Quantum State of Neutrons in Magnetic Thin Films and Superlattices

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An experiment which describes the quantum states of neutrons in magnetic thin films and superlattices is reviewed.

While specular polarized neutron reflectivity (PNR) is widely recognized as a powerful tool for the investigation of magnetization profiles in magnetic nanostructures, there is still a confusion concerning the quantum state of neutrons in a magnetic sample. We have now shown unambiguously that the neutron has to be treated as a spin 1/2 particle in each homogeneous magnetic layer. This is at variance with the conventional description of neutron reflectivity, which often considers the neutron magnetic moment as a classical dot product.

Neutrons interact with a magnetic thin film via the Fermi nuclear potential and via the magnetic induction. Thus, the neutron-film interaction hamiltonian includes both contributions: $V = V_n + V_m = (\hbar^2/2m)4\pi Nb - \mu B$, where $m$ is the neutron mass, $N$ is the particle density of the material, $b$ is the coherent scattering length, $|\mu|$ is the magnetic moment of the neutron, and $|B|$ is the magnetic induction of the film. Unconventionally, however, neutron reflectivity treats the dot product between the magnetic induction and neutron magnetic moment classically: $V_m = -\mu \cdot B = \pm |\mu||B|\cos(\theta)$, where $\theta$ is the angle between the incoming neutron polarization direction and the direction of the magnetization 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 $-|\mu||B|$ to $+|\mu||B|$. 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.

On the other hand we know that there are only two eigenstates 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. 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, quantum mechanics predicts that there are only two critical angles for the total reflection: one corresponding to the $R^+$ and one to the $R^-$ reflectivity.

 Obviously there is a contradiction between the quantum mechanical prediction and the prediction based on the classical representation of the magnetic potential: quantum mechanics predicts that the spin state of the neutron is 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 state of the neutrons is fixed by the incident polarization axis (see Fig. 1).

To resolve this contradiction we have carried out an experiment which provides direct and unambiguous evidence for the spin state of neutrons in magnetic media. The goal was to find a system where the angle between the neutron polarization and direction of the magnetization inside of the film can be fixed and controlled. Then we measure the $R^+$ and $R^-$ reflectivities and determine whether the position of the critical edges changes as a function of the angle $\theta$, or whether the critical edges stay fixed, and only intensity redistributes between reflections $R^+$ and $R^-$ with change of $\theta$. The easiest way to control the angle $\theta$ is to rotate the magnetic film and therefore the magnetization direction with respect to the neutron spin polarization, which remains fixed in space outside

![FIG. 1: Two possible orientations of the neutron spin in a film with a homogeneous magnetization $M$, which makes an angle $\theta$ against the polarization axis: either the spin orientation is maintained parallel to the polarization axis, or the spin is reoriented according to the new quantization axis in the film. Maintaining the spin orientation corresponds to a classical description of the interaction of neutrons with a magnetic potential, reorientation is required by quantum mechanics for a particle with spin 1/2.](image)
of the sample. This requires that the film should have a high remanent 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.

The experiment was performed at the ADAM reflectometer (ILL). As magnetic film we have chosen a 100 nm thick Fe layer deposited by rf-sputtering on a Si substrate. The remanence obtained by measuring the hysteresis curve is about 97.5%. In the rotation experiment the Fe layer was first magnetized parallel to the neutron polarization direction and then the magnet was removed. A small guiding field was still present at the sample position in order to maintain the neutron polarization. Subsequently a series of $R^+$ and $R^-$ reflectivities were measured for several in-plane rotation angles of the sample. The results are shown in Fig. 2. We observe two characteristics of the reflectivities: first, the critical edges are fixed and independent of the in-plane rotation angle $\theta$ of the magnetization vector, and second the $R^-$ intensity continuously increases at the expense of the $R^+$ intensity as a function of the $\theta$ angle. The plain experimental results as well as a detailed fit unambiguously show that the critical edges $Q^{c+}$ and $Q^{c-}$ for total reflection of the two spin states are fixed independent of the orientation of the magnetization vector in the film. This confirms that the neutron spin inside of the sample is oriented according to the quantization axis parallel or antiparallel to the magnetization vector.

So far so good for magnetic thin films. But what is the orientation of polarized neutrons in magnetic multilayers with periodically varying magnetization direction? Does the spin orientation of the neutron follow the rapid change from layer to layer, or does the neutron interact with an average potential? To answer this question we have simulated the reflectivity profile of a Fe(6 nm)/Cr(0.8 nm) superlattice containing 40 repeats. The thicknesses of the Fe and Cr layers chosen are typical for this class of materials. For the simulation we used the freeware code PolarSim, which is based on the generalized matrix method. In Fig. 3 simulations are shown for $R^+$ and $R^-$ reflectivities and for three angles $\gamma$ between the magnetization vectors of adjacent Fe films: 0, 100, and 170°, the last one being close to an antiferromagnetic orientation of the Fe layer. Our focus is on the behaviour of the critical scattering vector for total reflection. We observe that for $\gamma = 180°$ (not shown here) there is no difference between the $R^+$ and $R^-$ reflectivities in the vicinity of the critical scattering vector. 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. Thus, unlike the single magnetic film, in case of magnetic multilayers the critical edges move. However, they do not shift according to the angle between the average magnetization vector and the polarization axis (as expected in the classical case), but according to the angle between the magnetization vectors between adjacent magnetic layers. Thus, from a practical point of view it is sufficient to determine the difference between the critical edges $Q^2 = (Q^{c+})^2 - (Q^{c-})^2$ to measure the angle between the magnetization vectors in magnetic superlattices, which then follows from: 

$$\cos(\gamma/2) = \Delta Q^2 / (2\mu_0|B|).$$

If we now choose a fixed coupling angle $\gamma$ between the magnetization vectors of adjacent layers and rotate the sample as in the previous case of a single magnetic layer, then the critical edges behave again in accordance with the neutron spin states in homogeneous magnetic media, i.e. $Q^{c+}$ and $Q^{c-}$ remain constant, but the intensity varies with the rotation angle between the average magnetization vector and the polarization direction.

This finally solves a long lasting controversy about the dependence of the critical edges for total reflection of polarized neutrons. The apparent shift of the critical edges is solely due to the opening angle between the magnetization vectors in successive layers of a magnetic multilayer. As soon as this angle is fixed anywhere between a ferro- to an antiferromagnetic state, the critical angles are fixed independent of the orientation of the superlattice with respect to the polarization direction of the neutrons. In the single homogeneous magnetic layer as well as in the superlattice, the polarized neutron within the sample assumes new eigenstates according with the
FIG. 3: Simulation of polarized neutron reflectivities ($R_{\uparrow}$ and $R_{\downarrow}$) for a Fe/Cr multilayer as a function of coupling angle ($\gamma$) between the magnetization vectors of adjacent Fe layers. In the inset the superlattice is shown with a cut-away view of the top magnetic layer. The critical angle for total reflection is sensitive to the coupling angle ($\gamma$) but not to the orientation of the average magnetization, with respect to the polarization axis of the neutrons. Also shown are the halforder peak from the antiferromagnetic component of the superlattice at $Q \approx 0.05\,\text{Å}^{-1}$ and the first order ferromagnetic peak at $Q \approx 0.095\,\text{Å}^{-1}$.

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