Neutron scattering from functional magnetic nano-patterns

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Recent review on the topic:

H. Zabel, K. Theis-Bröhl, B.P. Toperverg,

“Polarized neutron reflectivity and scattering of magnetic nanostructures and spintronic materials“,
Handbook of Magnetism and Advanced Magnetic Materials,

H. Kronmüller & S. Parkin (Eds.), NY, Wiley 2007, p. 1237-1288

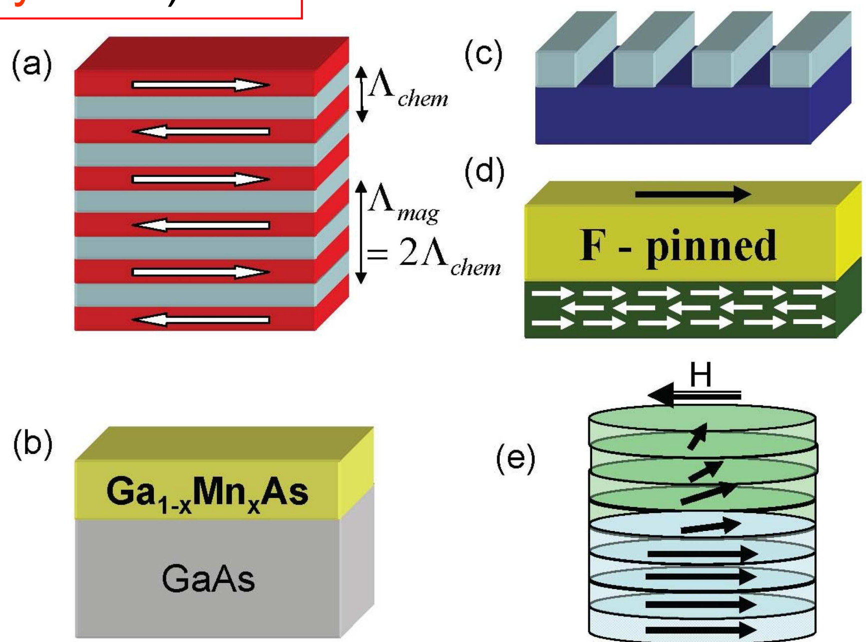
Grazing incidence neutron scattering from magnetic nanostructures

1. Examples of nano-systems under studies with neutrons
2. Coherence length and time in neutron experiment
3. Born approximation and Born series
4. Specular reflection of polarized neutrons (PNR)
5. Distorted wave Born approximation (DWBA)
6. Off-specular Bragg diffraction and diffuse scattering
7. Small angle scattering at grazing incidence (GISANS)
8. AC reflectometry
9. Inelastic reflection

Handmade nano-materials for electronics under routine investigations with neutrons

a) Exchange coupled bi-layers and superlattice with antiferromagnetic ordering (GMR and TMR systems)

c) Laterally patterned magnetic films, nano-wires



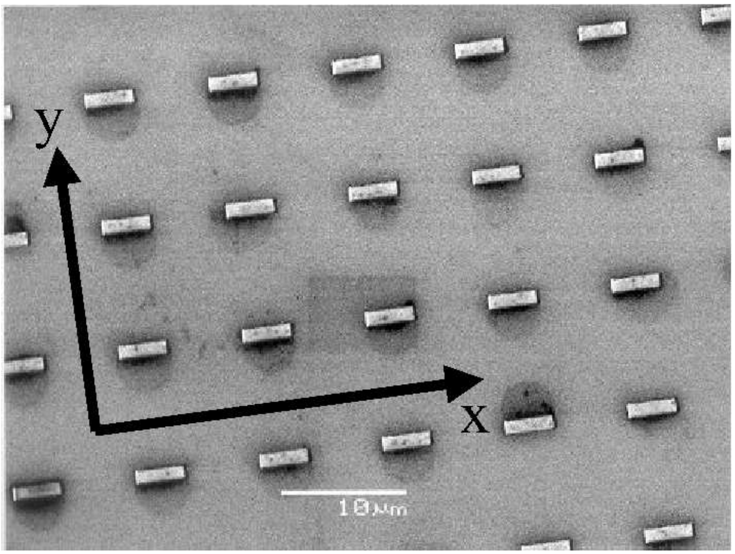
d) Ferromagnetic films on antiferromagnetic substrates with Exchange bias through common interfaces

b) Dilute magnetic semiconductors as spin-injectors in semiconductor heterostructures

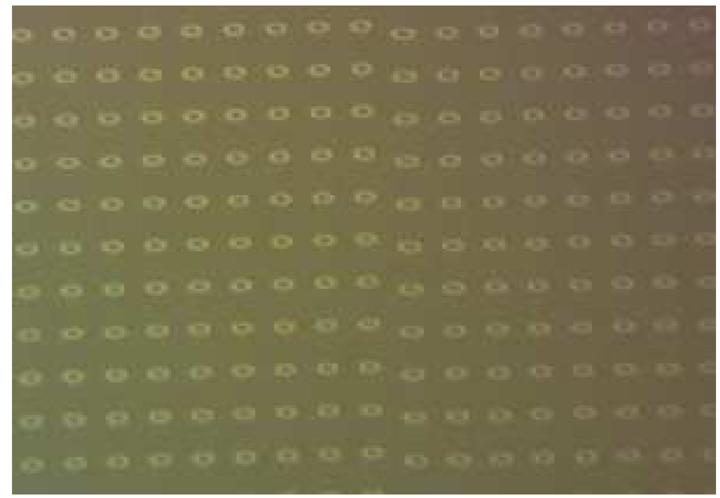
e) Spring magnets: soft magnetic layer exchange coupled to a magnetically hard layer (spin valves)

2D lateral patterns: bars, discs etc.

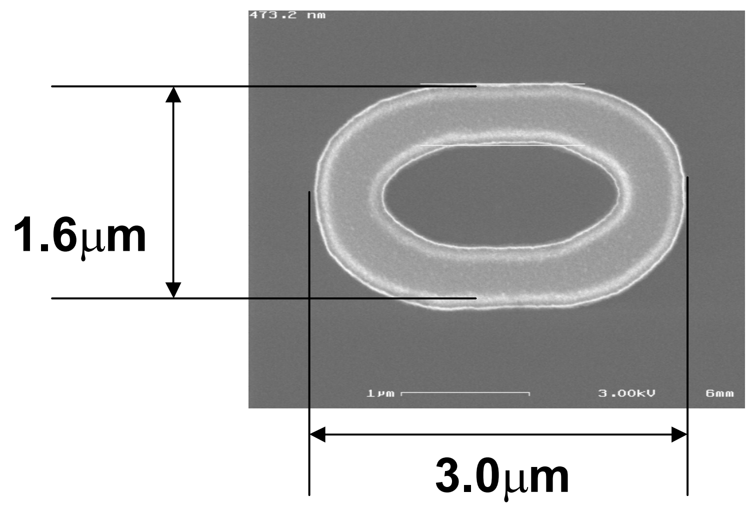
Cobalt bars on Si wafer (10 microns)



NiFe Ellipse arrays: thickness 20nm



Cobalt discs

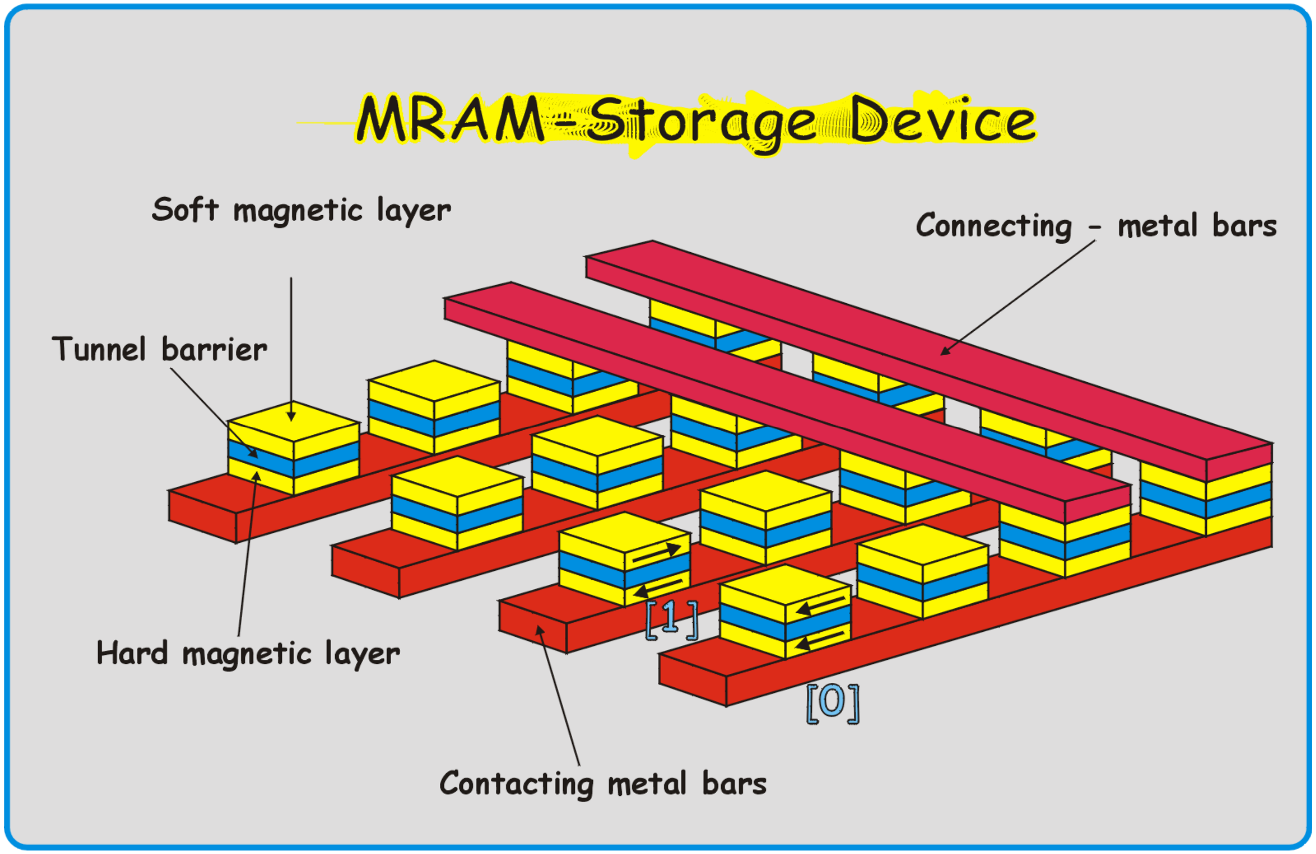


To-day spintronics application:
Magnetic Random Access Memory (MRAM)

- → "read-write" 2D arrays of spin valves
  - Writing with weak magnetic field
  - Reading with electric current

Neutrons:

Revealing spin configurations in nano-elements, nature and role of magnetic interactions, interfacial properties, etc.

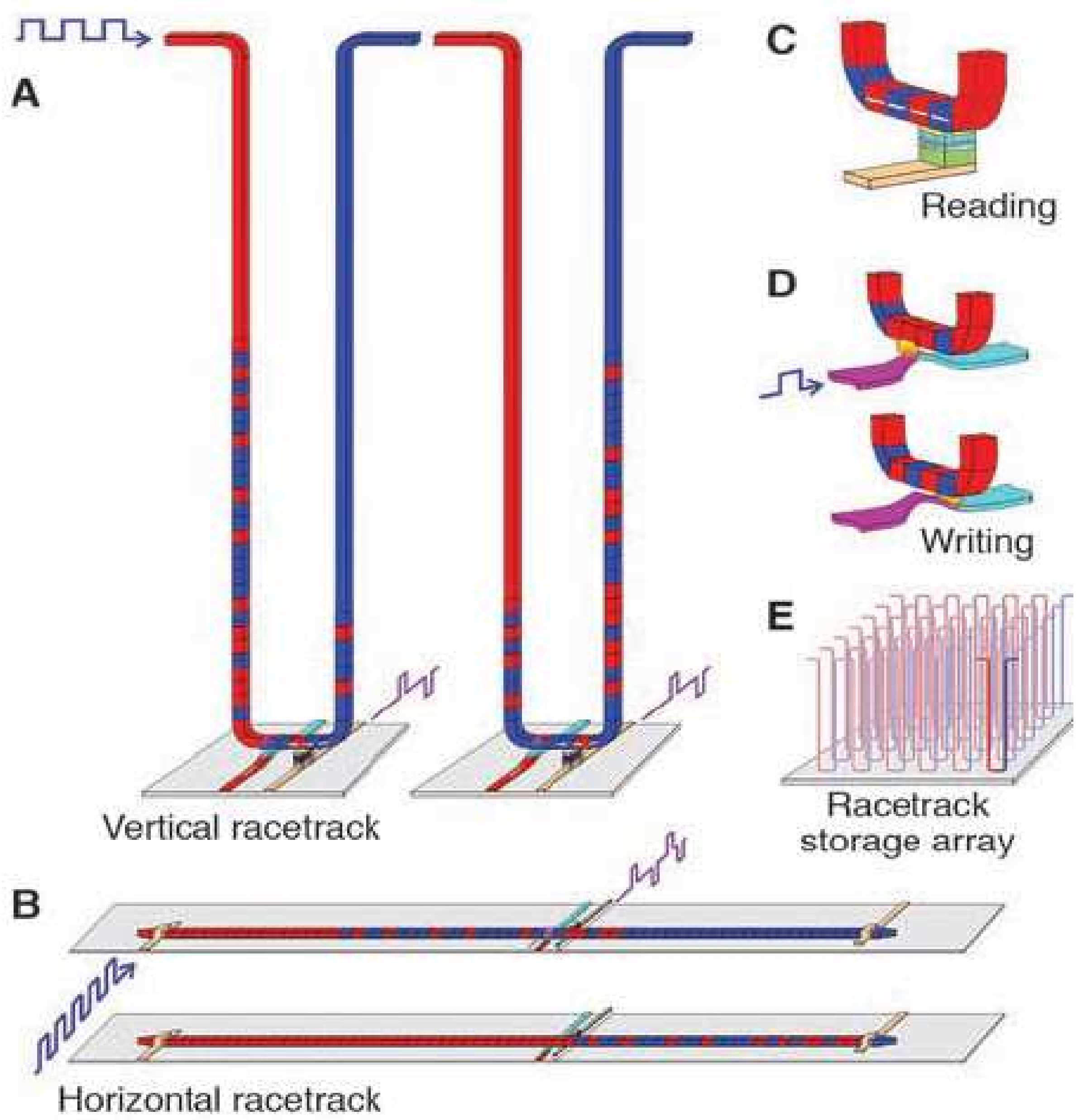


Forthcoming to-morrow?

Review: 11 APRIL 2008 VOL 320 SCIENCE

Magnetic Domain-Wall Racetrack Memory

Stuart S. P. Parkin,* Masamitsu Hayashi, Luc Thomas



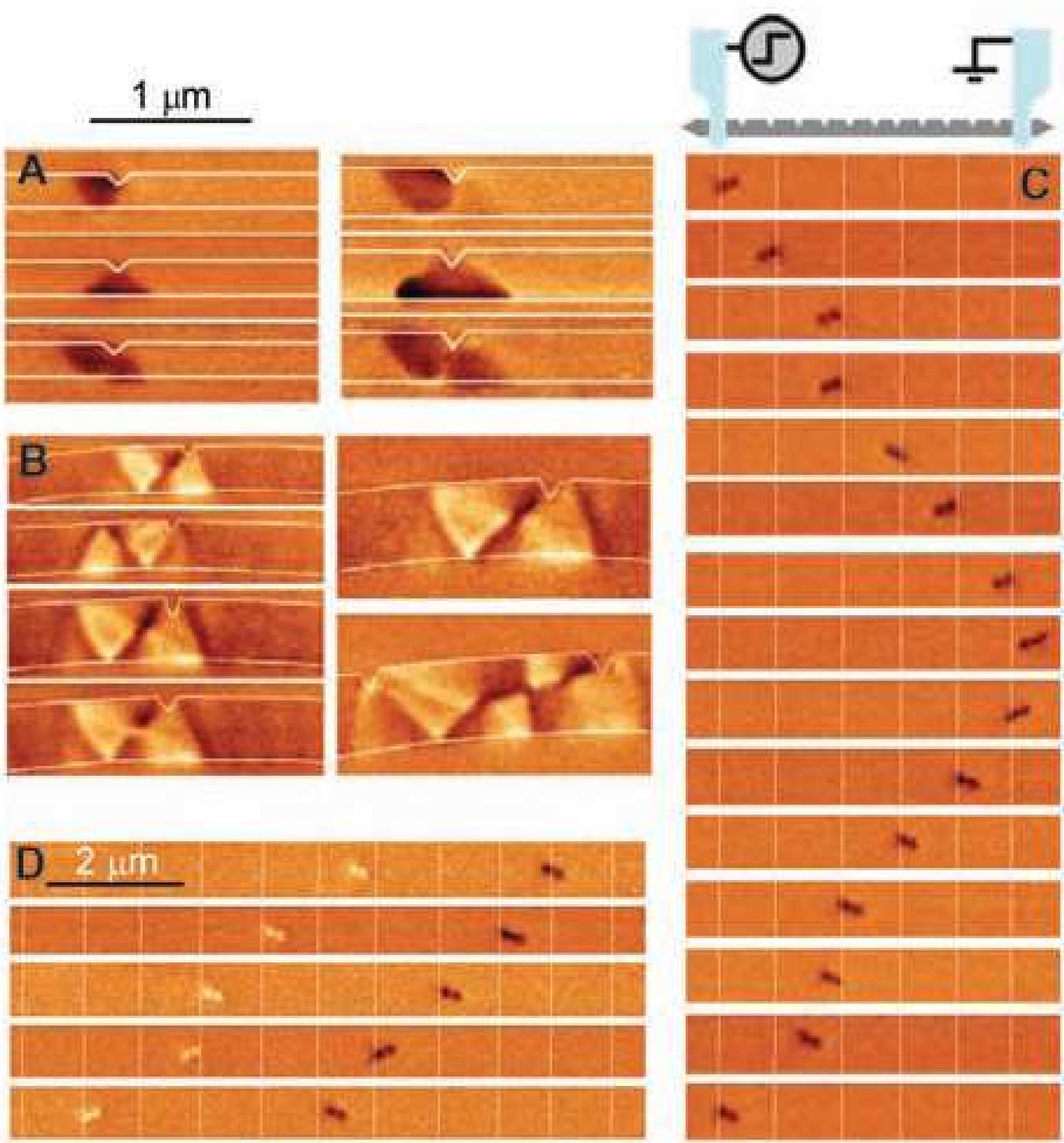
Domain propagation
in nano-wires:
New challenge for
neutrons

Exploiting 3D:
Books versus
one sheet newspapers?

Review: 11 APRIL 2008 VOL 320 SCIENCE

Magnetic Domain-Wall Racetrack Memory

Stuart S. P. Parkin,* Masamitsu Hayashi, Luc Thomas



- Domain propagation in nano-wires:

New challenge for neutrons

What do we want and can learn from neutron scattering at grazing incidence?

1. Specular reflectometry:

a) Distribution of materials and magnetization across the multilayer stack (layer-by-layer vector magnetometry)
b) Properties of interfaces (interdiffusion, roughness, e.g. magnetic, etc.)

2. Off specular scattering:

a) Lateral parameters of microstructures
b) In- and out-of-plane distribution of magnetization vector, e.g. over lateral magnetic domains

3. Small angle scattering at grazing incidence (GISANS) and diffraction (GID): nano and atomic scale structural parameters

4. Time resolved and inelastic PNR: re-magnetization times, domain wall nucleation, velocity and relaxation, spin excitations spectrum

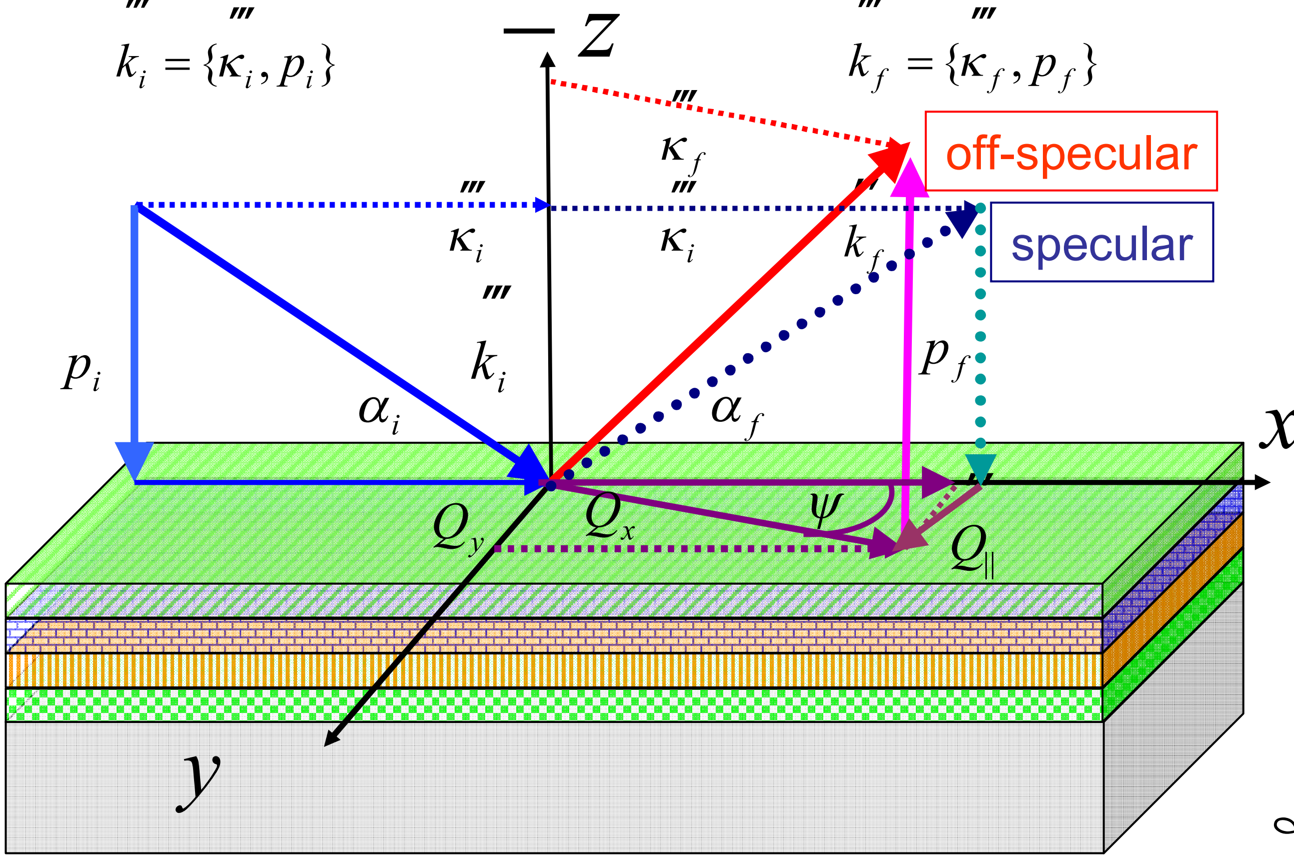
General scattering kinematics at grazing incidence

Incident wave vector:

$$\vec{k}_i = \{\vec{\kappa}_i, p_i\}$$

Outgoing wave vector:

$$\vec{k}_f = \{\vec{\kappa}_f, p_f\}$$



Wave vector transfer:

$$\vec{Q} = \vec{k}_f - \vec{k}_i = \{Q^x, Q^y, Q^z\}$$

$$\vec{Q}_\parallel = \vec{\kappa}_f - \vec{\kappa}_i$$

$$Q^x = \kappa_f^x - \kappa_i^x$$

$$Q^y = k \sin\psi \approx \frac{2\pi}{\lambda}\psi$$

$$Q^z = p_f - p_i$$

$$\hbar k_i^2 = 2m\varepsilon_i$$

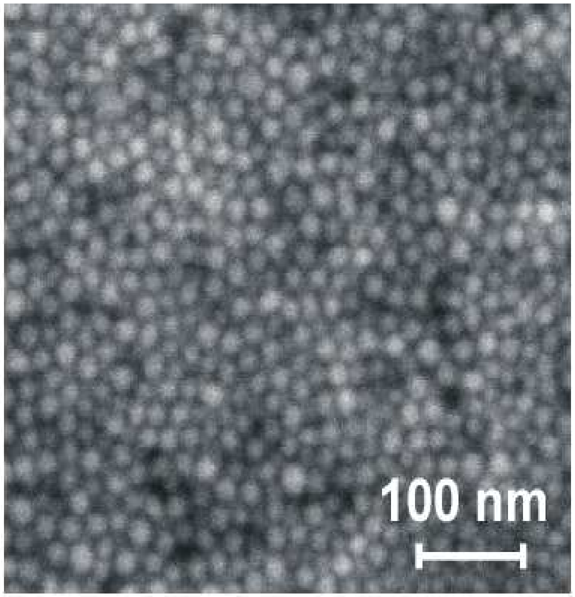
$$\hbar k_f^2 \approx 2m\varepsilon_i(1 + \omega / 2\varepsilon_i)$$

$$Q^z = k_f \sin\alpha_f + k_i \sin\alpha_i \approx \frac{2\pi}{\lambda_i}(\alpha_f + \alpha_i);$$

$$Q^x = k_f \cos\alpha_f \cos\psi - k_i \cos\alpha_i \approx \frac{\pi}{\lambda}(\alpha_f^2 - \alpha_i^2 - \psi^2 - \omega/\varepsilon) << Q^z; Q^y$$

