



**TUCAN**

# Development of high-performance UCN polarization analyzers at J-PARC and JRR-3

nEDM2023 - The 5th Workshop on Searches for a Neutron  
Electric Dipole Moment

2023-11-07

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for the TUCAN collaboration and M. Hino

# Outline

- **Introduction**

- Principle
- Review of previous works
- Our motivations
- Test methods: UCN transmission measurement, cold-neutron reflectometry

- **Recent results**

- UCN measurement at J-PARC
- Cold-neutron reflectometry measurement at JRR-3

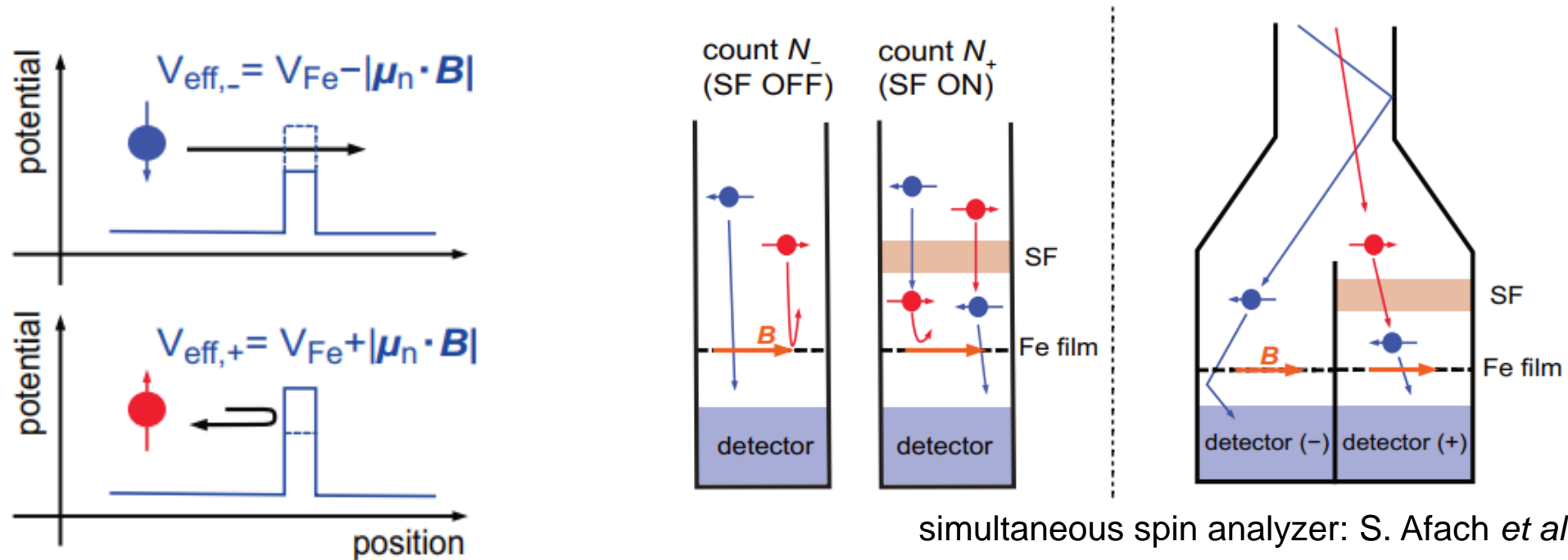
- **Summary**

# Principle

- UCN interaction with magnetic material:

$$V_{\text{eff},\pm} = V_{\text{F}} \pm |\boldsymbol{\mu}_{\text{n}}| \cdot \mathbf{B} = 209 \text{ neV} \pm 60 \frac{\text{neV}}{\text{T}} \cdot B \quad (\text{for Fe})$$

- Magnetized Fe films ( $\sim 2 \text{ T}$ ) ( $V_{+}=329 \text{ neV}$ ,  $V_{-}=89 \text{ neV}$ )  $\Rightarrow$  UCN polarizers
- Used together with spin flippers and detectors  $\Rightarrow$  UCN spin analyzers
- Fidelity of spin-state identification directly influences the visibility of the Ramsey fringes



# Review of previous works

- State-of-the art performance:  $\geq 0.90$
- Sputtering vs vacuum deposition:
  - Sputtering became more common.
  - Advantages: strong adherence of film, thin layers possible ( $\rightarrow$  low absorption)
- Common substrates: Al or Si
- Magnetic field used: vary from  $\sim 10$  mT to  $\sim 100$  mT
  - In principle, fully magnetized films are fine
  - Can confirm the performance only after UCN measurement
  - Permanent magnet system required for high field

Authors (year)	Mtehod	Substrate	Fe thickness (nm)	Magnetic field (mT)	Polarization
Egorov et al. (1974)	VD	Ti (1.5 $\mu\text{m}$ ) / cover glass (100 $\mu\text{m}$ )	200	?	$p'=0.75$
Herdin et al. (1978)	VD	NaCl	150 / 300	60	$p = 0.95$ (2) / $0.98$ (3)
Rogel (2009)	VD/IBS	Al foil (13-100 $\mu\text{m}$ )	200-1000	40	$p=0.90$
Lauer (2012)	IBS	Al foil	150	10	$p=0.96$ (3)
H�elaine (2014)	VD/IBS	Al foil (25 $\mu\text{m}$ )	400	120	$p'= 0.90$ (3), $0.91$ (3)
Baker et al. (2014)	VD	Si	1000	100	?
Zechlau (2016)	MS	Si	300	10	$p'=0.96$ (3)
Zechlau (2016)	MS	FeSi (supermirror)	15.3-128 (supermirror)	10	$p'=0.99$ (2)
Schreyer (2017)	MS	Al foil (100 $\mu\text{m}$ )	150	27	$p=0.960$ (8)

• We propose polarized cold neutron reflectometry as a test method of Fe films complementary to UCN tests

VD: vacuum deposition  
 IBS: ion-beam sputtering  
 MS: magnetron sputtering  
 p, p': see next page

# UCN transmission measurement

- Matrix formalism

Basis:  $|-\rangle = \begin{pmatrix} 1 \\ 0 \end{pmatrix}$ ,  $|+\rangle = \begin{pmatrix} 0 \\ 1 \end{pmatrix}$

Polarizer/analyzer:  $A = \begin{pmatrix} a_{11} & a_{12} \\ a_{21} & a_{22} \end{pmatrix}$ , ideally:  $\begin{pmatrix} 1 & 0 \\ 0 & 0 \end{pmatrix}$

Detector:  $D = \begin{pmatrix} 1 & 1 \end{pmatrix}$ , SFs:  $F_1 = \begin{pmatrix} \epsilon_1 & 1 - \epsilon_1 \\ 1 - \epsilon_1 & \epsilon_1 \end{pmatrix}$ ,  $F_2 = \begin{pmatrix} \epsilon_2 & 1 - \epsilon_2 \\ 1 - \epsilon_2 & \epsilon_2 \end{pmatrix}$ .

