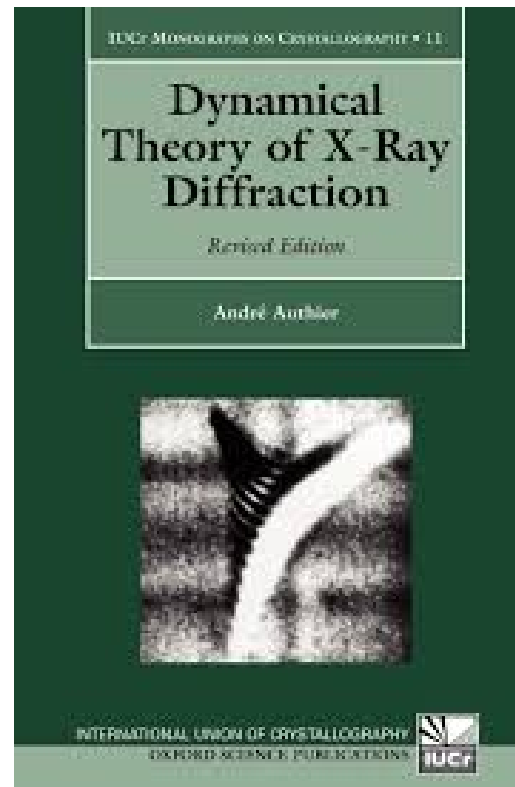
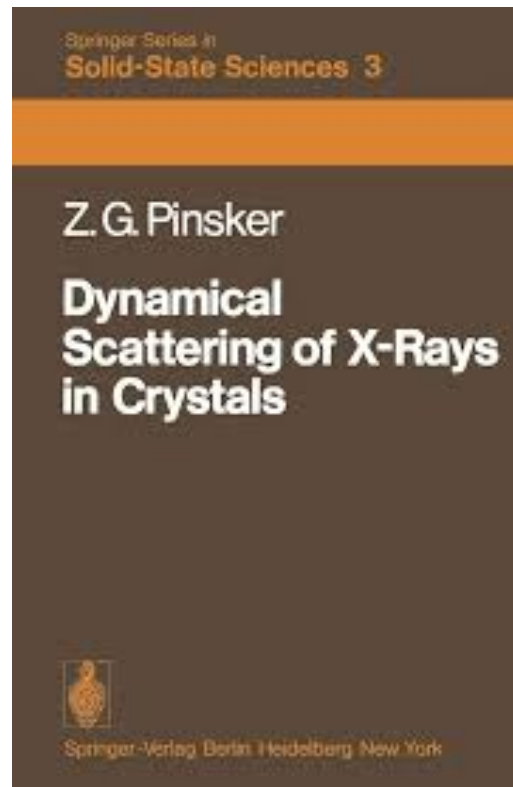
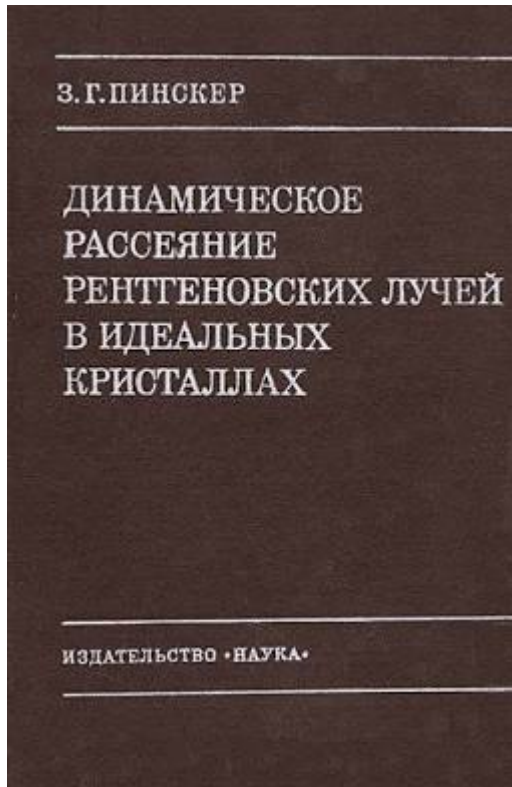


DYNAMICAL DIFFRACTION OF X-RAYS

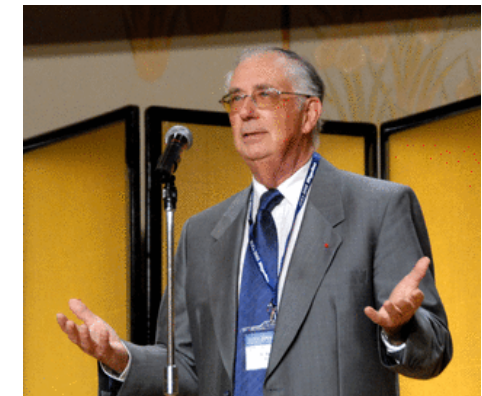
quick-and-dirty theory essentials for (maybe reluctant) beginners

Václav Holý

Department of Condensed Matter Physics
Charles University Prague



André Authier (1932-2023)



A. Authier, *Dynamical Theory of X-Ray Diffraction*, Oxford Univ. Press, Oxford 2001, ISBN-10: 0198559607

Everything begins from the Maxwell equations. In a monochromatic case (all quantities are $\sim e^{-i\omega t}$) we obtain the wave equation for the $\mathbf{E}$ -wave in a crystal

$$(\Delta + K^2)\mathbf{E} = \nabla(\nabla \cdot \mathbf{E}) - K^2\chi(\mathbf{r})\mathbf{E} \equiv \hat{V}\mathbf{E}, \quad K = 2\pi/\lambda$$

The crystal polarizability (dielectric susceptibility) is proportional to the **total** electron density $\rho(\mathbf{r})$: $\chi(\mathbf{r}) = -\frac{\lambda^2}{\pi} r_{el} \rho(\mathbf{r})$

How to solve the wave equation?

1. Successive iterations: $\mathbf{E}^{(n)}(\mathbf{r}) = \mathbf{E}^{(0)}(\mathbf{r}) + \int d^3\mathbf{r}' G_0(\mathbf{r} - \mathbf{r}') \hat{V}(\mathbf{r}') \mathbf{E}^{(n-1)}(\mathbf{r}')$, $(\Delta + K^2)E^{(0)} = 0$, $(\Delta + K^2)G_0(\mathbf{r} - \mathbf{r}') = \delta(\mathbf{r} - \mathbf{r}')$

The first iteration $\mathbf{E}^{(1)}(\mathbf{r})$ is the kinematical approximation. The iterations converge very slowly and the higher-order formulas are too complicated ☹

2. Bloch waves:

If the crystal is perfectly periodic (it means infinite !!!!), the solution of the wave equation is the Bloch wave

$$\mathbf{E}(\mathbf{r}) = \underbrace{e^{ik_0 \cdot \mathbf{r}}}_{\text{phase factor}} \underbrace{\sum_{\mathbf{g}} \mathbf{E}_{\mathbf{g}} e^{i\mathbf{g} \cdot \mathbf{r}}}_{\text{periodic function}} \equiv \sum_{\mathbf{g}} \mathbf{E}_{\mathbf{g}} e^{i\mathbf{k}_{\mathbf{g}} \cdot \mathbf{r}}, \quad \mathbf{k}_{\mathbf{g}} = \mathbf{k}_0 + \mathbf{g}, \quad \mathbf{k}_0 \in 1BZ$$

and the polarizability is expressed as a Fourier series $\chi(\mathbf{r}) = \sum_{\mathbf{g}} \chi_{\mathbf{g}} e^{i\mathbf{g} \cdot \mathbf{r}}, \quad \chi_{\mathbf{g}} = -\frac{\lambda^2 r_{el}}{\pi V_c} \underbrace{F(\mathbf{g})}_{\text{g-th structure factor}}$

Amplitude equations:

$$K^2 \mathbf{E}_{\mathbf{g}} - \mathbf{k}_{\mathbf{g}} \times (\mathbf{E}_{\mathbf{g}} \times \mathbf{k}_{\mathbf{g}}) = -K^2 \sum_{\mathbf{p}} \chi_{\mathbf{p}} \mathbf{E}_{\mathbf{g}-\mathbf{p}}$$

