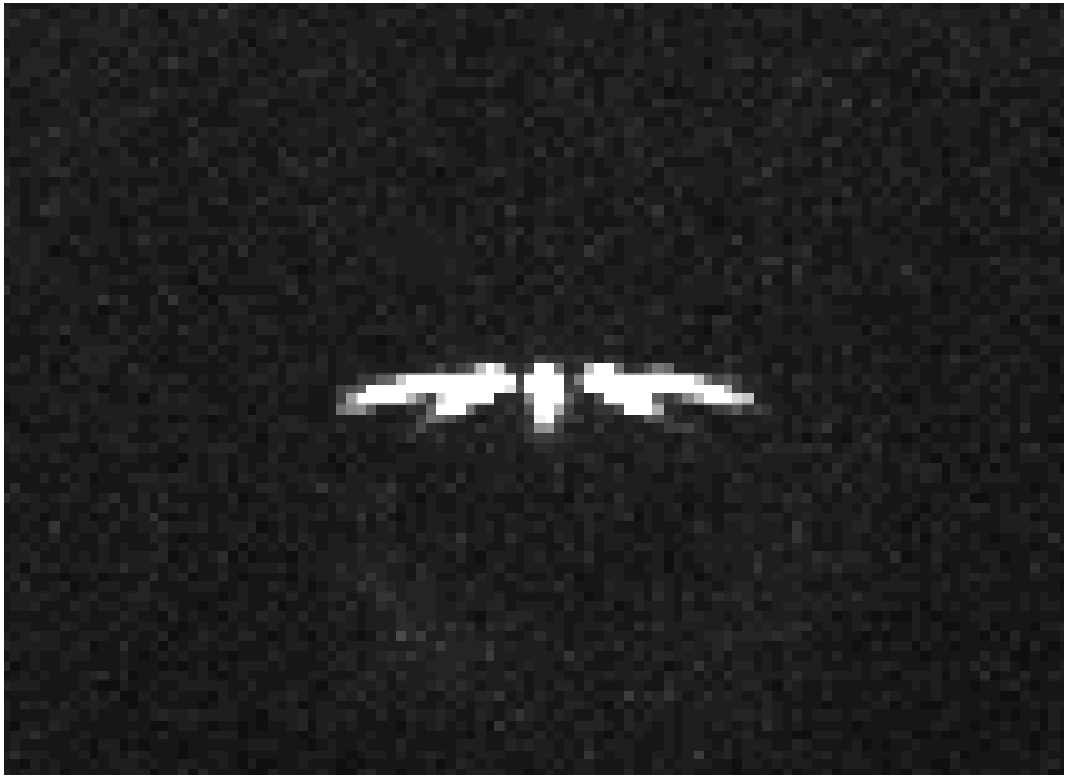
VCN interferometry: Some history, ongoing and (near) future projects

Jürgen Klepp, Faculty of Physics, University of Vienna, Austria



Outline

- Some VCN-related properties, some advantages and disadvantages
- (Shortened) history part: The COW experiments, VCN contribution
- Our way of making VCN diffraction gratings
- Some ideas for future no-two-path VCN interferometry methods
- A project for a VCN interferometer at PF2 of the ILL, possible designs
- Summary

Neutrons, VCN application related properties

- All wave phenomena known from light optics are available for VCN too (refraction, diffraction, total reflection,...)
- **Crystals do not work for long wavelengths so that the Bragg condition**

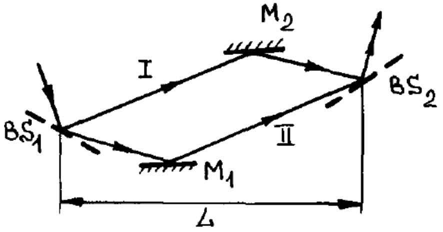
  $2\Lambda \sin\theta_B = \lambda$ cannot be fulfilled if $\lambda/(2\Lambda) > 1$

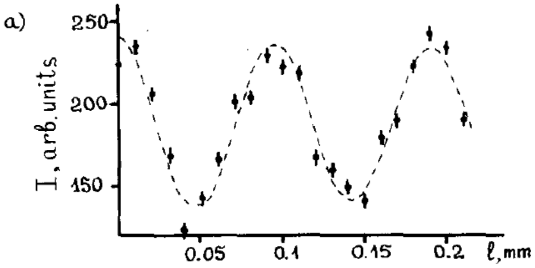
  Therefore: custom-made structures as optical elements (gratings, for instance; intercalated crystals?), such as in:

DIFFRACTION-GRATING NEUTRON INTERFEROMETERS

A.I. IOFFE

Physica B 151 (1988) 50–56
North-Holland, Amsterdam





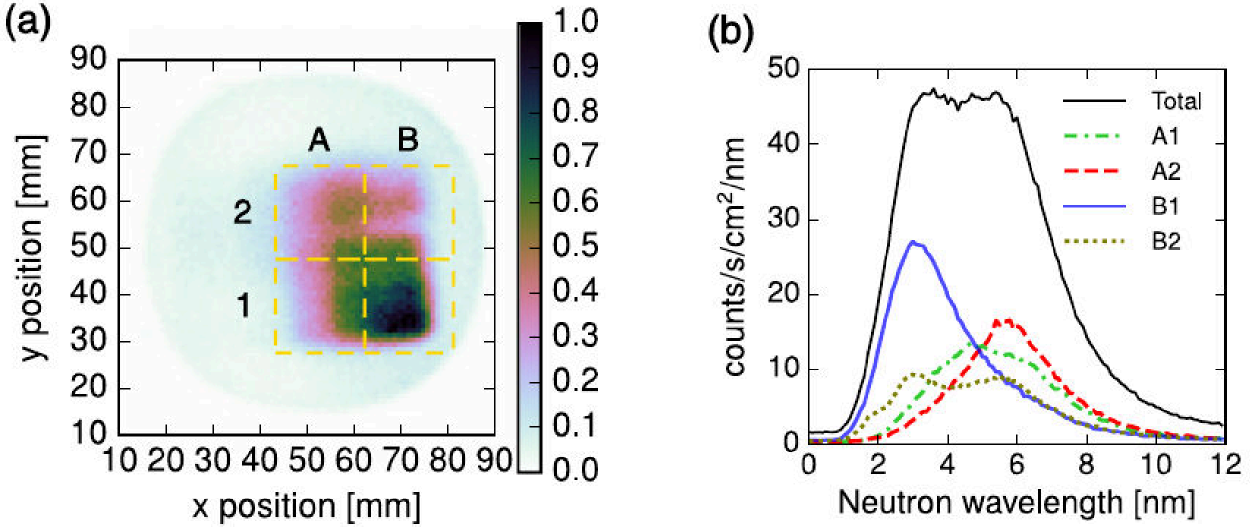
Very cold neutrons (VCN) -- Advantages

- VCN are slow and exhibit long interaction times with whatever we mount in their path (example: gravity: v ~ 100m/s --> falling down 4 mm at 3 m horizontal path).
- VCN can still be considered to come in beams, so that beam optics methods can be used.

VCN -- Drawbacks

- VCN "typically" come in broad wavelength distributions and large-divergent beams.
- VCN are slow enough to even get lost by neutron-air scattering and absorption, can be prevented by He or vaccum tubes, makes instruments somewhat inflexible.

→ VCN are nice for doing physics, but they are a scarce good!

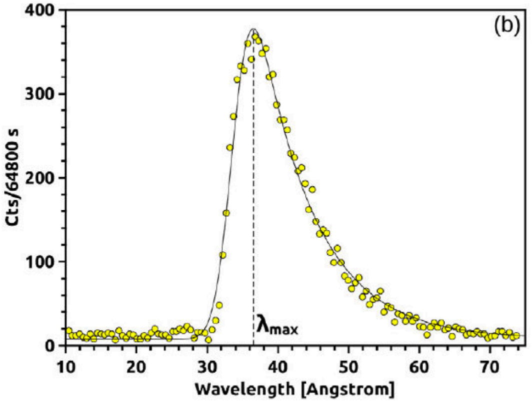


PF2 at the ILL (before refurbishment)

Typical divergence angle:
(size aperture 1 + size aperture 2)/

distance between them ~ 2 mrad

for grating diffraction experiments --> 1 n/s



T. Oda et al., Nucl. Instrum. Methods A 860, 35 (2017) at the Institute Laue Langevin, PF2/VCN
M. Blaickner et al., Nucl. Instrum. Methods A 916, 154 (2019)

Some neutron interferometry experiments (examples)

- Thermal neutrons (λ ~ 2 Å): Perfect crystal neutron interferometry and polarimetry [1,2]
- Cold neutrons (λ ~ 5 Å): Grating and multilayer interferometers and polarimetry [1,2,3,4]
- Very cold neutrons (VCN, λ ~ 4 nm): Grating interferometers [1,2,5]
- Ultra cold neutrons (λ ~ 65 nm): Gravity resonance spectroscopy [6]

[1] H. Rauch, S.A. Werner, Neutron Interferometry, Oxford University Press, UK (2000,2015)

[2] J. Klepp, S. Sponar, Y. Hasegawa, Prog. Theor. Exp. Phys. 082A01 (2014)

[3] C. Pruner et al., Nucl. Instrum. Methods Phys. Res. A 560, 598 (2006)

[4] Ebisawa et al., Phys. Rev. A 57, 4720 (1998)

[5] G. van der Zouw et al., Nucl. Instrum. Methods Phys. Res. A 440, 568 (2000)

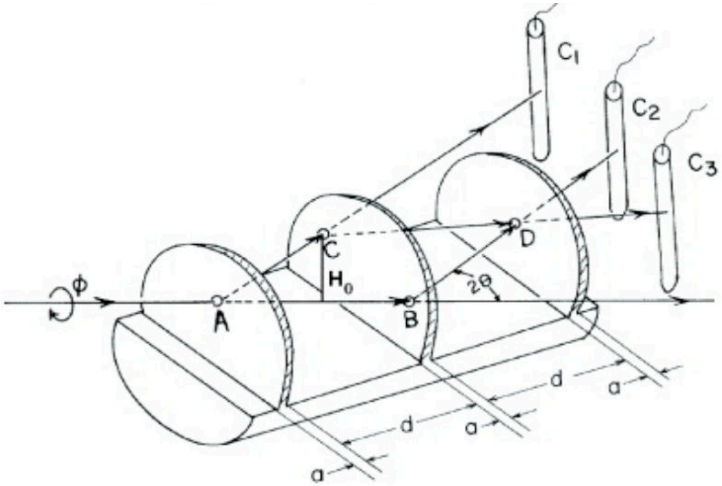
[6] T. Jenke et al., Phys. Rev. Lett. 112, 151105 (2014)