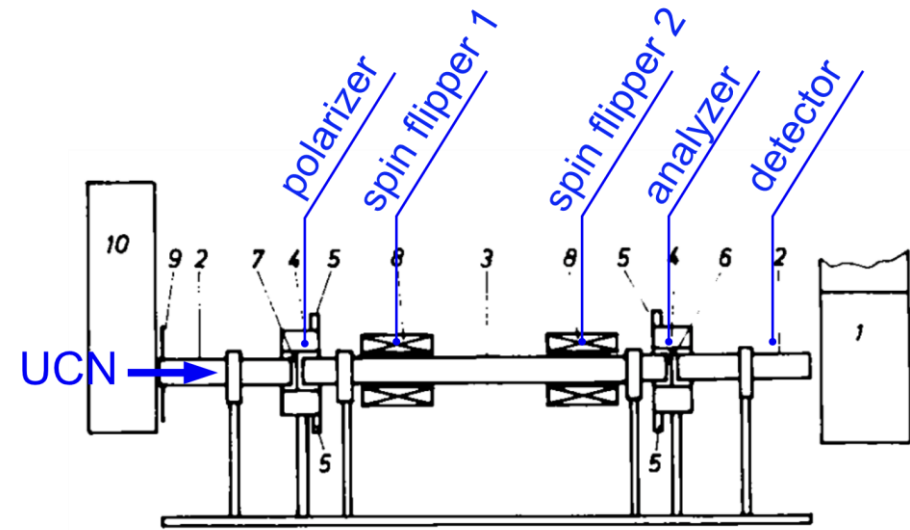
- Experimental observables: (SF1, SF2)=(0,0), (0,1), (1,0), (1,1) → 6 unknown parameters for 4 exp. configs

$$N_{ij} = DA(F_2)^j(F_1)^iA \begin{pmatrix} 1 \\ 1 \end{pmatrix} \quad (i, j \in \{0, 1\})$$

- Assumptions on  $A$ :

- Herdin et al.*  
 $A = \begin{pmatrix} a_{11} & 0 \\ a_{21} & 0 \end{pmatrix} \Rightarrow p = \frac{a_{11}}{a_{11} + a_{21}} = \frac{N_{00} - N_{10}}{f_1 \cdot N_{00} + N_{10}}; f_1 = \frac{N_{11} - N_{10}}{N_{00} - N_{01}}$

- Egorov et al.*  
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R. Herdin et al. NIM A 148, 353(1978)

J. Byrne, NIM A 167, 355 (1999) :

The assumption of Herdin et al. needs to different polarization vector amplitudes between polarizer (p) and analyzer (1).

The assumption of Egorov et al is more natural

⇒ UCN transmission experiment cannot fully distinguish different mechanisms of depolarization

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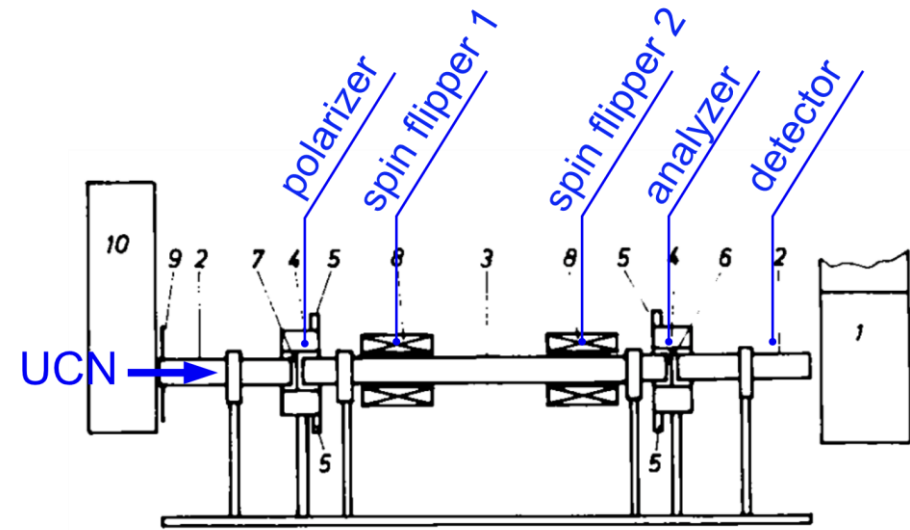
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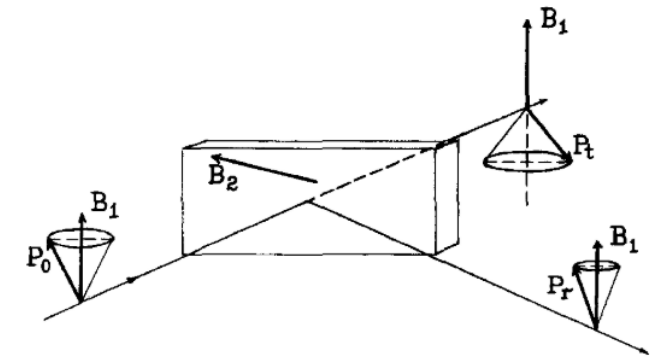
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# Polarized cold-neutron reflectometry

- Early studies on the magnetic materials with cold neutrons
- Reflection on a non-collinear magnetic induction induces depolarization  
→ averaged over domains experienced by a neutron

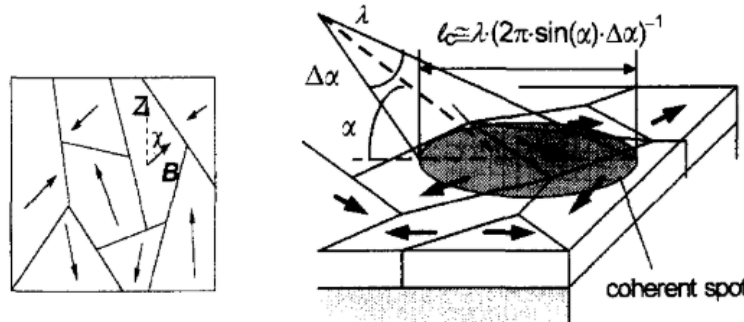


$$P = D(\mathbf{n}, \varphi) P_0,$$

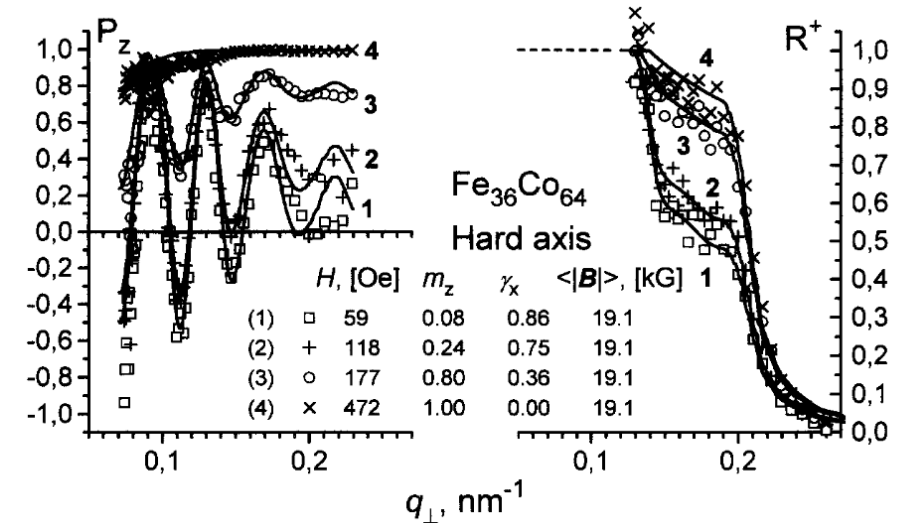
NK. Pleshanov. Z. Phys, B **94**, 233 (1994)

