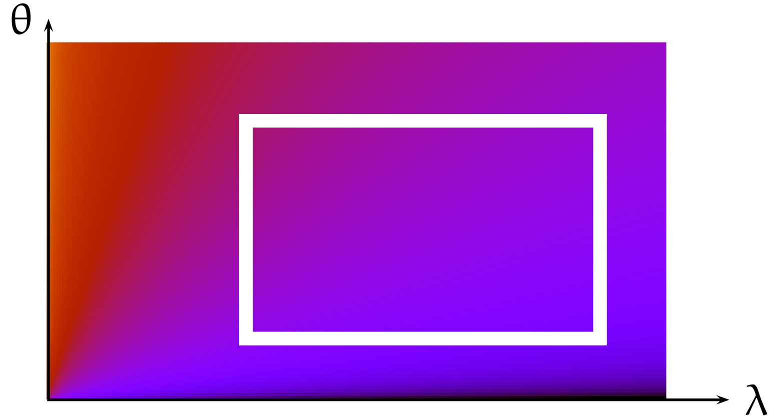
energy-dispersive reflectometry



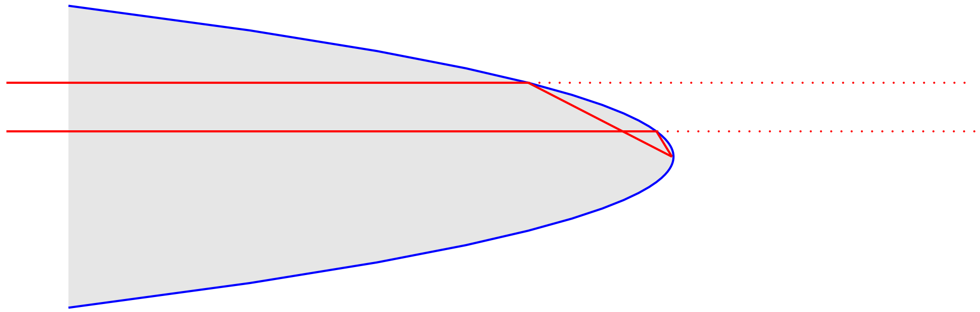


angle- and energy-dispersive reflectometry



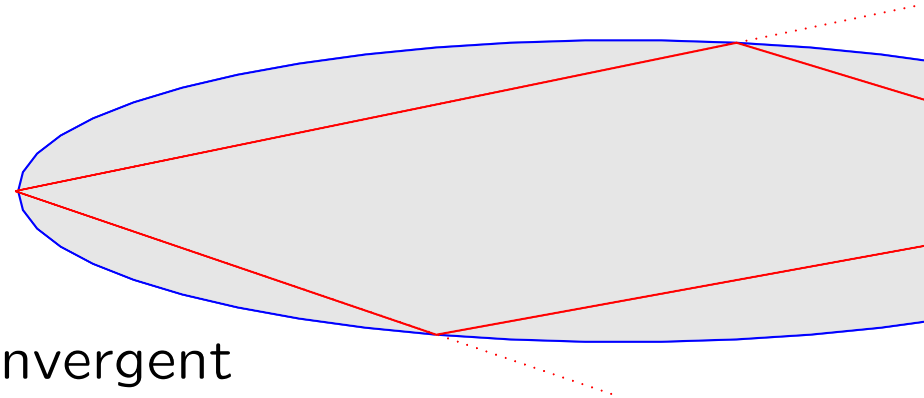


reflective focusing optics



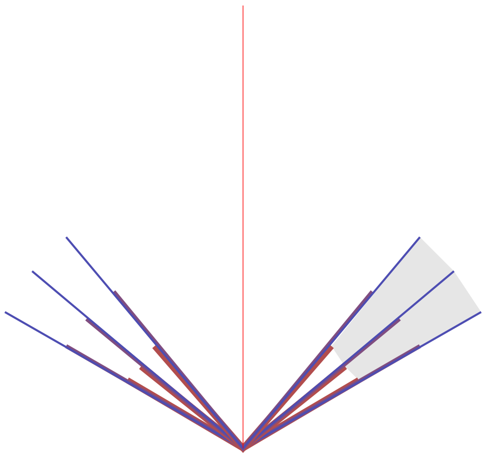
parabolic

parallel to convergent



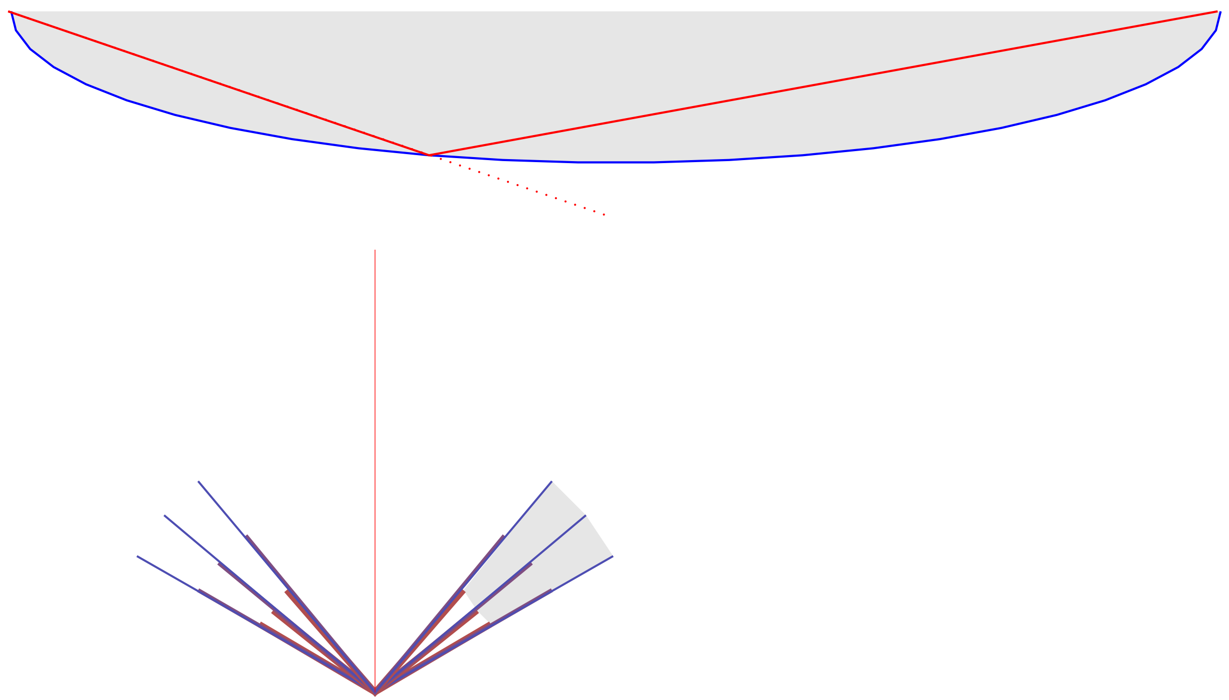