History part

Some past VCN interferometry experiments COW experiment (general)

$$E_0 = \frac{\hbar^2 k^2}{2m_I} + m_g\, g\, z$$

g gravitational acceleration
m_g gravitational mass
m_I inertial mass



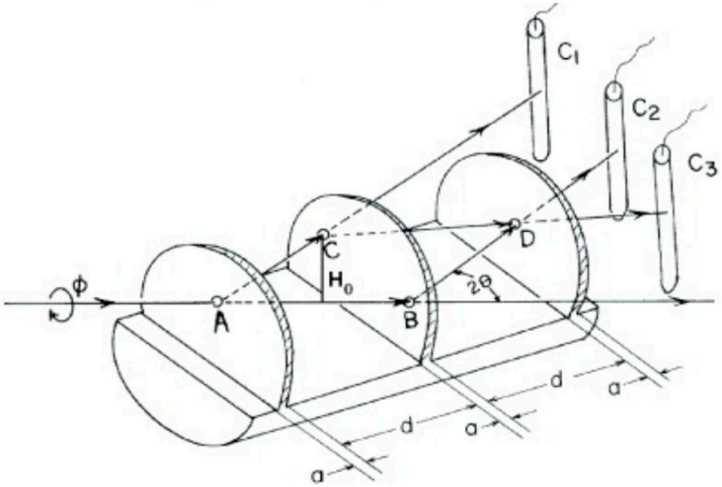
induced phase:

$$\triangle\Phi_{grav} = \Phi_{ACD} - \Phi_{ABD}$$

$$\triangle\Phi_{\mathrm{grav}} = -2\pi\lambda\frac{m_I m_g}{\hbar^2} g A_0$$

R. Collela, A.W. Overhauser, S.A. Werner, PRL 34, 1472 (1975),...
G. van der Zouw, M. Weber, J. Felber, R. Gähler, P.Geltenbort, A. Zeilinger,
Nucl. Instrum. Methods Phys. Res. A 440, 568 (2000)
H. Rauch, S.A. Werner, Neutron Interferometry, Oxford University Press, UK (2015)

Some past VCN interferometry experiments
COW experiment (general)



$$\triangle\Phi_{\mathrm{grav}} = -2\pi\lambda\frac{m_I m_g}{\hbar^2} g A_0$$

g gravitational acceleration
m_g gravitational mass
m_I inertial mass

COW experiments test the strong equivalence principle in the quantum regime: Viola and Onofrio have shown that the average fall time of a quantum object depends (apart from the height and g) on the ratio of gravitational and inertial mass. Fall time variance depends on gravitational mass only (apart from 1/width of the wave packet and 1/g). However, variance of the average travel time for a system accelerated by a=-g (by other means than gravity) depends on inertial mass only. For the strong equivalence principle to hold, inertial and gravitational masses must be the same. Otherwise, one could distinguish gravity from other acceleration by measuring fall time variance.

L. Viola, R. Onofrio, PRD 55, 455 (1997)
H. Rauch, S.A. Werner, Neutron Interferometry, Oxford University Press, UK (2015)

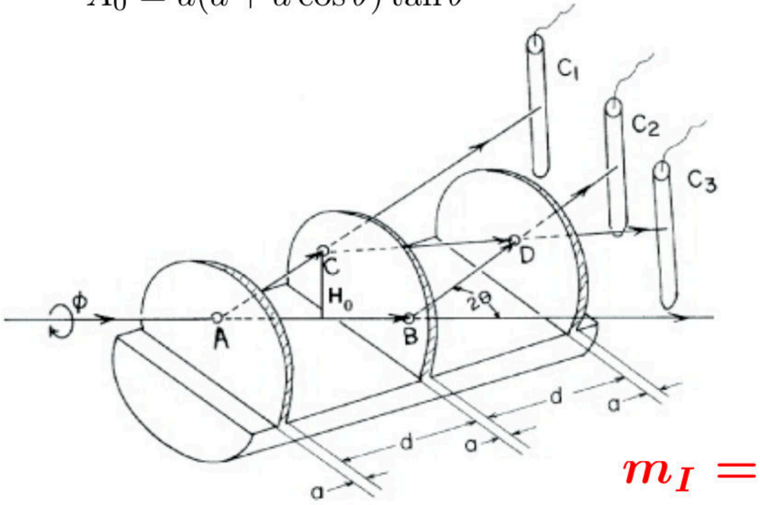
Some past VCN interferometry experiments
COW experiment (perfect-crystal IFM)

$$\triangle\Phi_{COW} = -2\pi\lambda\frac{m_I m_g}{\hbar^2} g A_0 \sin\phi$$
$$\triangle\Phi_{COW} = -q_{grav}\sin\phi$$

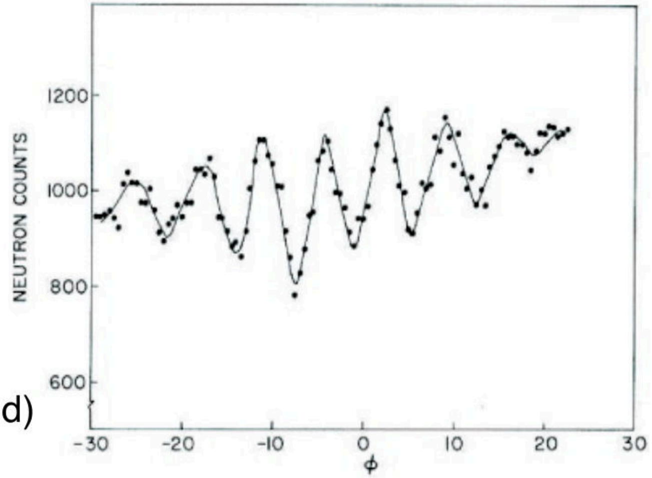
additional terms:

$$\triangle\Phi_{COW} = (-q_{grav} - q_{bend})\sin\phi + q_{Sagnac}\cos\phi$$

$$A_0 = d(d + a\cos\theta)\tan\theta$$



$q_{bend} \rightarrow$ X-ray measurements
$q_{Sagnac} = 2\pi m_I \omega_{Earth} A_0/\hbar\cos\theta_L$
θ_L colatitude angle



$$m_I = m_g$$

deviation ~ 4% (1% repeated)

R. Collela, A.W. Overhauser, S.A. Werner, PRL 34, 1472 (1975),...
H. Rauch, S.A. Werner, Neutron Interferometry, Oxford University Press, UK (2015)

Why COW with VCN?

Problems with perfect-crystal IFM for COW:

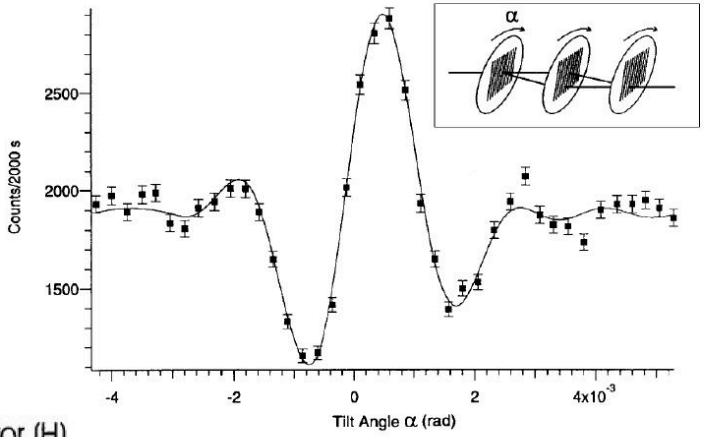
- Phase instabilities when scanning tilt angles (same for gratings). Improvement by phase shifter interferograms
- Since the crystals are heavy, they bend (not for grating IFM). Has been tried to correct by parallel in situ X-ray measurements. Abandoned later due to differences between neutrons and X-rays.
- It was found that corrections to results were needed (~5%) due to dynamical diffraction theory (DDT) effects. More paths than just two must be taken into account. Not needed for gratings that are not described by DDT.
- Some discrepancy remained ---> development of a VCN IFM at PF2 (ILL) for a COW experiment
- Area of VCN is ~same as for perfect-crystals, but wavelength is longer-->sensitivity to gravity phase is larger --> one doesn't have to tilt to large angles to measure large phase shifts.

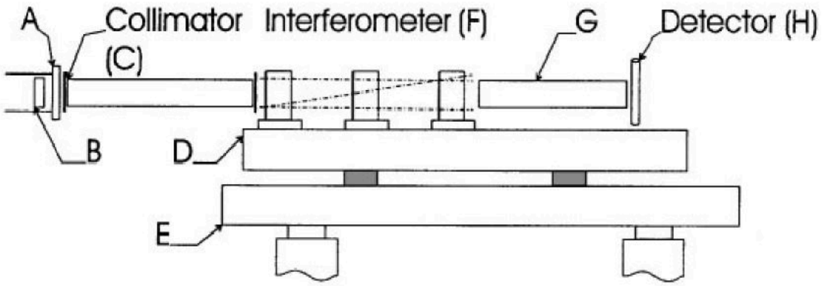
H. Rauch, S.A. Werner, Neutron Interferometry, Oxford University Press, UK (2015)

Some past VCN interferometry experiments
COW experiment (VCN)

$$\triangle\Phi_{grav} = -2\pi\lambda\frac{m_I m_g}{\hbar^2}gA(\lambda)\sin\phi$$

- mean wavelength : 100 Å
- $A(\lambda)$: separation $\geq 0.5\,\mathrm{mm}$, $\mathrm{L} = 1\,\mathrm{m}$
- low countrate : $0.6\,\mathrm{s}^{-1}$





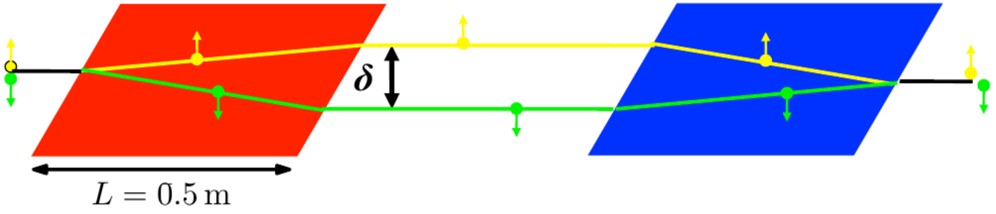
deviation: 1%

$$m_I = m_g$$

G. van der Zouw, M. Weber, J. Felber, R. Gähler, P.Geltenbort, A. Zeilinger,
Nucl. Instrum. Methods Phys. Res. A 440, 568 (2000)
H. Rauch, S.A. Werner, Neutron Interferometry, Oxford University Press, UK (2015)

