



1866-13

#### School on Pulsed Neutrons: Characterization of Materials

15 - 26 October 2007

**Polarized Neutrons** 

Peter Boeni Technische Universitat Munchen Fakultat fur Physik E21 Munchen Germany

## Methods and Techniques: Polarized Neutrons

**Lecture Notes** 

#### Peter Böni

Physik-Department E21 Technische Universität München D-85747 Garching, Germany

E-mail: peter.boeni@frm2.tum.de Web: www.ph.tum.de

1

ICTP School on Pulsed Neutrons, October 15 - 26 2007, Trieste

## Topics

- 1. Introduction
- 2. Need for polarized neutrons
- 3. Basics of polarized beam technique
- 4. Ancillary equipment for polarization analysis
- 5. Heusler monochromators
- 6. Polarizing supermirrors
- 7. Spin Filters: Protons, <sup>3</sup>He
- 8. Instruments
- 9. Applications for Polarized Neutrons
- 10. Focusing Techniques
- 11. Conclusions

## 1. Introduction

## **Comparison Neutrons – X-rays**



scattering length is random substitution by isotopes no charge magnetic moment

- distinction between light and heavy elements
- $\rightarrow$  deuteration

 $\rightarrow$ 

- $\rightarrow$  large penetration depth  $\rightarrow$  volume sensitiv
- $\rightarrow$  magnetic properties

## **Extreme sample environment**



Sample environment is essential (not only the neutrons)!

## Landmarks in Polarized Neutron Scattering

• 1939: Halpern and Johnson: polarized neutrons - magnetic moments

(O. Halpern and M. R. Johnson, Phys. Rev. 55, 898 (1939))

$$\mathbf{P'}=f(\mathbf{Q}\cdot\mathbf{P})$$

- 1957: Nathans et al.: experiment with polarized neutrons on Cr<sub>2</sub>O<sub>3</sub> scattered neutrons are analyzed with magnetized block of Fe (R. Nathans, T. Riste, G. Shirane, and C. G. Shull, Bull. Am. Phys. Soc. 2, 1, FA4 (1957))
- 1969: Moon, Riste and Koehler: measurement of  $\sigma^{++}$ ,  $\sigma^{-}$ ,  $\sigma^{+-}$ ,  $\sigma^{+-}$



## Measurement of $\sigma^{++}$ , $\sigma^{--}$ , $\sigma^{+-}$ , $\sigma^{-+}$



Combine monochromatization with polarization.

## Landmarks in Polarized Neutron Scattering

• 1939: Halpern and Johnson: polarized neutrons – magnetic moments

(O. Halpern and M. R. Johnson, Phys. Rev. 55, 898 (1939))

 $\mathbf{P'}=f(\mathbf{Q}\cdot\mathbf{P})$ 

- 1957: Nathans et al.: experiment with polarized neutrons on Cr<sub>2</sub>O<sub>3</sub> scattered neutrons are analyzed with magnetized block of Fe (R. Nathans, T. Riste, G. Shirane, and C. G. Shull, Bull. Am. Phys. Soc. 2, 1, FA4 (1957))
- 1969: Moon, Riste and Koehler: measurement of σ<sup>++</sup>, σ<sup>-</sup>, σ<sup>+-</sup>, σ<sup>+</sup>, σ<sup>+</sup>
- 1972: Mezei: neutron spin echo spectrometer: high *E*-resolution
- 1976: Mezei: polarizing supermirror (see also Turchin 1967) (F. Mezei, Communications on Physics 1, 81 (1976), V. F. Turchin, Deposited Paper, At. Energy 22 (1967))
- 1980: Ziebeck and Brown: inelastic measurements on 3-d magnets

## **Polarizers**

• Fe in magnetic field:

neutrons with spin down  $(b+p)^2$ : large cross section/small transmission neutrons with spin up  $(b-p)^2$ : small cross section/good transmission

• monochromators:

<sup>57</sup>Fe, FeCo Heusler Cu<sub>2</sub>MnAl (similar *d*-spacing as HOPG)

• Artificial multilayers:

FeGe multilayers supermirrors: Co/Ti, Fe/Si, Fe<sub>50</sub>Co<sub>48</sub>V<sub>2</sub>/TiN<sub>x</sub>

• Spin filters:

polarized protons polarized <sup>3</sup>He SmCo<sub>5</sub>

## 2. Need for polarized neutrons

## **Example 1: Measurement of Form Factors**



Polarization analysis is important to enhance signal:

## **Example 2: Magnetic Structures**



Janoschek et al., J. Phys.: Cond. Matter 17, L425

(P. J. Brown et al. J. Phys.: Condens. Matter 18 (2006) 10085-10096)

Spherical polarization analysis distinguishes between structures

## **Example 3: Nuclear and Magnetic Excitations**

Inelastic neutron scattering from Ni: magnons - phonons?



Martínez et al., Phys. Rev. B 32, 7037 (1985

## **Example 4: Separation of magnetic modes**



Polarization analysis can distinguish between various magnetic modes

## **Example 5: Chirality**



Polarization analysis distinguishes between left- and right-handed spirals

## **Example 6: Dynamics of Deuterium in Nb**

Further examples: polymers, hydrogen storage ( $\rightarrow$  Zabel)



## **Example 7: High-Resolution / Spin Echo**



High resolution with polarized neutrons (1µeV)!