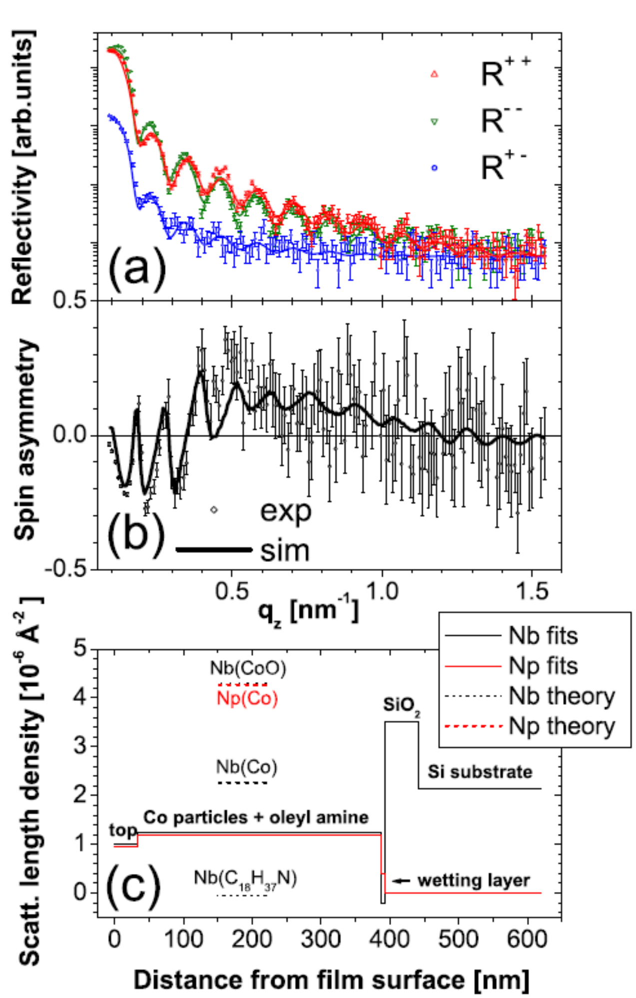
B.P. Toperverg, PNPI Winter School, 14/03/2009

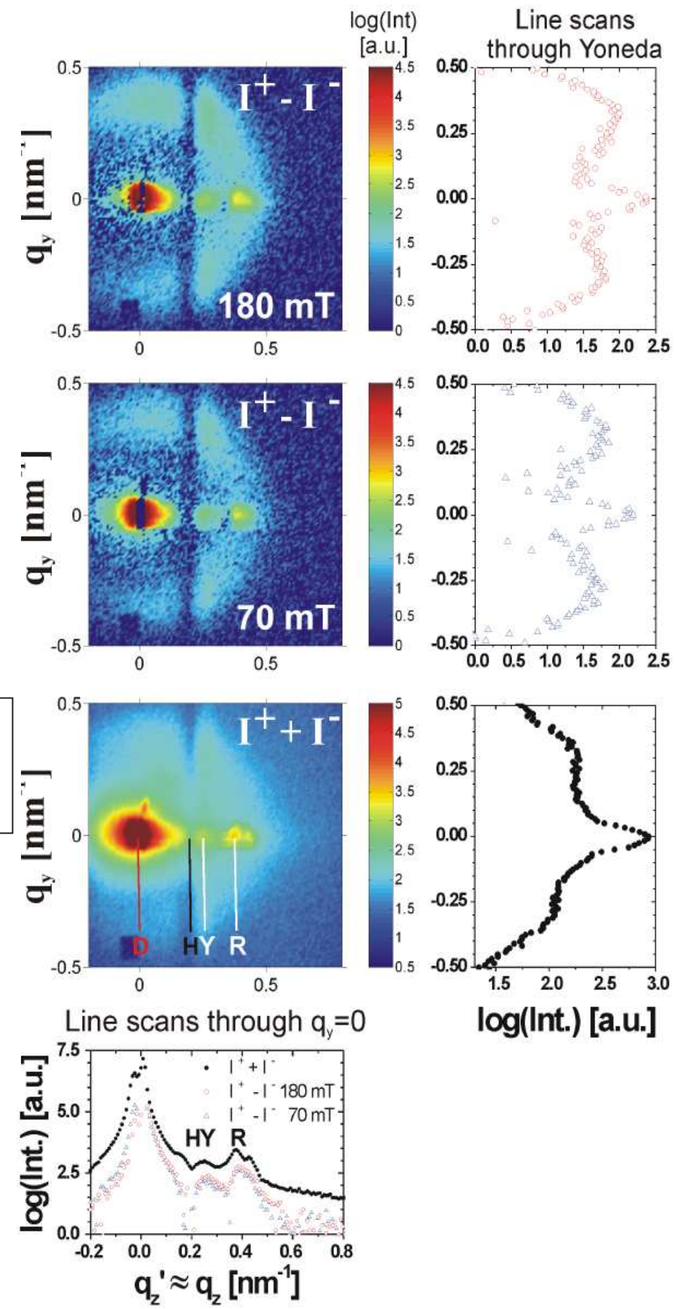
GISANS for Co multilayer of Co nano- particles (Bochum, D22 ILL)



SEM image of the Co nanoparticl

SELF-ORDERING OF NANOPARTICLES IN MAGNETO-…

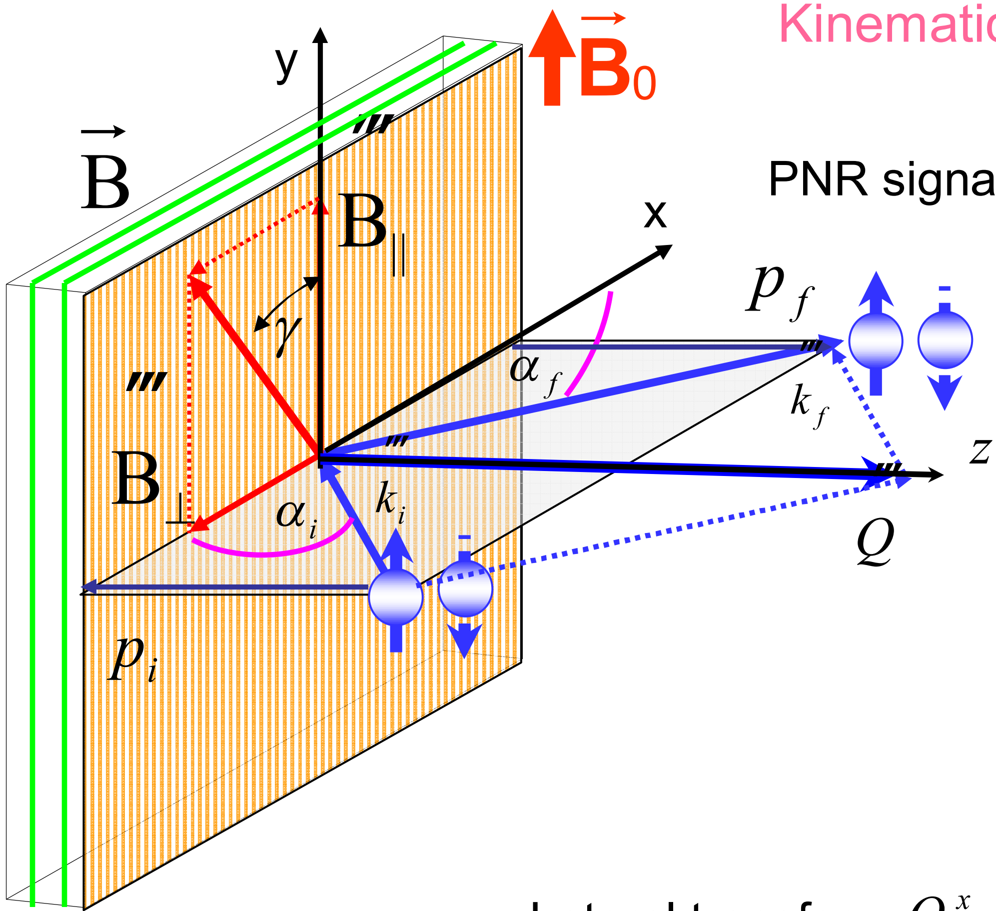




PRB 78, 134426 (2008)

B.P. Toperverg, PNPI Winter School, 26/02/2008

Kinematics of polarized neutron reflectometry (PNR)



PNR signal is integrated over $Q^y = k \sin\psi$

2D Wave vector transfer: $\vec{Q} = \{Q^x, Q^z\}$

Transverse component:

$$Q^z = p_f - p_i \approx \frac{2\pi}{\lambda}(\alpha_f + \alpha_i)$$

Lateral transfer: $Q^x = p_i + p_f \approx \frac{\pi}{\lambda}(\alpha_f^2 - \alpha_i^2 + \omega/\varepsilon) << Q^z$

specular reflection at: $Q^x = 0 \Rightarrow \alpha_f = \alpha_i$ Snell's law: zero order Bragg diffraction

off-specular scattering: $Q^x \neq 0, \Rightarrow \alpha_i \neq \alpha_f$ from lateral structure

Formalism of DWBA (stationary case)

$$\left\{-\frac{\hbar^2}{2m}\nabla^2+\hat{U}_0(z)+\Delta\hat{U}(\vec{r})-E\right\}\left|\Psi(\vec{r})\right\rangle=0$$

$$\text{Schrödinger equation for spinor}\left|\Psi(\vec{r})\right\rangle=\begin{pmatrix}\Psi_+\\ \Psi_-\end{pmatrix}$$

$$\hat{U}(\vec{r})=\hat{U}_{\text{nucl}}(\vec{r})+\hat{U}_{\text{magn}}(\vec{r});\qquad \hat{U}_{\text{nucl}}=\frac{2\pi\hbar^2}{m}Nb\ \ \text{(scattering length density - SLD)}$$

$$\hat{U}_{\text{magn}}(\vec{r})=\hat{\vec{\mu}}\vec{B}(\vec{r})=\mu\begin{pmatrix}B_z & B_x-iB_y\\ B_x+iB_y & -B_z\end{pmatrix};\qquad \hat{\vec{\mu}}=\mu\hat{\vec{\sigma}}\ \ \text{(neutron magnetic moment)}$$

$$\hat{U}_0(z)=\left\langle\hat{U}(\vec{r})\right\rangle\ \ \text{is mean potential averaged over}\ \ l_{x,y}^{\text{coh}}$$

$$\Delta\hat{U}(\vec{r})=\hat{U}(\vec{r})-\left\langle\hat{U}(\vec{r})\right\rangle\ \ \text{is perturbation}$$

$$\text{Solution}:\quad \left|\Psi(\vec{r})\right\rangle=\left|\Psi_0(z)\right\rangle e^{i\vec{\kappa}\vec{\rho}}+\left|\Psi_s(\vec{r})\right\rangle$$

$\left|\Psi_0(z)\right\rangle$ is the reference spinor (reflection, birefringence, transmission)

$\left|\Psi_s(\vec{r})\right\rangle$ correction (off - specular scattering)

Reference solution

$$\left\{-\frac{\hbar^2}{2m}\left(\nabla^2-\kappa^2\right)-E+\hat{U}_0(z)\right\}\left|\Psi_0(z)\right\rangle=0$$

if $\hat{U}_0(z)=\hat{U}_0$ then $\left|\Psi_0(z)\right\rangle=\hat{t}e^{i\hat{p}z}+\hat{r}e^{-i\hat{p}z}$ with

$\hat{p}=\sqrt{p_0^2-\hat{p}_c^2}$ is the matrix of refracted wave numbers;

$p_0=(2\pi/\lambda)\sin\alpha$; $\hbar p_0^2=2mE - \hbar\kappa^2$;

$\hbar\hat{p}_c^2=2m(U_{\mathrm{nucl}}+\hat{\vec{\mu}}B)$ is the matrix of total reflection numbers

$\hat{t}=t\left(\hat{\vec{\sigma}}\right)$ is the transmittance, and $\hat{r}=r\left(\hat{\vec{\sigma}}\right)$ the reflectance matrix

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In vacuum $\left|\Psi_0(z)\right\rangle=\left\{e^{ip_0z}+\hat{\mathrm{R}}e^{ip_0z}\right\}\left|\Psi_i\right\rangle$

$\hat{R}=R(\hat{\vec{\sigma}})$ is the matrix of reflection amplitudes

$$\frac{\partial\sigma}{\partial\Omega}=S(\alpha_i)\left|\left\langle\Psi_i\right|\hat{R}\left|\Psi_f\right\rangle\right|^2\delta(\alpha_i-\alpha_f);\quad S(\alpha_i)\ \text{is the area}$$

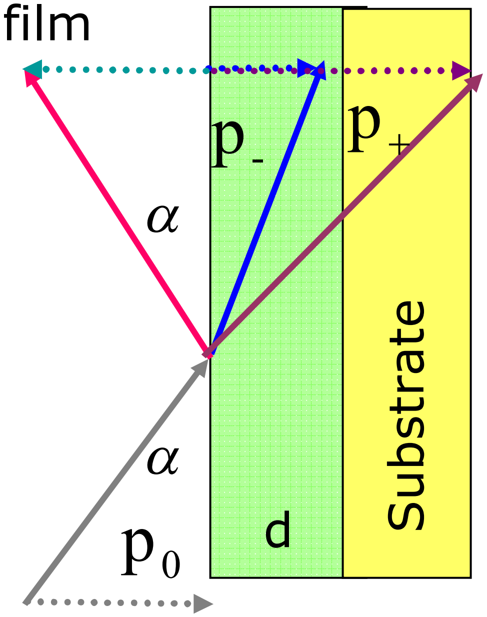
Birefringence in mean magnetic field

Magnetic (effective) film

Reflected

$$p_0 = k_0 \sin\alpha \;;$$

Incident



Two refracted (birefringence) wave numbers:

$$p_{\pm}^{2} = p_{0}^{2} - p_{c\pm}^{2}$$

$$p_{c\pm}^{2} = 4\pi(Nb_n \pm Nb_m)$$

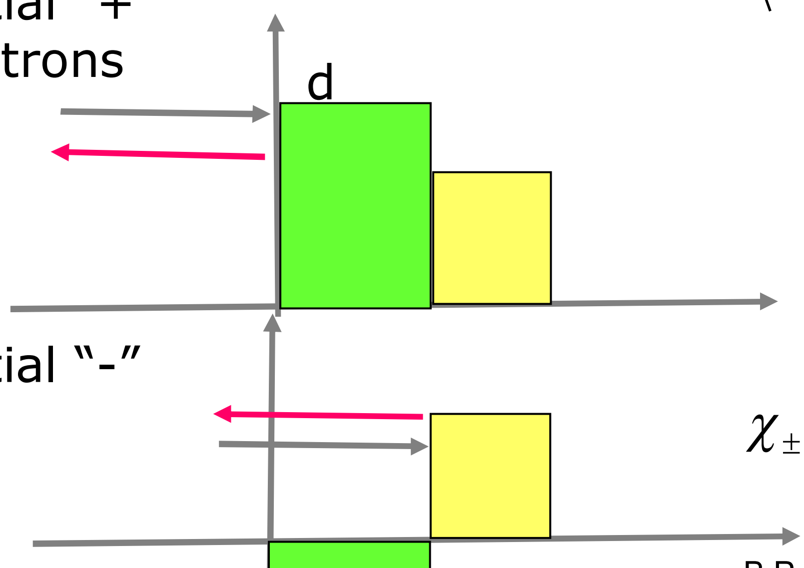
$$\sin\alpha_{c\pm} = \lambda\sqrt{\frac{Nb}{\pi} \pm \frac{m\mu B}{2\pi^{2}\hbar^{2}}} = \frac{\lambda}{\lambda_{c\pm}}$$

critical angle of total reflection

$B = 4\pi\langle M\rangle_{\mathrm{coh}}$ mean magnetization

Reflection potential "+" for "spin up" neutrons

Reflection potential "-" for spin "down"



$$\mathrm{R}_{\pm} = |\mathrm{R}_{\pm}|\exp(i\chi_{\pm}).$$

are eigenvalues of 2x2 reflectance matrix:

$$\hat{\mathrm{R}} = R(\hat{\sigma})$$

$\chi_{\pm}$ Phase shifts due to, e.g. film thickness d

General form for polarization dependency

$$\frac{\partial\sigma}{\partial\Omega} = \Sigma_0 + (\vec{\Sigma}_i \vec{P}_i) + (\vec{\Sigma}_f \vec{P}_f) + \Sigma_{if}{}^{\mu\nu} P_i^{\mu} P_f{}^{\nu},$$

where P^x, P^y, P^z are Cartesian projections of polarization vectors

$$\Sigma_0 = \frac{1}{4}\left[|R_+|^2 + |R_-|^2 \right]$$

$$\vec{\Sigma}_i = \vec{\Sigma}_f = \frac{\vec{b}}{4}\left[|R_+|^2 - |R_-|^2 \right]$$

$$\Sigma^{\mu\nu} = \Sigma_0 b^{\mu} b^{\nu} + \frac{1}{2}\mathrm{Re}\left(R_+{}^* R_-\right)\left(\delta^{\mu\nu} - b^{\mu} b^{\nu}\right) + \frac{1}{2}\mathrm{Im}\left(R_+{}^* R_-\right)\varepsilon^{\mu\nu\xi} b^{\varsigma},$$

where $\vec{b}$ is the unit vecto r in the direction of magnetic induction *averaged o ver the co herence ra nge*

SF and NSF reflection coefficients for ideal polarization

$$\mathbf{R}^{++} = \frac{1}{4}\left|R_+(1+\cos\gamma) + R_-(1-\cos\gamma)\right|^2$$

Non-spin-flip (NSF)

$$\mathbf{R}^{--} = \frac{1}{4}\left|R_+(1-\cos\gamma) + R_-(1+\cos\gamma)\right|^2$$

Non-spin-flip (NSF)

$$\mathbf{R}^{+-} = \mathbf{R}^{-+} = \frac{1}{2}|R_+ - R_-|^2 \sin^2\gamma$$

Spin-flip (SF)

From 1D PNR sense of the magnetization tilt cannot be determined!

Averaging over large domains:

$$\mathbf{R}^{++} = \frac{1}{4}\left[(|R_+|^2 + |R_-|^2)(1+\langle\cos^2\gamma\rangle) + 2(|R_+|^2 - |R_-|^2)\langle\cos\gamma\rangle + 2|R_+R_-|\cos\chi_{+-}\langle\sin^2\gamma\rangle\right]$$

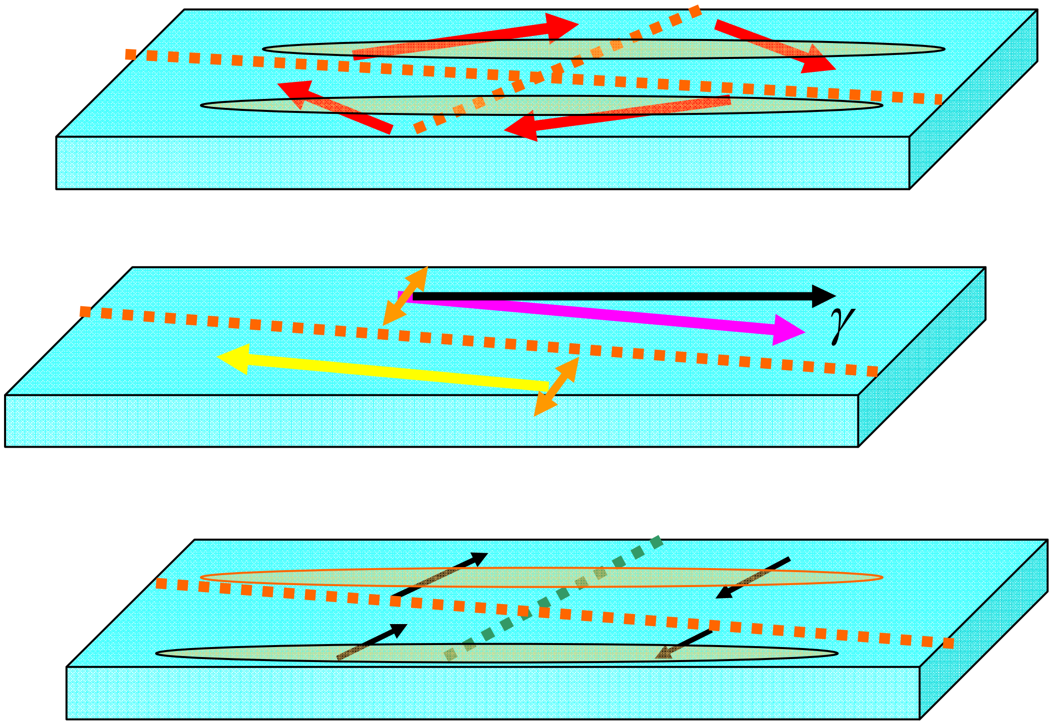
$$\mathbf{R}^{--} = \frac{1}{4}\left[(|R_+|^2 + |R_-|^2)(1+\langle\cos^2\gamma\rangle) - 2(|R_+|^2 - |R_-|^2)\langle\cos\gamma\rangle + 2|R_+R_-|\cos\chi_{+-}\langle\sin^2\gamma\rangle\right]$$

$$\mathbf{R}^{+-} = \mathbf{R}^{-+} = \frac{1}{2}\left[|R_+|^2 + |R_-|^2 - 2|R_+R_-|\cos\chi_{+-}\right]\langle\sin^2\gamma\rangle$$

Reflectivities are determined 2 functions: $|R_\pm| = |R(p_{c\pm})|$, phase shift $\chi_{+-} = \chi_+ - \chi_-$

and 2 constants: mean values $< \cos\gamma >$ and $\langle\sin^2\gamma\rangle = 1 - \langle\cos^2\gamma\rangle$

Example of domain structure

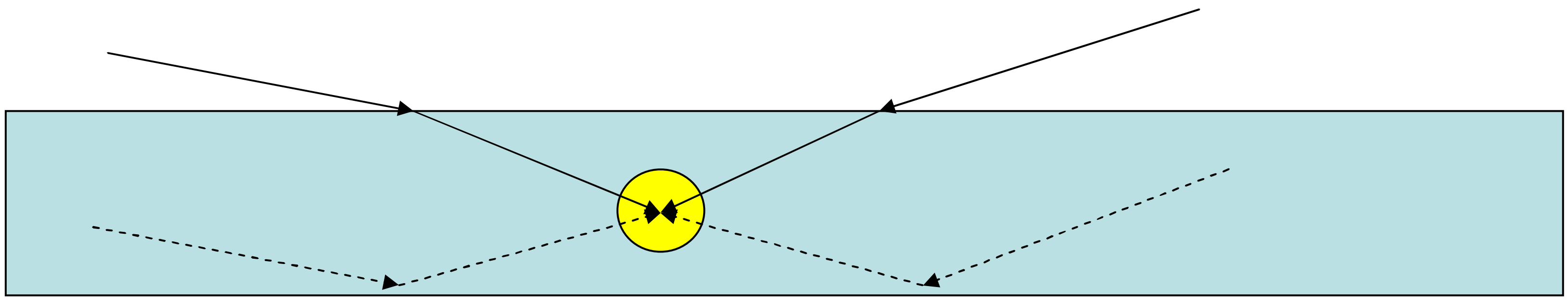


=

+ SF and NSF reflectionn

Off-specular

Scattering amplitude matrix in DWBA



The wave at the scatterer position $\left|\Psi_i(\vec r)\right\rangle = e^{i\vec\kappa_i\vec\rho}\hat S_i(z)\left|\Psi_{0i}\right\rangle$

is transmitted through the medium by the $\hat S = (\hat t e^{i\hat p z} + \hat r e^{-i\hat p z}) - matrix$

The scattering wave function $\left|\Psi_s(\vec r)\right\rangle = \dfrac{e^{ikr}}{r}\hat F\left|\Psi_i\right\rangle$

where $\hat F = F(\hat p_f, \hat p_i, \vec Q_\parallel) = -\dfrac{m}{2\pi\hbar^2}\int d\vec r e^{i\vec Q_\parallel\vec\rho}\hat S_f(z)\Delta\hat U(z,\vec\rho)\hat S_i(z)$

is the DWBA scattering amplitude matrix :

$\hat F = F(\hat{\vec\sigma}) = F_0 + (\vec F\hat{\vec\sigma})$

General structure of off-specular scattering cross section

$$\frac{\partial\sigma}{\partial\Omega} = \Sigma_0 + (\vec{\Sigma}_i \vec{P}_i) + (\vec{\Sigma}_f \vec{P}_f) + \Sigma_{if}{}^{\mu\nu} P_i^{\mu} P_f{}^{\nu},$$

is determined by one scalar, two vectors and one 3D tensor

$$\Sigma_0 = \frac{1}{2}\left\langle | F_0 |^2 + | \vec{F} |^2 \right\rangle$$

$$\vec{\Sigma}_i = \mathrm{Re}\left\langle F_0{}^{*} \vec{F} \right\rangle + \frac{1}{2}\mathrm{Im}\left\langle [\vec{F}^{*} \times \vec{F}] \right\rangle$$

