COMMON OPTICAL APPROACH TO POLARIZED NEUTRON – AND SYNCHROTRON MÖSSBAUER REFLECTOMETRY

L. Deák, L. Bottyán and D. L. Nagy

KFKI Research Institute for Particle and Nuclear Physics, P.O.B. 49, H–1525 Budapest, Hungary

OUTLINE

I. Introduction

II. General considerations

(Scattering problem → wave equation)

III. Common optical formalism (specular scattering)

Neutron reflectometry Mössbauer reflectometry &

x-ray reflectometry

IV. off-specular scattering

V. Applications

VI. Summary

II. GENERAL CONSIDERATIONS

(M. Lax: Rev. Mod. Phys. 23. (1951) 287.)

single scatterer: inhomogeneous wave equation

$$\left[\left(\Delta + k^2\right)I - U(\mathbf{r})\right]\Psi_1(\mathbf{r}) = 0 \qquad (1)$$

$U(\mathbf{r})$	scattering potential	<u>not specialized</u>
$\Psi_1(\mathbf{r})$	amplitude of scattered wave	<u>not specialized</u>
k	vacuum wave number	
$I = \begin{pmatrix} 1 & 0 \\ 0 & 1 \end{pmatrix}$	unity matrix	

many scatterers: homogeneous three dimensional wave equation for the coherent field $\Psi(\mathbf{r})$

$$\left[\left(\Delta + k^2\right)I + 4\pi N\bar{f}\,\right]\Psi(\mathbf{r}) = 0 \qquad (2)$$

$\bar{f}$	coherent forward scattering amplitude
N	density of scattering centers

stratified media: one dimensional wave equation

$$\Psi''(z) + k^2 \sin\Theta\left[I\sin\Theta + \frac{\chi}{\sin\Theta}\right]\Psi(z) = 0 \qquad (3)$$

$\chi \equiv \dfrac{4\pi N}{k^2}\bar{f}$	susceptibility
Θ	glancing angle

III. COMMON OPTICAL FORMALISM

Using matrix notation and the definition $\Phi'(z) \equiv \Psi''(z)$ we get from Eq. (3) a system of first order differential equations

$$\frac{\mathrm{d}}{\mathrm{d}z}\begin{pmatrix}\Phi\\ \Psi\end{pmatrix} = ikM(z)\begin{pmatrix}\Phi\\ \Psi\end{pmatrix}, \text{ where} \tag{4}$$

$$M(z) = \begin{pmatrix} 0 & I\sin\Theta + \dfrac{\chi}{\sin\Theta} \\ I\sin\Theta & 0 \end{pmatrix}. \tag{5}$$

In the case of s different homogeneous layers with thickness d_l: ($l=1...s$, layer s is the substrate)

$$M(z) = M_l = const. \text{ for the } l^{th} \text{ layer,} \tag{6}$$

so the solution may be given by the 4×4 characteristic matrix L, that is the product of the characteristic matrices $L_l = \exp(ikd_lM_l)$ of the individual layers

$$L = L_s \cdot ... \cdot L_2 \cdot L_1 = \exp(ikd_sM_s) \cdot ... \cdot \exp(ikd_2M_2) \cdot \exp(ikd_1M_1). \tag{7}$$

Denoting the 2×2 submatrices of L with $L^{[ij]}$ (ij=1,2) the 2×2 reflectivity matrix r reads

$$r = \left[L^{[11]} - L^{[12]} - L^{[21]} + L^{[22]}\right]^{-1}\left[L^{[11]} + L^{[12]} - L^{[21]} - L^{[22]}\right]. \tag{8}$$

The reflected intensity I^r is

$$I^r = Tr\left[r^+ r\rho\right], \tag{9}$$

and ρ is the 2×2 polarization density matrix of the incident radiation.

