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Neutron magnetic resonance and non-specular reflection from a magnetic film placed in an oscillating magnetic field

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Abstract. Experimental results on neutron reflection from a magnetic film placed in an oscillating magnetic field are reported. We found evidence for neutron spin resonance in the film and observed a spatial beam-splitting. The beam-splitting finds its origin in the exchange of an energy quantum $\hbar\omega$ between the oscillating field and the neutron.

1. Introduction

Acceleration and deceleration of neutrons in a magnetic field were experimentally demonstrated in the case of a constant [1] and an oscillating [2] magnetic field. In 1978, V.K. Ignatovich predicted [3] the effect of spatial beam-splitting upon reflection and refraction from media in which the magnetic induction is non-collinear with respect to the applied magnetic field. This effect was observed for reflected [4,5] and refracted [6] beams. The magnitude of the beam-splitting and its relation to the sign of the spin-flip transitions corresponded to the gain or loss of Zeeman energy by the neutron during a spin-flip event in a rather large applied magnetic field (~ 10 kOe). In references [7-9], beam-splitting upon reflection and refraction in a low magnetic field (about 100 Oe) applied parallel to the film surface was observed. In this case the magnitude of the beam-splitting and the types of neutron spin transitions involved found their origin in the presence of magnetic domains and domain walls with non-collinear magnetization.

Recently, spatial beam-splitting upon reflection and refraction was predicted [10-14] in the case of a magnetic film placed in an oscillating magnetic field. We report here on the experimental observation of this effect.

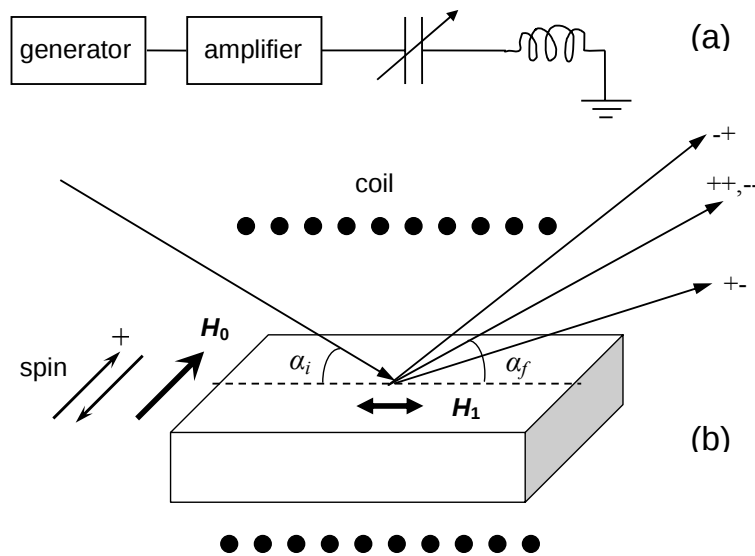


Figure 1. Experimental setup. (a) Schematic of the electric circuit generating the oscillating field. (b) Geometry of the experiment: H_0 is the constant magnetic field and H_1 is the oscillating magnetic field produced by the rectangular coil.

2. Experimental setup

The experimental setup is presented in figure 1. A radio-frequency generator (Rohde&Schwarz SMC100A) and a power amplifier (100 W CW, American Microwave Technology M3200/M3000) were used to create an oscillating field in a resonant LC circuit. The adjustable capacity could be varied in the range 5 – 100 pF. The magnetic coil had a length of 50 mm, a cross-section of $5 \times 30 \text{ mm}^2$, and consisted of 50 turns of 1 mm diameter Cu wire.

The experiment was conducted at the polarized neutron reflectometer N-REX⁺ (Forschungsneutronenquelle Heinz Maier-Leibnitz, FRM II, Garching, near Munich, Germany) with the sample surface oriented horizontally. The neutron wavelength λ was 4.26 Å (FWHM = 1 %), the angular resolution of the incident beam was 0.006°. A supermirror in transmission mode was used as polarizer. The polarization of the incident beam was 97 %. The polarization of the reflected neutrons was not analyzed. A Mezei type spin-flipper of 100 % efficiency was used to flip the beam polarization. The reflectivity was measured using a two-dimensional ³He position-sensitive detector (PSD) with a spatial resolution of 3 mm. The distance between the collimating diaphragm and the sample was 2200 mm, the distance between the sample and the detector was 2500 mm. The sample was a permalloy (Py) film of 5000 Å thickness deposited on a Si substrate with a size of $25 \times 25 \times 1 \text{ mm}^3$. The nominal composition of the permalloy was Fe(20.6 % at.)Ni(79.4 % at.).