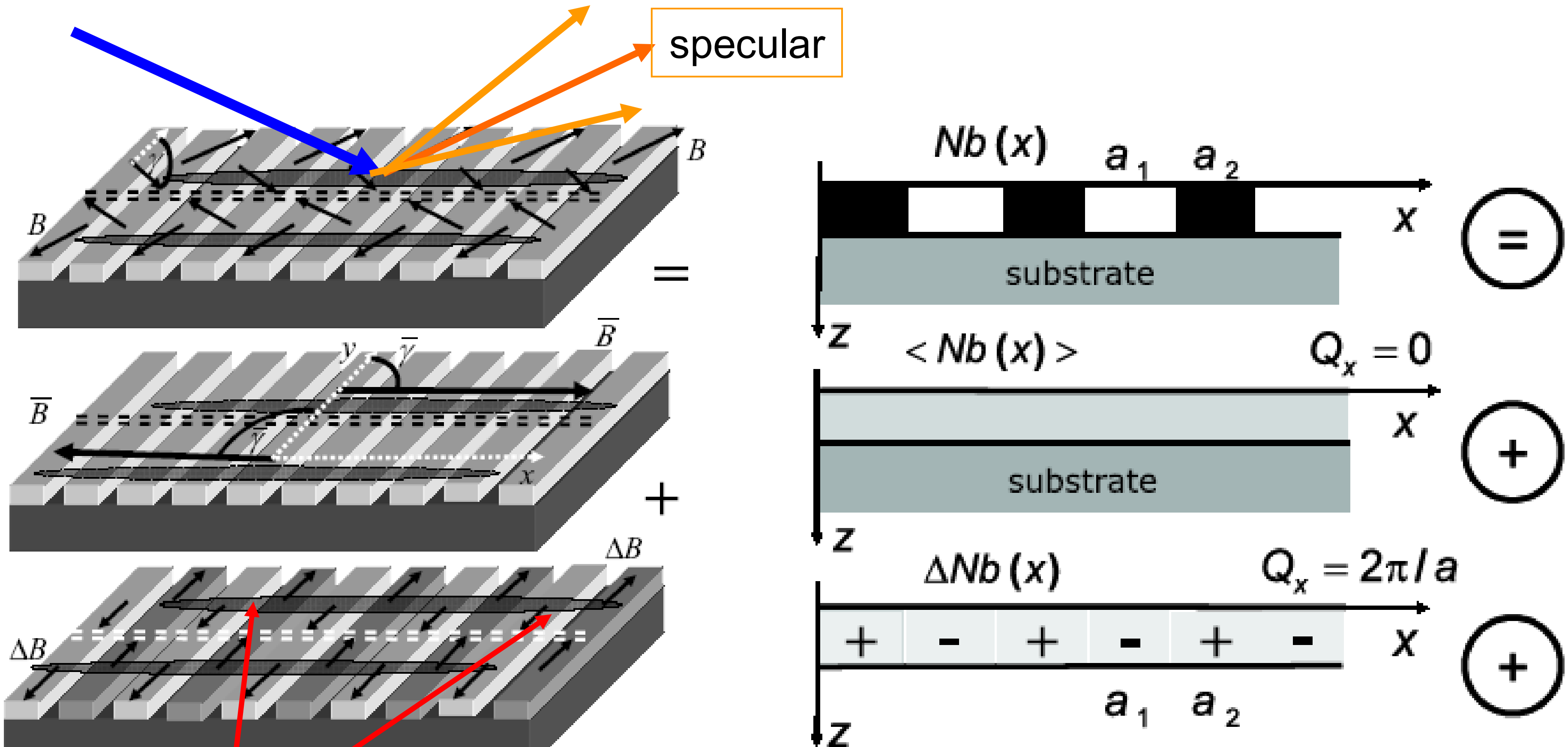
$$\vec{\Sigma}_f = \mathrm{Re}\left\langle F_0{}^{*} \vec{F} \right\rangle - \frac{1}{2}\mathrm{Im}\left\langle [\vec{F}^{*} \times \vec{F}] \right\rangle$$

$$\Sigma_{if}{}^{\mu\nu} = \frac{1}{2}\left\langle | F_0 |^2 - | \vec{F} |^2 \right\rangle \delta^{\mu\nu} + \mathrm{Re}\left\langle F^{\mu *} F^{\nu} \right\rangle - \frac{1}{2}\mathrm{Im}\left\langle F_0{}^{*} F^{\gamma} \right\rangle \varepsilon^{\mu\nu\gamma}$$

Reciprocity principle:

$$\left(\frac{d\sigma}{d\Omega}\right)_{if}^{+-} = \left(\frac{d\sigma}{d\Omega}\right)_{fi}^{-+}$$

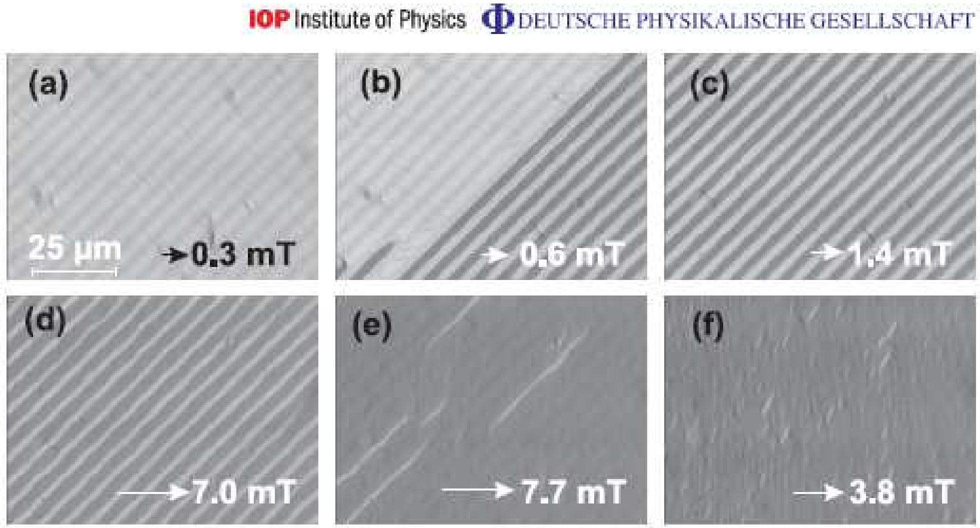
Logics of Distorted Wave Born Approximation (DWBA)

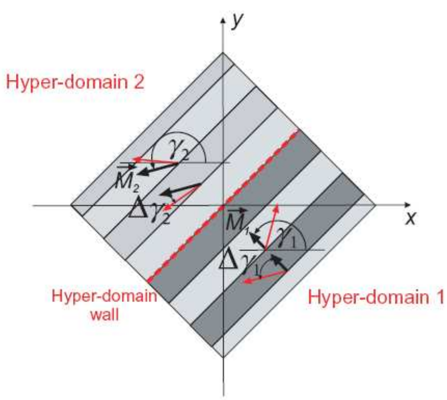


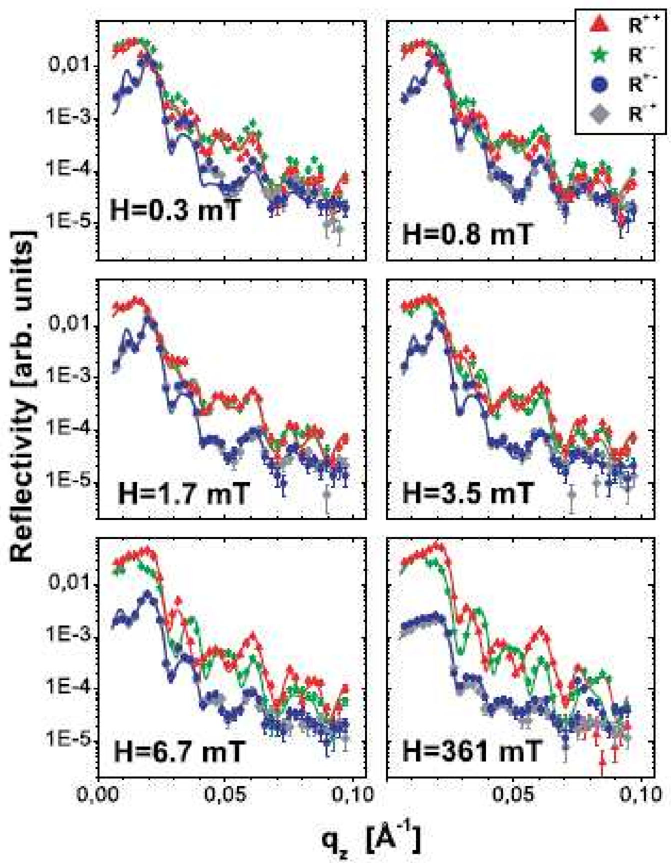
Coherent ellipsoids

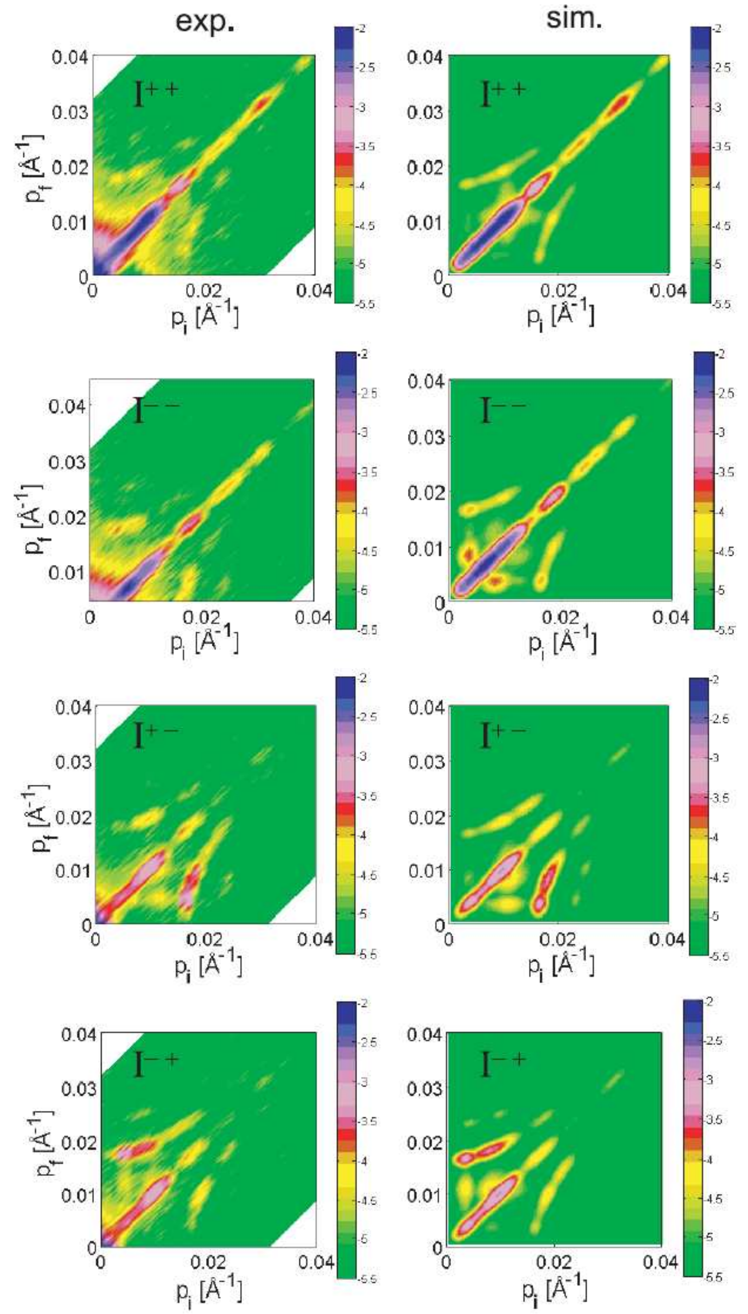
deviations from mean potential

$$\langle \Delta Nb \rangle_{coh} = 0$$







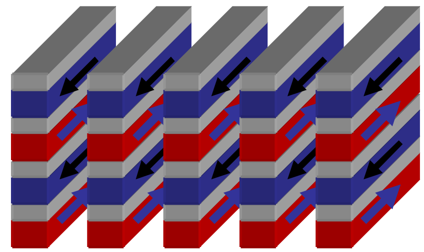


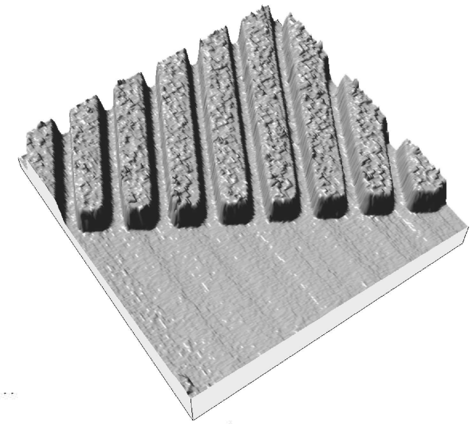
Exchange bias stripe & hyper Domains.
Phys. Rev. B73 (2006);
New Journ. Phys. (2008)

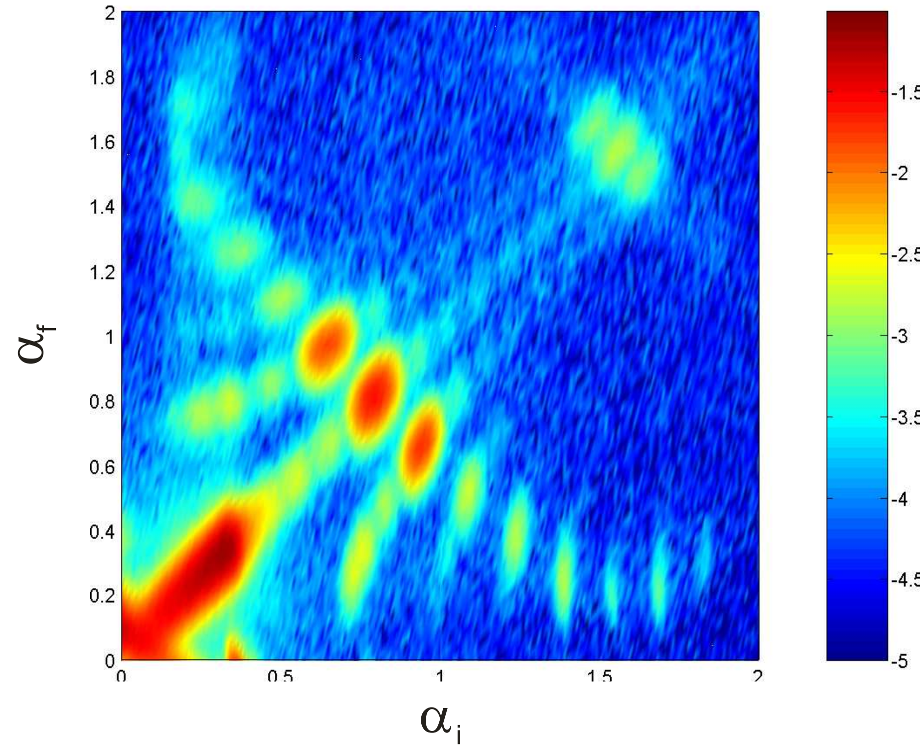
Reflectivity on striped multilayer

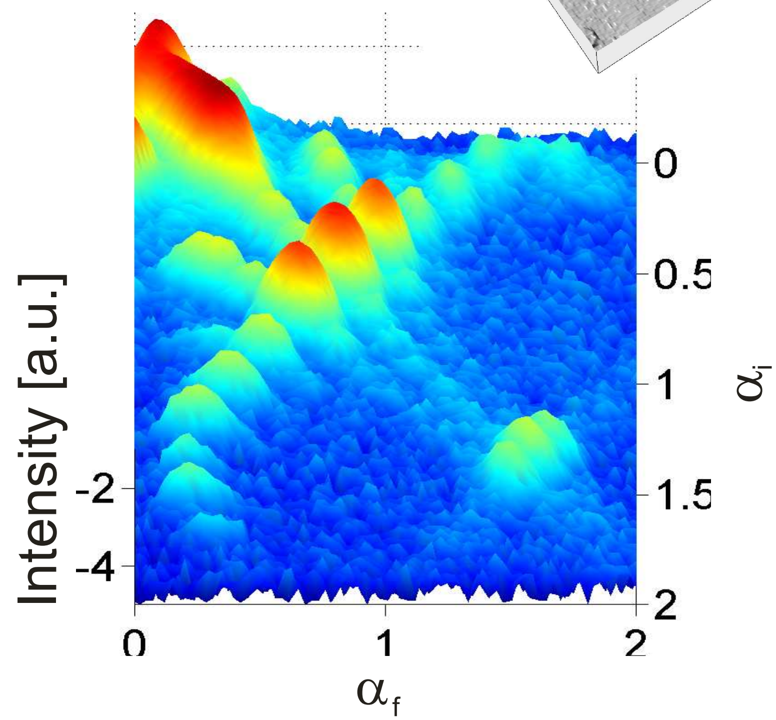
$[Cr/Co/Cr/Fe]_8$ Λ=16 nm

Stripe period: 6 μm









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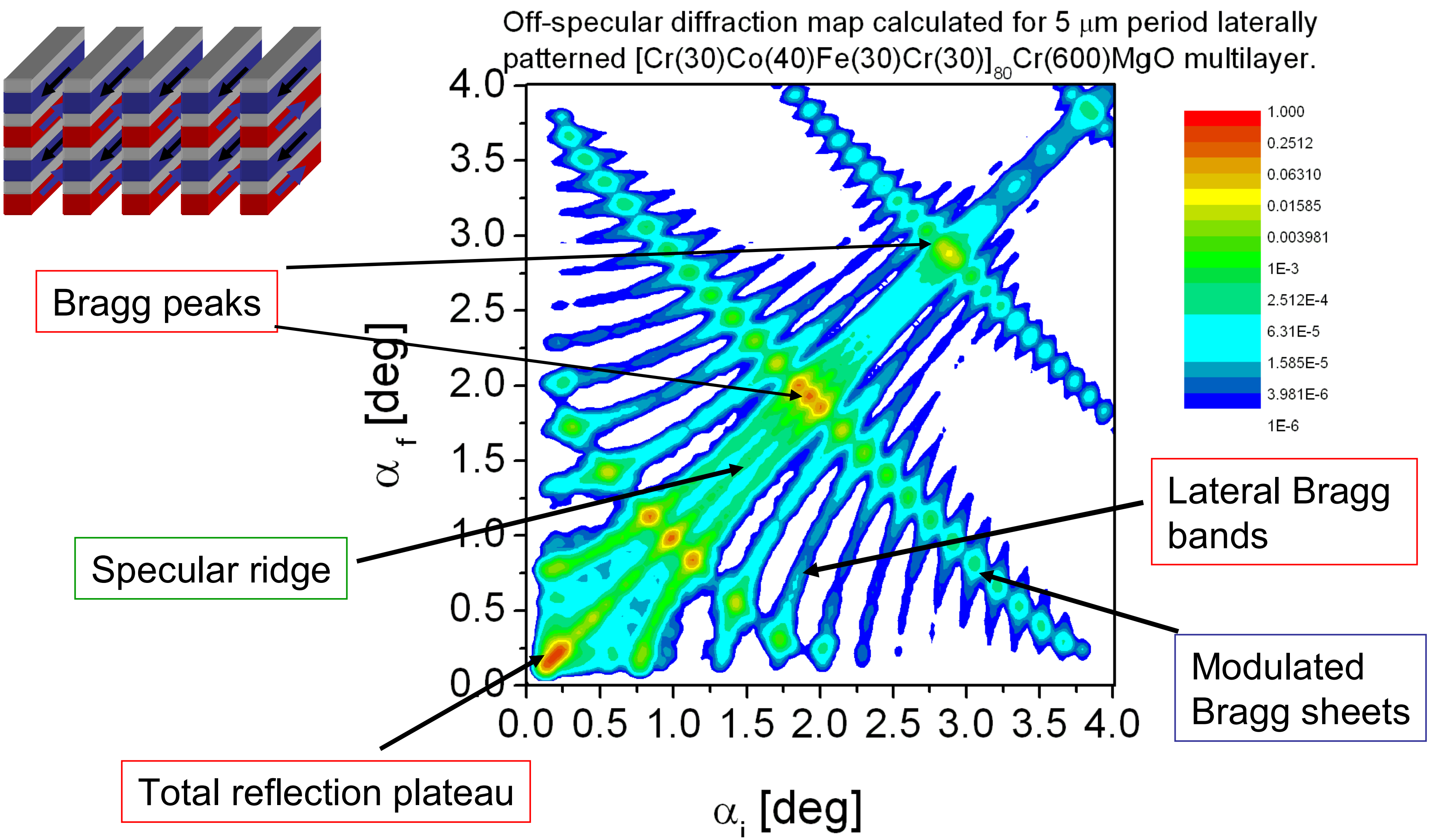
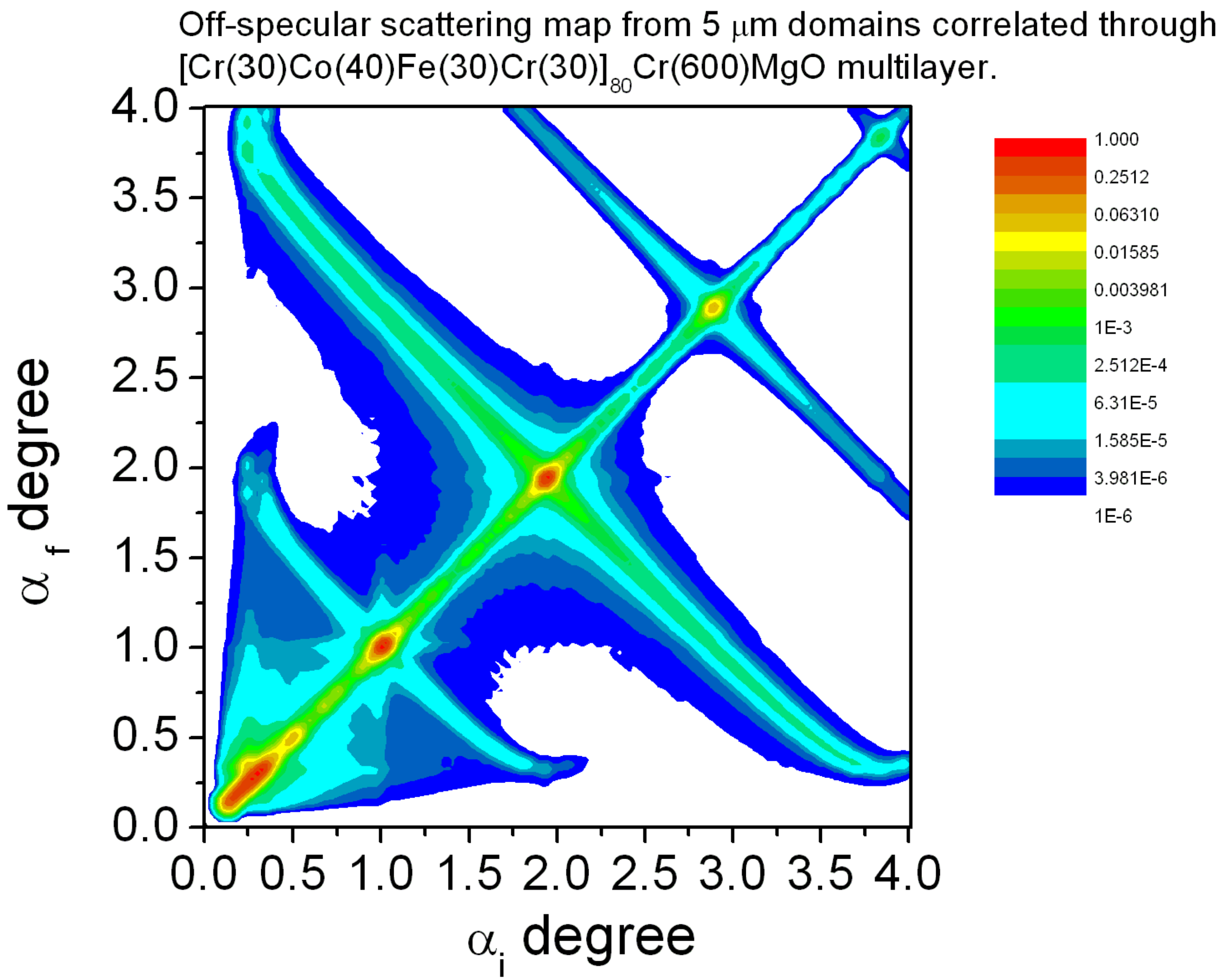
Off-specular scattering map for multilayered stripes



Off-specular scattering map for laterally random, but correlated through multilayer domains



