Quantum States of Neutrons in Magnetic Thin Films

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INTRODUCTION

Neutron reactivity goes back to 1946 when it was first used by Fermi and Zinn for the determination of coherent scattering lengths [1]. Subsequently inserting a polarizer and analyzer to produce a polarized neutron beam, Hughes and Burgy started to perform polarized neutron experiments as early as 1951 [2]. It was foreseen by the authors of this work that beam s of completely polarized neutrons will be useful in the study of magnetic and nuclear properties. The next major achievement of Polarized Neutron Reflectometry (PNR) was the prediction of spin-polarized neutron reactivity by Ignatovich [1978] [3] and the pioneering experiments on magnetic surfaces by Felcher [1981] [4]. While specular PNR is widely recognized as a powerful tool for the investigation of magnetic structures in magnetic heterostructures [5], the description of o-scattering from magnetic domains is still under development [6]. In spite of these important developments there is still confusion concerning the quantum states of neutrons in a magnetic sample. Here we show unambiguously that the neutron has to be treated as a spin 1/2 particle [1,2] in each homogenous magnetic layer. This is at variance with the conventional description of neutron reactivity, which often considers the neutron magnetic potential as a classical dot product [3,4,12,13].

Neutrons interact with a magnetic field via the Fermi nuclear potential and via the magnetic induction. Thus, the neutron - interaction hamiltonian includes both contributions: \( H = V_n + V_m = ( -2m) N b \cdot B \), where \( m \) is the neutron mass, \( N \) is the particle density of the material, \( b \) is the coherent scattering length, \( j \) is the magnetic moment of the neutron, and \( B \) is the magnetic induction of the field. Unconventionally, however, neutron reactivity treats the dot product between the magnetic induction and neutron magnetic moment classically: \( V_m = B \cdot j B \cos(\theta) \), where \( \theta \) is the angle between the incoming neutron polarization direction and the direction of the magnetic field inside the film. Writing the magnetic potential as a classical dot product implies that the neutron energies in the magnetic layer have a continuous distribution from \(-j B \) to \(+j B \). This predicts that the critical angle for total reflection depends on the angle between the direction of polarization and the direction of the magnetic field inside the layer:

\[
4 \sin(\frac{\theta}{2}) = \frac{N_n j B \cos(\theta)}{2m}, \quad \text{where} \quad \theta \quad \text{is the glancing angle to the surface,} \quad \text{is the wavelength of the neutrons, and} \quad Q_c \quad \text{is the critical scattering vector.} \]

From the Stern-Gerlach experiment we know that there are only two eigen states for the spin 1/2 particles in a magnetic field. Therefore, the eigen wave number of a neutron in a magnetic thin film has two proper values. After solving the Schrödinger equation one obtains two eigen wave numbers for neutrons in a magnetic field: \( k^2 = \frac{2m}{\hbar^2} N_n j jB \). They correspond to two possible states of spin orientation: one for the case, when the spin is parallel to the magnetic induction, and the other for the antiparallel orientation. It follows that there are only two possible energies and consequently only two values for the index of refraction corresponding to the spin-up and spin-down states of the neutrons. Therefore,
QM predicts that there are only two critical angles for the total reflection: one corresponding to the $R^+$ and one to the $R^-$ reflection

$$4 \sin (\varphi) = Q_c = \frac{r}{2m} (N_n j |j| \langle \psi | \rangle$$

Obviously there is a contradiction between the quantum mechanical prediction (Eq.2) and the prediction based on the classical representation of the magnetic potential (Eq.1). Quantum mechanics predicts that the spin states of the neutrons are determined by the magnetic induction in the sample, whereas classical representation of the magnetic potential, supported by experiments on magnetic multilayers, assert that the spin states of the neutrons is excited by the incident polarization axis.

Here we describe an experiment which provides direct and unambiguous evidence for the spin states of neutrons in magnetic media. The goal is to find a system where the angle between the neutron polarization and direction of the magnetization inside of the film can be varied and controlled. Then we measure the $R^+$ and $R^-$ reflection and determine whether the position of the critical edges changes as a function of the angle, or whether the critical edges stay fixed, and only intensity redistributes between reflections $R^+$ and $R^-$ with change of . The easiest way to control the angle is to rotate the magnetic film and therefore the magnetization direction with respect to the neutron spin polarization, which remains fixed in space outside of the sample. This requires that the film should have a high remnant magnetization. Additionally, the film thickness should exceed the average neutron penetration depth . The last requirement is essential in order to avoid neutron tunnelling effects which will hinder the precise determination of the critical edges.

