

# Neutron Scattering for Condensed Matter Studies

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Week 9: Lecture 24C

Keywords: Polarization analysis, Spin flip, Cobalt-gadolinium multilayer

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Without polarization analysis, one measures  $R^+$  and  $R^-$

Polarization of the neutron beam parallel ( $R^+$ ) and antiparallel ( $R^-$ ) to the sample magnetization. The up (+) and down (-) beams see different magnetic potentials.

$$V = \begin{bmatrix} V_+ \\ V_- \end{bmatrix} = \frac{2\pi\hbar^2}{m} \rho \begin{bmatrix} b_{coh} + b_M \\ b_{coh} - b_M \end{bmatrix} \quad \theta_c^\pm = \sqrt{\frac{\rho(b_{coh} \pm b_M)}{\pi}} \lambda$$

$$\rightarrow \frac{d^2\psi_+(Z)}{dZ^2} + \frac{2m}{\hbar^2} [E - V_+(Z)]\psi_+(Z) = 0$$

$$\rightarrow \frac{d^2\psi_-(Z)}{dZ^2} + \frac{2m}{\hbar^2} [E - V_-(Z)]\psi_-(Z) = 0$$

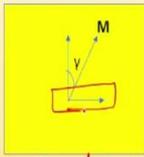
Schrodinger eqn.s for neutron propagation with spin parallel (+) or anti-parallel (-) to the film magnetization



we consider a potential with 4 components in the matrix form

$$\begin{bmatrix} V_{++} & V_{+-} \\ V_{-+} & V_{--} \end{bmatrix} \quad \text{Compare with} \rightarrow \quad V = \begin{bmatrix} V_+ \\ V_- \end{bmatrix} = \frac{2\pi\hbar^2}{m} \rho \begin{bmatrix} b_{coh} + b_M \\ b_{coh} - b_M \end{bmatrix}$$

The expression for the four components of the potential can be written in terms of the coherent nuclear scattering length density, ' $\rho b_{coh}$ ' and components of the magnetic moment along the applied field and normal to it



The magnetism 'M' comes from a magnetic scattering length ' $b_m$ ' and its density

*SF is purely magnetic*

In terms of magnetic scattering length density components:

$$V_{++} = \frac{2\pi\hbar^2}{m} \rho [b_{coh} + b_y] = \frac{2\pi\hbar^2}{m} \rho [b_{coh} + b_m \cos \gamma]$$

$$V_{--} = \frac{2\pi\hbar^2}{m} \rho [b_{coh} - b_m \cos \gamma], V_{\pm\mp} = \frac{2\pi\hbar^2}{m} \rho b_M \sin \gamma$$


Let me remind you, when we discussed about experiments without polarization analysis, we had two component potential,  $V_+$  and  $V_-$ , and I also showed you two Schrodinger wave equations for the two polarizations. Now, with one step ahead, I have got a four-component potential. That means there is a potential for non-spin flip reflection, given by  $V_{++}$ ,  $V_{--}$ , and two potentials for spin flip,  $V_{+-}$ , and  $V_{-+}$ . Compare this with the two-component potential (earlier). I have got four components now. Why four components?

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$$V_M \sim \frac{2\pi\hbar^2}{m} \rho b_m$$

$$M \cos \gamma \rightarrow \underline{\underline{NSF}}$$

$$\underline{\underline{M \sin \gamma \rightarrow SF}}$$

$$b_m$$

we consider a potential with 4 components in the matrix form

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*SF is purely magnetic*

Let me quickly justify to you the physics (behind these potentials). You imagine, there is a sample with an in-plane magnetization direction. Now suppose the in-plane magnetization is not parallel with the magnetic field but at an angle, say at an angle  $\gamma$ . Let us consider this is the neutrons spin, this is the angle  $\gamma$  which the in-plane magnetization  $M$  makes with the applied field. I can resolve  $M$  in 2-components,  $M \cos \gamma, M \sin \gamma$ . This  $M$  comes from magnetic scattering length  $b_M$ . We know  $V_M$  can be written as,  $V_M \sim \frac{2\pi\hbar^2}{m} \rho b_M$ , but now it has 2-components. Component  $M \cos \gamma$  has magnetic origin and it does get added (subtracted) from the nuclear potential. Now spin of neutrons can be either up or down (with respect to applied field) and hence can be either parallel or antiparallel to this component of magnetic moment. And because they are parallel, this magnetic field will not cause any spin flip for the neutron. So, that means  $M \cos \gamma$  will not give any spin flip component. However,  $M \sin \gamma$  allows precession of the magnetic moment around  $H + M \cos \gamma$ , and this causes spin flip for the reflectivity.