- Experiment with Fe<sub>36</sub>Co<sub>64</sub> film:
  - Small applied magnetic field: low reflectivity, low polarization of reflected beam  
→ increased as the film magnetized

VM. Pusenkov et al. J. Mag. Mag. Mat. **175**, 237 (1997)



$$P_z = (R_{++} - R_{+-}) / (R_{-+} + R_{+-}), R^+ = R_{-+} + R_{++}$$

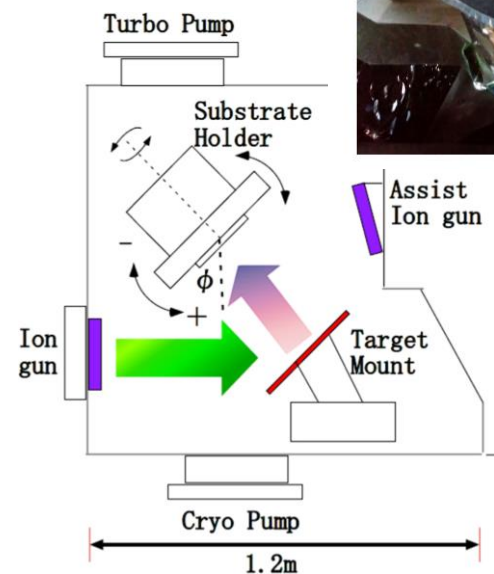
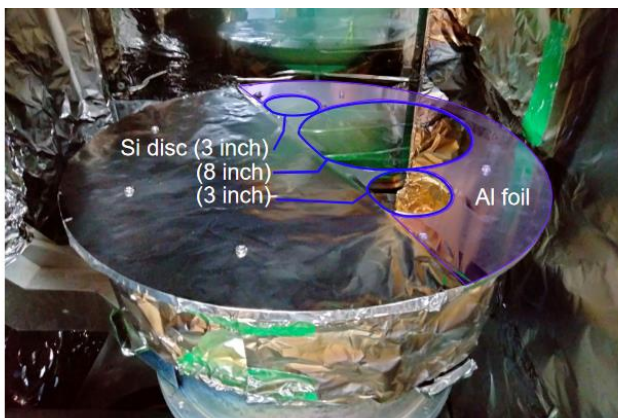


⇒ Give complementary information to the UCN measurement (in principle  $\sim (1 - a_{22})$  of matrix  $A$ )

# Our strategies

## Production

- In-house ion-beam sputtering at KURNS  
M. Hino et al. NIM A 797, 265 (2015)
- Substrate materials:  
Al (t 20–30  $\mu\text{m}$ ) or Si (t 0.1–2.0 mm)
- Fe layer thickness: 100–300 nm

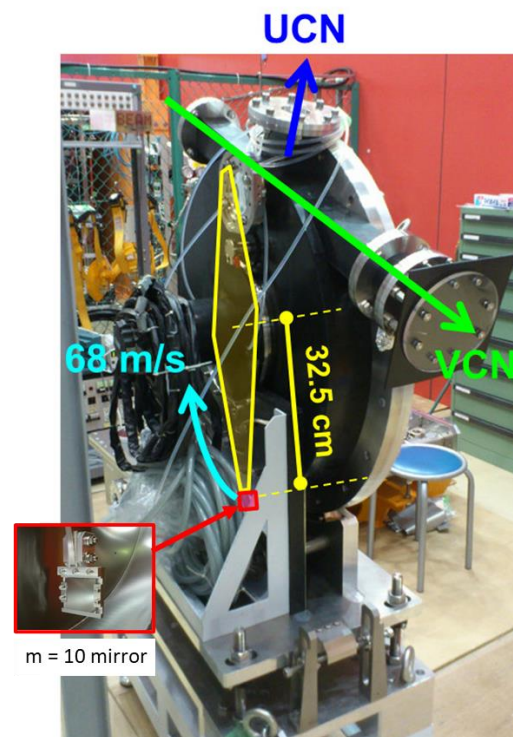


## Testing

### UCN transmission

→ Pulsed UCN source at J-PARC

S. Imajo et al. PTEP 2016, 013C22 (2016)



T. Higuchi, nEDM2023, 2023-11-07

### Cold neutron reflectometry (for Si substrate)

→ JRR-3 MINE2

( $\lambda=0.8$  nm,  $\Delta\lambda/\lambda=2.8\%$  monochromatic)



H. Akatsuka et al. JPS Conf. Proc. 37, 020801 (2022)

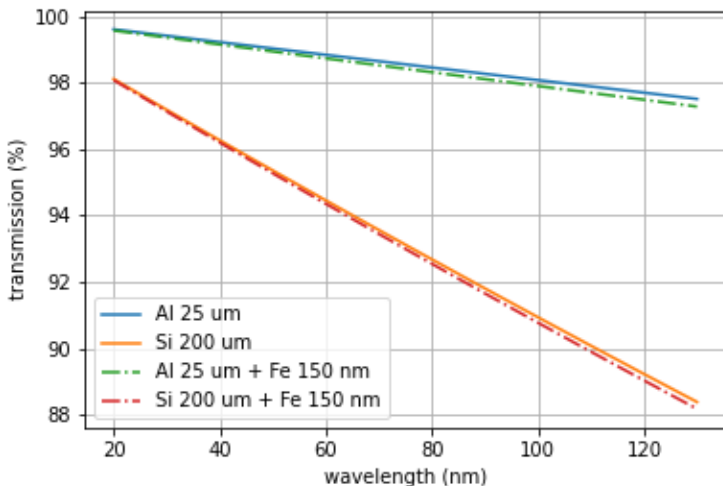


# Al and Si as substrate for Fe films

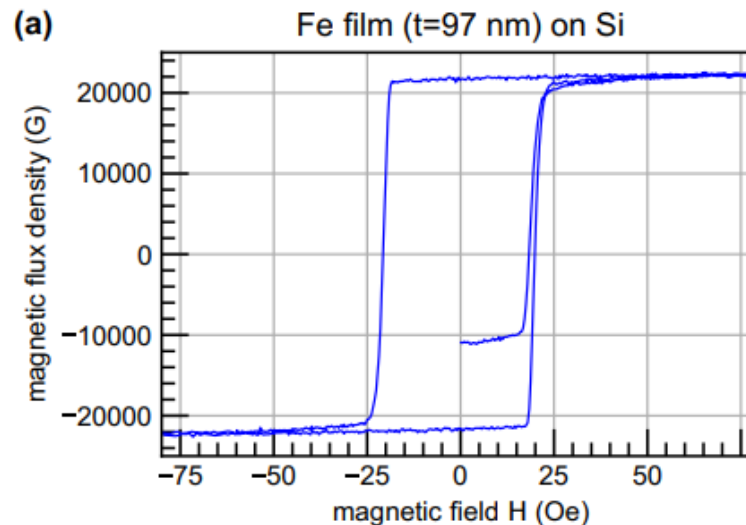
- **Si:** mirror-surface wafers available:  
 → can be characterized by cold-neutron reflectometry
- **Al:** can be made thin (~20  $\mu\text{m}$ ) → small absorption
  - Absorption for  $\lambda=58\text{ nm}$  (243 neV): Si 0.2 mm: 5.4%, Al 25  $\mu\text{m}$ : 1.2%
- **B-H curve measurements by vibrating-sample magnetometry (VSM):**  
 Fe on Si saturates more easily than Fe on Al: inverse magnetostriction effects (deformation induces magnetization)