17

### **Example 8: Polarized Neutrons for Particle Physics**

#### Time inversion invariance experiments:

- $n \rightarrow p + e + v + \gamma$  (asymmetry of n-decay)
- electric dipole moment



(from O. Zimmer, Techn. Univ. Munich)

## Where are polarized beams available?

- Polarized triple axis spectrometers at reactors: ILL, HMI, NIST, FRM-II, ... and PSI (spallation source)
- Time-of- Flight: ILL (D7), OSIRIS (ISIS), HYSPEC (SNS, planned)
- Neutron Spin Echo: ILL, FRM II, HMI, NIST, LLB, JAEA
- Reflectometers: almost all facilities
- Small angle neutron scattering: HMI, PSI, FRM II, JAEA ...
- Particle physics: ILL

polarized neutrons mostly at continuous sources Urgent need for:

- ToF
- dedicated beam lines



# **3. Basics of Polarized Beam Technique**

## **Scattering Cross Section**

(see also W. Fischer's talk)

• Fermi's golden rule (= 1<sup>st</sup> Born approximation):

$$\frac{d^2\sigma}{d\Omega dE_f} = \left(\frac{m}{2\pi\hbar^2}\right)^2 \frac{k_f}{k_i} \sum_{\lambda_i} p_{\lambda_i} \sum_{\lambda_f} \left| \left\langle k_f, \lambda_f \right| \widetilde{\mathbf{U}} \right| k_i, \lambda_i \right\rangle \right|^2 \delta \left( E_{\lambda_i} - E_{\lambda_f} + \hbar \omega \right)$$

•  $|\lambda_i\rangle$ : initial state of sample

•  $p_{\lambda i}$ : probability that initial state is occupied

 $\frac{2\pi\hbar^2}{m}b\delta(\mathbf{r}-\mathbf{r}_j)$ 

•  $|\lambda_f\rangle$ : final state of sample

- U: interaction potential neutron-sample
- matrix element for nuclear scattering:

$$\left\langle \mathbf{k}_{f}, \boldsymbol{\lambda}_{f} \middle| \mathbf{\widetilde{U}} \middle| \mathbf{k}_{i}, \boldsymbol{\lambda}_{i} \right\rangle = \left\langle f \middle| \mathbf{\widetilde{U}} \middle| i \right\rangle = \left\langle \lambda_{f} \middle| \int_{j} \sum_{j} e^{-i\mathbf{k}_{f}\mathbf{r}} V_{j}(\mathbf{r} - \mathbf{r}_{j}) e^{i\mathbf{k}_{i}\mathbf{r}} dr \middle| \lambda_{i} \right\rangle$$

is essentially proportional to  $\sum_{i} \int b_{i} e^{i\mathbf{Q}\cdot\mathbf{r}_{i}}$ 

Message: scattering given by the square of the Fourier transform of potential

## **Introducing the Scattering Function**

• scattering intensity is proportional to square of scattering amplitude

$$\sum_{j} \int b_{j} e^{i\mathbf{Q}\cdot\mathbf{r}_{j}}$$

• a more detailed treatment yields:

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{1}{2\pi\hbar} \frac{k_f}{k_i} \sum_{jj'} b_j b_{j'} \int \left\langle e^{-i\mathbf{Q}\cdot\mathbf{r}_{j'}(0)} e^{i\mathbf{Q}\cdot\mathbf{r}_{j}(t)} \right\rangle e^{-i\omega t} dt$$

• in terms of the scattering function:

$$\frac{d^2\sigma}{d\Omega dE_f} = \frac{\sigma}{4\pi} \frac{k_f}{k_i} NS(\mathbf{Q}, \omega)$$

• which is given by  $S(\mathbf{Q},\omega) = \frac{1}{2\pi\hbar} \int G(\mathbf{r},t) e^{i(\mathbf{Q}\cdot\mathbf{r}-\omega t)} d\mathbf{r} dt$ 

• where the pair correlation function: 
$$G(\mathbf{r},t) = \left(\frac{1}{2\pi}\right)^3 \frac{1}{N} \int \sum_{jj'} e^{-i\mathbf{Q}\cdot\mathbf{r}} \langle b_{j'} e^{-i\mathbf{Q}\cdot\mathbf{r}_{j'}(0)} b_j e^{i\mathbf{Q}\cdot\mathbf{r}_{j}(t)} \rangle d\mathbf{Q}$$

## **Magnetic Interaction**

polarization of neutron

• magnetic interaction operator:

$$\breve{\mathbf{U}}_m = -\mathbf{\mu} \cdot \mathbf{B} = -\gamma \mu_N \mathbf{\sigma} \cdot \mathbf{B}$$

• <i>γ</i> = -1.913:	gyromagnetic ratio
• $\mu_N$ :	nuclear magneton
• µ:	magnetic moment of neutron
• <b>o</b> :	Pauli spin operator
• B:	magnetic field (sample, external field, pseudo)

• field of unpaired electron at position  $\mathbf{r}_i$  (dipolar approximation):



## **Magnetic Scattering Length**

• Fourier transform of dipolar interaction yields magnetic scattering length:

$$p = -\gamma r_0 \boldsymbol{\sigma} \cdot \left( \hat{\mathbf{Q}} \times (\mathbf{S} \times \hat{\mathbf{Q}}) + \frac{i}{\hbar |\mathbf{Q}|} \left( \mathbf{p}_e \times \hat{\mathbf{Q}} \right) \right) = -\gamma r_0 \frac{g}{2} \boldsymbol{\sigma} \cdot \left( \hat{\mathbf{Q}} \times (\mathbf{S} \times \hat{\mathbf{Q}}) \right)$$
  
compare  
with b  
• classical radius of electron:  $r_0 = 0.2818 \cdot 10^{-12} \text{ cm}$   
• normalised scattering vector:  $\hat{\mathbf{Q}} = \mathbf{Q} / |\mathbf{Q}|$   
• Landé factor:  $g = 1 + \frac{J(J+1) + S(S+1) - L(L+1)}{2J(J+1)}$ 

• note: 
$$p$$
 depends on vector quantities  
 $p$  is comparable to  $b$   
 $\sigma_{mag:}$  replace b by p in Fourier transform