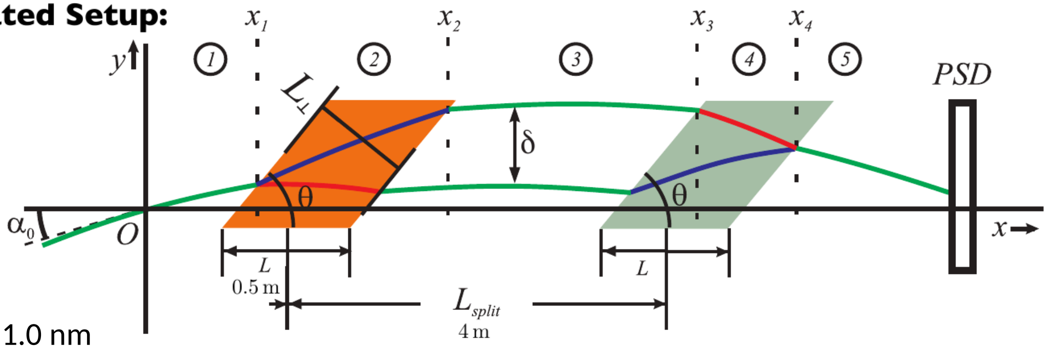
Some past VCN interferometry experiments
COW experiment (instrument OffSpec at ISIS, UK)

Top view:



$$\delta = \frac{\gamma_L B L \lambda^2 \cot\theta_0}{2\pi h} \qquad \delta = 10\,\mathrm{nm} \ldots 20\,\mu\mathrm{m}$$

Rotated Setup:



Wavelength 0.2 - 1.0 nm
Result: gravitational phase shift measured to 0.1 % accuracy
Some problems with Sagnac effect corrections and unexpected correction that should also apply to perfect-crystal IFM COW, see discussion of:

R. Gähler, R. Golub, K. Habicht, T. Keller, J. Felber, Physics B 229, 1 (1996)
V.-O- de Haan, J. Plomp, A.A. vanWell, M. T. Rekveldt, Y. Hasegawa, R.M. Dalgliesh, N.-J. Steinke, PRA 89, 063611 (2014)

Was VCN IFM for COW a success?

In principle yes (gravitational phase shift in agreement with theory-->mi=mg), but:

- It turned out that the neutron spectrum could not be understood (modelled) satisfacorily and also not measured (TOF!) with sufficient statistics. Exp. uncertainty is still large.

- ALSO: Meanwhile atom IFM had evolved. Much higher accuracy (6 orders of magnitude) than neutron IFM.

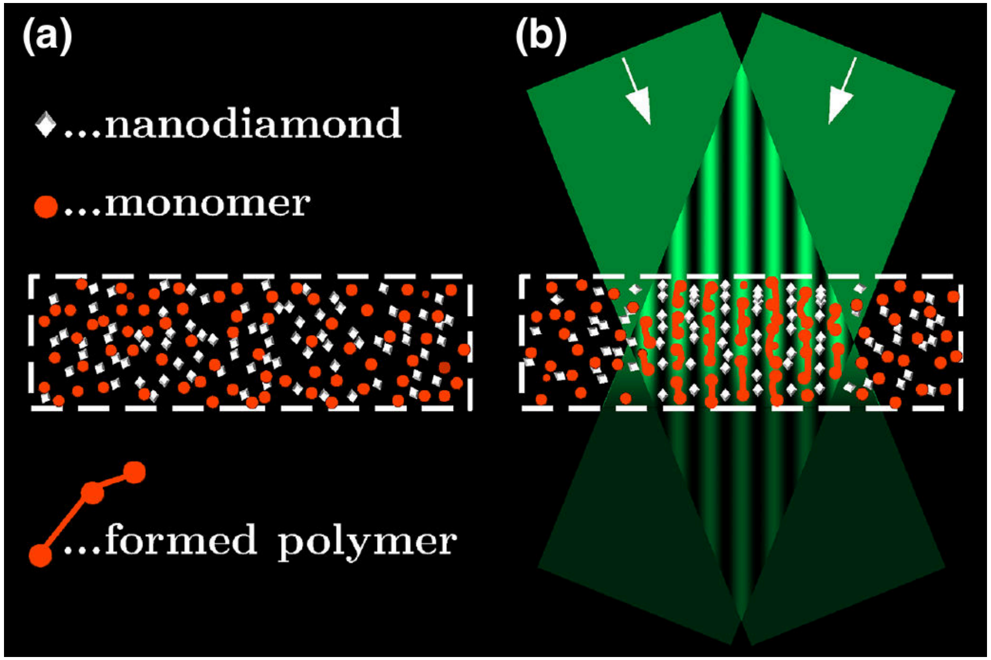
H. Rauch, S.A. Werner, Neutron Interferometry, Oxford University Press, UK (2015)

End of history part

Our way of making VCN diffraction gratings

J. Klepp et al., Materials 5, 2788 (2012)

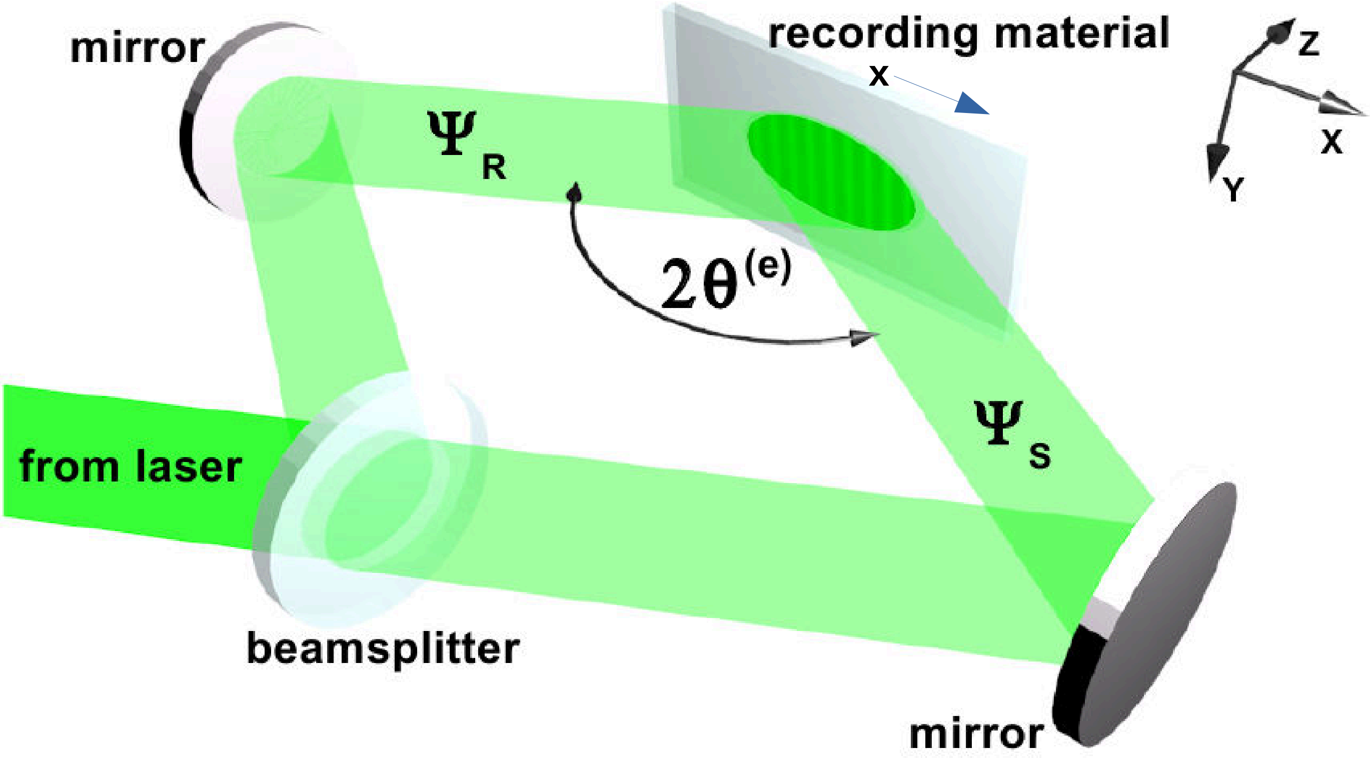
Optical holography with photosensitive materials to make VCN diffraction gratings



$$\Lambda = \frac{2\pi}{K} = \frac{\lambda}{2\sin\theta^{(e)}} \qquad \Delta n(x) = \sum_j \Delta n_j \cos(jKx+\varphi_j) \approx \Delta n_0 + \Delta n_1 \cos(Kx+\varphi_1)$$

Y. Tomita et al., Phys. Rev Applied 14, 044056 (2020)

Optical holography with photosensitive materials



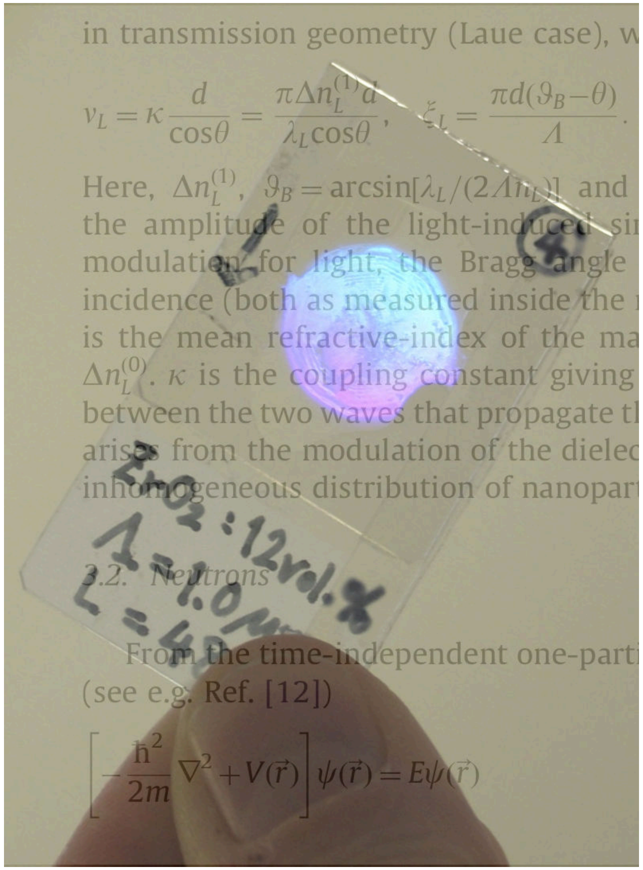
$$\Lambda = \frac{2\pi}{K} = \frac{\lambda}{2\sin\theta^{(e)}}$$

$$\Delta n(x) = \sum_j \Delta n_j \cos(jKx + \varphi_j) \approx \Delta n_0 + \Delta n_1 \cos(Kx + \varphi_1)$$