That's all. All other formulas are just mere technicalities

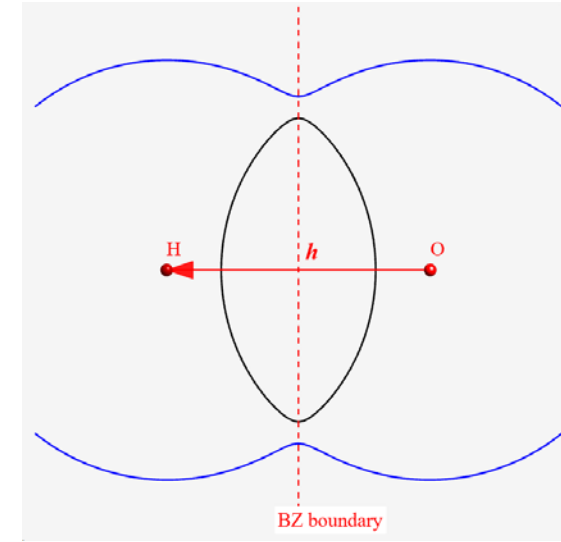
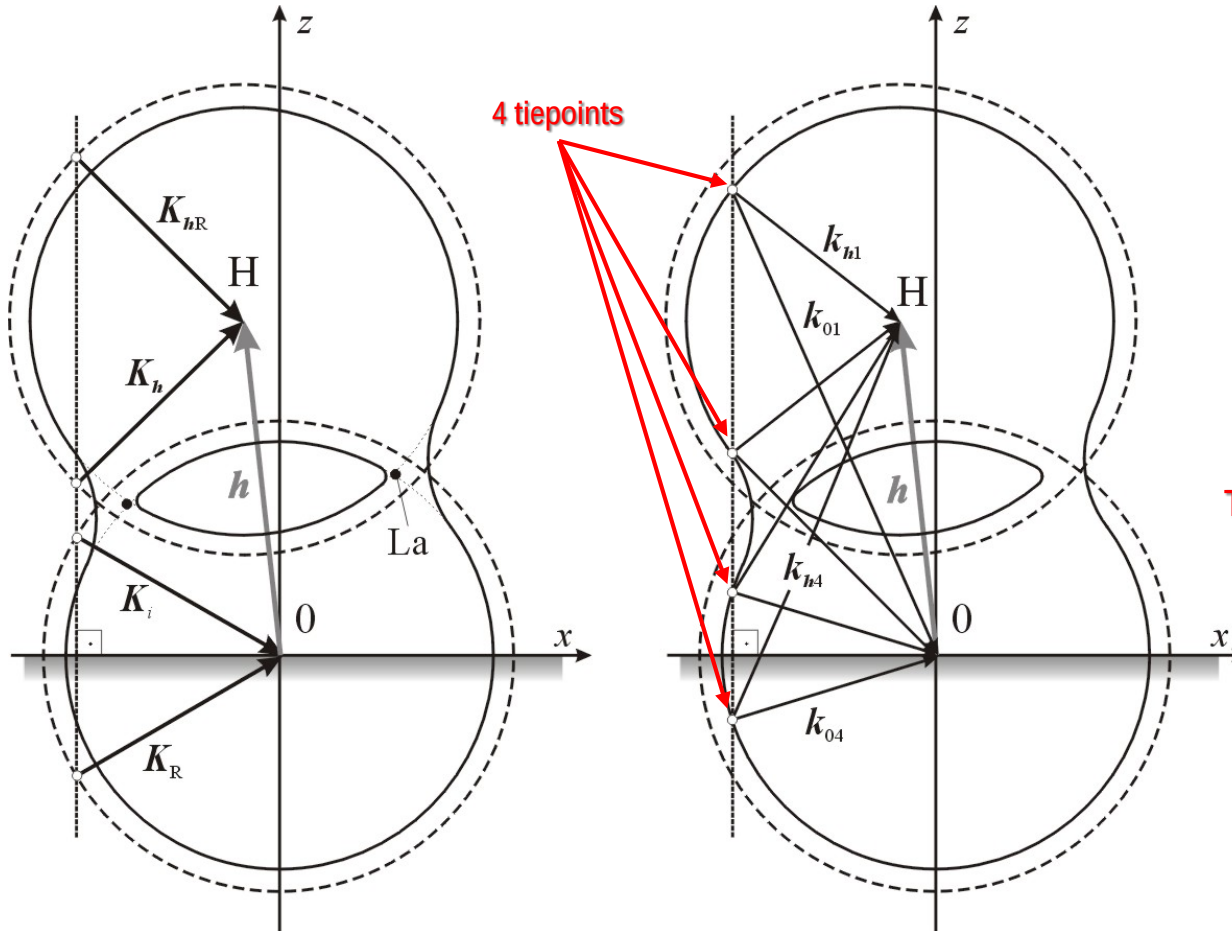
Usually two components of the Bloch wave are considered $\mathbf{g} = 0, \mathbf{h} \Rightarrow$ **two-beam case**

Boundary conditions:

This is the weak point of the Laue theory. Tangential components of the wave vectors and of the polarization vectors $\mathbf{E}$ are **continuous at the interface**

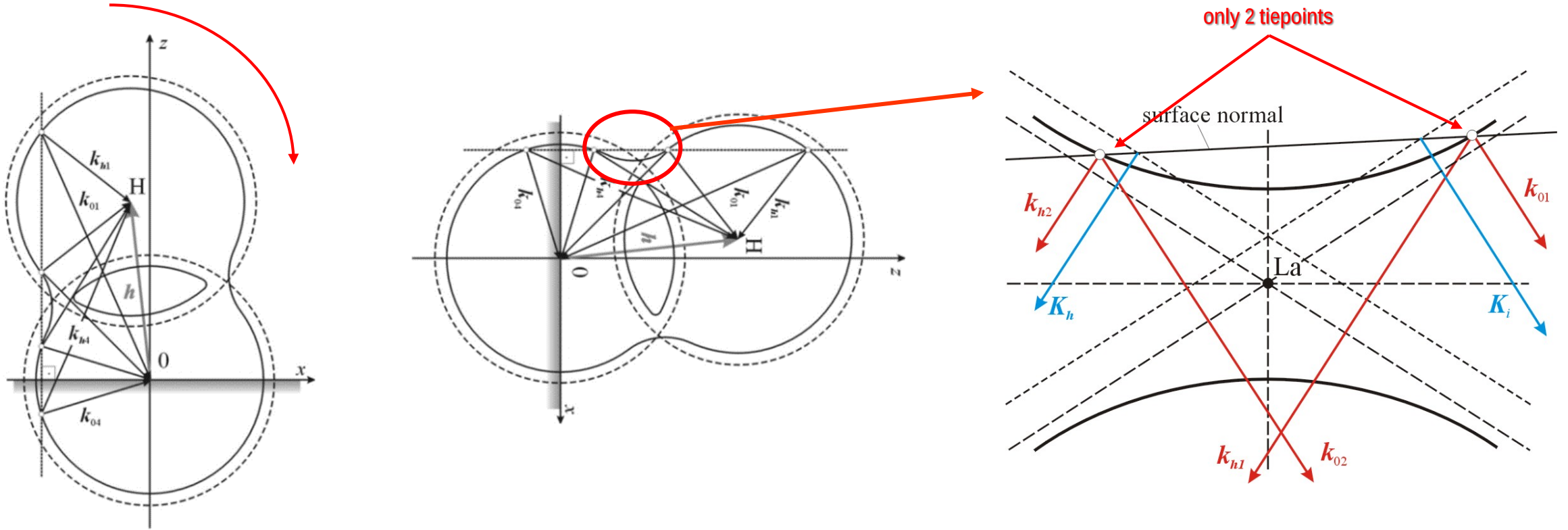
The amplitude equations build a set of homogeneous linear equations for $\mathbf{E}_g$. The condition of the existence of a non-trivial solution is the zero of the determinant $\text{Det}(\mathbf{k}_0, K, \vec{\chi}) = 0$, which defines the dispersion surface. In the two-beam case the dispersion surface looks like this

With the boundary conditions (continuity of the tangential components of the wave vectors):



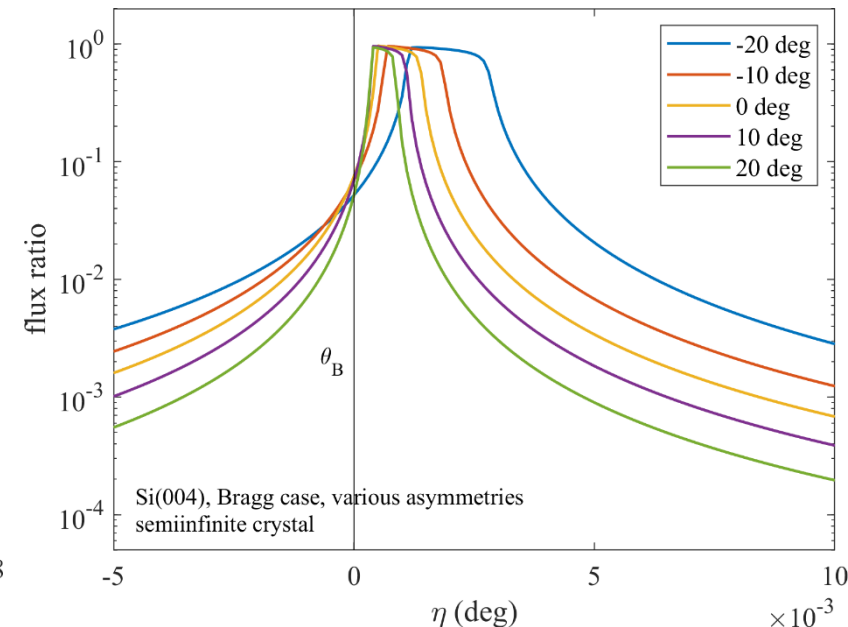
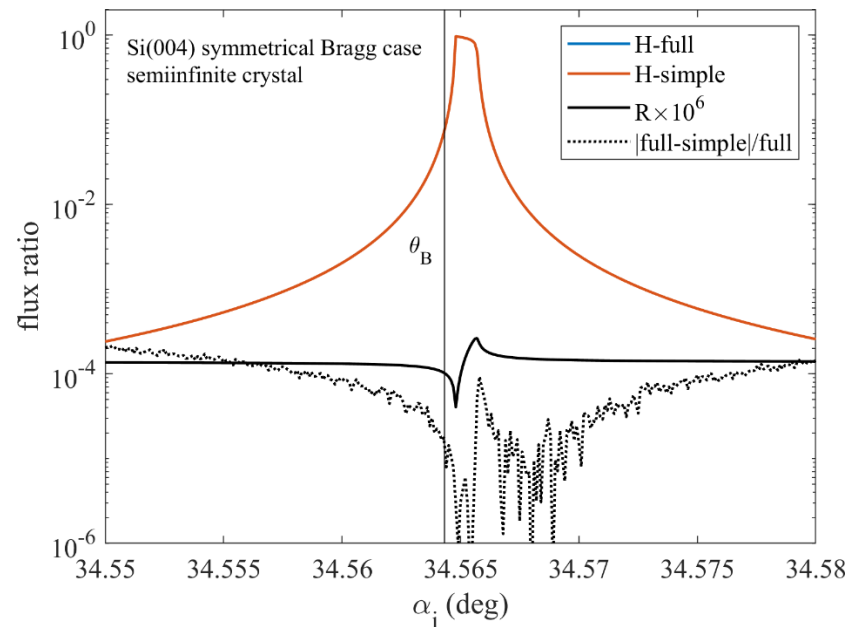
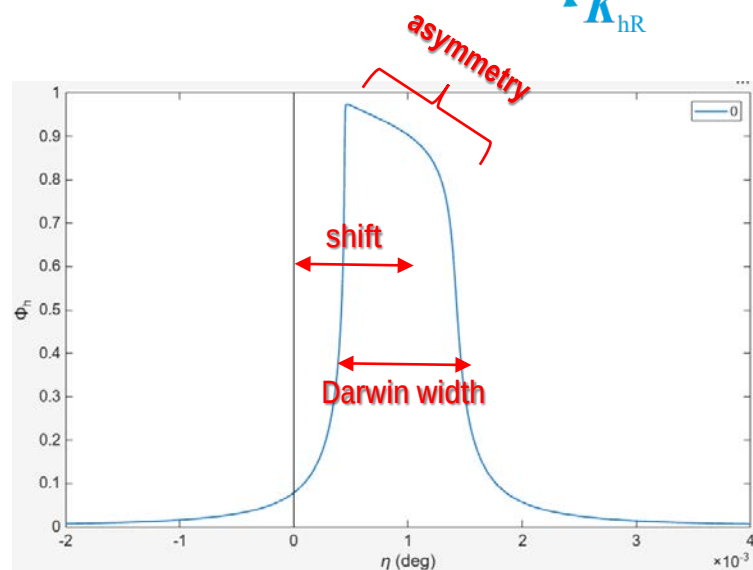
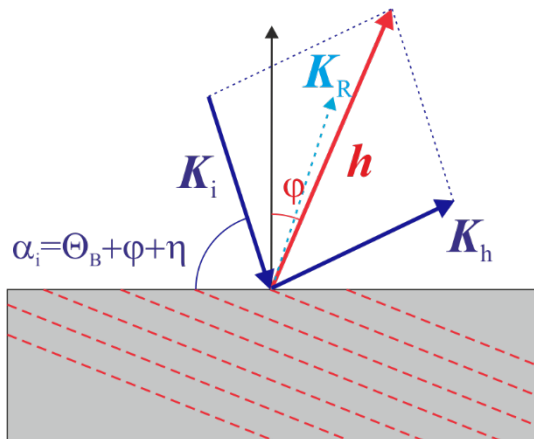
The dispersion surface is a surface of 4th order