	$V_F$ (neV)	$\sigma_{\text{abs}}$ (b) @ 2200 m/s
<b>Al</b>	53.92	0.231
<b>Si</b>	54.21	0.171

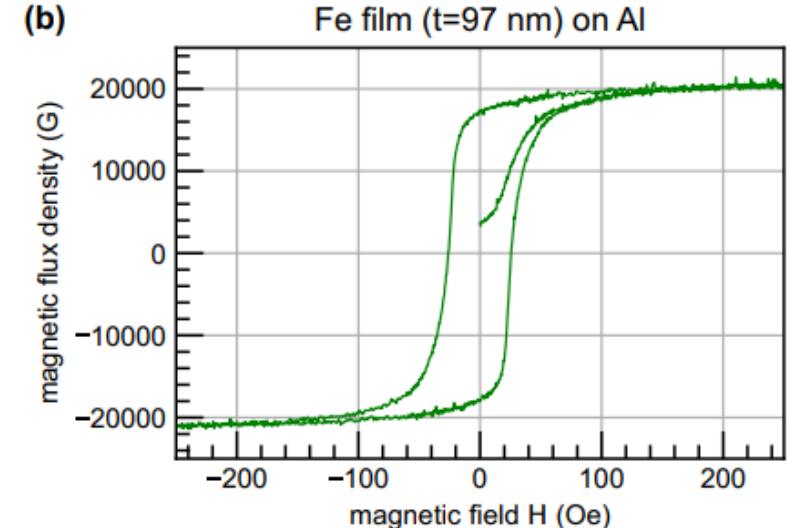
## Transmission comparison



## B-H curves



Require ~50 Oe (5 mT) for saturation



Require ~200 Oe (20 mT) for saturation

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- **Recent results**

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- Polarized cold-neutron reflectometry measurement at JRR-3

- **Summary**

# Recent results from J-PARC pulsed UCN source

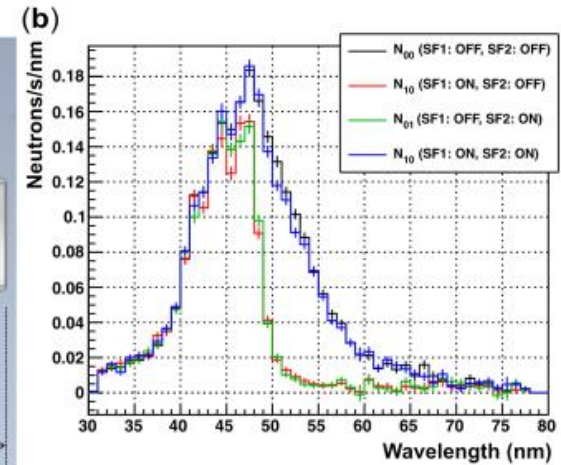
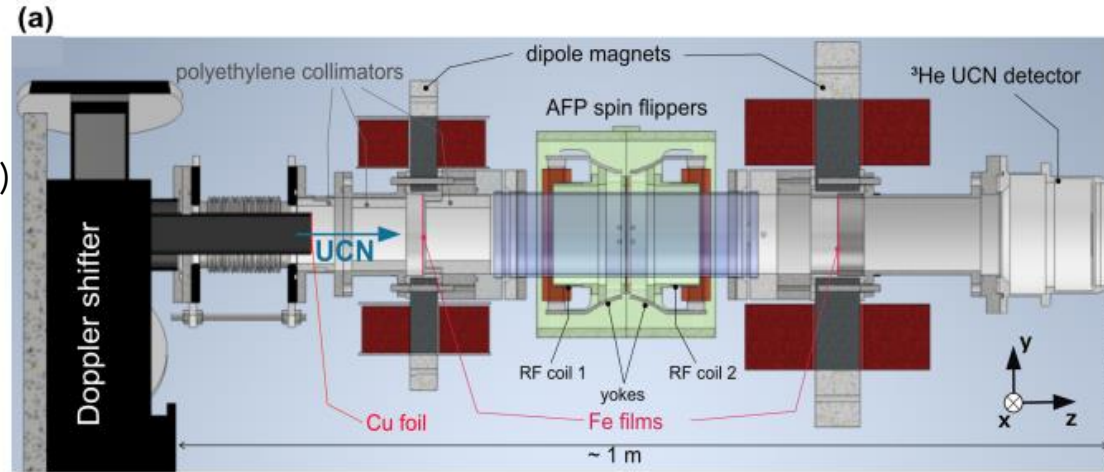
## ■ Setup:

- Doppler shifter produces UCNs every 120 ms
- Dipole electromagnets (up to 30 mT)
- Two AFP SFs

## ■ Samples:

- Fe (97 nm) on Al (25  $\mu\text{m}$ )
- Fe (97 nm) on Si (0.2 mm)

## ■ Results



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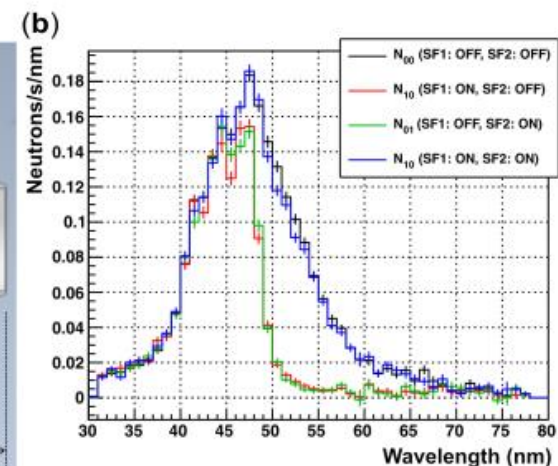
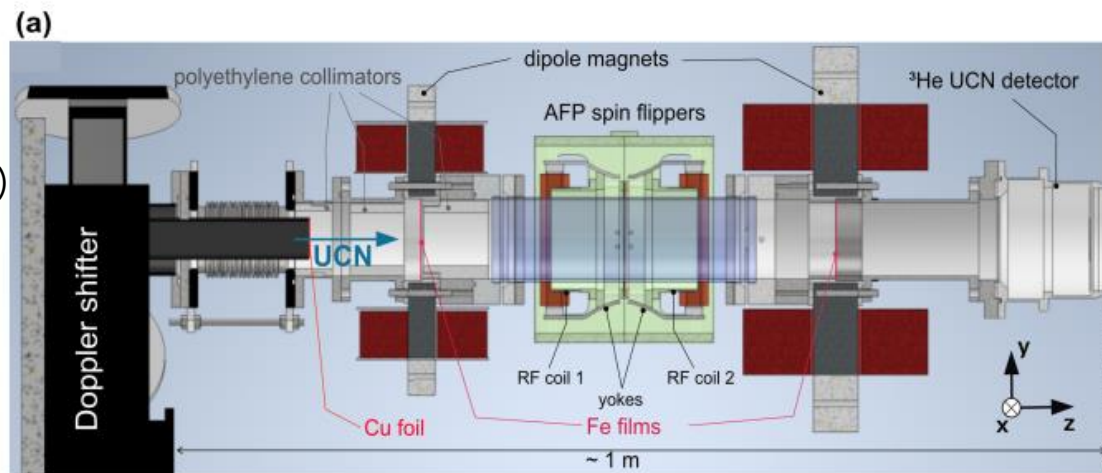
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## Results