In this four-component potential, we have

$$V_{++} = \frac{2\pi\hbar^2}{m} \rho [b_{coh} + b_y] = \frac{2\pi\hbar^2}{m} \rho [b_{coh} + b_m \cos \gamma]$$

$$V_{--} = \frac{2\pi\hbar^2}{m} \rho [b_{coh} - b_m \cos \gamma]$$

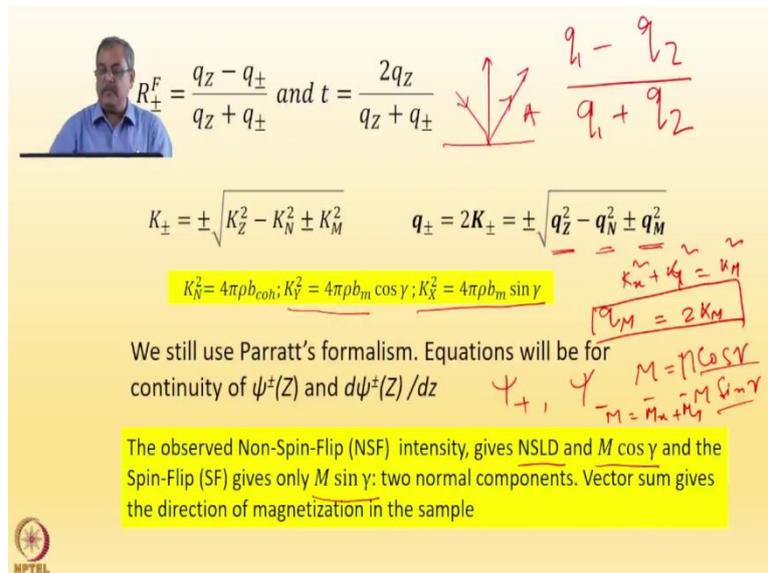
and we also have two spin flip components,

$$V_{\pm\mp} = \frac{2\pi\hbar^2}{m} \rho b_m \sin \gamma$$

This spin flip component is of purely magnetic origin.

Why I mention this? Because you can see the non-spin flip part has nuclear as well as magnetic parts, either plus or minus. This is similar to what I showed you for  $V_+$  and  $V_-$ . But here, the spin flip potential has its origin only in the normal component of the magnetic moment and it is purely magnetic in origin.

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$R_{\pm}^F = \frac{q_z - q_{\pm}}{q_z + q_{\pm}}$  and  $t = \frac{2q_z}{q_z + q_{\pm}}$

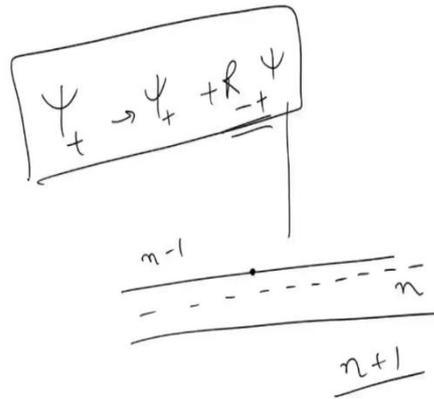
$K_{\pm} = \pm \sqrt{K_z^2 - K_N^2 \pm K_M^2}$        $q_{\pm} = 2K_{\pm} = \pm \sqrt{q_z^2 - q_N^2 \pm q_M^2}$

$K_N^2 = 4\pi\rho b_{coh}$ ;  $K_Y^2 = 4\pi\rho b_m \cos \gamma$ ;  $K_X^2 = 4\pi\rho b_m \sin \gamma$

We still use Parratt's formalism. Equations will be for continuity of  $\psi^{\pm}(Z)$  and  $d\psi^{\pm}(Z)/dz$

The observed Non-Spin-Flip (NSF) intensity, gives NSLD and  $M \cos \gamma$  and the Spin-Flip (SF) gives only  $M \sin \gamma$ : two normal components. Vector sum gives the direction of magnetization in the sample

Handwritten notes:  $q_1 - q_2$  over  $q_1 + q_2$ ;  $K_N + K_Y = K_M$ ;  $M = M \cos \gamma$ ;  $M = M_x + M_y$



Now, I can write down the Fresnel reflectivity as

$$R_{\pm}^F = \frac{q_Z - q_{\pm}}{q_Z + q_{\pm}}$$

where  $q_{\pm} = \pm \sqrt{q_Z^2 - q_N^2 \pm q_M^2}$ . Here,  $q_Z$  is propagation vector in air and  $q_{\pm}$  are the propagation vectors in the medium.  $q_M$  has got two parts, nuclear as well as magnetic. We know  $q_M = 2K_M$  where  $K_M^2 = K_x^2 + K_y^2 = 4\pi\rho b_m \cos \gamma + 4\pi\rho b_m \sin \gamma$ .