Usually, the problem is simplified replacing the exact two-beam dispersion surface by a surface of 2nd order – hyperbolic cylinder



This approximation is valid only in a close vicinity of the Brillouin-zone boundary, i.e. in a small region around the diffraction maximum. This approximation does not describe the reflected beams and it fails also for very small incidence and/or exit angles.

Example 1: single semiinfinite crystal, coplanar reflection geometry

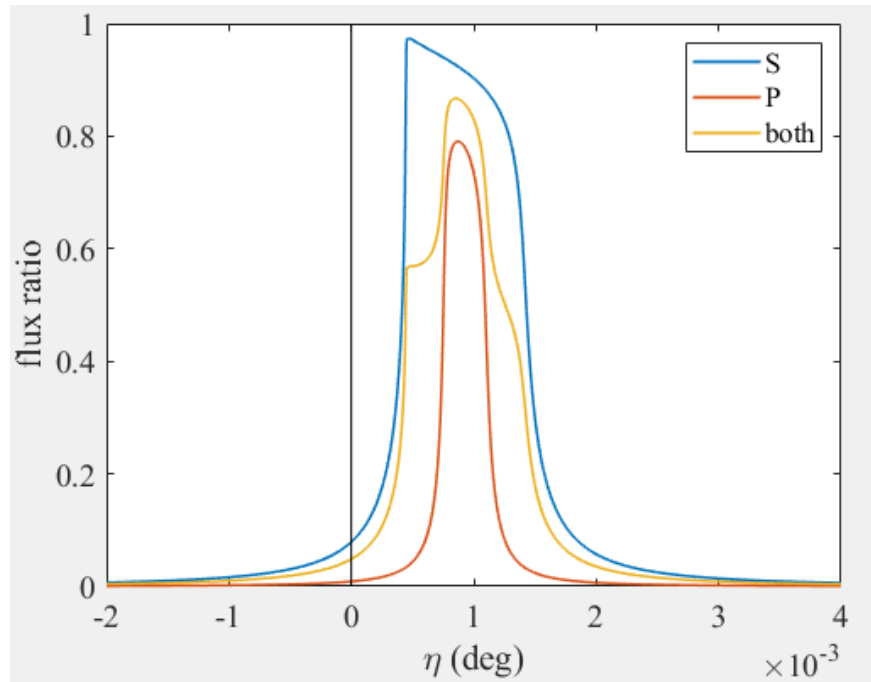


Shift from the kinematical Bragg position (due to refraction): $\eta_{\max} = -\frac{\text{Re}(\chi_0)}{2 \sin(2\Theta_B)} \left(1 - \frac{\gamma_h}{\gamma_0}\right) > 0$

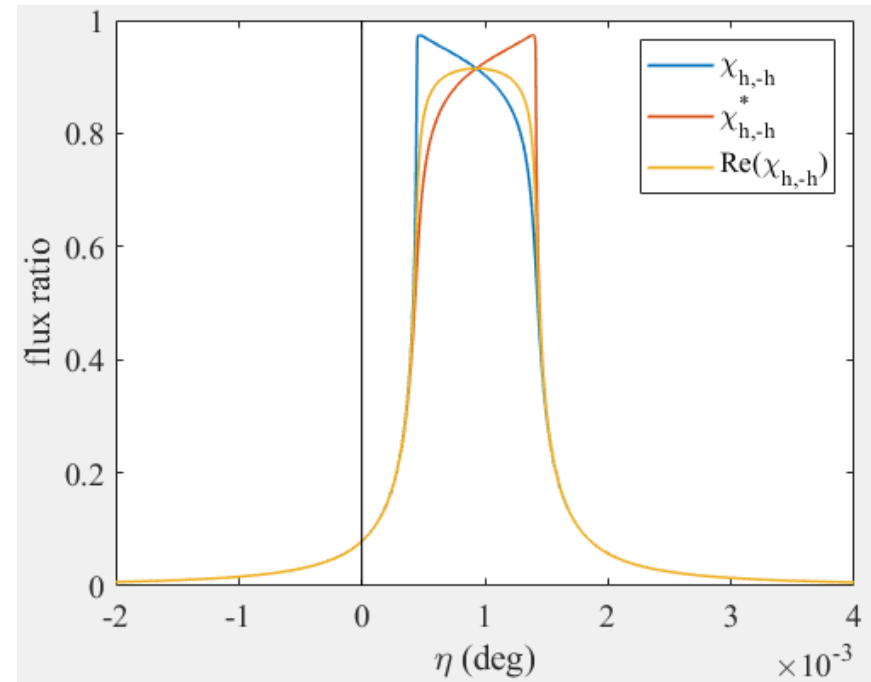
Finite width of the diffraction curve (even if T goes to infinity): $\Delta\eta = \frac{2C\text{Re}(\sqrt{\chi_h\chi_{-h}})}{\sin(2\Theta_B)} \left|\frac{\gamma_h}{\gamma_0}\right|$

dependence of the diffraction asymmetry

Asymmetry of the maximum – due to $\text{Im}(\chi_h)$ – the Borrmann effect

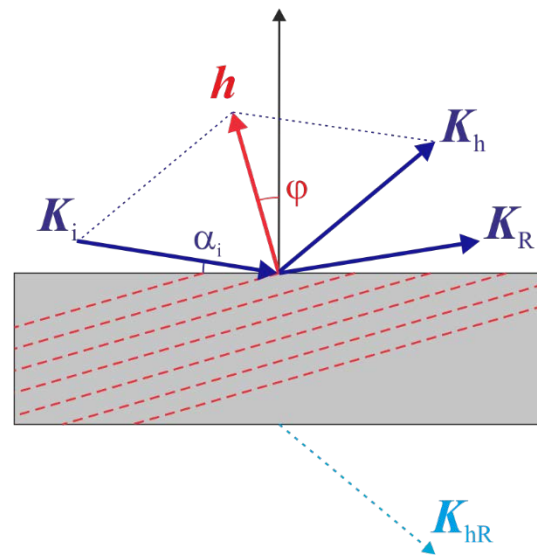


The curve 'both' depicts the simulation of a measurement with a 2x220Ge channel-cut monochromator

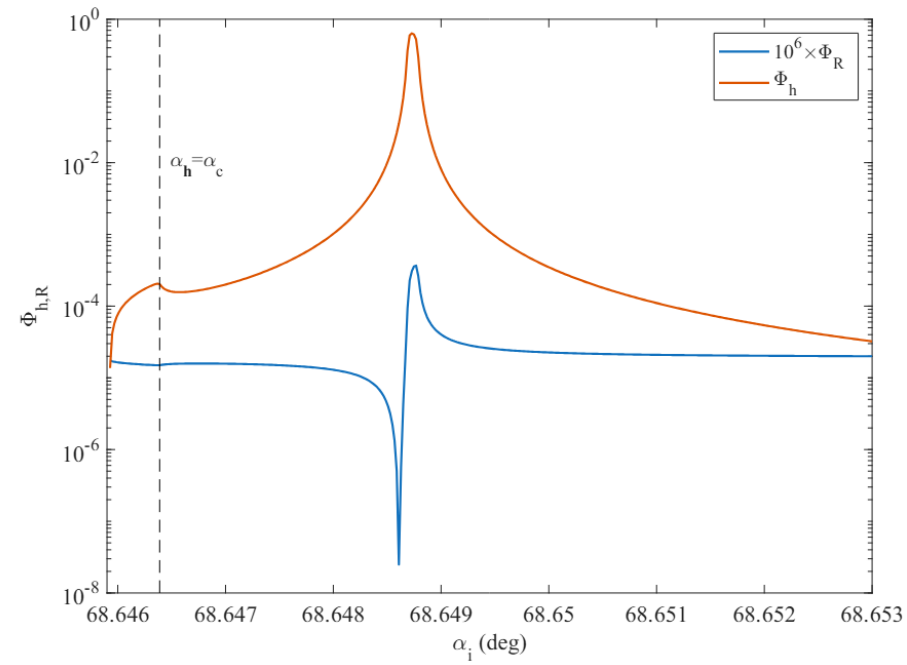
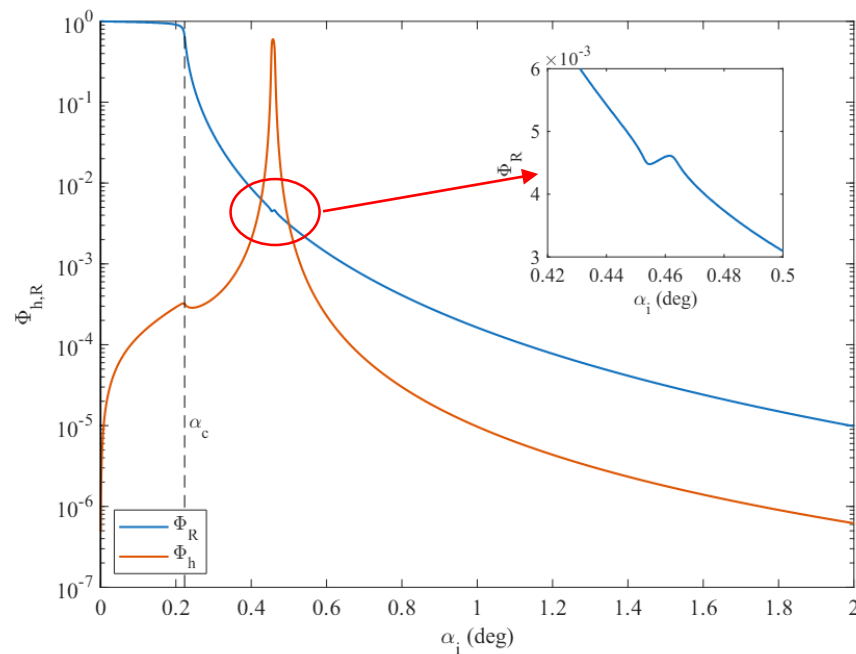


