Development of polarized neutron reflectometry

Introduction:

Polarized-neutron specular reflectometry (PNR) was developed in the 1980s as a means of measuring depth-resolved magnetization in flat films with characteristic thicknesses from 2 to 5000 Å. PNR has been widely used to study homogeneous and heterogeneous magnetic films, as well as superconductors. Neutron reflectivity is a method for measuring the special profile of the neutron interaction potential with the medium. The interaction potential is the sum of neutron interaction potentials with separate isotopes of the medium. The determined neutron interaction potential with the structure unit, secondary radiation is simultaneously registered with a neutron. Channels for registering secondary radiation of charged particles gamma rays and neutrons have experienced spin–flip.

Due to its relevance to studying the phenomena of proximity occurring at the interface between media, superconductor, and ferromagnetic. Due to the mutual impact of ferromagnetic and superconductors, caused by the finite values of the coherent wavelengths of the phenomena, there is a significant modification of magnetic phenomena and superconducting properties. This becomes obvious, in a change of the spatial distribution of magnetization. In this case, it is important to establish the correspondence of magnetic spatial profile with isotopes of the contacting layers. For determining the spatial magnetic profile, standard reflectometry of polarized neutrons is used. In this regard, standard neutron reflectometry did not allow one to identify to what kind of elements the changes of the interaction potential and in particular, of the magnetic profile, are related.
The following figure show the sketch of the superconductor/ferromagnetic bilayer.
(a) When the layers are separated from each other, the magnetic field exists only inside the F layer.
(b) In contrast, when the layers are put in contact, the magnetization inside the ferromagnetic becomes the source of the long-range magnetic field in the superconductor. In both panels, the orange solid (blue dashed) curves schematically represent the profile of the magnetic field when the total current inside the F-layer is diamagnetic.

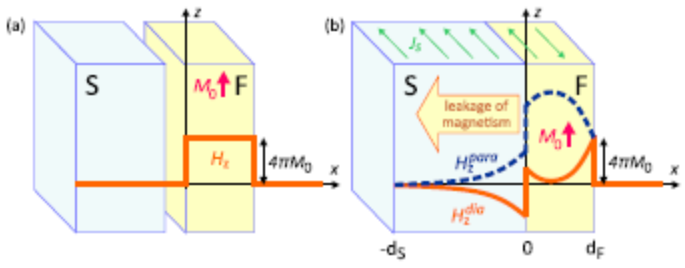


Figure No. 1 – the sketch of the superconductor/ferromagnetic bilayer

Starting from simple profiles, and gradually solving structures of greater complexity, PNR has been used to observe or clarify phenomena as diverse as the magnetism of very thin films, the

penetration of fluxoids in superconductors, and the magnetic coupling across non-magnetic spacers. Although PNR is considered to be a probe of depth-dependent magnetic structure, laterally averaged in the plane of the film, the development of new scattering techniques promises to enable the characterization of lateral magnetic structures. Retaining the depth-sensitivity of specular reflectivity, off-specular reflectivity can resolve in-plane structures over nanometer to micron length scales. Presently limited by the neutron fluxes available, neutron reflectivity is expected to blossom in the next century, thanks to the increased brightness of the neutron beams, due not only to continuing improvements in neutron optics, but especially to the advent of second-generation spallation neutron sources.

Polarized neutron reflectometry (pnr) is one such probe that is particularly well suited for determining the nanostructures of magnetic thin films and multilayers. Together with magnetic x-ray scattering, pnr provides a unique means of "seeing" the vector magnetization with extraordinary spatial detail well beneath the surface. For neutrons this sensitivity to atomic magnetic moments comes about because the neutron itself possesses a magnetic moment and neutrons can be obtained with a wavelength comparable to interatomic distances. More specifically, the specular reflection of polarized neutrons, namely, coherent elastic scattering for which the angles of incidence and reflection of the neutron wave vector relative to a flat surface are of equal magnitude, can be analyzed to yield the in-plane average of the vector magnetization depth profile along the surface normal.

Basic Relations:

1- Neutron Wave Vector :

$$K < \pi/d$$

d : is the distance between atoms of medium, neutrons propagation in the medium.

2- the neutron reflectivity:

$$R^{++} = \frac{1}{4I_0 f_1 f_2 P_p P_a}[I^{\text{off,off}}(F_1 P_p + 1)(F_2 P_a + 1) + I^{\text{on,on}}(P_p - 1)(P_a - 1) + \\ + I^{\text{off,on}}(F_1 P_p + 1)(P_a - 1) + I^{\text{on,off}}(P_p - 1)(F_2 P_a + 1)],$$

$$R^{--} = \frac{1}{4I_0 f_1 f_2 P_p P_a}[I^{\text{off,off}}(F_1 P_p - 1)(F_2 P_a - 1) + I^{\text{on,on}}(P_p + 1)(P_a + 1) + \\ + I^{\text{off,on}}(F_1 P_p - 1)(P_a + 1) + I^{\text{on,off}}(P_p + 1)(F_2 P_a - 1)],$$