**Sample Characterization by MOKE and PNR**

To fill the aforementioned requirements, we have chosen a 100 nm thick polycrystalline Fe film deposited by rf-sputtering on a Si substrate. The thickness of the Fe film was about 4 times larger than the average penetration depth $d = 2m V_n = 2$. The Fe film was covered with thin Co and CoO layers, the latter one protecting the Fe film from oxidation. For sample characterization at room temperature we first recorded hysteresis loops with the magneto-optical Kerr effect (MOKE). A series of hysteresis loops were taken with the field parallel to the film plane but with different azimuth angles of the sample. A typical hysteresis loop is shown in Fig.1.

The coercive field is about 20 Oe and the remanence is high. A plot of the ratio between the remnant magnetization and saturation magnetization $M_{\text{rem}} = M_{\text{sat}}$ versus the rotation angle about the sample normal is shown in

FIG.1: Bottom: hysteresis loop of the polycrystalline Fe/Si sample measured by MOKE. Top: the behavior of the remnant magnetization as a function of the rotation angle extracted from hysteresis loops.

FIG.2: (Color online). Top: Polarized neutron reflectivity curves $R^+$ (solid black circles) and $R^-$ (open black circles) of the Fe/Si sample. The black line is the simulated $R^+$ reflectivity and the red (light gray) line is the simulated $R^-$ reflectivity. The applied magnetic field was 2000 Oe. In the inset, the magnetic profile obtained from fitting the data is shown. Bottom: Experimental (open black symbols) and simulated (black line) spin asymmetry $(R^+ - R^-)/(R^+ + R^-)$ are plotted for the same sample in saturation. All lines in the top and bottom panels are fits to the data points using the $G M M$ (for more details see text). The abscissa is the wave vector transfer: $Q = 4 \sin (\varphi)$.
rectivity curves were measured for four different angles between the neutron polarization along \( B_0 \) and the direction of the magnetic induction (\( B \)) which lies in the sample plane. The guiding \( \angle \) is \( B_0 \parallel 1000 \text{ Oe} \). The blue (thick dark gray) lines are the simulated \( R^+ \) rectivities and the red (thin light gray) lines are the simulated \( R^- \) rectivities. In the right side the experimental geometry is shown. The figure shows that the critical edges \( Q^+ \) and \( Q^- \) are not sensitive to the angle.

**TABLE I**: Parameters of the Fe/Si sample obtained by fitting to the \( R^- \) and \( R^+ \) data shown in Fig.2. \( d \) is the layer thickness, \( \sigma \) is the rms roughness, SLD is the scattering length density, and \( B \) is the magnetic induction in the \( \mathrm{Fe}\) magnetic \( \mathrm{m} \) s.

| Layer     | \( d \) [Å] | \( R \) | SLD | \( B \) [Oe] |
|-----------|-------------|--------|-----|-------------|
| CoO \( \gamma \) | 50          | 11.5   | 4.99e-6 | 0           |
| CoFe \( x \) | 39          | 3      | 5.0518e-6 | 15563.4     |
| Fe        | 987         | 5      | 8.024e-06 | 21600       |
| substrate | non         | 6      | 2.073e-06 | 0           |

From Table I and the magnetic characterization (Fig.1) we conclude that the sample fulfills the requirements as concerns thickness, anisotropy, and remanence as required for our experiment.

**RO TATION EXPERIMENT**

The rotation experiment was performed as follows: the Fe layer was magnetized parallel to the neutron polarization direction and then the magnet was remagnetized. A small guiding \( \angle \) (\( \gamma_c \)) is still present at the sample position in order to maintain the neutron polarization. Subsequently a series of \( R^+ \) and \( R^- \) rectivities, shown in Fig.2, were measured for several in-plane rotation angles of the sample. We observe two characteristics of the rectivities: (1) the critical edges are gapped and independent of the in-plane rotation angle (see Eq.2), and (2) the \( R^- \) intensity continuously increases at the expense of the \( R^+ \) intensity as a function of the \( \angle \). Using the parameters obtained from the \( d \) to the saturation data (see Table I and using the rotation angle \( \angle \) as set during the experiment, we have simulated \( 14 \) the \( R^- \) and \( R^+ \) rectivities using the GM Method approach \( 7 \) which transparently predicts the behavior of the critical edges described by Eq.2. The simulated curves are plotted together with the
experim ental data in Fig.3. These are no free parameters for these simulations, providing an excellent description of the experim ental results. The xed critical edges $Q^+$ and $Q^-$ can easily be interpreted in the context of the neutron spin states in homogenous magnetic media as discussed in the introduction.