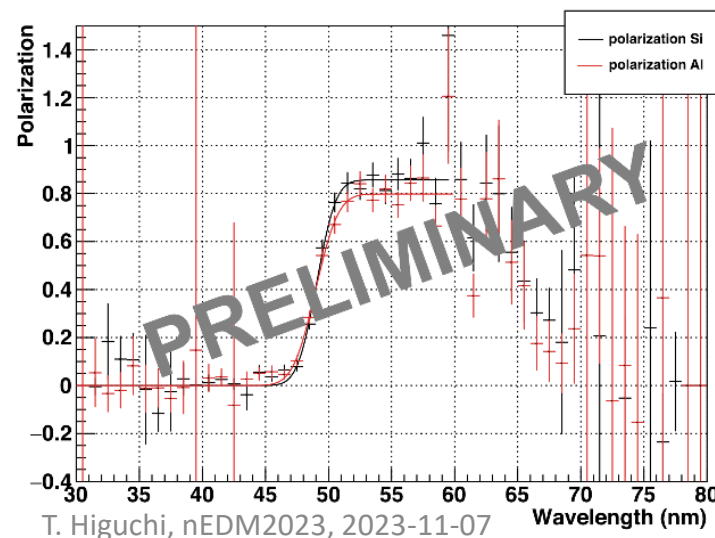
Sample	H (mT)	$\rho$	$\rho'$
Fe on Si	6	0.78 (3)	0.88 (1)
Fe on Si	12	0.85 (3)	0.92 (1)
Fe on Si	30	0.86 (3)	0.93 (1)
Fe on Al	12	0.80 (2)	0.89 (1)

\*BG neutron counts not fully accounted  
 $\rightarrow$  would increase the polarization value finally

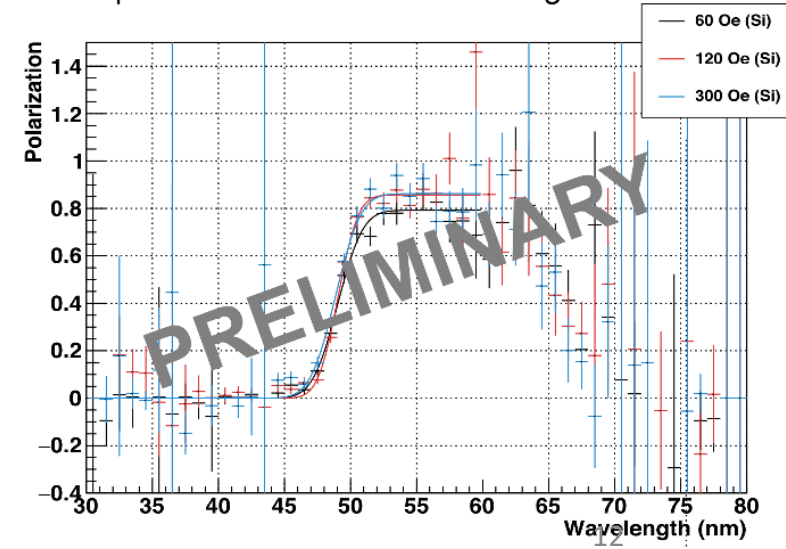
- Rise of  $\rho$  between 60 and 120 Oe for Si
- Higher  $\rho$  at 120 Oe of Si than Al (more field needed for Al)



a comparison between different samples

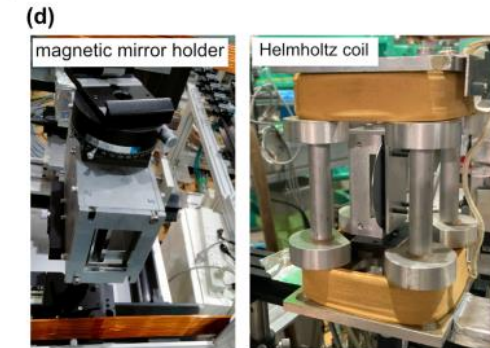
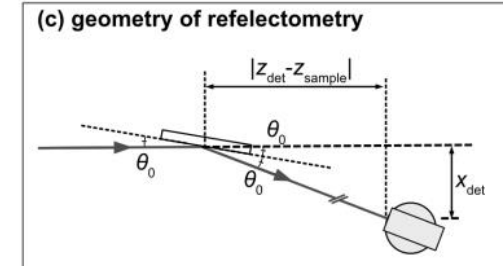
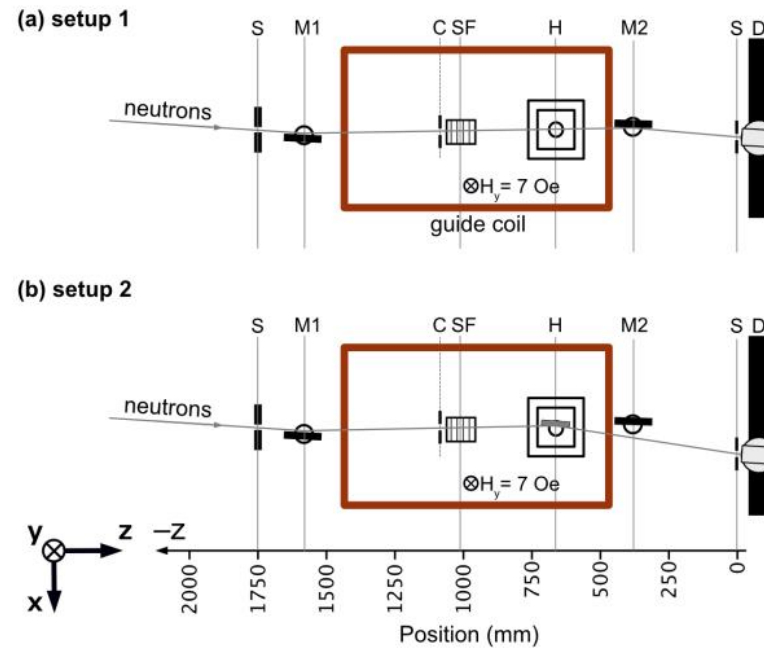


b comparison between different magnetic fields



# Recent results from JRR-3 MINE2

- **Test setup:**
  - 0.88-nm neutrons polarized by a Fe/SiGe magnetic multilayer mirror to 0.97 polarization
  - $\theta$ - $2\theta$  scan with a rotation stage and a X stage
  - Up to 5.5 mT: used a Helmholtz coil (H)
  - 30 mT: used a magnetic mirror holder (M2)
- **Sample:**
  - Fe (97 nm) on Si (2 mm)
- **Results:**