elliptic

divergent to convergent



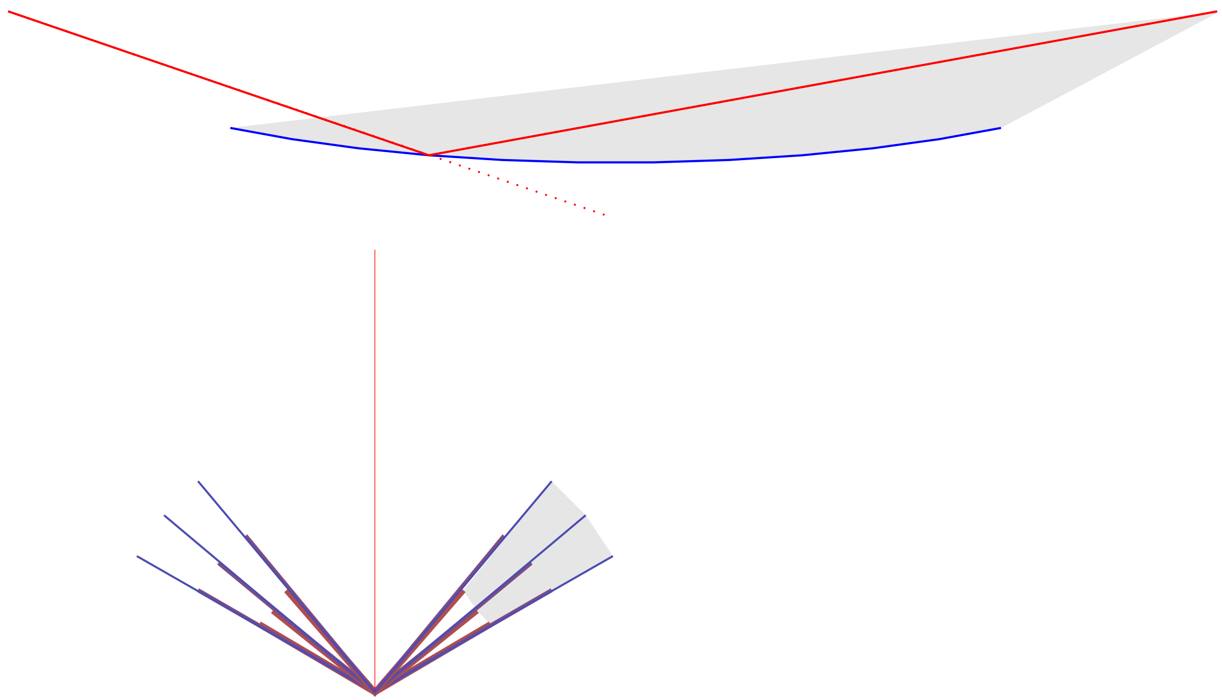
reflective focusing optics

elliptic



reflective focusing optics

elliptic



- Lithium Transport Through Thin Silicon Films
- high-intensity specular reflectometry
- the ***Selene*** guide
- Amor & ***Selene***
- reduction of the Li transport data
- ***Estia***