We will still use the Parratt's formalism and, as I told you, there will be equations for continuity of  $\psi^{\pm}(z)$  and their differentials (with respect to 'Z'). Now in the continuity of  $\psi^+$  where  $\psi^+ \rightarrow \psi^+ + R_{-+}^{\psi}$  in the medium. If you remember, in a Parratt's formalism, I considered continuity at  $n-1$  and  $n^{\text{th}}$  layer and  $n+1^{\text{th}}$  layer came in the recursion relation. But here, when I talk about  $\psi^+$  here, I also have a refracted component of  $\psi$  in the medium. So now in Parratt's formalism, the continuity equation will have one extra term, which I am not writing over here, but that is straightforward.

Again, let me highlight that the observed non-spin flip intensity gives nuclear scattering length density and  $M \cos \gamma$ , and the spin flip component gives only  $M \sin \gamma$ . And from these two components, we can rebuild the magnetic moment (magnitude and direction) as  $M$  equal to vector sum of  $M \cos \gamma$  and  $M \sin \gamma$ . I can say  $\bar{M} = \bar{M}_x + \bar{M}_y$ , which is basically  $M^2 = M^2 \cos^2 \gamma + M^2 \sin^2 \gamma$ , so we can get the value of  $M$ , if I can determine  $M \cos \gamma$  and  $M \sin \gamma$ . Basically, we have to determine the angle at which the magnetic moment is oriented, from the spin flip intensity.

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Helical magnetic ordering in Co-Gd multilayers

Si/[Gd(140 Å/Co(80 Å)]x 8, eight bilayers ✓

Co and Gd both ferromagnets. Co is magnetic at room temperature ( $T_c \sim 1400$  K), Gd is ferromagnetic just below room temperature ( $T_c \sim 292$  K)

Co and Gd are antiferromagnetically → How? Magnetic hysteresis loop (200 K) in field-cooled sample

MPTEL

Surendra Singh et. al. Phys. Rev. (Rap. Comm.) B 100, 140405 (R) (2019)

*XRD*

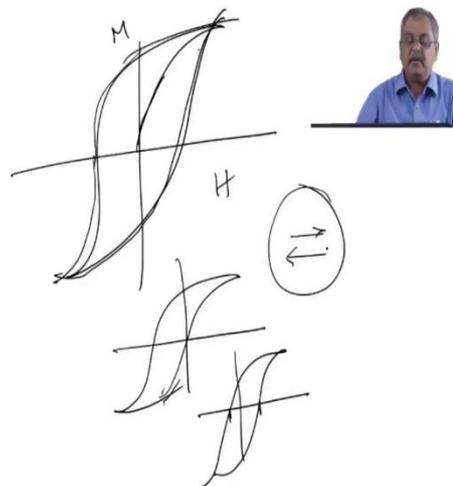
*XRR*

*M-H*

I will use only one example over here (for magnetic structure in) cobalt-gadolinium multilayer. This is the model structure for this (symbolically). We have gadolinium and cobalt bilayers of 140 Å and 80 Å thickness (respectively) and there are 8 (such) bilayers on silicon. Both of them are ferromagnetic. Co is magnetic at room temperature and has got a Curie temperature of around 1400 K. Gadolinium is ferromagnetic just below room temperature at around 290 K.

Here, in this example that I have chosen, what we observed actually is a very interesting (magnetic) structure in cobalt-gadolinium multilayer. Here, again we obtained additional information from other measurements. One is XRD which says that this is a polycrystalline sample. XRR gives me the scattering length density or the physical density as a function of depth in the sample and magnetic hysteresis loop gives bulk magnetization. I just show you the hysteresis loop at 5 K, under a field cooling mode where fields of +500 Oe and -500 Oe were used.

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You can see the shift in the hysteresis loop.

We know that the hysteresis loop should look like this. Initially when you start applying the field, the sample gets magnetized and reaches saturation magnetization. When we reduce it (applied field), magnetization goes to remnant magnetic moment, then to coercive field and then the loops go from the positive to negative applied field. It (the loop in the negative applied field) goes to the same structure.

