OPPORTUNITIES IN THE USE OF VERY COLD NEUTRONS FOR REFLECTOMETRY TECHNIQUES.

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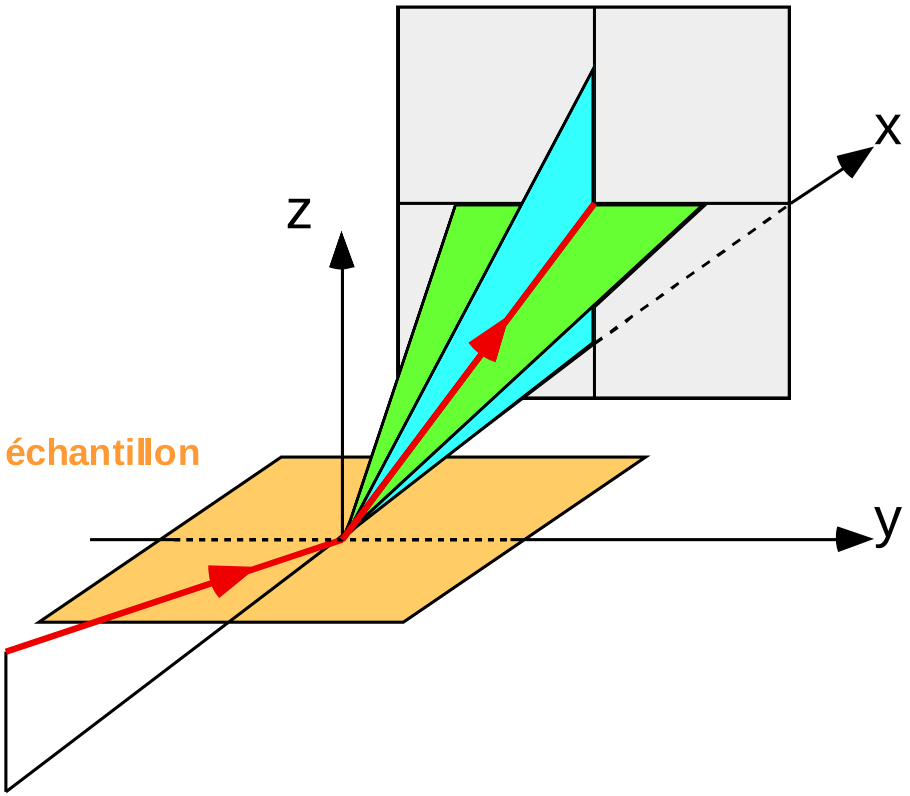
WHAT DO WE AIM AT PROBING ?

What science ?

- → nanoscale (1-10nm) structure of interfaces
  - Lower values : not accessible
  - Higher values : of little interest
- Only a structural method (so far)
- Polymer films interfaces, solid liquid interfaces, liquid surfaces, magnetic films



- Specular reflection
  $0.06 < Q_z < 3\ nm^{-1}$
  $1\ nm < \xi < 100\ nm$
- Incidence plane (Off-specular)
  $10^{-4} < Q_x < 10^{-2}\ nm^{-1}$
  $600\ nm < \xi < 60\ \mu m$
- ⊥ incidence plane (GISANS)
  $10^{-4} < Q_y < 3\ nm^{-1}$
  $3\ nm < \xi < 100\ nm$