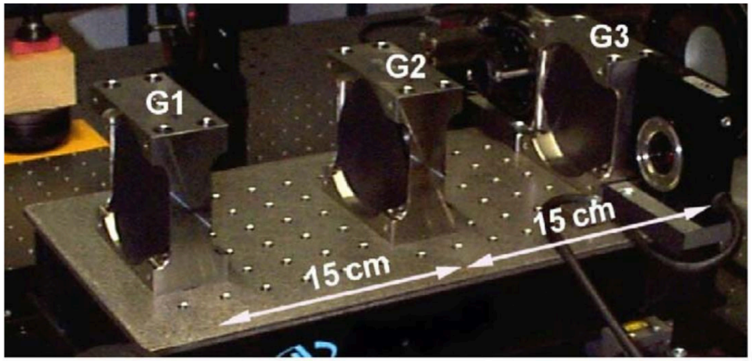
First application for neutron optics: R. Rupp et al., PRL 64, 301 (1990)

Optical holography with photosensitive materials

**Versatile:
can be made
for specific
requirement,
in little time**



Some past VCN (CN?) interferometry experiment
A deuterated PMMA grating IFM (D11, ILL, France)



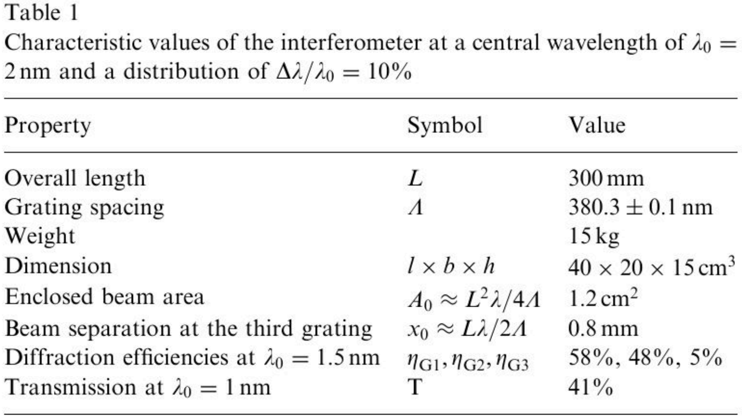
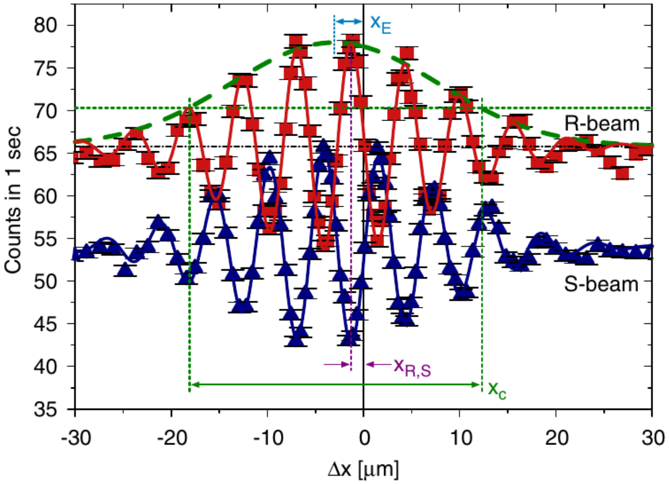


Table 1
Characteristic values of the interferometer at a central wavelength of $\lambda_0 =$ 2 nm and a distribution of $\Delta\lambda/\lambda_0 = 10\%$

Property	Symbol	Value
Overall length	L	300 mm
Grating spacing	Λ	380.3 ± 0.1 nm
Weight		15 kg
Dimension	$l \times b \times h$	$40 \times 20 \times 15\,\mathrm{cm}^3$
Enclosed beam area	$A_0 \approx L^2\lambda/4\Lambda$	$1.2\,\mathrm{cm}^2$
Beam separation at the third grating	$x_0 \approx L\lambda/2\Lambda$	0.8 mm
Diffraction efficiencies at $\lambda_0 = 1.5$ nm	$\eta_{G1}, \eta_{G2}, \eta_{G3}$	58%, 48%, 5%
Transmission at $\lambda_0 = 1$ nm	T	41%



U. Schellhorn, R.A. Rupp, S. Breer, R.P. May, Physica B 234, 1068 (1997)
C. Pruner, M. Fally, R.A. Rupp, R.P. May, J. Vollbrandt, NIMA 560, 598 (2006)
H. Rauch, S.A. Werner, Neutron Interferometry, Oxford University Press, UK (2015)

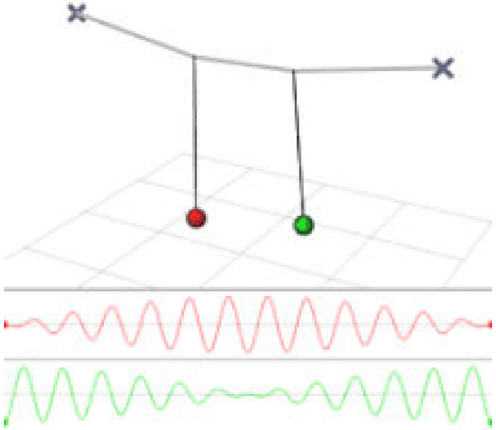
In addition to traditional interferometer experiments, we think it is worthwhile to develop/test/improve more VCN-economic interferometric methods, for low-coherence beams, using most of the wavelength distr., beams as divergent as possible (little collimation)

--> No TOF, no narrow peaks, no high background, avoid absolute measurements, avoid sensitivity on vibrations, temperature, humidity,…

A first idea: Pendellösung effect

One can observe the periodic energy transfer (beating) between two coupled mechanical pendulums, which is analogous to the energy transfer between diffracted beams originating from waves propagating and interfering within a periodic potential.

The energy is transferred back and forth between diffraction orders as a function of thickness of the potential region, potential strength, wavelength, deviation from the Bragg condition. Ewald [1] coined the term "Pendellösung" (pendulum solution).



https://de.wikipedia.org/wiki/Gekoppelte_Pendel

[1] P.P. Ewald, Rev. Mod. Physics 37, 46 (1965) and the References therein

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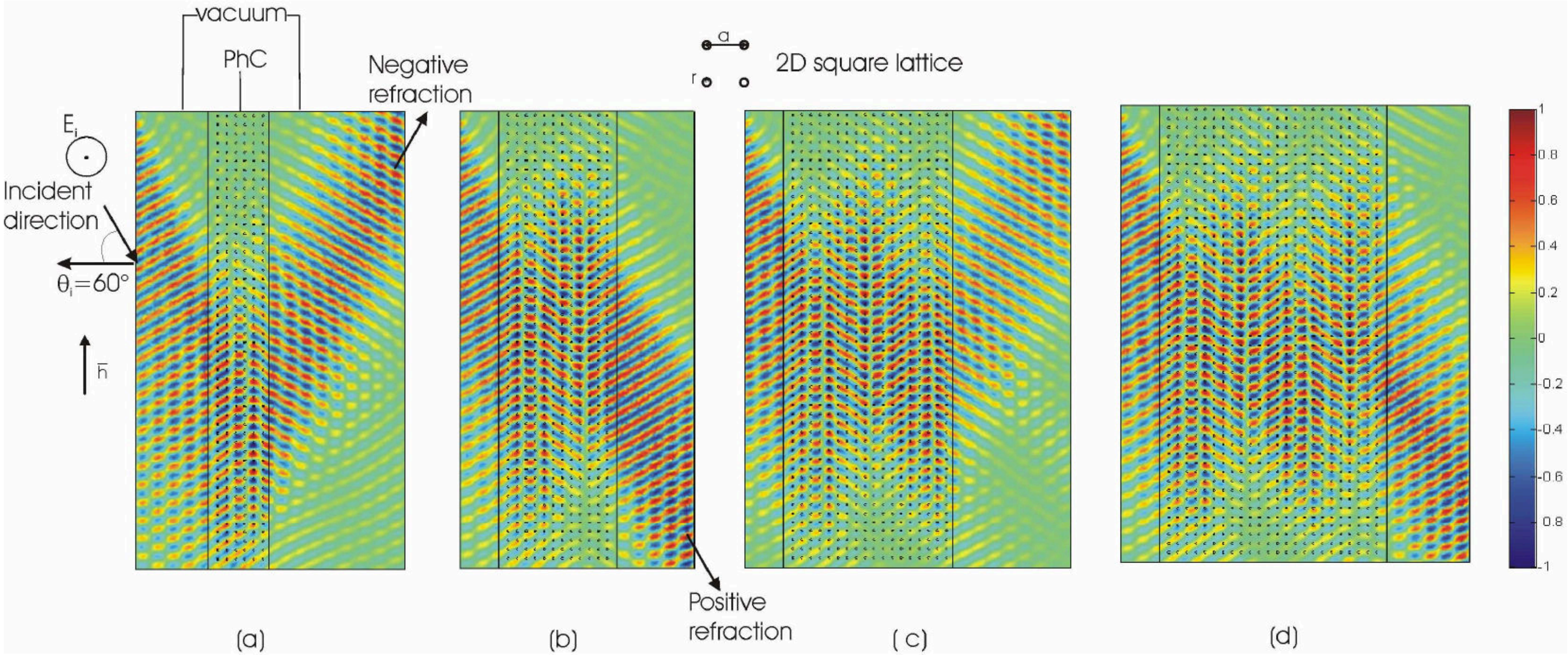
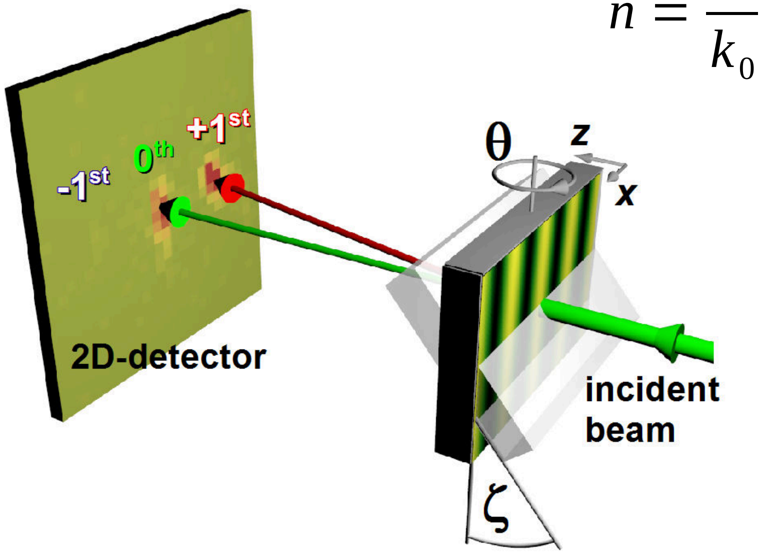
Pendellösung effect in photonic crystals - FEM simulation