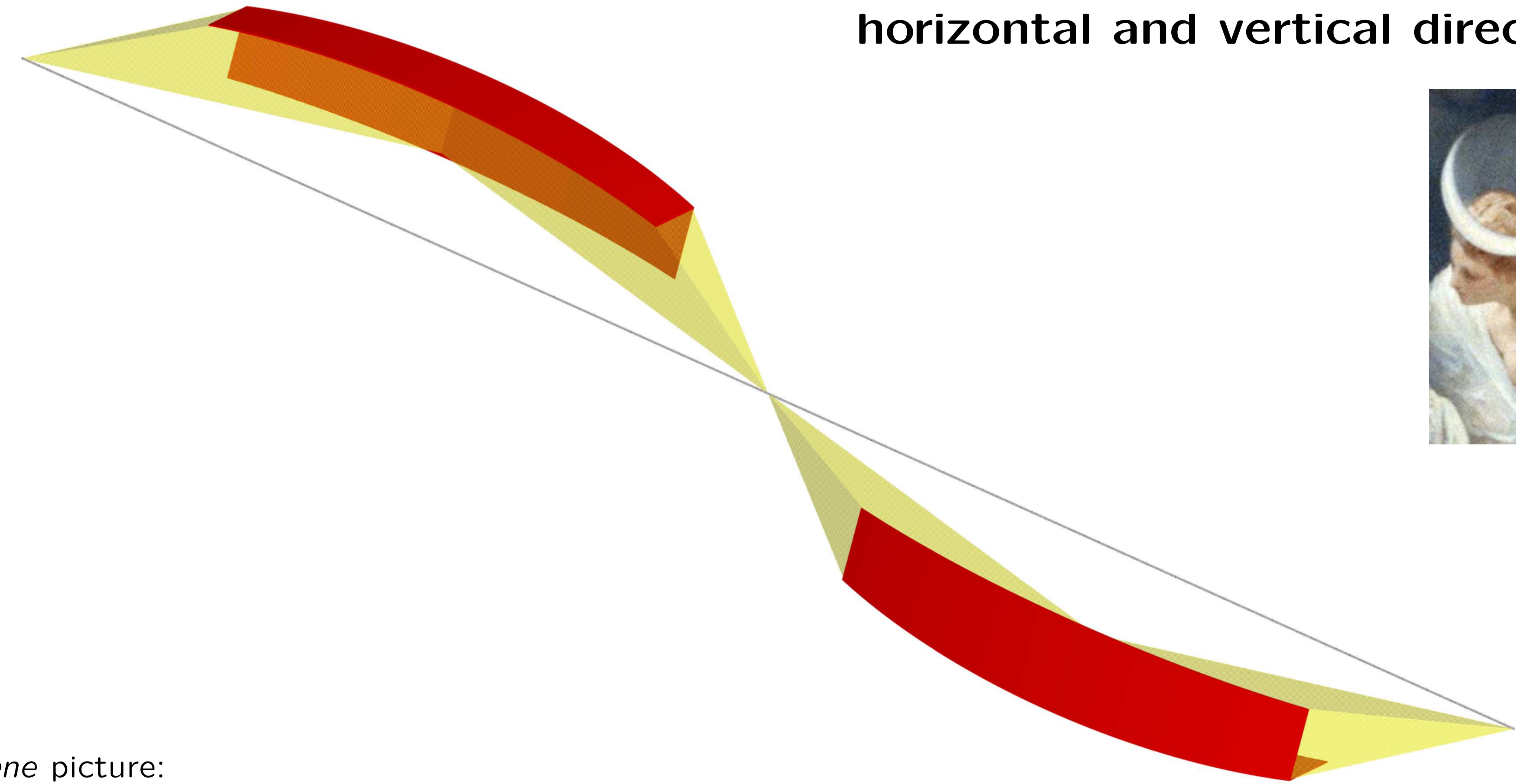
point-to-point focusing

with

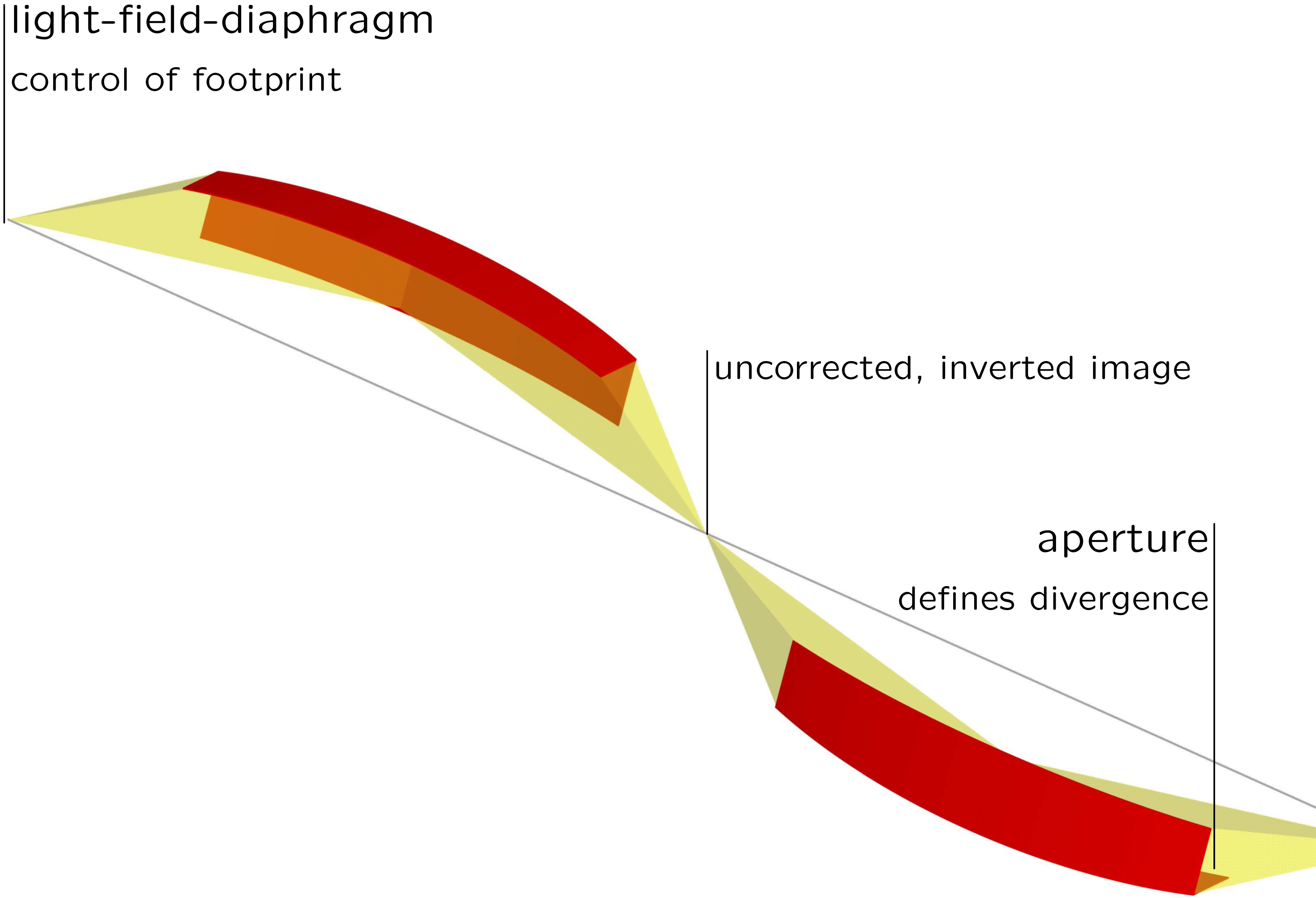
2 subsequent elliptical reflectors

for

horizontal and vertical direction

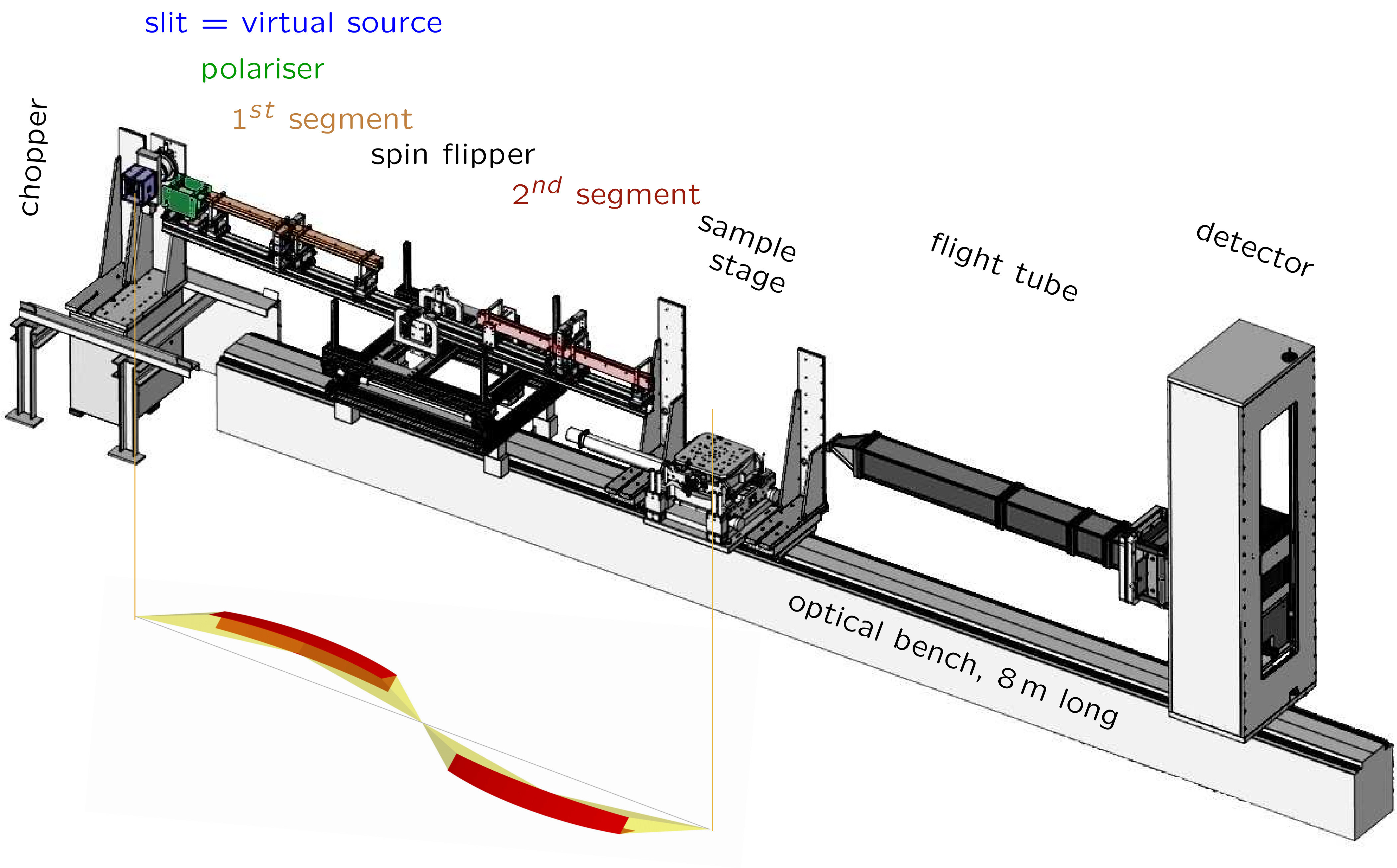


Selene picture:
ceiling painting in the Ny Carlsberg Glyptotek, København


light-field-diaphragm