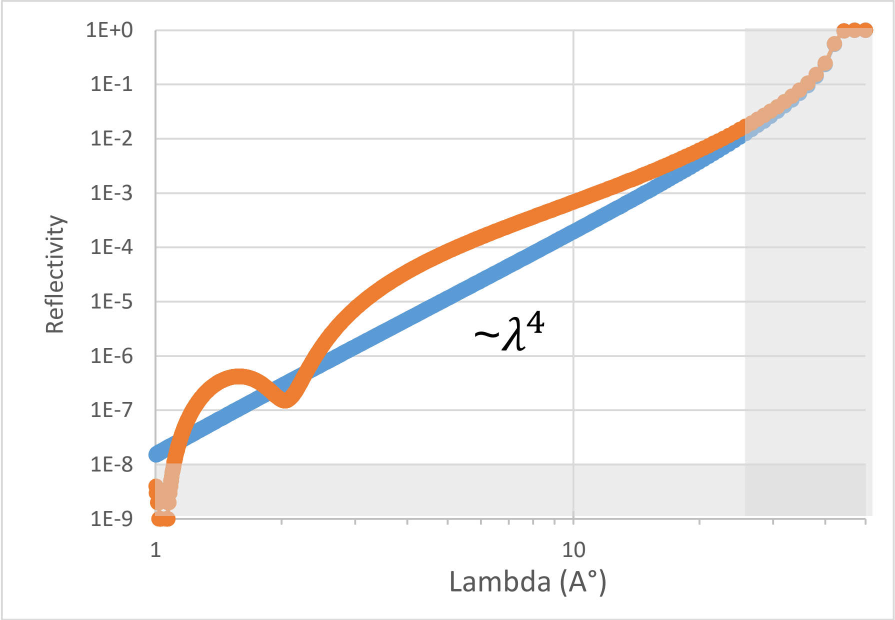
REFLECTOMETRY IS ESPECIALLY SUITED TO TOF

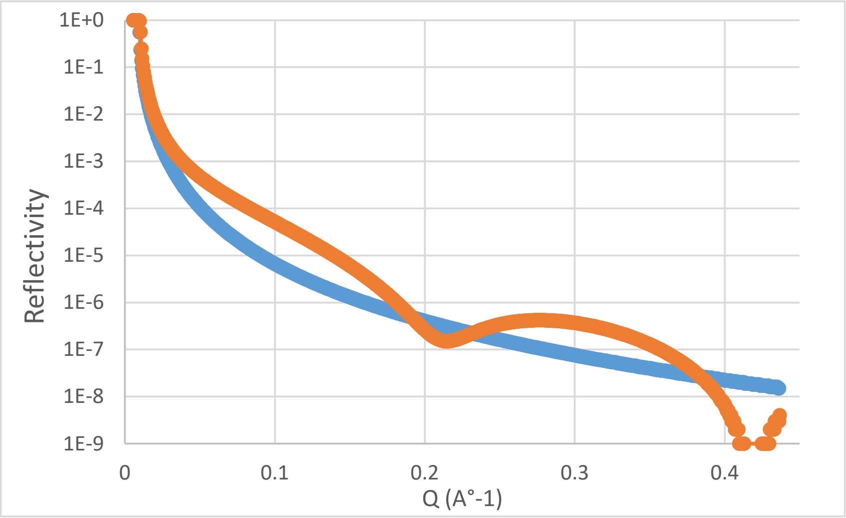
NOTA: the presentation focusses on VNC use at pulsed sources

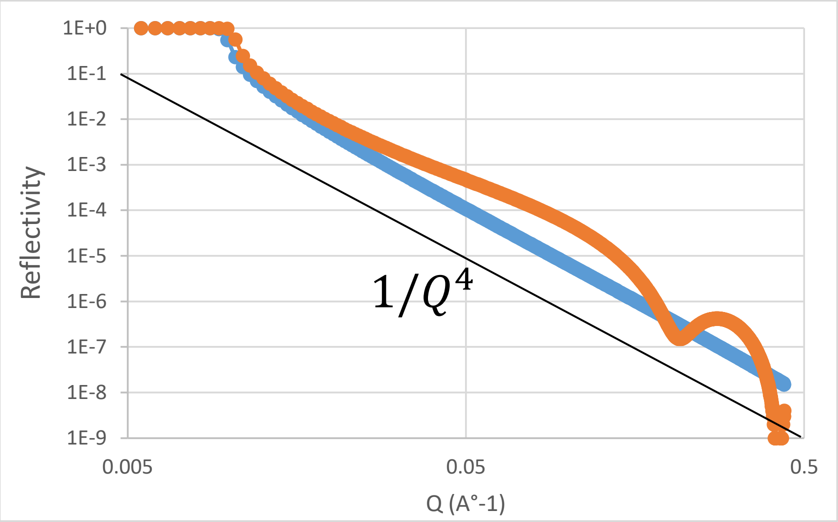
- Sample reflectivities follow a $1/Q^4$ trend with modulations around this general trend

$$R(Q) \propto 1/Q^4$$

- In « experimental space » the measurement looks as (incidence angle of 2°)