The geometry of the experiment is presented in figure 1b. The constant magnetic field $H_0 = 20 \text{ Oe}$ was produced by an electromagnet and applied perpendicular to the reflection plane. The polarization of the incident neutron beam was directed parallel ('+') or antiparallel ('-') with respect to this quantization field H_0 . The sample was mounted on a glass holder inside the magnetic coil. The oscillating magnetic field $H_1(t) = H_1 \cos \omega t$ was applied in the film plane, perpendicular to the constant magnetic field H_0 and the neutron spin. The amplitude of the oscillating magnetic field was $H_1 = 10 \text{ Oe}$. The frequency of the magnetic field was varied in the interval $f = 25 \dots 35 \text{ MHz}$. The grazing incidence angle of the neutron beam was fixed at $\alpha_i = 0.4^\circ$. The reflected neutrons were measured as a function of the exit angle using the position-sensitive detector. The magnetic state of the sample was prepared by saturating it in an H_0 field of 1000 Oe and subsequently releasing the field to 20 Oe, where it was held throughout the measurements.

3. Sample characterization

The sample was characterized by polarized neutron reflectometry (PNR) and vibrating sample magnetometry (VSM) in CEA Saclay, France. In figure 2, the hysteresis loop of the film is presented. The film was close to magnetic saturation above 500 Oe and reached the full saturation equal to 9.5 kG for an applied field of 4 kOe. The coercive field of the film is of the order of about ± 60 Oe. The film is partially demagnetized for low fields and the average magnetization is 4 kG for an applied field of 20 Oe. VSM measurements show that in this state there is a small magnetization component perpendicular to the film plane (data not shown here). The fact that a small perpendicular anisotropy appears in thick permalloy films is documented in literature [15].

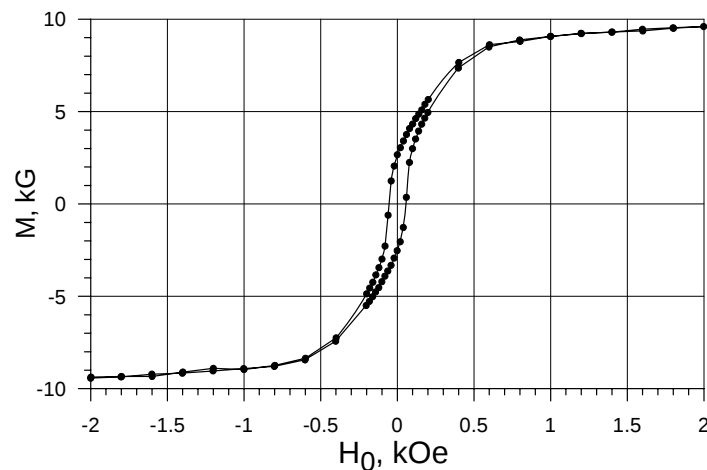


Figure 2. Hysteresis loop measured by VSM.

The characterization by neutron reflectometry was carried out at the polarized neutron reflectometer PRISM [16] installed at the reactor ORPHEE (LLB, Saclay, France). The neutron wavelength was set to 4.0 Å (FWHM = 0.37 Å), the angular resolution was 0.04°. In figure 3a, the spin-resolved experimental reflectivities R^{++} (solid symbols) and R^{--} (open symbols) are shown as a function of the vertical component of the wave-vector of the incident beam, $q = 2\pi \sin \alpha_i / \lambda$, for an applied field of 20 Oe (after saturation in a field of 4 kOe). The dashed line indicates the value of q corresponding to $\lambda = 4.26$ Å and $\alpha_i = 0.4^\circ$ as used during the resonance experiment performed at N-REX⁺. The solid lines correspond to fits obtained with the program *SimulReflec* [17]. The fit result $U = 211.2$ neV for the nuclear potential corresponds to 89 % of the theoretical value $U = 237.3$ neV for the nominal sample stoichiometry Fe(20.6 % at.)Ni(79.4 % at.). The layer density is reduced during the sample preparation which is the most likely explanation since reduced densities are usually observed for sputtered films. The fit result for the magnetization is $M = 4.0$ kG. This value is in agreement with the VSM data.