## **Magnetic Scattering Function**

• selection rule for magnetic scattering:

scattering function

$$\left(\frac{d\sigma}{d\Omega dE_f}\right)_{mag} = \frac{k_f}{k_i} \left(\gamma r_0 \frac{g}{2} F(\mathbf{Q})\right)^2 \sum_{\alpha\beta} \left(\delta_{\alpha\beta} - \hat{Q}_{\alpha} \hat{Q}_{\beta}\right) S_{\alpha\beta}(\mathbf{Q}, \omega)$$



- → spin fluctuations along Q do not contribute to the scattering
- → the spin components are projected on a plane perpendicular to Q

## **Magnetic Scattering Function**

• magnetic scattering function (nuclear scattering not included): (non-polarized neutrons)

$$S^{\alpha\beta}(\mathbf{Q},\omega) = \frac{1}{2\pi\hbar} \int \sum_{jj'} \left\langle S_{j'\alpha}(0)S_{j\beta}(t) \right\rangle e^{i\mathbf{Q}\cdot(\mathbf{r}_j - \mathbf{r}_{j'})} e^{-i\omega t} dt$$

 $S^{\alpha\beta}(\mathbf{Q},\omega)$  corresponds to the **Fourier transform** of the magnetic pair correlation function that gives the probability to find a magnetic moment at position  $\mathbf{r}_j$  at time *t* with a spin component  $S_{j\beta}(t)$  and the same or another moment at position  $\mathbf{r}_{j'} = 0$  at time t = 0 with a component  $S_{j'\alpha}(0)$ .

• note: projection operator  $\sum_{\alpha\beta} \left( \delta_{\alpha\beta} - \hat{Q}_{\alpha} \hat{Q}_{\beta} \right)$  is included in the scattering cross section

## **Imaginary Part of the Magnetic Susceptibility**

• fluctuation-dissipation theorem ( $\rightarrow$  W. Fischer's talk):

$$S^{\alpha\beta}(\mathbf{Q},\omega) = \frac{\hbar}{\pi} \frac{1}{1 - e^{-\hbar\omega/k_BT}} \Im\chi^{\alpha\beta}(\mathbf{Q},\omega)$$

imaginary part of the generalised susceptibility

Interpretation: The magnetic moment of the neutron acts on the sample like a frequency and wavevector dependent magnetic field  $B(Q,\omega)$ .

## **Magnetic Susceptibility**

The magnetic properties of a material (bulk, thin films etc.) can be determined by means of various techniques:

- bulk measurements:  $\chi(0,\omega)$  (susceptometer, ESR, etc.)
- nuclear techniques:  $\int \chi(\mathbf{Q},\omega) d\mathbf{Q}$  (NMR,  $\mu$ SR)
- neutron scattering:  $\chi(\mathbf{Q},\omega)$

 $\rightarrow$  Information of magnetic properties on various scales in time and space

Magnetic properties "understood" if all measurements are consistent with each other

## **Polarization Analysis**

We have seen, it is difficult to:

- separate magnetic from nuclear scattering
- distinguish between magnetic modes
- coherent from incoherent scattering

So far neglected:

- spin of the neutron
- $\rightarrow$  polarization analysis

## **Polarization analysis: Nuclear Scattering**







## **Polarization analysis: Magnetic Scattering**



 $\rightarrow$  is **not** a magnetic (dipolar) interaction  $\rightarrow$  no projection operator

 $\rightarrow$  spin-incoherence can be detected with polarized neutrons!

## **Pauli Spin Matrices**

• general structure of interactions:

$$\mathbf{\sigma} \cdot \mathbf{A} = \sigma_x A_x + \sigma_y A_y + \sigma_z A_z$$

$$\sigma_{x} = \begin{pmatrix} 0 & 1 \\ 1 & 0 \end{pmatrix}$$
$$\sigma_{y} = \begin{pmatrix} 0 & -i \\ i & 0 \end{pmatrix}$$
$$\sigma_{z} = \begin{pmatrix} 1 & 0 \\ 0 & -1 \end{pmatrix}$$

$$\sigma_{x}|\uparrow\rangle = |\downarrow\rangle \quad \sigma_{x}|\downarrow\rangle = |\uparrow\rangle$$

$$\sigma_{y}|\uparrow\rangle = i|\downarrow\rangle \quad \sigma_{y}|\downarrow\rangle = -i|\uparrow\rangle$$

$$\sigma_{z}|\uparrow\rangle = |\uparrow\rangle \quad \sigma_{z}|\downarrow\rangle = -|\downarrow\rangle$$

 $\sigma_z$  is already diagonal eigenfunctions:  $|\uparrow\rangle = \begin{vmatrix} 1\\0 \end{pmatrix}$  and  $|\downarrow\rangle = \begin{vmatrix} 0\\1 \end{pmatrix}$ 

## **Magnetic Interaction**

$$p \propto \mathbf{\sigma} \cdot \mathbf{M}_{\perp} = \sigma_{x} M_{\perp,x} + \sigma_{y} M_{\perp,y} + \sigma_{z} M_{\perp,z}$$

only spin-flip scattering possible



only non-spin-flip scattering possible for  $M_{\perp,z}$ (same is true for incoherent scattering:  $A+BI_z$ )