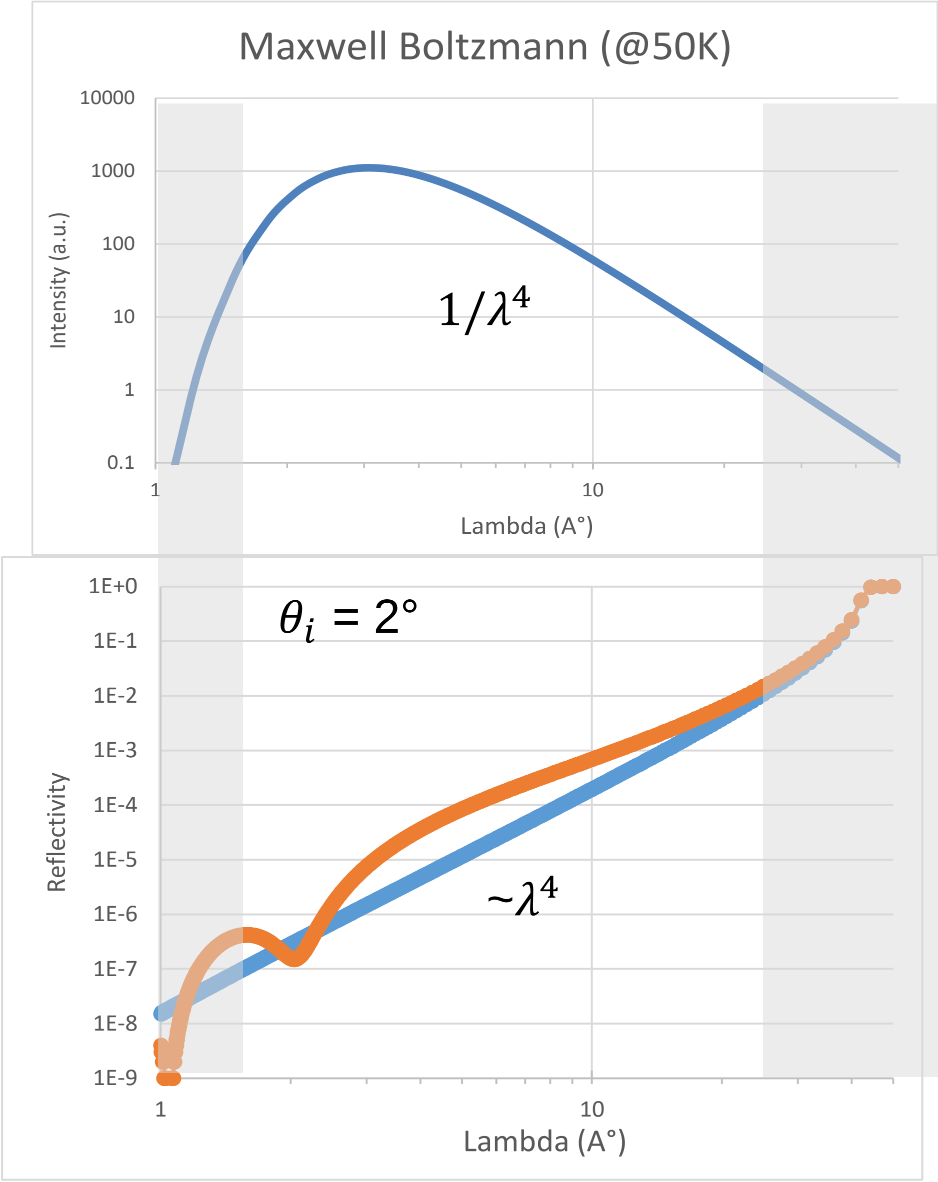






Si substrate and SiO_2//Si

MATCHING THE INCIDENT SPECTRUM AND THE REFLECTIVITY RANGE OF INTEREST



Reflectometry measurements on a long pulse source are using neutrons very efficiently in TOF mode.

- All neutrons provide information with a roughly equivalent statistics over a wide Q range (within a factor 3)

$$Q = \frac{4\pi}{\lambda}\sin\theta_i$$

The probed Q-range of interest can be tuned by simply changing the incidence angle θ_i

WHAT IMPACT OF USING VERY COLD NEUTRONS ?

- 1°/ The physics probed by the neutrons does not depend on the wavelength

- 2°/ For a given Q range of interest
  - If the wavelength spectrum is shifted by a factor α, it involves using incidence angles θ_i which are α times larger.
  - Assuming a constant $\delta\theta/\theta_i$, the angular resolution can be relaxed by a factor α (by simply relaxing the collimation slits)
  - There are second order corrections if the sample is over-illuminated or under-illuminated