TREATMENT OF THE NUMERICAL PROBLEMS

Problem #1: *Calculation of the* 4×4 *matrix exponentials* $L_l = \exp(ikd_l M_l)$

$$L_l = \begin{pmatrix} \cosh(F_l) & \frac{1}{x_l} F_l \sinh(F_l) \\ x_l F_l^{-1} \sinh(F_l) & \cosh(F_l) \end{pmatrix}, \tag{10}$$

where $F_l = kd_l \sqrt{-I\sin^2\Theta - \chi_l}$ and $x_l = ikd_l \sin\Theta$. ✔ **OK**

Problem #2: the calculation of the 2×2 square root matrix F_l from the problem #1

from the Cayley–Hamilton theory for any 2×2 matrices G

$$G^{1/2} = \frac{G + I\sqrt{\det G}}{\sqrt{\mathrm{Tr}G + 2\sqrt{\det G}}} \quad (\text{if } G \sim I\text{, then } G^{1/2} = I(\det G)^{1/4}) \tag{11}$$

✔ **OK**

Problem #3: *Calculation of the* 2×2 *(sinh and cosh)* $\rightarrow$ *exp functions* in problem #1

Using the identity:

$$\exp G = \exp\left(\frac{1}{2}\mathrm{Tr}\,G\right)\left[\cos\sqrt{\det\overline{G}}\,I + \frac{\sin\sqrt{\det\overline{G}}}{\sqrt{\det\overline{G}}}\sqrt{\overline{G}}\right], \tag{12}$$

where $\overline{G} = G - \frac{1}{2} I\,\mathrm{Tr}\,G$ ✔ **OK**

Problem #4: The substrate:

The characteristic matrix of a semi-infinite layer $L_s \to L_\infty$ is calculated by taking the $d_s \to \infty$ limes. From Eq. (10) we get

$$L_\infty = \begin{pmatrix} I & a\sqrt{I + \dfrac{\chi_s}{\sin^2\Theta}} \\ a\left(\sqrt{I + \dfrac{\chi_s}{\sin^2\Theta}}\right)^{-1} & I \end{pmatrix}, \tag{13}$$

where $a = \operatorname{sgn}[\operatorname{Re}(\operatorname{Tr} F_\infty)]$ is the sign of the real part of the trace of $F_\infty = \sqrt{-I\sin^2\Theta - \chi_s}$.

✓ **OK**

Problem #5: Interface and surface roughness:

In the case of rough interfaces the characteristic matrix L_l of layer l has to be modified:

$$L_l \to \begin{pmatrix} L_l^{[11]}\left[I + k^2\left(\sigma_l^2 - \sigma_{l+1}^2\right)\chi_l\right] & L_l^{[12]}\left[I - k^2\left(\sigma_l^2 + \sigma_{l+1}^2\right)\chi_l\right] \\ L_l^{[21]}\left[I + k^2\left(\sigma_l^2 + \sigma_{l+1}^2\right)\chi_l\right] & L_l^{[22]}\left[I - k^2\left(\sigma_l^2 - \sigma_{l+1}^2\right)\chi_l\right] \end{pmatrix}, \tag{14}$$

where

σ_l and σ_{l+1}: *RMS surface roughness* at the top and bottom of the layer.

We assume $d_l << \sigma_l, \sigma_{l+1}$.

The approximation is in the order of $(k\sigma)^2\|\chi\|$.

✓ **OK**

NEUTRON REFLECTOMETRY:

Using the potential $U(\mathbf{r}) = U_p(\mathbf{r}) + U_m(\mathbf{r})$ as the sum of the isotropic nuclear potential

$$U_p(\mathbf{r}) = 4\pi b\delta(\mathbf{r})I \tag{15}$$

and the anisotropic magnetic potential

$$U_m(\mathbf{r}) = -\frac{2m}{\hbar^2} g\mu_n \boldsymbol{\sigma} \cdot [\mathbf{B}_a(\mathbf{r}) + \mathbf{B}_{ext}] = -\frac{2m}{\hbar^2} g\mu_n \boldsymbol{\sigma} \cdot \mathbf{B}(\mathbf{r}), \tag{16}$$

where

$\mu_N = 5.050 \times 10^{-27}\,\mathrm{Am}^2$,
$g = -1.9132$,
$\boldsymbol{\sigma} = (\sigma_\xi, \sigma_\eta, \sigma_\varsigma)$ is the Pauli operator,
b: is the nuclear scattering length, and
$\mathbf{B}_a(\mathbf{r})$ and $\mathbf{B}_{\mathrm{ext}}$ are the atomic and the external magnetic field.