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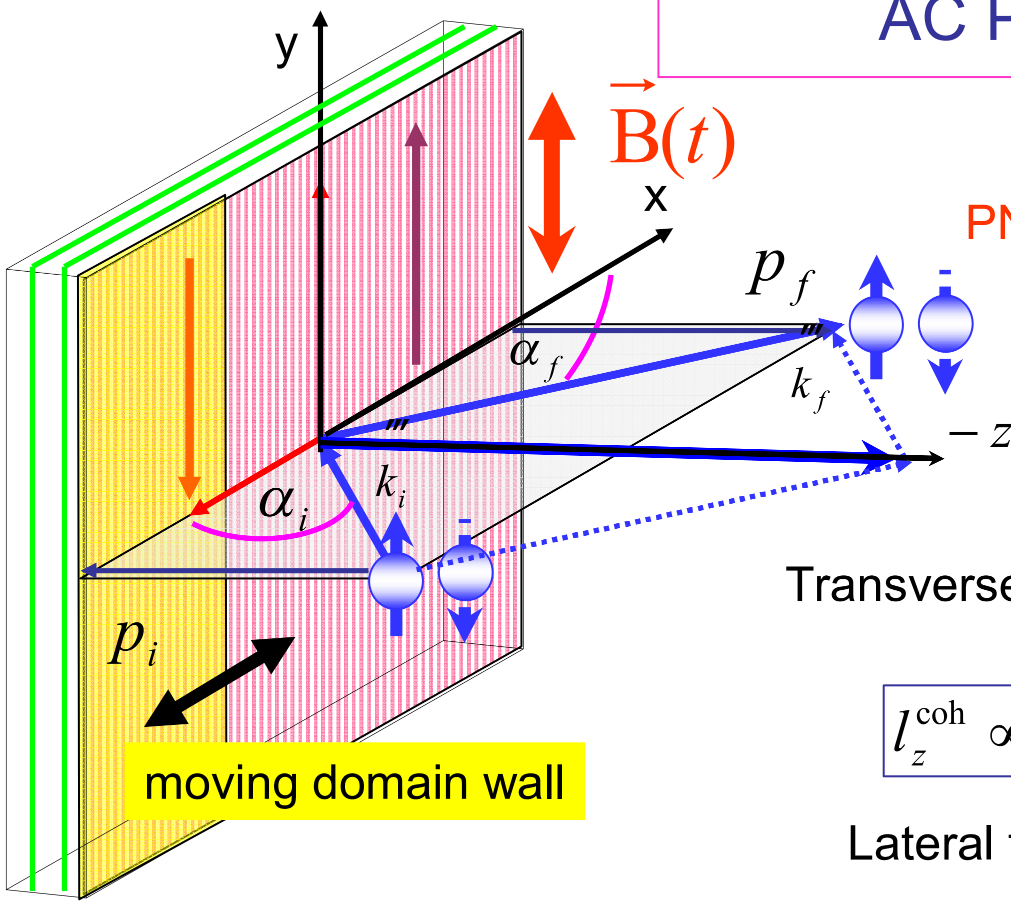
AC PNR – real time reflectometry



PNR signal is integrated over ω

2D Wave vector transfer:

$$\vec{Q} = \{Q^x, Q^z\}$$

Transverse component: $Q^z \approx \frac{2\pi}{\lambda}(\alpha_f + \alpha_i)$

$l_z^{coh} \propto \lambda / \Delta\alpha$ Transverse coherence length

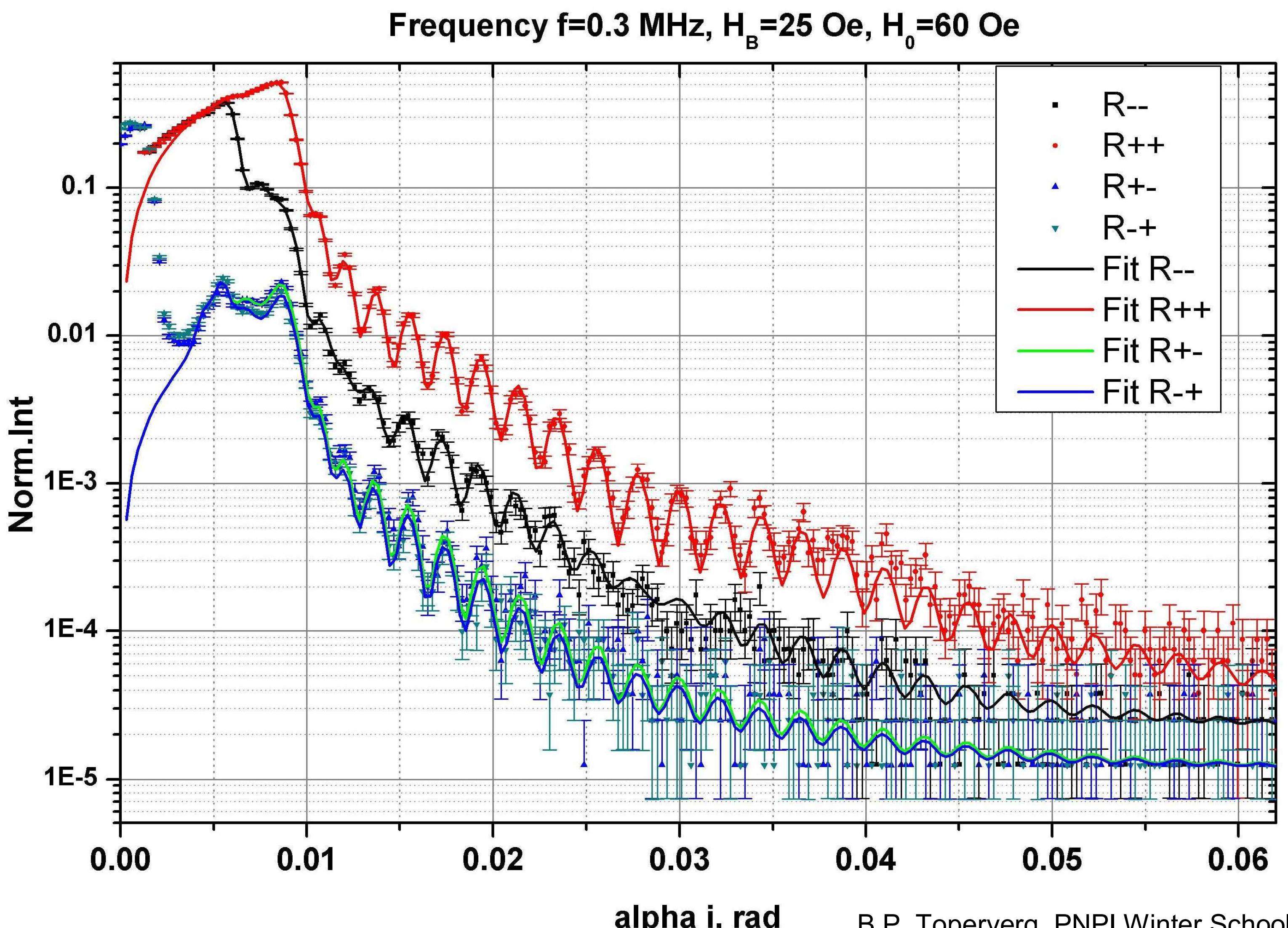
Lateral transfer: $Q^x \approx \frac{\pi}{\lambda}(\alpha_f^2 - \alpha_i^2) << Q^z$

$l_x^{coh} \propto \lambda / \alpha\Delta\alpha = l_z^{coh} / \alpha >> l_z^{coh}$ Lateral coherence length

specular reflection: $R = R(t)$

off-specular scattering: $Q^x \neq 0, \Rightarrow \alpha_f \neq \alpha_i$ from small domains, etc.

PNR from MBE grown 100 nm iron film in 60 Oe AC field
(Zhernenkov et al, ADAM, fall of 2008)



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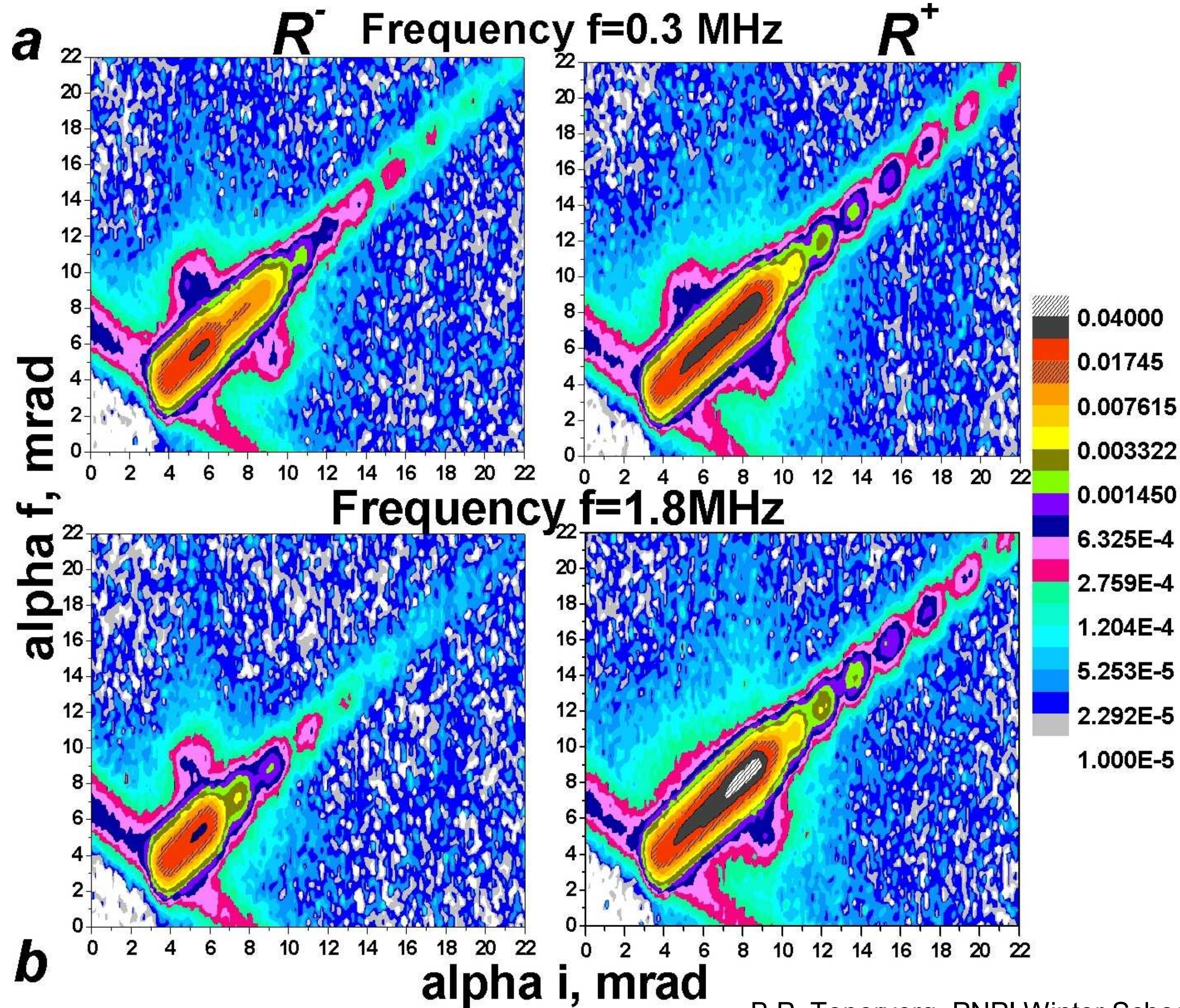
Specular reflection and off specular scattering from Fe film
(Zhernenkov et al, ADAM 2008)



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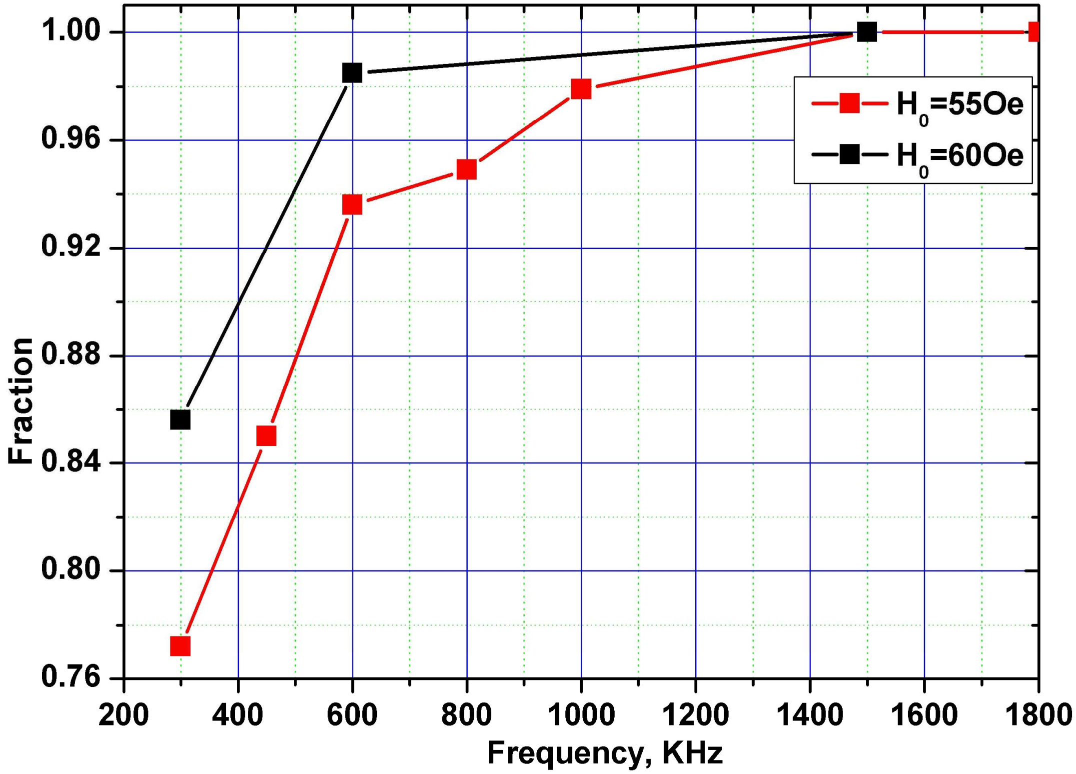
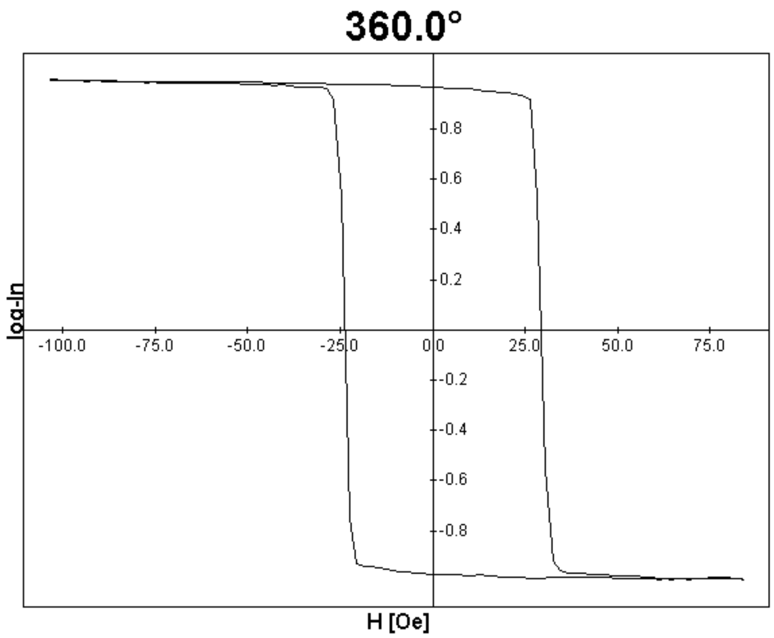
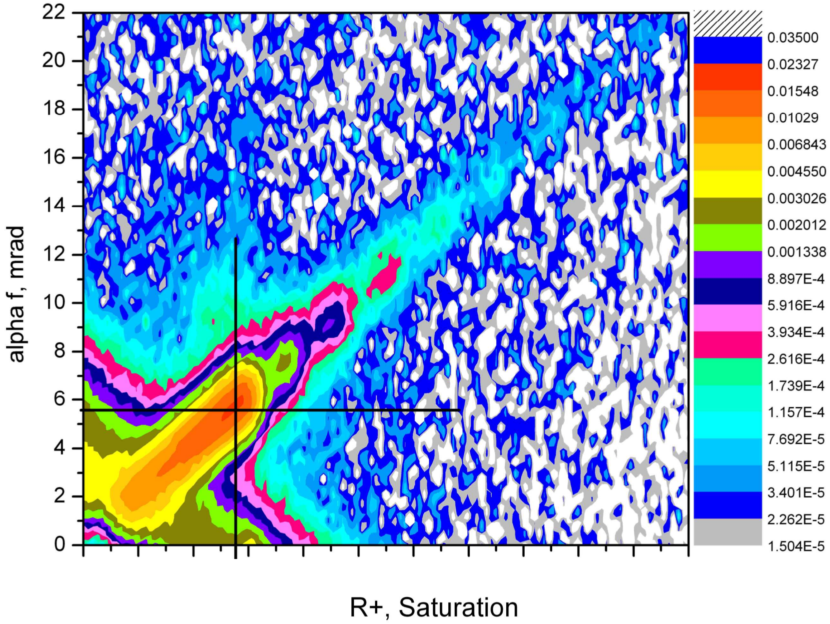
Fraction of non-flipped domains under AC and DC bias field

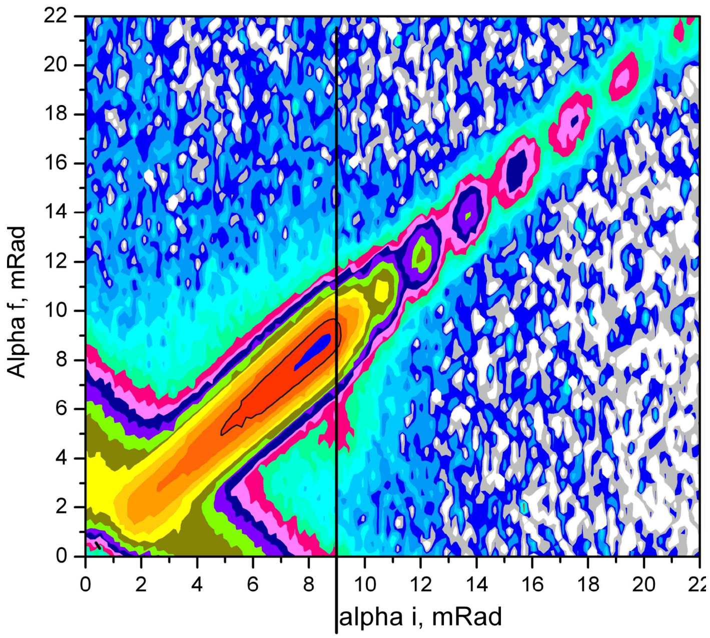


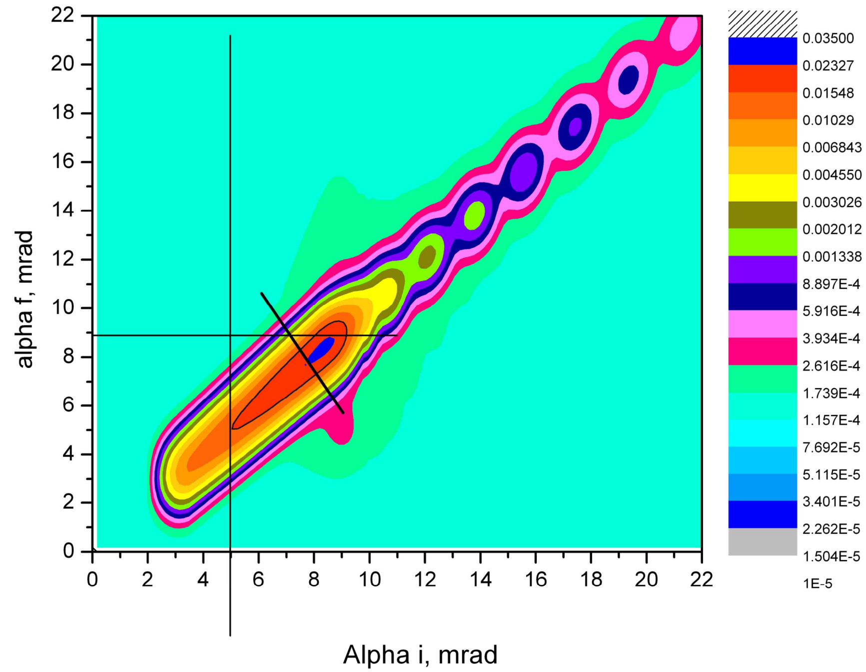
MOKE hysteresis



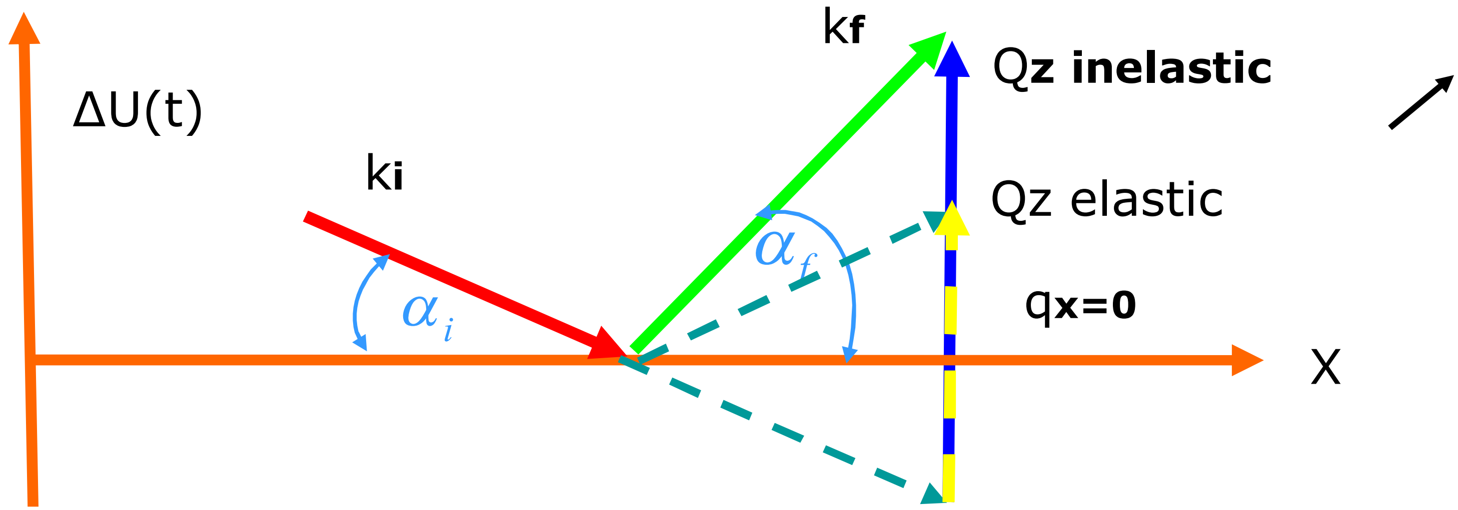


Specular reflection
and off-specular scattering
from 100 nm Fe film
in saturation

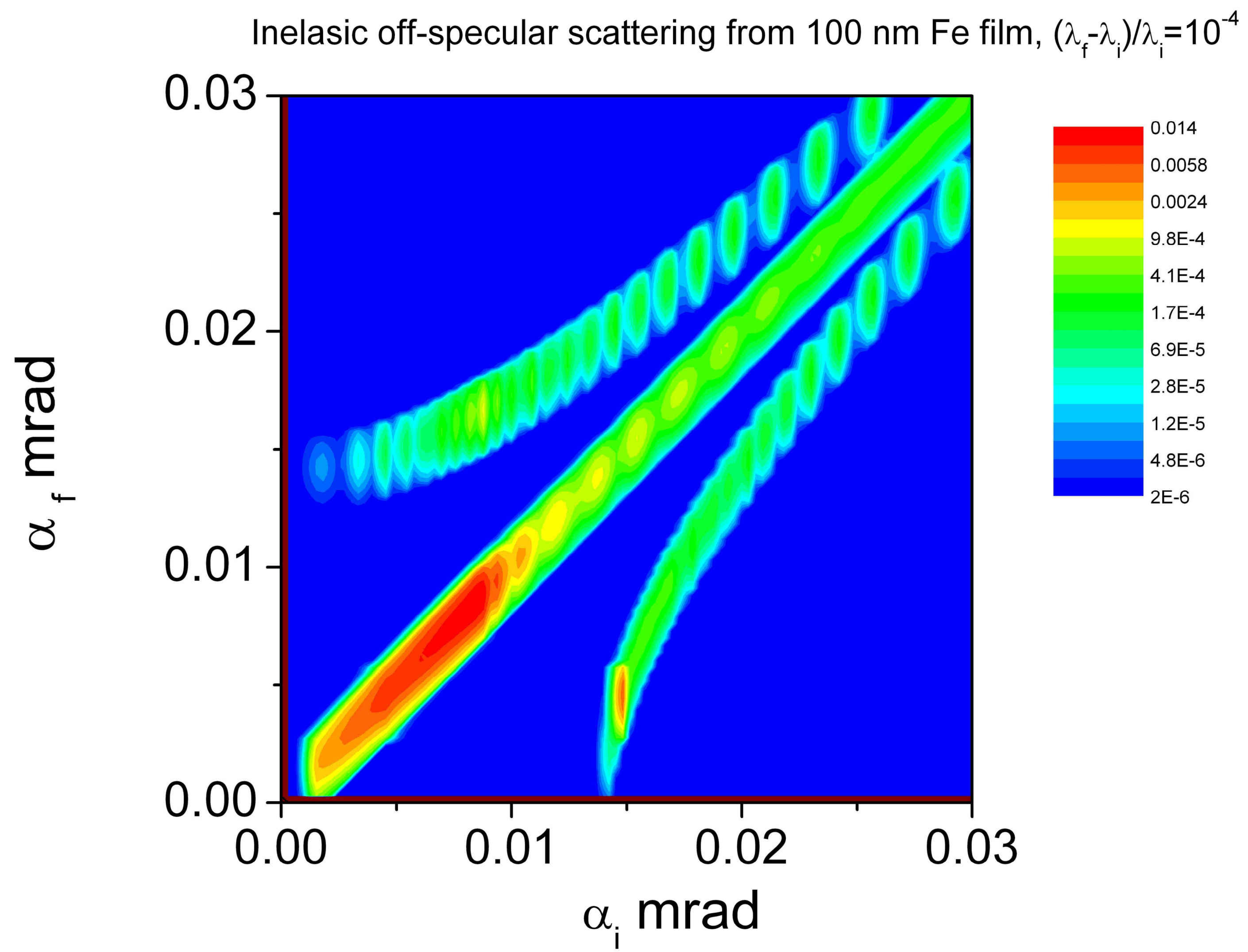




Inelastic off-specular reflection on dynamic homogeneous deviations $\Delta U(t)=U(t)-U$