The reflectivities for a higher external magnetic field of 4.4 kOe are shown in figure 3b. The fit result for the nuclear potential U was the same as for the low field. The fit result for the magnetization is $M = 9.0$ kG. This is again very close to the VSM data. One can conclude that during the resonance experiments performed at $H_0 = 20$ Oe the sample was partially demagnetized, and that it was initially saturated to a magnetization of about 9.0 kG by applying a high magnetic field of 1000 Oe.

4. Results

The first proof-of-principle experiment on beam-splitting at reflection from a permalloy film (2500 Å) placed in an oscillating magnetic field was carried out in 2006 at the time-of-flight polarized neutron reflectometer REMUR at the reactor IBR-2 (FLNP, JINR, Dubna, Russia). At a fixed frequency of

33.5 MHz, the neutron spin resonance was examined by scanning the constant external magnetic field in the range 100 – 6000 Oe. These results are not discussed here and will be published elsewhere.

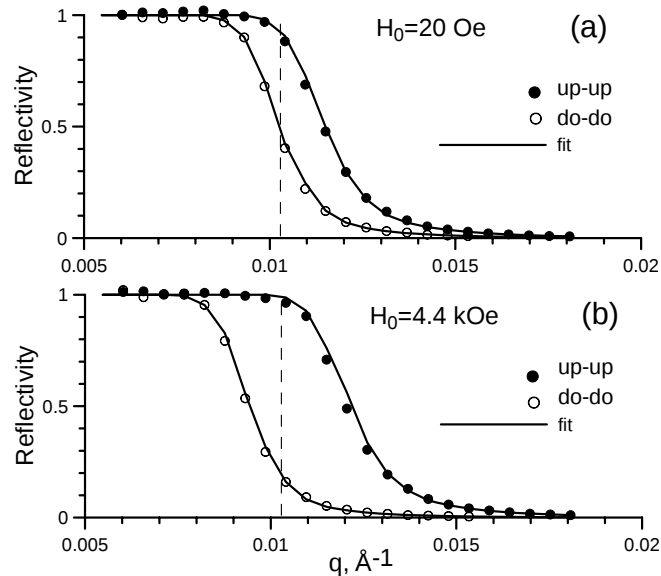


Figure 3. Experimental neutron reflectivities in the up-up (solid symbols) and down-down (open symbols) channel for a neutron wavelength of 4.0 Å. Solid lines are fit curves obtained with the program *SimulReflec* [17].

(a) demagnetized state in a field of 20 Oe; (b) saturated state in a field of 4.4 kOe. The dashed line corresponds to an incidence angle of 0.4° and a neutron wavelength of 4.26 Å as used during the resonance experiments at N-REX⁺.