The asymmetry of the curve depends on the sign of the imaginary part of $\chi_{h,-h}$ - illustration of the Borrmann effect

Strongly asymmetric diffraction



The general two-beam diffraction theory must be used (dispersion equation of the 4th order)



Si crystal, diffraction 400, asymmetry angles $\phi = -34^\circ$ (left) and $\phi = 34^\circ$ (right)

Umweganregung effect – extinction of the diffracted beam due to specular reflection

Example 2: single semiinfinite crystal, general scattering geometry, GID

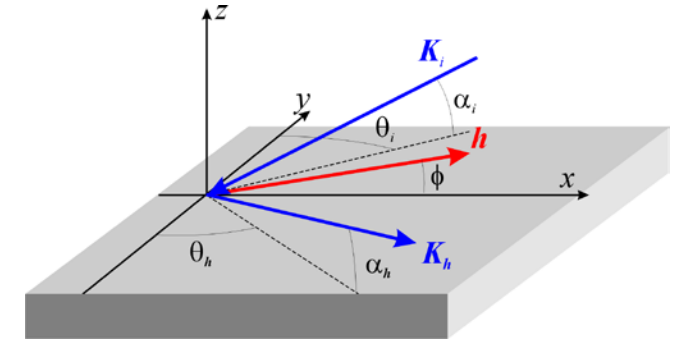
Lateral diffraction condition: $\cos \alpha_h \sin \theta_h + \cos \alpha_i \sin \theta_i = 2 \sin \theta_B \cos \varphi = 2 \sin \theta_{B\parallel}, \cos \alpha_h \cos \theta_h = \cos \alpha_i \cos \theta_i$

The collimating effect of the GID geometry: $\Delta \alpha_h = \frac{\Delta \theta_i}{\alpha_h} \sin(2\theta_{B\parallel})$

Thus a small azimuthal divergence of the primary beam results in a large vertical divergence of the diffracted beam

The penetration depth of the primary beam can be tuned by changing α_i

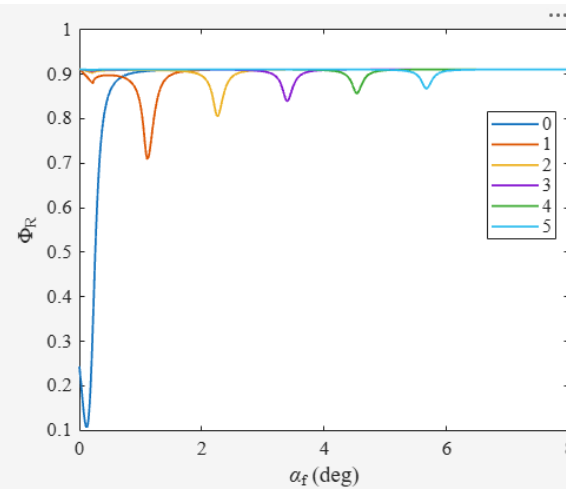
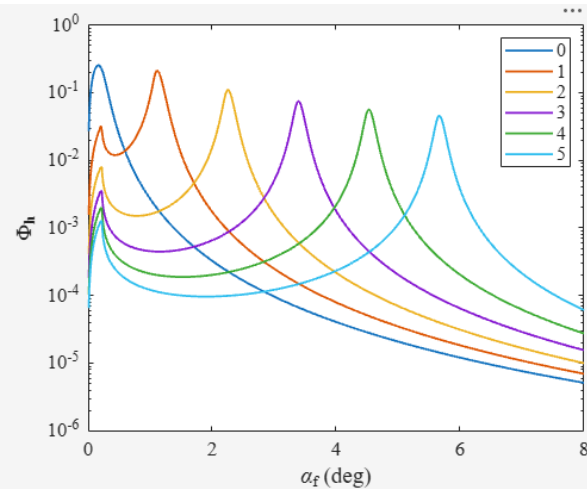
The primary beam must be collimated in both directions $\Rightarrow$ synchrotron radiation is necessary



Various asymmetry angles $\varphi, \alpha_i = 0.2\text{deg}$

diffracted

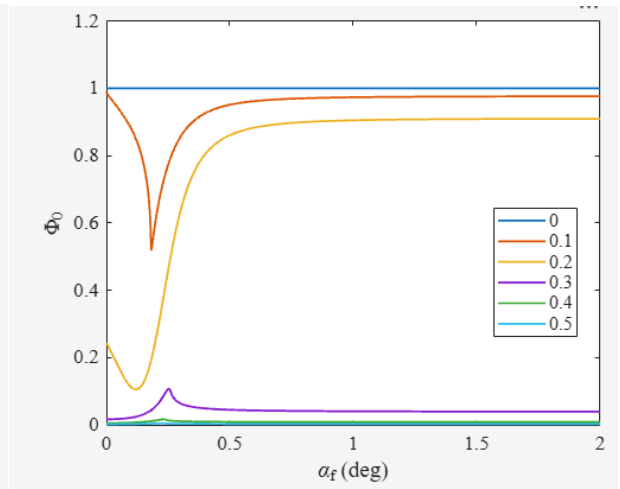
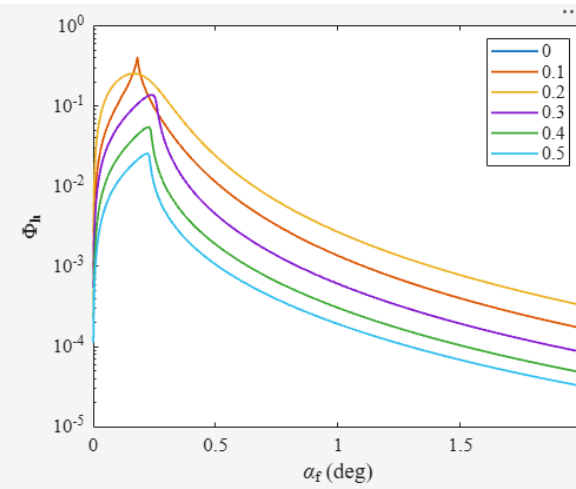
reflected



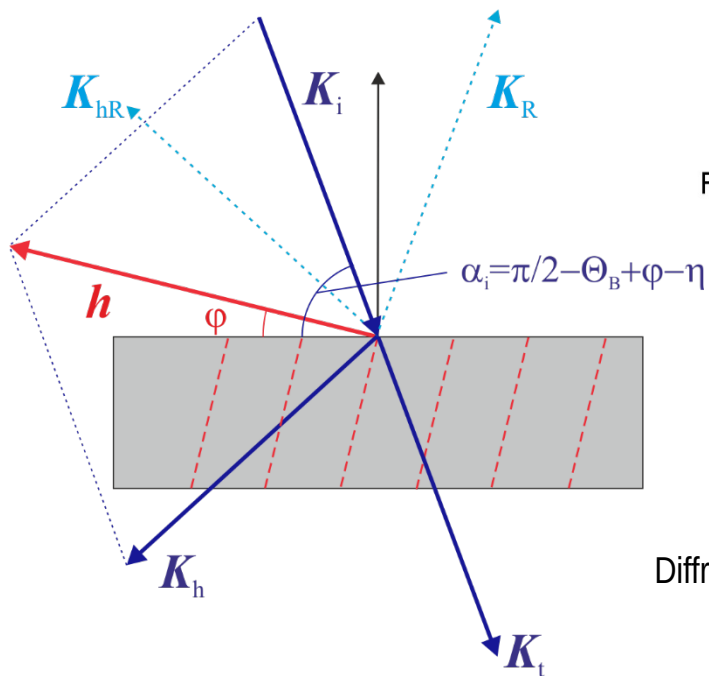
Various incidence angles $\alpha_i, \varphi = 0$