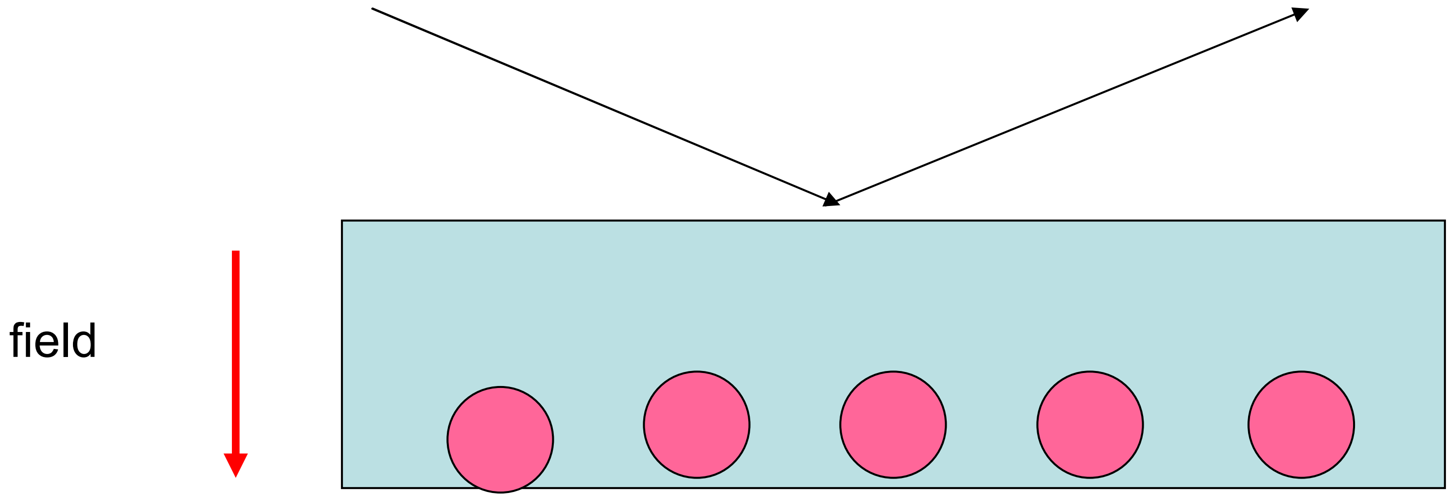


Dynamical fluctuations with size greater coherence length

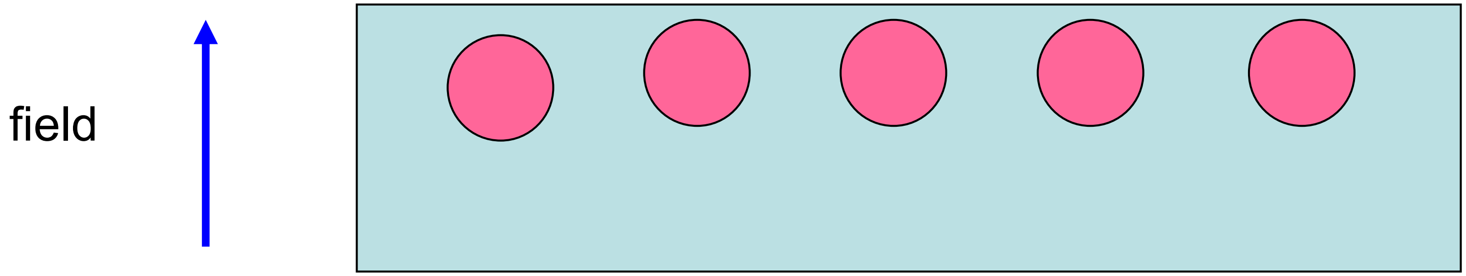


Inelasic off-specular scattering from 100 nm Fe film, $(\lambda_f-\lambda_i)/\lambda_i=10^{-4}$
0.03
0.02
0.01
0.00
α_f mrad
0.00
0.01
0.02
0.03
α_i mrad
0.014
0.0058
0.0024
9.8E-4
4.1E-4
1.7E-4
6.9E-5
2.8E-5
1.2E-5
4.8E-6
2E-6

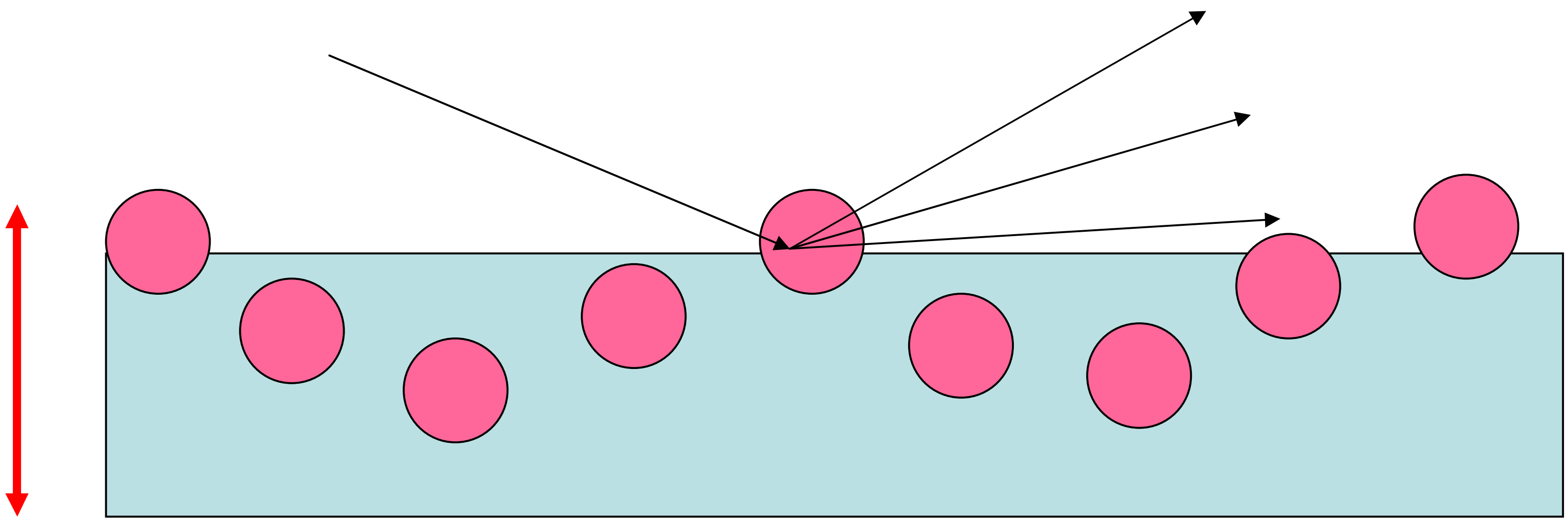
Adiabatic AC specular reflectometry



SLD depth profile variation via reflectivity: $R = R(q_z, \bar{t}, \Omega)$ $0 \leq \Omega \leq GHz$



Adiabatic AC off-speqular scattering



Lateral instabilities caused by AC transverse field

displacements $r^{\mu}(\bar{t}) \propto \int dt \chi^{\mu\nu}(\bar{t}-t)E^{\nu}(t)$

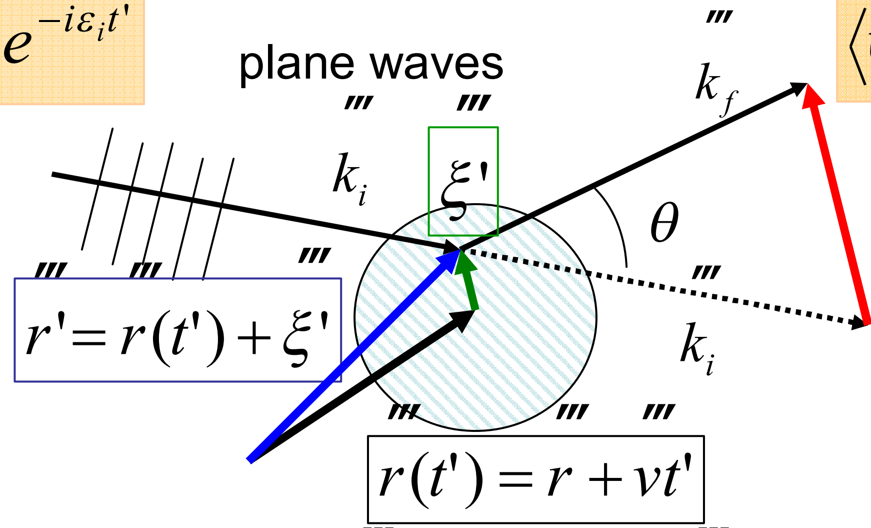
$\widetilde{S}(q_z, q_x, \bar{t}, \Omega)$ Is non-linear function of field $0 \leq \Omega \leq GHz$

Elementary scattering theory: amplitude

$$f(q,\omega) \propto \int dr' dt' \langle \psi_f(r',t') | U(r',t') | \psi_i(r',t') \rangle$$

$$|\psi_i(r',t')\rangle = e^{ik_i r'} e^{-i\varepsilon_i t'}$$

$$\langle \psi_i(r',t') | = e^{-ik_f r'} e^{i\varepsilon_i t'}$$



$$\omega = \varepsilon_f - \varepsilon_i$$

frequency transfer

$$q = k_f - k_i$$

wave vector transfer

$$f(q,\omega) \propto \int dt' e^{-i(\omega - vq)t'} \int d\xi' e^{iq(r+\xi')} U(\xi',t')$$

for hard sphere: $U(\xi',t') = U_0 \vartheta(|\xi'| - r_0)$

$$f(q) \propto \frac{[\sin(qr_0) - (qr_0)\cos(qr_0)]}{(qr_0)^3} e^{iqr} \delta(\omega - vq)$$

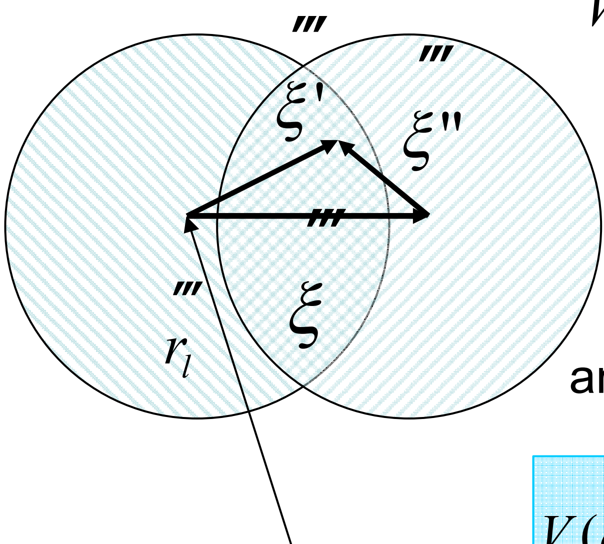
r_0 is radius of sphere

Hard sphere autocorrelator

$$\left|f_0(\vec{q})\right|^2 \propto \int d\vec{\xi}\,' d\vec{\xi}\,'' e^{i\vec{q}(\vec{\xi}\,'-\vec{\xi}\,'')} \vartheta(|\vec{\xi}\,'|-r_0)\vartheta(|\vec{\xi}\,''|-r_0)$$

$$\left|f_0(\vec{q})\right|^2 \propto V(\vec{q}) = \int d\vec{\xi} e^{i\vec{q}\vec{\xi}} V(\vec{\xi})$$ where

$$V(\vec{\xi}) \propto \int d\vec{\xi}\,' \vartheta(|\vec{\xi}\,'|-r_0)\vartheta(|\vec{\xi}\,'+\vec{\xi}|-r_0)$$



is volume of self-intersection:

$$V(\vec{\xi}) = V(0)\{1 - 2.5x + 2x^2 - 0.5x^3\}$$ with $$x = \frac{\xi}{2r_0}$$

and asymptotics $V(0) = \frac{4}{3}\pi r_0^3$ $\quad V(r_0) = 0$

$$V(\vec{q}) = \int d\vec{\xi} e^{i\vec{q}\vec{\xi}} V(\vec{\xi}) \propto \frac{[\sin(qr_0) - (qr_0)\cos(qr_0)]^2}{(qr_0)^6}$$

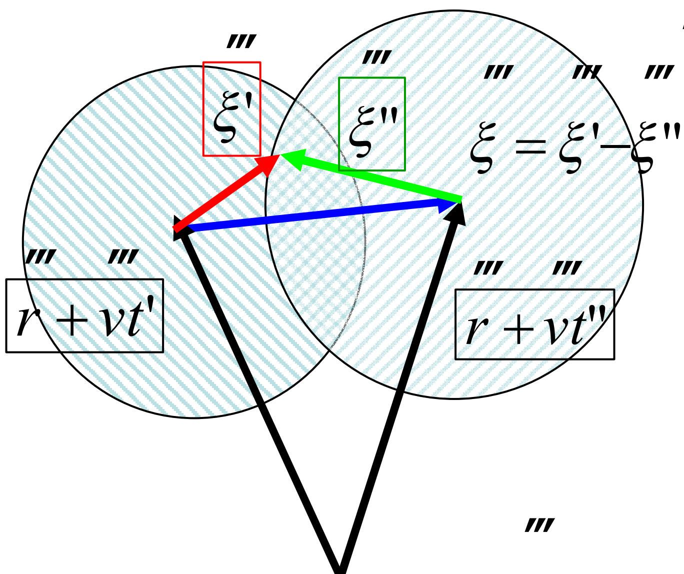
For many spheres $$\left|f(\vec{q})\right|^2 \propto \sum_{ll'} V(\vec{q}) e^{i\vec{q}(\vec{r}_l - \vec{r}_{l'})}$$

Scattering cross section

$$\frac{\partial^2\sigma}{\partial\omega\partial\Omega} = S(\vec{q},\omega) = \frac{1}{T}\left|f(\vec{q},\omega)\right|^2$$

$$S(\vec{q},\omega) \propto \frac{1}{T}\int dt'dt''e^{-i\omega(t'-t'')}\int d\vec{r}\,'d\vec{r}\,''e^{i\vec{q}(\vec{r}\,'-\vec{r}\,'')}U(\vec{\xi}\,',t')U(\vec{\xi}\,'',t'')$$

$$\vec{r}\,'=\vec{r}+\vec{v}t'+\vec{\xi}\,' \qquad \vec{r}\,''=\vec{r}+\vec{v}t''+\vec{\xi}\,''$$



$$\vec{r}\,'-\vec{r}\,''=\vec{\xi}+\vec{v}\tau \qquad \vec{\xi}=\vec{\xi}\,'-\vec{\xi}\,'' \qquad \tau=t'-t''$$

cross section via correlator:

$$S(\vec{q},\omega) \propto \int d\tau d\vec{\xi}e^{-i(\omega-\vec{v}\vec{q})\tau}e^{i\vec{q}\vec{\xi}}K(\vec{\xi},\tau)$$

$$K(\vec{\xi},\tau) = \frac{1}{T}\int dt'\int d\vec{\xi}\,'U(\vec{\xi}\,',t')U(\vec{\xi}\,'+\vec{\xi},t'+\tau)$$

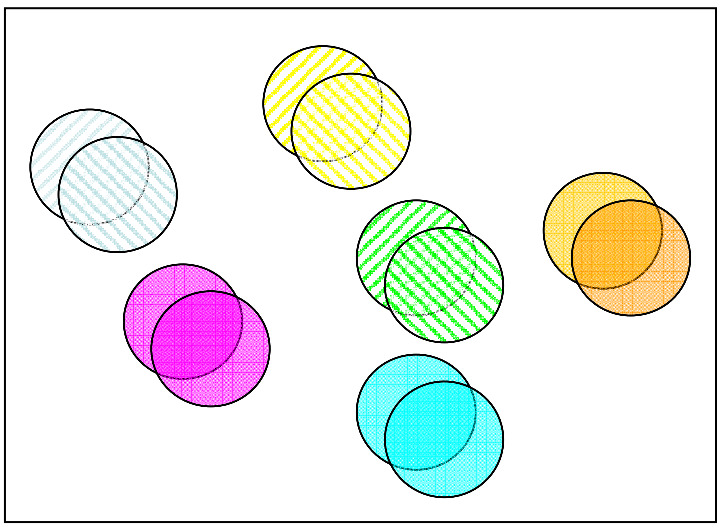
for hard sphere $K(\vec{\xi},\tau) \propto V(\vec{\xi})\vartheta(\tau_r-|\tau|)$ with $\tau_r = 2r_0/v$

$$\tilde{S}(\varpi,\vec{q}) \propto V(\vec{q})\int_{-\tau_r}^{\tau_r} d\tau e^{-i[\varpi-\vec{q}\vec{v}_l]\tau} = 2\pi V(\vec{q})\Delta[\varpi-\vec{q}\vec{v}_l]$$

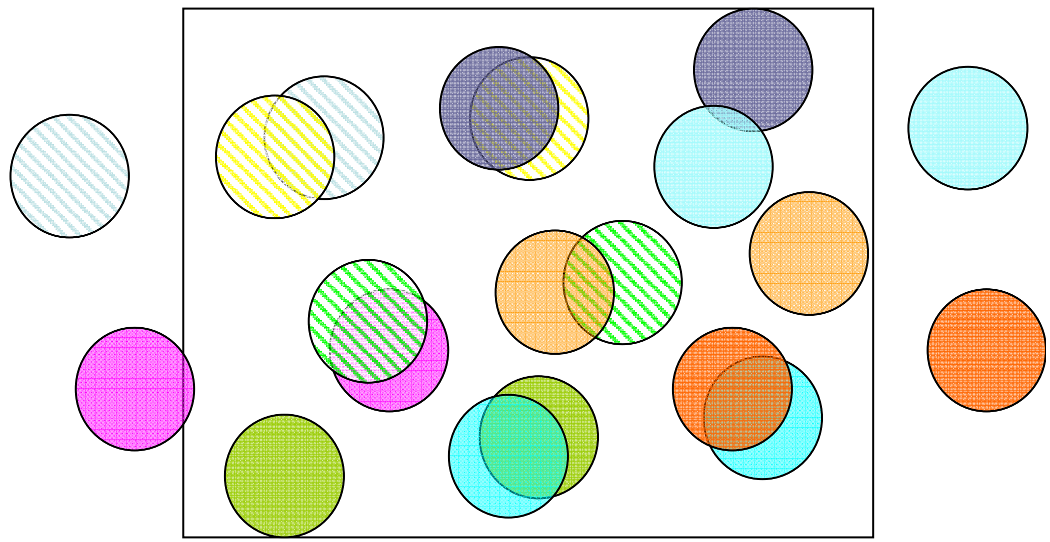
$$\Delta[\varpi-\vec{q}\vec{v}_l] \Rightarrow \delta(\varpi-\vec{q}\vec{v}_l)$$

• Scattering from Brownian diffusion

short time: autocorrelations



long time: hetero-correlations



coherence volume

$$\tilde{S}(\overline{\omega},\vec{q};\bar{t}_l)=\int d\tau e^{-i\overline{\omega}\tau}<e^{i\vec{q}(\vec{R}_l'-\vec{R}_l'')}>F(\vec{q})$$

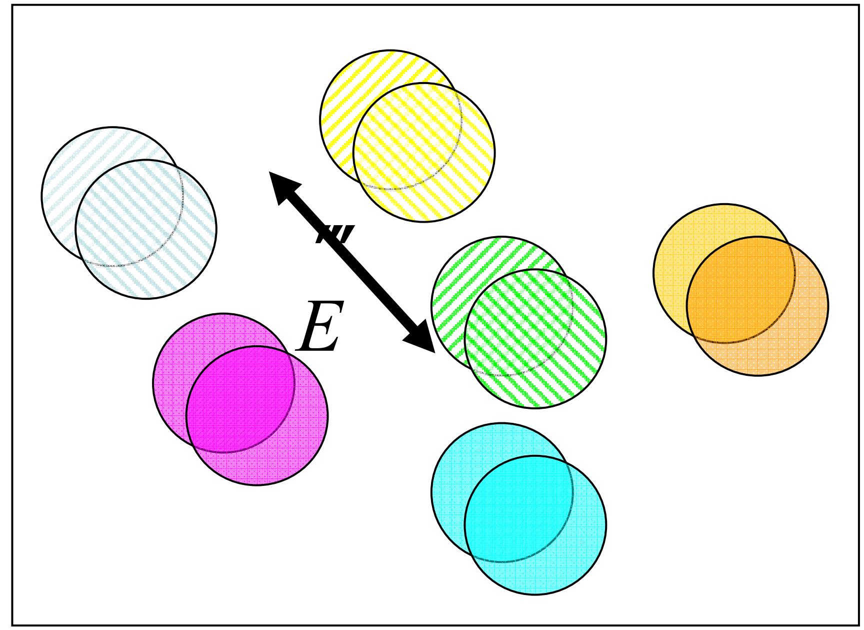
after long random walk $$<e^{i\vec{q}(\vec{R}_l'-\vec{R}_l'')}>\sim e^{-Dq^2|\tau|}$$ and

$$\tilde{S}(\overline{\omega},\vec{q};\bar{t}_l)\propto F(q)\frac{Dq^2}{\overline{\omega}^2+D^2q^4}$$

with $\overline{\omega}\sim Dq^2<<vq$

Scattering from particles in small oscillating field

$$\tilde{S}(\overline{\omega}, q; \bar{t}_l) = \int d\tau e^{-i\overline{\omega}\tau} e^{iqa_0 \cos\Omega\tau} F(q)$$

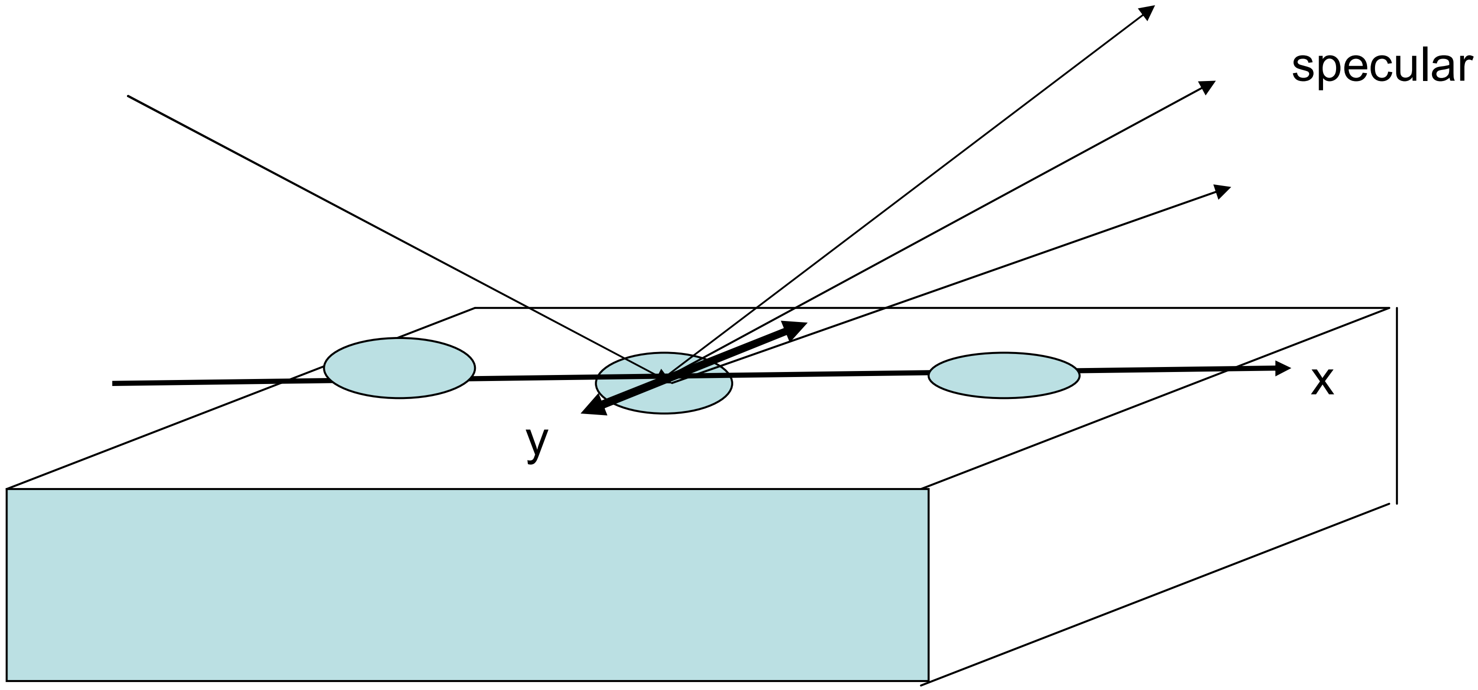


$a \le r_0$ is amplitude of particle displacements

if $qa << 1$

$$\begin{aligned} &\tilde{S}(\overline{\omega}, q; \bar{t}_l) - \tilde{S}(0, q; \bar{t}_l) \\ &\propto F(q)\int d\tau e^{-i\overline{\omega}\tau}(qa_0)[i\cos(\Omega\tau) - \frac{1}{2}\cos^2(\Omega\tau) + ...] \\ &\propto (qa)[\delta(\overline{\omega} - \Omega) + \delta(\overline{\omega} + \Omega)] + (qa_0)^2\delta(\overline{\omega} - 2\Omega) + ... \end{aligned}$$