3- Relationship between polarized neutrons and magnetic structure:

$$M_i = \int \left(p_{ii}^2(z) + p_{if}^2(z)\right) k_w^2(z) dz / k_{0z}$$

Where

$$p_{ii}(z) = \psi_i/\psi_{0i},\ p_{if}(z) = \psi_f/\psi_{0i}.$$

4- the interaction potential of the neutron with the medium:

$$U(z) = V - iW = \alpha \Sigma_g N_g b_g + \boldsymbol{\mu B} - i\left[\beta\left(\Sigma_g N_g \Sigma_p \sigma_{gp} + N_n \sigma_n + \right.\right. \\ \left.\left. N_m \sigma_m + N_{nm} \sigma_{nm}\right) + \gamma_{ne} \sigma_{ne}\right]$$

Where:

B is the magnetic field induction vector, $\alpha = 2\pi\hbar^2/m$, $\beta = \hbar v/2$, ħ is the Planck constant, μ is the neutron magnetic moment, m is the neutron mass, v is the neutron velocity.

5- Relationship between magnetic impact and reflectivity :

$$\left(-\frac{\partial^2}{\partial x^2} + u_0 \theta(x > 0) + 2\boldsymbol{\sigma B}\theta(x > 0) - k^2\right)|\boldsymbol{\Psi}(x)\rangle = 0$$

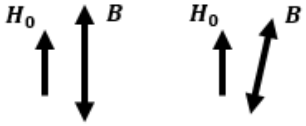
$$U = U_{nuc} + U_{mag}$$

$$U_{mag} = -\boldsymbol{\mu B} = |\mu_n|\boldsymbol{\sigma B}$$

$$M = 0 \rightarrow R_{++} = R_{--};\ R_{+-} = R_{-+} = 0$$

$$M \neq 0; B \parallel M \rightarrow R_{++} \neq R_{--};\ R_{+-} = R_{-+} = 0$$

$$M \neq 0; B \nparallel M \rightarrow R_{++} \neq R_{--};\ R_{+-} \neq 0;\ R_{-+} \neq 0$$



Based on the above mentioned, the following tasks were performed with the MATLAB code (prepared by Dr. Vladimir Zhaketov) as well as the X-ray software, and a report of its results is presented as follows:

Task 1. Fit of experimental data

Scope :

In this task, we are going to draw a reflective neutron diagram with MATLAB code and using the following formulas, and also calculate the asymmetric spin of neutrons caused by radiation with an angle of 6 radians (to the material surfaces of the model below) Continue with the help of "..." software to plot the experimental data obtained for temperatures of 12 degrees Kelvin and 1.2 degrees Kelvin and compare the results.

Method of Calculation:

At first, using the following formula, we have drawn the asymmetric spin diagram of the irradiated neutrons (with an angle of 6 radians) according to their wavelength, and then, with the help of the Spectrum Viewer software, the contour diagrams obtained from the experimental tests for the temperatures of 12 Kelvin and 1.2 Kelvin, as in the figure No. 3 and finally, using the following relations, we have drawn the spin asymmetry of neutrons according to their wavelength.

- Plot reflectivity for plus and minus states:

$$R_+ = \frac{I_+}{I_{dir}}; R_- = \frac{I_-}{I_{dir}}$$

- Calculate spin asymmetry for experimental data:

$$SA = \frac{R_+ - R_-}{R_+ + R_-}$$

- Modeling of reflectivity for nominal structure:

  Al2O3 / Nb(100nm) / Gd(3nm) / V(70nm) / Nb(15nm)

- Try to fit experimental data (make better correspondence between experiment and modeling)

- Below T<Tc(Nb,V) superconducting layers close to ferromagnet layer must be magnetized (see ‘inverse proximity effect’)

Formula for lambda calculation for experimental data:

$$\lambda[\text{Å}] = \frac{(\mathrm{N} + 35 - 12) * 3.956 * 256}{34030}$$

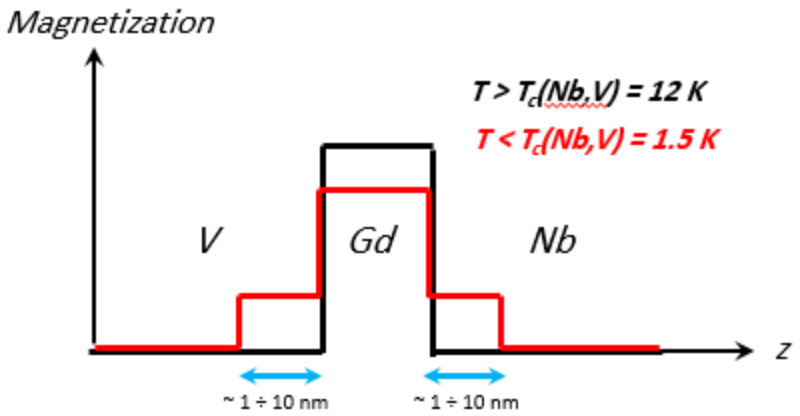


Figure No .2 – Relationship between Temperature and magnetization

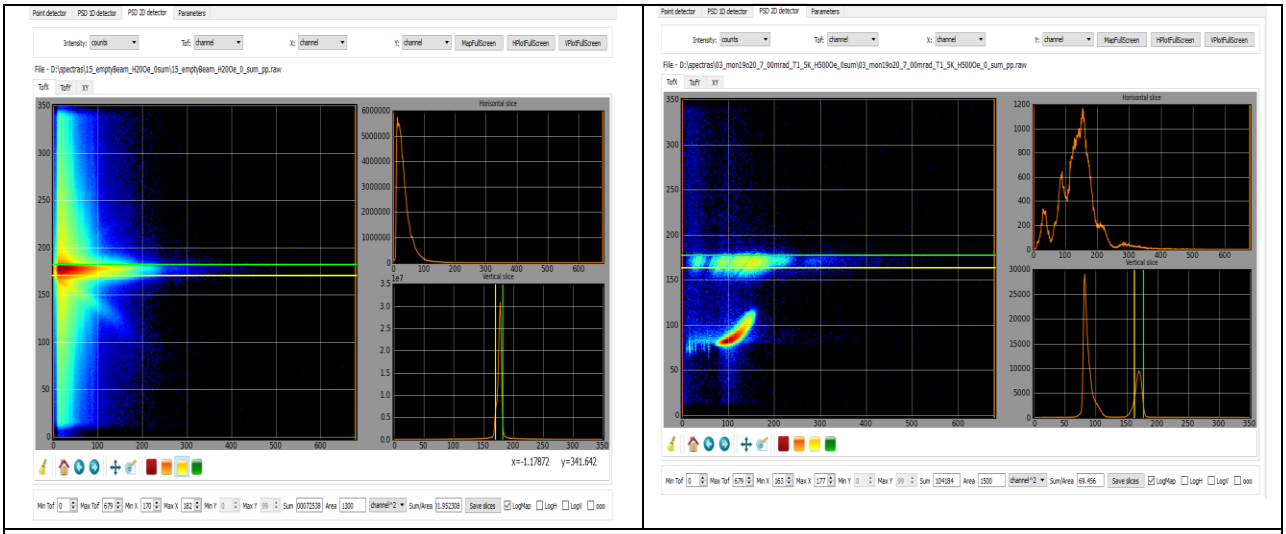


Figure No. 3 – the Experimental Data by Spectrum Viewer Software

The figure of the results related to this problem are presented as follows:

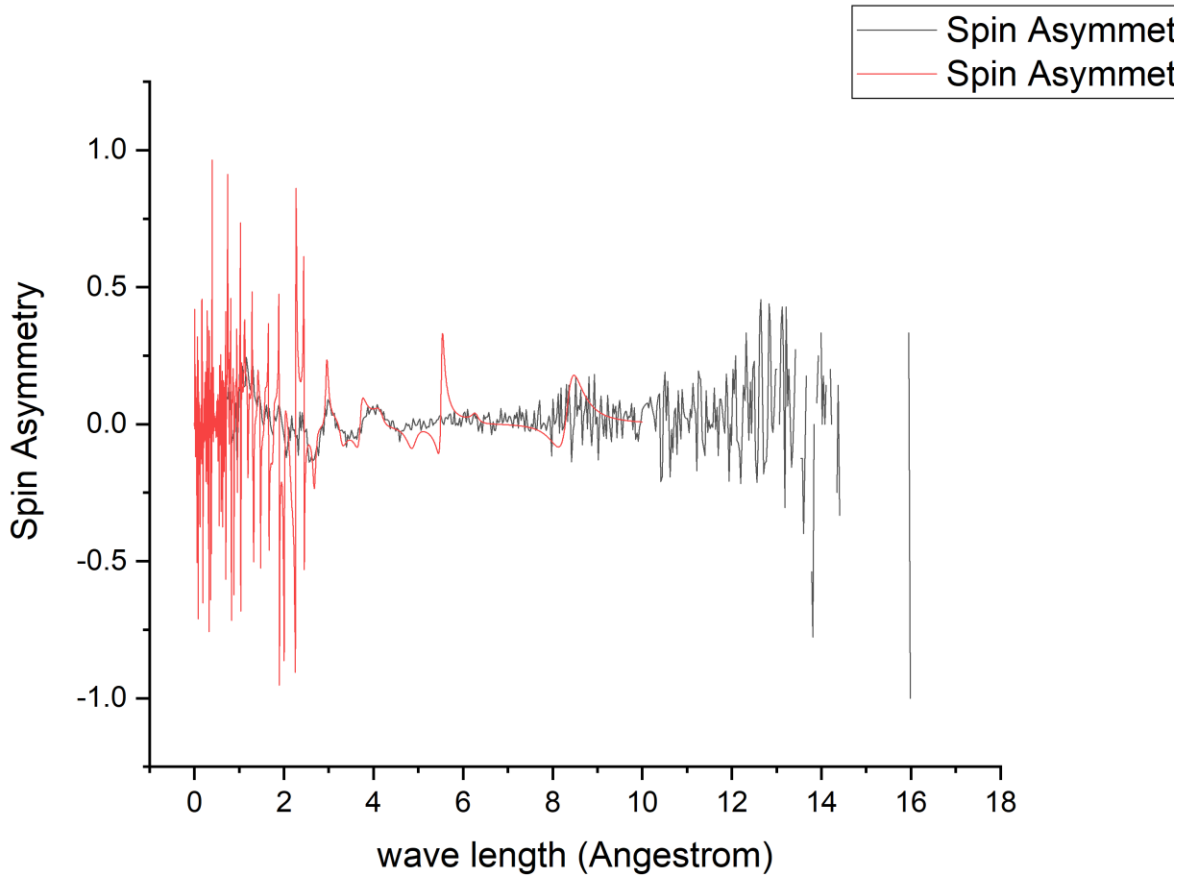


Figure No.4 - Fit of experimental and Calculated data for T=12 K

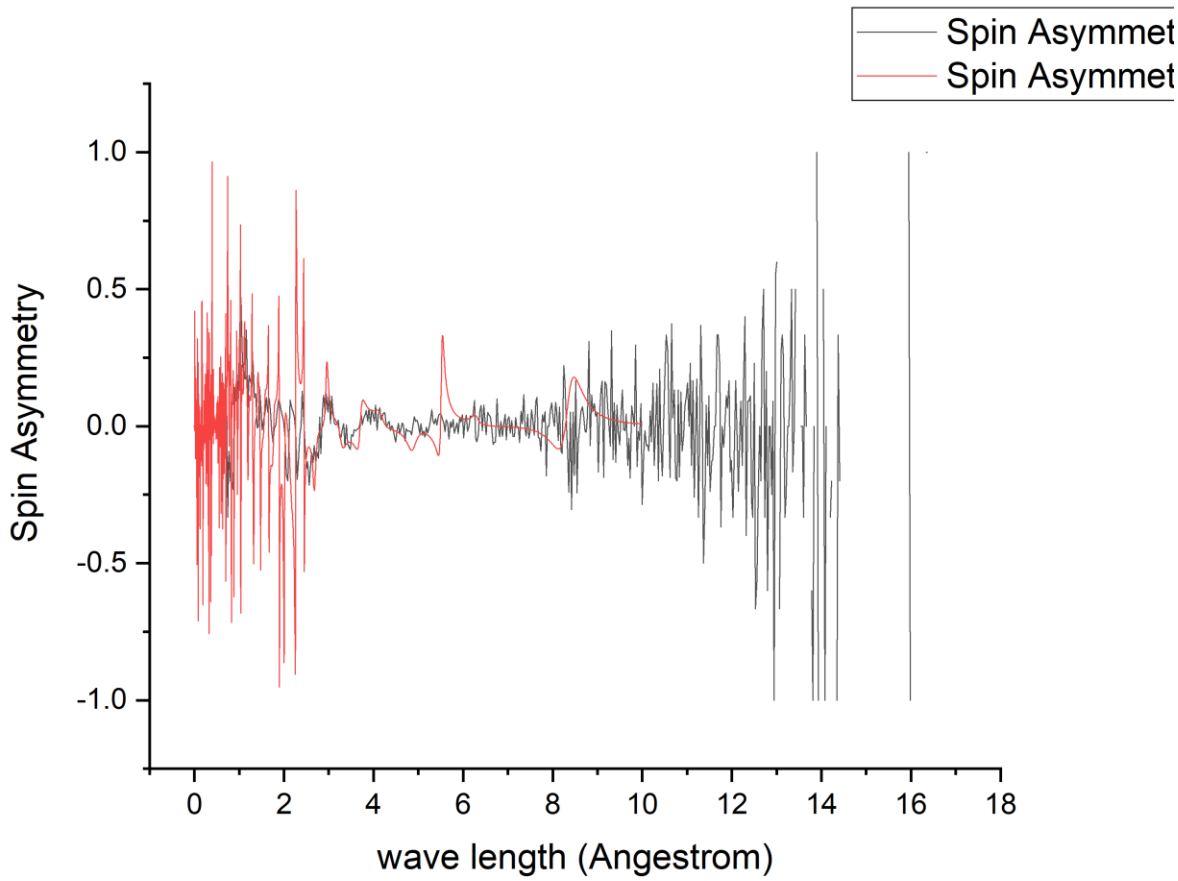


Figure No.5 - Fit of experimental and Calculated data for T=1.2 K

Conclusion:

Based on the graphs drawn below, it can be concluded that the experimental data obtained with numerical calculations have a low relative error. Of course, there is a small relative error in the wavelength of 4 angstroms. Also, for wavelengths higher than 9 angstroms, we see a mismatch between experimental data and numerical calculations.

