JOINT INSTITUTE FOR NUCLEAR RESEARCH
Frank Laboratory of Neutron Physics

POLARIZED NEUTRON REFLECTOMETRY WITH VERY COLD NEUTRONS

Zhaketov Vladimir

UCN and VCN Source at the Institute of Nuclear Physics, Kazakhstan and their applications.
Institute of Nuclear Physics, Almaty, Kazakhstan, 8–11 Apr 2024.

OUTLINE

1. Motivation
2. Geometric advantages
3. Possibilities for effects increasing with wavelength
4. Technical design
5. Type of layered systems
6. Advantages for inhomogeneous systems with $\xi > 10\ nm$

THE QUESTION OF USING VERY COLD NEUTRONS (≈15-100 Å) FOR POLARIZED NEUTRON REFLECTOMETRY IS OFTEN RAISED

FOUR REFLECTOMETERS WITH POLARIZATION ANALYSIS PROPOSED FOR NEW SOURSE IN DUBNA (NEPTUN), BUT "WHITE SPOT“ IS NEUTRON REFLECTOMETRY WITH VCN

No.	REFLECTOMETER	PURPOSE OF THE SPECTROMETER
1	HIGH FLUX STANDARD TYPE REFLECTOMETER	MAGNETIC STRUCTURE OF BILAYERS AND MULTISTRUCTURES; MAGNETIC AND NUCLEAR STRUCTURE OF THE SURFACE AND THIN MAGNETIC LAYERS
2	REFLECTOMETER FOR STUDYING LIQUIDS WITH HORIZONTAL GEOMETRY	FREE SURFACE OF LIQUIDS, MAGNETIC LIQUIDS, BIOLOGICAL SYSTEMS, POLYMER FILMS
3	REFLECTOMETER WITH LARMOR PRECESSION USING	DIFFUSION AND VIBRATIONS OF MACROMOLECULES AND CLUSTERS ON THE SURFACE AND IN THE LAYERS OF THE STRUCTURE
4	HIGH-RESOLUTION REFLECTOMETER WITH REGISTRATION OF SECONDARY RADIATION	MAGNETIC AND NUCLEAR STRUCTURE OF A SINGLE INTERFACE OF A LAYERED STRUCTURE

AGAIN QUESTION RAISED FOR UCN AND VCN SOURCE AT THE INSTITUTE OF NUCLEAR PHYSICS IN KAZAKHSTAN

The first thoughts when talking about very cold neutrons for polarized neutron reflectometry (not only):

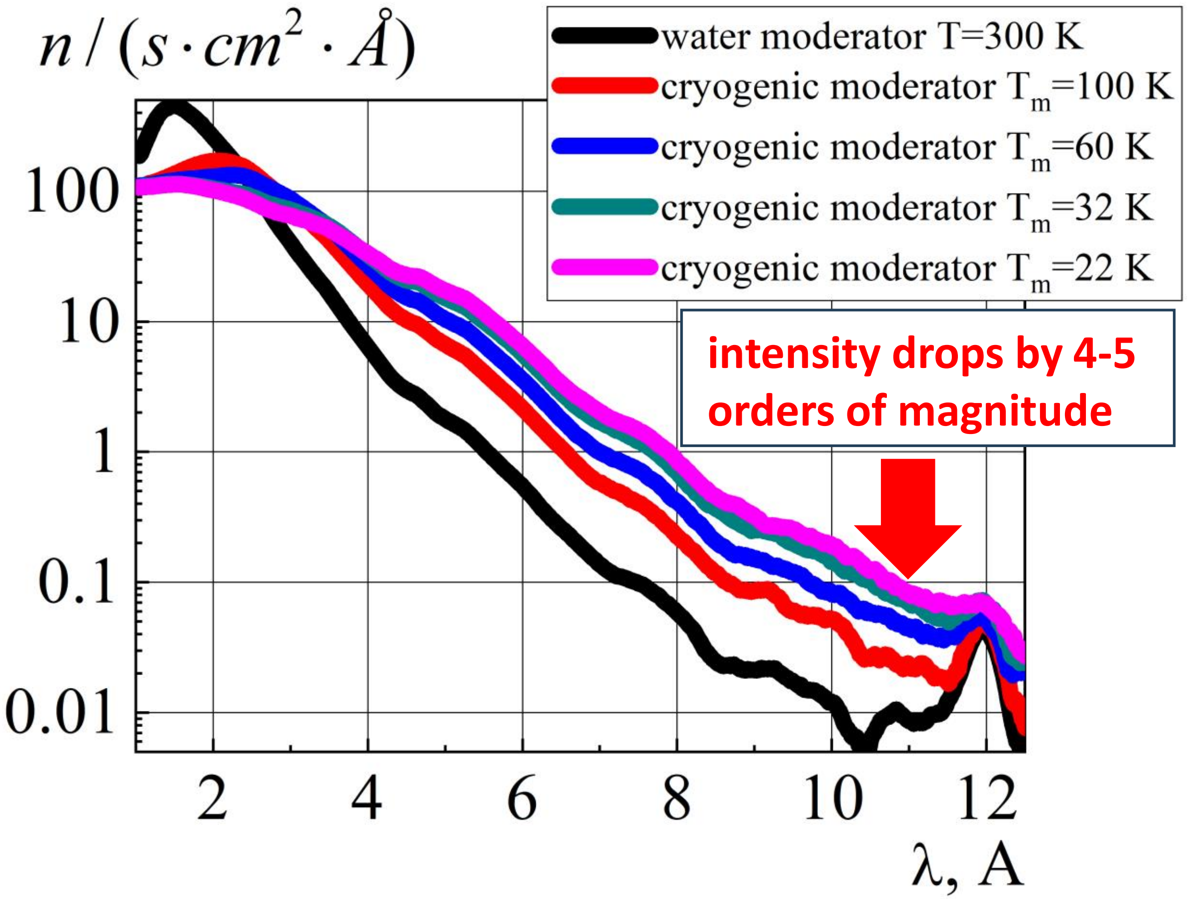
- **Maxwellian losses (low flux)**
- **Corrections for free fall in the Earth's gravitational field**

$L = 60\ m$, $\lambda = 60$ Å → $\Delta h = \frac{g}{2}\left(\frac{m_n \cdot L \cdot \lambda}{2 \cdot \pi \cdot \hbar}\right)^2 \approx 4\ m$!!!

(overlapping at $\nu = \frac{3.96 \cdot 10^3}{L \cdot \lambda} > 1\ Hz$)

$\nu' = 5\ Hz$, $L = 15\ m$ → $\lambda_{max} = 52.8$ Å → $\Delta h \approx 0.2\ m$

- **But in further discussions we will ignore these disadvantages and consider the advantages of using very cold neutrons for PNR**
- **Outside report detector systems**
  Oliver Kirstein et al. // arXiv:1411.6194v1
  λ_{max} **=23 Å ?**



Dependence of neutron flux on wavelength on the REMUR spectrometer at different temperatures of the cryogenic moderator.

Known literature on the use of VCN for PNR

Yu. N. Pokotilovski.
On the Possibility of Investigations with Very Cold Neutrons at Pulsed Sources //
Physics of Particles and Nuclei Letters, 2018, Vol. 15, No. 1, pp. 83–91.

Frédéric Ott.
Opportunities in the use of Very Cold Neutrons in reflectometry techniques //
Journal of Neutron Research 24 (2022) 211–221.

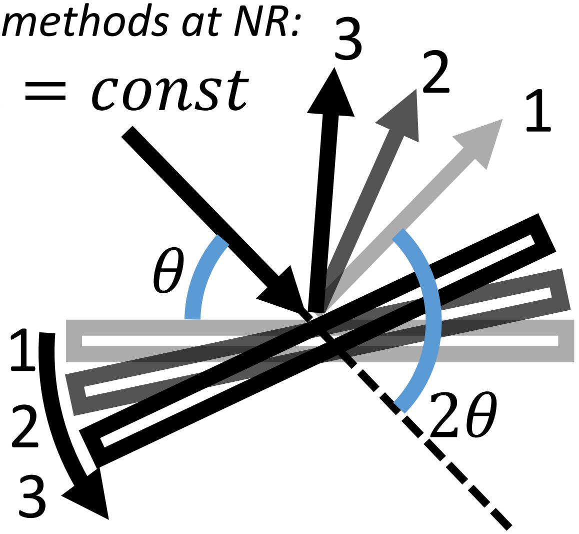
Ferenc Mezei.
Very cold neutrons in condensed matter research //
Journal of Neutron Research 24 (2022) 205–210.

A.K. Petukhov, V.V. Nesvizhevsky, T. Bigault, P. Courtois, D. Jullien, T. Soldner.
Advanced broad-band solid-state supermirror polarizers for cold neutrons //
arXiv:1606.01960.