In the 1st order Born approximation the coherent forward scattering amplitude is $\bar{f} = -\frac{1}{4\pi}\int \mathrm{d}^3\mathbf{r}\, U(\mathbf{r})$, and $\chi \equiv \frac{4\pi N}{k^2}\bar{f}$

$$\chi = \frac{1}{k^2}\left[\frac{2m}{\hbar^2} g\mu_N \boldsymbol{\sigma} \cdot \overline{\mathbf{B}} - 4\pi \sum_i \alpha_i b_i I\right], \tag{17}$$

where

α_i is the abundance of the i^{th} type nuclear scattering center,
$\overline{\mathbf{B}} = \frac{1}{\Omega}\int_\Omega \mathrm{d}^3\mathbf{r}\, \mathbf{B}(\mathbf{r})$, and Ω is the volume of the interaction.

NEUTRON OPTICS

$$\frac{\mathrm{d}}{\mathrm{d}z}\begin{pmatrix}\Phi\\ \Psi\end{pmatrix} = ik\begin{pmatrix}0 & I\sin\Theta + \dfrac{\chi}{\sin\Theta}\\ I\sin\Theta & 0\end{pmatrix}\begin{pmatrix}\Phi\\ \Psi\end{pmatrix} = i\begin{pmatrix}0 & I\dfrac{Q}{2} + \dfrac{2K}{Q}\\ I\dfrac{Q}{2} & 0\end{pmatrix}\begin{pmatrix}\Phi\\ \Psi\end{pmatrix} \tag{18}$$

where:

momentum transfer:

$$Q=2k\sin\theta$$

scattering length density:

$$K = \frac{2m}{\hbar^2} g\mu_N \begin{pmatrix}B_x & B_y - iB_z\\ B_y + iB_z & -B_x\end{pmatrix} - 4\pi\sum_i \alpha_l b_l I$$

magnetic field:

$$(B_x, B_y, B_z) = \overline{\mathbf{B}}.$$

External magnetic field $\Rightarrow K \neq 0 \Rightarrow$ vacuum is an *anisotropic medium*

$\Downarrow$

Eq. (8) invalid!!!

The solution is well known in (photon) optics: impedance tensor

$$\gamma = \sqrt{I + \frac{4K}{Q^2}} = \frac{1}{Q}\begin{pmatrix}\sqrt{Q_+} & 0\\ 0 & \sqrt{Q_-}\end{pmatrix}, \text{ with } Q_\pm^2 = Q^2 \pm \frac{8m}{\hbar^2} g\mu_N \left|\mathbf{B}_{ext}\right|$$

And finally the reflectivity matrix:

$$r = \left[\left(L^{[11]} - L^{[21]}\right)\gamma - L^{[12]} + L^{[22]}\right]^{-1}\left[\left(L^{[11]} - L^{[21]}\right)\gamma + L^{[12]} - L^{[22]}\right]. \tag{19}$$

MÖSSBAUER REFLECTOMETRY:

FROM THE DYNAMICAL THEORY:

*J.P. Hannon et al, Phys. Rev B **32**, 6363 (1985)*

The grazing incidence limit of the dynamical theory of Mössbauer radiation in matrix form:

$$\frac{\mathrm{d}}{\mathrm{d}z}\begin{pmatrix} T \\ R \end{pmatrix} = i\begin{pmatrix} g_0 I + G & G \\ G & g_0 I + G \end{pmatrix}\begin{pmatrix} T \\ R \end{pmatrix}, \qquad (20)$$

where T and R are the amplitudes of waves incident from above and below, respectively. With $g_0 = k\sin\Theta$ and $G = (4\pi N/2k\sin\Theta)\overline{f}$

$$\frac{\mathrm{d}}{\mathrm{d}z}\begin{pmatrix} T \\ R \end{pmatrix} = ik\begin{pmatrix} I\sin\Theta + \frac{\chi}{2\sin\Theta} & \frac{\chi}{2\sin\Theta} \\ -\frac{\chi}{2\sin\Theta} & -I\sin\Theta - \frac{\chi}{2\sin\Theta} \end{pmatrix}\begin{pmatrix} T \\ R \end{pmatrix}. \qquad (21)$$

Applying the unitary transformation $C = \frac{1}{\sqrt{2}}\begin{pmatrix} I & -I \\ I & I \end{pmatrix}$ we get

$$\frac{\mathrm{d}}{\mathrm{d}z}\begin{pmatrix} T \\ R \end{pmatrix}' = ik\begin{pmatrix} 0 & I\sin\Theta + \frac{\chi}{\sin\Theta} \\ I\sin\Theta & 0 \end{pmatrix}\begin{pmatrix} T \\ R \end{pmatrix}', \qquad (22)$$

and again: $\frac{\mathrm{d}}{\mathrm{d}z}W(z) = ikM(z)W(z)$.

Validity: $\Theta < 10\,\mathrm{mrad}$

FROM THE OPTICAL THEORY:

L. Deák et al, Phys. Rev B ***53****, 6158 (1996):*

The 3×3 nuclear susceptibility tensor (Afanas'ev and Kagan):

$$\hat{\chi} = -\frac{4\pi}{c^2 k^2} \frac{N}{2I_g + 1} \sum_{m_e m_g} \frac{\mathbf{J}_{m_g m_e} \circ \mathbf{J}^*_{m_e m_g}}{E_k - E_{m_e m_g} + i\Gamma/2}, \qquad (23)$$

where: E_k: the energy of the photon,

$E_{m_e m_g}$: the energy difference between the nuclear excited and ground states,

Γ: the natural width of the excited states,

$\mathbf{J}$: the current density operator.

$\circ$: the symbol of the dyadic vector product

Using the anisotropic optical formalism (Borzdov–Barkovskii–Lavrukovich) and the Andreeva approximation, the Maxwell equations transform into

$$\frac{\mathrm{d}}{\mathrm{d}z}\begin{pmatrix} T \\ R \end{pmatrix} = ik \begin{pmatrix} 0 & I\sin\Theta + \dfrac{\chi}{\sin\Theta} \\ I\sin\Theta & 0 \end{pmatrix} \begin{pmatrix} T \\ R \end{pmatrix},$$

where

$$\chi = \frac{4\pi N}{k^2}\bar{f} \qquad (24)$$

Conclusion: common differential equations for different scattering processes

Validity: $1\,\mathrm{mrad} < \Theta < 10\,\mathrm{mrad}$

IV. OFF-SPECULAR SCATTERING

inhomogeneous wave equation

$$\left[\Delta + k^2 I\right]\Psi(\mathbf{r}) = -k^2 \chi(\mathbf{r})\Psi(\mathbf{r}), \qquad (25)$$

where $\chi(\mathbf{r}) = \sum_{l=1}^{S} \chi_l(\mathbf{r}_{\mathrm{II}})$ and l is the layer index.