V. Mocella, Optics Express 13, 1361 (2005)

Described by dynamical diffraction theory (DDT) [1] or similar theories (Kogelnik [2,3])



$$n = \frac{k}{k_0} = \sqrt{1 - \frac{V}{E_0}} \rightarrow n(x) \simeq 1 - [\rho + \Delta\rho(x)] \frac{\lambda^2 b_C}{2\pi}$$

$$\eta = \frac{I_D}{I_D + I_F}$$

$$\eta(\mathrm{x},\mathrm{y}) = \frac{\sin^2\left(\mathrm{y}\sqrt{\mathrm{x}^2+1}\right)}{\mathrm{x}^2+1}$$

$$\mathrm{x} = \frac{2\pi\cos\theta_B}{\lambda\Lambda b_C \Delta\rho}(\theta_B - \theta), \quad y = \frac{\lambda d b_C \Delta\rho}{2\cos\theta}$$

Assuming transmission- (or Laue-) geometry, vicinity of Bragg-angle, two interfering waves only

[1] V. F. Sears, Neutron Optics, Oxford University Press (1989), [2] H. Kogelnik, Bell System Tech. J. 48 2909 (1969), [3] J. Klepp et al., Nucl. Instrum. Methods A 634, S59 (2011)

How does the graph of this function typically look like?

$$\eta(x,y) = \frac{\sin^2\left(y\sqrt{x^2+1}\right)}{x^2+1}, \quad x = \frac{2\pi\cos\theta_B}{\lambda\Lambda b_C\Delta\rho}(\theta_B-\theta), \quad y = \frac{\lambda d b_C \Delta\rho}{2\cos\theta}$$

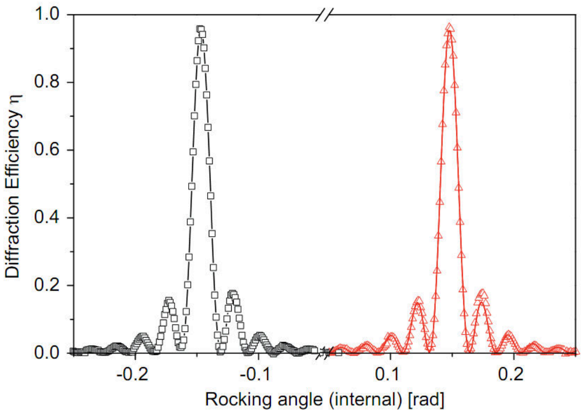


Fig. 2. ± 1st order rocking curves (left/right) of a nanoparticle holographic grating with 20 vol% SiO_2 nanoparticles, $d \simeq 50\,\mu m$ and $\Lambda = 1\,\mu m$ measured with laser light of wavelength $\lambda_L = 458$ nm.

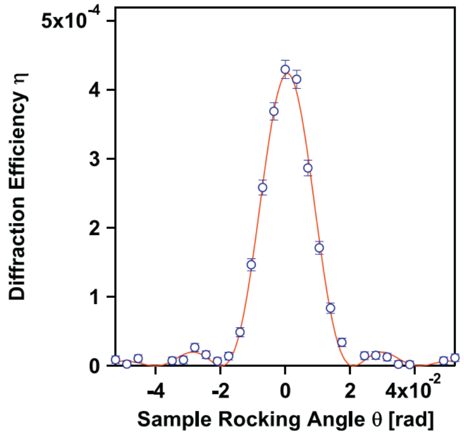
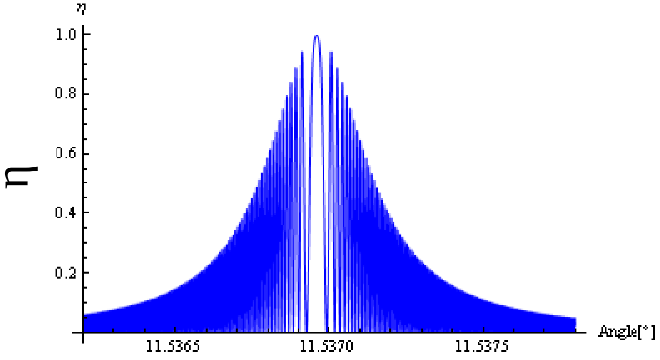
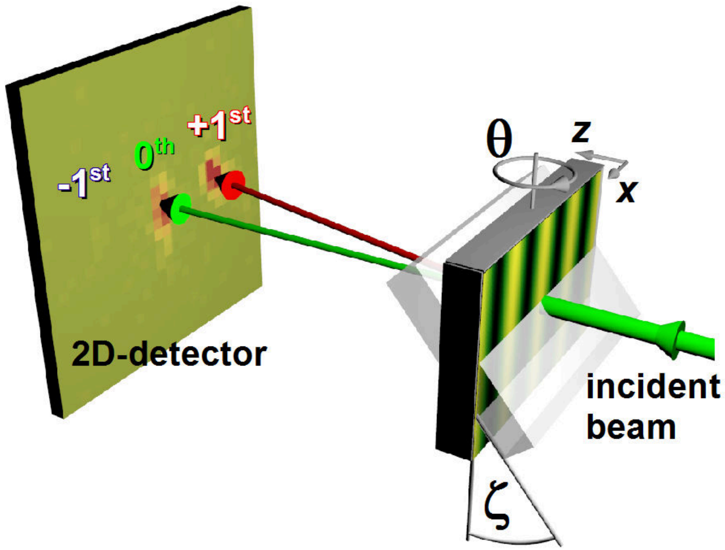


Fig. 4. Rocking curve of the same sample with analyzer crystal at − 1st order peak position (c.f. Fig. 3).



J. Klepp et al., Nucl. Instrum. Methods A 634, S59 (2011)

Why the tilt (angle ζ)?



$$\eta(x,y) = \frac{\sin^2\left(y\sqrt{x^2+1}\right)}{x^2+1}$$

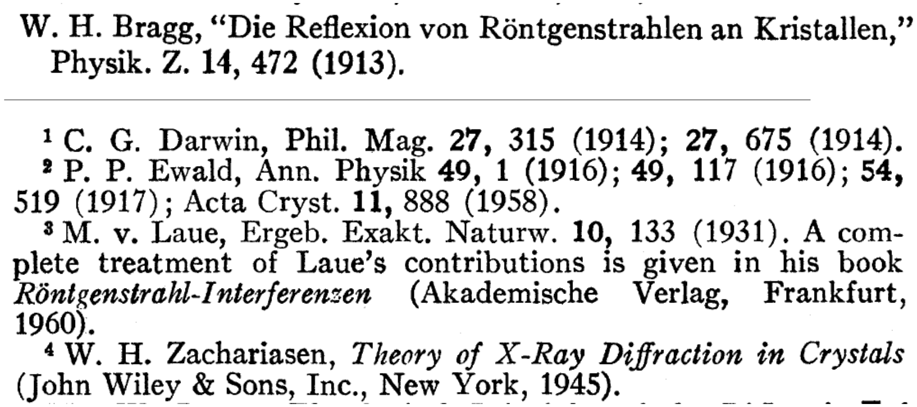
$$x = \frac{2\pi\cos\theta_B}{\lambda\Lambda b_C\Delta\rho}(\theta_B-\theta),\quad y = \frac{\lambda d b_C\Delta\rho}{2\cos\theta}$$

$$\textit{At Bragg angle}:\ \eta(0,y) = \sin^2 y$$

The diffraction efficiency at the Bragg angle (so that x = 0) will oscillate as a function of the wavelength, the thickness and the grating strength. Pendellösung interf.

$$\eta = \sin^2\left(\frac{\lambda d b_C \Delta\rho}{2\cos\zeta\cos\theta_B}\right)$$

Previous works, X-rays and γ -rays (examples):



W. H. Bragg, “Die Reflexion von Röntgenstrahlen an Kristallen,” Physik. Z. **14**, 472 (1913).

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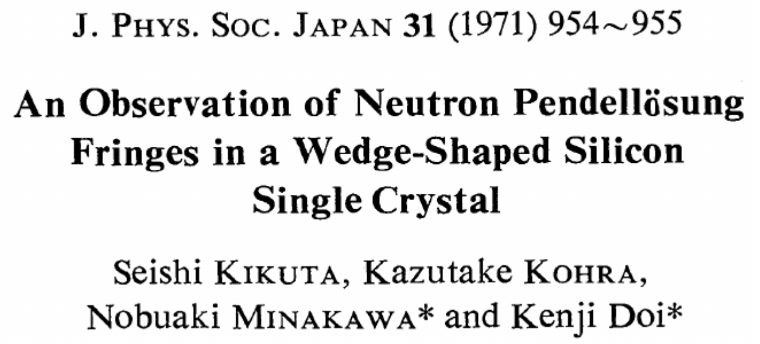
[1] C. G. Darwin, Phil. Mag. **27**, 315 (1914); **27**, 675 (1914).

[2] P. P. Ewald, Ann. Physik **49**, 1 (1916); **49**, 117 (1916); **54**, 519 (1917); Acta Cryst. **11**, 888 (1958).

[3] M. v. Laue, Ergeb. Exakt. Naturw. **10**, 133 (1931). A complete treatment of Laue’s contributions is given in his book *Röntgenstrahl-Interferenzen* (Akademische Verlag, Frankfurt, 1960).