NEUTRON REFLECTOMETRY AT PULSED SOURCE

Two measurement methods at NR:

$\lambda = const$

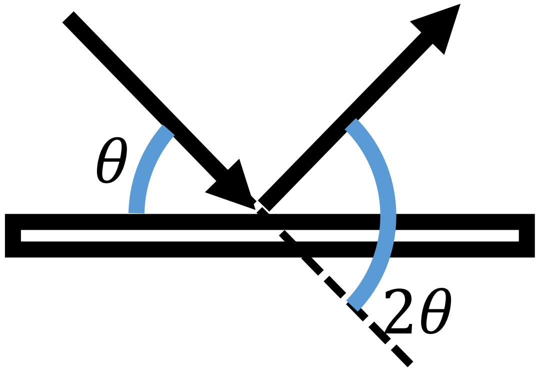


$$R(Q) = {}^{I_R}/_{I_0}$$

$$Q = \frac{4\pi}{\lambda}\sin(\theta)$$

TOF (time-of-flight)

$\theta = const$



$$\lambda \sim \frac{TOF}{L}$$

- On a constant source, both methods give approximately the same intensity
- Time-of-flight technology is more convenient for in situ measurements in real time
- TOF - A reflectometer on a pulsed source is more effective than on a stationary source
- Two thirds of the world's neutron reflectometers are time-of-flight installations

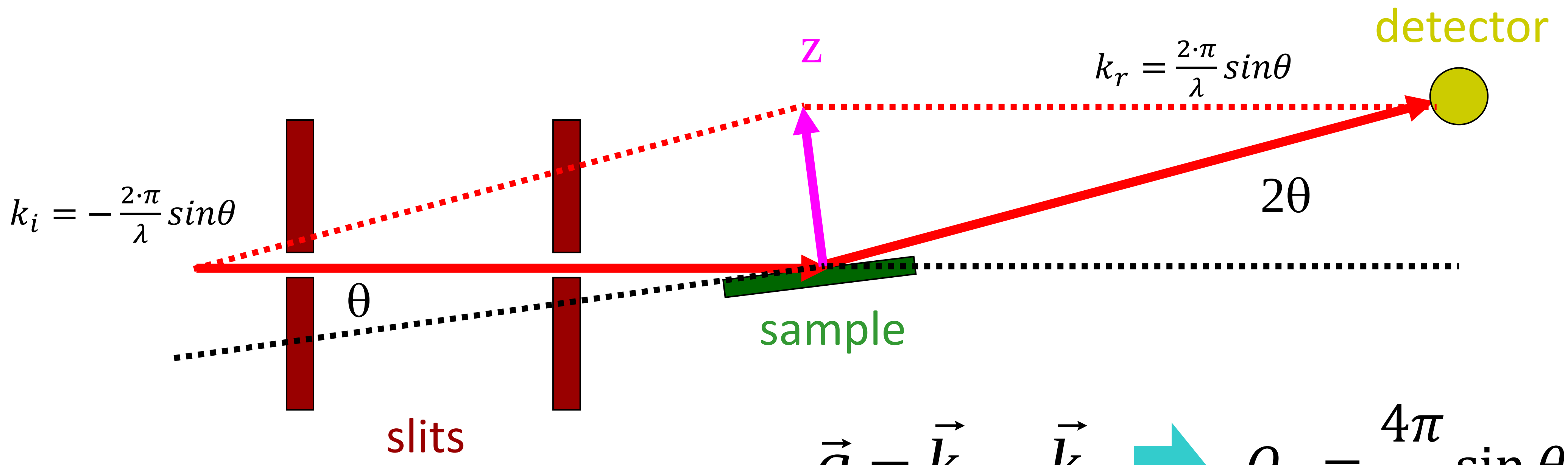
V.I. Bodnarchuk et al. // Crystallography, 2022, vol. 67, №1, pp. 57-71.
V.D. Zhaketov et al. // JETP, 2017, Vol. 125, No. 3, pp. 480–494.
Devishvili A. et al. // Review of scientific instruments 84, 025112 (2013)
R.A. Campbell et al. // Eur. Phys. J. Plus (2011) 126: 107.
F. Piscitelli. // arXiv:1701.07623v1 (2017)
Roland Garoby et al. // Phys. Scr. 93 (2018) 014001 (121pp).

Parameters	SuperADAM (ILL)	FIGARO (ILL)	ESTIA (ESS)	REMUR (IBR-2)
Neutron flux at the sample, $n{\cdot}cm^{-2}{\cdot}s^{-1}$	$8{\cdot}10^4$	$4{\cdot}10^4$	$(1÷5){\cdot}10^7$	$3{\cdot}10^5$
Average power, MW	58.3	58.3	5	2

So, neutron reflectometry measurements on a pulse source (or steady flux source with fermi-choppers) are very efficiently in TOF mode

REFLECTOMETRY EXPERIMENT

- method for studying thin films and interfaces with 1 nm resolution
- magnetic and superconducting heterostructures
- polymer films, biology systems, surface of liquids, magnetic liquids



$$\vec{q} = \vec{k}_r - \vec{k}_i \Rightarrow Q_z = \frac{4\pi}{\lambda}\sin\theta$$

q → momentum transfer

2ϑ → scattering angle

ESTIMATES FROM SIMPLE CONSIDERATIONS

Typical parameters for neutron reflectometry:

$$\theta \approx 3 \div 15\ mrad$$

$$\lambda \approx 1 \div 8\ Å$$

It correspond to momentum transfer:

$$Q_z \approx 0.05 \div 1.9\ nm^{-1}$$

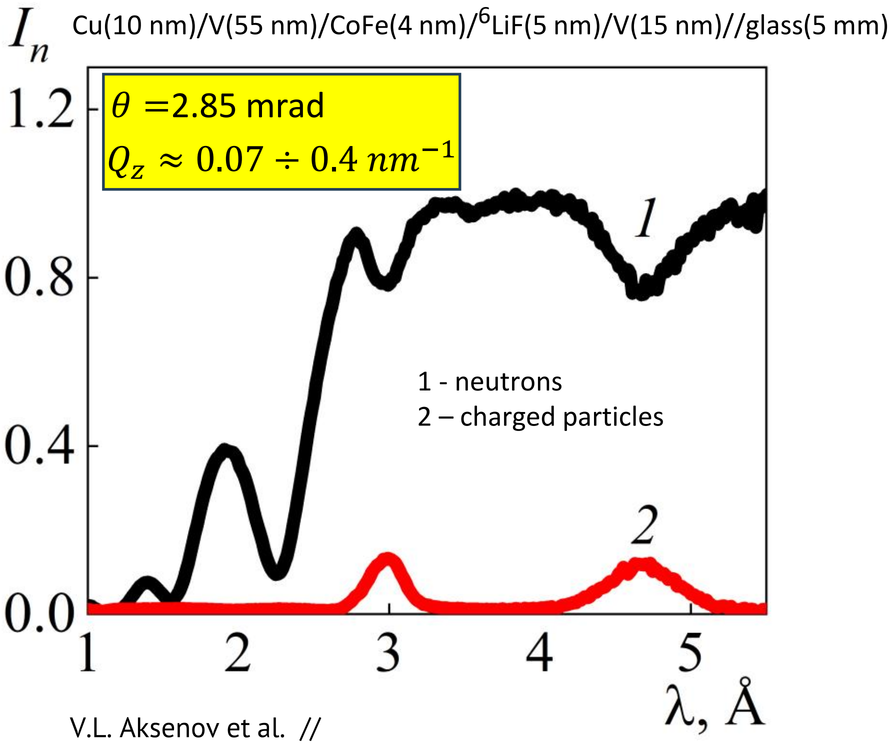
Wavelength of VCN:

$$\lambda \approx 15 \div 100\ Å$$

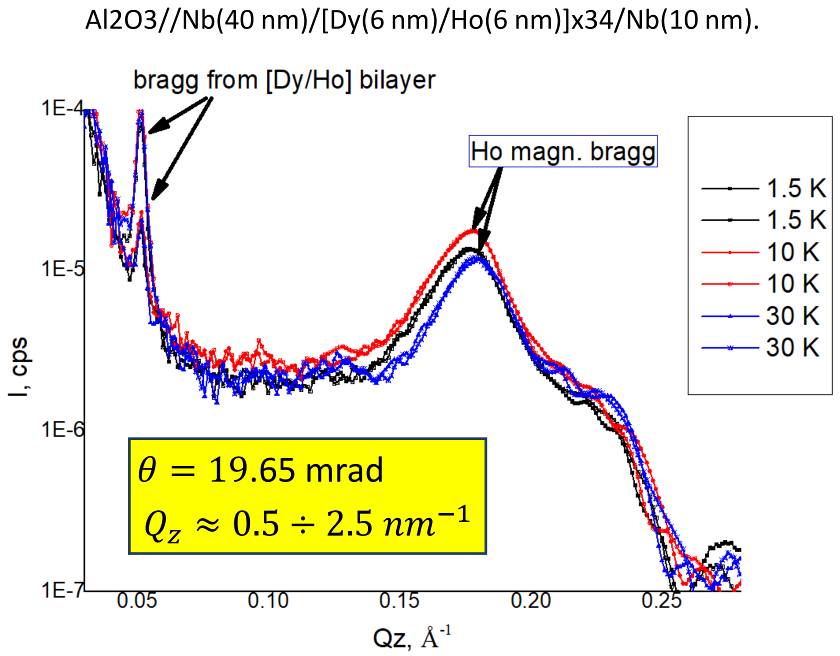
For the same Q_z, we get:

$$\theta \approx 6 \cdot 10^0 \div 1.5 \cdot 10^3\ mrad$$

SOME REFLECTIVITY FROM REMUR AS EXAMPLE



V.L. Aksenov et al. //
Physics of Particles and Nuclei, Vol. 54, No. 4, pp. 756-775 (2023).