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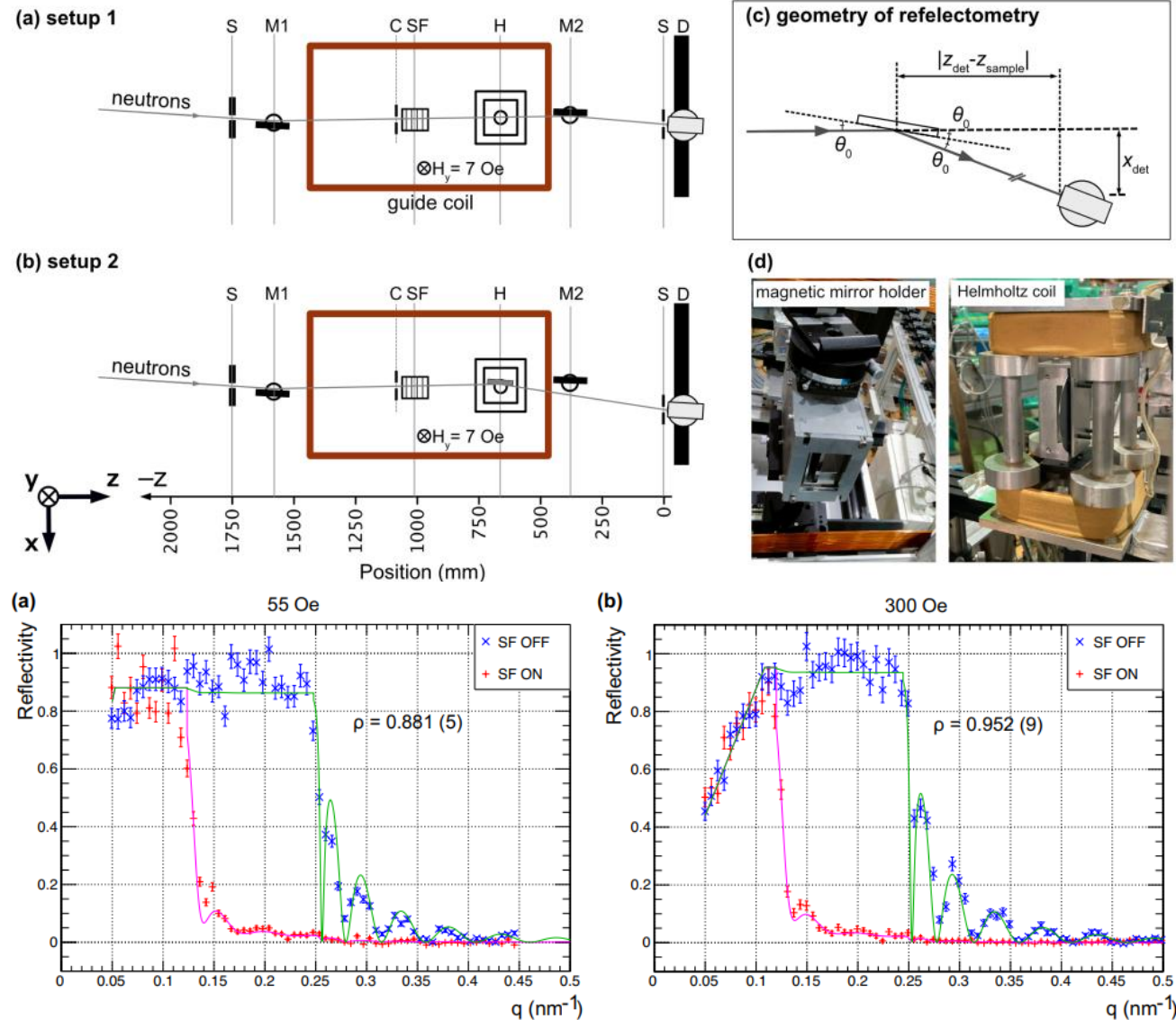
- Fe (97 nm) on Si (2 mm)

## Results:

- Data with spin on/off simultaneous fitted with a model
- The reflectivity in different applied magnetic fields

H (mT)	$R_0$
1.7	0.86 (6)
2.5	0.87 (7)
3.2	0.89 (7)
5.5	0.88 (6)
30	0.95 (9)

Rise of reflectivity between 5.5 and 30 mT



# Summary



- Introduced polarized cold-neutron reflectometry as a testing method of Fe films complementary to UCN transmission measurement
- Preliminary results imply that the magnetic field required to operate Si-substrate Fe film is larger than 5 mT as indicated by VSM
- Spin-analyzers for TUCAN EDM experiment: films with analyzing efficiency of 90% (12 mT) developed. Further evaluation & development planned

More details will be on: T. Higuchi et al. J. Phys. Soc. Jpn. (submitted)

## Acknowledgements



*JST-funded PhD position(s) available at Kyoto U.!*

***Thank you for your attention!***

# Backup



# MINE2 polarized cold-neutron reflectometry

- Fit model

$$R(q|q_{1,\pm}, q_2, d) = \begin{cases} 1 & (q < q_{1,\pm}) \\ \left| \frac{r_{10} - r_{12} \exp(iq_{1,\pm}d)}{1 - r_{12}r_{10} \exp(iq_{1,\pm}d)} \right|^2 & (q \geq q_{1,\pm}) \end{cases}$$

with  $r_{10} \equiv \frac{q - q_{1,\pm}}{q + q_{1,\pm}}$ ,  $r_{12} \equiv \frac{q_2 - q_{1,\pm}}{q_2 + q_{1,\pm}}$ . (15)

$$q_{1,\pm} = \sqrt{q^2 - \frac{8m_n V_{\text{eff},\pm}}{\hbar^2}}, \quad q_2 = \sqrt{q^2 - \frac{8m_n V_{\text{Si}}}{\hbar^2}}.$$