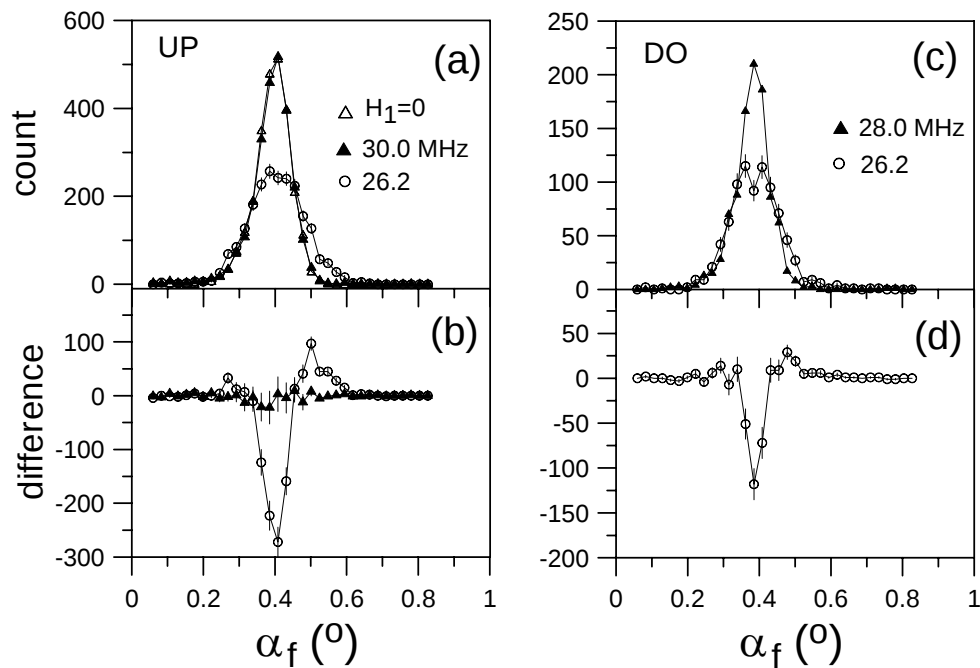


Figure 4. (Top row) Neutron counts versus reflection angle α_f without oscillating field ($H_1=0$ Oe) and with oscillating field ($H_1=10$ Oe) for the frequencies 26.2, 28.0 and 30.0 MHz. (Bottom row) Difference datasets obtained from the data plotted in the upper panels by subtraction of reference exit angle profiles as follows: In the spin up case (left column) the measurement for $H_1=0$ Oe was subtracted, and in the spin down case (right column) the measurement for 28 MHz was subtracted. Spin up and spin down refer to the polarization of the incident beam. The measuring time was 20 s per point.

In the experiment reported here, we scanned the neutron resonance by variation of the frequency of the oscillating magnetic field. In figure 4, the reflected intensity is plotted as a function of the

reflection angle α_f for different oscillating magnetic fields. The measuring time was 20 s per point. Left and right panels correspond to up and down polarization of the incident beam, respectively. Figures 4a and 4c show raw neutron counts as a function of the reflection angle α_f for different frequencies (26.2, 28.0 and 30.0 MHz) of the oscillating field ($H_1 = 10$ Oe) and without an oscillating field ($H_1 = 0$ Oe). Figure 4b shows the difference of the signals obtained with oscillating field (26.2 and 30.0 MHz) and without oscillating field ($H_1 = 0$ Oe) for spin up polarization of the incident beam. Figure 4d shows the difference of the signals obtained at 26.2 MHz and at 28.0 MHz for the spin down case. In figure 4 one can distinguish three exit angle regimes:

- $\alpha_f = 0.338 \dots 0.455^\circ$, specular region ($\alpha_f \cong \alpha_i$)
- $\alpha_f = 0.199 \dots 0.338^\circ$, off-specular region I ($\alpha_f < \alpha_i$)
- $\alpha_f = 0.455 \dots 0.641^\circ$, off-specular region II ($\alpha_f > \alpha_i$)

One can see that the spin up signal at the frequency 30.0 MHz coincides with the signal without the oscillating field ($H_1 = 0$). At the frequency 26.2 MHz the signal decreases in the specular region and increases in the off-specular regions, in particular in region II. This observation holds for both initial beam polarizations.

The reflected signals shown in figure 4, namely the integral number of counts and the peak intensity in the specular region, are plotted as a function of the frequency of the oscillating field in figure 5. Left and right panels correspond to up and down polarization of the incident beam, respectively. One can identify a resonance feature at the frequency $f_{\text{res}} = 26.2$ MHz at which the reflected signal is minimized. The full width at half maximum (FWHM) of the resonance is $\Delta f_{\text{res}} = 1.2$ MHz. The relative signal reduction at the resonance is similar for up and down polarization of the incident beam: 25 % in case of the integrated counts and about 60 % in case of the peak maximum.