[4] W. H. Zachariasen, *Theory of X-Ray Diffraction in Crystals* (John Wiley & Sons, Inc., New York, 1945).



J. Phys. Soc. Japan **31** (1971) 954∼955

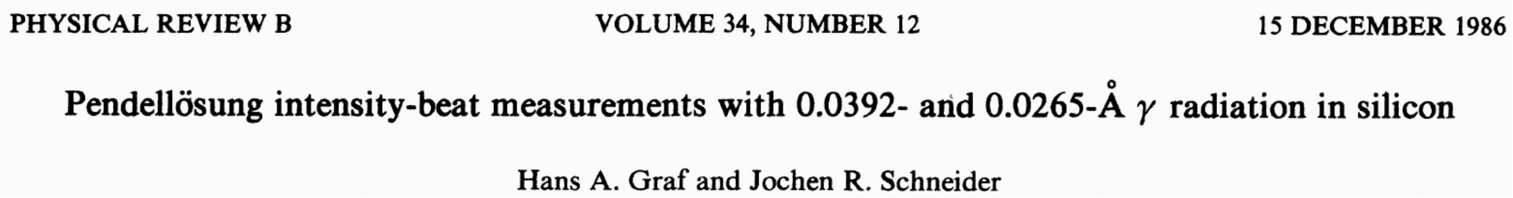
An Observation of Neutron Pendellösung Fringes in a Wedge-Shaped Silicon Single Crystal

Seishi Kikuta, Kazutake Kohra,
Nobuaki Minakawa* and Kenji Doi*

Acta Cryst. (1959). **12**, 787

A Study of Pendellösung Fringes in X-ray Diffraction

By N. Kato and A. R. Lang



PHYSICAL REVIEW B VOLUME 34, NUMBER 12 15 DECEMBER 1986

Pendellösung intensity-beat measurements with 0.0392- and 0.0265-Å γ radiation in silicon

Hans A. Graf and Jochen R. Schneider

B. Batterman, H. Cole, RMP 36, 681 (1964) and

P.P. Ewald, Rev. Mod. Physics 37, 46 (1965) and the references therein

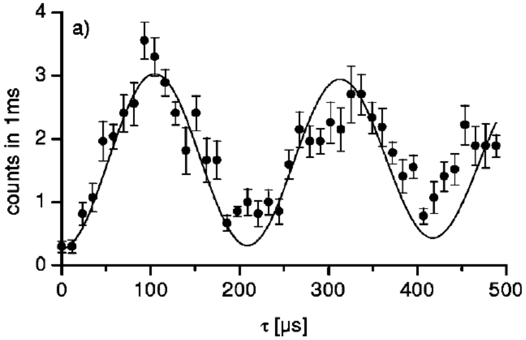
Previous works, atoms:

PHYSICAL REVIEW A VOLUME 60, NUMBER 1 JULY 1999

Dynamical diffraction of atomic matter waves by crystals of light

M. K. Oberthaler,[1,2] R. Abfalterer,[1] S. Bernet,[1] C. Keller,[1] J. Schmiedmayer,[1] and A. Zeilinger[1]

[1] Bragg refl. of atoms from standing light wave as fct. of interaction time, S. Dürr, G. Rempe, PRA 59, 1495 (1999)



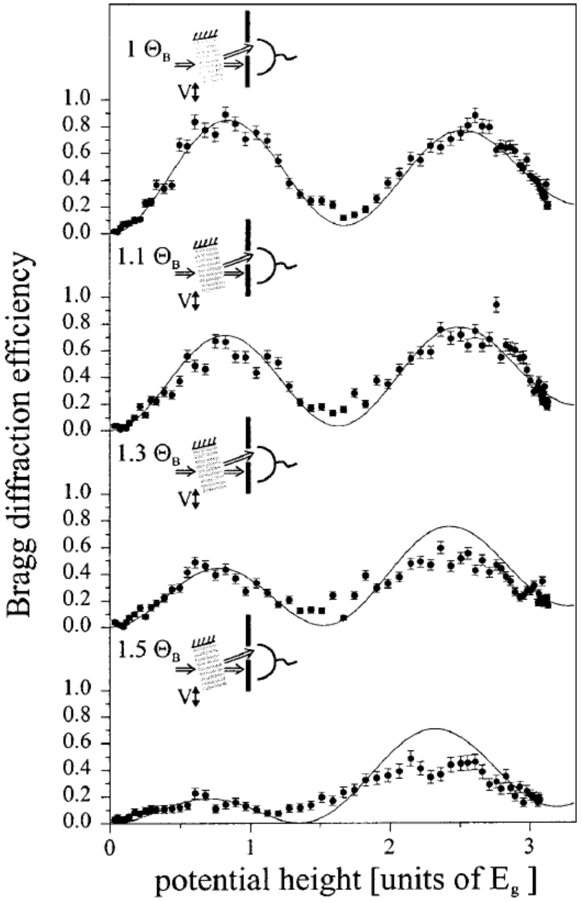
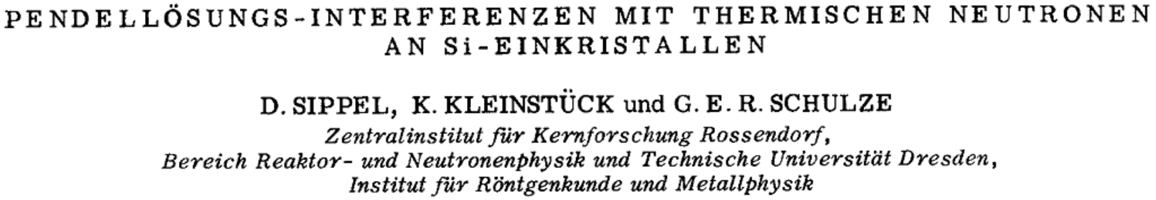


FIG. 12. Experimental demonstration of the Pendellösung phenomenon as a function of potential height for different incidence angles: The solid lines represent the predictions of dynamical diffraction theory.

Previous works, neutrons:



PENDELLÖSUNGS-INTERFERENZEN MIT THERMISCHEN NEUTRONEN
AN Si-EINKRISTALLEN

D. SIPPEL, K. KLEINSTÜCK und G. E. R. SCHULZE
Zentralinstitut für Kernforschung Rossendorf,
Bereich Reaktor- und Neutronenphysik und Technische Universität Dresden,
Institut für Röntgenkunde und Metallphysik

$$\eta = \sin^2\left(\frac{\lambda\, d\, b_C\, \Delta\rho}{2\cos\zeta\cos\theta_B}\right)$$

variation of thickness
Phys. Lett.(1965)

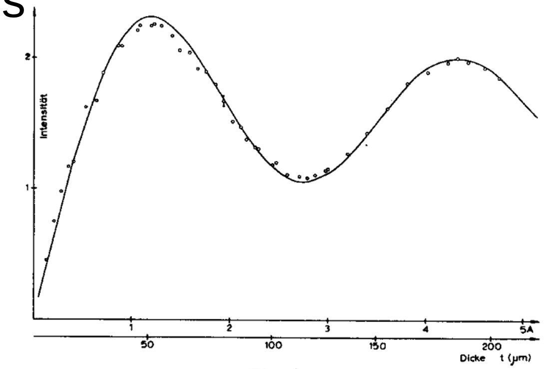


Fig. 1.

Somenkov et al., Solid State Communications, Vol. 25, pp. 593–595, 1978.

variation of thickness

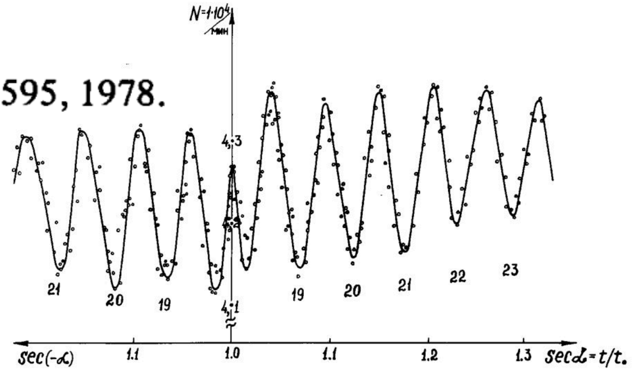


Fig. 2. Experimental dependence of the "peak intensity" as a function of sec α for reflection (2$\bar{2}$0) of Ge monocrystal having 1050 μm in thickness with λ = 1.45 Å obtained on the diffractometer with mosaic monochromator.

Previous works, neutrons II:

OBSERVATION OF PENDELLÖSUNG FRINGE STRUCTURE IN NEUTRON DIFFRACTION*

C. G. Shull

PRL 21, 1585 (1968)

variation of wavelength

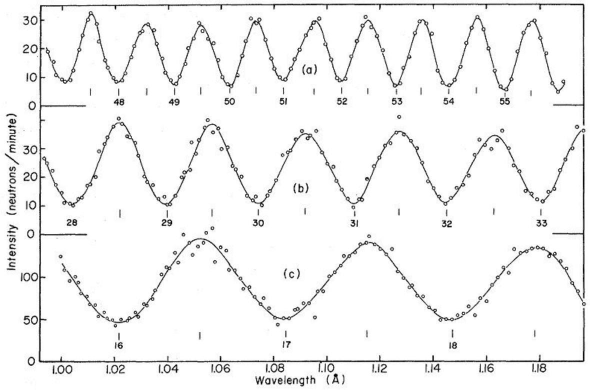
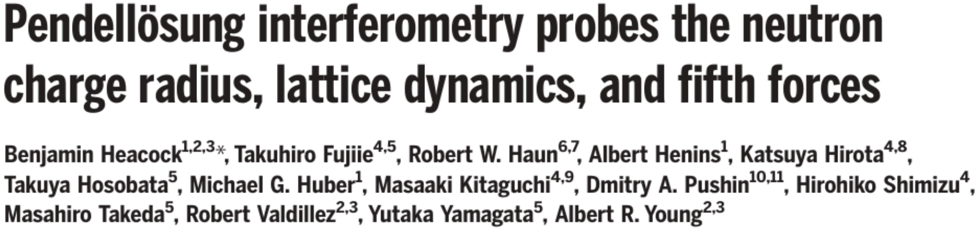


