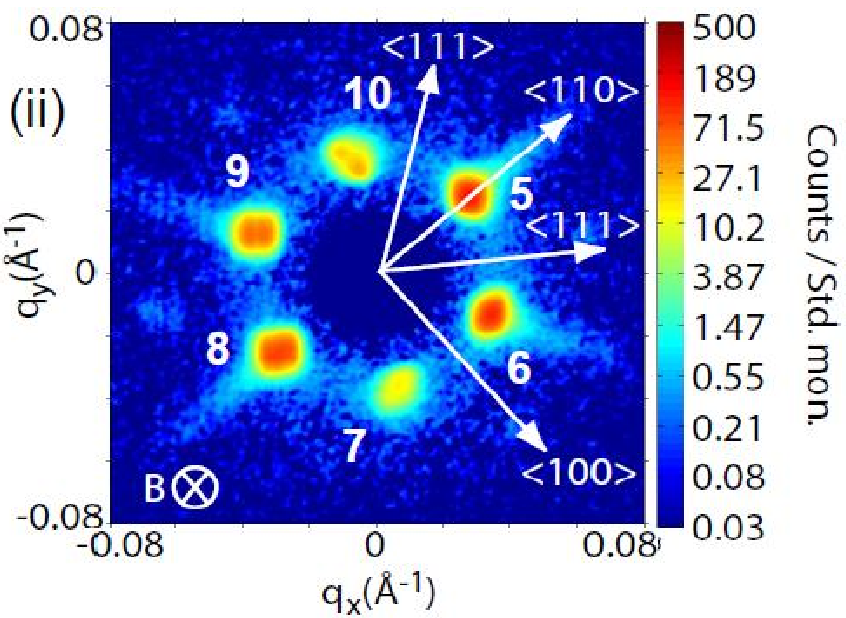
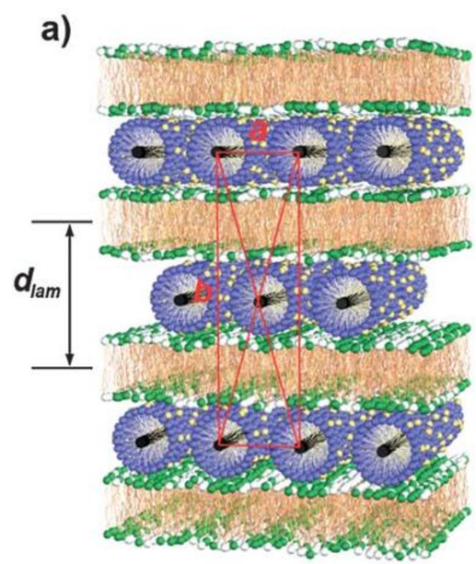
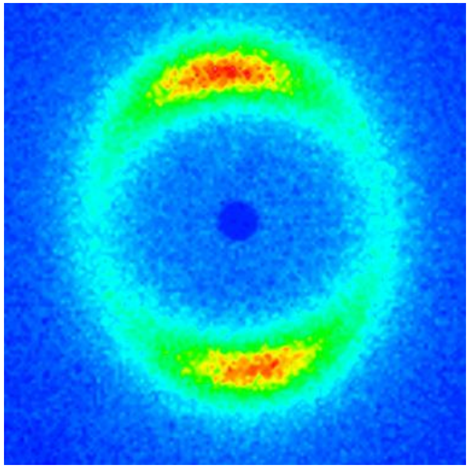
Physics with Neutrons II, SS 2016







Lecture 5, 30.5.2016

MLZ is a cooperation between:

JÜLICH FORSCHUNGSZENTRUM

Helmholtz-Zentrum Geesthacht
Zentrum für Material- und Küstenforschung

Technische Universität München

Plan for SS 2016

- VL1: Repetition of winter term, basic neutron scattering theory
- VL2: SANS, theory and applications
- VL3: Neutron optics
- VL4: Reflectometry and dynamical scattering theory
- VL5: Diffuse neutron scattering
- VL6: Magnetic elastic scattering (diffraction)
- VL7: Magnetic structures and structure analysis
- VL8: Polarized neutrons and 3d-polarimetry
- VL9: Inelastic scattering on magnetism
- VL10: 4.7.2016 (8:30!!)Phase transitions and critical phenomena as seen by neutrons
- VL11: Spin echo spectrocopy

Exam: Please register until 30.6.2016

Reminder:
Dynamical scattering theory:
Index of refraction and reflectometry

Dynamical scattering theory

Born approximation: Intensity would diverge for Q->0

$$I(Q_z) \sim \frac{1}{Q_z^4} \left| \int \frac{\mathrm{d}V(z)}{\mathrm{d}z} \exp(-iQ_z z) \mathrm{d}z \right|^2$$

Complete description: Dynamical scattering theory

Derivation using Schrödinger equation for neutron wave in medium

Single, smooth layer using QM

$$\left[-\frac{\hbar^2}{2m_n} \Delta + V(r) \right] \Psi(r) = E\Psi(r)$$

Refractive index for neutrons (very small)

$$n_t = 1 - \frac{\lambda^2}{2\pi} \sum_j b_j \varrho_j = 1 - \delta_t$$

Transmitted and reflected wave

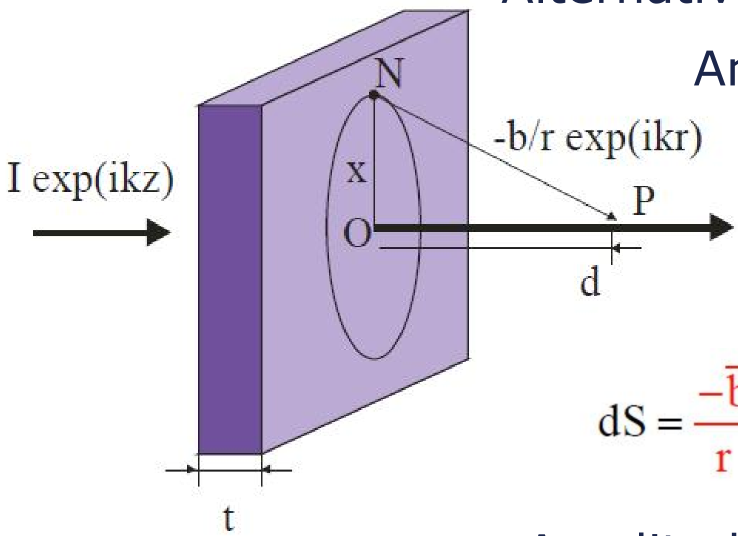
$$r_t = \frac{2k_z}{k_z + k_{t,z}} \qquad r_f = \frac{k_z - k_{t,z}}{k_z + k_{t,z}}$$

Absorption: Imaginary part

$$n_t = 1 - \delta_t + \mathrm{i}\beta_t$$

Dynamical scattering theory

Alternative derivation of refractice index



Amlitude at point P produced by spherical waves scattered from N

$$S_N = \frac{-b}{r}\exp(ikr)$$

Contribution of circle of radius x

$$dS = \frac{-\bar{b}}{r}\exp(ikr)\,\rho\; t2\pi x\,dx = -2\pi\bar{b}\rho t \exp\left(ik\sqrt{x^2+d^2}\right)\frac{x}{\sqrt{x^2+d^2}}dx$$

Amplitude of scattered wave at point P

$$S = -2\pi\bar{b}\rho t\int_0^{\infty}\exp\left(ik\sqrt{x^2+d^2}\right)\frac{x}{\sqrt{x^2+d^2}}dx = -i\frac{2\pi}{k}\bar{b}\rho t\exp(ikd)$$

Scattered and direct beam retarded with respect to incident beam

$$S + I = \left(1 - i\bar{b}\lambda\rho t\right)\exp(ikd) \approx \exp(ikd+\phi) \qquad \phi = -\bar{b}\lambda\rho t$$

Retardation can be expressed in terms of an refraction index

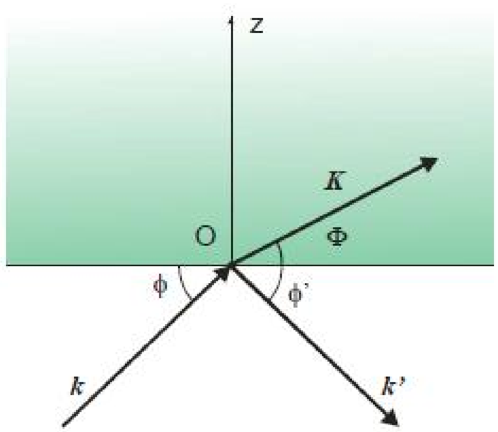
$$\phi = \frac{2\pi}{\lambda}(1-n)t \qquad \boxed{n = \frac{k_{medium}}{k_{vacuum}} = 1 - \frac{1}{2\pi}\rho\lambda^2\bar{b}}$$