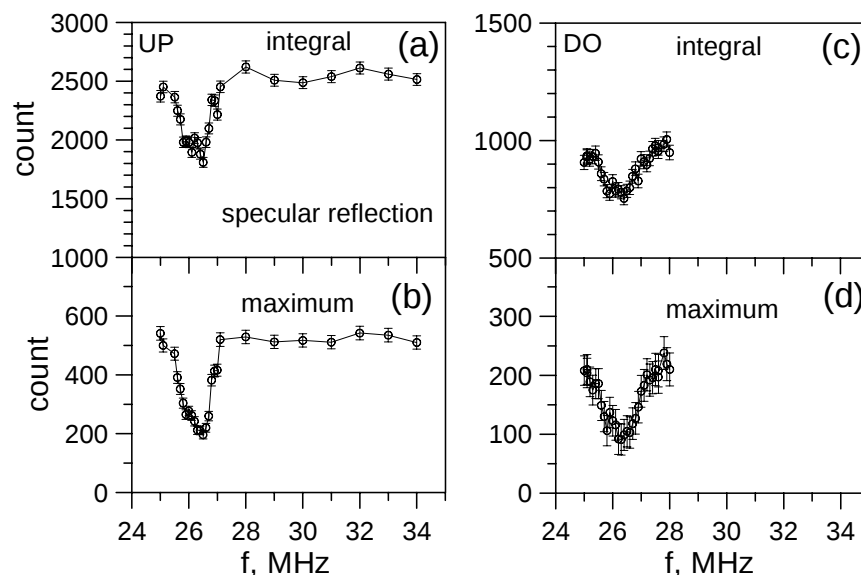


Figure 5. Reflected number of neutrons as a function of the frequency of the oscillating magnetic field. (top) integrated reflected signal, (bottom) maximum of the reflected peak, (left column) initial polarization spin up, (right column) initial polarization spin down.

The frequency dependence of the integrated signals as shown in figures 5a and 5c for the specular region is plotted again for the case of integration over the two off-specular regions I and II in figures 6a and 6b, for up and down polarization of the incident beam, respectively. Closed symbols

correspond to region I ($\alpha_f < \alpha_i$), open symbols to region II ($\alpha_f > \alpha_i$). As opposed to figures 5a and 5c, however, reference datasets (obtained without oscillating field for spin up and for an oscillation frequency of 28.0 MHz for spin down, respectively) have been integrated over regions I and II and the results subtracted as a baseline before plotting the data in figure 6. Let us quantify our results by normalizing the integrated off-specular excess counts at the resonance as plotted in 6a and 6b by the integrated specular counts outside the resonance as plotted in figures 5a and 5c (i.e. 2500 counts for spin up, 1000 counts for spin down). In these terms, the spin-flip effect amounts to 3 % for region I ($\alpha_f < \alpha_i$) and 12 % for region II ($\alpha_f > \alpha_i$), both for up and down polarization of the incident beam. Similar numbers are obtained when comparing the neutron counts at the peak maxima of the specular and non-specular reflected beams accordingly. In this case, however, the statistical errors are larger since the contributing numbers of neutron counts are lower.