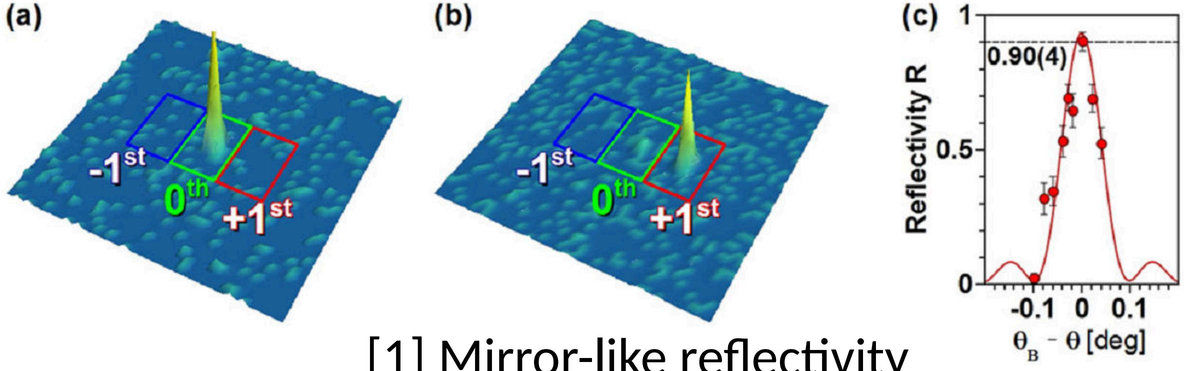
FIG. 3. Fringe development at the center of the Bragg reflection as the wavelength is increased. The three fringe patterns correspond to different values of crystal thickness: (a) 1.0000 cm, (b) 0.5939 cm, and (c) 0.3315 cm. Fringe order numbers are shown at the minima positions.



Pendellösung interferometry probes the neutron charge radius, lattice dynamics, and fifth forces

Benjamin Heacock[1,2,3*], Takuhiro Fujiie[4,5], Robert W. Haun[6,7], Albert Henins[1], Katsuya Hirota[4,8], Takuya Hosobata[5], Michael G. Huber[1], Masaaki Kitaguchi[4,9], Dmitry A. Pushin[10,11], Hirohiko Shimizu[4], Masahiro Takeda[5], Robert Valdillez[2,3], Yutaka Yamagata[5], Albert R. Young[2,3]

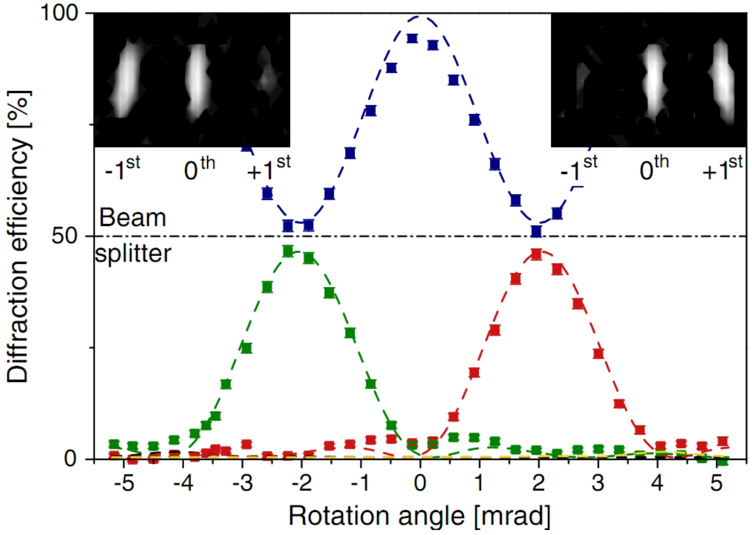
Heacock *et al.*, *Science* **373**, 1239–1243 (2021)

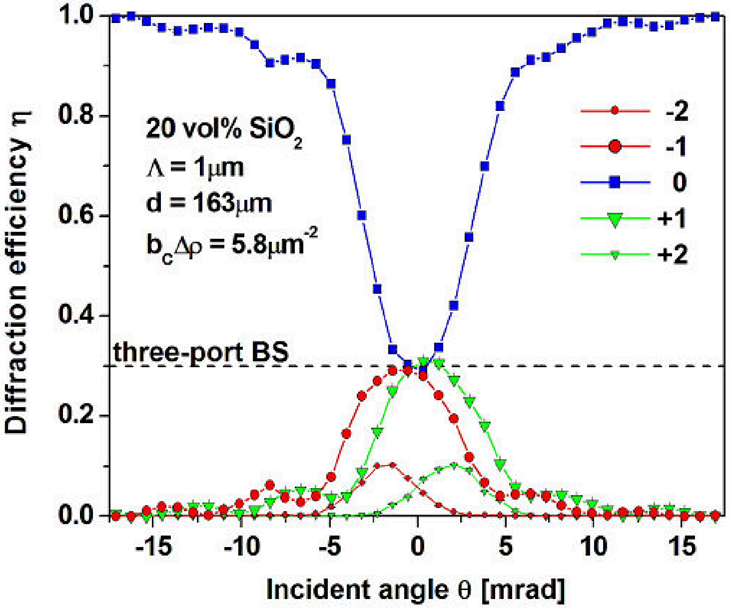


[1] Mirror-like reflectivity

[3] Three port beam splitter

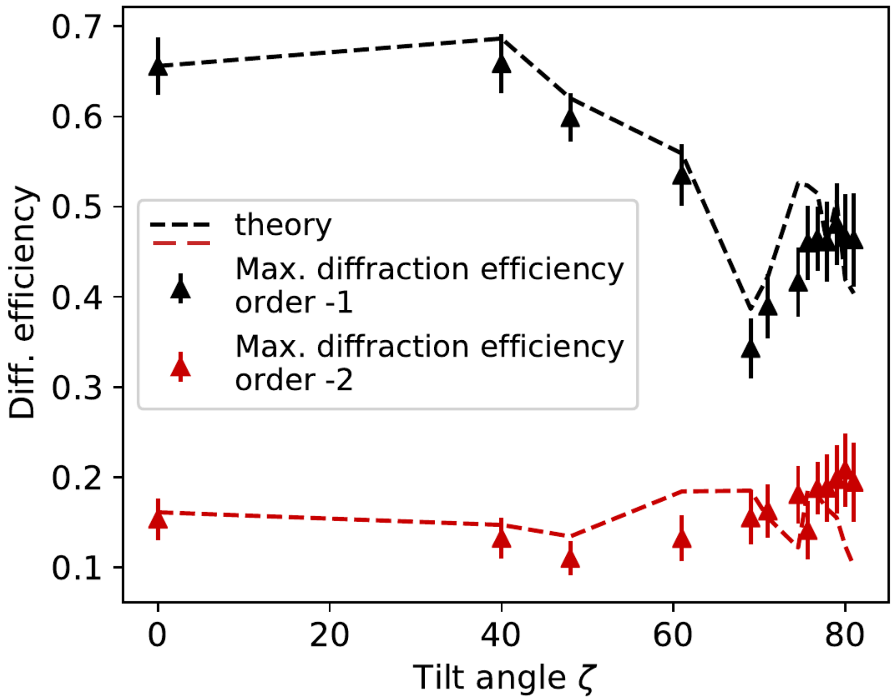
[2] Beam splitter





[1] J. Klepp et al., Appl. Phys. Lett. 100, 214104 (2012), [2] M. Fally et al., Phys. Rev. Lett. 105, 123904 (2010), [3] J. Klepp et al., Appl. Phys. Lett. 101, 154104 (2012)

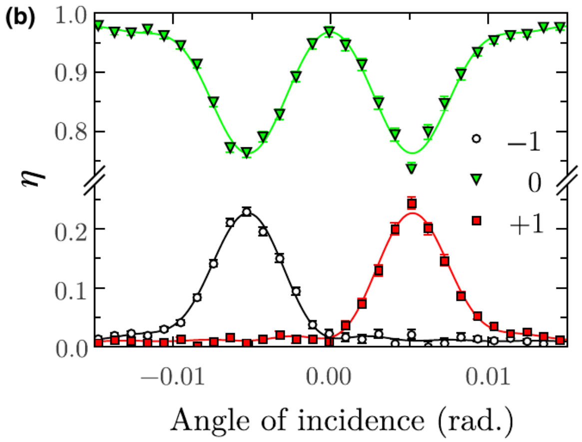
Some preliminary Pendellösung results:



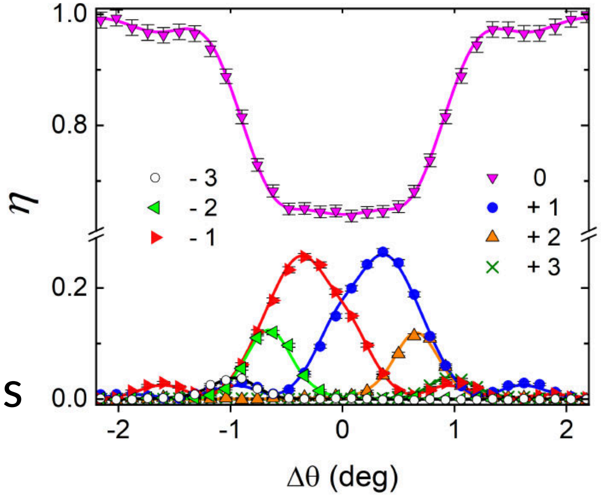
I. V. Masiello, Master thesis, University of Aarhus, University of Vienna (2022)

Some more results for holographic gratings:

[1] Nanodiamond gratings: high η, low angle selectivity

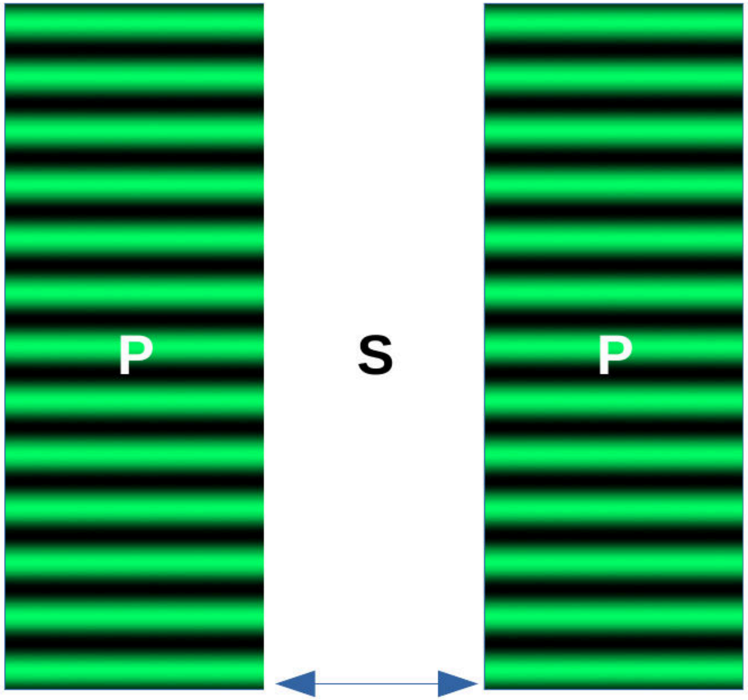


[2] Grating strength record for nanodiamond composite materials

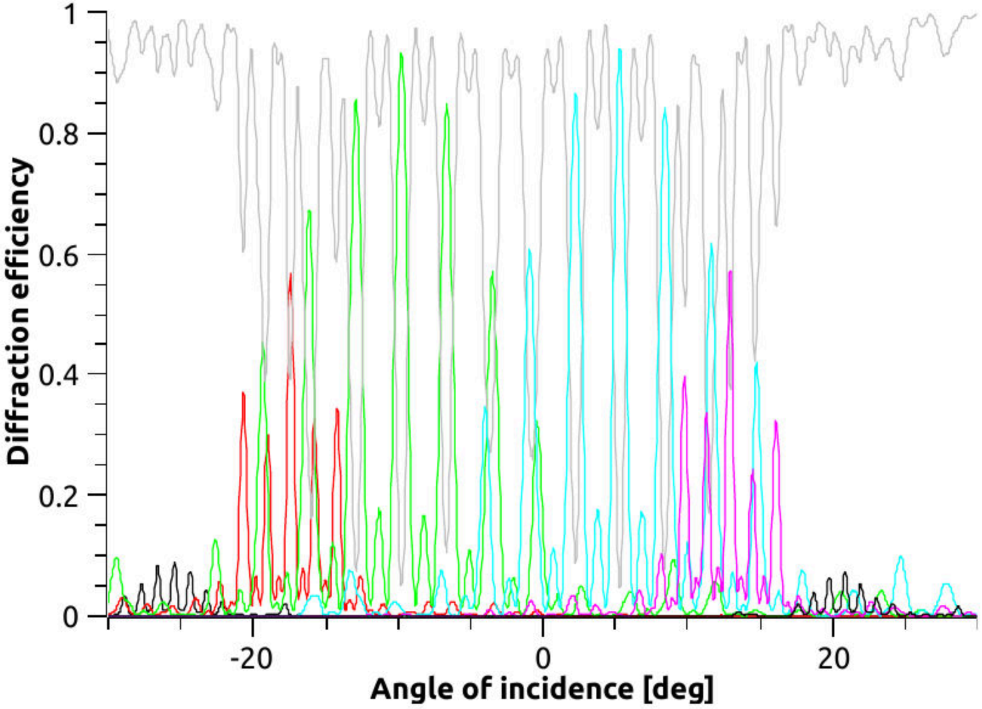


[1] Y. Tomita et al., Phys. Rev. Applied 14, 044056 (2020), [2] M. Fally et al., Optics Express 29, 16153 (2021)

Another idea for VCN-economic IFM methods….



Multilayer holographic grating

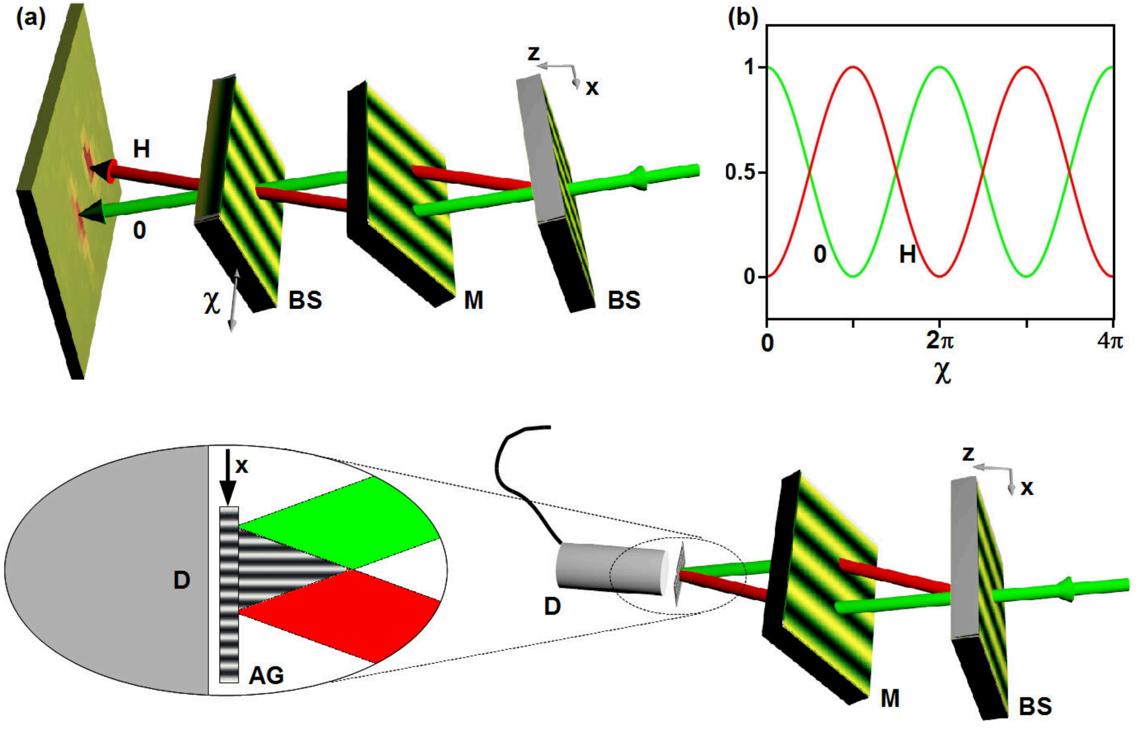


A device of a couple of cm² area and ~100 µm thickness, neutrons interfering within, no space for putting stuff in one of the beampaths, but little adjustment, no feedback-loop readjustment, no temperature control, no vibrational damping,… necessary

Austrian Science Fund Infrastructure programm "Quantum Austria - NextPi" partly (150 kEUR) for design and prototype construction of a VCN IFM at PF2, ILL, Grenoble

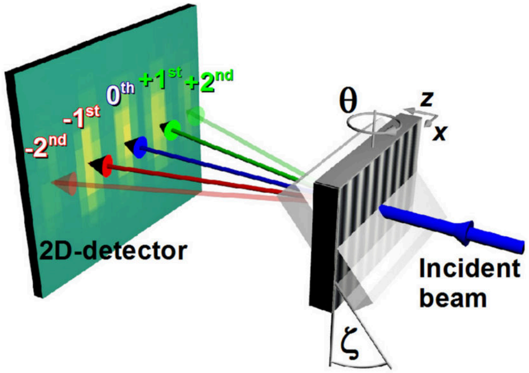
Proj. No. FO999896034

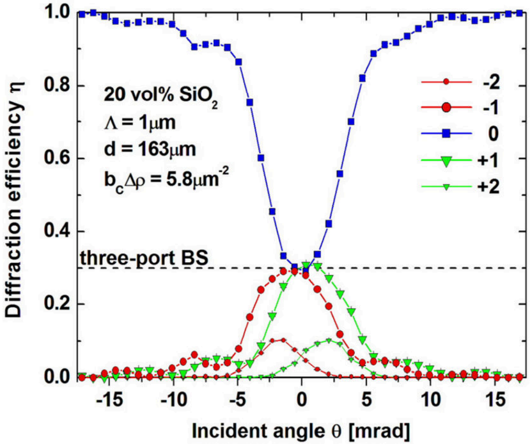
Quantum Austria: VCN IFM layouts using holographic gratings

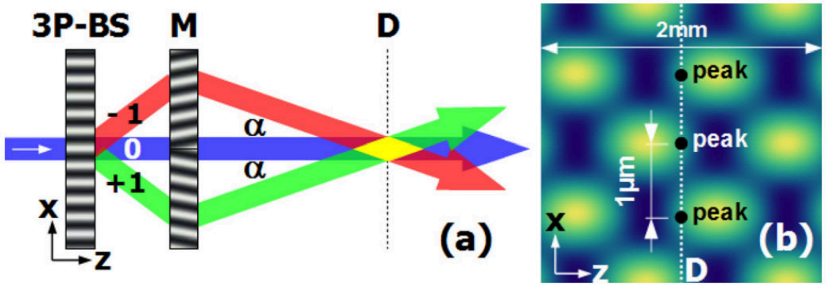


J. Klepp et al., Materials 5, 2788 (2012)

Quantum Austria: VCN IFM layouts using holographic gratings







Suppose we require to maintain at least 1 cm separation between the three beams (of 1 mm width, say) at point M to conveniently insert a sample into the central beam. From geometrical considerations, it is found that for an incident neutron wavelength of 8 nm and a three-port beam splitter with a grating spacing of 0.5 μm, the interference pattern formed along the x direction at D has a periodicity of 1 μm at a beam splitter-to- detector distance of about 3.75 m. The pattern is spread 25 cm along the optical axis and at most 1 mm along the x direction.

J. Klepp et al., Appl. Phys. Lett. (2012)

Summary:

- Very cold neutrons still come in beams and exhibit long interaction times (compared to other parts of the neutron spectrum forming beams)
- Very cold neutrons are scarce!
- A little history: The COW experiment and the VCN contribution
- Holographic diffraction gratings for VCN
- A project to build a new VCN IFM at PF2 VCN of the ILL
- Try to develop interferometric methods that make use of divergent beams and wide wavelength distribution
  - Multilayer gratings
  - Develope VCN methods to use the Pendellösung interference effect

Proj. No. FO999896034

Der Wissenschaftsfonds.
Proj. No. P35597

Martin Fally[1], Hanno Filter[2], Peter Geltenbort[2], Elhoucine Hadden[2,] Tobias Jenke[2], Joachim Kohlbrecher[3], Christian Pruner[4], Yasuo Tomita[5], Jürgen Klepp[1]

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[2]Institut Laue Langevin, Grenoble, France

[3]Paul Scherrer Institut, Villigen, Switzerland

[4]Department of Material Science and Physics, University of Salzburg, Austria

[5]Department of Engineering Science, University of Electro-Communications, Tokyo, Japan

Thank you!