control of footprint
uncorrected, inverted image
aperture
defines divergence
image
sample

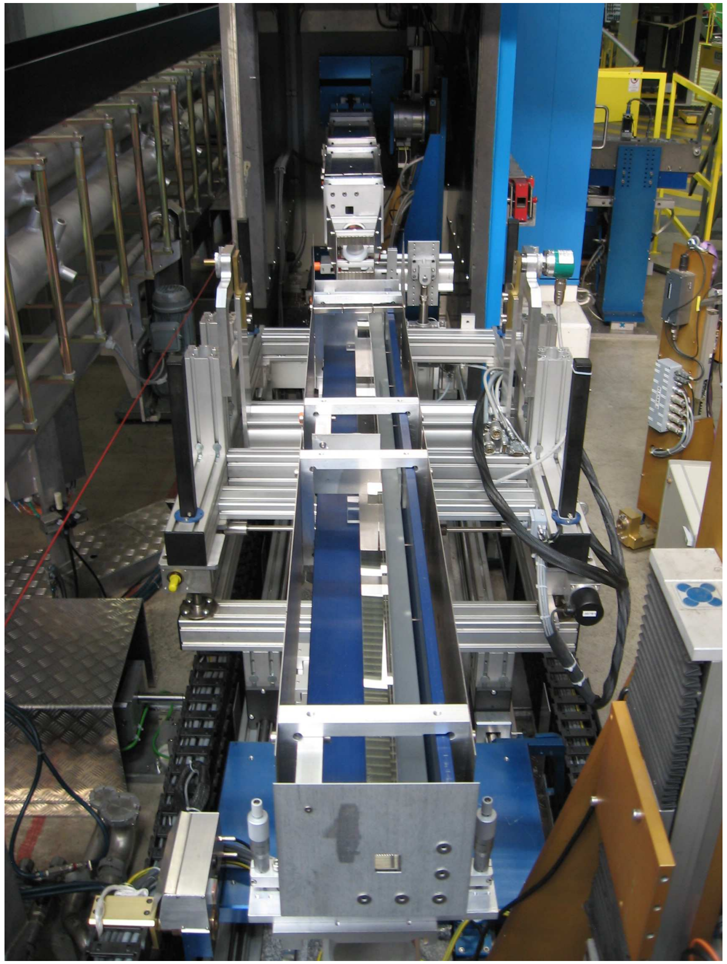
- Lithium Transport Through Thin Silicon Films
  - high-intensity specular reflectometry
    - the ***Selene*** guide
      - Amor & ***Selene***
        - reduction of the Li transport data
          - ***Estia***