diffracted

reflected

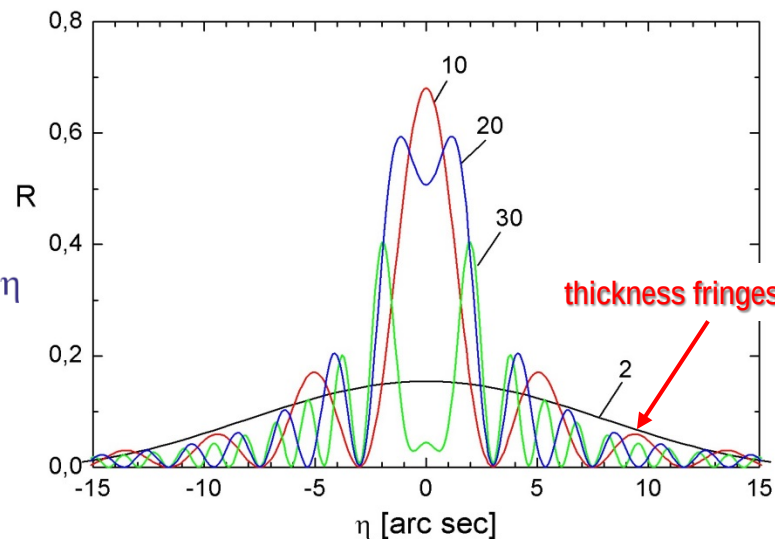


Example 3: single plane-parallel crystal, coplanar transmission geometry

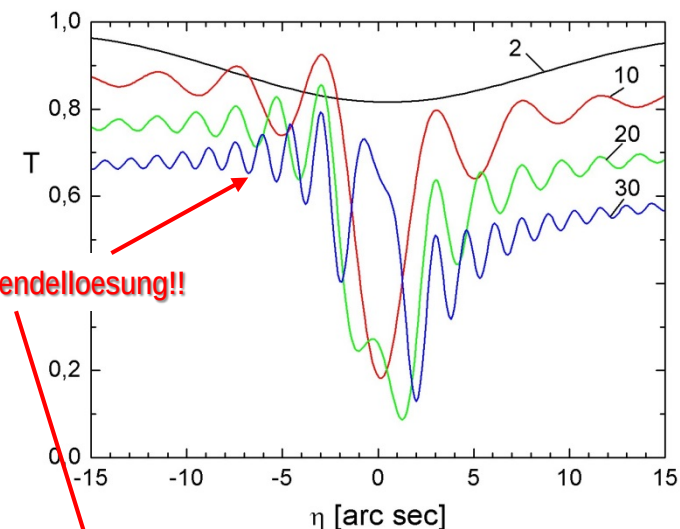


Symmetric Laue case 220 diffraction on Si,
CuK α_1 , various thicknesses in μm

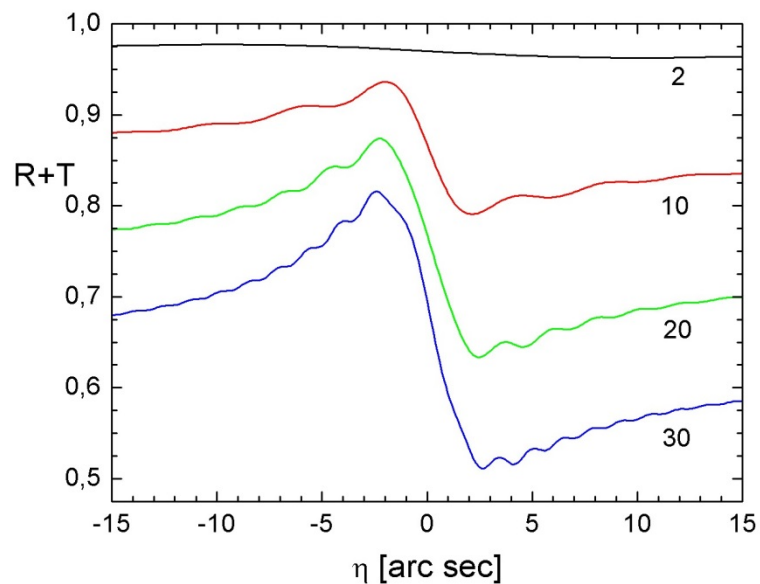
Diffracted flux



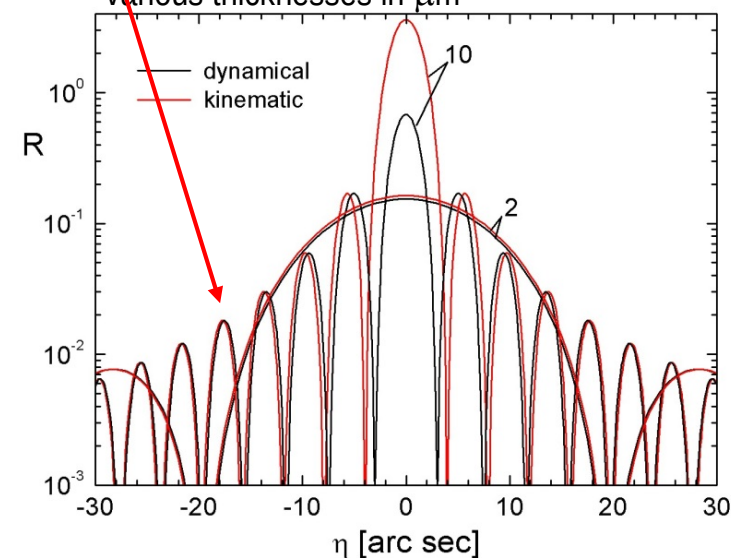
Transmitted flux



Diffracted + transmitted

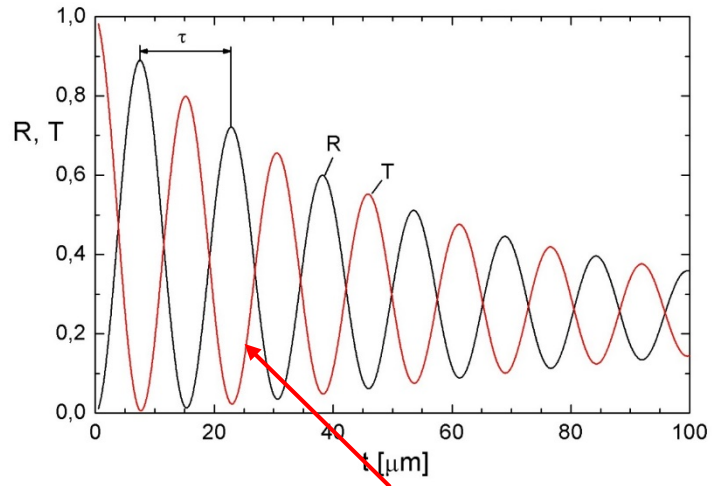


Comparison dynamical vs kinematical simulations,
various thicknesses in μm



The oscillation period is $\sim \lambda/T$

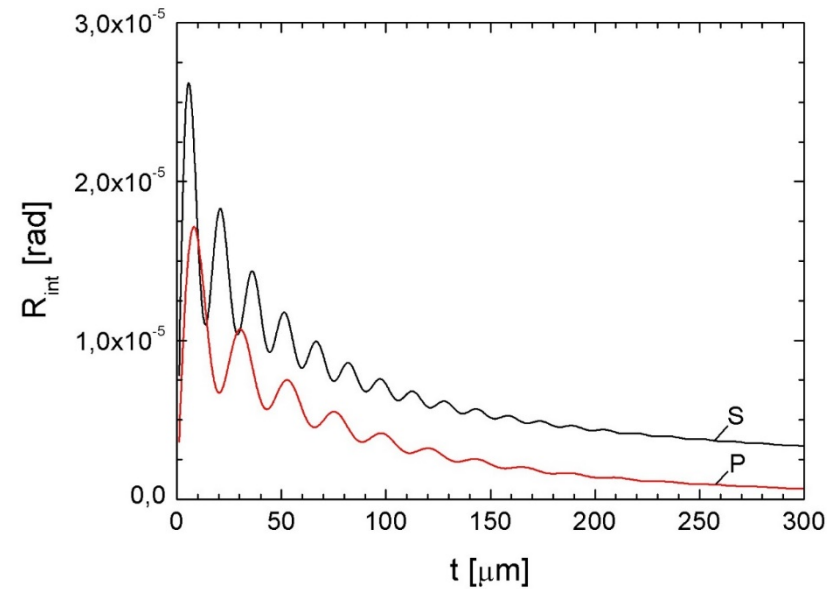
Thickness dependence of the diffracted and transmitted intensities (in the diffraction maximum):



The oscillation period is the extinction length: $\tau = \frac{\lambda}{C} \text{Re} \sqrt{\frac{\gamma_0 \gamma_h}{\chi_h \chi - h}}$ (few μm)