AC field induced off specular scattering



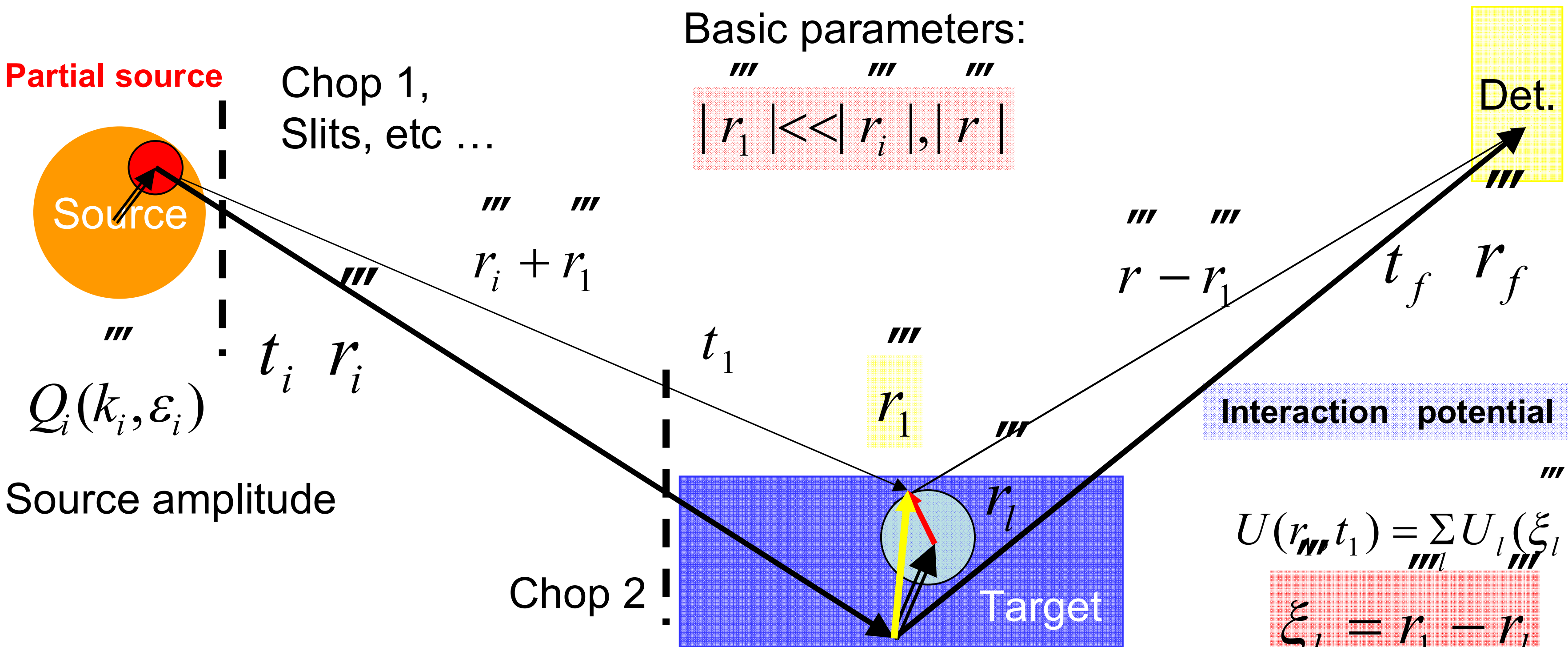
$$< \tilde{S}'''(\bar{\omega}, q; \bar{t}_l) - \tilde{S}'''(0, q; \bar{t}_l) >_{q_y, \bar{\omega}}$$

$$\propto < (q_y a_0)^2 > [\delta(\alpha_f{}^2 - \alpha_f{}^2 - 4\Omega / \omega_i) + \delta(\alpha_f{}^2 - \alpha_f{}^2 + 4\Omega / \omega_i).]$$

$$\alpha_i{}^2 \sim \alpha_f{}^2 \sim 10^{-8}$$

$$\Omega \sim GHz$$

Scattering kinematics: far-field-limit



Interaction potential

$$U(\vec{r}_1, t_1) = \sum_l U_l(\vec{\xi}_l, t_1)$$

$$\vec{\xi}_l = \vec{r}_1 - \vec{r}_l$$

Incident unit vector

$$\vec{n}_i = \vec{r}_i / |\vec{r}_i|$$

Final unit vector

$$\vec{n}_f = \vec{r}_f / |\vec{r}_f|$$

Basic approximations

$$|\vec{r}_i + \vec{r}_1| \approx r_i + \vec{r}_1\vec{n}_i - (\vec{r}_1\vec{n}_{i\perp})^2 / 2r_i \ldots$$

$$|\vec{r} - \vec{r}_1| \approx r - \vec{r}_1\vec{n}_f + (\vec{r}_1\vec{n}_{f\perp})^2 / 2r_f$$

Incident wave function:

$$\Psi_i(\vec{r}_1, t_1) = \int d\varepsilon_i G(\vec{r}_1 + \vec{r}_i, \varepsilon_i) e^{-i\varepsilon_i (t_1 - t_i)} Q_i(\vec{k}_i, \varepsilon_i)$$

is a superposition of plane waves with spherical wave amplitudes:

$$\Psi_i(\vec{r}_1, t_1) \propto \int d\varepsilon_i \frac{e^{ik_i r_i}}{r_i} e^{i\vec{k}_i \vec{r}_1} e^{-i\varepsilon_i (t_1 - t_i)} Q_i(\vec{k}_i, \varepsilon_i)$$

Finally scattered w.f. is:

$$\Psi_f(\vec{r}, t) \propto \frac{1}{r_i r_f} \int d\varepsilon_f d\varepsilon_i e^{i(k_f r - \varepsilon_f t)} U(\vec{q}, \omega) e^{i(k_i r_i - \varepsilon_i t_i)} Q(\vec{k}_i, \varepsilon_i)$$

with scattering amplitude

$$U(\vec{q}, \omega) = \int d\vec{r}_1 dt_1 e^{-i(\vec{k}_f - \vec{k}_i)\vec{r}_1} e^{i(\varepsilon_f - \varepsilon_i) t_1} U(\vec{r}_1, t_1)$$

Fourier transform in space and time

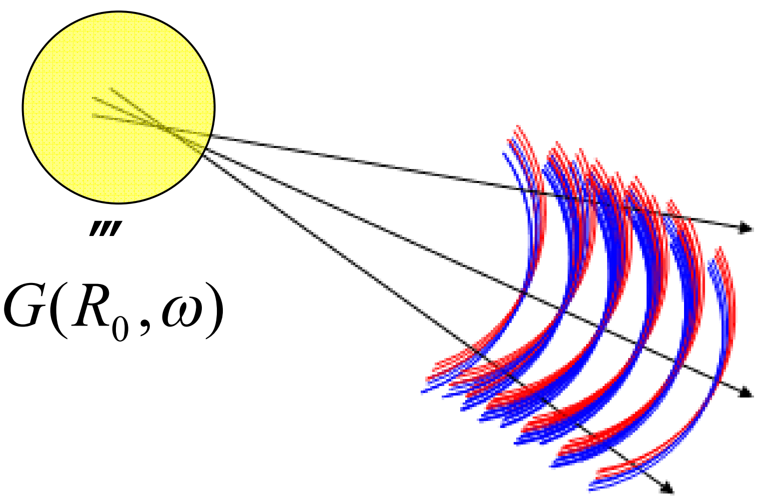
Incident wave field:

$$\psi_i(\vec{R}_i, t_l') = \int d\tau_s' d\omega_i' G(\vec{R}_i, \omega_i) e^{-i\omega_i(t_l' - t_s' + \tau_s')} Q_i(\tau_s')$$

is a superposition

$$\psi_i(\vec{R}_i, t_l) = \int d\omega_i G(\vec{R}_i, \omega_i) e^{-i\omega_i(t_l - t_s)} Q_i(\omega_i - \omega_0) \vartheta(t_l' - t_s')$$

of spherical waves



$$G(\vec{R}_i, \omega) \propto \frac{e^{ik_i|\vec{R}_0 + \vec{r}_l' - \vec{r}_s'|}}{R_0} \approx e^{i\vec{k}_i(\vec{r}_l' - \vec{r}_s')} G(\vec{R}_0, \omega)$$

approximated with plane waves with spherical wave amplitude

$$\vec{k}_i = \frac{\vec{R}_0}{R_0} k_i$$ is incident wave vector

Link back to direct space

Probability flux density:

$$\vec{J}(\vec{r},t) = \frac{i\hbar}{2m}\{(\nabla\Psi^*)\Psi - \Psi^*(\nabla\Psi)\} = \frac{i\hbar}{2m}\lim_{r\to r'}(\nabla_r - \nabla_{r'})\Psi^*(r)\Psi(r')$$

where

$$\Psi_f(\vec{r},t) = \int d\vec{r}_1 \int dt_1 G(\vec{r}-\vec{r}_1, t-t_1) Q_s(\vec{r}_1, t_1)$$

with

$$Q_s(\vec{r}_1, t_1) = U(\vec{r}_1, t_1)\Psi_i(\vec{r}_1, t_1)$$

is the secondary source amplitude and

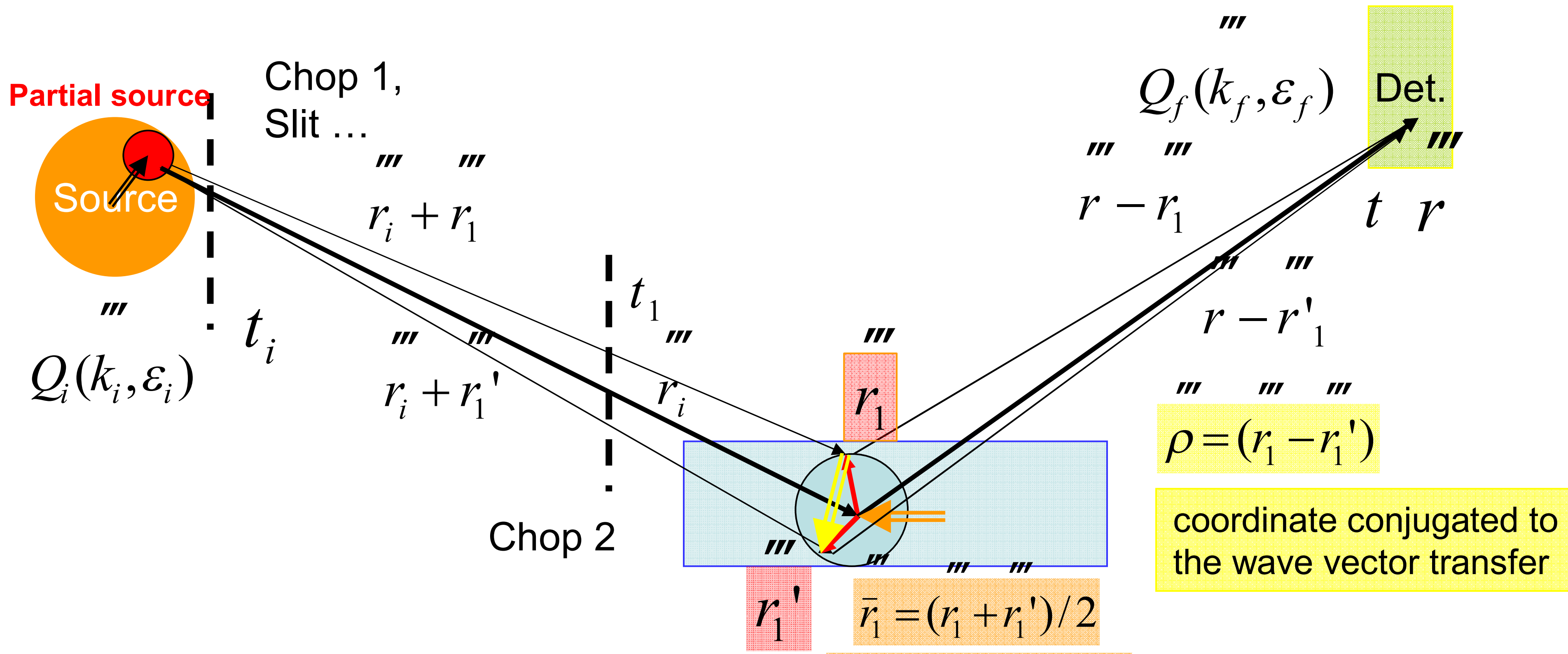
$$\Psi_f(r_f, t) \propto \int d\vec{r}_1 \int dt_1 \int d\varepsilon_f e^{i\Phi_f(|\vec{r}_f - \vec{r}_1|, t-t_1)} Q_s(\vec{r}_1, t_1)$$

with

the phase: $\Phi_f = k_f |\vec{r} - \vec{r}_1| - \varepsilon_f (t - t_1) >> 1$ generally very large

$$|\Psi_f(\vec{r},t)|^2 \propto \int d\vec{r}_1 d\vec{r}_1' dt_1 dt_1' \int d\varepsilon_f d\varepsilon_f' e^{i(\Phi_f - \Phi_f')} Q_s^*(\vec{r}_1, t_1) Q_s(\vec{r}_1', t_1')$$

Wave field correlation range



Mean incident wave vector

$$\kappa_i = (k_i + k_i')/2$$

$n_i = r_i / |r_i|$ Mean incident direction

Mean final wave vector

$$\kappa_f = (k_f + k_f')/2$$

$n_f = r / |r|$ Mean outcome direction

Group velocity and neutron path

the principle contribution into the integral: $\int d\varepsilon_f d\varepsilon_f' e^{i(\Phi_f-\Phi_f')}$ is due to minimum of

$$\Delta\Phi = \Phi_f - \Phi_f' = [k_f\,|\vec{r} - \vec{r}_1| - k_f'\,|\vec{r} - \vec{r}_1'|] - [\varepsilon_f(t-t_1) - \varepsilon_f'(t-t_1')] \le 1$$

Introducing variables:

“moment” of time and “coordinate” of interaction: $\bar{t}_1 = (t_1 + t_1')/2$ and $\bar{\vec{r}}_1 = (\vec{r}_1 + \vec{r}_1')/2$

mean neutron energy and wave vector: $\bar{\varepsilon}_f = (\varepsilon_f + \varepsilon_f')/2$ and $\vec{\kappa}_f = (\vec{k}_f + \vec{k}_f')/2$

One approximates the phase shift $\Delta\Phi \approx \tilde{\omega}_f[(t-\bar{t}_1) - |\vec{r} - \bar{\vec{r}}_1|/\bar{v}_f] - [\bar{\varepsilon}_f\tau - \vec{\kappa}_f\vec{\rho}] \le 1$

where $\bar{v}_f = \bar{\kappa}_f/m$ is group velocity, and $\tau = (t_1 - t_1')$ $\vec{\rho} = (\vec{r}_1 - \vec{r}_1')$

Integration over $\tilde{\omega}_f = (\varepsilon_f - \varepsilon_f')$ finally yields:

$$\int d\bar{\varepsilon}_f d\tilde{\omega}_f e^{i\Delta\Phi(\bar{\varepsilon}_f,\tilde{\omega}_f)} = 2\pi\int d\bar{\varepsilon}_f e^{i(\bar{\varepsilon}_f\tau - \vec{\kappa}_f\vec{\rho})}\delta[(t-\bar{t}_1) - \left|\vec{r} - \bar{\vec{r}}_1\right|/\bar{v}_f]$$

Tracing along neutron path

$$G_f^{\,*} G_f \propto \int d\bar{\omega}_f e^{-i(\bar{\omega}_f \tau - \vec{\kappa}_f \vec{\rho})} \int d\tilde{\omega}_f e^{i\tilde{\omega}_f (t - \bar{t}_1 - \bar{s}_f / \bar{v}_f)}$$

$$= \int d\bar{\omega}_f \, 2\pi\delta(t - \bar{t}_1 - \bar{s}_f / \bar{v}_f) e^{-i(\bar{\omega}_f \tau - \vec{\kappa}_f \vec{\rho})}$$ Outgoing trace

$$G_i^{\,*} G_i \propto \int d\bar{\omega}_i \delta(t - \bar{t}_1 - \bar{s}_i / \bar{v}_i) e^{i(\bar{\omega}_i \tau - \vec{\kappa}_i \vec{\rho})}$$ Incoming trace

$\bar{s}_i = |\vec{r}_i + \vec{\bar{r}}_1| \approx r_i$ is flight path from source, $\bar{s}_f = |\vec{r} - \vec{\bar{r}}_1| \approx r$ is flight path from the target

$$J_i(\vec{r},t) \propto \int d\vec{\bar{r}}_1 d\bar{t}_1 d\bar{\omega}_f d\bar{\omega}_i \delta(t - \bar{t}_1 - \bar{s}_f / \bar{v}_f) \tilde{S}(\bar{\omega}, \vec{\bar{q}}; \vec{\bar{r}}_1, \bar{t}_1) \delta(\bar{t}_1 - \bar{t}_i - \bar{s}_i / \bar{v}_i) |Q_i(\vec{\kappa}_i, \omega_i)|^2$$

with $$\tilde{S}(\bar{\omega}, \vec{\bar{q}}; \vec{\bar{r}}_1, \bar{t}_1) = \int d\vec{\rho} d\tau e^{-i(\bar{\omega}\tau - \vec{\bar{q}}\vec{\rho})} U(\vec{\bar{r}}_1 + \frac{\vec{\rho}}{2}, \bar{t}_1 + \frac{\tau}{2}) U(\vec{\bar{r}}_1 - \frac{\vec{\rho}}{2}, \bar{t}_1 - \frac{\tau}{2})$$

After integration over $\bar{\omega}_f$ and $\bar{\omega}_i$ at fixed $\bar{t}_i$ and $\bar{t}_1 - \bar{t}_i$

$$J_i(\vec{r},t) \propto \langle \tilde{S}_i(\bar{\omega}, \vec{\bar{q}}; \vec{\bar{r}}_1, \bar{t}_1) \rangle_{\Delta t_1 \Delta r_1}$$ where now $\bar{\omega} = \bar{\omega}_f - \bar{\omega}_i$

with $\tilde{\omega}_{f(i)} = \frac{m\bar{v}^2_{f(i)}}{2}$ $\bar{v}_f = \bar{s}_f / (t - \bar{t}_1)$ $\bar{v}_i = \bar{s}_i / (\bar{t}_1 - \bar{t}_i)$ classical velocities before and after scattering event

Homogeneity, ergodicity, equilibrium & linearity

$$\bar{J} = \frac{1}{N}\sum_i^N J_i(t) \propto \frac{1}{N}\sum_i^N \tilde{S}_i(\omega, q; t_i) = \bar{S}(\omega, q)$$

$$\bar{S}(\omega, q) = \langle \int d\omega_1 d\omega_1' e^{i(\omega_1+\omega_1')t} \rangle_t \int d\tau e^{-i(\omega-\bar{\omega}_1)\tau} U(\omega_1) U(\omega_1')$$

$$\propto \langle |U(\omega)|^2 \rangle$$

$$\overline{\omega_1} = \frac{\omega_1 + \omega_1'}{2}$$

here $U(q, \omega) = \int dt e^{i\omega t} e^{iHt} U(q) e^{-iHt}$

In equilibrium $\bar{S}(\omega, q) \Rightarrow \sum_{nm} \rho_n |U_{nm}(\bar{q})|^2 \delta(E_n - E_m - \bar{\omega})$ Expansion over exact states of the Hamiltonian

Example: $U(q, \omega) = S_n m_\perp(q, \omega)$ (magnetic interaction)

$\bar{S}(\omega, q) \propto \langle m^\alpha(q, \omega) m^\beta(-q, -\omega) \rangle$ is pair correlation function

$\bar{S}(\omega, q) \propto -n(-\omega)\chi_{\alpha\beta}''(q, \omega)$ via FDT related to:

$\chi_{\alpha\beta}(t) = -i\langle [m^\alpha(t) m^\beta(0) - m^\beta(0) m^\alpha(t)] \rangle \vartheta(t)$ linear response function

Higher harmonics: non-linear response for external field

$$m(t) \propto \{\cos(\Omega_0 t)\chi'(\Omega_0) + \sin(\Omega_0 t)\chi''(\Omega_0)\}B_0$$

$$m_2(t) \propto \{\cos(2\Omega_0 t)\chi_2'(2\Omega_0) + \sin(2\Omega_0 t)\chi_2''(2\Omega_0)\}B^2{}_0$$

$$m_3(t) \propto \{\cos(3\Omega_0 t)\chi_3'(3\Omega_0) + \sin(3\Omega_0 t)\chi_3''(3\Omega_0)\}B^3{}_0$$

etc ....

Harmonic analysis of scattering in ac field:

$$\Delta J(\Omega_0, \omega, t) \propto \{\cos(\Omega_0 t)\chi_3'(\omega - \Omega_0, \omega + \Omega_0) + \sin(\Omega_0 t)\chi_3''(\omega - \Omega_0, \omega + \Omega_0)\}B_0 + \ldots$$

Higher order correlations

$$\langle \tilde{U}(\omega)\tilde{U}(\omega')\rangle \propto \langle m_0(\omega)m_0(-\omega)\rangle\delta(\omega-\omega') + \langle m_0(\omega)m_0(\omega \pm \Omega)m_0(\omega \mp \Omega)\rangle B_0 + \ldots$$

Frequency mixing via incident flux modulation

Reflection and GID in ac field?