## **Selection Rules for Polarization Analysis**

Note: The polarization of the neutron defines the *z*-axis (so called longitudinal polarization analysis)

$$\left\langle \uparrow \left| \boldsymbol{\sigma} \cdot \boldsymbol{\breve{M}}_{\perp} \right| \uparrow \right\rangle = \boldsymbol{\breve{M}}_{\perp,z}$$
 
$$\begin{array}{c} \delta \mathbf{S} & \text{non-spin flip} \\ \bullet & \bullet & \bullet \\ \hline \boldsymbol{\nabla} \cdot \boldsymbol{\breve{M}}_{\perp} \right| \downarrow \rangle = -\boldsymbol{\breve{M}}_{\perp,z} \\ \hline \left| \boldsymbol{\sigma} \cdot \boldsymbol{\breve{M}}_{\perp} \right| \downarrow \rangle = -\boldsymbol{\breve{M}}_{\perp,z} \\ \end{array}$$

$$\left\langle \downarrow \left| \boldsymbol{\sigma} \cdot \breve{\mathbf{M}}_{\perp} \right| \uparrow \right\rangle = \breve{M}_{\perp,x} + i\breve{M}_{\perp,y} = \breve{M}^{+}$$

$$\left\langle \uparrow \left| \boldsymbol{\sigma} \cdot \breve{\mathbf{M}}_{\perp} \right| \downarrow \right\rangle = \breve{M}_{\perp,x} - i\breve{M}_{\perp,y} = \breve{M}^{-}$$

$$\left\langle \uparrow \left| \boldsymbol{\sigma} \cdot \mathbf{M}_{\perp} \right| \downarrow \right\rangle = \mathbf{M}_{\perp,x} + i \mathbf{M}_{\perp,y}$$

## "Half-Polarized" Beam Technique

see M. Blume, Phys. Rev. 130, 1670 (1963):

$$\sigma = N \cdot N^* + \mathbf{M} \cdot \mathbf{M}^* + \mathbf{P}_i \cdot (N \cdot \mathbf{M}^* + N^* \cdot \mathbf{M}) + i\mathbf{P}_i \cdot (\mathbf{M} \times \mathbf{M}^*)$$

$$M_{\perp} = \hat{\mathbf{Q}} \times (\mathbf{M}(\mathbf{Q}) \times \hat{\mathbf{Q}})$$

$$M_{\perp} = \hat{\mathbf{Q}} \times (\mathbf{M}(\mathbf{Q}) \times \hat{\mathbf{Q}})$$

$$M(\mathbf{Q}) = \int \mathbf{M}(\mathbf{r})e^{i\mathbf{Q}\cdot\mathbf{r}}d\mathbf{r}$$
nuclear scattering magnetic scattering operator chiral term

**Special case:** ferromagnet with  $|\mathbf{M}| = N$ :

• chiral term is 0

•  $P_i = -1: \sigma = 0$ 

useful for polarizing neutrons

•  $P_i = +1$ :  $\sigma$  finite
### **Rules for Polarization Analysis 1**

• nuclear scattering (excluding nuclear spin incoherence):

no Pauli spin matrices involved

 $\rightarrow$  all scattering is non-spin-flip



• (room) **background**:

 $\rightarrow$  contributes to all scattering channels

### **Rules for Polarization Analysis 2**

(special case: isotropic ferromagnet!)

• paramagnetic scattering in a vertical field:  $\mathbf{Q} \perp \mathbf{B}$ 



### **Rules for Polarization Analysis 3**



• at reasonable temperatures: (nuclear spins disordered)

$$\langle I_x^2 \rangle = \langle I_y^2 \rangle = \langle I_z^2 \rangle = \frac{1}{3}I(I+1)$$

• contribution of spin-incoherent:

$$I_{NSI}^{nsf} = \frac{1}{3}\sigma_{NSI} \qquad I_{NSI}^{sf} = \frac{2}{3}\sigma_{NSI} \qquad 38$$

### **Summary: Polarization Analysis**

	non-spin-flip	spin-flip
Q // B	$\sigma_{N} + 0\sigma_{m} + \frac{1}{3}\sigma_{NSI} + \sigma_{bg}$	$\sigma_m + \frac{2}{3}\sigma_{NSI} + \sigma_{bg}$
$\mathbf{Q} \perp \mathbf{B}$	$\sigma_N + \frac{1}{2}\sigma_m + \frac{1}{3}\sigma_{NSI} + \sigma_{bg}$	$\frac{1}{2}\sigma_m + \frac{2}{3}\sigma_{NSI} + \sigma_{bg}$

Measurement of all cross sections allows the determination of individual scattering contributions.

**Example:** Paramagnetic scattering from ferromagnetic material:

$$\frac{1}{2}\sigma_{mag} = I_{Q\perp B}^{nsf} - I_{Q//B}^{nsf}$$

$$\frac{1}{2}\sigma_{mag} = I_{Q/B}^{sf} - I_{Q\perp 3B}^{sf}$$

# 4. Ancillary equipment for polarization analysis

### **Guiding the Polarization of Neutrons**

• Larmor precession:

$$\hbar \frac{d\mathbf{S}}{dt} = \mathbf{M}_{mec} = \gamma \hbar \mathbf{S} \times \mathbf{B}$$

gyromagnetic ratio:  $\gamma_1 = -2\pi \cdot 2916 \frac{\text{rad}}{\text{Gs}} = -183.2461 \frac{\text{rad}}{\text{Ts}}$ 



• spin precession in **B**-field: 
$$\mathbf{S}(t) = S \begin{pmatrix} \cos \omega_L t \\ \sin \omega_L t \\ 0 \end{pmatrix}$$

• Larmor frequency:  $\omega_L = -\gamma B$ 

• change of phase:  $\varphi = (\varphi(t) - \varphi(0)) = \omega_L t = -\gamma_I B t = -\gamma_I B \frac{s}{v}$ 

• example: • B = 100 Gauss • s = 100 mm • v = 1000 m/s •  $\varphi = -183$  rad  $\cong 29$  revolutions

Neutron beam quickly depolarises!