$R(q|q_{1,\pm}, q_2, d)$  by

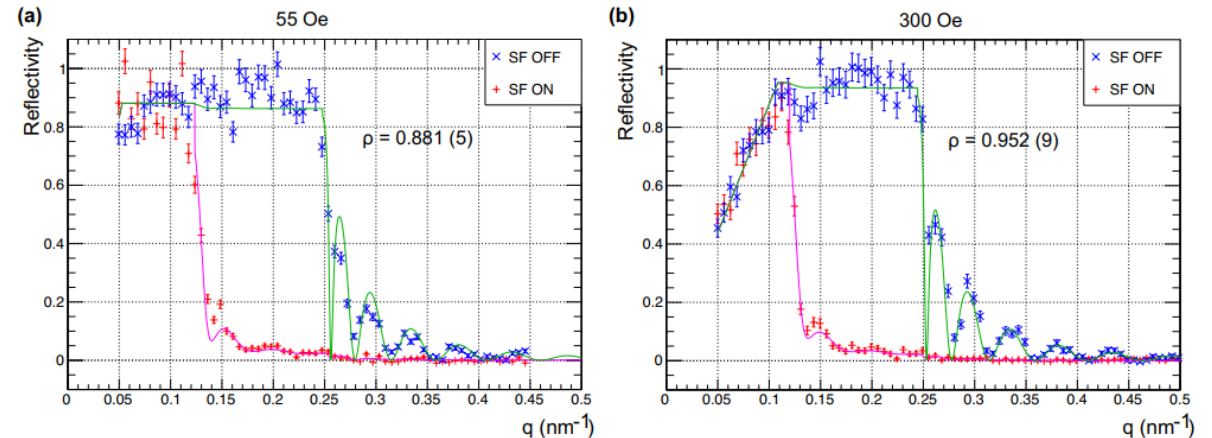
$$R_0 = \frac{1 + p_3}{2} R(q|q_{1,+}, q_2, d) + \frac{1 - p_3}{2} R(q|q_{1,-}, q_2, d)$$

$$R_1 = \frac{1 - p_3}{2} R(q|q_{1,+}, q_2, d) + \frac{1 + p_3}{2} R(q|q_{1,-}, q_2, d).$$

$$\eta(q|q_{\text{th}}) = \begin{cases} q/q_{\text{th}} & (q < q_{\text{th}}) \\ 1 & (q \geq q_{\text{th}}). \end{cases}$$

$$R'_0(q|q_{1,+}, q_{1,-}, q_2, q_{\text{th}}, d, p, \rho) = \rho \eta(q|q_{\text{th}}) \left[ \frac{1 + p_3}{2} R(q|q_{1,+}, q_2, d) + \frac{1 - p_3}{2} R(q|q_{1,-}, q_2, d) \right], \quad (18)$$

$$R'_1(q|q_{1,+}, q_{1,-}, q_2, q_{\text{th}}, d, p, \rho) = \rho \eta(q|q_{\text{th}}) \left[ \frac{1 - p_3}{2} R(q|q_{1,+}, q_2, d) + \frac{1 + p_3}{2} R(q|q_{1,-}, q_2, d) \right]. \quad (19)$$



# MINE2 polarized cold-neutron reflectometry

- Fit results

$H$ (Oe)	$q_{1,+}$ (nm <sup>-1</sup> )	$q_{1,-}$ (nm <sup>-1</sup> )	$q_2$ (nm <sup>-1</sup> )	$q_{th}$ (nm <sup>-1</sup> )	$d$ (nm)	$\rho$	$p_3$	$\chi^2/ndf$
17	0.1275 (4)	0.2472 (1)	0.117 (3)	0.059 (1)	96.5 (2)	0.855 (5)	0.935 (4)	855/123
25	0.2470 (1)	0.1225 (7)	0.109 (7)	0.052 (1)	96.1 (2)	0.870 (6)	0.891 (5)	808/123
32	0.2475 (1)	0.1255 (6)	0.110 (7)	0.058 (1)	97.8 (2)	0.890 (6)	0.910 (4)	838/123
55	0.2479 (1)	0.124 (2)	0.110 (2)	0.053 (2)	97.2 (2)	0.881 (5)	0.958 (4)	918/123
300	0.2445 (2)	0.1200 (5)	0.110 (4)	0.108 (2)	95.5 (3)	0.952 (9)	0.962 (4)	394/139

# VM. Pusenikov et al. J. Mag. Mag. Mat. 175 , 237 (1997)

$$P = D(n, \varphi) P_0,$$

$$\varphi = \varphi_+ - \varphi_-,$$

$$\varphi_+ = 2 \arccos\{[E_{\perp}/V^+]^{1/2}\} \quad (E_{\perp} < V^+),$$

$$\varphi_- = 2 \arccos\{[E_{\perp}/V_g]^{1/2}\} + 2k_{\perp}d$$

$$(V^- < E_{\perp} < V_g).$$

$$P_z = D_{zz} = \langle 1 - n_x^2 (1 - \cos(\varphi)) \rangle.$$

$$m_z \equiv \langle n_z \rangle = \langle f(\chi) \cos(\chi) \rangle = \langle B_z \rangle / B,$$

$$\gamma_x \equiv \langle n_x^2 \rangle = (1 - \langle f(\chi) \cos(2\chi) \rangle) / 2.$$

$$R^+ = R_{-+} + R_{++} \cong \langle \cos^2(\chi/2) \rangle = \frac{1}{2} (1 + m_z), \quad (9a)$$

$$P_z \cong 1 - \frac{1}{2} \langle \sin^2(\chi) \rangle \langle \cos^{-2}(\chi/2) \rangle$$

$$= 1 - \gamma_x / (1 + m_z). \quad (9b)$$

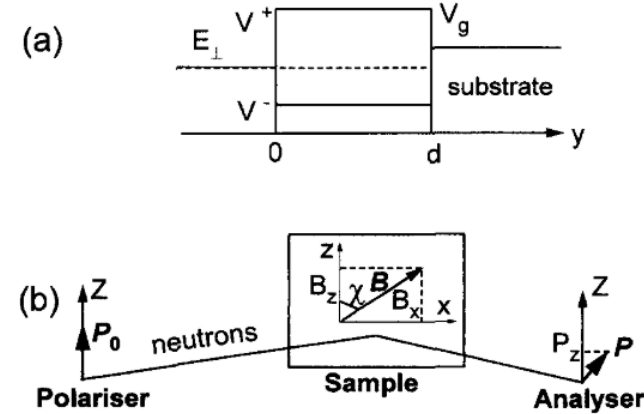


Fig. 1. (a) The potentials of a magnetic film. (b) The experimental scheme. The directions of the quantisation axis ( $Z$ ), the guide field ( $H$ ), and the magnetic induction in the film ( $B$ ) are shown; the coordinate system ( $x, y, z$ ) is connected with the sample.

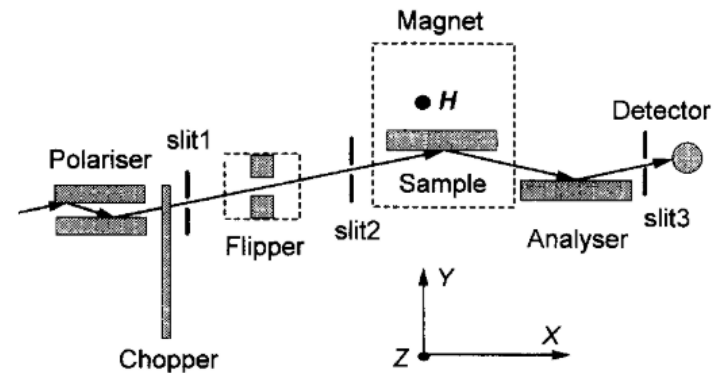


Fig. 4. A scheme of the neutron reflectometer ZINA.