Task 2. Comparing reflectivity at different grazing angles (Calculation only neutron reflectivity)

SCOPE:

Based on the following formula, the reflectivity of neutrons is related to the radiation angle, so in this task we intend to evaluate the effect of the radiation angle on surfaces:

$$Q = \frac{4\pi}{\lambda}\theta$$

Method of Calculation:

By changing the radiation angles in the MATLAB code and for the following model, we have evaluated the effect of these changes in reflectivity.

- Comparing reflectivity at different grazing angles by calculation only neutron reflectivity for:

  Al2O3 / Nb(100nm) / Gd(3nm) / V(70nm) / Nb(15nm) and θ = 3, 6, 12 mrad

The figure of the results related to this problem are presented as follows:

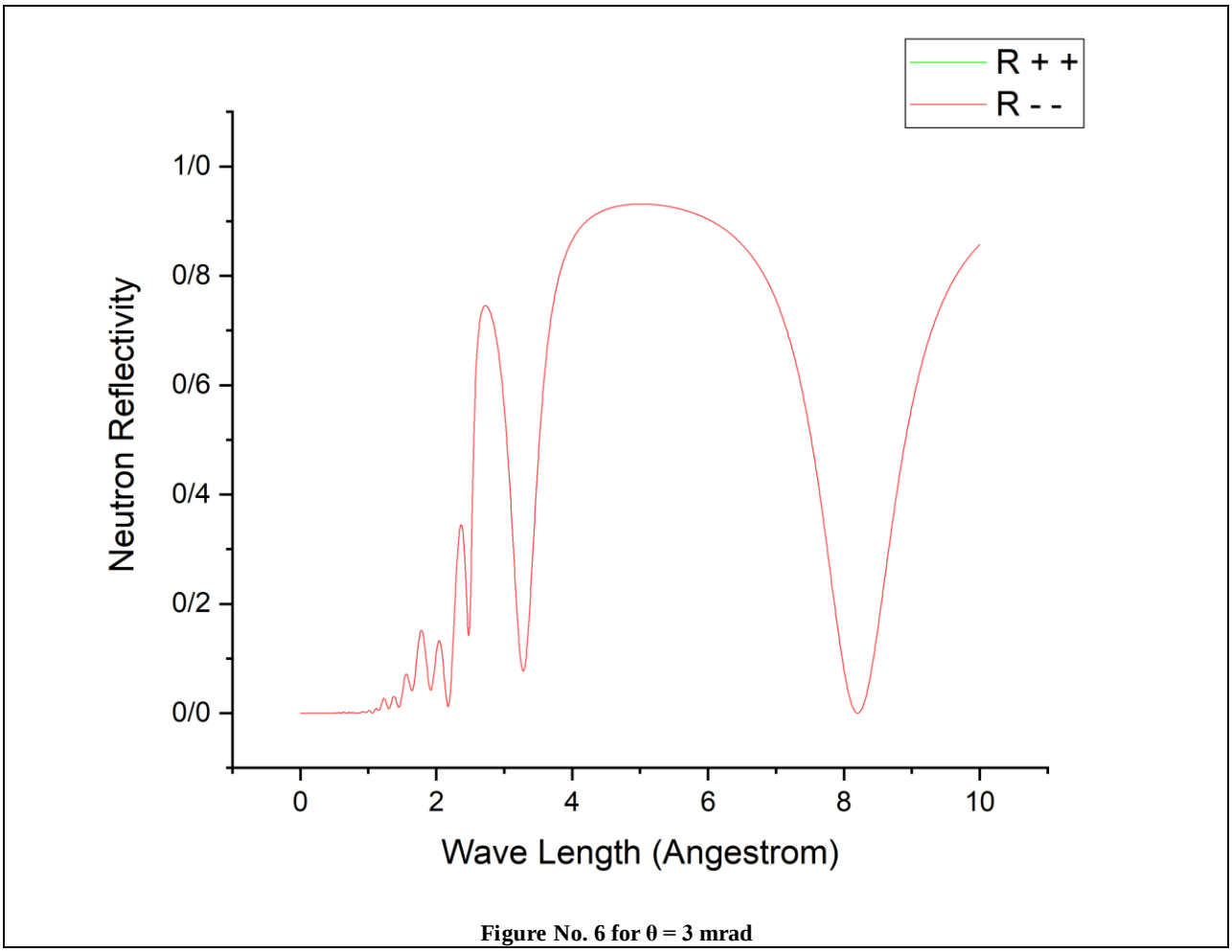


Figure No. 6 for θ = 3 mrad

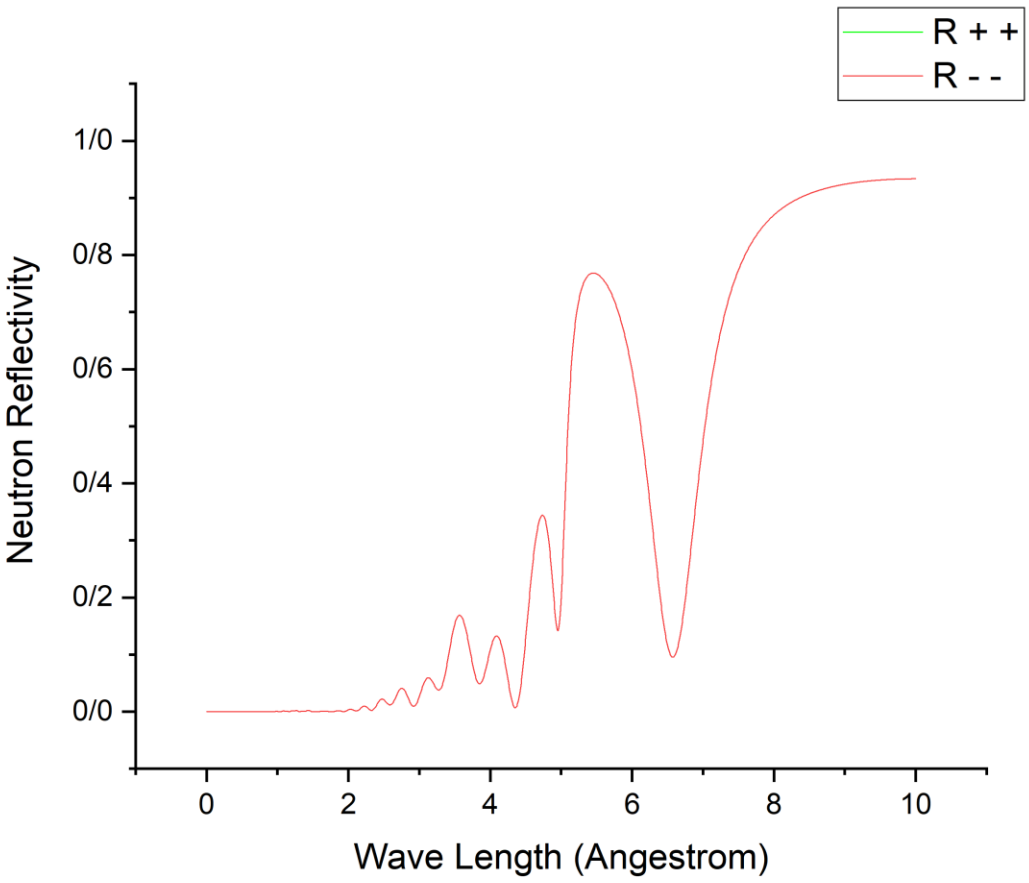


Figure No. 7 for θ = 6 mrad

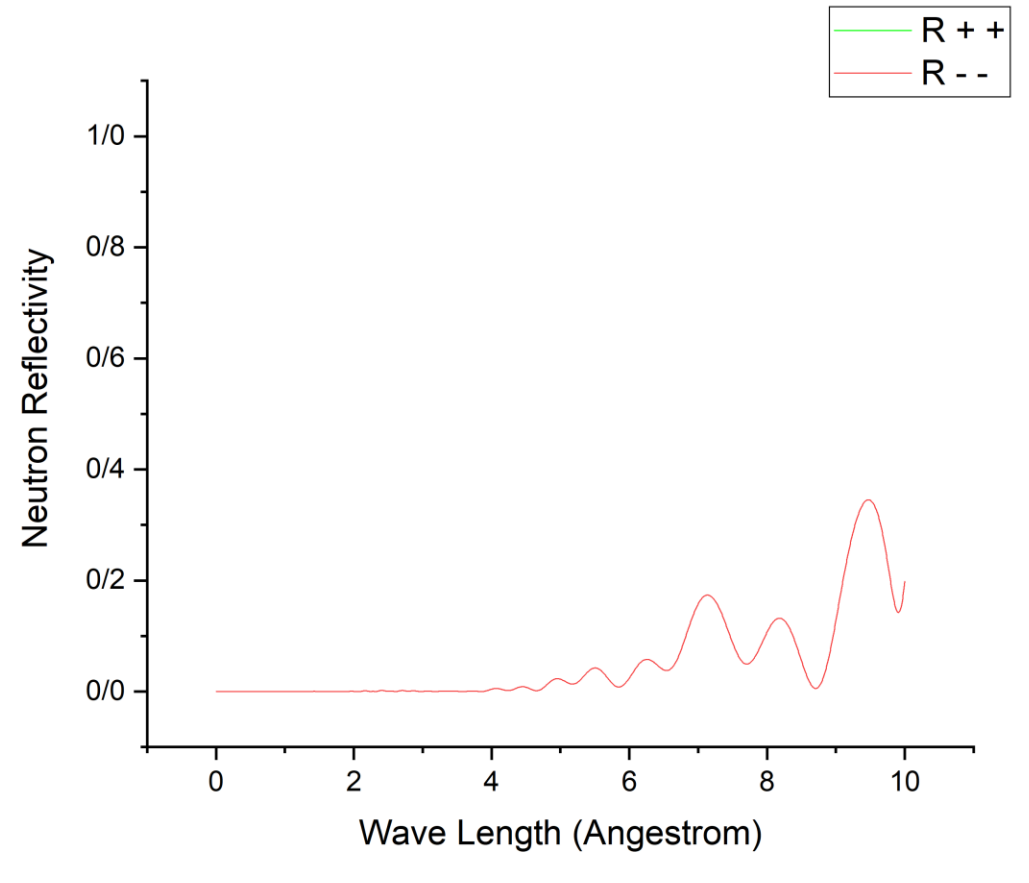


Figure No. 8 for θ = 12 mrad

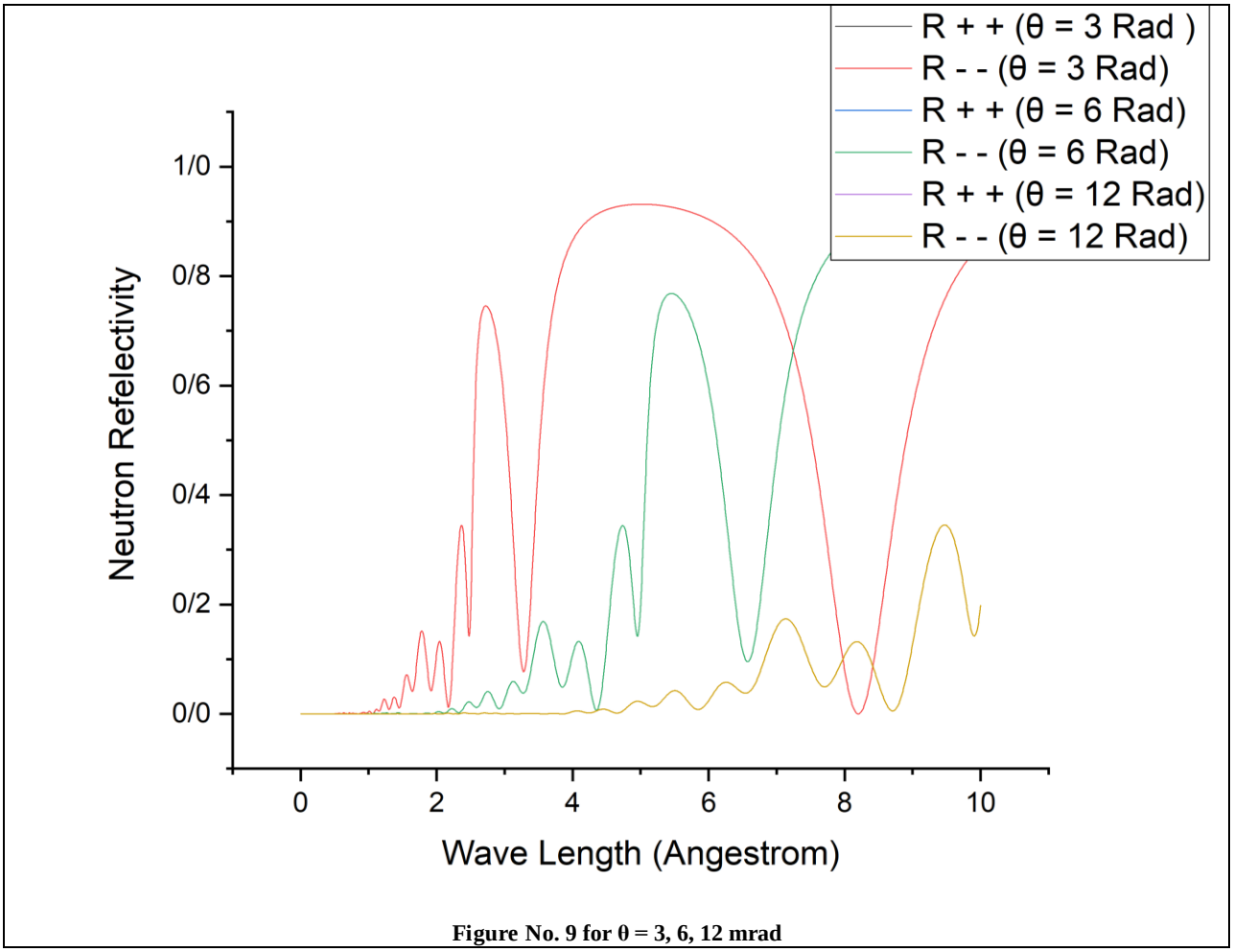


Figure No. 9 for θ = 3, 6, 12 mrad

Conclusion:

As you can see in the forms No. 6 to 9, with the increase of the radiation angle, the peaks of the graph are smaller and subsequently the reflectivity has also decreased. So much so that the maximum "peak" reflectivity for the angle of 3 radians is almost 3 times the reflectivity of the angle of 12 radians. The red and green curves corresponding to the "plus" and "minus" of neutrons overlap almost exactly, which is due to not taking into account magnetic effects.