41

### Adiabatic – Sudden Transitions

#### • maintain direction of polarization:

→ guide fields: define quantization axis, **B** (2 mT) >>  $\mathbf{B}_{earth}$  (0.1 mT)

adiabatic transition:

rotation of  $\mathbf{B} \ll \omega_L$ : **P** follows **B** 

sudden transition:

**B** changes abruptly,  $\rightarrow$  **P** does not follow **B** 

(Nb-shield, current sheet)

$$\begin{array}{c} \bullet B = 10 \text{ Gauss} \\ \bullet B = 1 \text{ mm} \\ \bullet v = 1000 \text{ m/s} \end{array}$$

 $\rightarrow$  Choose **B** = 0  $\rightarrow$  no precession  $\rightarrow$  no depolarisation



## **Changing Polarisation: Flat Coil Spin Flipper**



### **Principle of the Flat Coil Spin Flipper**

 $\mathbf{B}_{flip}$  is chosen such that neutron makes a  $\pi$ -rotation, when passing the flipper coil



by negative compensation

field:  $\mathbf{B}_{vert} = -\mathbf{B}_{guide}$ 

45

### **Polarized Beam Experiment**



### **Classical Polarization Analysis**

- polarization precesses around magnetic field
- guide field parallel to incident polarization will prevent depolarization!

### However:

- Depolarization of components of scattered polarization perpendicular to guide field
- only component parallel to initial polarisation can be measured
   → longitudinal polarisation analysis
- Loss of information: diagonal elements  $P_{ij}$  ( $i \neq j$ ) are lost

$$\overrightarrow{P'} = \begin{pmatrix} P^{X'} \\ P^{Y'} \\ P^{Z'} \\ P^{Z'} \end{pmatrix} = \begin{pmatrix} P_{XX} & P_{yX} & P_{zX} \\ P_{Xy} & P_{yy} & P_{zy} \\ P_{Xz} & P_{yz} & P_{zz} \end{pmatrix} \begin{pmatrix} P^{X} \\ P^{Y} \\ P^{Z} \\ P^{Z} \end{pmatrix} + \begin{pmatrix} \hat{P}^{X} \\ \hat{P}^{Y} \\ \hat{P}^{Z} \\ \hat{P}^{Z} \end{pmatrix}$$

### **Spherical Polarization Analysis**

### Solution:

- P is conserved in ZERO magnetic field
- complete polarization tensor can be measured





### MuPAD installed on TASP @ SINQ



# 5. Heusler Monochromators

### **Polarizing Single-Crystals**

Ferromagnet in saturating field **B**:

- all moments parallel to **B**
- note: **S** is antiparallel to  $\mu$

Bragg intensity of magnetic crystal:

• given by adding or subtracting *p* 

$$I(Q) \propto \left(\sum_{i} (b_i \pm p_i) e^{i\mathbf{Q}\cdot\mathbf{r}_i}\right)^2$$

• Bragg peaks appear at reciprocal lattice points  $G_{hkl}$ 

$$I_{hkl} \propto \delta(Q - G_{hkl})$$

### **Polarizing Heusler**

$$I(Q) \propto \left(\sum_{i} (b_i \pm p_i) e^{i\mathbf{Q}\cdot\mathbf{r}_i}\right)^2$$



**Heusler** (Cu<sub>2</sub>MnAl):  $\mu$  = 4.12  $\mu$ <sub>B</sub>

• flipping ratio *R*:

$$R = \frac{I_{111}^{++}}{I_{111}^{--}} = \frac{(b+p)^2}{(b-p)^2} = \frac{4b^2}{0} = \infty$$

• polarisation *P*:

$$P = \frac{R-1}{R+1} = 1$$

Compare with expression for halfpolarized set-up with N = M:

$$\boldsymbol{\sigma} = N \cdot N^* + \mathbf{M} \cdot \mathbf{M}^* + \mathbf{P}_i \cdot (N \cdot \mathbf{M}^* + N^* \cdot \mathbf{M})$$

Experiment:  $P \cong 90\% - 95\%$ 



TASP analyzer at SINQ (PSI, Roessli/Böni)

Selection of wavelength and polarization

### **Implementation:** Triple-Axis Spectrometer



Combine monochromatization with polarization.

# 6. Polarizing supermirrors

### **Total Reflection from Surfaces**

Schrödinger equation: 
$$\frac{\hbar^2}{2m} \nabla^2 \Psi + V \Psi = E \Psi$$
  
• in vacuum:  $V = 0$   $E = \frac{\hbar^2 k_0^2}{2m_n}$   
• in material:  $V = \frac{2\pi\hbar}{m} \rho(b \pm p)$   $\rightarrow E - V = \frac{\hbar^2 k^2}{2m_n}$   $\rho$ : number density  
b: coherent scattering length  
m: mass of neutron  
• refractive index:  $n^2 = \frac{k^2}{k_0^2} = 1 - \frac{V}{E}$   
 $\Rightarrow n \approx 1 - \frac{1}{2\pi} \lambda^2 \rho(b \pm p)$   $\Rightarrow \theta_c = \lambda \sqrt{\frac{\rho(b \pm p)}{\pi}}$  Example: Ni  
 $\theta_{C,Ni}(^0) = 0.1m\lambda$  (Å)

*"m* = 1"

# Total Reflection from Fe<sub>50</sub>Co<sub>48</sub>V<sub>2</sub>

(O. Schärpf, J. Penfold, 1970'ies)

$$b \cong p$$
  $\theta_c = \lambda \sqrt{\frac{\rho(b \pm p)}{\pi}}$ 

- no reflection for one spin state
- Strong reflection for opposite spin state

Film transparent for one spin state:



- free standing: transmission polarizer
- reflection angles **too small** (Mezei, Turchin) supermirrors

### **Diffraction from Multilayers**



• non-magnetic layer:  $G_{nm} = n_{nm}b_{nm}$ 

### How Does a Supermirror Work

(Turchin, Mezei 1976)



### **TEM of a Polarizing Supermirror**



produced at PSI:

60

### **Remanent Supermirrors**



(measured by: K. Pappas, HMI)

### **Towards Large m**



m -value

# **Spin Selection - Supermirror**



 $\downarrow$  short field pulse of 200 G



### Advantages:

- +high polarization: > 95 %
  - + white beam polarization
  - + remanent polarizing supermirror
    - $\Leftrightarrow$  no spin flipper necessary
  - + compact devices
- + polarizing neutron guide

### **Disadvantages:**

- diffuse scattering
- precise alignment necessary
- limited divergence
- cobalt!!