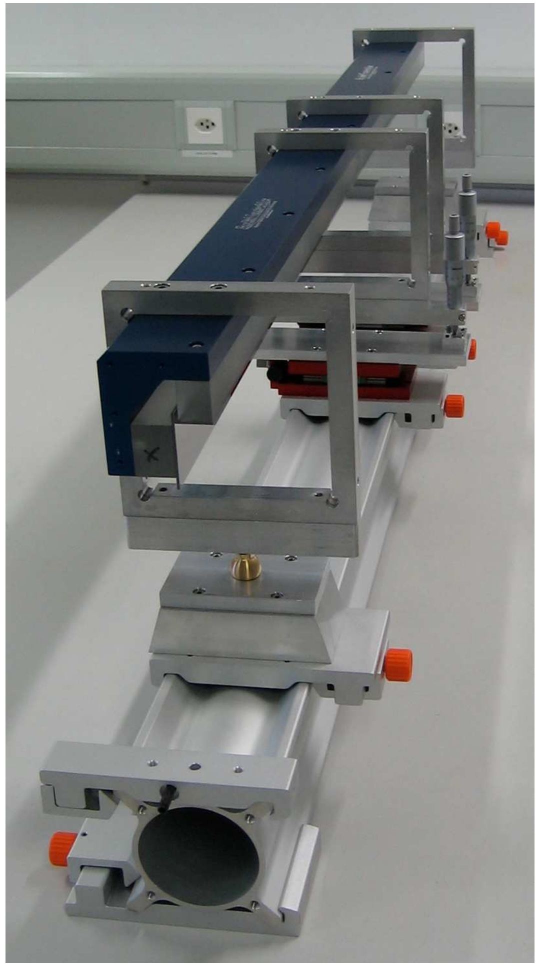
prototype guide on Amor@PSI



prototype guide on Amor@PSI

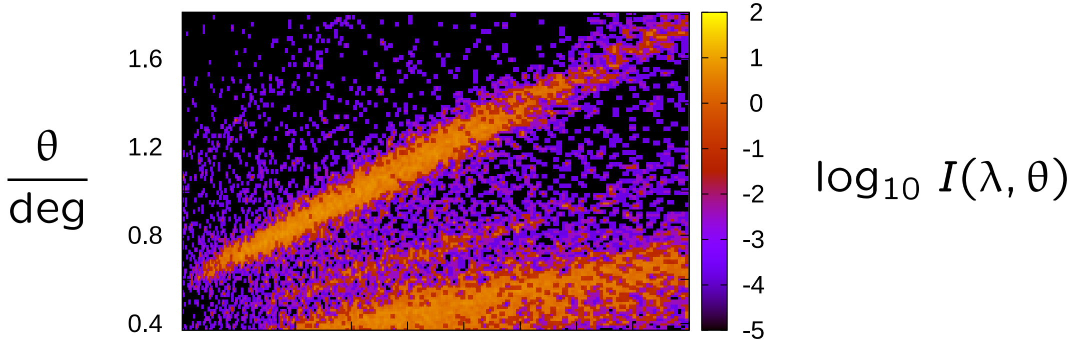
- total length $= 4\,\mathrm{m}$
- max spot size $\approx 2 \times 2\,\mathrm{mm}^2$
- divergence $\approx 1.8^\circ \times 1.8^\circ$





- Lithium Transport Through Thin Silicon Films
  - high-intensity specular reflectometry
    - the ***Selene*** guide
      - Amor & ***Selene***
        - reduction of the Li transport data
          - ***Estia***

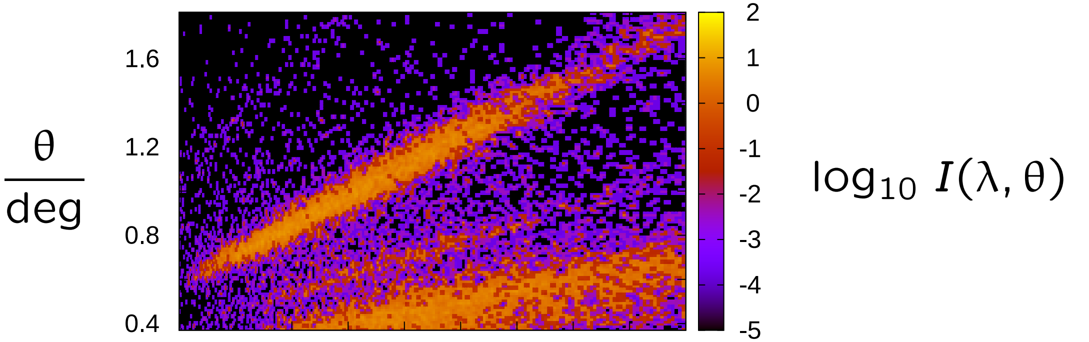
raw data



^{6}Li/Si/^{7}Li/Si multilayer
counting time 1.5 min

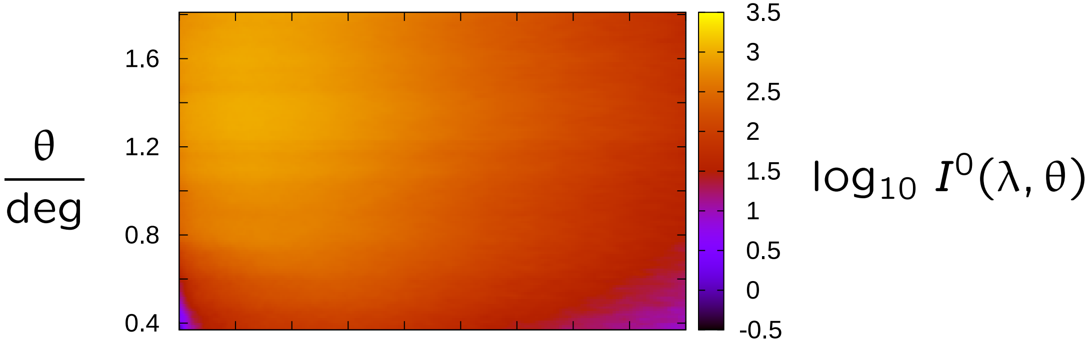
reduction of Li-transport data

raw data



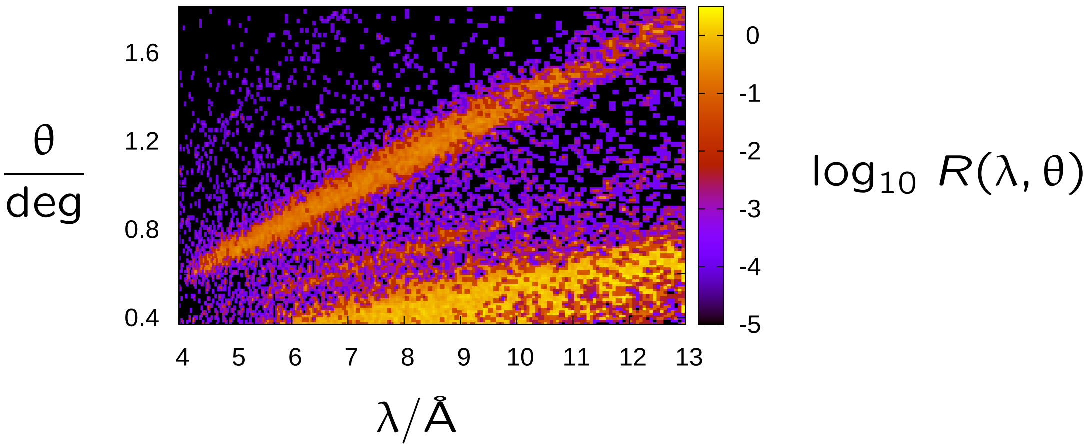
^{6}Li/Si/^{7}Li/Si multilayer
counting time 1.5 min