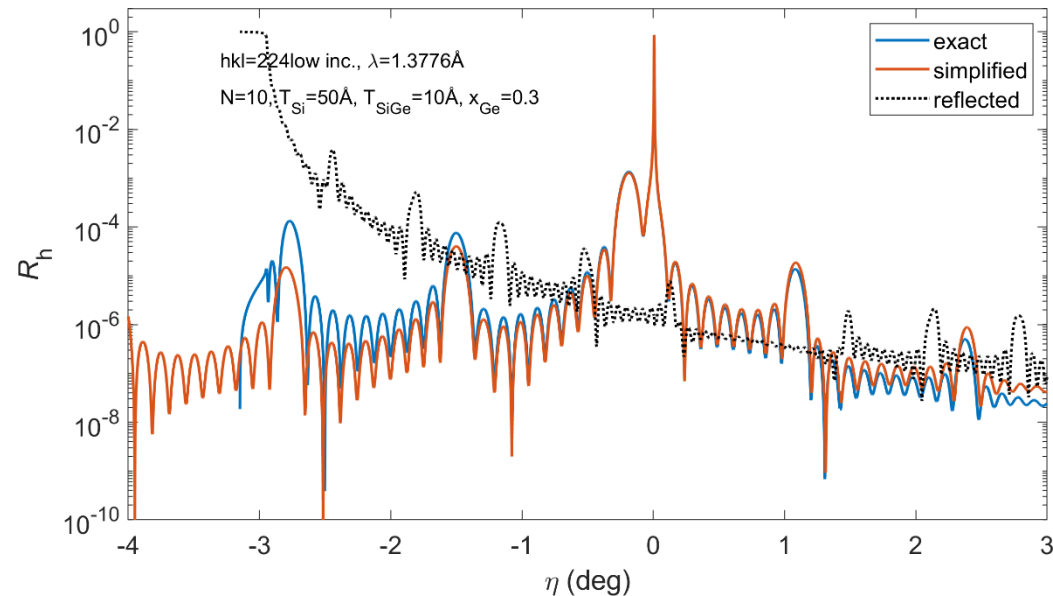
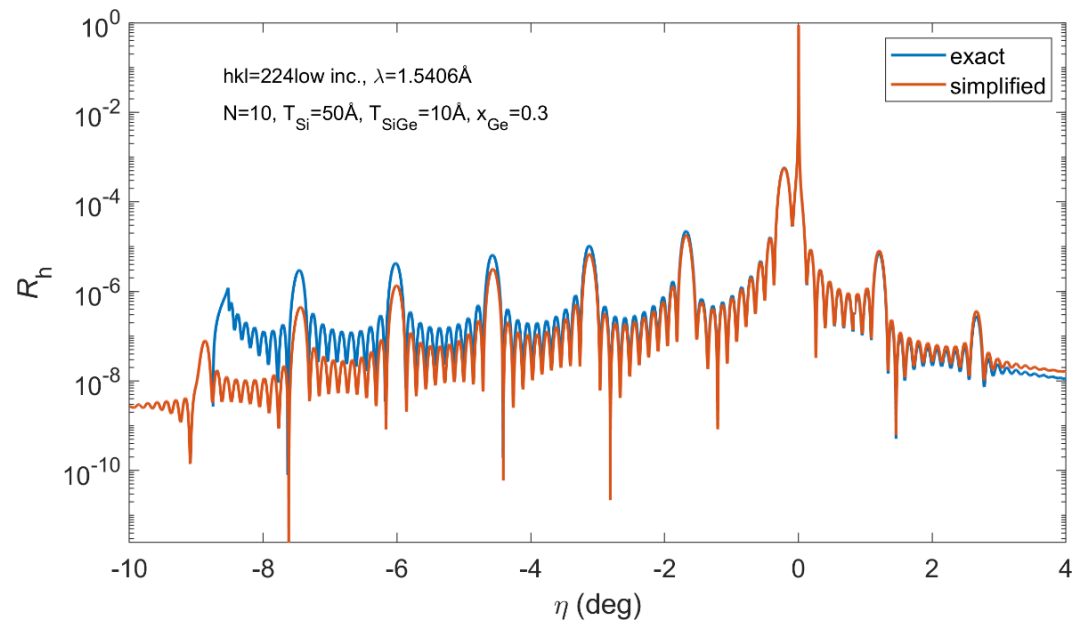
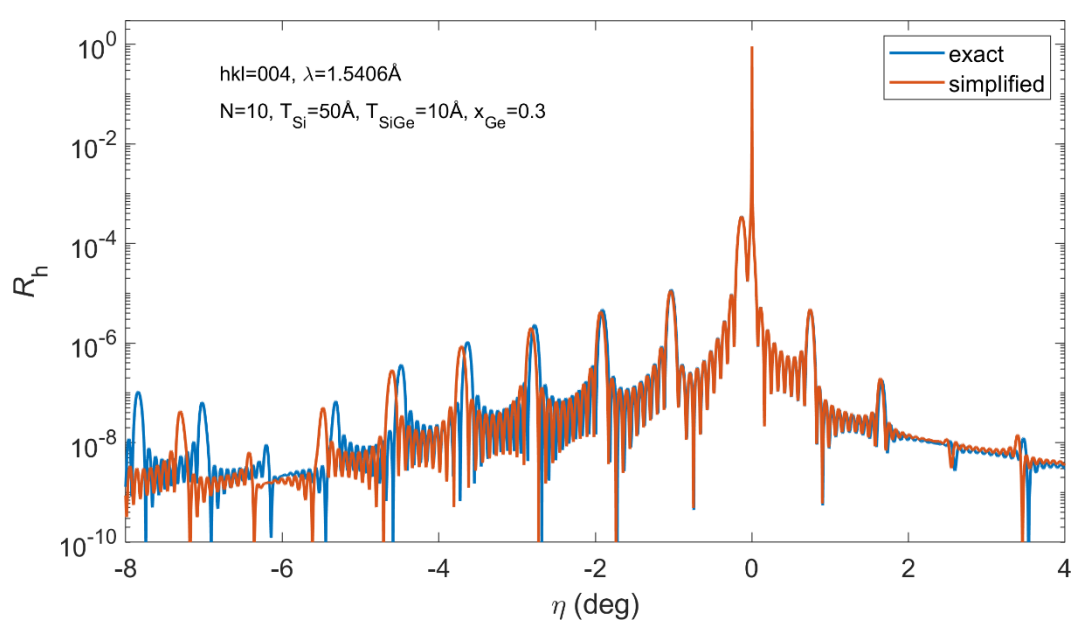
the kinematical approximation is valid for $T \ll \tau$

Integrated intensity: $\mathcal{R}_{int} = \int_{-\infty}^{\infty} d\eta \mathcal{R}(\eta)$

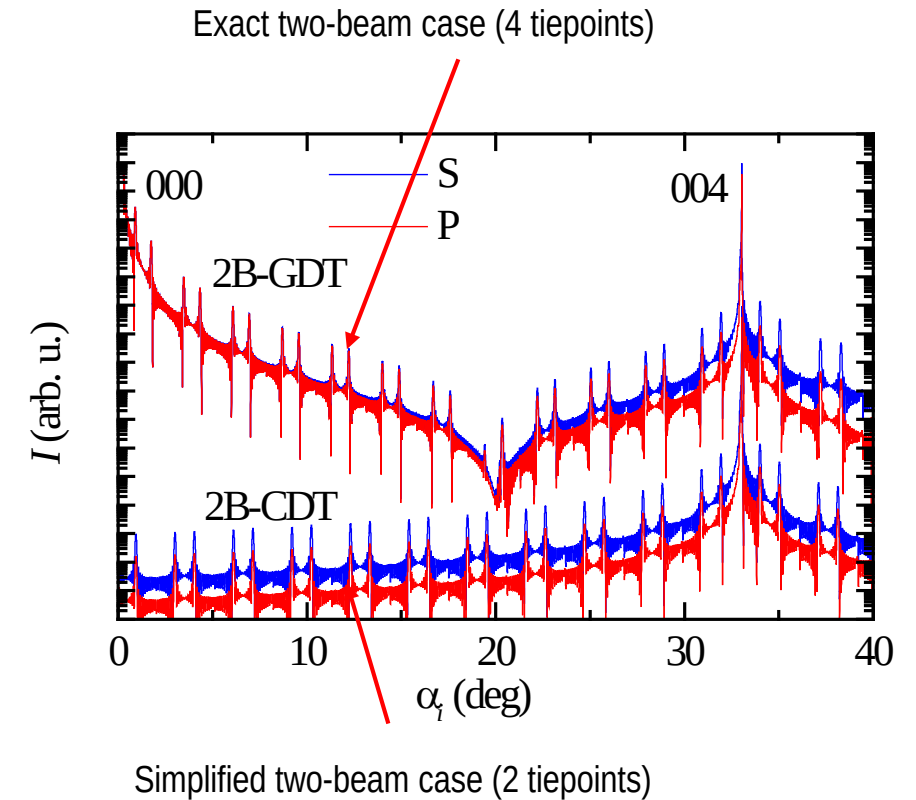
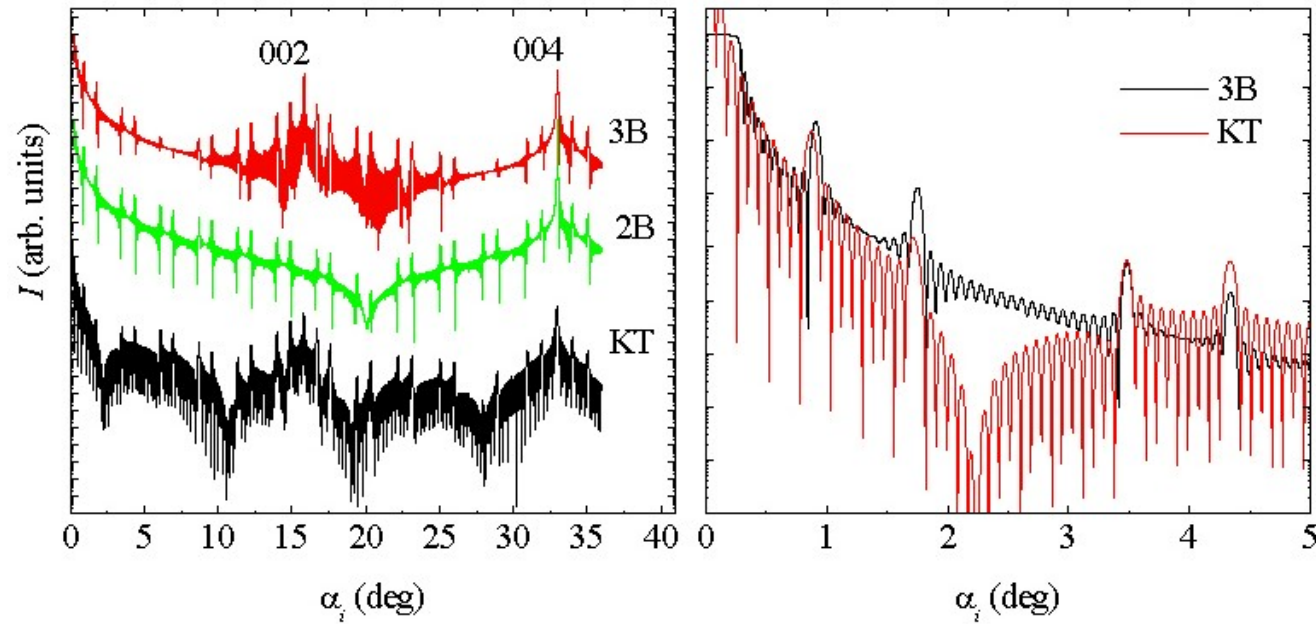


Example 4: Si/Ge superlattice, coplanar reflection geometry

The dynamical theory correctly describes only the pseudomorph case, i.e., no plastic relaxation, no misfit dislocations



How good is the two-beam approximation? Is the three-beam case an improvement?
 Let us consider a pseudomorph GaAs/AlAs superlattice with orientation (001):