$$Q = \frac{4\pi}{\lambda}\sin\theta_i \approx \frac{4\pi}{\lambda}\theta_i$$

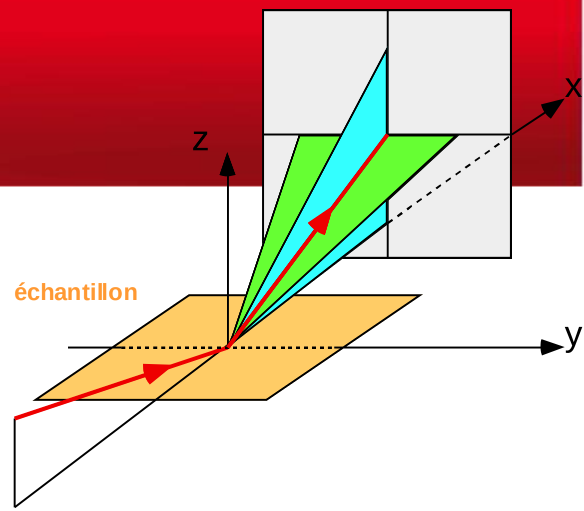
➔ The gain in flux is proportionnal to the wavelength spectrum shift α

- Note that for large samples (ex. Langmuir trough 100mm long), increasing the incidence angle from typically 4° to 12°, requires designing collimations which are on the order of 12m to fully benefit from the new wavelength spectrum → It is not a technical challenge but it is an unusual design for reflectometers.

- 3°/ The absorption effects remain usually negligible

- 4°/ There are no « multiple scattering » issues. The measurement remains as « clean and simple » as before whatever the incident wavelength spectrum.

OTHER IMPACTS OF VCN



Specular Reflectometry is 1D technique

Q directions perpendicular to the sample surface are probed

Optics improvement

- There are no phase space constrains in the direction perpendicular to the incidence plane Hence the beam can be focussed as much as possible in this direction
- The efficiency of reflective optics scales as *m x 0.1° / Å* where *m* is the reflectivity enhancement factor of the super mirrors compared to nickel mirrors.

- If the wavelength spectrum is shifted by a factor α, the angular phase space which can be used (perpendicular to the incidence plane) is also mutiplied by a factor α

- Beware that this has practical limitations
  - With a highly focussing optics in the plane perpendicular to the incidence plane, one has to ensure that all neutrons are collected by the detector.
  - Assuming for example an operation with wavelengths up to 50 *Å, and m=4* optics, the critical angle of reflexion is on the order of 20°. If the detector is set at 1.5m from the sample (=focal point), the footprint at the detector position is 1500 x 2 x sin(20°) = 1000mm !
- Assuming these technical constrains can be overcome, **the gain in flux is proportional to α**

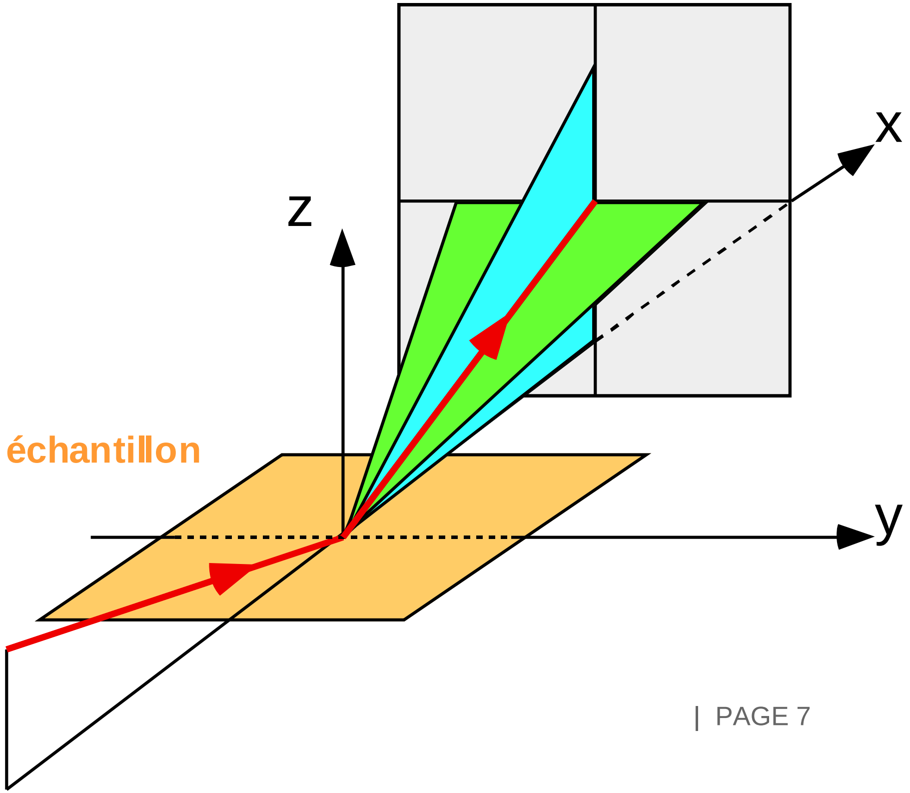
SPECIFIC CASE OF OFF-SPECULAR AND GISANS