Moving from units of milliradians to units of degrees, the adjustment process becomes easier

PERFORMANCE IMPROVEMENTS RELATED TO GEOMETRY ON A REFLECTOMETER USING VERY COLD NEUTRONS

Frédéric Ott. Opportunities in the use of Very Cold Neutrons in reflectometry techniques // Journal of Neutron Research 24 (2022) 211–221.

$$\frac{\delta\theta}{\theta} = const$$

for VCN:

$$\theta \uparrow \;\Rightarrow\; \delta\theta \uparrow$$

Close to REMUR parameters:

$$\frac{\delta\theta}{\theta} = 5\% \;\Rightarrow\; \begin{array}{|l} \theta = 5\,mrad \\ \delta\theta = 0.25\,mrad \end{array}$$

$$\frac{2W}{L} = \delta\theta \qquad L = 2\,m$$

distance between slits

$$W = \frac{L\cdot\delta\theta}{2} = \frac{2\cdot 0.25\cdot 10^{-3}}{2} = 0.25\,mm$$

slit

for VCN: $\theta = 50\,mrad$

$$\delta\theta = 2.5\,mrad$$

$$W' = \frac{2\cdot 2.5\cdot 10^{-3}}{2} = 2.5\,mm$$

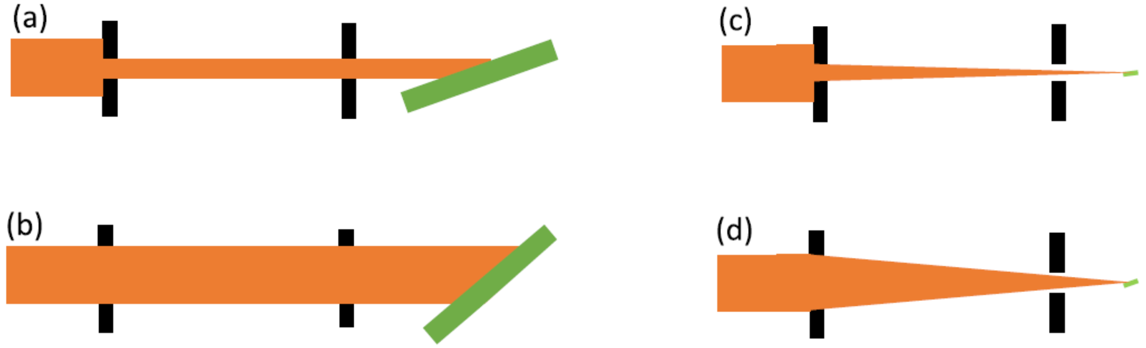


Fig. 4. (a) and (b): Illumination scheme of large samples with cold (a) and very cold (b) neutrons, leading to equivalent resolution. (c) and (d): Illumination scheme of small samples with cold (c) and very cold (d) neutrons, leading to equivalent resolution. Sample is shown from the side.

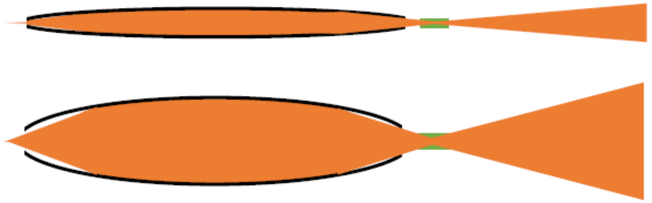


Fig. 5. If the wavelength spectrum is shifted to longer wavelengths, the focusing optics can be designed to accommodate a large transverse divergence $\delta\theta_y$ (sample seen from the top).

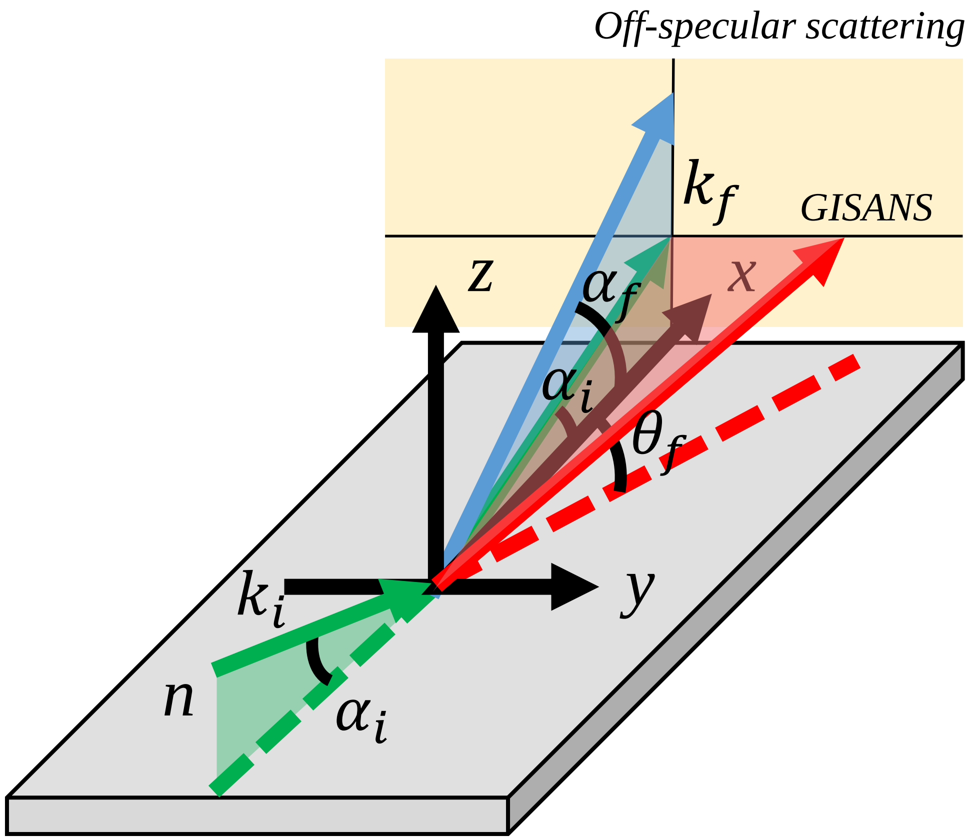
$$\theta \uparrow \;\Rightarrow\; \delta\theta \uparrow \quad W \uparrow \quad d_{eff} \uparrow \;\Rightarrow$$

this approach allows to benefit for the reflected neutron flux (10-100 times ?)

"The performances of reflective optics scale as m × 0.1° / Å where m is the reflectivity enhancement factor of the supermirrors compared to nickel mirrors, hence the focusing capabilities of optics is proportional to λ."

NEUTRON SCATTERING IN GRAZING INCIDENCE

- ➡ Specular reflection $\xi_z \approx 1 \div 100\ nm$
- ➡ Off-specular scattering $\xi_x \approx 1 \div 100\ \mu m$
- ➡ Grazing incidence small angular (diffraction) neutron scattering $\xi_y \approx 1 \div 100\ nm$



$$Q_z = (2\pi/\lambda)(sin\alpha_f + \sin\alpha_i)$$
$$Q_x = (2\pi/\lambda)(\cos\alpha_f \cdot \cos 2\theta_f - \cos\alpha_i)$$
$$Q_y = (2\pi/\lambda)(\cos\alpha_f \cdot \sin 2\theta_f)$$

⬇

$$Q_z = (2\pi/\lambda)(\alpha_f + \alpha_i)$$
$$Q_x = (2\pi/\lambda)(\alpha_i^2 - \alpha_f^2 - \theta_f^2)/2 \qquad \sim\alpha_f^2$$
$$Q_y = (2\pi/\lambda) \cdot \theta_f \qquad \sim\theta_f^1$$

Denis Korolkov et al. // J. Appl. Cryst. (2012). 45, 245–254

$\lambda \uparrow$ ➡ For the same position at the detector $Q_{x,y} \downarrow$ ➡ $d \sim 2\pi \cdot Q^{-1}$ ➡ $d \uparrow$

Scattering cross section on clusters $\sim\lambda^2$, which is better for studying matter at large scales (100-1000 Å)