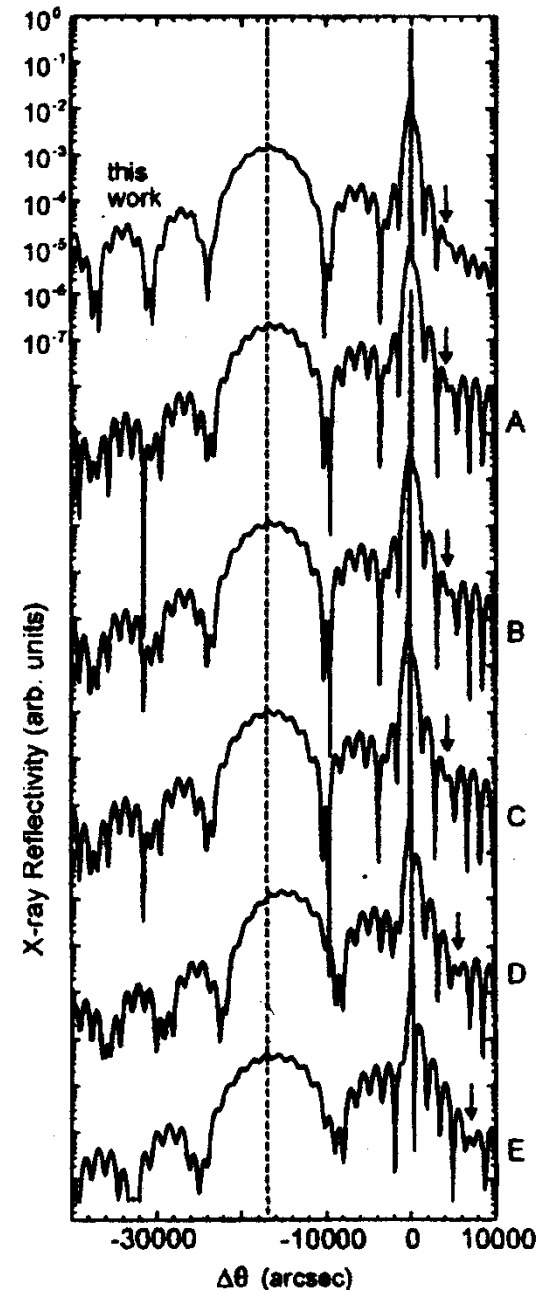
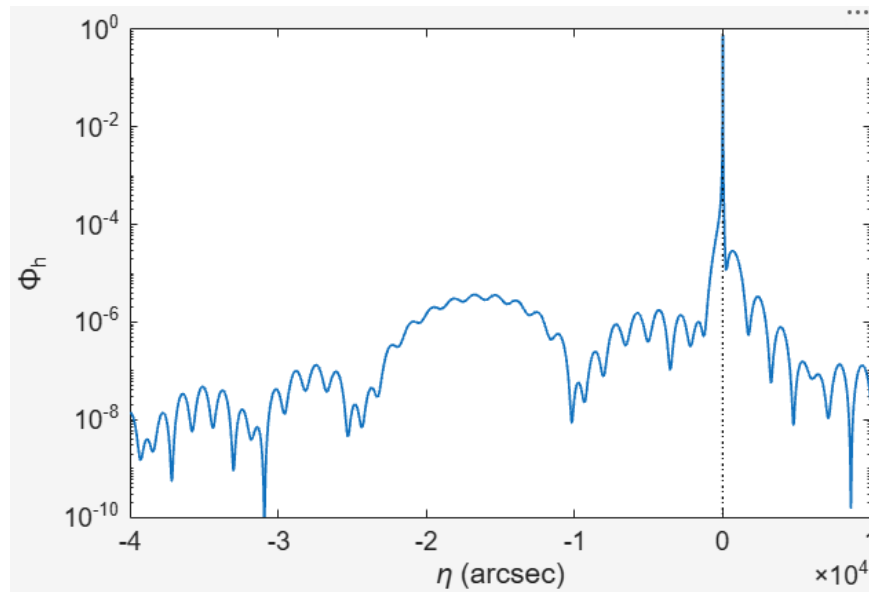


Between the diffraction maxima the 2beam dynamical description fails, since no two strong beam are present in the sample. Instead, simple kinematical approximation (i.e., Fourier transformation of the electron density) is more appropriate

M. Grundmann, A. Krost, phys. stat. sol. (b) 218, 417 (2000)

Fig. 1. Simulations for 8 ML InAs on GaAs (001) capped with 40 ML GaAs with our AKI model and five different commercial programs based on dynamical diffraction theory (DT,A to DT,E) [5]. $\Delta\theta \approx 0$ denotes the substrate (004) Bragg reflection

My result



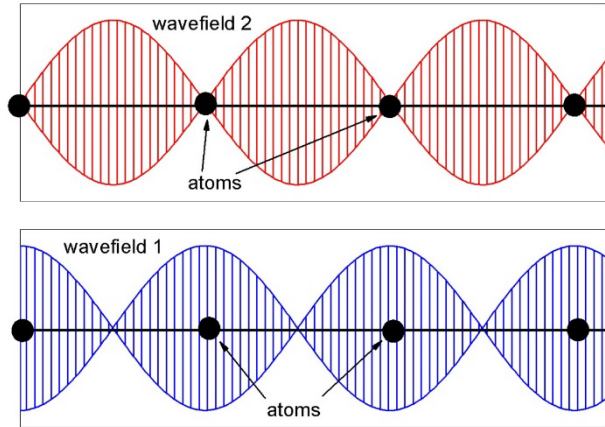
Don't trust the commercial software!!!!

Using two-beam conventional diffraction and S polarization, the wavefield in a diffracting crystal is superposition of four plane waves:

$$E(\mathbf{r}) = E_{01}e^{ik_{01} \cdot \mathbf{r}} + E_{02}e^{ik_{02} \cdot \mathbf{r}} + E_{h1}e^{ik_{h1} \cdot \mathbf{r}} + E_{h2}e^{ik_{h2} \cdot \mathbf{r}}, \mathbf{k}_{hn} = \mathbf{k}_{0n} + \mathbf{h}$$

We can make the superposition **in this way**:

$$E(\mathbf{r}) = (E_{01}e^{ik_{01} \cdot \mathbf{r}} + E_{h1}e^{ik_{h1} \cdot \mathbf{r}}) + (E_{02}e^{ik_{02} \cdot \mathbf{r}} + E_{h2}e^{ik_{h2} \cdot \mathbf{r}}) = E_1(\mathbf{r}) + E_2(\mathbf{r})$$



Therefore, the wavefield #1 is anomalously absorbed and #2 is anomalously transmitted – the Borrmann effect

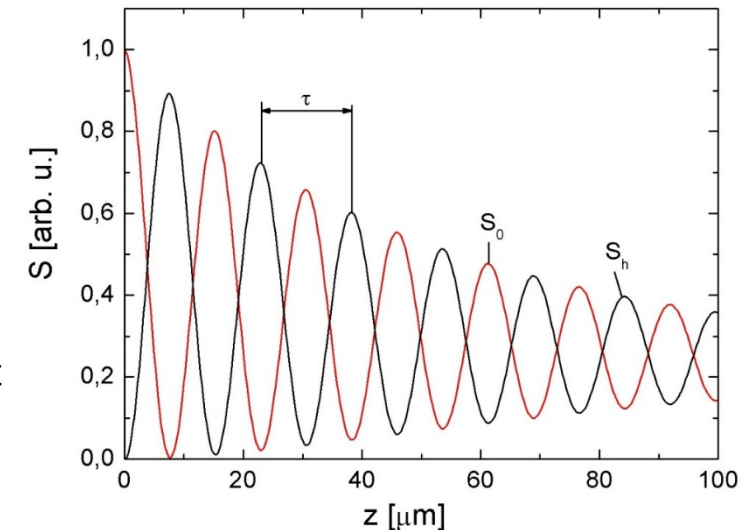


Gerhard Borrmann, 1908-2006

Or in this way: $E(\mathbf{r}) = (E_{01}e^{ik_{01} \cdot \mathbf{r}} + E_{02}e^{ik_{02} \cdot \mathbf{r}}) + (E_{h1}e^{ik_{h1} \cdot \mathbf{r}} + E_{h2}e^{ik_{h2} \cdot \mathbf{r}}) = E_0(\mathbf{r}) + E_h(\mathbf{r})$

The Poynting vector $\mathbf{S}(\mathbf{r}) = \mathbf{S}_0(\mathbf{r}) + \mathbf{S}_h(\mathbf{r})$ $S_0 \propto \left| \cos\left(\frac{\pi z}{\tau}\right) \right|^2$, $S_h \propto \left| \sin\left(\frac{\pi z}{\tau}\right) \right|^2$

where $\tau = \frac{\lambda}{C} \operatorname{Re} \sqrt{\frac{\gamma_0 \gamma_h}{\chi_h \chi - h}}$ is the extinction length (few μm). The consequence is the Pendellösung effect



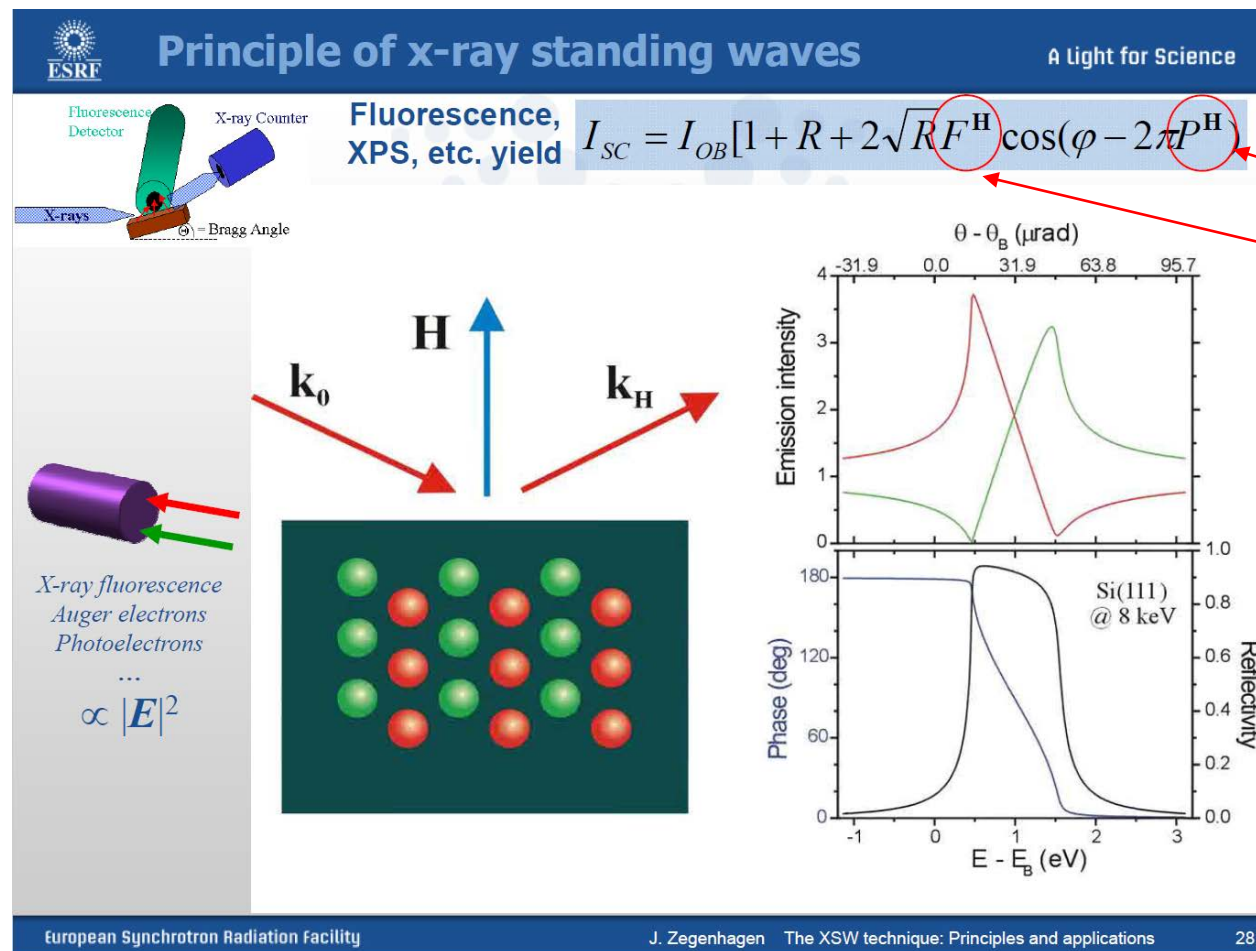
Do we have an x-ray detector inside the material???