## **Polarizing Guide / Bender**

Idea: each neutron is reflected at least once:



## Implementation: TRISP @ FRM II

(T. Keller, B. Keimer @ MPI Stuttgart)





Loss: typically factor of two due to polarization
 → most powerful thermal beam at FRM-II

### **Polarizing Guide: iNSE (JAEA)**

(M. Nagao)



P = 93%

### **Focusing Spin Selector: iNSE (JAEA)**



### **Neutron Spectrometer iNSE (JAEA)**

polarizing

guide



polarizing analyser

69

coils

### Small Activation → Supermirror Fe/Si

$$G_{-} = G_{\rm nm} > 0$$



# **Transmission Polarizer:** $G_{-} = G_{nm} > 0$

stacked Si-wafers coated with polarizing supermirror



watch out: neutrons are absorbed while transmitting the stack of Si-wafers <sup>71</sup>
#### Polarizer for NSE @ SNS



#### **Transmission/Reflection of Bender**

 $\lambda = 4.7 \text{ Å}$ Bender C2, Si-Transmission 60mm som=5° 8000 8000 3500 no bender direct beam (spin up) 7000 3000 6000 neutron Counts with bender: 6000 Integrated Intensity 2500 neutron counts approx. 5000  $T \cong 71\%$ 4000 2000 4000 1500 3000 2000 1000 bender (spin up) 2000 500 0 0.5 1 1.5 1000 -2 -0.5 0 2 -1.5 -1 2θ 0 ° 0 5 0 3 0 2 4 -1 **2**θ

- transmitted beam: small divergence
- reflected beam broadened
- excellent: polarization

#### **Neutron Beam Polarization: Cavity**



λ / nm

# 7. Spin Filters: Protons, <sup>3</sup>He

#### **Properties of Spin Filter**



#### properties of filter:

- high transmission for one spin state
- zero transmission for other spin state
- does not change phase space of neutrons

### **Polarized Proton Target**

(Van den Brandt, Hautle, Konter: PSI)

• scattering is strongly spin dependent





- + homogeneous: area detectors
- + no change of phase space
- + "*E*-independent": ideal for TOF
- complicated technique

#### <sup>3</sup>He-Polarizers

• absorption is strongly spin dependent



 $\sigma \cong 0 \text{ (5 barns)}$ 

```
\sigmalarge (5925 barns Å/\lambda)
```



+ homogeneous: area detectors+ no change of phase space+ "*E*-independent": TOF

79

- complicated technique

#### **Production of Polarized <sup>3</sup>He**

#### Metastability Exchange Optical Pumping





#### **Spin Exchange Optical Pumping**







#### <sup>3</sup>He-Polarizers: MEOP



Abbildung 2: Der metastabile Zustand von <sup>3</sup>He  $(1s2s^{3}S_{1}, F = \frac{1}{2})$ , bzw.  $(1s2s^{3}S_{1}, F = \frac{3}{2})$  wird über die  $C_{8}$ - oder  $C_{9}$ -Linie auf den Zustand  $(1s1p^{3}P_{0}, F = \frac{1}{2})$  gehoben und polarisiert. Die Absorption von rechtszirkular-polarisierten Photonen führt letztlich zu einer Zunahme der magnetischen Sublevel mit hoher magnetischer Quantenzahl. [2]

## Production of Polarized <sup>3</sup>He: MEOP

(Mainz, ILL, HMI, FRM II)

**Metastability Exchange Optical Pumping** 



from: S. Masalovich, FRM II, TUM

 $P_n = \tanh(\eta P_{He}) \qquad T_n = e^{-\eta} \cdot \cosh(\eta P_{He})$ opacity:  $\eta = 0.0732 \cdot pL\lambda$ 



≅ 100%

quality factor:  $Q = P \cdot \sqrt{T}$ 

- not continuous: filters have to be exchanged
- compression necessary

#### **Production of Polarized <sup>3</sup>He: SEOP**

(NIST, Jülich, Argonne, Oak Ridge)

#### **Spin Exchange Optical Pumping: Experiment in March 06 at IPNS**



V.O. Garlea, G.L. Jones, B. Collett, W.C. Chen, T.R. Gentile, P.M.B. Piccoli, M.E. Miller, A.J. Schultz, H.Y. Yan, X. Tong, M. Snow, B.C. Sales, S.E. Nagler, W.T. Lee, C. Hoffmann

<sup>3</sup>He polarization: 60% In 2007: 70%

- continuous
- high pressure

# <sup>3</sup>He-Polarizers: Discussion



Quartz / single crystal silicon cells coated with cesium

- + white beam polarizaton
- + good efficiency for  $\lambda < 1$  Å
- wide angle analysis
   off specular reflectometry
   SANS
- + transmission geometry ⇔ straight beam path
- + no precise alignment necessary
- + decoupling of divergence and polarization
- + calibration of polarization
- decay of efficiency with time (MEOP)
- neutron beam absorption
- complicated (manpower, etc.)<sub>84</sub>

# 8. Instruments

#### **Polarized Triple-Axis Spectrometer**



Combine monochromatization with polarization.

#### **Polarized ToF-Spectrometer**



incident beam is polarized: guide, cavity

Analyzer for white beam, high energy, large area.