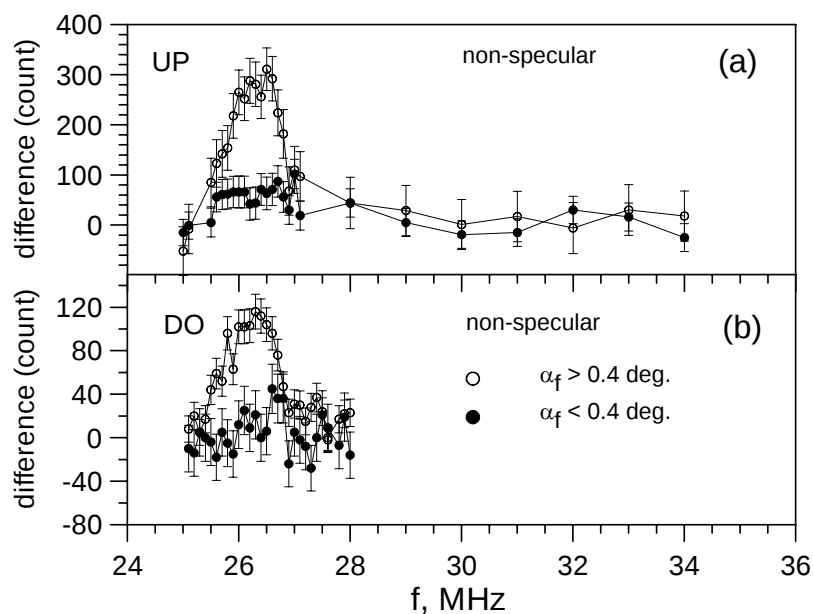


Figure 6. Excess counts of non-specularly reflected neutrons integrated over the exit angle intervals 0.455° - 0.641° (open symbols) and 0.199° - 0.338° (solid symbols) as a function of the frequency of the oscillating magnetic field. These excess counts were determined from the integrated raw data by subtraction of the corresponding integrated counts measured (a) without oscillating field for spin up and (b) at an oscillation frequency of 28.0 MHz for spin down polarization of the incident beam, respectively.

The observed fact is that the resonant off-specular intensities are proportional to the respective off-resonance specular reflectivities. A detailed quantitative explanation of the origin of this observation requires a more careful theoretical consideration. In this context it has to be investigated if the resonance phenomenon can be treated as a perturbation or if this observation is related to more general concepts such as particle flux conservation.

Having concluded that reflected neutrons are partially redirected into non-specular directions close to the resonance condition, we will now discuss the value of the measured resonance frequency f_{res} . It is clear that a neutron spin-flip should take place inside the film when the frequency of the applied oscillating field is close to the neutron Larmor precession frequency f_L inside the magnetic film. This Larmor frequency depends on the magnetic induction B inside the film as $f_L = \gamma B$, where $\gamma = 2\mu/\hbar = 2916 \text{ G}^{-1}\text{s}^{-1}$, μ is the magnetic moment of the neutron, and $\hbar = h/(2\pi)$ is the reduced

Planck constant. The resonance frequency $f_{\text{res}} = 26.2$ MHz found in our experiments corresponds to Larmor precession in a magnetic induction $B = 9.0$ kG. This value coincides with the saturation magnetization $M = 9.0$ kG obtained by VSM and by neutron reflectometry at PRISM under an external magnetic field $H_0 = 4.4$ kOe. The external magnetic field in the neutron spin resonance experiment at N-REX⁺, however, was only $H_0 = 20$ Oe. In this low field the film was partially demagnetized and the average magnetization was $M = 4.0$ kG (as determined from VSM and reflectometry). However, at low external field the magnetic state of the permalloy film is characterized by large (several tens of μm) saturated domains in which the local magnetization is 9.0 kG, but whose orientations are distributed such that the average magnetization is reduced to 4.0 kG and oriented along the constant external field and the neutron beam polarization. This shows that the observed resonance phenomenon is caused by interaction of the neutrons with the local field in individual magnetic domains.

Let us finally discuss the magnitude of the observed beam-splitting. The Zeeman energy in the constant magnetic field H_0 is equal to $\pm\mu H_0$, where the signs ‘+’ and ‘−’ correspond to spin up and spin down neutrons, respectively. For $H_0 = 20$ Oe, the Zeeman energy is thus 0.12 neV which is totally negligible compared to the energy quantum $\hbar\omega_{\text{res}} = hf_{\text{res}} = 108.54$ neV of the oscillating magnetic field at the resonance frequency $f_{\text{res}} = 26.2$ MHz. The initial kinetic energy of the neutrons is $E = \frac{\hbar^2 k_i^2}{2m} = \frac{\hbar^2 k_{i\perp}^2}{2m} + \frac{\hbar^2 k_{i\parallel}^2}{2m}$, where m is the neutron mass, and where $k_{i\perp} = k_i \sin \alpha_i$ and $k_{i\parallel} = k_i \cos \alpha_i$ are the wave vector components perpendicular and parallel to the sample surface. During reflection and refraction from a homogeneous medium, the parallel wave vector component does not change: $k_{i\parallel} = k_{f\parallel}$. From energy conservation it thus follows for spin-flip reflection in the presence of an oscillating magnetic field of frequency f :

$$\frac{\hbar^2 \sin^2 \alpha_f}{2m\lambda^2} = \frac{\hbar^2 \sin^2 \alpha_i}{2m\lambda^2} \pm hf, \quad (1)$$

where the ‘+’ sign corresponds to the spin-flip from the (Zeeman) low-energy ‘−’ state (spin down) to the high-energy ‘+’ state (spin up), and vice versa. Since incidence and exit angle are small ($\sin \alpha \approx \alpha$), we can write:

$$\alpha_f = \left(\alpha_i^2 \pm f \frac{2m\lambda^2}{h} \right)^{\frac{1}{2}} \quad (2)$$