Yes, we have ..

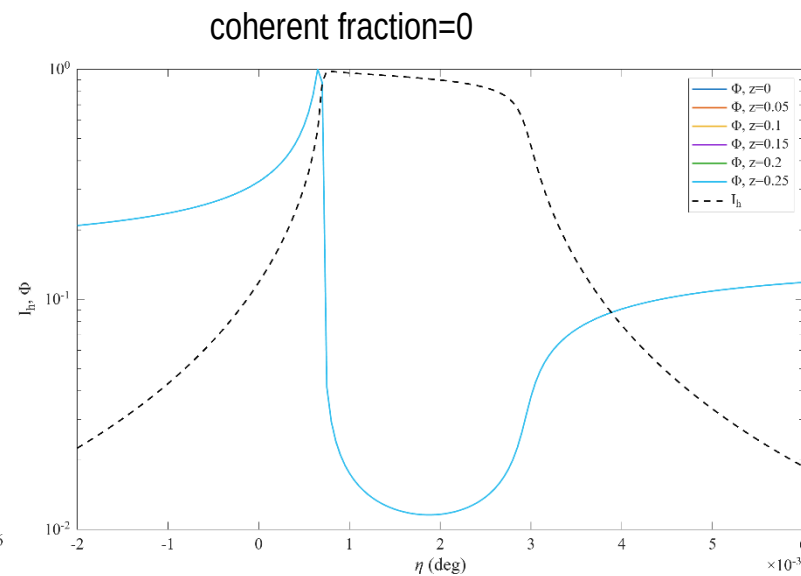
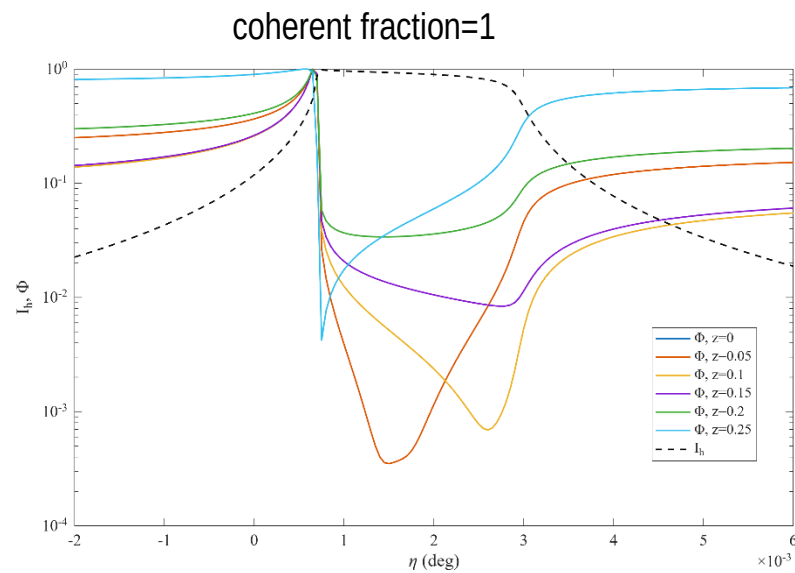
Practical application of these formulas is the **x-ray standing-wave effect (XSW)**. For instance, in a semi-infinite crystal and Bragg case, we have only one physically relevant tiepoint and one component of the Poynting vector. The maximum of this component shifts within the unit cell and the shift depends on the angular deviation η of the incoming beam from the maximum position. The intensity of the secondary effects (fluorescence, photoelectrons) then depends on η as well and this dependence is different for different positions of the source of the secondary effect in the unit cell.

Pioneering paper: B. W. Batterman, Phys. Rev. 133, A759 (1964), see also the review paper by Joerg Zegenhagen, Surface Science Reports 18, 199-271 (1993).

From the lecture by J. Zegenhagen and A. Kazimirov

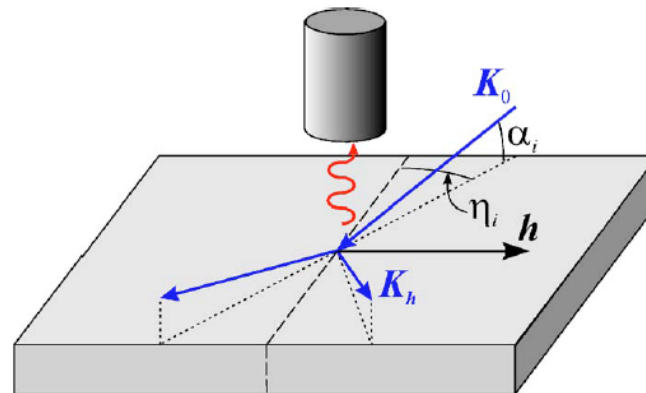
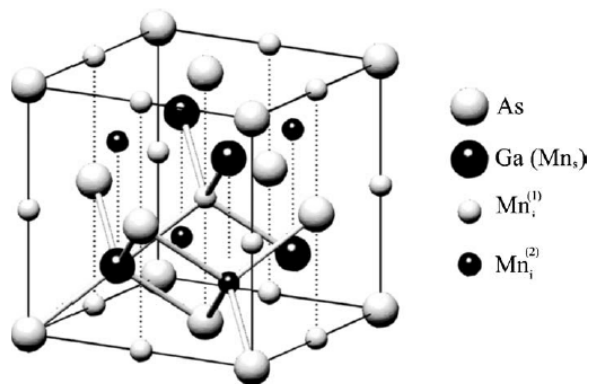


Numerical example: GaAs 004 symmetrical Bragg case, various positions z of the fluorescing atoms



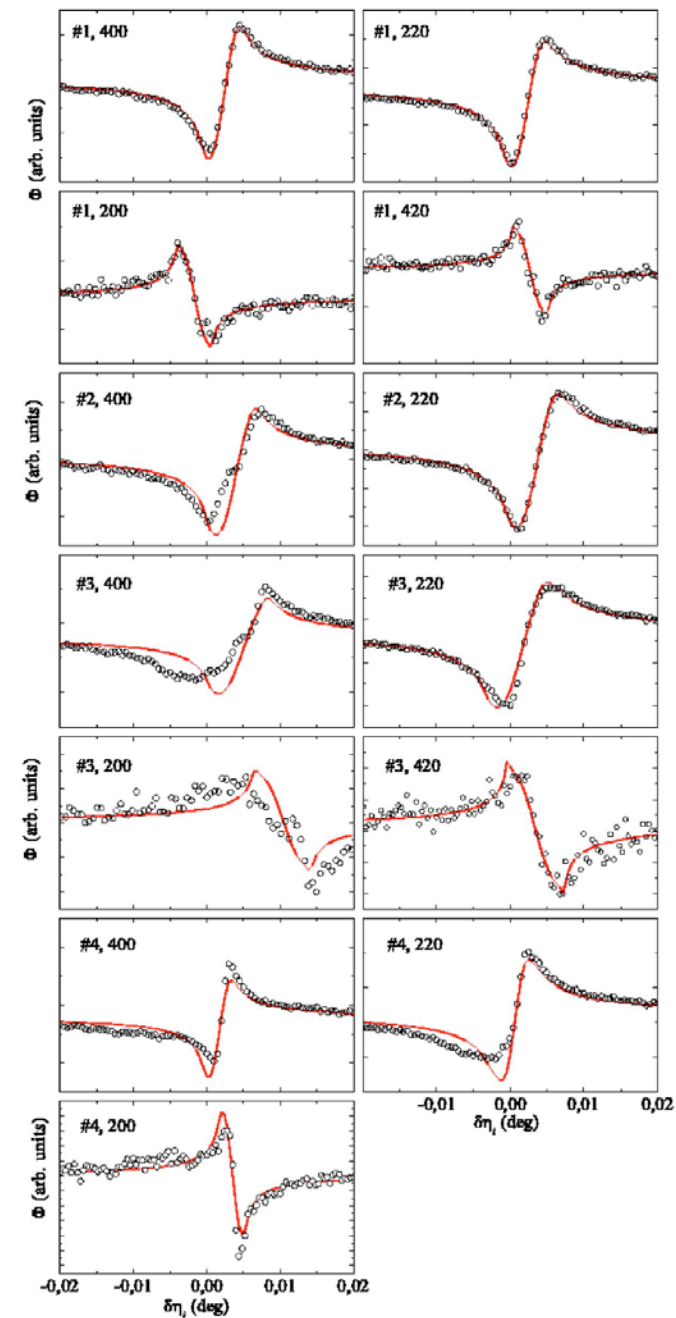
Our old result: *XSW in GID geometry from GaMnAs thin layers*

V. Holý et al. Phys. Rev. B 74, 245205 (2006)



GID experimental arrangement

SFEL Stará Lesná, 2026



There is a lot of other interesting stories about dynamical x-ray diffraction, but I know when to stop ...

THANK YOU FOR YOUR KIND ATTENTION