#### **Polarized SANS Diffractometer**



Area detector necessary  $\rightarrow$  analyzer should not disturb phase space  $\rightarrow$  <sup>3</sup>He-polarizer is well suited

#### **Neutron Spin Echo**

**Goal:** improve the *E*-resolution of a spectrometer:

- Triple axis spectrometer: insert collimations, decrease neutron energy
   → decrease of intensity, dynamic range restricted
- ToF spectrometer: shorten the neutron pulses  $\rightarrow$  decrease in intensity
- Idea: use spin degree of freedom of neutron as a clock



Energy resolution decoupled from momentum resolution

89

#### **Neutron Spin Echo – Neutron Resonance SE**

sample exchanges energy with sample:

- quasielastic scattering (diffusion)
- inelastic scattering: phonons, spin waves

 $\rightarrow \varphi_{ges}(v) \neq 0$ 

 $\rightarrow$  degree of polarization is a measure of energy transfer to neutron

Technical realisation: Resonance spin flippers

Static precession field:





#### **Technical Realisation: NSE + TAS**



# 9. Applications for Polarized Neutrons

#### **Example 1: Deuterium in Nb**

- investigation of the interstitial diffusion process
- interesting for superionic conductors (like AgI)
- deuterium:  $\sigma_{coh}$  = 5.6 barns,  $\sigma_{inc}$  = 2.0 barns

- 1<sup>st</sup> approach: theoretical separation, high quality data necessary
- 2<sup>nd</sup> approach: polarization analysis
  - → spectrometer: D7 at Institute Laue-Langevin, E<sub>i</sub> = 3.52 meV (TOF at a continuous source!)

#### **D7: From Yellow Book** @ ILL



#### **Deuterium in Nb**

$$I_{NSI}^{nsf} = \frac{1}{3}\sigma_{NSI} \qquad I_{NSI}^{sf} = \frac{2}{3}\sigma_{NSI}$$

J. C. Cook et al., J. Phys. Condens. Matter 2, 79 (1990).



96

#### **Deuterium in Nb**



- separation successful
- good agreement with theory
- incoherent-coherent residence times:  $\tau_{coh} = 0.49 \tau_{inc}$

97

#### **Ex. 2: Quantum Phase Transition in PdNi**<sub>x</sub>



# **Experiment: Radiography**

Idea: neutrons are depolarized by ferromagnetic domains



#### **Experimental Results**



Neutrons are depolarized by ferromagnetic domains  $\rightarrow$  blue regions 100

#### **Short Introduction into Spin Waves**

$$\mathbf{H} = -\sum_{jj'} J_{jj'} \mathbf{S}_{j} \cdot \mathbf{S}_{j'}$$

Fourier transform (compare with diffraction!)

• linear spin wave theory: 
$$\hbar \omega_q = 2S(J(0) - J(\mathbf{q}))$$

where: 
$$J(\mathbf{q}) = \sum_{jj'} J_{jj'} e^{i\mathbf{q}\cdot(\mathbf{r}_j - \mathbf{r}_{j'})}$$

• most simple case:  $J_{ij} = J$  (see Charles Kittel: Solid State Physics)

$$E_q = 4JS(1 - \cos qa)$$
 small  $q$   $E_q = Dq^2$ 

101



#### **Spin Waves**





#### What can we learn?

- exchange interactions: information on electronic structure
- phase transitions: critical exponents, universality
- importance of additional terms of magnetic interaction: anisotropies (xy-like, Ising)



evolves into critical scattering close to and above  $T_C$ 

#### **Example 3: Paramagnetic Scattering in Fe**



Distinction of magnetic from nuclear scattering (phonons, incoherent) difficult

### **Experiment: TAS with Pol. Analyzis**

Application of:

- vertical field  $\mathbf{B}_{v}$
- horizontal field  $\mathbf{B}_h$



Extract magnetic scattering by measuring four spindependent cross sections:

Momentum transfer:  $\mathbf{Q} = \mathbf{k}_i - \mathbf{k}_f$ 



#### **Experimental Result**


## **Interpretation of Paramagnetic Scattering**

 $\rightarrow$  no spin waves above  $T_C$ 

 $\rightarrow$  time evolution of magnetic fluctuations can be measured:

 $\rightarrow \Gamma = Aq^{2.5}$ 

→ behavior similar as a simple Heisenberg ferromagnet

$$S(\mathbf{Q},\omega) = \chi(0,0) \frac{\kappa^2}{\kappa^2 + q^2} \frac{1}{\pi} \frac{\Gamma}{\omega^2 + \Gamma^2}$$

Wicksted et al., PRB 30, 3655 (1984)



# **Example 4: Longitudinal Fluctuations**

• visualisation of spin waves (magnons) at low temperature:



• close to  $T_C$ : magnon-magnon interaction



# **Experiment: Polarized TAS**



#### **Experimental realisation:**

- Q along [200] direction  $\rightarrow$  fluctuations perpendicular to Q contribute to scattering cross section:  $\delta \mathbf{S}_z^T$ ,  $\delta \mathbf{S}_{sw}^T$
- magnetic field **B** is applied vertical to scattering plane
- **P** does not change ("non spin flip scattering"): longitudinal fluctuations  $\delta \mathbf{S}_z^T$
- **P** is inverted ("spin flip scattering"): scattering by spin waves  $\delta \mathbf{S}_{sw}^{T}$
- $\rightarrow$  measure spin flip and non spin flip scattering to distinguish the modes <sup>110</sup>

### **Experimental Result**

#### Spin waves:

#### Longitudinal fluctuations:



#### **Polarized Neutrons Necessary? YES**



Polarization analysis: Method to distinguish magnetic modes

# Ex. 5: Conventional Superconductivity (BCS)

BCS-theory: The electron-phonon interaction couples to electrons and produces a Cooper-Pair:  $\{p_{\uparrow}, -p_{\downarrow}\}$ 



## **Triple-Axis with Spin-Echo: TRISP**



# **Damping of Phonons in Pb**

Method: Triple axis spectrometer combined with neutron spin echo



Open question: Mechanism in high- $T_C$  compounds?