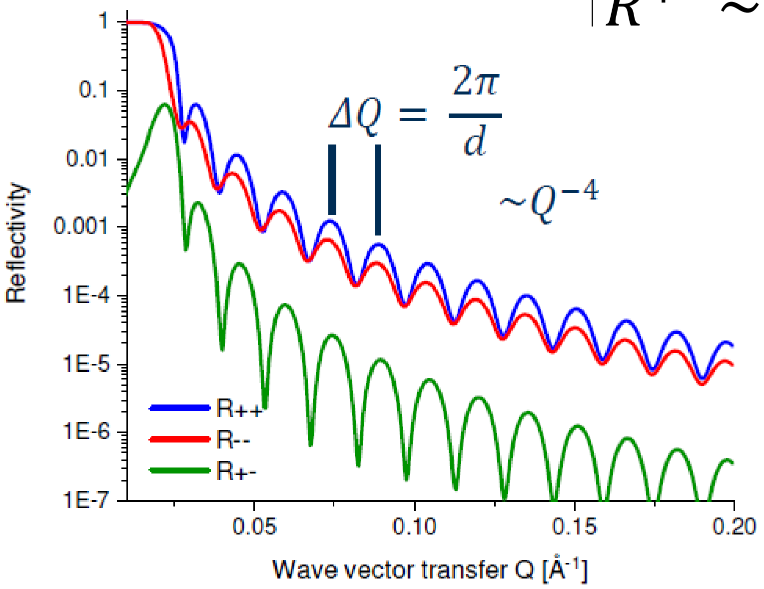
POLARIZED NEUTRON REFLECTOMETRY

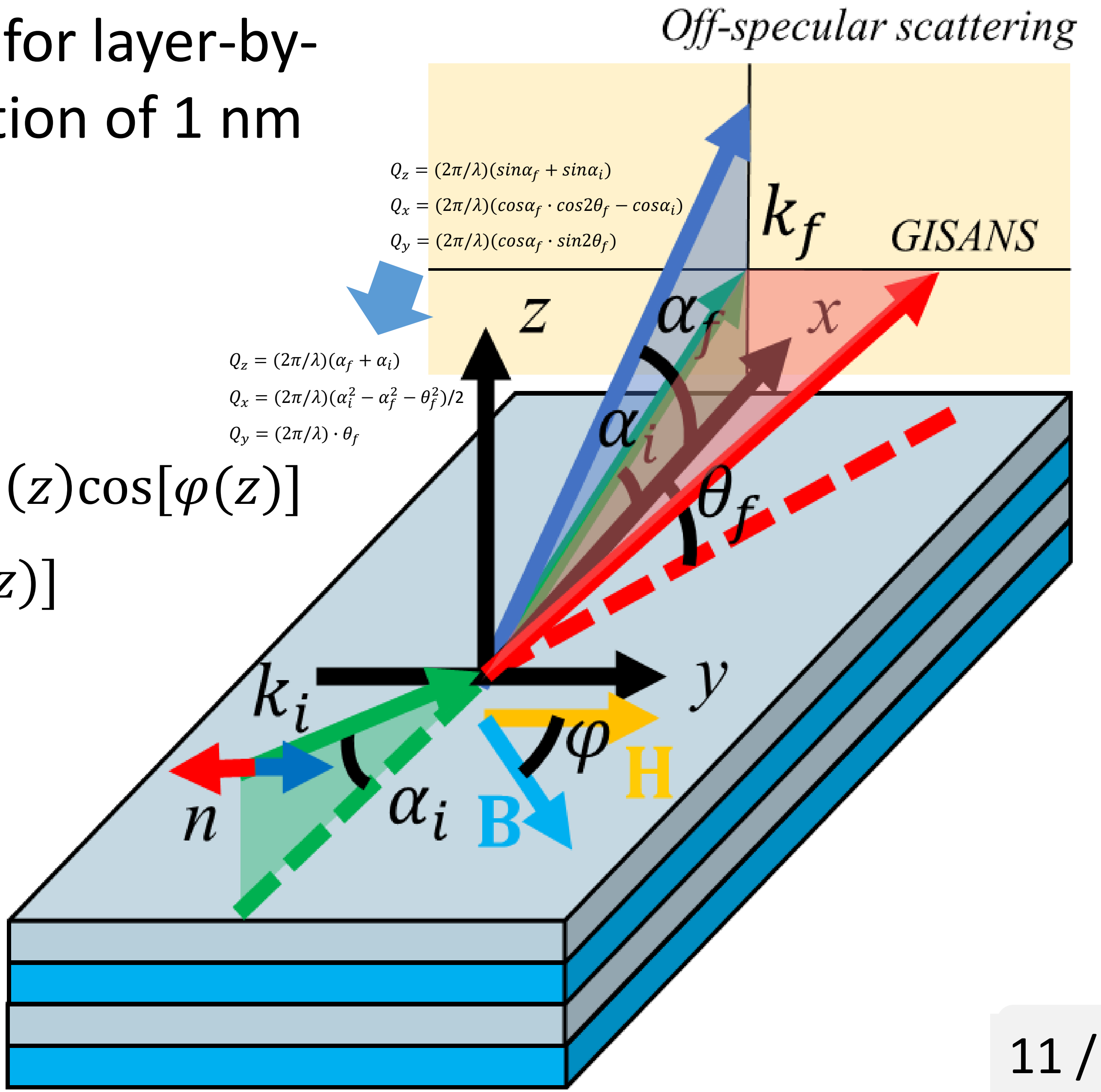
The use of polarized neutrons allows for layer-by-layer magnetic analysis with a resolution of 1 nm

$$U = U_{nuc} + U_{mag}$$

$$U_{mag} = -\boldsymbol{\mu B} = |\mu_n|\boldsymbol{\sigma B}$$

$$r = \begin{pmatrix} r_{++} & r_{+-} \\ r_{-+} & r_{--} \end{pmatrix} \Rightarrow \left| \begin{array}{l} R^{\pm\pm} \sim SLD(z) \pm B(z)\cos[\varphi(z)] \\ R^{+-} \sim B(z)\sin[\varphi(z)] \end{array} \right.$$





$Q_z = (2\pi/\lambda)(sin\alpha_f + sin\alpha_i)$

$Q_x = (2\pi/\lambda)(cos\alpha_f \cdot cos2\theta_f - cos\alpha_i)$

$Q_y = (2\pi/\lambda)(cos\alpha_f \cdot sin2\theta_f)$

$Q_z = (2\pi/\lambda)(\alpha_f + \alpha_i)$

$Q_x = (2\pi/\lambda)(\alpha_i^2 - \alpha_f^2 - \theta_f^2)/2$

$Q_y = (2\pi/\lambda) \cdot \theta_f$

Due to larger neutron phase shift (change in phase difference between two spin states) in matter and magnetic field ($\sim\lambda$) VCN have better sensitive to small contrast

Additiobal advantages exist also for grazing incidence neutron spin echo and the neutron depolarization method

Yu. V. Nikitenko. Grazing Incidence Spin-Echo Neutron Spectrometer. //
Journal of Surface Investigation. Xray, Synchrotron and Neutron Techniques, 2016, Vol. 10, No. 1, pp. 169–176.

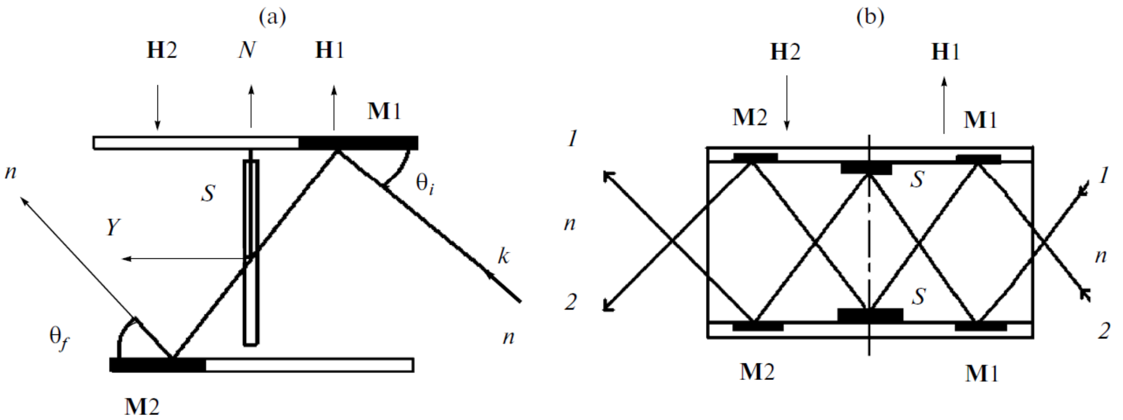


Fig. 9. Schemes of spin-echo spectrometers for measuring the (a) bulk and (b) surface properties of sample *S*. Magnetic fields **H**1 and **H**2; mirrors with magnetization **M**1 and **M**2. A neutron beam "*n*" with wave vector **k** directed at grazing angle θ to the mirrors.

$$\delta\Delta\varphi = -\rho \cdot H \cdot d \cdot \delta(1/k_{\perp})$$

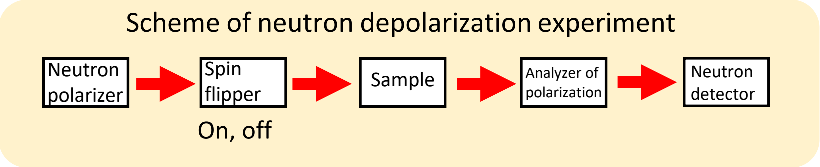
$$\rho = \frac{2 \cdot m \cdot \mu}{\hbar^2}$$

$$\Rightarrow \quad \lambda\uparrow \quad \Rightarrow \quad \delta\Delta\varphi$$

V.K. Aksenov et al. // Physica B 213&214 (1995) 100-106

K. Krezhov et al. // J. Phys.: Condens. Matter 5 (1993) 9277-9286.

D. Patroi et al. // Optoelectronics and Advanced Materials – Rapid Communications,
Vol. 9, No. 9-10, September – October 2015, p. 1328 - 1331



Scheme of neutron depolarization experiment

Neutron polarizer → Spin flipper → Sample → Analyzer of polarization → Neutron detector

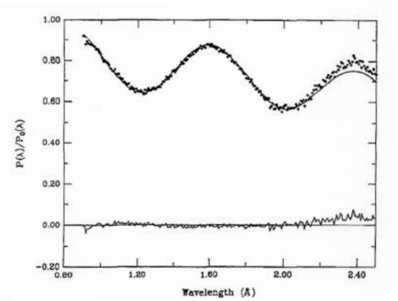
On, off

$$P = \frac{I_{off} - I_{on}}{I_{off} + I_{on}}$$

P – polarization of the beam after passing through the sample

P_0 – polarization without sample

Coefficient $D = \frac{P - P_0}{P + P_0}$ proportional to magnetization and has oscillation dependence



$$\delta\Delta\varphi = C \cdot B_i \cdot d \cdot \lambda$$

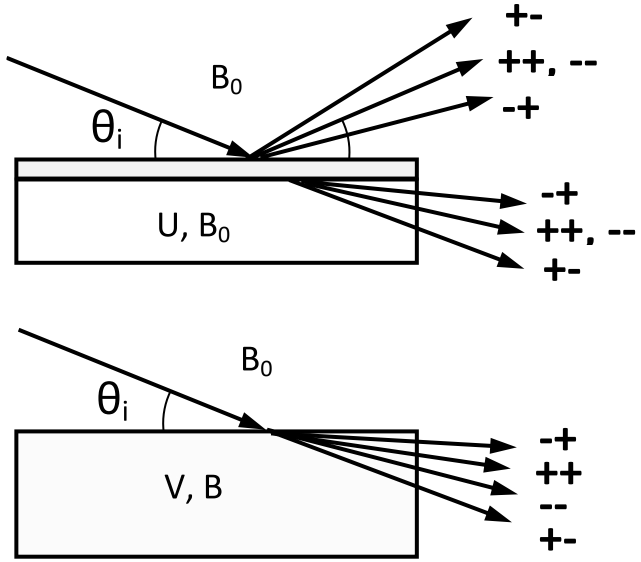
$$C = 4.66 \cdot 10^{-5}$$

SPATIAL BEAM SPLITTING & NEUTRON CHANNELING IN NEUTRON REFLECTOMETRY

S. V. Kozhevnikov, V. D. Zhaketov, A. V. Petrenko et al.
Application of a Cryogenic Moderator in the REMUR Neutron Reflectometer. //
Journal of Surface Investigation. X-ray, Synchrotron and Neutron Techniques, 2016, Vol. 10, No. 1, pp. 1–9.