**EXPERIMENTAL DETERMINATION OF MAGNETIZATION ORIENTATION**

The sensitivity to the in-plane rotation angle of the magnetization is seen very clearly seen in the reflected intensities $R^+$ and $R^-$ plotted in Fig.3. It has been shown theoretically [14] that, for a single magnetic layer, the normal alized spin asymmetry $nSA(\theta)$ is directly related to the angle through the following expression:

$$nSA(\theta) = \frac{SA(\theta)}{SA(0)} = \cos(\theta) \quad (3)$$

Now, we use our experim ental data shown in Fig.3 to confirm the validity of this equation. In Fig.4 is shown the experim ental normal alized spin asymmetry and the cosine of the experim ental edges. The agreement between the experim ental normal alized spin asymmetry (sym bol s) and the cosine of the angles (lines) set during the experim ent is excellent over the whole wave vector transfer range. It follows that the magnetization orientation of a single magnetic layer with respect to the neutron polarization outside the layer can be easily extracted experim entally using Eq. (3). For more complicated systems a numerical tit is still necessary. The $nSA$ is an import ant measure of hysteresis loops. It was shown in Ref. [14] that $nSA$ can be written, generally, as:

$$nSA = M_{sat}^{r} / M_{sat}$$

where $M_{sat}$ is the remnant magnetization. This implies that $nSA$ reproduces the hysteresis loops as measured by SQUID or MOKE. Here we can con firm the validity of the $nSA$ for determining the magnetization reversal via coherent rotation and via domain wall motion. This is important for samples which contain a single magnetic layer. Comparing MOKE or SQUID hysteresis loops with $nSA$ is a very useful tool for the evaluation of magnetic domain state and/or a reduced magnetization within the layer.

**MULTILAYERS**

Our next topic is to investigate the neutron spin states in multilayers with noncollinear magnetization of adjacent layers. We simulated the reactivity profile of a Fe(60Å)/Cr(8Å))$_n$/Si superlattice, with thicknesses of the Fe and Cr layers which are typical for many real superlattices [13]). For the simulation we used the freeware code PolSim [14] based on GMM for the calculation of the reaction and transmission coefficients together with a full quantum mechanical description of the spin states [7]. In the simulation the choice of a Si substrate has the advantage that it does not obscure the critical edge of the neutron state. In the top panel of Fig.5, we show simulations of $R^+$ and $R^-$ reactivities for three angles between the magnetization vectors of adjacent Fe layers: $\theta = 0$ (or FeIron magnetic alignment); $\theta = 100$; and $\theta = 170$ (close to antiferromagnetic alignment). Our focus is on the behavior of the critical scattering vector for total reflection. We observe that for $\theta = 0$ the (+) and (-) critical scattering vectors are well separated and that they contain information about the saturation magnetization. When the value increases, the critical edges approach each other. For an angle of $\theta = 180$ (not shown here) there is no difference between the $R^+$ and $R^-$ reactivities. The main result from this simulation is the observation that the separation of the critical edges is a continuous function of the angle between the in-plane adjacent magnetization vectors. The critical edge positions satisfy the following relation:

$$4 \sin^2 (\theta_c) = 2 \sum \left( \frac{V_{n,eff}}{m} \right) J_B \cos (\theta_c)$$

where $V_{n,eff}$ is the effective nuclear potential. Clearly, for this geometry the angles and $\theta_c$ coincide if the neutron...
polarization is parallel to the average field \( \langle B_1 + B_2 \rangle = 2 \), where \( B_1 \) and \( B_2 \) are the magnetic field inductions of adjacent layers. Therefore, numerically, the eqs. 1 and 3 are almost identical. However, there is a fundamental difference: similarly to the single layer, the angle does not in essence the position of critical edges, whereas the angle is only responsible for the continuous shift.