But if there is an anti-ferromagnetic coupling at the interface, then the coupling needs to be broken in the hysteresis loop. This anti-ferromagnetic coupling indicates a shift in the hysteresis loop in either direction along the field axis and the coercive field in the positive and negative directions are not same and that is what is shown here. That means there is an antiferromagnetic coupling at the interfaces that we can try to estimate from the magnetic hysteresis loop in our Co-Gd sample. Another interesting fact is that, gadolinium and cobalt both of them are strong neutron absorbers, so this experiment was really difficult (regarding intensity of reflected neutrons).



This experiment was done at OFFESPEC spectrometer at ISIS. I will show you the results now. At 300 K, the sample had ferromagnetic cobalt while gadolinium was non-magnetic. So, at room temperature, cobalt should show a ferromagnetic nature while gadolinium should not. When we plot the room temperature magnetic scattering length data along with the nucleus scattering data it looks like this: so NSLD is the left side (Y-axis), and magnetic scattering length density (MSLD) has been plotted (right side Y-axis). There is no (spin) analysis of the reflected beam, I will find out just the magnetic moment density profile.

At 300 K plot (red curve) of MSLD, you can see that wherever it comes to gadolinium layer, the magnetic moment density is 0, and whenever we go to cobalt layer, I can see a positive magnetic moment scattering length density.

When we go to 200 K, there is a shift in the hysteresis loop. And from the MSLD profile, we found that Co and Gd are antiferromagnetically coupled at the interface. In the PNR plot, I have shown non-spin flip components  $R^{++}$ ,  $R^{--}$  and non-spin flip components plotted for up and down neutrons. This is how they looked at 300 K and this is how they looked at 200 K. But please pay attention to the spin flip plot which is plotted in (combined) form of  $(R^{+-} + R^{-+})/2$ , average of two spin flip components. Here the spin flip intensity, with the given model of gadolinium and cobalt where one is antiferromagnetically coupled to the other, is very small. It means that there is not much of spin flip. That indicates, if this is the magnetic field direction the cobalt is aligned almost along it and the gadolinium is aligned opposite to it. So, cobalt layer and the gadolinium layer are antiferromagnetically coupled at the interface which makes the hysteresis loop shift. This 200 K structure is between two ferromagnets, which are antiferromagnetically coupled. And the spin flip intensity at 200 K is almost 0, that means there is no normal component (perpendicular to the field).

Now, let us go down further to 125 K. Here, you can see that spin-flip intensity has increased as compared to 200 K. To understand this, know that the magnetic moment is having an angle with respect to the applied field which can be broken up into two components, one along the field which is the cause for this non-spin flip reflectivity, and it has a normal component which causes the spin flip (reflection) and we can fit these two components. Now, please look at this structure which you obtain at 125 K, we get something like a two-dimensional domain wall structure.

That means, if I consider this is a gadolinium layer, and this is a cobalt layer, one (phenomena) is that this Gd (magnetic moment) is rotating (in the layer) and does a  $2\pi$  rotation inside the

layer, that is why you call that this layer behaves like a 2D domain wall. And, in case of cobalt it is again antiferromagnetic coupled (to Gd). Then it rotates around the field and finally, it comes back to the same orientation here. So, these are  $2\pi$  domain wall (like) structure, and this is what we show here. Actually, you can see in this plot where the (rotating) magnetic moment direction has been plotted as a function of the thickness as you go from the cobalt to gadolinium layer.

This is the in-plane direction of the applied magnetic field, which is 500 Oe. And you can see the rotation of the (atomic spin). The helix getting created in this cobalt gadolinium structure. This is a very interesting result in which by polarization analysis at 125 K, you have got these components (parallel and normal to applied field). I fitted this data, and after fitting the spin flip and the non-spin flip components we deciphered this structure. A helix which is forming in this cobalt-gadolinium multilayer. It is a very interesting structure and possibly this (neutron reflectometry) is the only technique which can determine this structure without destroying the sample, experimentally.

So, this is a two-dimensional domain wall, and in case of 5 K data, we see the (helical) structure only in the gadolinium, and the entire cobalt layer remains antiferromagnetically coupled to the gadolinium layer (with parallel spins). At 5 K, this helix we see only in the gadolinium layer, and we do not see the helix in the cobalt layer.

That means (in summary) at 300 K cobalt is ferromagnetic, and at 200 K both of them (Co and Gd) are ferromagnetic, but coupled anti ferromagnetically at the interface, that is why in both these cases we have only non-spin flip data. Then at 125 K, we have got the helix in the cobalt layer as well as in the gadolinium layer. At 5 K, we have the helix in the gadolinium layer, and not in the cobalt layer. This result shows the strength of polarized neutron reflectometry with polarization analysis. And with this, I will stop here.