1. Circling along hysteresis loop. Nonlinear response.

2. Larmor precession of magnetization of finite elements in the dc field normal to the surface.

3. Doppler scan of domain walls propagation in films.

4. Excitations in lateral structures.

Frequency labeling of pulses and time frame overlap problem

$$J(t) \propto \sum_i \int_\Delta dt^i{}_1 d\omega_f d\omega_i \delta(t - t_1{}^i - s_f / v_f) \widetilde{S}(\omega, t_1{}^i) \delta(t_1{}^i - t_i - s_i / v_i) \cos\Omega_i t_1{}^i$$

Each pulse "*i*" is labeled by its own frequency Ω_i if $\Omega_i \Delta >> 1$

Δ is the pulse width

$$J(t) \propto \sum_i \int_\Delta dt^i{}_1 \widetilde{S}(\bar{\omega}_f - \bar{\omega}_i, t_1{}^i) \cos\Omega_i t_1{}^i$$

where $\bar{\omega}_f \approx \frac{m\bar{v}^2{}_f}{2} \approx \frac{ms^2{}_f}{2(t - \bar{t}_1)^2}$ $\bar{\omega}_i \approx \frac{m\bar{v}^2{}_i}{2} \approx \frac{ms^2{}_i}{2(\bar{t}_1 - \bar{t}_i)^2}$

$\bar{t}_1 - t_i = \langle t^i{}_1 \rangle - t_i$ is the value fixing incoming velocity $\bar{v}_i$

Intensity at fixed arriving time is a sum $J(t) \propto \sum_i J_i(t, \Omega_i)$

Correlation of flux fluctuations:

$$G(\tau)=\frac{1}{N}\int dt J(t+\tau)J(t)\propto\langle S(\omega,\vec{q};\tau)S(\omega,q;0)\rangle_t$$

Fourier transform:

$$G(\Omega)=\int d\tau e^{i\Omega\tau}G(\tau)\propto\langle|U(\omega+\Omega,\vec{q})|^2|U(\omega-\Omega,\vec{q})|^2\rangle$$

Fluctuations - differential effect $\Delta G\propto G(\Omega)-G(0)$

$$\Delta G(\Omega)\propto\langle[|U(\omega+\Omega,\vec{q})|^2-\langle|U(\omega,\vec{q})|^2\rangle][|U(\omega-\Omega,\vec{q})|^2-\langle|U(\omega,\vec{q})|^2\rangle]\rangle$$

Four point correlation function:

$$\langle m^{\alpha}(\vec{q},\omega+\Omega)m^{\beta}(-\vec{q},-\omega+\Omega)m^{\gamma}(\vec{q},\omega-\Omega)m^{\delta}(-\vec{q},-\omega-\Omega)\rangle$$

is expressed via non-linear susceptibility

Non-linear response for ac field

Scattering on system under external ac field. Hamiltonian $H_{ext}(t)$

$$\tilde{\vec{U}}(\vec{q},t) = \hat{S}^{-1}(t)\vec{U}(\vec{q},t)\hat{S}(t)$$ where $U(t) = e^{iH_0 t} U e^{-iH_0 t}$ Heisenberg representation: Hamiltonian with no field

S-matrix: $$\hat{S}(t) = \hat{T}\exp\{-i\int dt H_{\text{int}}(t)\} = 1 - i\int_{-\infty}^{t} dt_1 H_{\text{int}}(t_1) - \int_{-\infty}^{t} dt_1 \int_{-\infty}^{t_1} dt_2 H_{\text{int}}(t_1) H_{\text{int}}(t_2) + \ldots$$

with $$\tilde{U}(t) = U(t) - i\int_{-\infty}^{t} dt_1 \{H_{\text{int}}(t_1)U(t) - U(t)H_{\text{int}}(t_1)\} + \ldots$$ and

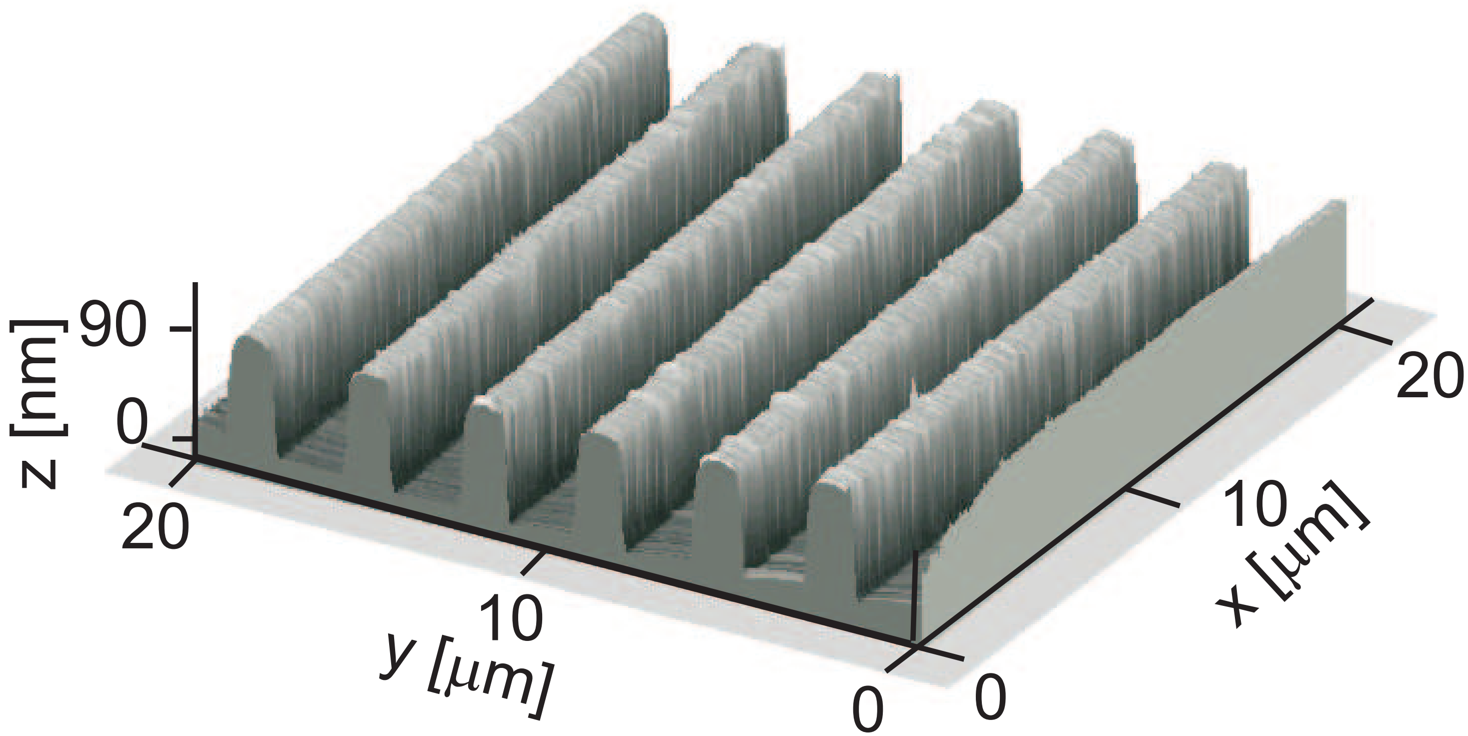
$$H_{\text{int}}(t) = e^{iH_0 t} H_{ext}(t) e^{-iH_0 t}$$ external field in the "interaction" representation

Example: external ac *magnetic* field: $$H_{ext}(t) = \vec{m}_0 \vec{B}(t) = \vec{m}_0 \vec{B}_0 \cos(\Omega_0 t)$$

$$H_{\text{int}}(t) = \vec{m}_0(t)\vec{B}(t) = \vec{m}_0(t)\vec{B}_0 \cos(\Omega_0 t)$$ where $$\vec{m}_0(t) = e^{iH_0 t}\vec{m}_0 e^{-iH_0 t}$$

$$\langle \tilde{U}(t)\tilde{U}(t')\rangle \propto \langle m_0(t) m_0(t')\rangle - i\langle \int_{-\infty}^{\infty} dt_1 \{\chi_0(t-t_1) m_0(t') - m_0(t)\chi_0(t_1 - t')\}\rangle B_0(t_1) + \ldots$$

$$\chi_0(t) = -i[m_0(t)m_0(0) - m_0(0)m_0(t)]\vartheta(t)$$ is retarded commutator



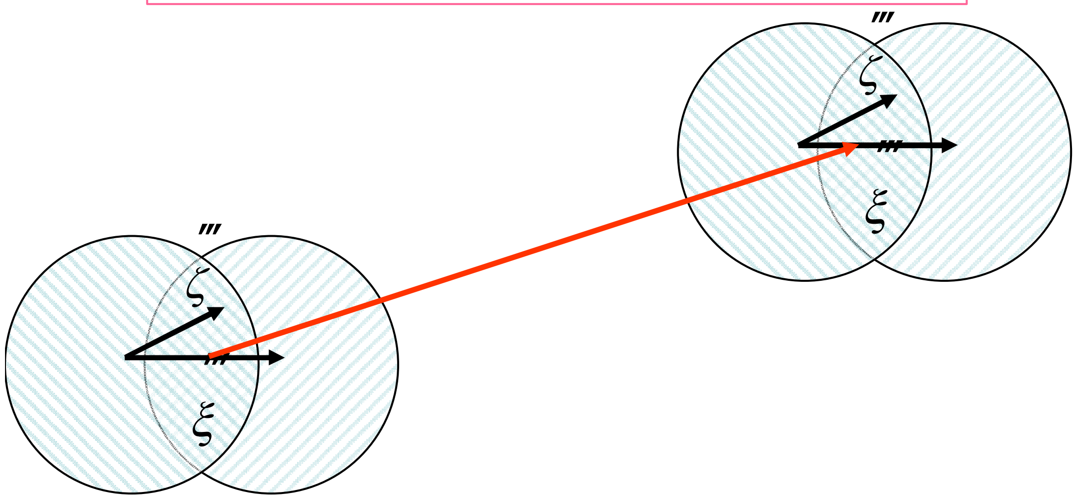
z [nm]
90
0
20
10
y [μm]
0
0
10
20
x [μm]

Doppler shift from hard sphere moving with constant speed

$$\tilde{S}(\overline{\omega}, q) \propto V(q) \int_{-\tau_r}^{\tau_r} d\tau e^{-i[\overline{\omega} - q v_l]\tau} = 2\pi V(q)\Delta[\overline{\omega} - q v_l]$$

$$\Delta[\overline{\omega} - q v_l] \Rightarrow \delta(\overline{\omega} - q v_l)$$

Accounts for moving center of mass



is sphere
form-factor

In general $\vartheta(r_0 - v\,|\,\xi\,|)$ is particle shape function with $r_0 = r_0(t, n)$

$r_0 \sim 10^{-6}\, cm, v_T \sim 10^2\, cm/s, \tau_0 \sim 10^{-4}\, s, qv = \omega \sim 10^8\, s^{-1}$

GISANS from block co-polymer nano-composite multilayers (D22 ILL)

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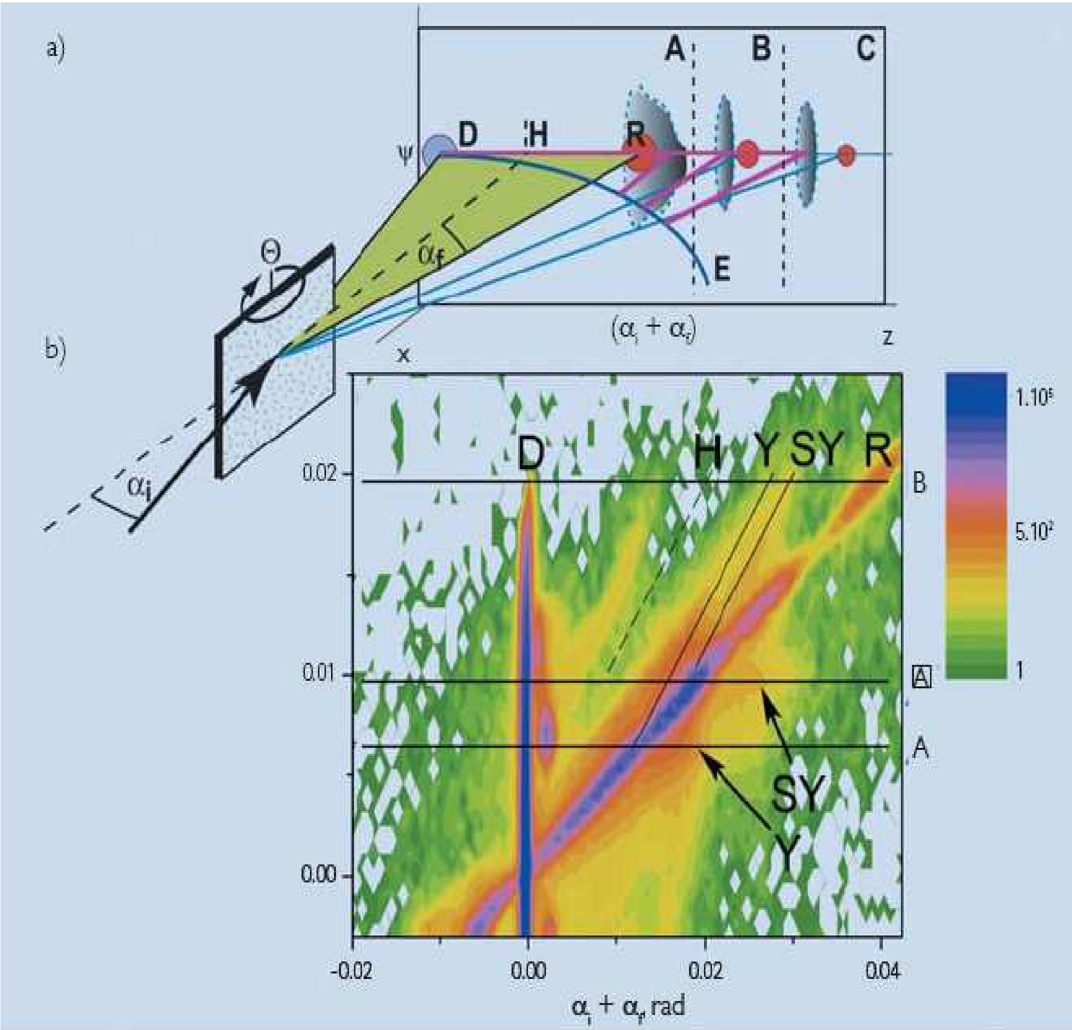
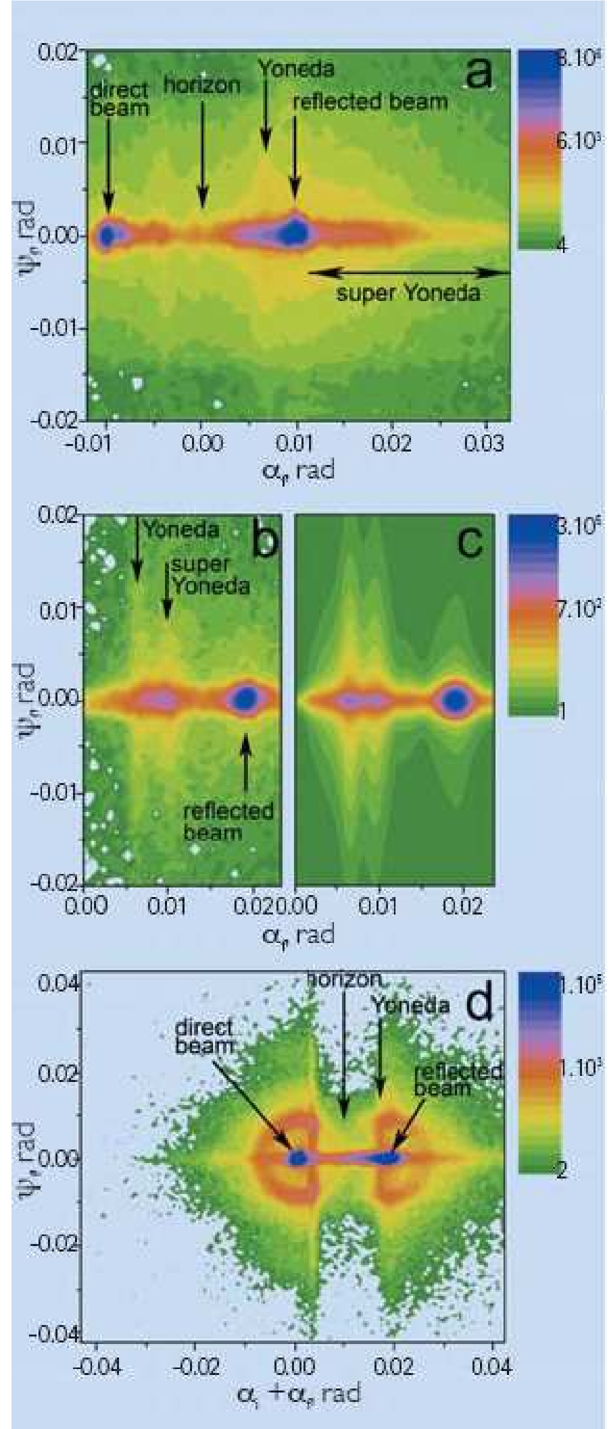
B. Toperverg (PNPI St. Petersburg and ILL)
M. Jernenkov and H. Lauter (ILL)

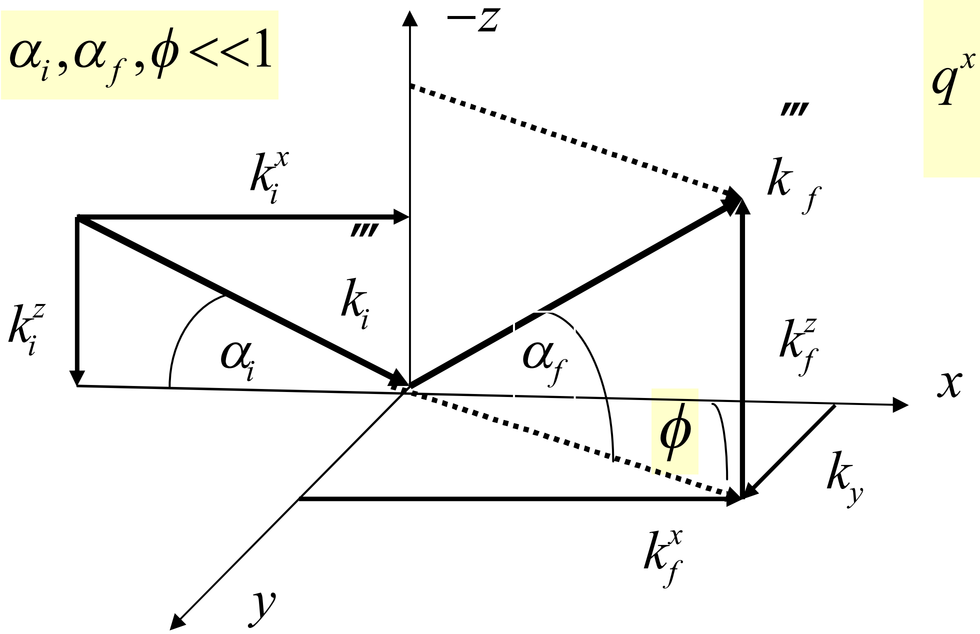




Grazing incidence kinematics

$$\tilde{S}(\overline{\omega},q;\bar{t}_l) \propto \int d\tau e^{-i\overline{\omega}\tau} \; < e^{iq^x(x_l'-x_l'')} > e^{iq^y a\cos\Omega\tau} F(q)$$

$$\alpha_i,\alpha_f,\phi<<1$$



$$q^x(\overline{\omega}) \approx \frac{\omega_i}{c}\left[\frac{\alpha_i^{\,2}-\alpha_f^{\,2}}{2}+\frac{\overline{\omega}}{\omega_i}-\frac{\phi^2}{2}\right] << q^z, q^y$$

$$q^z \approx \frac{\omega_i}{c}\left[\alpha_i+\alpha_f\right]$$

$$q^y \approx \frac{\omega_i}{c}\phi$$

and

where

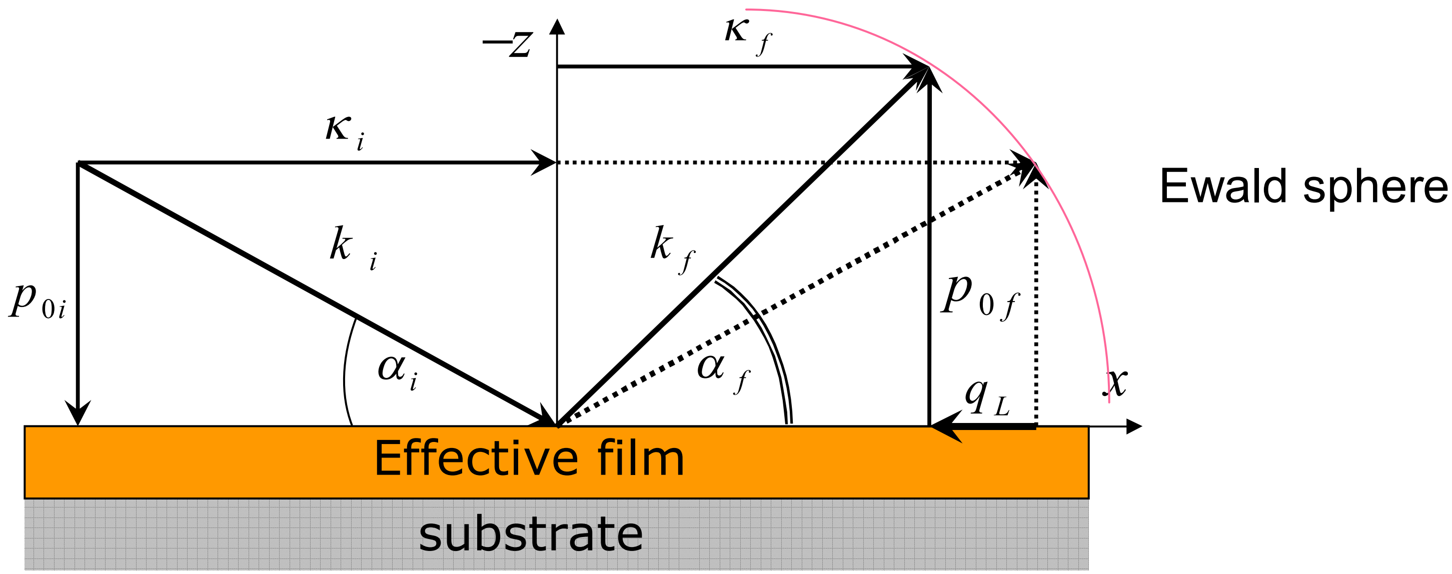
$$< e^{iq^x(x_l'-x_l'')} > \approx \delta(q^x) \propto \delta[\alpha_i^{\,2}-\alpha_f^{\,2}+2\overline{\omega}/\omega_i]$$

with

$$\tilde{S}(\overline{\omega},q;\bar{t}_l) = \int d\tau e^{-i\overline{\omega}\tau} e^{iqa_0\cos\Omega\tau} F(q)$$

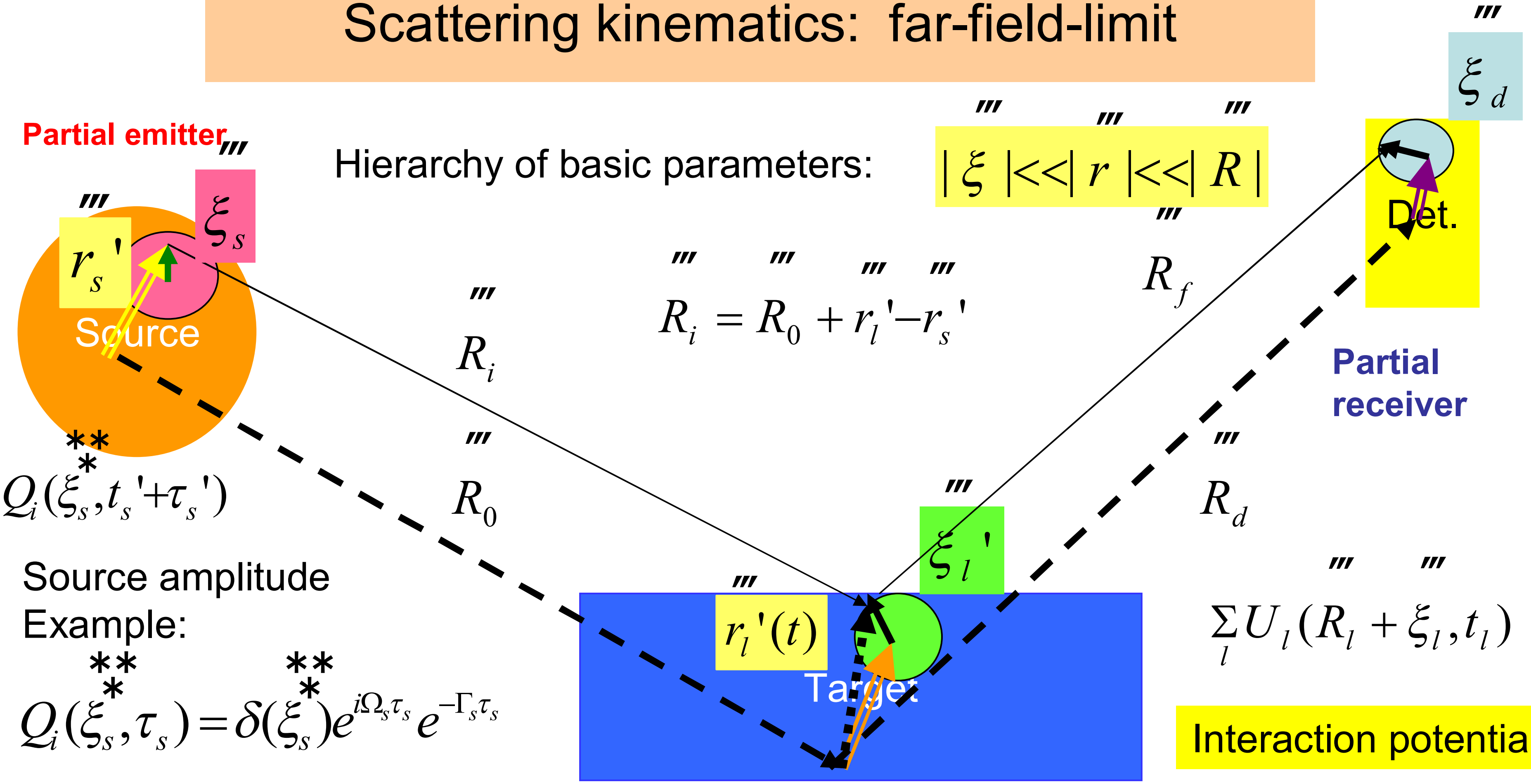
Specular reflection an off-specular scattering

- Specular:
  Energy+lateral momentum are conserved, transverse not conserved
- Off-specular: Lateral momentum is not conserved