Defining $\bar{\chi}_l = \langle \chi_l(\mathbf{r}_{\mathrm{II}}) \rangle$ we can separate the $\Psi_{coh}(\mathbf{r})$ (coherent) specular and $\Psi_{off}(\mathbf{r})$ off-specular fields

$$\left[\Delta + k^2 I\right]\Psi(\mathbf{r}) = -k^2 \sum \bar{\chi}_l \Psi(\mathbf{r}) - k^2 \sum \left(\chi_l(\mathbf{r}_{\mathrm{II}}) - \bar{\chi}_l\right)\Psi(\mathbf{r}) \qquad (26)$$

$\Downarrow$

with $\Psi(\mathbf{r}) = \Psi_{coh}(\mathbf{r}) + \Psi_{off}(\mathbf{r})$

$$\left[\Delta + k^2 I\right]\Psi_{off}(\mathbf{r}) = -k^2 \sum \left(\chi_l(\mathbf{r}_{\mathrm{II}}) - \bar{\chi}_l\right)\Psi_{coh}(\mathbf{r}) - k^2 \sum \chi_l(\mathbf{r}_{\mathrm{II}})\Psi_{off}(\mathbf{r}) \qquad (27)$$

$\Downarrow$

known

$$\begin{aligned}\Psi_{coh}(\mathbf{r}) &= \Psi_{coh}(\mathbf{r}_\perp)\exp(i\mathbf{k}_{\mathrm{II}} \cdot \mathbf{r}_{\mathrm{II}}) \\ &= \left[L^{[21]}(\mathbf{r}_\perp) + L^{[22]}(\mathbf{r}_\perp) r\right] \cdot \Psi^{in} \exp(i\mathbf{k}_{\mathrm{II}} \cdot \mathbf{r}_{\mathrm{II}})\end{aligned} \qquad (28)$$

1st DWBA: neglecting the second term in Eq. (27)

$$\Psi_{off}(\mathbf{r}) = \frac{k^2}{4\pi}\sum_l \int \frac{\exp(ikR)}{R}\left(\chi_l(\mathbf{r}'_{\mathrm{II}}) - \overline{\chi}_l\right)\Psi_{coh}(\mathbf{r}')\mathrm{d}^3\mathbf{r}' \qquad (29)$$

where $R = |\mathbf{r} - \mathbf{r}'|$

In the Fraunhofer approximation $\frac{\exp(ikR)}{R} \approx \frac{\exp(ikr)}{r}\exp(i\mathbf{k}'\cdot\mathbf{r}')$ and the scattered intensity is proportional to:

$$I_{off} = \frac{k^4}{(4\pi r)^2}\int\int\left(\Psi_{coh}\left(\mathbf{r}''_{\perp}\right)\sum_{l,l'} C_{l,l'}(\mathbf{K}_{\mathrm{II}})\cdot\Psi_{coh}\left(\mathbf{r}'_{\perp}\right)\right)\cdot \exp\left[i\mathbf{k}'_{\perp}\left(\mathbf{r}'_{\perp} - \mathbf{r}''_{\perp}\right)\right]\mathrm{d}\mathbf{r}'_{\perp}\mathrm{d}\mathbf{r}''_{\perp} \qquad (30)$$

$C_{l,l'}(\mathbf{K}_{\mathrm{II}})$ is the two dimensional Fourier transformation of the cross correlation function of $\left(\chi_l(\mathbf{r}'_{\mathrm{II}}) - \overline{\chi}_l\right)$ and **K**=**k’**-**k**