reference



$m = 5$ supermirror

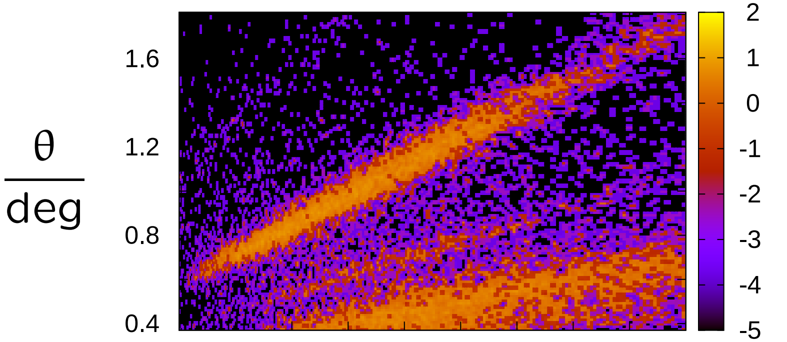
$R(\lambda, \theta)$



quotient

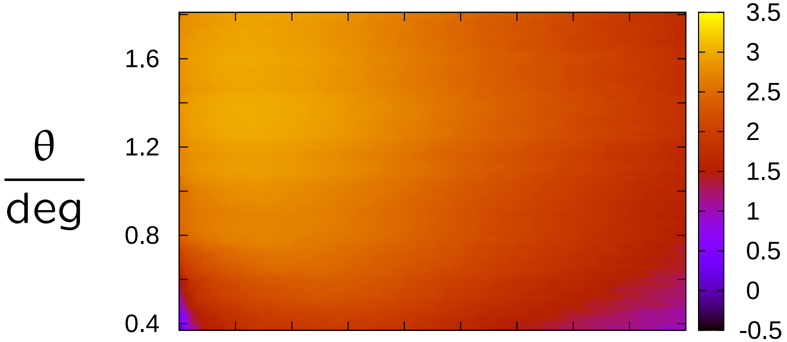
reduction of Li-transport data

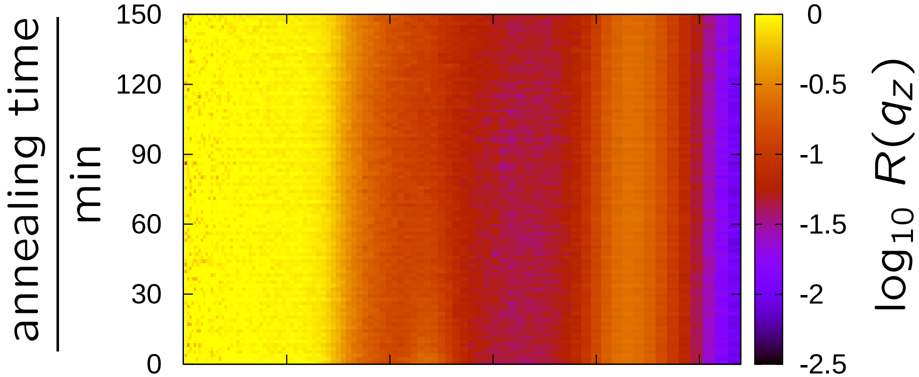
raw data



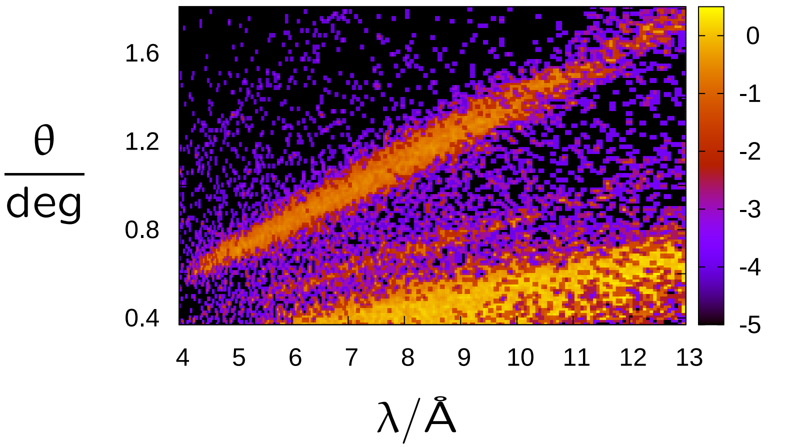
^{6}Li/Si/^{7}Li/Si multilayer
annealed at 240°C
continuously measured
with 1.5 min resolution

reference

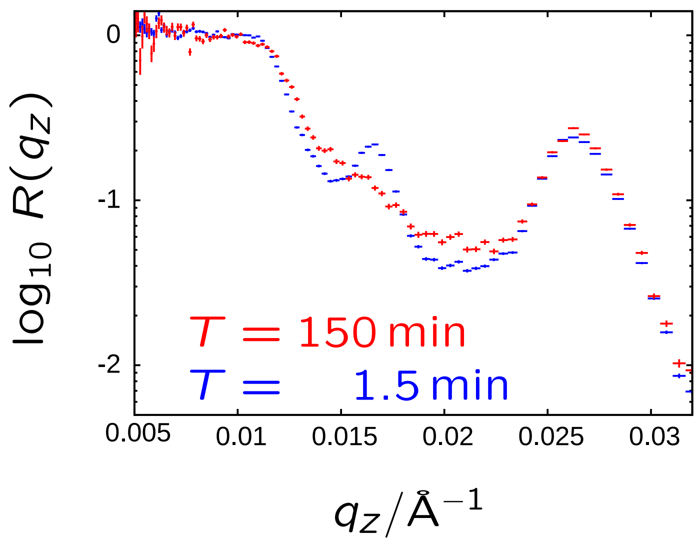




$R(\lambda, \theta)$



averaging along q_z



preliminary results

- 10 nm amorphous Si

permeability of Li in Si $P = (1.28 \pm 0.25) \cdot 10^{-16}\,\mathrm{cm}^2\,\mathrm{s}^{-1}$