To shed more light on how the and angles affect the critical edges for polarized neutron reactivity at the multilayers we simulated numerically the rotation experiment performed on the single layer. For a fixed coupling angle of \( \phi = 90^\circ \), as it can be achieved also experimentally via biquadratic exchange coupling, the reactivities \( R' \) and \( R \) are plotted as a function of the angle. Here the angle is the angle between the incoming neutron polarization and the direction of the average magnetization vector of two adjacent ferromagnetic layers. The results are shown in the bottom panel of Fig. 5. We observe a similar behavior of the critical edges and intensities as for the single layer. While the positions of the critical scattering vectors \( Q^r \) and \( Q^c \) remain fixed for a constant coupling angle \( \phi \), the \( R' \) intensity increases on the expense of the \( R \) intensity with increasing angle. With this simulations we lift the contradiction stated in the introduction by showing that Eq. 4 is a particular case of Eq. 3 which, in turn, is in agreement with the description of the neutron spin states in magnetic media Eq. 2. The different behaviors of the critical edges for the case of a single homogeneous ferromagnetic layer and for a multilayer with alternating directions of the layer magnetization vectors now become obvious: in the multilayer the neutrons are acted on by an average magnetic potential which depends on the relative orientation of the magnetic induction in the individual layers. However, in both cases, single as well as multilayer, the magnetic potential of the individual layers \( V_m = \langle B \rangle j \) enters the algorithm for calculating the reactivities.

It should be noted that the dependence of \( Q^r \) and \( Q^c \) on the angle \( \theta \) in a multilayer is a general property of the periodic potential with different direction and magnitude. It is natural to expect that such a sample is a noncollinear ferromagnetic with ferromagnetic eddy \( B_\parallel = \langle B_1 + B_2 \rangle = 2 \), where

\[
B_\parallel = B_1 + B_2 = B \cos(\theta) = 2, \tag{5}
\]

and with antiferromagnetic eddy \( B_{af} = B_1 - B_2 = 2 \),

\[
B_{af} = B_1 - B_2 = B \sin(\theta) = 2 \tag{6}
\]

Then, the critical edges can be expected to be given by Eq. 4.

To further stress the origin of the effective nuclear potential \( V_{n,eff} \) term in Eq. 4, let us consider the critical edge for non-polarized neutrons when scattered at a \( Fe(Ax)(Cr(yA)) \) multilayer. Naively, we may expect that the critical edge to be given by the Fermi interaction potential of Fe as it is higher than the potential of Cr. This is, however, not the case. For a finite thickness \( x \) of the Fe layer and zero thick Cr layer, indeed the critical edge is equal to the critical edge of a single thick Fe layer. Vice versa, for zero thickness of Fe layer and finite thickness \( y \) for the Cr layer the critical edge is given by the Fermi potential of Cr. However, when both layers have finite thicknesses the critical edge of the multilayer will vary from the value for pure Fe to the value for pure Cr. Therefore, the critical edge for non-polarized neutrons reflected from a multilayer not only depends on the Fermi potential of the two separate layers, but also on their individual thicknesses.
CONCLUSIONS

In summary, we have analyzed the behavior of the critical scattering vectors $Q_c^+$ and $Q_c^-$ for total external reflection of a polarized neutron beam for the case of homogeneous ferromagnetic films and for antiferromagnetically coupled multilayers. For a single film we have observed experimentally and shown theoretically that the critical edges do not change as a function of the angle between the neutron polarization and the direction of the magnetic spins inside the film. They fulfill the relation $Q_c = \frac{2\pi}{V_n} j \beta_s j$, which directly reflects the spin states of the neutron beam in magnetic thin films. For multilayers we found that the critical edges for total external reflection move towards each other as a function of the coupling angle. Their position is well reproduced by the Eq. $Q_c = \frac{2\pi}{V_{eff}} j \beta_s j \cos(-2\theta)$. The $\cos(-2\theta)$ dependence is not related to the neutron spin states in the magnetic media, but it is the result of the presence of a ferromagnetic field direction along the average field in the noncollinear ferromagnet. By choosing a fixed coupling angle between the magneticization vectors of adjacent layers and rotating the sample, the critical edges behave again in accordance with the neutron spin states in homogeneous magnetic media. Practically, the coupling angle in non-collinear superlattices can be inferred directly from the experimental data through the separation of the critical edges. For a single layer the orientation of the magneticization can be extracted experimentally from the spin asymmetry.

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