V. APPLICATIONS

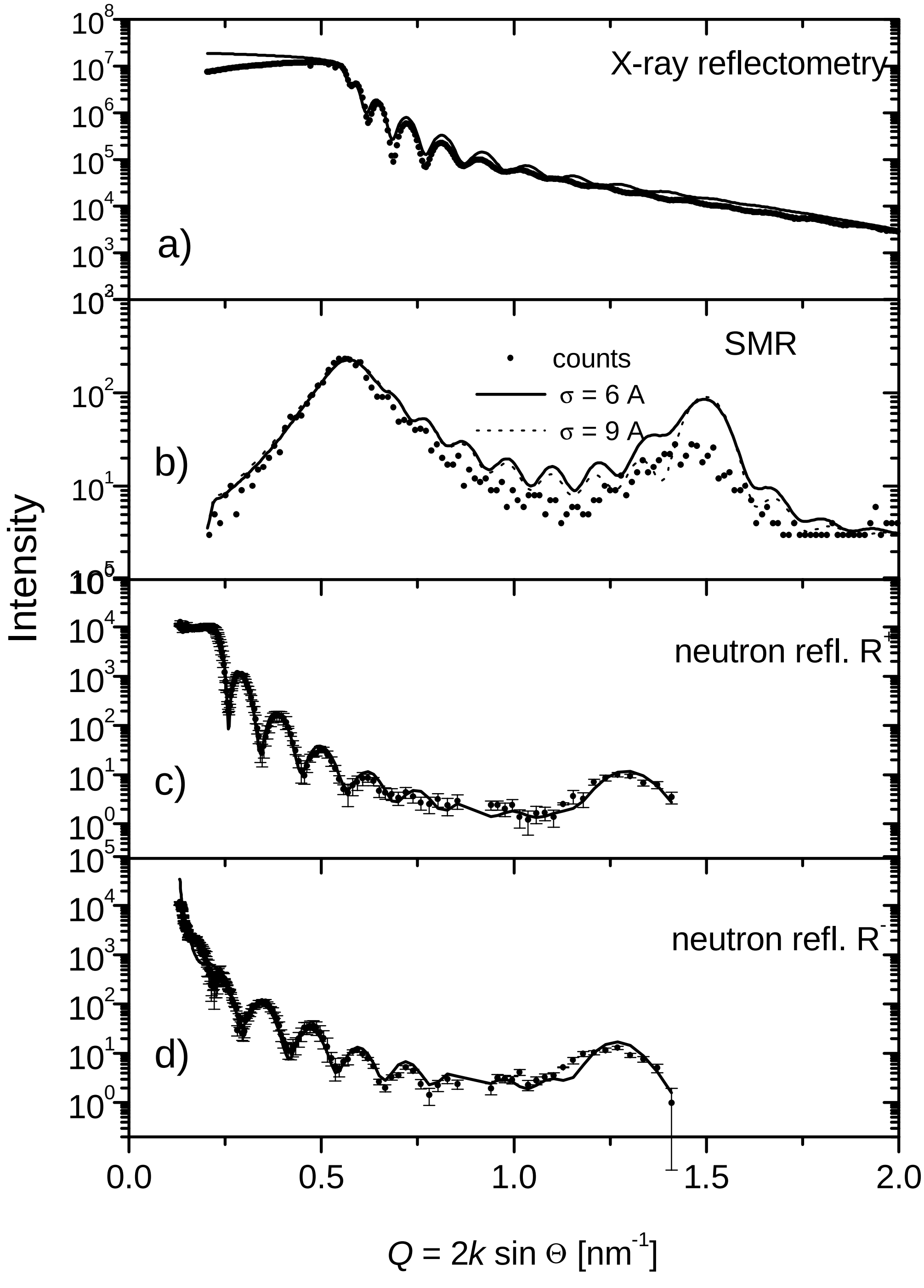

Fig.: Different type of reflectometry measurements of $[^{57}Fe(2.33\ nm)/^{nat}Fe(2.33\ nm)]_{10}$/ZERODUR multilayer. The data evaluation was made in terms of the common optical algorithm.