Dynamical scattering theory

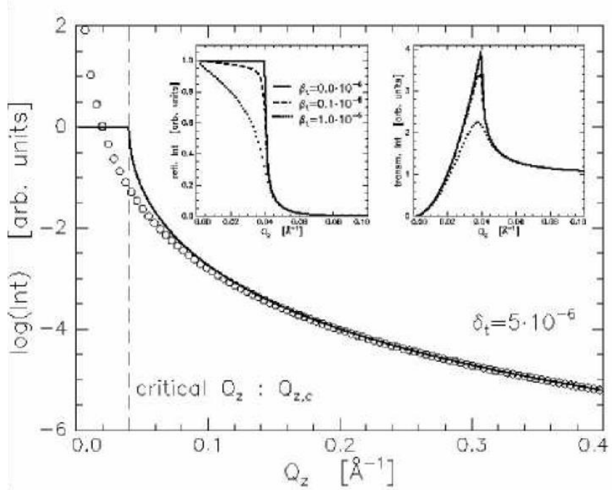
Consequences for neutron reflectivity

Fresnel formulas for reflected and transmitted beam

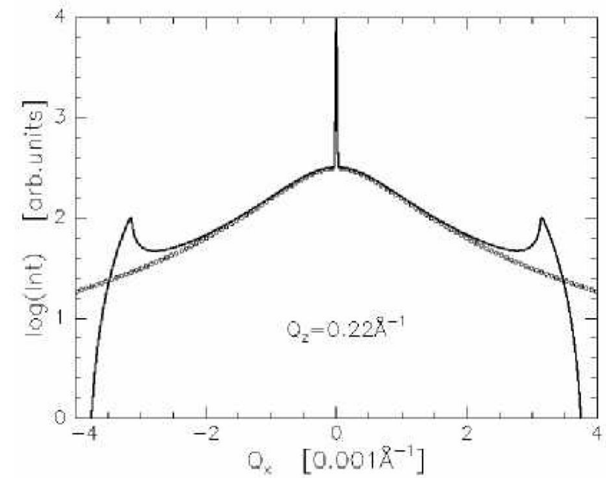
$$r_t = \frac{2k_z}{k_z + k_{t,z}} \qquad r_f = \frac{k_z - k_{t,z}}{k_z + k_{t,z}}$$

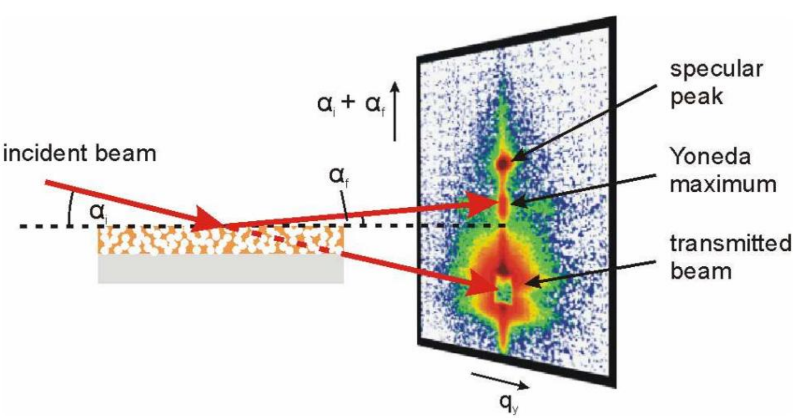


Plateau of total external reflection for $\Theta<\Theta_c$



Evanescent wave for $\Theta=\Theta_c$ (Yoneda peak)



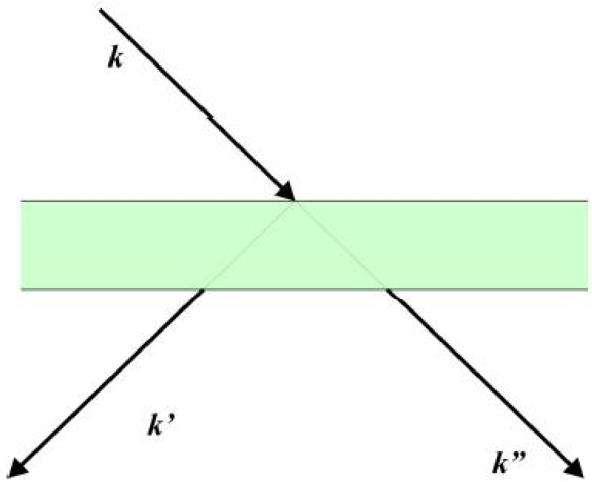


⇨ Full QM treatment only possible for specular reflectivity

Reminder:
Dynamical scattering theory:
Modifications to diffraction

Dynamical scattering theory

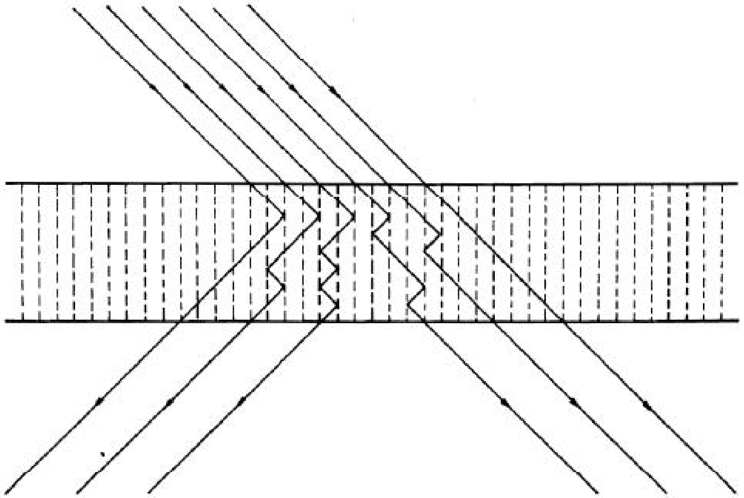
Neutron conservation? Multiple scattering?



$$I_{hkl} \propto N \frac{(2\pi)^3}{V_0} |F_{hkl}|^2$$

Diffracted intensity proportional to sample thickness.

→ Transmission?



→ Rigorous treatment of multiple scatting

Dynamical scattering theory

Full treatment only using Schrödinger equation

Periodic potential $V(r) = \frac{2\pi\hbar^2}{m_n}\langle\sum b_j\delta(r-R_j)\rangle$ with $R_j = l + d + u(^l_d)$

Use lattice sum identities to rewrite potential

$$\langle\sum b_j\delta(r-R_j)\rangle = \frac{1}{v_0}\sum_\tau F_\tau e^{-i\tau r} \qquad F_\tau = \sum_d \bar{b}_d e^{i\tau d} e^{-W_d}$$

$$W_d = \frac{1}{2}\langle(\tau u(^l_d))^2\rangle$$

Insert into Schrödinger equation:

$$\frac{\hbar^2}{2m_n}\nabla^2\Psi(E-V(r))\Psi = 0$$

Solution: Bloch waves $\Psi = \sum_\tau a_\tau e^{i(k-\tau)r}$

Coefficients: $a_\tau(k_0^2 - (k-\tau)^2) = \sum_{\tau'}\frac{4\pi}{v_0}F_{\tau'}a_{\tau-\tau'}$

Something happens close to Bragg peaks:

Dynamical scattering theory

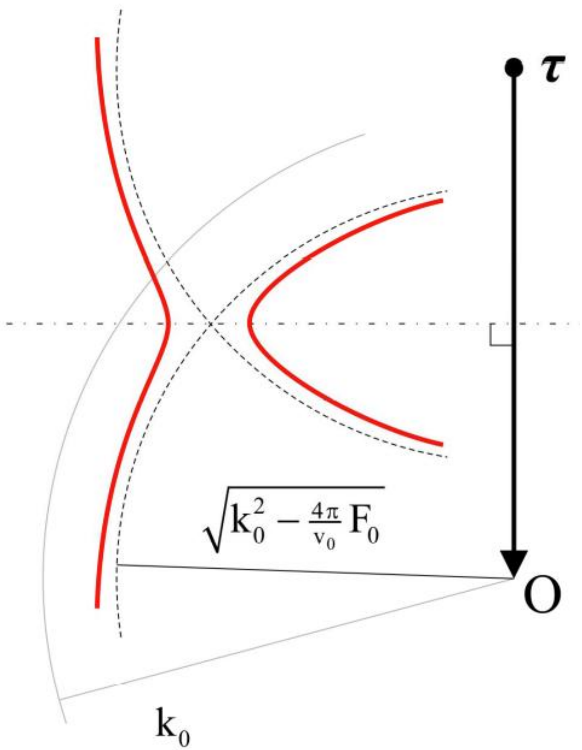
So far: Eigenstate of a neutron in a crystal is a plane wave