“Two examples of applying a combined neutron moderator including a cryogenic moderator for experiments using a reflectometer were presented: the formation of a neutron microbeam with a layered waveguide structure and spatial beam splitting upon reflection from a magnetically noncollinear film.”

Kozhevnikov et al., JAC **45**, 814 (2012)
Aksenov et al., Physica B **297**, (2001)
Felcher et al., Nature **377**, 409(1995)



Spin-flip probability

$$R^{+-} \sim \sin^2 \alpha$$

Energy conservation

$$\frac{\hbar^2 p_i^2}{2m} + \mu B_0 = \frac{\hbar^2 (p_f^{+-})^2}{2m} - \mu B_0$$

$$(p_f^{+-})^2 = p_i^2 + 2\mu B_0 \frac{2m}{\hbar^2}$$

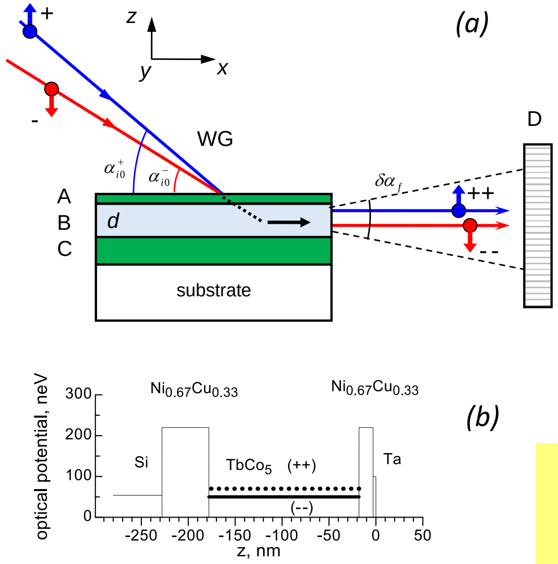
$$(p_f^{-+})^2 = p_i^2 - 2\mu B_0 \frac{2m}{\hbar^2}$$

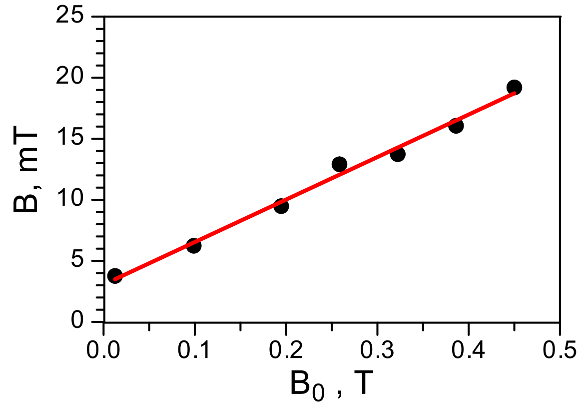
$$(\theta_f^{+-})^2 = \theta_i^2 + 2\mu B_0 \lambda^2 \frac{2m}{h^2}$$

$$(\theta_f^{-+})^2 = \theta_i^2 - 2\mu B_0 \lambda^2 \frac{2m}{h^2}$$

$$\lambda \uparrow \Rightarrow \Delta\theta$$

Kozhevnikov et al., JETP Lett. **103**, 36 (2016)
NREX, Garching



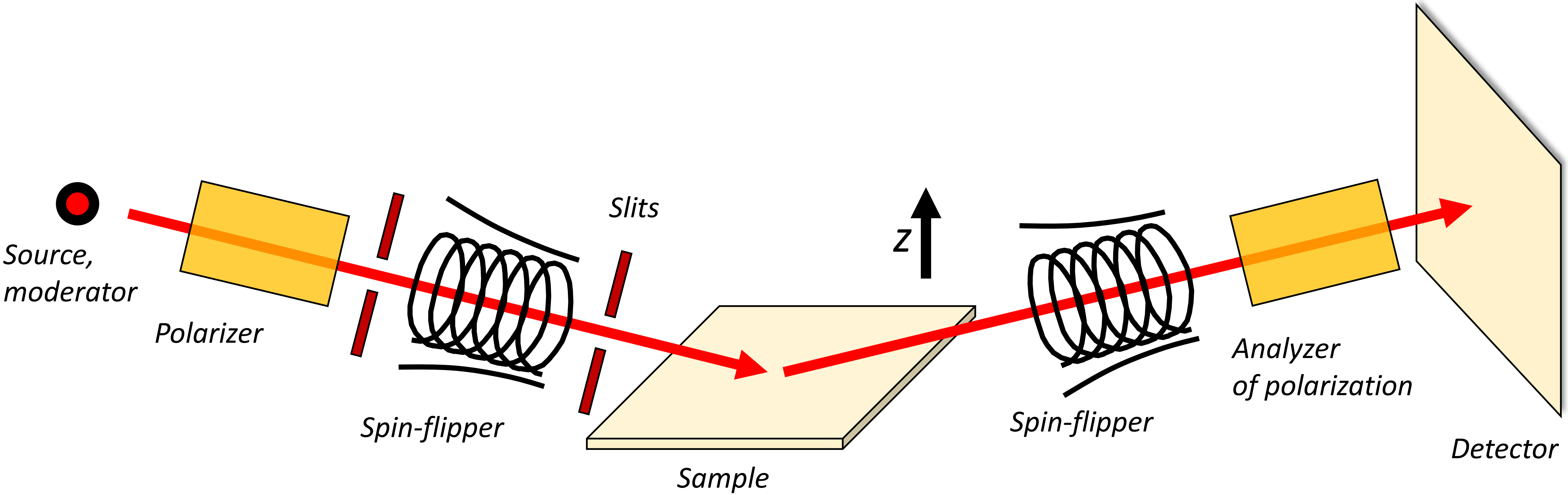


$$B = \frac{\hbar^2}{4\mu\, m}\left(\frac{2\pi}{\lambda}\right)^2 \left[\sin^2(\alpha_{i0}^{+}) - \sin^2(\alpha_{i0}^{-})\right]$$

$$\lambda \uparrow \Rightarrow \Delta\alpha \uparrow$$

@ materials provided by Sergey Kozhevnikov

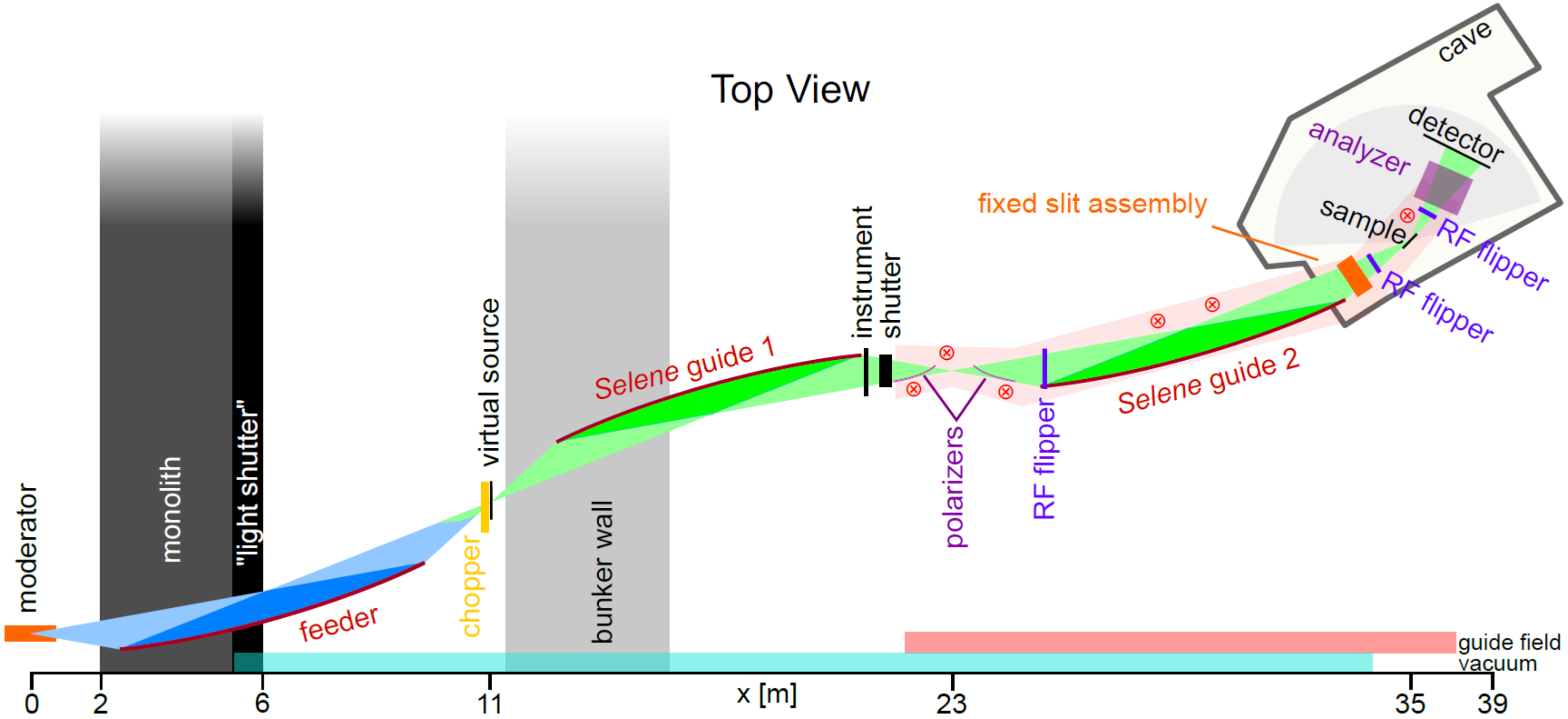
SCHEME OF A REFLECTOMETRY EXPERIMENT WITH FULL POLARIZATION ANALYSIS



- For VCN the same technic can be used for full polarization analysis: supermirrors & radio-frequency flippers
- The total reflection angle $\sim\lambda$, this gives better reflection from mirror, from supermirrors with large m up to angles 10-20 degrees
- Some detailes for SM discussed in work of A.K. Petukhov et al. // arXiv:1606.01960.
- The requirement for supermirror quality increases on the wavelength scale of very cold neutrons
- Some detailes for RF flippers discussed at this work: A.N. Bazhenov el al. // NIM Research A332 (1993) 534-536
- The condition of adiabatic spin rotation is easier to maintain for VCN

$$S_{LP} = \frac{2\cdot\pi\cdot\hbar}{m\cdot\lambda\uparrow}\cdot\frac{1}{\gamma\cdot B} \ll S'$$