# J. Byrne, NIM A 167, 355 (1999)

- Introduced 3-dimensional polarization vector ( $\leftrightarrow$  Stokes vector of light polarization)
- Re-formulate the transfer matrix in three-dimensional form, T: polarizer/analyzer, R: spin flipper

$$P_0 = I, \quad P = P_0 \langle \sigma \rangle = P_0 \mathbf{p}$$

$$P' = TP$$

$$\mathbf{p}_p = \frac{T_{x0}}{T_{00}}, \frac{T_{y0}}{T_{00}}, \frac{T_{z0}}{T_{00}}, \quad \mathbf{p}_a = \frac{T_{0x}}{T_{00}}, \frac{T_{0y}}{T_{00}}, \frac{T_{0z}}{T_{00}}.$$

$$R = \begin{pmatrix} 1 & 0 \\ 0 & M \end{pmatrix}$$

$$P' = TRTP$$

$$P'_0(0, \phi) = C_1 \quad \text{and} \quad P'_0(\pi, \phi) = C_2$$

$$\frac{C_1 - C_2}{C_1 + C_2} = \frac{T_{z0} T_{0z}}{T_{00}^2} = \mathbf{p}_p \cdot \mathbf{p}_a.$$

The result [eq. (1)] of Egorev et al.<sup>2)</sup> follows immediately from the assumption  $\mathbf{p}_p \equiv \mathbf{p}_a$  whereas the results of Herdin et al.<sup>2)</sup> hold only in the limit that  $\mathbf{p}_a$  and  $\mathbf{p}_p$  are parallel and that  $|\mathbf{p}_a| \equiv 1$ . This implies that the active element, although an imperfect polarizer is yet a perfect analyser. Such a notion, although tenable in theory, seems somewhat unlikely in practice. Indeed one might comment that, since Herdin et al.<sup>1)</sup> limit their discussion to a formulation in terms of  $2 \times 2$  transfer matrices, effectively suppressing the  $x$  and  $y$  components of the polarization, the possible non-parallelism of  $\mathbf{p}_a$  and  $\mathbf{p}_p$  lies outside the range of their treatment.

# Kim & Oliveria, J. Appl. Phys. 74, 1233–1241 (1993)

- Dependence of magnetic properties of sputtered Fe film

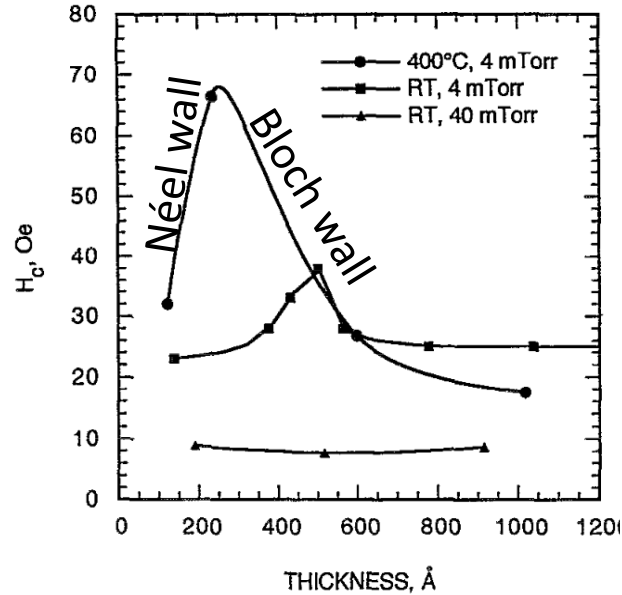
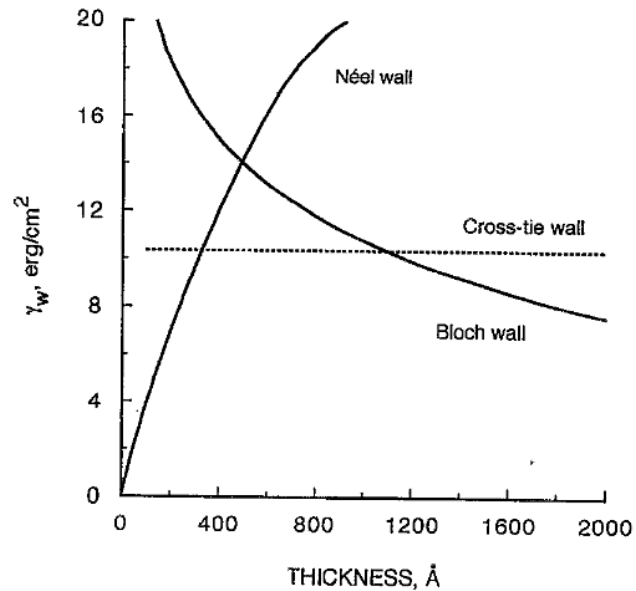
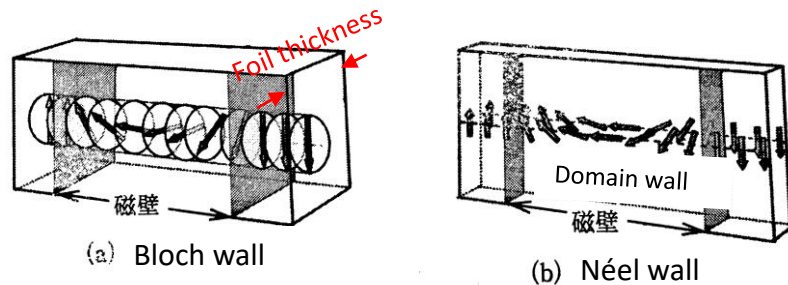


FIG. 11. Coercivity,  $H_c$ , as a function of film thickness



Critical thickness  $\delta$ :  
 $\delta \sim \sqrt{A/K}$   
 $A$ : exchange stiffness,  
 $K$ : magnetic anisotropy constant  
 For Fe,  $\delta \sim 40$  nm

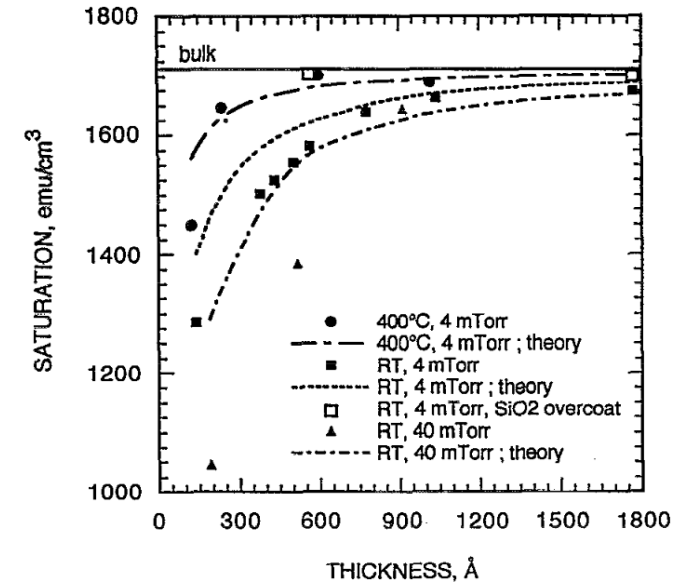


FIG. 4. Saturation magnetization,  $M_s$ , as a function of film thickness.

$$M_s^{\text{tot}}(\text{theory}) = pM_s^{\text{Fe}}(\text{bulk}) + (1-p)M_s^{\text{Fe}_3\text{O}_4}(\text{bulk})$$