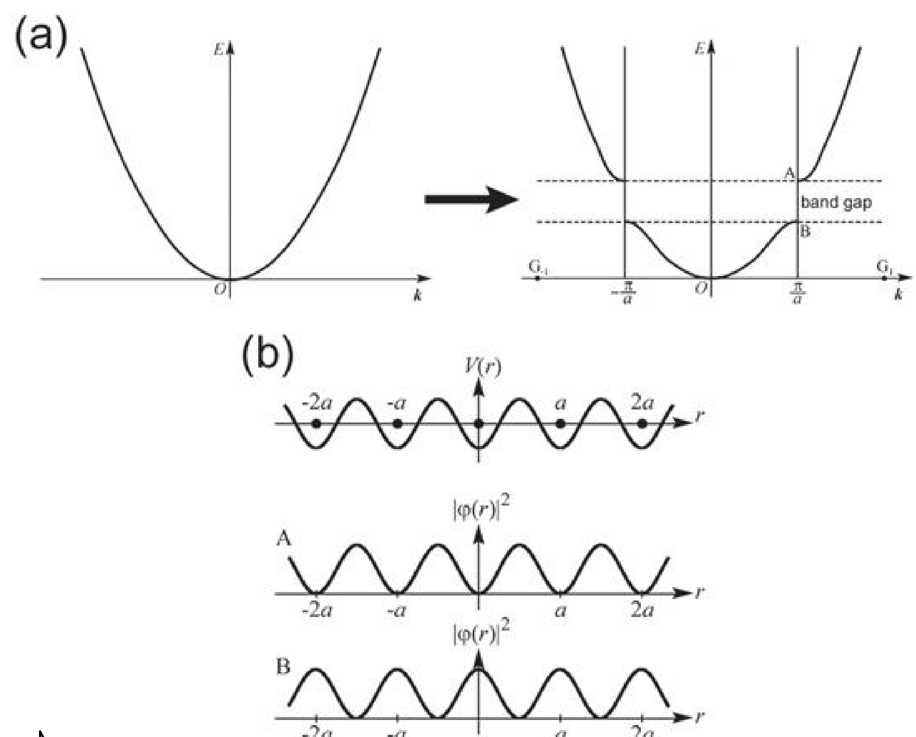
Rigorous treatment: Neutron as some kind of Bloch waves

➡ Dispersion surfaces: Manifold of k-values of a Bloch wave where energy is equal to:

$$E = \frac{\hbar^2}{2m} k_0^2$$

Electrons in solids: Bloch waves

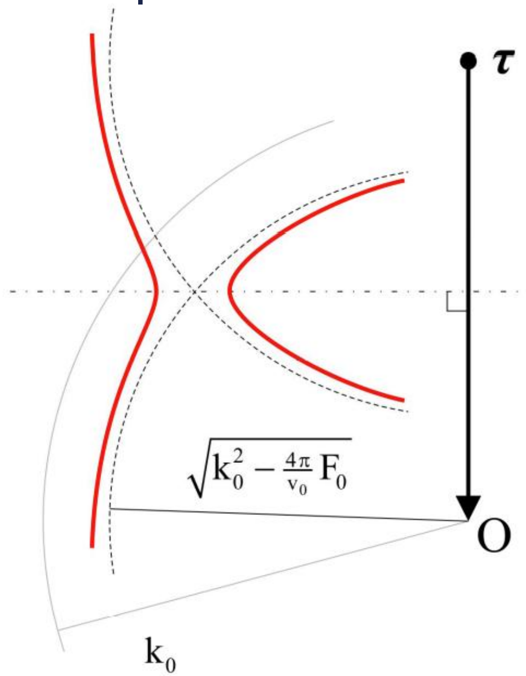




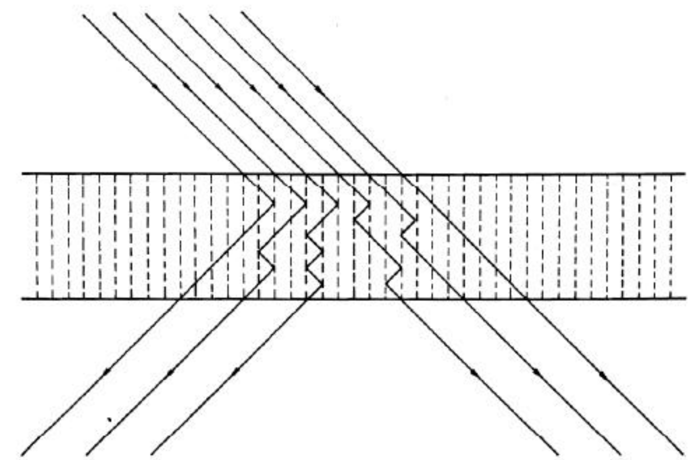
➡ Band gap opens at the zone boundary

Dynamical scattering theory

Dispersion surfaces



Multiple scattering



Consequences of dynamical scattering theory:

Far away from Bragg peaks

Close to Bragg peaks
in high quality single Xtals

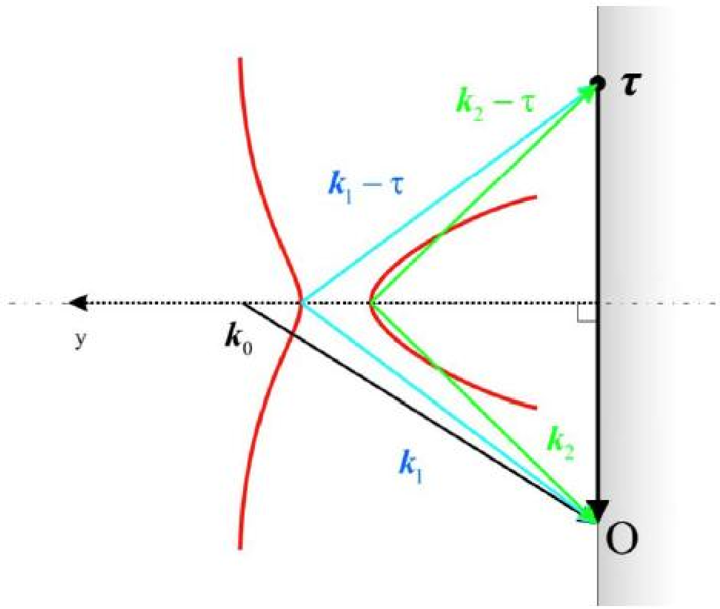
Kinematic description (Born approximation) gives right result

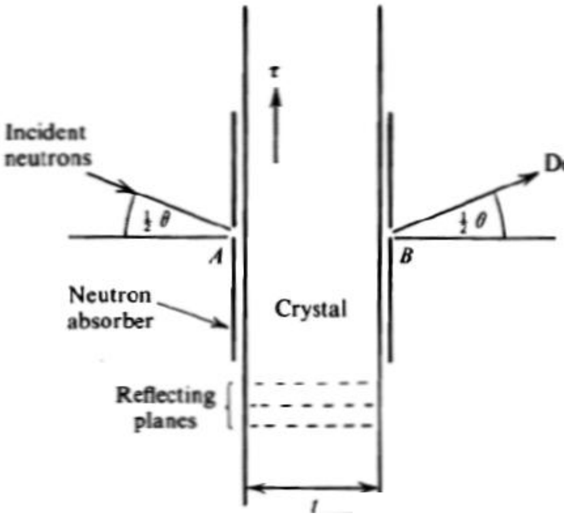
Kinematic description (Born approximation) is strongly modified

Dynamical scattering theory

Consequences of dynamical scattering theory:

Pendellösung oscillations (Kato fringes)



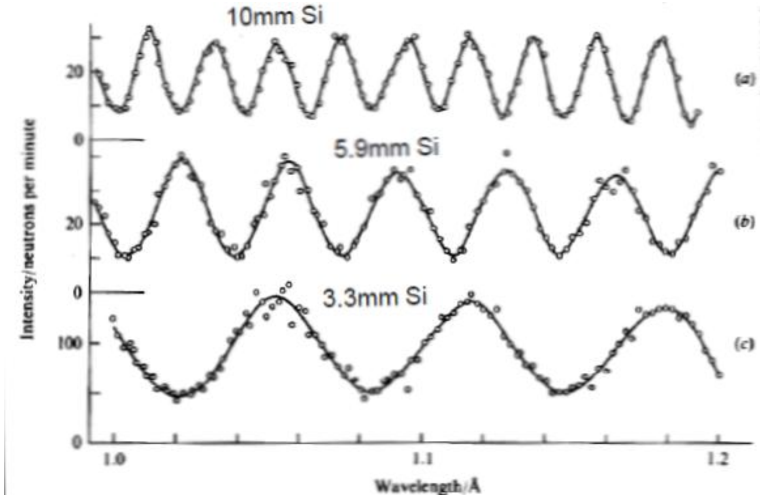


Interference fringes between k_1 and k_2:

⇨ Intensity oscillations

Energy is constantly redistributed from one wave to the other.