Formation of a neutron beam on the ESTIA (ESS) reflectometer



Estia instrument overview

Distance moderator-sample	35 m
Beam-line	Elliptical feeder guide at 2m Selene guide m=4
Chopper	14 Hz at 10.7 m
Slit after chopper	Height up to 20 mm Width 60 µm ÷ 5 mm
Distance sample-detector	4 m
Detector	2D PSD 500 x 250 mm^2 Resolution 0.5 x 2 mm Multi-Blade Boron system

Wavelength	Band 6.9 Å between 4 and 25 Å Resolution 0.3 Å
Q-range	0.005 Å^{-1} – 3.0 Å^{-1}
Polarization	Supermirror m=4
Flippers	RF
Analyzer	Fan
Minima sample	1 x 1 mm^2
Sample environment	TEFI & FLUCO
Reflectivity	10^{-7}

- Optics efficiency is proportional to the wavelength (*m* x 0.1° / Å)
- Technical implementation is much easier

@ Frédéric Ott

TYPE OF LAYERED SYSTEMS

- Enhanced wave field mode
- Periodic superlattice
- Fibonacci systems

NEUTRON RESONATORS

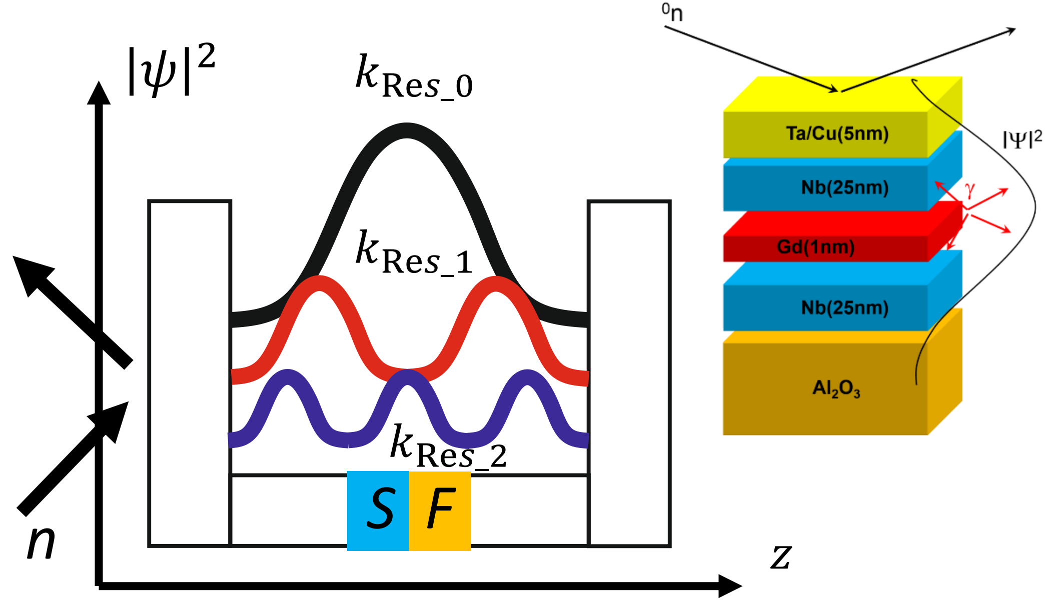
$$|\psi(z,k)|^2 = |\psi_d(z,k) + \psi_b(z,k)|^2$$

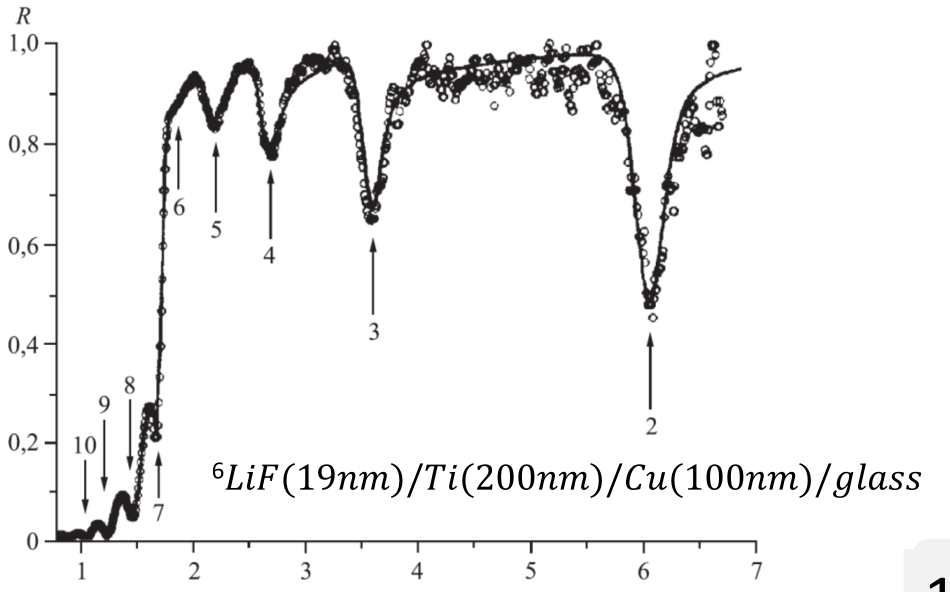
$$\eta_{\max}(nucl.\ density) \approx 10^5$$

$$W = 10^{-5} \div 10^{-3}\ V$$

Secondary radiation calculation:

$$J_{i,j}(k_{z0}) \sim \int \frac{|\psi(z,k_{z0})|^2 \cdot Im(u)}{|\psi_0(k_{z0})|^2 \cdot k_0} N_i(z)dz$$





V.L. Aksenov et al. // Physica B 276-278 (2000) 946-947.

REGISTRATION OF SECONDARY RADIATION

$$W = \sum W_{ij} \propto \sum N_i \sigma_{ij}$$

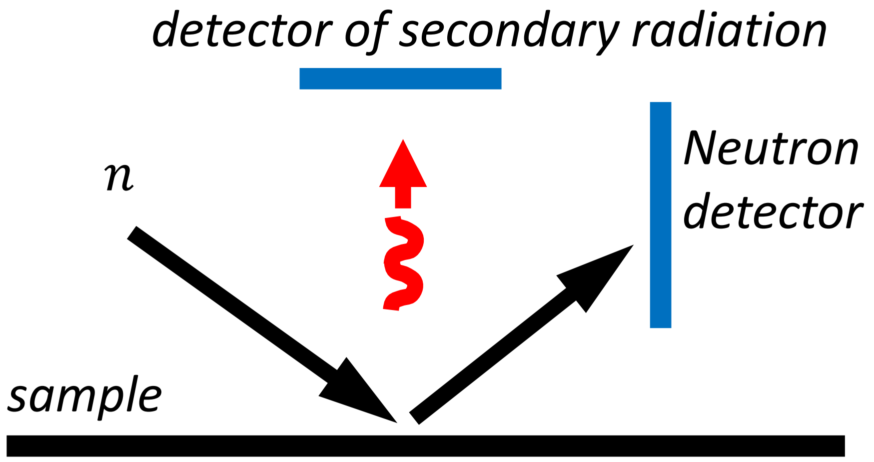
i – isotope, j – type of secondary radiation

Characteristic secondary radiation

- *Charged particles (n, α); (n, t); (n, p)*
- *Gamma-quanta (n, γ)*
- *Fission fragments (n, f)*

Some secondary processes

- *Spin-flip neutrons*
- *Noncoherent scattered neutrons by nuclei*
- *Inelastically scattered neutrons*
- *Diffusely scattered neutrons on medium inhomogeneities*



$$I_{i,j}(k_{z0}) \sim \int \frac{|\psi(z,k_{z0})|^2 \cdot Im(u)}{|\psi_0(k_{z0})|^2 \cdot k_0} N_i(z)dz$$