- P and D depend strongly on Si film thickness

d_{Si} / nm	4	7	9	11	15	20
full intermixing after t / s	< 30	180	4 800	9 000	54 000	$> 90\,000$

- The Li-O-Si interface has no significant influence

◦ data analysis in progress

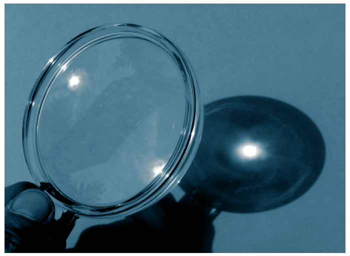
◦ further experiments are planned

- Lithium Transport Through Thin Silicon Films
  - high-intensity specular reflectometry
    - the ***Selene*** guide
      - Amor & ***Selene***
        - reduction of the Li transport data
          - ***Estia***

Estia a polarised focusing reflectometer for small samples

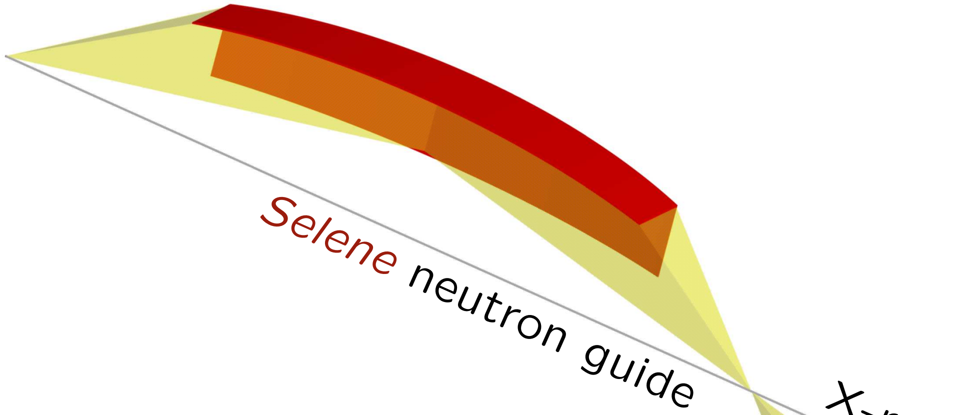
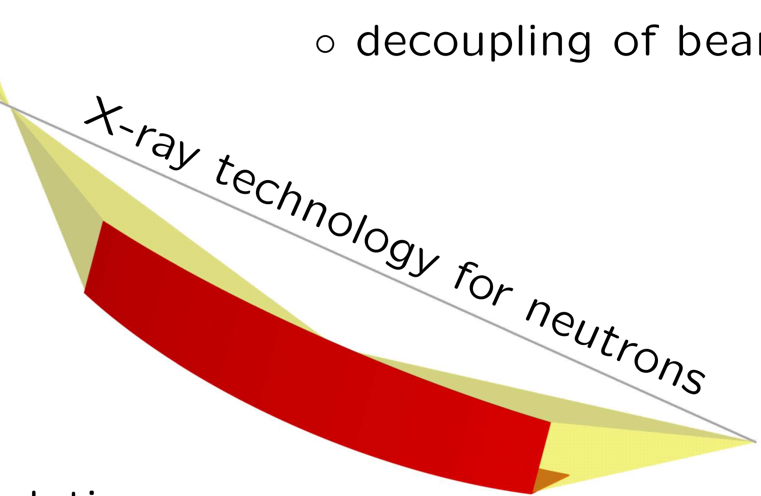
for the investigation of chemical and magnetic depth-profiles near surfaces and of lateral correlations and structures

functional devices	*spin-valves, spintronics*
diffusion processes	*Li batteries, corrosion protection*
multifunctional materials	*interface-coupled electric and magnetic properties*
towards *real* materials	*raster-scanning of bent, faceted or multi-domain surfaces*



loupe-like **neutron guide**

- new operation modes
- low background
- high flexibility
- decoupling of beam size and divergence