Note that this beam-splitting due to resonance with an oscillating field is opposite to the Zeeman beam-splitting. In the oscillating field we have $\alpha_f^{+-} < \alpha_i$ and $\alpha_f^{++} > \alpha_i$, while in case of the Zeeman splitting one has $\alpha_f^{-+} > \alpha_i$ and $\alpha_f^{--} < \alpha_i$ [6].

Figure 7 shows the beam-splitting as calculated from Eq. (2) for our experimental parameters, i.e. for a neutron wavelength of 4.26 Å and an external field oscillating at the resonance frequency $f_{\text{res}} = 26.2$ MHz. The two experimental data points obtained for $\alpha_i = 0.4^\circ$ agree very well with the calculated exit angles for the spin-flipped neutrons. Note that the strong beam-splitting can clearly be attributed to the Zeeman energy difference $2\mu B$ related to the strong magnetic induction $B = 9.0$ kG in saturated domains, while the interaction energy $2\mu H_0$ with the constant external field would be way too small to explain the observed effect. On the other hand, the observed beam-splitting is of similar magnitude as the one reported in references [7-9] for reflection from a certain domain structure and domain wall morphology. In this case the external magnetic field was also low (20-100 Oe), but the local magnetic induction was reported to be high (20 kG), similar as in our case. Note, however, that this type of beam-splitting due to a certain static configuration of internal magnetic fields is fundamentally different from our observations, because in the study presented here the beam-splitting

is absent outside the resonance condition. This proves that the strong beam splitting we observed is entirely due to local neutron spin resonance in saturated single domains.

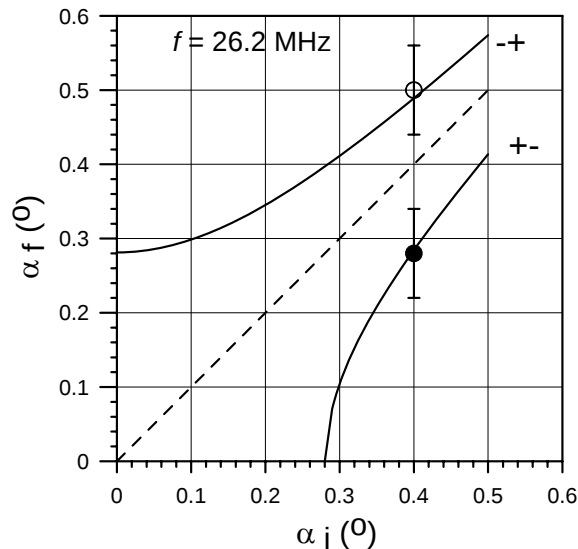


Figure 7. Calculated beam-splitting due to internal spin-flip induced by an external magnetic field oscillating at 26.2 MHz. The involved spin transitions, ‘-+’ and ‘+-’, are indicated. The symbols correspond to our experimental results. The error bars are defined by the spatial resolution of our position-sensitive detector.

5. Conclusions

We have obtained new experimental results from neutron reflection on a thin permalloy film placed in an oscillating magnetic field. The following key conclusions can be drawn.

- Neutron magnetic resonance takes place inside the film.
- The spatial beam-splitting is measurable only when the frequency of the oscillating external magnetic field is close to the Larmor precession frequency corresponding to the local magnetic induction in the sample.
- Non-specular reflected beams occur for both initial spin states of neutrons, but the intensities of the two non-specular beams are not identical.
- The resonant off-specular intensities are proportional to the respective off-resonance specular reflectivities.

The frequency of the oscillating magnetic field as applied in the experiments reported here can be used in the future as a new parameter for investigations of local magnetic properties in layered magnetic structures, and it may also be extended to the study of dynamic magnetic processes in such systems. This method can be called Neutron Resolved Magnetic Resonance.

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