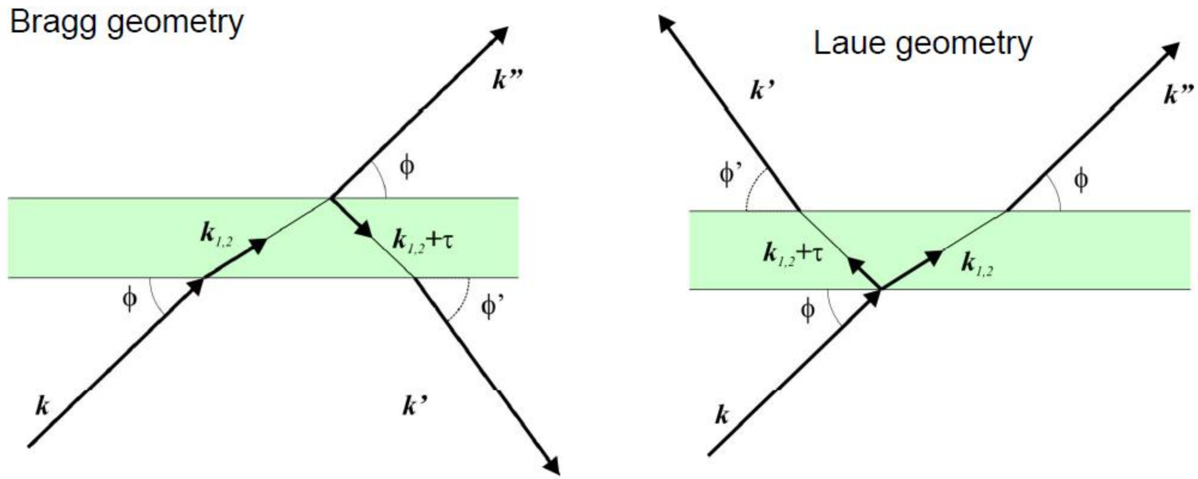
⇨ Anomalous transmission

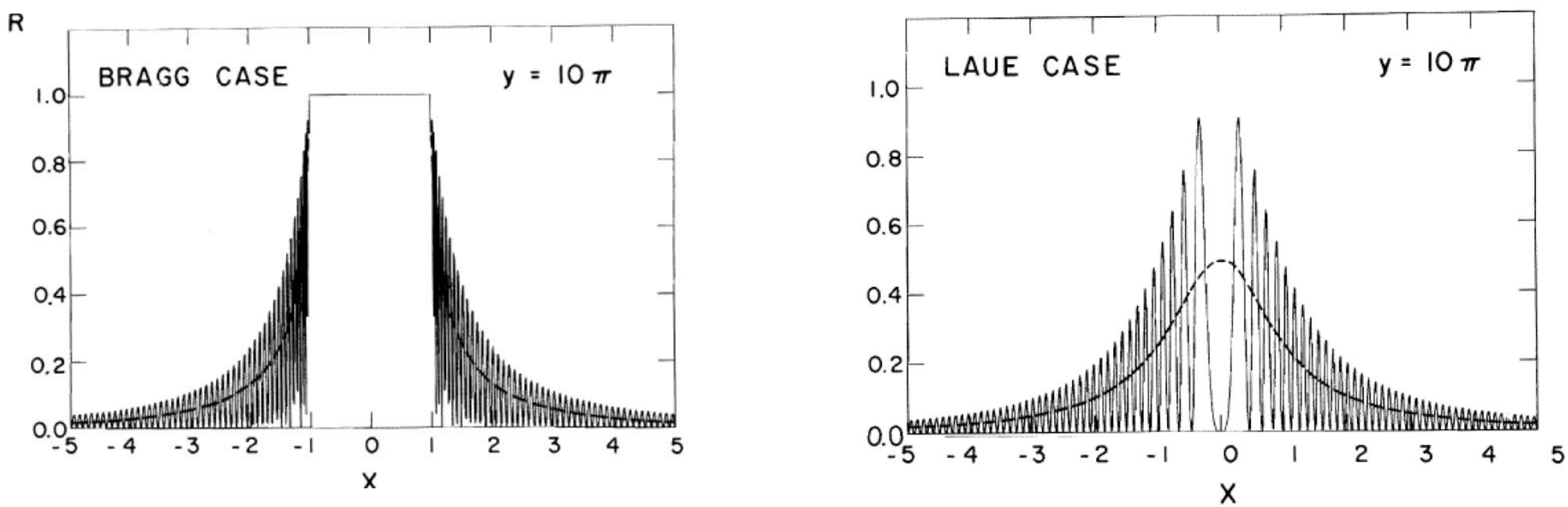


Dynamical scattering theory

Consequences of dynamical scattering theory:

Shape, width and intensity of Bragg peaks modified

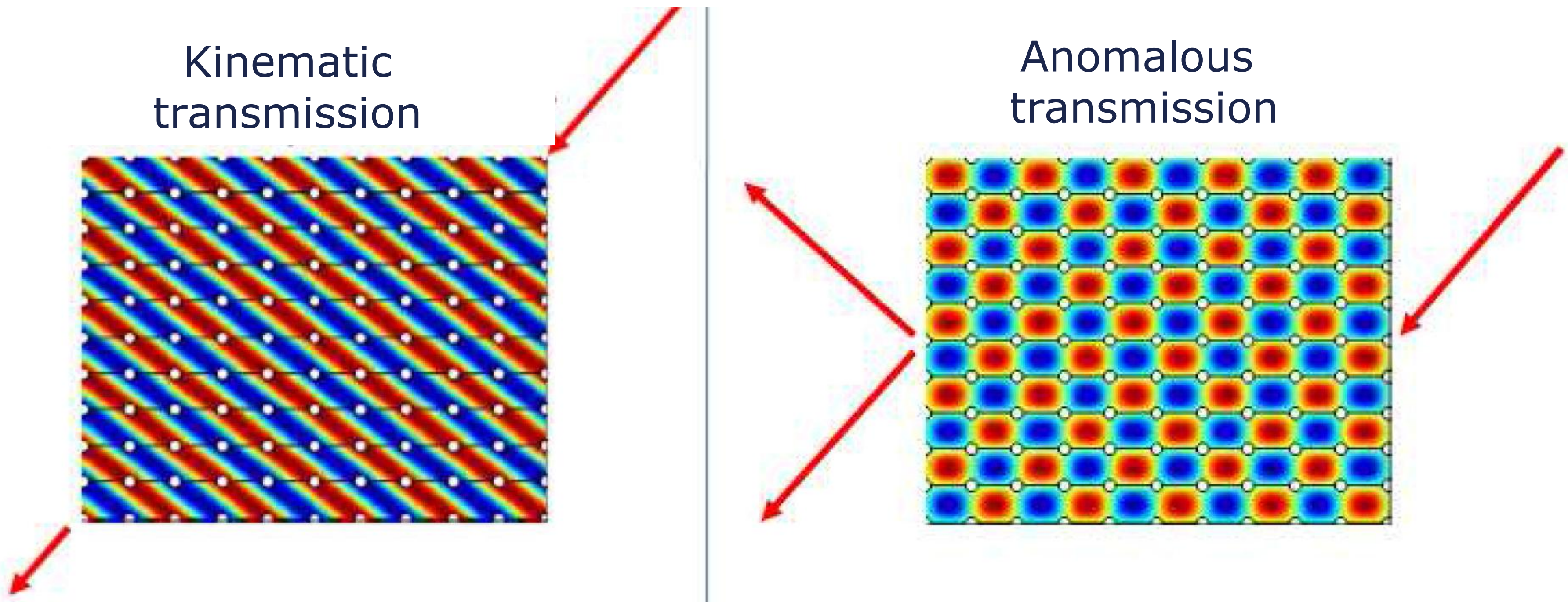




Dynamical scattering theory

Consequences of dynamical scattering theory:

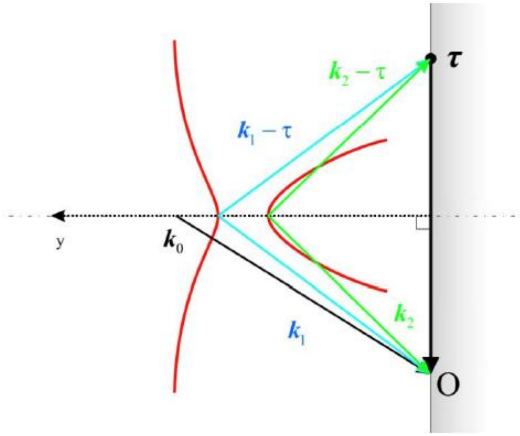
Anomalous transmission (Borrmann fan)



Dynamical scattering theory

Consequences of dynamical scattering theory:

Primary extinction

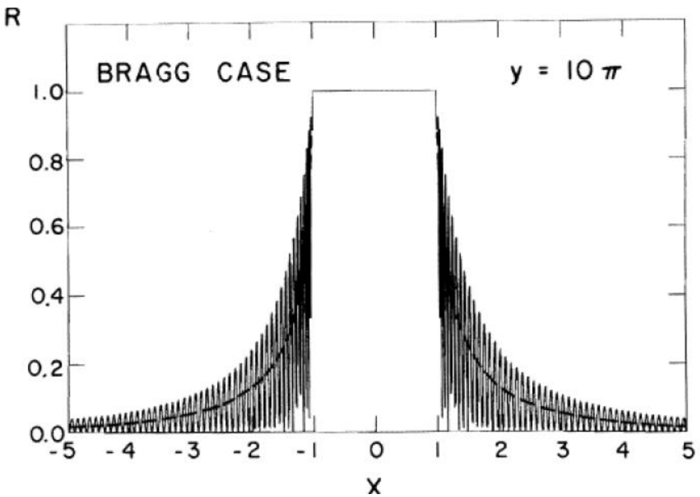


- Only the surface contritutes to Bragg reflections
- Intensity not given by structure factor (measurements are too low)

Define extinction length ξ

$$(k_1-k_2)\frac{\xi}{\cos\frac{1}{2}\theta}=1$$

Given by phase difference of k_1 and k_2



- Only several μm for good crystal samples
- Size ξ of the crystal reflects: Darwin width of peaks

$$\Delta\theta\approx\frac{1}{\tau\xi}$$

Diffuse Neutron Scattering: Looking between the Bragg spots

Diffuse Neutron Scattering

Master formula for elastic coherent scattering on crystals

$$\frac{d\sigma}{d\Omega}_{coh.el} = N_0 \frac{(2\pi)^3}{v_0} e^{-2W(\vec{\kappa})} \sum_{\vec{\tau}} |S_{\vec{\tau}}|^2 \, \delta(\vec{\kappa} - \vec{\tau})$$

Normalization   Debye-Waller   Structure factor   Bragg positions

Definition: Diffuse scattering is scattering that arises from departures of any kind from a perfect regular crystal lattice

- Phonons: Thermal diffuse scattering (TDS)
- Distortions due to defects: Huang scattering
- Static of dynamic displacements
- Stubstitution or interstitial defects
- Stacking faults
- (Micro-)Domains
- Magnetic effects
- ......

Diffuse neutron scattering can be static or dynamic

Diffuse Neutron Scattering

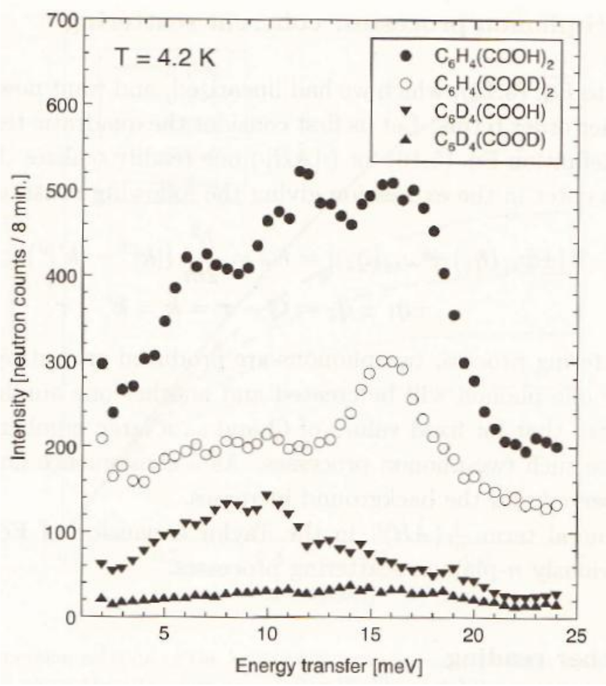
What we already know: Incoherent inelastic scattering

- Flat in Q (apart from DW)
- Gives Phonon DOS

$$\frac{d^2\sigma}{d\Omega d\omega} = \frac{1}{4M}\frac{k'}{k}\left(\langle b^2\rangle - \langle b\rangle^2\right) e^{-W(\boldsymbol{Q})} \times \langle(\boldsymbol{Q}\cdot e_s(\boldsymbol{q}))^2\rangle \cdot \frac{g(\omega)}{\omega} \cdot \left[\coth\frac{\hbar\omega}{2k_BT} \pm 1\right]$$

With phonon DOS g(ω)

$$\int_0^\infty g(\omega)d\omega = 3N$$



Diffuse Neutron Scattering

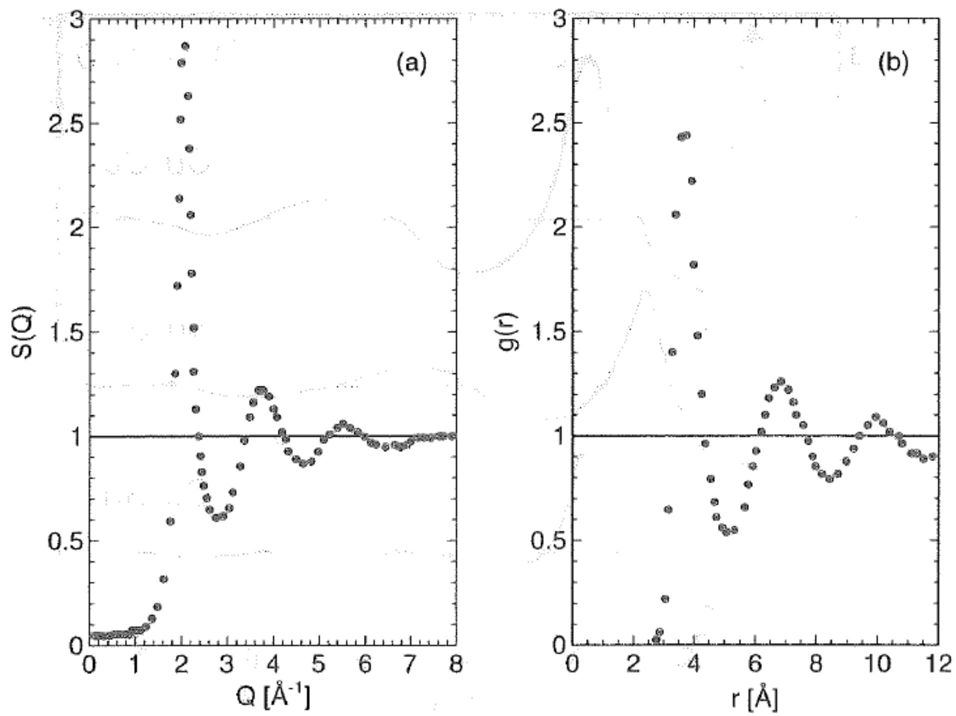
What we already know: Scattering on liquids and amorphous materials

➡ Measure of pair correlation function g(r)

➡ Quasielastic scattering: Diffusion

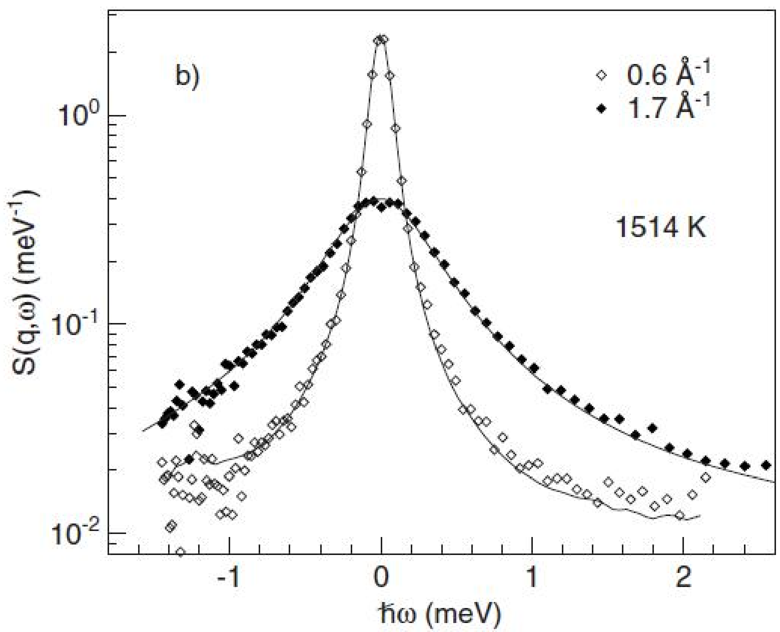
Static structure factor:
Deviations of the mean density n(r)

Dynamic structure factor:
Diffusive processes



Static structure factor: Scattering function

g(r) pair corr. function: Deviations from mean density n(r).