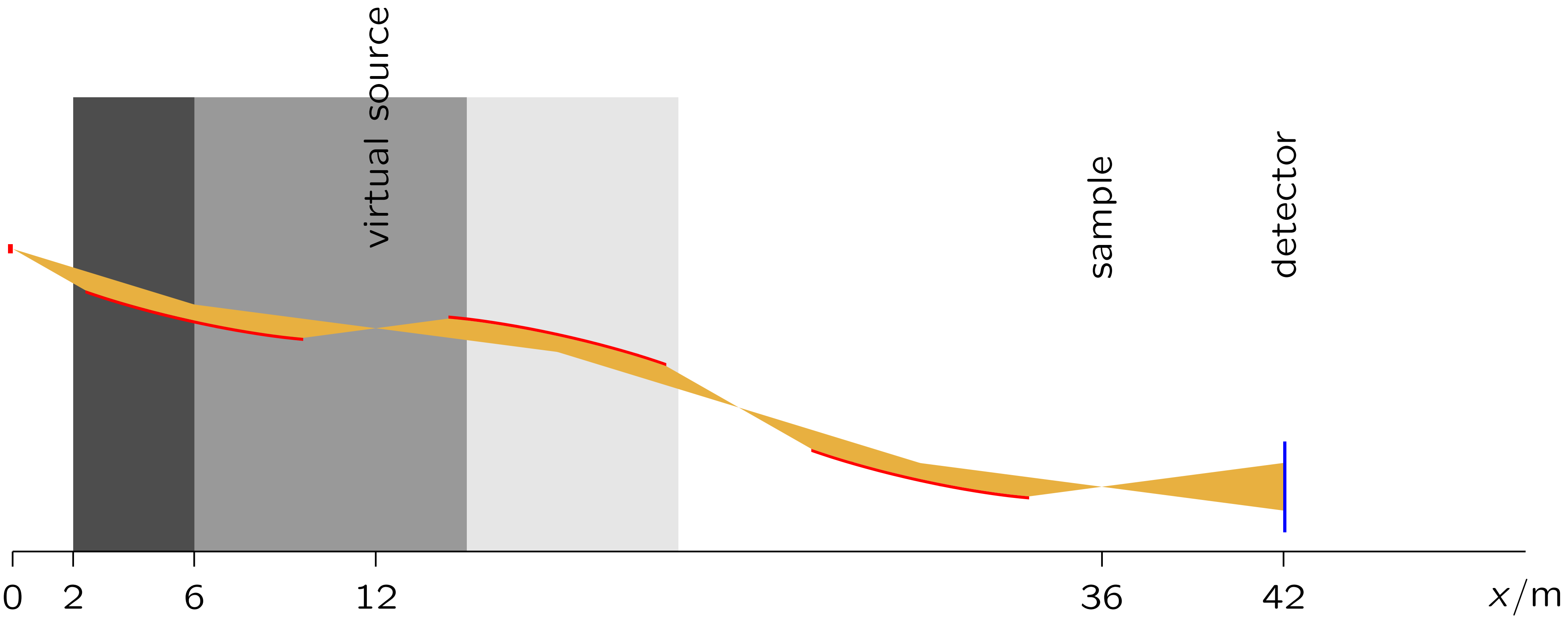
pushing the limits

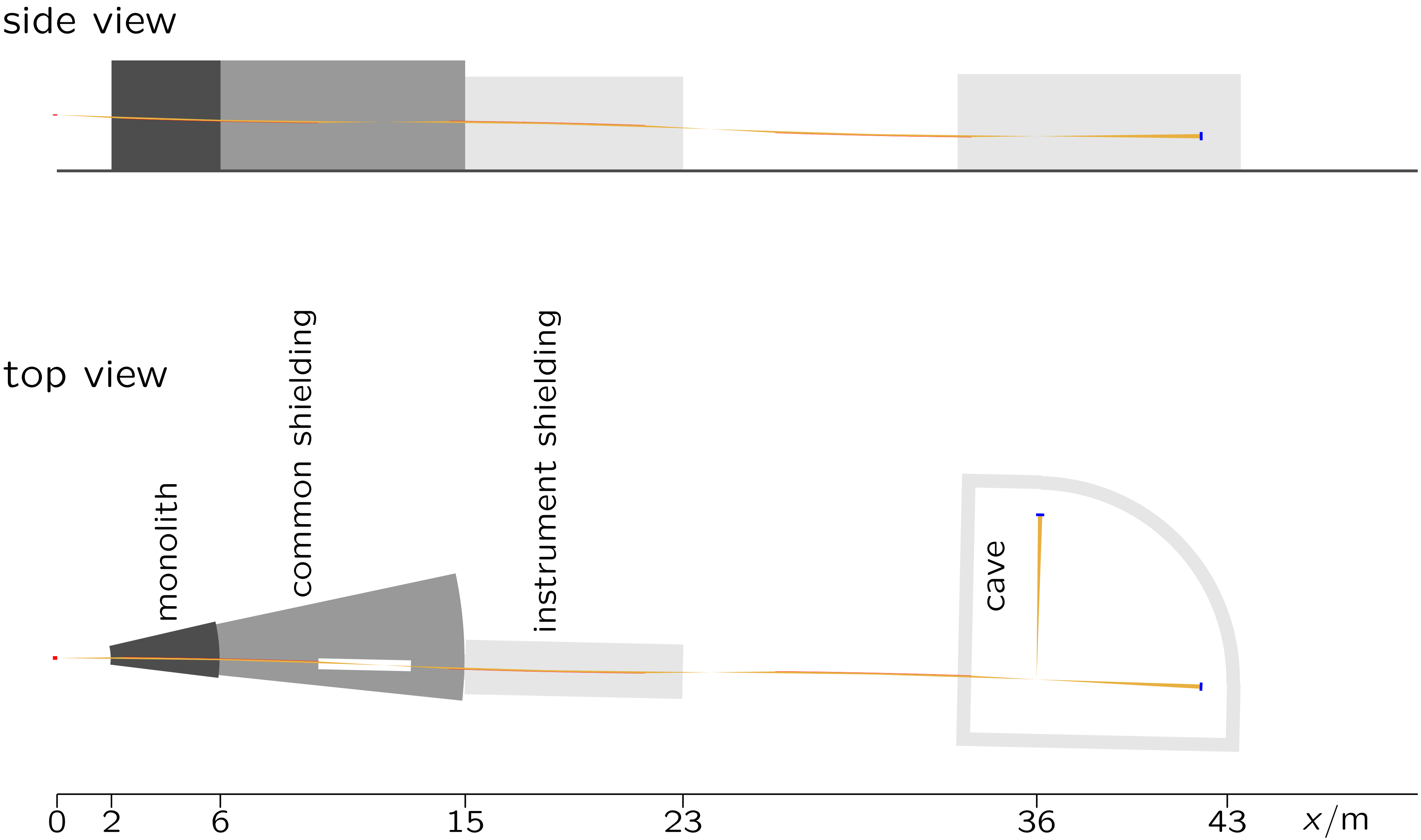
by 2 to 3 orders of magnitude for

- tiny samples ($< 1\,\mathrm{mm}^2$)
- fast measurements ($< 0.1\,\mathrm{sec}$)
- in-situ studies during growth or manipulation

TOF reflectometer for the ESS

- horizontal scattering plane
- sample size $< 10 \times 50\,\mathrm{mm}^2$
- divergence $1.5^\circ \times 1.5^\circ$
- $\lambda \in [4, 10]\,\text{Å}$





side view
top view
monolith
common shielding
instrument shielding
cave
0
2
6
15
23
36
43
x/m

comparison for high-intensity mode to *Selene* prototype on Amor

		factor
source flux	ESS / SINQ	150
guide transmission	Selene / Amor + Selene	4
footprint	(sample size $1\,\mathrm{cm}^2$)	10
in total		6 000

$\Rightarrow$ q_z-range $0.005\,\text{Å}^{-1}$ to $0.05\,\text{Å}^{-1}$ in one shot (0.07 s)

alternative use of flux:

- ∘ smaller samples ($< 1\,\mathrm{mm}^2$)
- ∘ higher q_z (smaller structures)
- ∘ off-specular scattering (lateral structures)

- Lithium Transport Through Thin Silicon Films
- high-intensity specular reflectometry
- the ***Selene*** guide
- Amor & ***Selene***
- reduction of the Li transport data
- ***Estia***

Thank you!