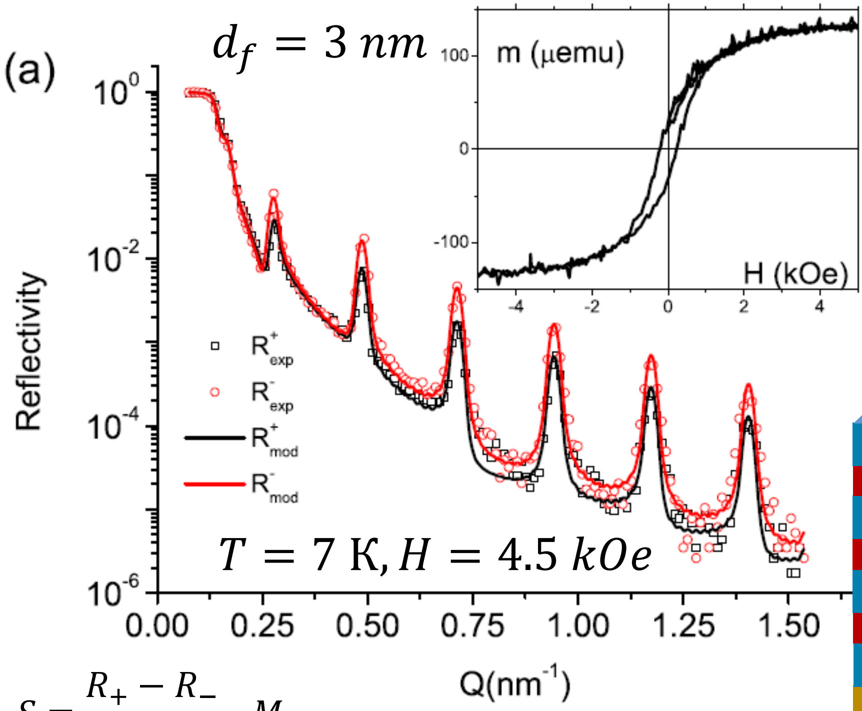
$$Im(b) = \frac{\sigma(\lambda)}{2 \cdot \lambda} \Rightarrow \lambda \uparrow \Rightarrow \sigma(\lambda) \uparrow \Rightarrow Im(b) = const$$

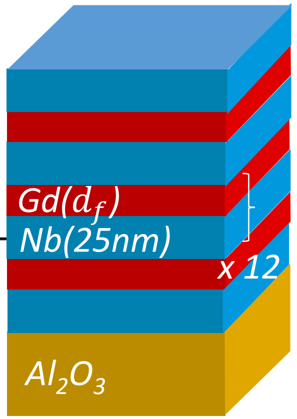
Registration of secondary radiation in neutron reflectometry makes it possible to determine the isotopic profiles of elements. This approach with VCN seemed to be efficient for scattering processes by slow motions with characteristic energy of VCN range, clusters with big size and other.

Zhaketov V.D. et al. // Journal of Surface Investigations, vol. 15, no. 3, pp. 549-562 (2021)
Zhaketov V.D. et al. // Journal of Surface Investigations, vol. 13, no. 3, pp. 478-487 (2019)

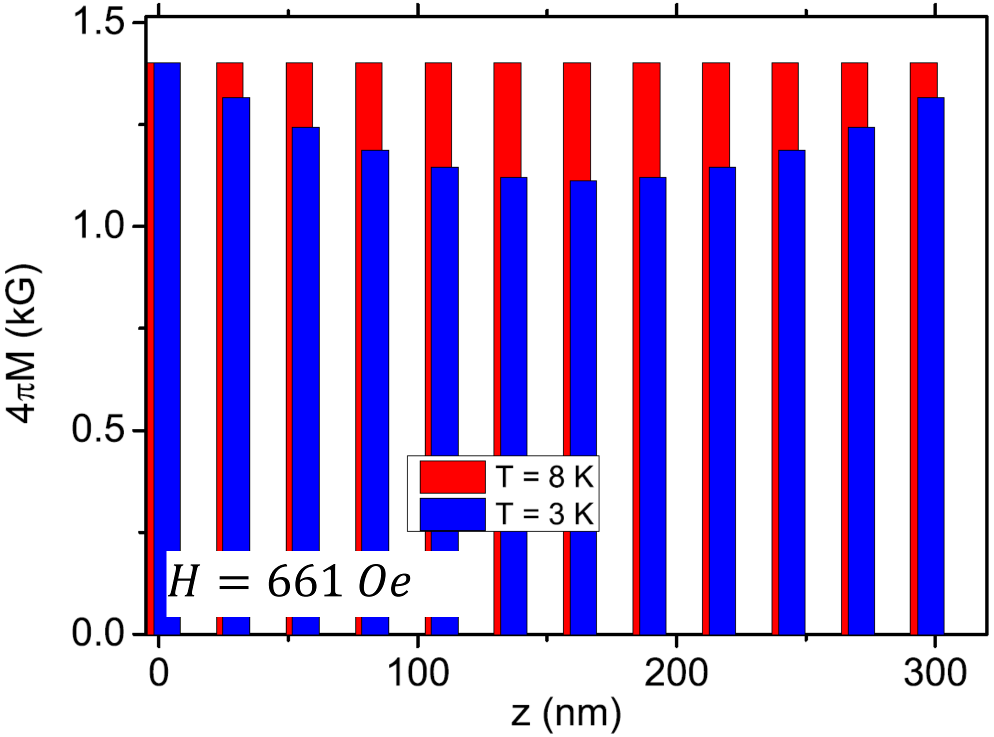
PERIODIC SYSTEMS

- Superlattice periodic systems – artifitial crystals with $d \sim \xi$
- Reducing the magnetic moment of structures *[Nb(25 nm)/Gd(x=1.2, 3, 5 nm)]x12* below T_c
- Transition of a ferromagnetic structure to superconducting state (artifitial magnetic superconductors)





$$S = \frac{R_+ - R_-}{R_+ + R_-} \sim M$$



Results of experimental data fitting

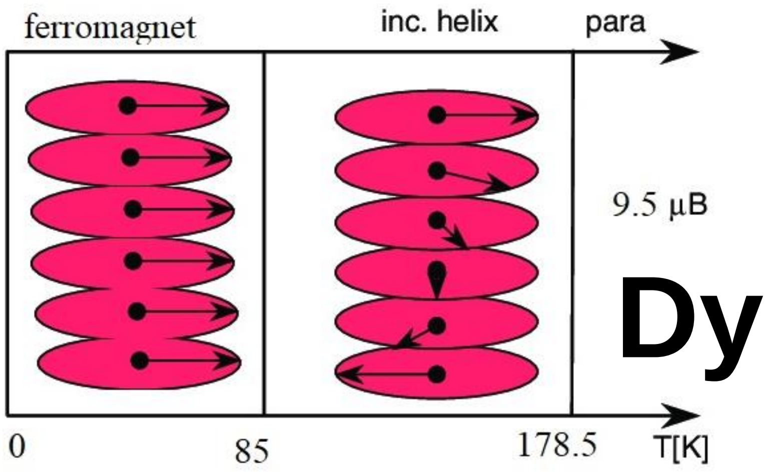
$$n \cdot \lambda = 2 \cdot d \cdot \sin\theta$$

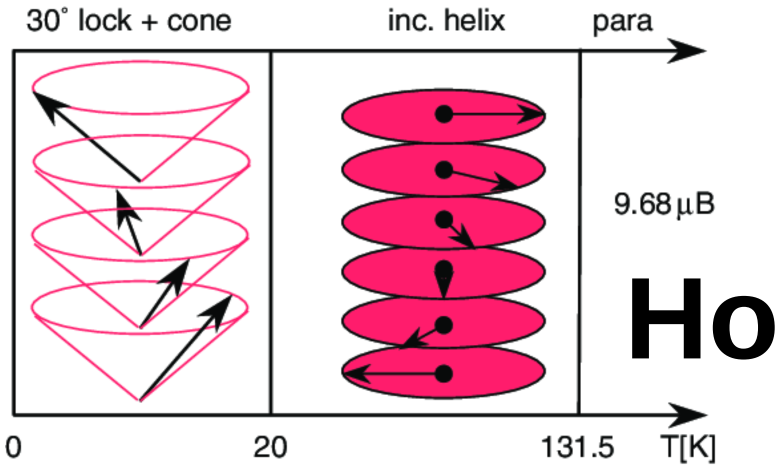
$$d \sim \xi$$

$\lambda \uparrow$ $\theta \uparrow$

Khaydukov Yu.N. et al. // Phys. Rev. B, 99, 140503 (2019)

PERIODIC SYSTEMS WITH NON-COLLINEAR MAGNETS



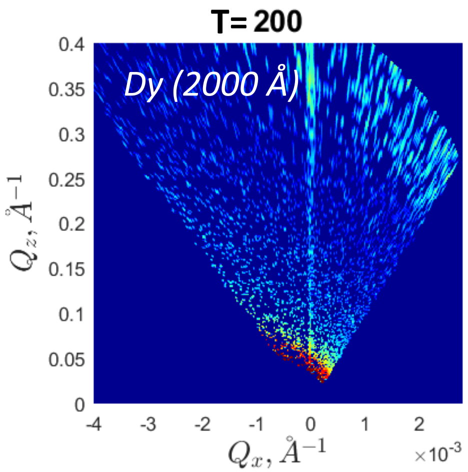


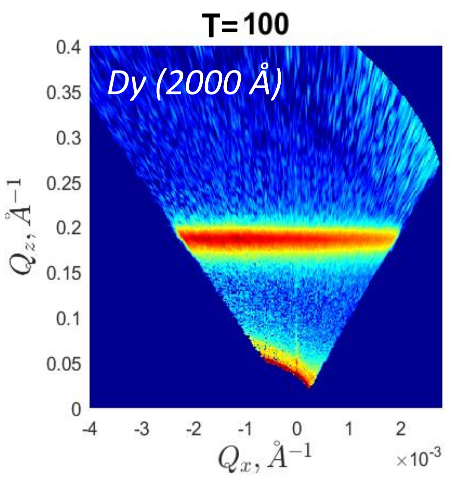
Investigated structures:

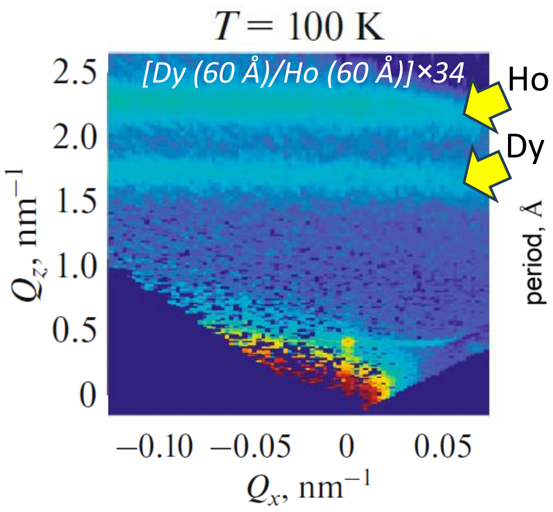
1) Dy (2000 Å)

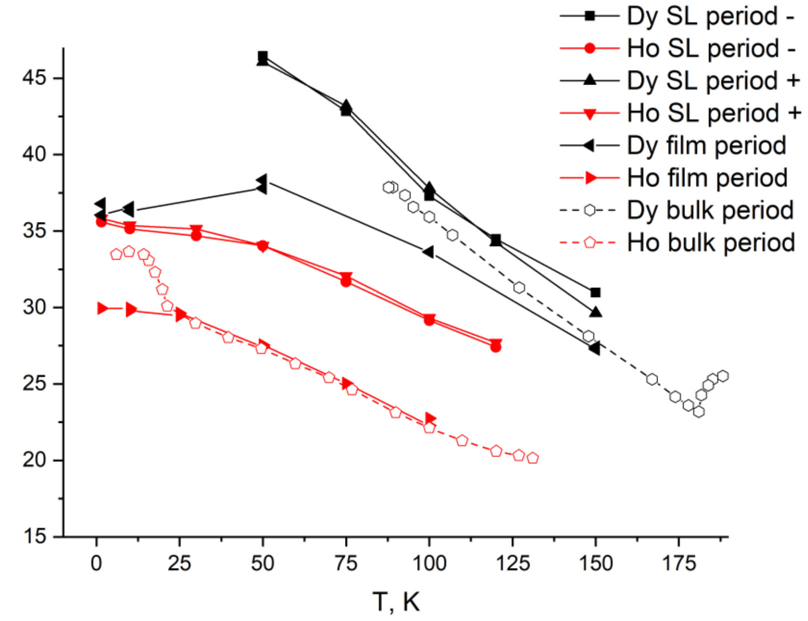
2) Ho (2000 Å)

3) [Dy (60 Å)/Ho (60 Å)]×34







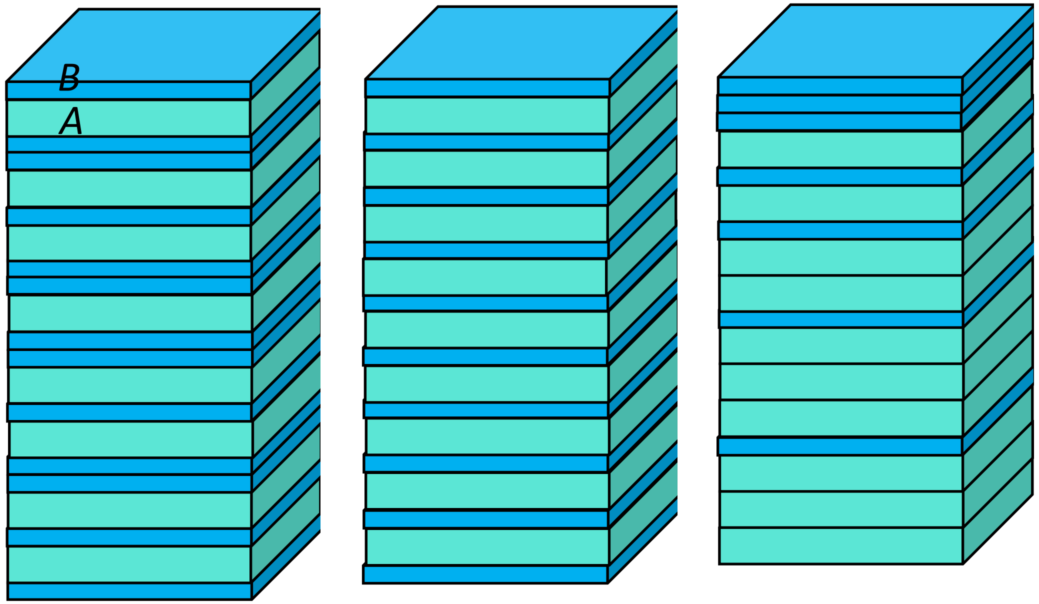


$$d_{he}\sim 30\ \text{Å} \rightarrow \theta\sim 20\ mrad \rightarrow \lambda=\frac{2\cdot d\cdot \sin\theta}{n}\sim 1.2\ \text{Å} \rightarrow \lambda\uparrow \rightarrow \theta\uparrow$$

Devyaterikov D.I. et al. // Journal of Surface Investigations, Vol. 16, No. 5, pp. 839-842 (2022)

Devyaterikov D.I. et al. // Physics of Metals and Metallography, Vol. 122, No. 5, pp. 465-471 (2021)

LAYERED ONE-DIMENTIONAL QUASICRYSTALS (FIBONACCI)



Quasiperiodic structure:
BABBABABBABBABABBABAB

Periodic structure:
BABABABABABA…

Disordered structure:
BBBABABAABAAA

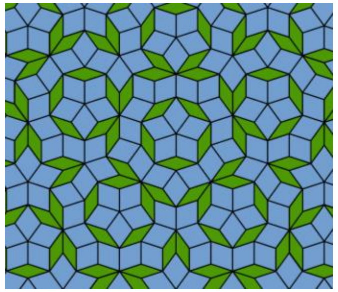
The figure shows: the quasiperiodic structure obtained for the Fibonacci series member S_7 ; periodic structure; and an disordered structure obtained using a random number generator.

$A \to B \quad B \to BA$

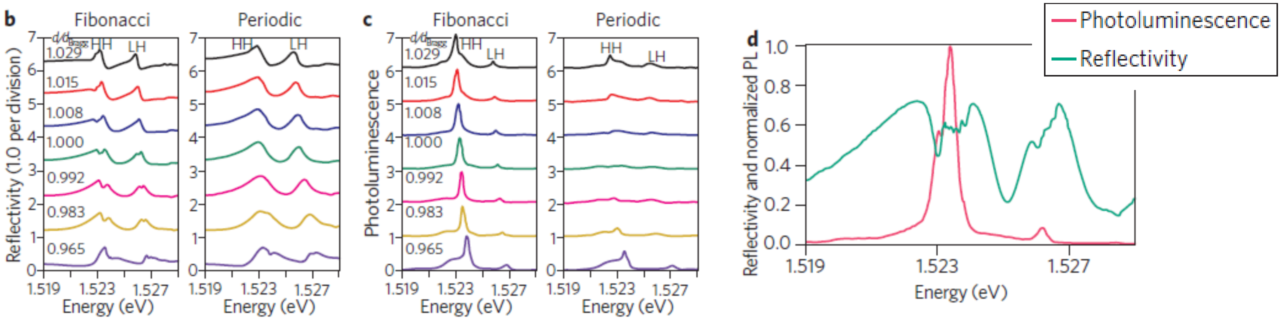
$$F(\boldsymbol{k}) = \lim_{N\to\infty} N^{-1} \sum_{\boldsymbol{z}_n} \exp(i\boldsymbol{k}\boldsymbol{z}_n)$$

$S_0 = A,$
$S_1 = B,$
$S_2 = BA,$
$S_3 = BAB,$
$S_4 = BABBA,$
$S_5 = BABBABAB,$
$S_6 = BABBABABBABBA,$
$S_7 = BABBABABBABBABABBABAB$

N – number of layers,
$\boldsymbol{z}_n$ – spatial position of layers,
$\boldsymbol{k}$ – wave-vector.



This talk about system like one-dimentional Penrose mosaic



Reflectivity and Photoluminescence for Fibonacci and Periodic structure
Z. Valy Vardeny et al. // Nature Photonics, Vol. 7, pp. 177-187 (2013)

$\lambda \uparrow$

$\theta \uparrow$

Artifitial 1D superconducting and magnetic quasicrystalls require $d{\sim}\xi$

But if take into account only rule $\frac{\lambda_B}{\lambda_B} = 1.618$ its calculations are needed to understand the advantages of such systems for VCN.

WHAT PHENOMENON ?

- Very cold neutrons with $\lambda = 15 - 100\ \text{Å}$ have certain advantages for studying matter at large spatial (100-1000 Å) and time scales
- Slow motion with characteristic energy of the VCN range are typical for example: vortexes in superconductors or magnetic systems (magnons, skyrmions) etc.

From the other hand, series of works demonstrated:

- Solid state layered systems inhomogeneus and exist clusters (and even groups of clusters with fractal ordering) with scales $\xi = 100 - 1000\ \text{Å}$
- Slow-motion relaxation at big times ($\sim 10^5\ s$) of superconducting vortexes and magnetic inhomogenities
- In focus of view dynamical processes in oscillating magnetic field
- Review of this phenomenon see in the work of:

Yu. V. Nikitenko and V. D. Zhaketov // Physics of Particles and Nuclei, 2022, Vol. 53, No. 6, pp. 1089–1125.

Analysys with VCN would be very usefull here!

CONCLUSION

1. The possibilities of using very cold neutrons for polarized neutron reflectometry are considered. Conclusions are drawn about the possible geometric advantages of using very cold neutrons. Neutrons of this kind are also useful for methods using Larmor neutron precession.
2. Very cold neutrons with $\lambda = 15 - 100\ \text{Å}$ have certain advantages for studying matter at large spatial (100-1000 Å) and time scales. Slow motion with characteristic energy in the VCN range are typical for example: vortexes in superconductors or magnetic systems (magnons, skyrmions) etc.

Thank you for your attention!