S(Q,w) ⟷ FT ⟷ G(r,t)

QENS, peaked at w=0

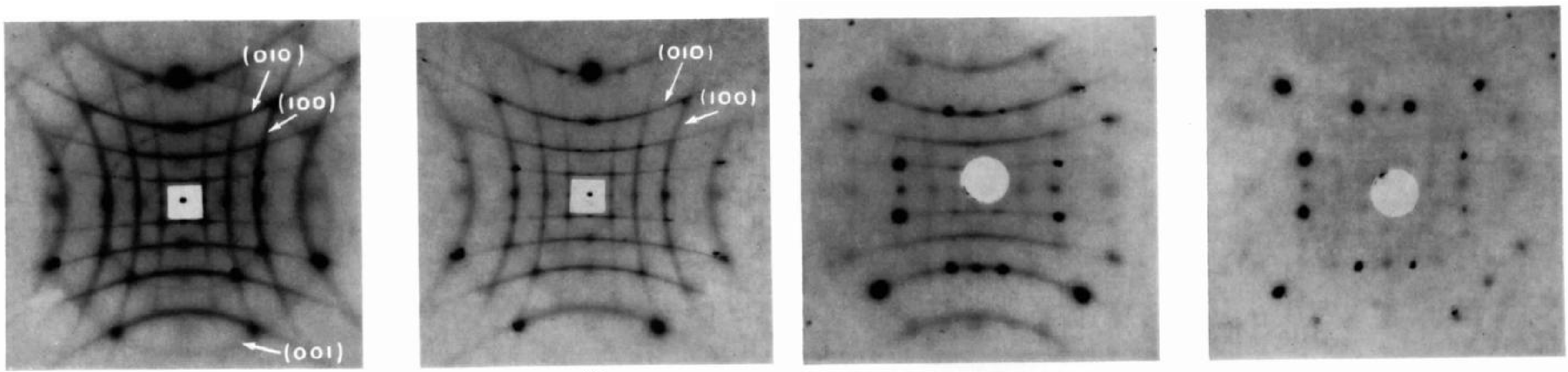
Example 1 — Ferroelectric Perovskite $KNbO_3$

Acta Cryst. (1970). A**26**, 244

Désordre Linéaire dans les Cristaux (cas du Silicium, du Quartz, et des Pérovskites Ferroélectriques)

PAR R. COMÈS, M. LAMBERT ET A. GUINIER

Service de Physique des Solides, Faculté des Sciences, 91-Orsay, France*



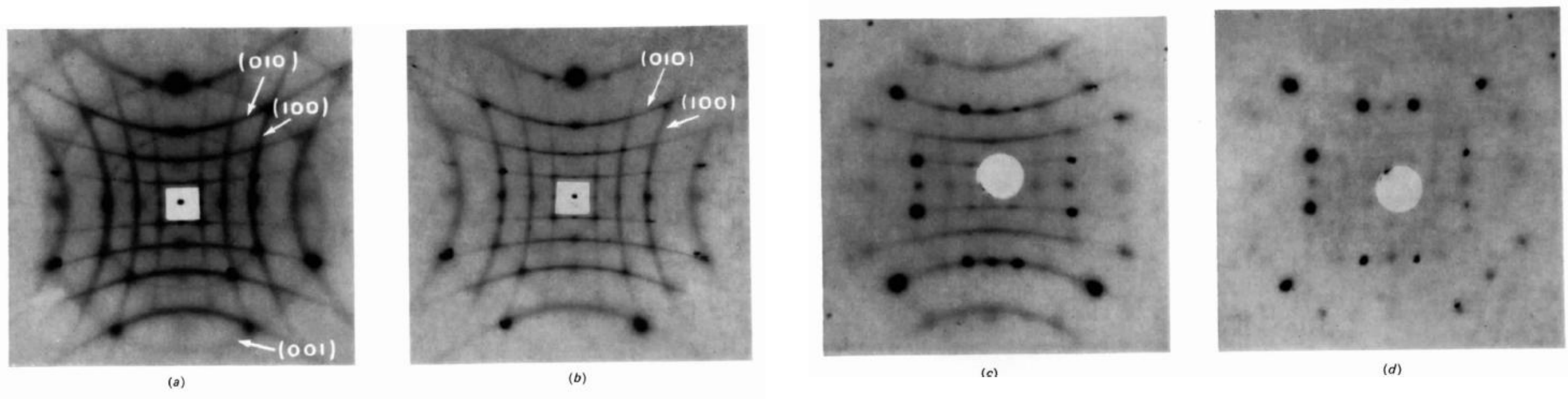
(*a*) (*b*) (*c*) (*d*)

Fig. 6. Diagrammes de diffusions obtenus avec $KNbO_3$ (axe *b* vertical, axe *c* presque parallèle au faisceau incident, λ = Mo $K\alpha$, exposition 2 heures). (*a*) Phase cubique: $T = 500\,^\circ C$. On observe des diffusions dans les *trois* famille de plans {100}; la légère désorientation de l'axe *c* par rapport au faisceau incident permet de s'assurer que les anneaux observés ne sont pas dus à une poudre parasite. (*b*) Phase tétragonale: $T = 250\,^\circ C$ [même cristal qu'en (*a*)]. La famille de plan (001) a disparue, les *deux* familles (100) et (010) subsistent. (*c*) Phase orthorhombique: $T = -56\,^\circ C$. Il ne subsiste qu'*une* seule famille de plans de diffusion: les plans (010). (*d*) Phase rhomboédrique: Il s'agit du même cristal que (*c*) et à la *même température* (grace à l'hystérésis thermique de la transition orthorhombique⇔rhomboédrique).

X-Ray diffraction (1970): What is going on here?
Phonons? Ionic chains?

Diffuse Neutron Scattering

Example 1 Ferroelectric Perovskite $KNbO_3$



Two alternative models for diffuse sheets:

- Formation of linear chains of correlated vibrational dispalcements due to a specific soft mode? Thermal diffuse scattering?
- Formation of linear chains of correlated local displacements?

Diffuse Neutron Scattering

Ferroelectric Perovskite $KNbO_3$

Compare to $PbTiO_3$ and $BaTiO_3$

??

Formation of linear chains of correlated vibrational displacements due to a specific soft mode?
Thermal diffuse scattering?

Soft mode similar for $PbTiO_3$, $BaTiO_3$ and $KnbO_3$

Expect similar results

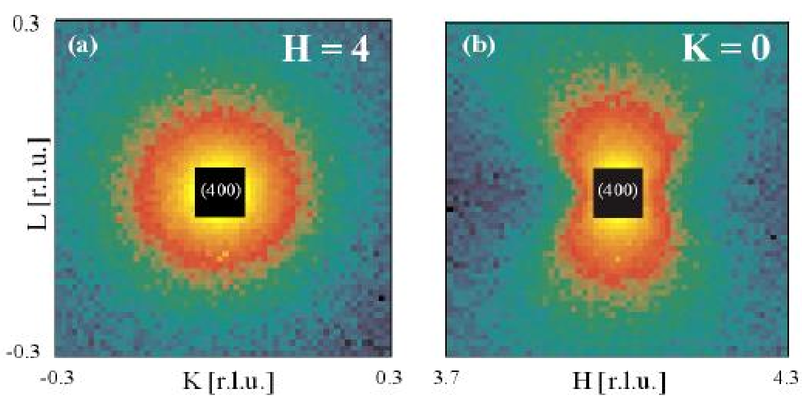
Formation of linear chains of correlated local displacements?