# **Example 6: Larmor Precession in MnSi**



#### Idea of experiment:

- measure precession of polarization in **B**-field before and after sample
- reciprocal lattice vector determines flight path of neutron through field
- time spent in magnetic field independent of wavelength of neutron
  - → number of precession for all diffracted neutrons identical



## **Thermal Expansion in MnSi**

C. Pfleiderer, P. Böni, T. Keller, U. K. Rössler, and A. Rosch, Science 316, 1871 (2007)



#### **Example 6: Chiral Fluctuations**

Recall: half polarized beam setup:



# **Magnetic Spiral in MnSi**



# Small $q \rightarrow SANS$







Roessli, Böni, Fischer and Endoh, PRL 88, 237204 (2002). paramagnetische Phase (0.5 meV) 0.30 0.70 0.80 0.90 1.00 1.10 1.20 1.30 0,20 MnSi

> Fluktuationen zeigen ebenfalls Chiralität!

# **Magnons in MnSi**

spin waves in incommensurate structures:  $E_{\blacktriangle}$  $E_{\blacktriangle}$  $E_{\perp}$ etc a qa Izyumov and Laptev helimagnet ferromagnet 450 H=0.6Tesla, T=28.2K E=1.5meV 400 Neutron Counts/15min. 350 300250 200H=0.1Tesla, T=28.2K 150E=1meV10050 B. Roessli et al., Physica 0 0.75 0.8 0.85 0.9 0.95 1.15 1.2 1.25 1.051.1B 345, 124 (2004) (q,q,q) (rlu)

## **Example 6: 3-***d* **Polarisation Analysis**

$$k = (0.017 \ 0.017 \ 0.017)$$

## **Example 7: Chirality in Multilayers**







$$\boldsymbol{\sigma}_{c} = i \mathbf{P}_{0} \cdot \left( \mathbf{S}_{bot} \times \mathbf{S}_{top} \right)$$



#### **Chirality in Multilayers**



# **10. Focusing Techniques**

## **Elliptic Guides - Large Critical Angles**



128

## **Intensity Gain with Elliptic Guides**



(C. Schanzer, P. Böni, and U. Filges, Nucl. Inst. and Meth. A 529, 63-68 (2004))

#### **Guides - Monochromators**



# **Elliptic Focusing: Set up**

• exit:

 $4 \times 8 \text{ mm}^2$ 

- largest cross-section: 10.59 x 21.17 mm<sup>2</sup>
- focal point: 80 mm



N. Kardjilov et al., NIMA 542 (2005) 248.

# **Experimental Results: Elliptic Focusing** $\lambda = 3 \text{ Å}$



collaboration with N. Kardjilov, HMI



# **Inelastic Scattering and Focusing**



- volume of sampe: 250 times smaller
- low background
- better resolution



Hradil, Mühlbauer, Böni 4. Oktober 2007

# Going to the limits



-



- volume of sampe: > 2000 times smaller
- low background
- better resolution

# **Future Applications**



Experiments are possible with very small samples (comp. synchrotron radiation):

- $\rightarrow$  extreme environment (*p*, **B**)
- $\rightarrow$  quantum phase transitions
- $\rightarrow$  combine with polarization of neutrons

## Elliptic Guides - NSE



- large beams  $\rightarrow$  low intensity
- large beams  $\rightarrow$  large divergence  $\rightarrow$  various  $\int Hds$  $\rightarrow$  decrease of resolution

# **Solution with Elliptic Guides**



- good resolution
- multi angle
- compact design

- low heat production
- compact design
- simple detector for MIEZE

# **11. Conclusions**

# Conlusions

- extraction of magnetic cross sections
- separation of magnetic modes
- itinerant antiferromagnets: Fermi surface topology
- chirality in itinerant magnets and multilayers
- next: 3-d polarization analysis of magnetic excitations
- high energy resolution with neutron spin echo
- extreme spatial resolution with Larmor diffraction
- large intensity gains possible by combining polarization analysis and focusing techniques

Missing: Polarized beam instruments at pulsed sources





# How to polarize neutrons:

- most efficient for polarizing neutrons: polarizing guides
  - $\rightarrow$  consider activation problems (Co)
- (linear) position sensitive detectors:
  - cold/thermal neutrons: supermirrors
  - hot/epithermal neutrons: <sup>3</sup>He or supermirrors with very large m
- area detectors: <sup>3</sup>He most convenient

Urgent need for polarized beam lines
in particular for time of flight instruments

### Acknowledgments

- C. Schanzer
- M. Janoschek
- S. Mühlbauer
- R. Gähler
- B. Roessli
- C. Pfleiderer
- and many others ...

**nmi** 









#### Literature

• Thomas Brückel and Werner Schweika (editors), "Polarized Neutron Scattering", Lectures of the 1st Summer School held a the Forschungszentrum Jülich from 10 to 14 September 2002. Schriften des Forschungszentrums Jülich, Volume 12, ISSN 1433-5506, ISBN 3-89336-314-9.

• G. L. Squires, "Introduction to the theory of thermal neutron scattering", Cambridge University Press, Cambridge (1978).

- R. M. Moon, T. Riste, W. C. Koehler, Phys. Rev. 181, 920 (1969).
- E. R. Pike and P. Sabatier, "Scattering and Inverse Scattering in Pure and Applied Science", edited by , S. 1242-1263, Academic Press 2002.
- W. G. Williams, "Polarized Neutrons", Oxford, New. York, 1988