Scattering kinematics: far-field-limit

Hierarchy of basic parameters: $|\vec{\xi}| \ll |\vec{r}| \ll |\vec{R}|$



$$\vec{R}_i = \vec{R}_0 + \vec{r}_l' - \vec{r}_s'$$

$$Q_i(\vec{\xi}_s^{\,*}, t_s' + \tau_s')$$

Source amplitude

Example:

$$Q_i(\vec{\xi}_s^{\,*}, \tau_s) = \delta(\vec{\xi}_s^{\,*}) e^{i\Omega_s \tau_s} e^{-\Gamma_s \tau_s}$$

$$\Gamma \ll \Omega_0 \sim 10^{18} - 10^{20} s^{-1}$$

$$R \sim 10^2 - 10^3 \, cm$$

$$r \sim 10^{-1} - 10^{-3} \, cm$$

$$\xi \sim 10^{-4} - 10^{-8} \, cm$$

$$\sum_l U_l(\vec{R}_l + \vec{\xi}_l, t_l)$$

Interaction potential

$$\left|\vec{R}_i\right| \approx R_0 + \vec{n}_0(\vec{r}_l' - \vec{r}_s')$$

$$\vec{n}_0 = \vec{R}_0 / |\vec{R}_0|$$

Wave equation with source

includes initial and boundary conditions

Formal solution: $\psi(\vec{r},t) = \int d\vec{r}\,' dt' G(\vec{r}-\vec{r}\,', t-t')Q(\vec{r}\,',t')$ integral form of the W.E.

where the retarded Green function $G(\vec{r},t) = \int d\varepsilon G(\vec{r},\varepsilon)e^{-i\varepsilon t}\vartheta(t)$

obeys W.E. with point-like instant source $Q(\vec{r},t) = \delta(\vec{r})\delta(t)$ is a superposition

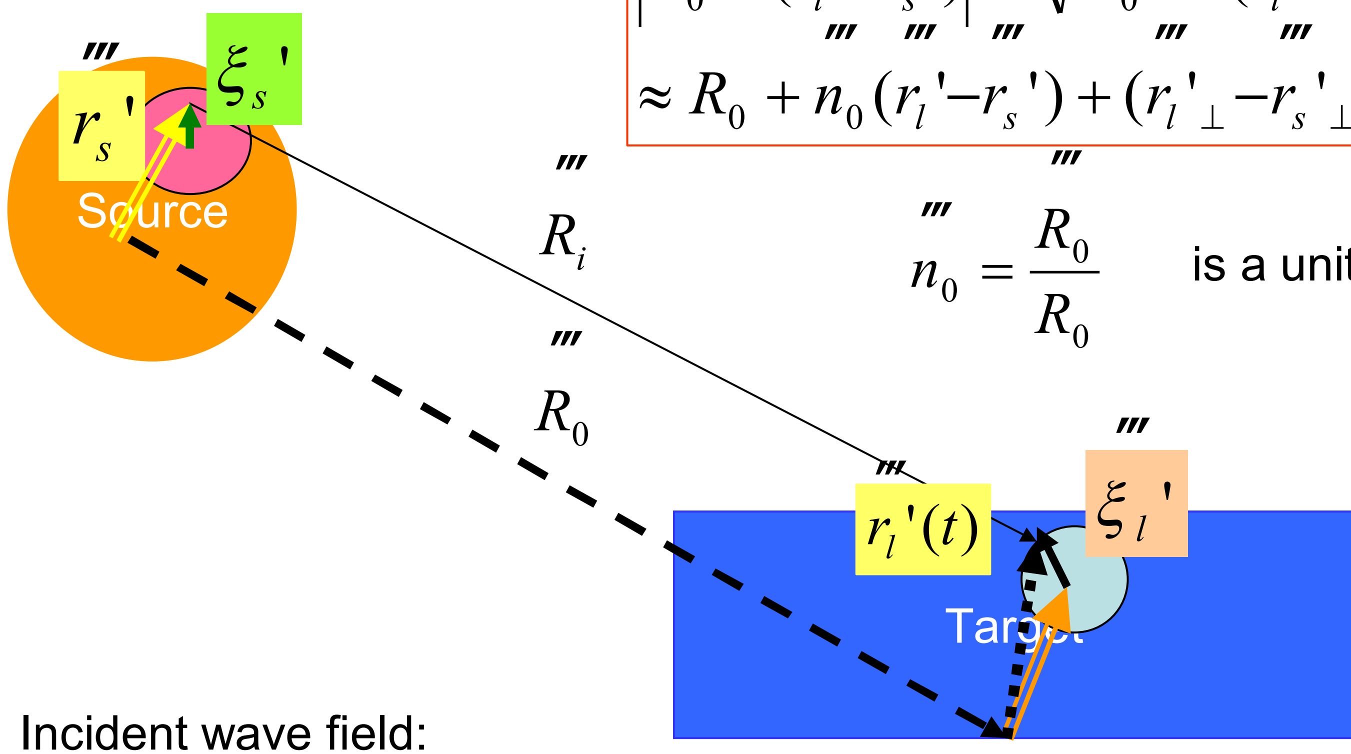
of divergent spherical waves $G(\vec{r},\varepsilon) \propto \dfrac{e^{ikr}}{r}$

with wave numbers due to $\hbar^2 k^2 = 2m\hbar\varepsilon$

Far field limit

$R_0 >> r_l, r_s \Rightarrow$

$$\left|\vec{R}_0 + (\vec{r}_l' - \vec{r}_s')\right| = \sqrt{R_0^{\,2} + (\vec{r}_l' - \vec{r}_s')^2 + 2\vec{R}_0(\vec{r}_l' - \vec{r}_s')}$$
$$\approx R_0 + \vec{n}_0(\vec{r}_l' - \vec{r}_s') + (\vec{r}_l'_{\perp} - \vec{r}_s'_{\perp})^2 / 2R_0 + \ldots..$$



$$\vec{n}_0 = \frac{\vec{R}_0}{R_0}$$ is a unit vector

$$(\vec{r}_{\perp}\vec{n}_0) = 0$$

determines coherence ellipsoid (resolution)

Incident wave field:

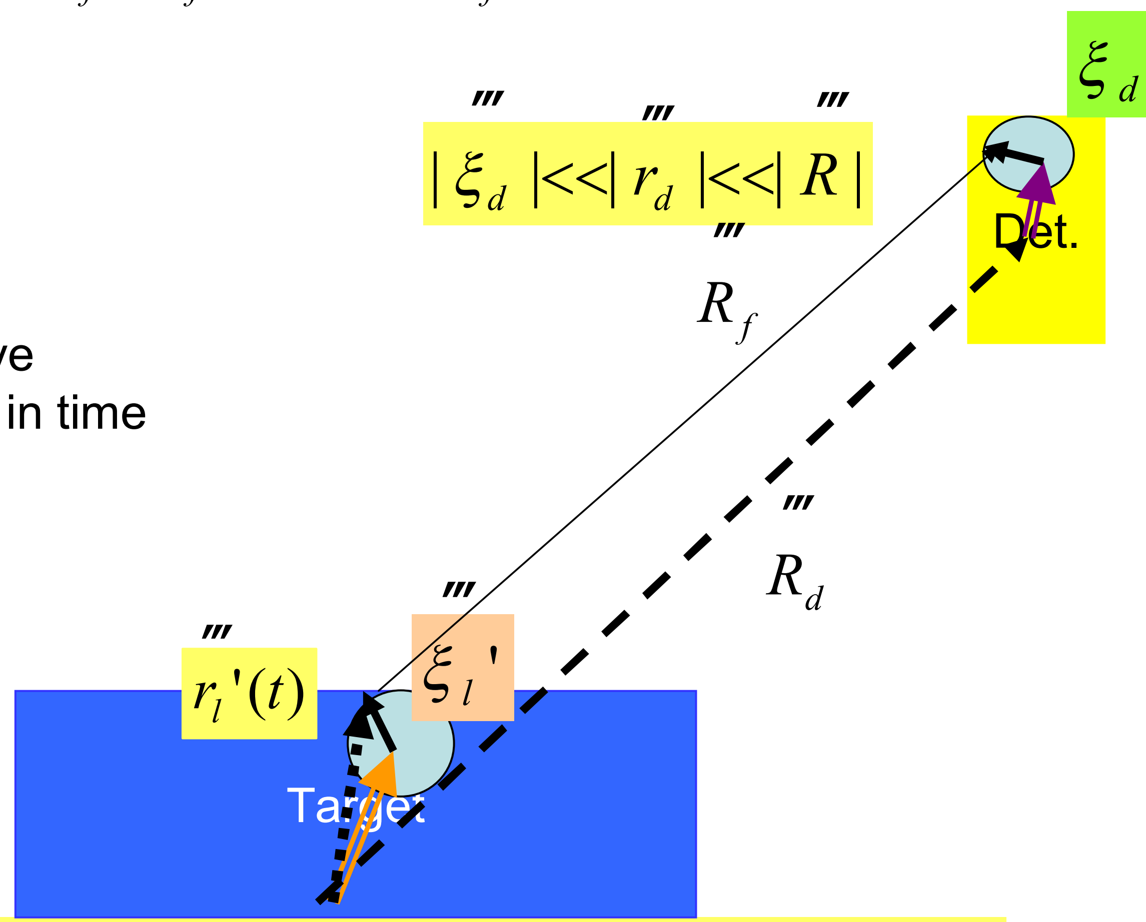
$$\psi_i(\vec{R}_i, t_l) \approx \int d\omega_i' e^{i\vec{k}_i\vec{r}_l'} e^{-i\omega_i(t_l - t_s)} G(R_0, \omega_i') Q_i(\omega_i' - \omega_0) \vartheta(t_l - t_s)$$

outgoing wave field (space, but not time reversed):

$$\psi_f(\vec{R}_f, t_d - t_l) = \int d\omega_f' Q_f(\omega_f') G(R_d, \omega_f') e^{-i\vec{k}_f \vec{r}\,'_l} e^{-i\omega_f'(t_d' - t_l')} \vartheta(t_d' - t_l')$$

$$\vec{k}_f = \frac{\vec{R}_d}{R_d} k_f = \vec{n}_f k_f$$

The same as incoming wave
reversed in space, but not in time



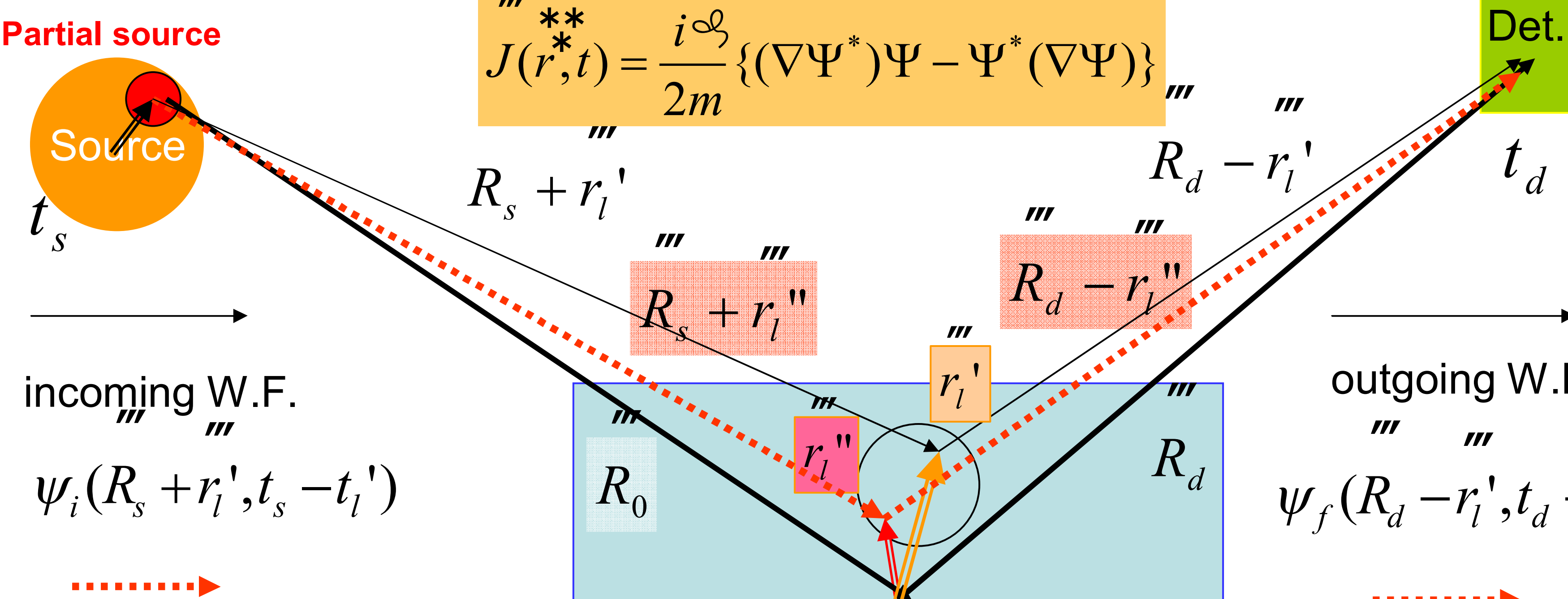
Transition
amplitude in BA:

$$\int_{t_s}^{t_d} dt_l d\vec{r}_l \psi_f(\vec{R}_d - \vec{R}_l, t_d - t_l) U(\vec{r}_l, t_l) \psi_i(\vec{R}_s + \vec{R}_l, t_l - t_s)$$

Observable

Flux density is bi-linear combination of W.F.

$$\vec{J}(\vec{r},t) = \frac{i\hbar}{2m}\{(\nabla\Psi^*)\Psi - \Psi^*(\nabla\Psi)\}$$



incoming W.F.

$$\psi_i(R_s + r_l', t_s - t_l')$$

outgoing W.F.

$$\psi_f(R_d - r_l', t_d - t_l')$$

time reversed incoming W.F.

$$\psi_i^*(R_s + r_l'', t_s - t_l'')$$

$$t_l' = t_l''$$

Equal time instant correlation

time reversed outgoing W.F.

$$\psi_f^*(R_d - r_l'', t_d - t_l'')$$

Incident wave field correlator

$$\Psi_i(t')\Psi_i^*(t'') \propto \int d\omega_i' d\omega_i'' e^{-i(\Phi_i' - \Phi_i'')} Q_s(\omega_i') Q_s^*(\omega_i'')$$

with (huge!) phases $\Phi_i' = \omega_i(t_l' - t_s - R_s/c) - \vec{k}_i \vec{r}_l'$ $\quad \Phi_i'' = \omega_i''(t_l'' - t_s - R_s/c) - \vec{k}_i \vec{r}_l''$

$\bar{t}_l = (t_l' + t_l'')/2$ mean time of propagation $\quad \tau = (t_l' - t_l'')$ time shift

$\overline{\omega}_i = (\omega_i' + \omega_i'')/2$ mean frequency $\quad \tilde{\omega} = (\omega_i' - \omega_i'')$ frequency shift

$$\boxed{\Psi_i(t_l')\Psi_i^*(t_l'') \propto \int d\overline{\omega}_i e^{-i\overline{\omega}_i \tau} e^{i\vec{k}_i(\vec{r}_l' - \vec{r}_l'')} W_i(\overline{\omega}_i, \bar{t}_l - t_s)}$$

with

$$W_i(\vec{k}_i; \overline{\omega}_i, \bar{t}_l) \propto \int d\tilde{\omega} e^{-i\tilde{\omega}(\bar{t}_l - R_s/c)} Q_i(\overline{\omega}_i + \tilde{\omega}/2) Q_i^*(\overline{\omega}_i - \tilde{\omega}/2)$$ ray tracing function

$$\boxed{(\bar{t} - R_s/c) \le \tilde{\omega}^{-1} \sim \tau_0}$$

Ray tracing function example:

Example: $$Q_i(\overset{*\!*}{\underset{*}{\xi}}_s,\tau_s)=\delta(\overset{*\!*}{\underset{*}{\xi}}_s)e^{i\omega_0\tau_s}e^{-\tau_s/\tau_0}$$

$$\overset{\prime\prime\prime}{W}_i(k_i;\overline{\omega}_i,\bar{t})\propto \overset{\prime\prime\prime}{W}_i(k_i;\overline{\omega}_i)\frac{\sin\left[(\overline{\omega}-\omega_0)(\bar{t}_l-R_s/c)\right]}{(\overline{\omega}-\omega_0)}e^{-2(\bar{t}-R_s/c)/\tau_0}\vartheta(\bar{t}-R_s/c)$$

Speckles in time

$$t\sim R/c\sim(10^{-10}-10^{-9})s$$

$$(\bar{t}-R_s/c)\leq 2\tau_0\sim 10^{-13}-10^{-15}s$$

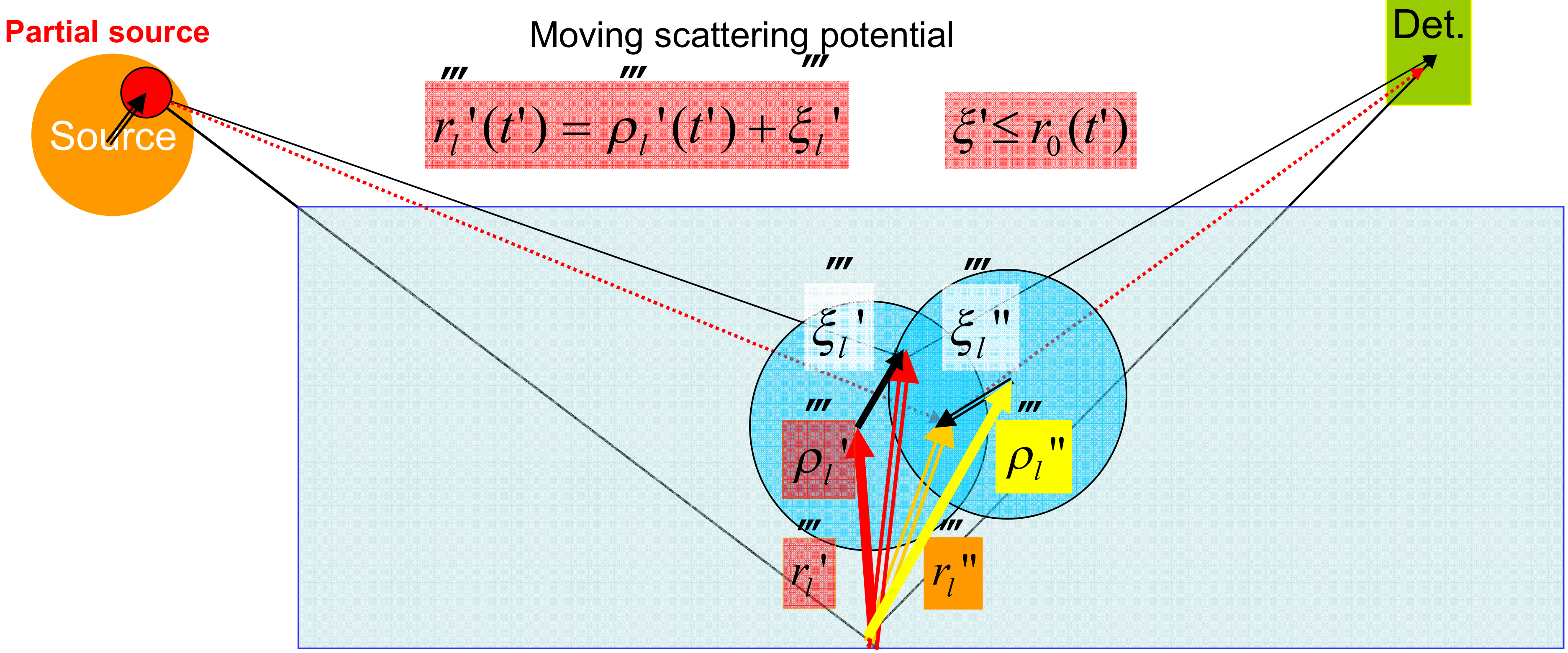
$$W_i(\overline{\omega}_i,\bar{t})\Rightarrow\delta(\bar{t}-R_s/c)\delta(\overline{\omega}_i-\omega_0)$$

Smeared over range:

$$\left|\overline{\omega}-\omega_0\right|\leq(\bar{t}-R_s/c)^{-1}\leq\tau_0^{-1}$$

Wave field correlations in space and time

$t_l' \neq t_l''$ $\qquad t_l' = \bar{t}_l + \tau_l/2$ $\qquad t_l'' = \bar{t}_l - \tau_l/2$



$$r_l''(t'') = \rho_l''(t'') + \xi_l''$$

$\xi'' \leq = r_0(t'')$ Interaction range

Intermediate scattering function via correlator

$$\tilde{S}(\overline{\omega},\vec{q};\bar{t}_l) = \int d\vec{r}_l' d\vec{r}_l'' d\tau e^{-i\overline{\omega}\tau} e^{i\vec{q}(\vec{r}_l'-\vec{r}_l'')} U(\vec{\xi}_l',t_l')U(\vec{\xi}_l'',t_l'')$$

where: $\vec{r}_l(t') = \vec{\rho}_l'(t') + \vec{\xi}_l'$ $\quad \vec{r}_l''(t'') = \vec{\rho}_l''(t'') + \vec{\xi}_l''$ and $\tau = t_l' - t_l''$

$$\tilde{S}(\overline{\omega},\vec{q};\bar{t}_l) = \int d\tau e^{-i\overline{\omega}\tau} e^{i\vec{q}(\vec{\rho}_l'-\vec{\rho}_l'')} K(\vec{q},\tau;\bar{t}_l)$$

$$K(\vec{q},\tau;\bar{t}_l) = \int d\vec{\xi} e^{i\vec{q}\vec{\xi}} K(\vec{\xi},\tau;\bar{t}_l)$$

$$K(\vec{\xi}'-\vec{\xi}'',\tau;\bar{t}_l) = \int d\vec{\zeta} U(\vec{\xi}',t_l')U(\vec{\xi}'',t_l'')$$

is autocorrelation function averaged over

$\vec{\varsigma} = (\vec{\xi}_l' + \vec{\xi}_l'')/2$ and depending on the difference $\vec{\xi} = \vec{\xi}_l' - \vec{\xi}_l''$ $\quad \bar{t} = (t_l' + t_l'')/2$

expanding $\vec{r}_l' \approx \vec{r}_l(\bar{t}_l) + \vec{v}_l'\tau/2$ and $\vec{r}_l'' \approx \vec{r}_l(\bar{t}) - \vec{v}_l''\tau/2$ then $\vec{r}_l' - \vec{r}_l'' = \vec{v}_l\tau$

$$\tilde{S}(\overline{\omega},\vec{q};\bar{t}_l) = \int d\tau e^{-i[\overline{\omega}-\vec{q}\vec{v}_l]\tau} K(\vec{q},\tau;\bar{t}_l)$$

$\vec{v}_l$ is velocity

Probability via intermediate scattering function

$$I(t) \propto \int d\bar{t}_l d\overline{\omega}_f d\overline{\omega}_i W_f(\overline{\omega}_f) \Delta(t_d - \bar{t}_l - R_d / c) \widetilde{S}(\overline{\omega}, \vec{q}; \bar{t}_l) W_i(\overline{\omega}_i) \Delta(\bar{t}_l - t_s - R_0 / c)$$

where $\overline{\omega} = \overline{\omega}_f - \overline{\omega}_i$ is frequency shift $\vec{q} = \vec{k}_f - \vec{k}_i$ is wave vector transfer

Neglecting smearing over uncertainties $\bar{t}_l \leq (\tau_s, \tau_d)$ and time of interaction

and time-of-flight functions:

$$\Delta(\bar{t}_l - t_s - R_0 / c) \Rightarrow \delta(\bar{t}_l - t_s - R_0 / c)$$

$$\Delta(t_d - \bar{t}_l - R_d / c) \Rightarrow \delta(t - \bar{t}_l - R_d / c)$$

$$I(t) \propto \delta[t - t_s - (R_s + R_d)/c] \int d\overline{\omega}_f d\overline{\omega}_i W_f(\overline{\omega}_f) \widetilde{S}(\overline{\omega}, \vec{q}; \bar{t}_l) W_i(\overline{\omega}_i)$$

$$\bar{t}_l \approx t_s + R_s / c$$

with spectral weight functions $W_i(\overline{\omega}_i)$ $W_f(\overline{\omega}_f)$

Way to reciprocal world

Scattered wave function $\Psi_f(\vec{r},t) = \int d\vec{r}_1 \int dt_1 G(\vec{r}-\vec{r}_1, t-t_1) U(\vec{r}_1, t_1) \Psi_i(\vec{r}_1, t_1)$

with $G(\vec{r}-\vec{r}_1, t-t_1) = \int d\varepsilon_f G(\vec{r}-\vec{r}_1, \varepsilon_f) e^{-i\varepsilon_f (t-t_1)}$

is a superposition of *plane waves* with spherical wave amplitudes

$$G(\vec{r}-\vec{r}_1, \varepsilon_f) \propto \frac{e^{ik_f|\vec{r}-\vec{r}_1|}}{|\vec{r}-\vec{r}_1|} \approx \frac{e^{ik_f r}}{r} e^{-i\vec{k}_f \vec{r}_1}$$

wave vectors $-\vec{k}_f = -\vec{n}_f k_f$ space (not time!) inversed

and frequencies due to $\hbar^2 k_f^2 = 2m\hbar\varepsilon_f$

Off-specular scattering map for uncorrelated domains in multilayer