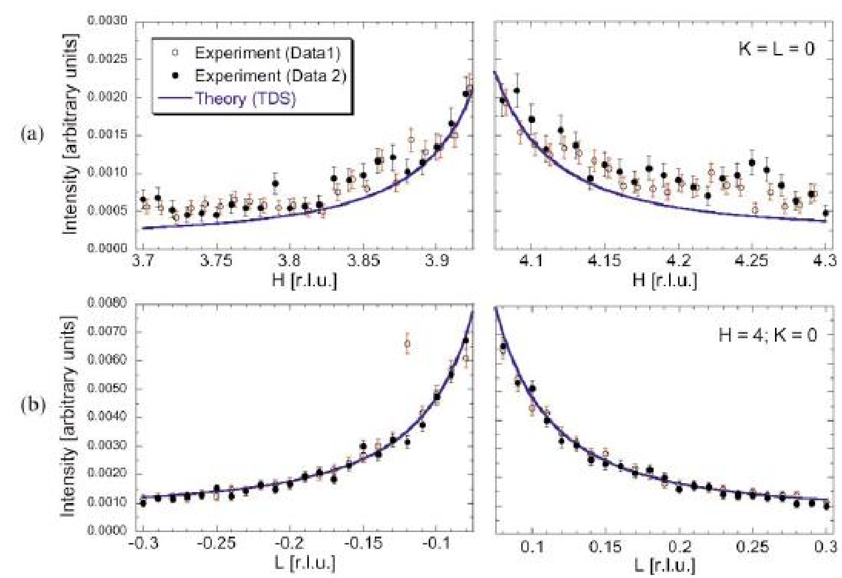
Different local symmetry

No diffuse sheets for $PbTiO_3$

Expect different result

Thermal diffuse scattering $PbTiO_3$





Diffuse intensity around Bragg positions

Diffuse Neutron Scattering

Ferroelectric Perovskite $KNbO_3$

Formation of linear chains of correlated local displacements

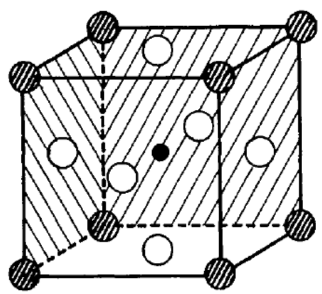
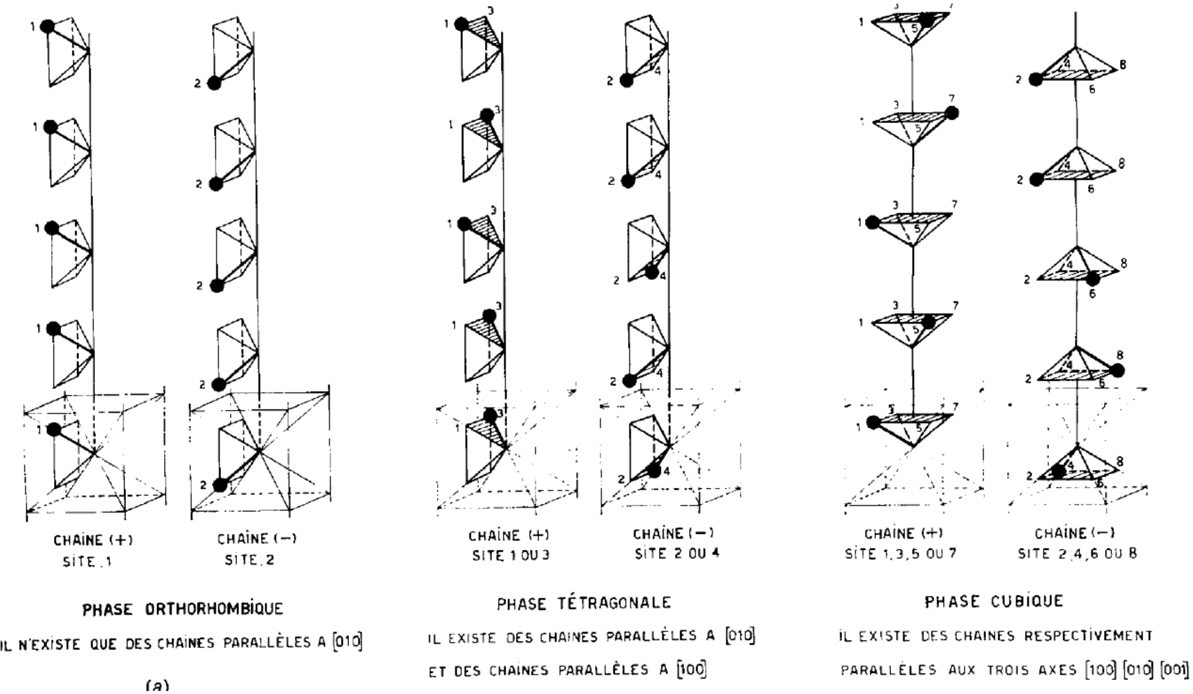


Fig. 5. La structure pérovskite idéale.



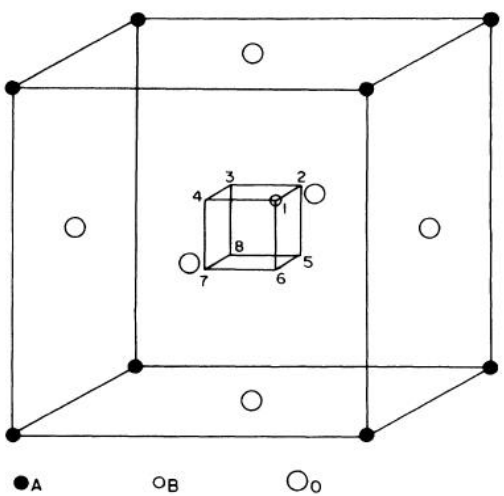
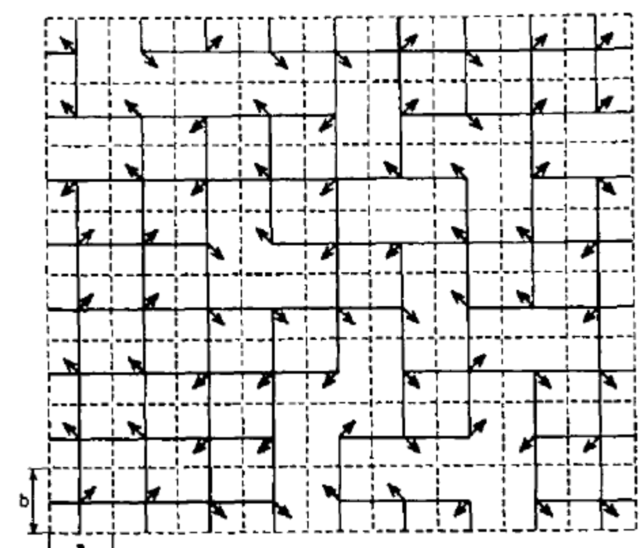


Fig. 10. Le croisement des chaines de corrélation en projection sur un plan (001) dans la phase tétragonale. Les flèches schématisent le déplacement de l'atome central projeté sur le plan (001); les mailles appartenant à une même chaine sont reliées en trait plein. En phase tétragonale la composante du déplacement perpendiculairement au plan de figure est constante; en phase cubique au contraire il existerait un système de chaines analogue suivant [001].



Diffuse Neutron Scattering

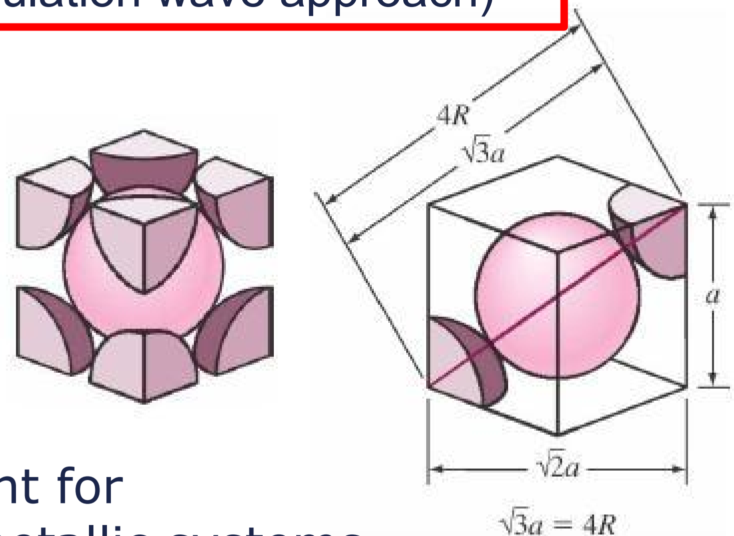
In general: Size of distortion ~L

Spread in Δq~2ϖ/L

Try to minimize parameter space

Short range features, spread over large Q Work in real space (correlation appoach, microdomain approach)	Long range features, spread over small Q Work in reciprocal space (Modulation wave approach)

Example 2: Simple binary alloy on a bcc lattice

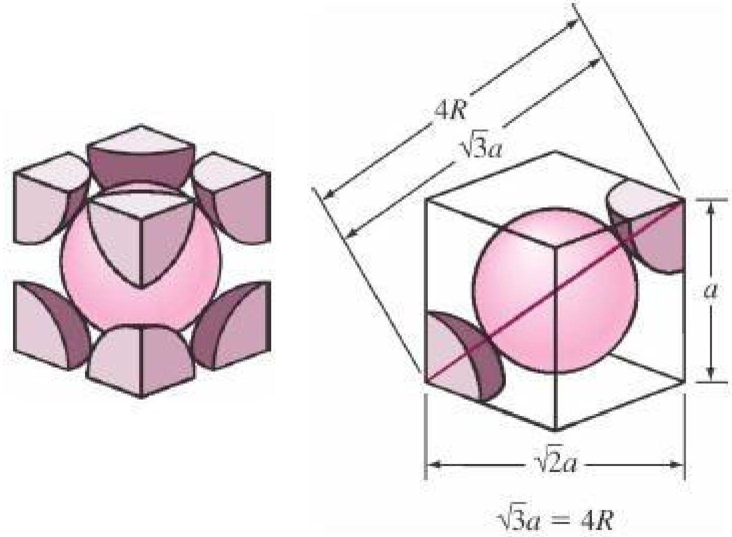


Crystallogaphic sice can be occuied by
- two specific atoms
- half occupacy
- random occupany

Relevant for
- metallic systems
- semiconducting alloys
- hydrogen in metals
- non-stoichiometric alloys

Diffuse Neutron Scattering

Example 2: Simple binary alloy on a bcc lattice

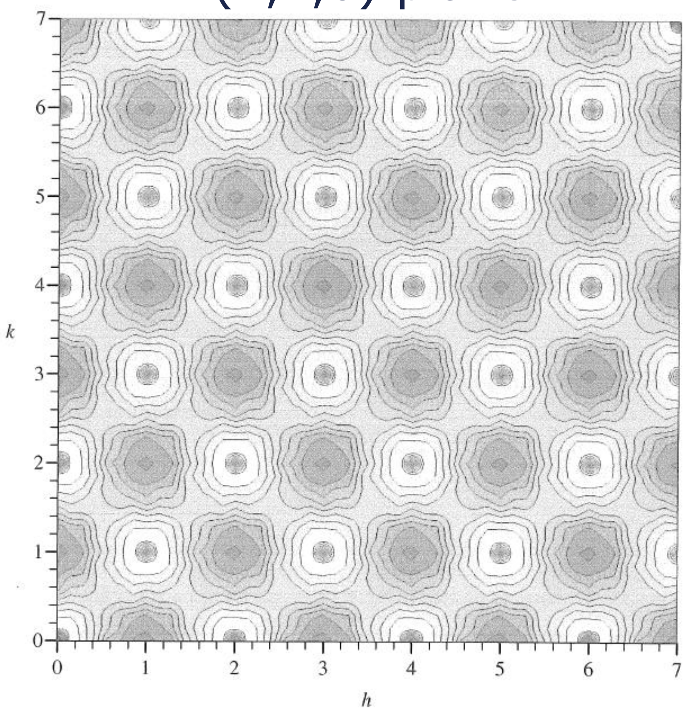


Simple rule:

- Similar atoms with scattering length b
- No imaginary part
- No variation of b
- Exactly one atom per unit call occupied, however, randomly distributed

„Random" distribution, flat in Q?