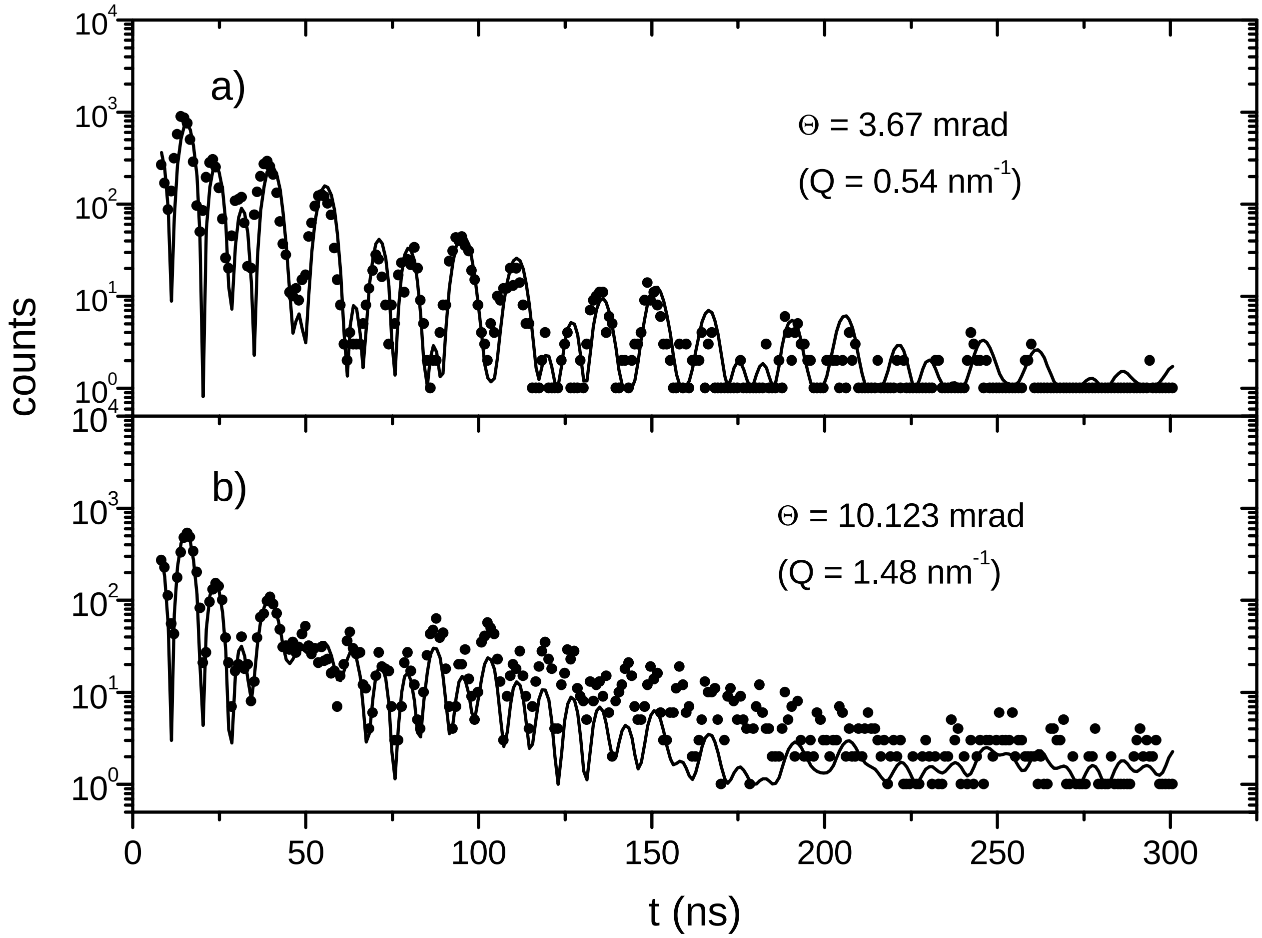

Fig.: Time spectra at the total reflection peak and the first Bragg peak in $[^{57}Fe(2.33\ nm)/^{nat}Fe(2.33\ nm)]_{10}$/ZERODUR multilayer. The data evaluation was made in terms of the common optical algorithm.

VI. SUMMARY

- *Without* specifying the scattering process
  a common formalism is derived, which
- simplifies to a 2×2 matrix algebra and which is
- *suitable* for x-ray-, Mössbauer- and n-reflectometry
- This analogy helped when treating the external magnetic field as an anisotropic optical medium (for neutrons).
- Common formalism $\Rightarrow$ common evaluation program (EFFINO - Environment For Fitting Nuclear Optics)