Off-specular scattering

→ the same considerations as for specular reflectometry apply

GISANS

→ the focussing optics cannot be used



THE ISSUE OF TIME RESOLUTION

The ESS long pulse / low repetition rate is especially suited to low resolution techniques such as reflectometry.

Using VCN introduces extra constrains

Assuming that the wavelength spectrum is shifted by a factor α, and that the pulse length is kept constant (at 2.6ms),

- The instrument length would be increased by a factor α to keep the same resolution
- Any increase in the pulse length would lead
  to a proportionnal increase of the instrument length to compensate

- When the pulse time structure become too sub-optimal,
  other implementations could be considered but they would be less efficient and reduce the Q-range which can be measured (for a given θ_i)

- At the extreme limit, using a continuous VCN source would lead to only marginal improvements compared to ILL instruments.

CASE OF A CONTINUOUS SOURCE

The above considerations apply also for reflectometry implemented on a continuous source

- Either on a fixed wavelength instrument where the operation wavelength can be shifted to higher values
- Or on a chopper TOF instrument for which the very same considerations apply

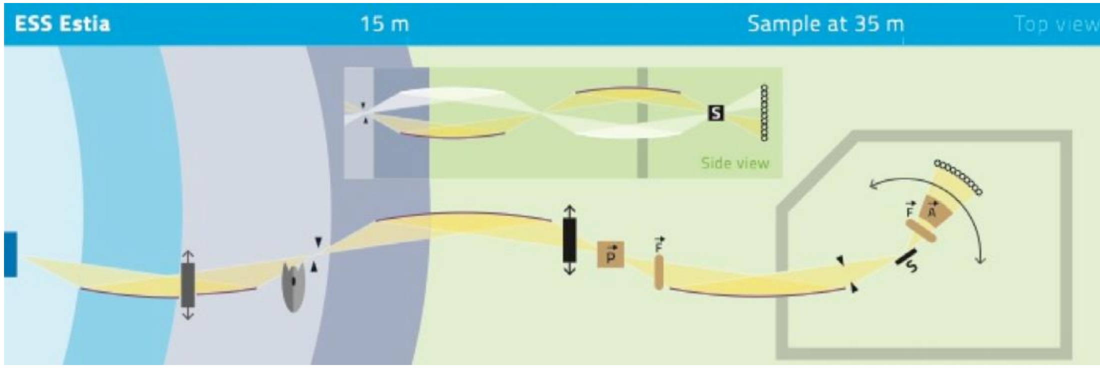
OTHER MORE EXOTIC TECHNIQUES

In the previous decades other implementation of reflectometry measurements have been proposed:

- **Energy analysis of a white beam** (via refraction or optics, ex. RAINBOW or EASYREF) Such techniques are efficient only on continuous sources but would strongly benefit from a shift to longer wavelengths
  - Optics efficiency is proportional to λ (*m x 0.1° / Å)*
  - Refraction efficiency is proportional to λ^2

- **SERGIS** techniques using spin-echo techniques
  - Refer to the discussion on the Spin-Echo techniques with VCN

- **SELENE type (→ ESTIA@ESS)**
  - Optics efficiency is proportional to the wavelength (*m x 0.1° / Å)*
  - Technical implementation is much easier
    - Either shorter optics or broader λ and angular range



SUMMARY

The time structure of ESS is ideally suited for reflectometry experiments → hard to compete

Assuming a temperature shift from 20K to 5K,

- The wavelength spectrum would be shifted by a factor 2

Technique	Gain	20K → 5K
Specular reflectivity	$\propto \lambda^2$	4
Off-specular reflectivity	$\propto \lambda^2$	4
GISANS	$\propto \lambda$	2

- Assuming that the integrated VCN flux is identical to the CN flux (brightness value)
- Assuming the time structure is not too dilated (pulse < 10ms)
- VCN neutrons could be practically useful up to a shift of a factor 3 in the wavelength spectrum.
- Beware that neutrons are falling in the gravity field
  → challenge to achieve clean horizontal instrument measurements