(h,k,o) plane



Diffuse scattering

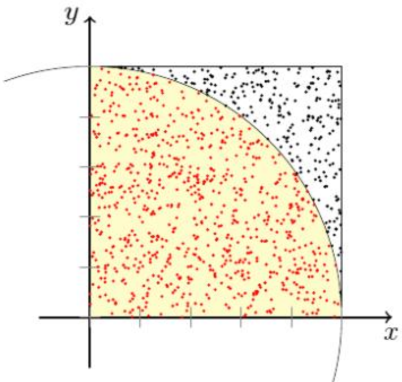
$$D^2 = \left| \sum_u s_u ib \sin(\mathbf{Q} \cdot \mathbf{d}) \exp(i\mathbf{Q} \cdot \mathbf{R}_u) \right|^2$$

+ Bragg scattering

$$\left(\frac{d\sigma}{d\Omega}\right)_{\text{coh}} = b^2 \sum_{j,j'} \exp(i\mathbf{Q} \cdot (\mathbf{R}_j - \mathbf{R}_{j'}))$$

Diffuse Neutron Scattering

Powerful tool for modeling diffuse scattering



Monte Carlo simulations (MC)

1) Choose appropriate descriptions of potentials

2) Generate arrangement of N atoms including boundary conditions

3) Calculate system energy

4) Randomly move atom(s)

5) Recalculate energy

6) Refine energy of the system with Monte Carlo method

⇨ Working from the potential to the structure

Reverse Monte Carlo simulations (RMC)

1) Generate arrangement of N atoms including boundary conditions and hard core potential to avoid overlap

2) Calculate F(Q) from arrangements

3) Randomly move atom(s)

4) Recalculate scattering signal F(Q)

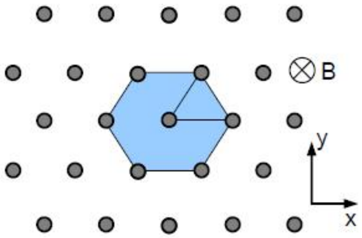
5) Refine F(Q) of the system with Monte Carlo method

⇨ Working from the scattering signal to the structure

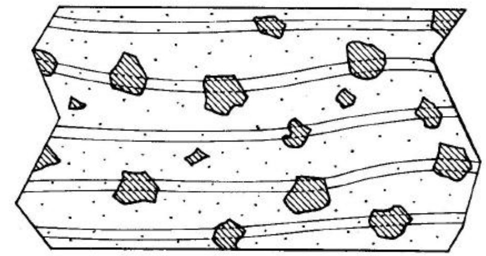
Diffuse Neutron Scattering

Application of RMC: Vortex lattice in the presence of weak pinning

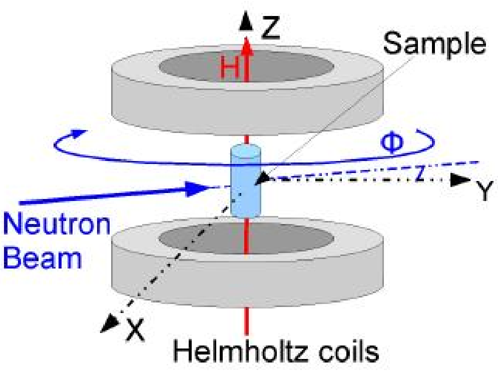
Ideal six-fold VL symmetry
Ginzbung Landau theory

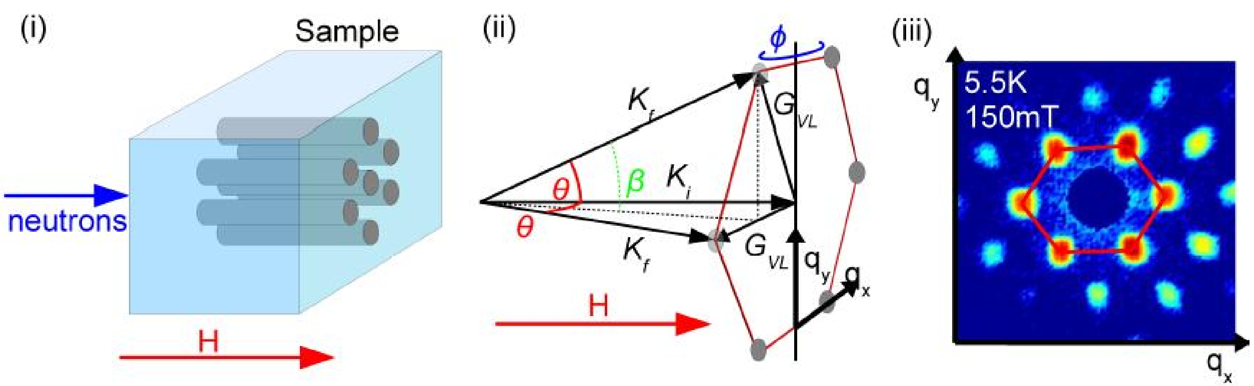


Pinning
Local distortion of the VL



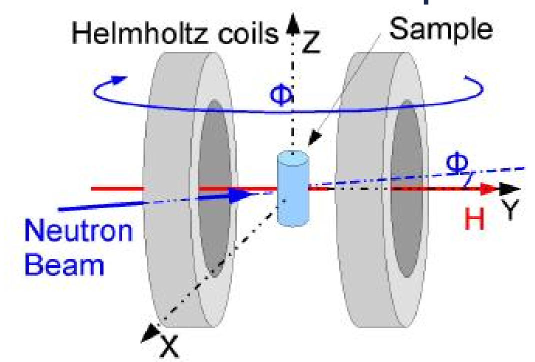
High resolution setup





Rocking scan:
Measurements of the “crystalline properies“ of the VL

Standard setup



Application of RMC: Vortex lattice in the presence of weak pinning

Uncovering Flux Line Correlations in Superconductors by Reverse Monte Carlo Refinement of Neutron Scattering Data

M. Laver,[1,2,3,4] E. M. Forgan,[2] A. B. Abrahamsen,[5] C. Bowell,[2] Th. Geue,[6] and R. Cubitt[3]

[1]*NIST Center for Neutron Research, Gaithersburg, Maryland 20899, USA*

[2]*School of Physics and Astronomy, The University of Birmingham, Birmingham B15 2TT, United Kingdom*

[3]*Institut Laue-Langevin, BP 156, F-38042 Grenoble, France*

[4]*University of Maryland, College Park, Maryland 20742, USA*

[5]*Risø National Laboratory, DTU, Frederiksborgvej 399, DK-4000 Roskilde, Denmark*

[6]*Paul Scherrer Institut, Villigen PSI, CH 5232, Switzerland*

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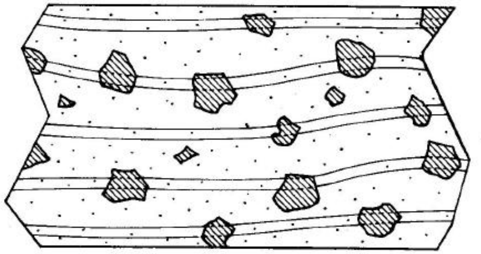
We describe the use of reverse Monte Carlo refinement to extract structural information from angle-resolved data of a Bragg peak. Starting with small-angle neutron scattering data, the positional order of an ensemble of flux lines in superconducting Nb is revealed. We discuss the uncovered correlation functions in the light of topical theories, in particular, the "Bragg glass" paradigm.

200x200 (N=40105) vortex system with RMC

Steps <0.001 vortex distance

Convergence after ~2000N steps

Pinning: Local distortion of the VL



Random distortions

Break-up in VL factures (domains)

Bragg-glass?

Diffuse Neutron Scattering

Application of RMC: Vortex lattice in the presence of weak pinning