Task 3. Comparing reflectivity at different magnetization (Calculation only neutron reflectivity)

SCOPE:

Considering that the potential considered in the Schrödinger equation is related to magnetic fields (according to the following formula), in this task we intend to evaluate the effect of the magnetic effect of materials on the reflectivity of neutrons.

$$i\hbar \frac{\partial}{\partial t}\Psi(\boldsymbol{r},t)=\left[-\frac{\hbar^2}{2m}\Delta+\mathrm{U}(\boldsymbol{r},t)\right]\Psi(\boldsymbol{r},t)$$

$$U = U_{nuc} + U_{mag}$$
$$U_{mag} = -\boldsymbol{\mu B} = |\mu_n|\boldsymbol{\sigma B}$$

Method of Calculation:

By using MATLAB code, and by changing the amount of magnetic effects of Gadolonium element, we have evaluated the reflectivity behavior as follows:

- Comparing reflectivity at different magnetization by calculation only neutron reflectivity for following data:

  Al2O3 / Nb(100nm) / Gd(3nm) / V(70nm) / Nb(15nm) AND θ = 6 mrad

  - Collinear case: Mz(Gd) = 100, 1000, 10000 Oe; Mx(Gd)=0, My(Gd)=0

  And

  - Non-collinear case: Mx(Gd) = 100, 500, 1000 Oe; Mz(Gd)=1000 Oe , My(Gd)=0

The figure of the results related to this problem are presented as follows:

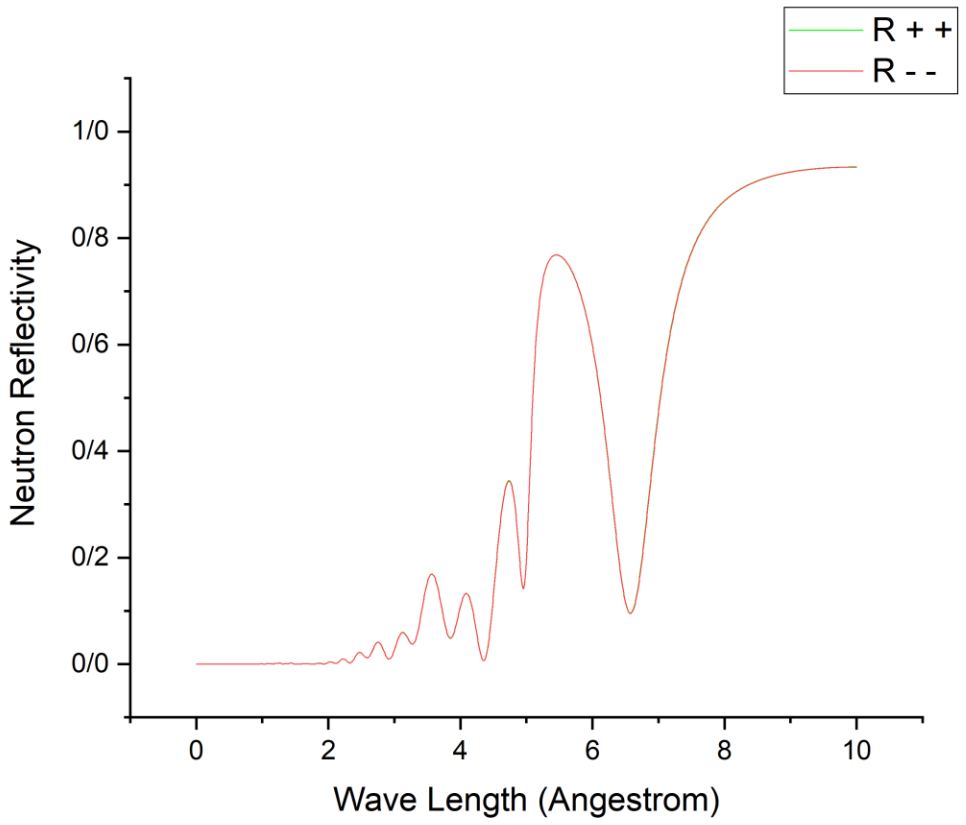


Figure No. 10 - Collinear case: Mz(Gd) = 100 , Mx(Gd)=0, My(Gd)=0

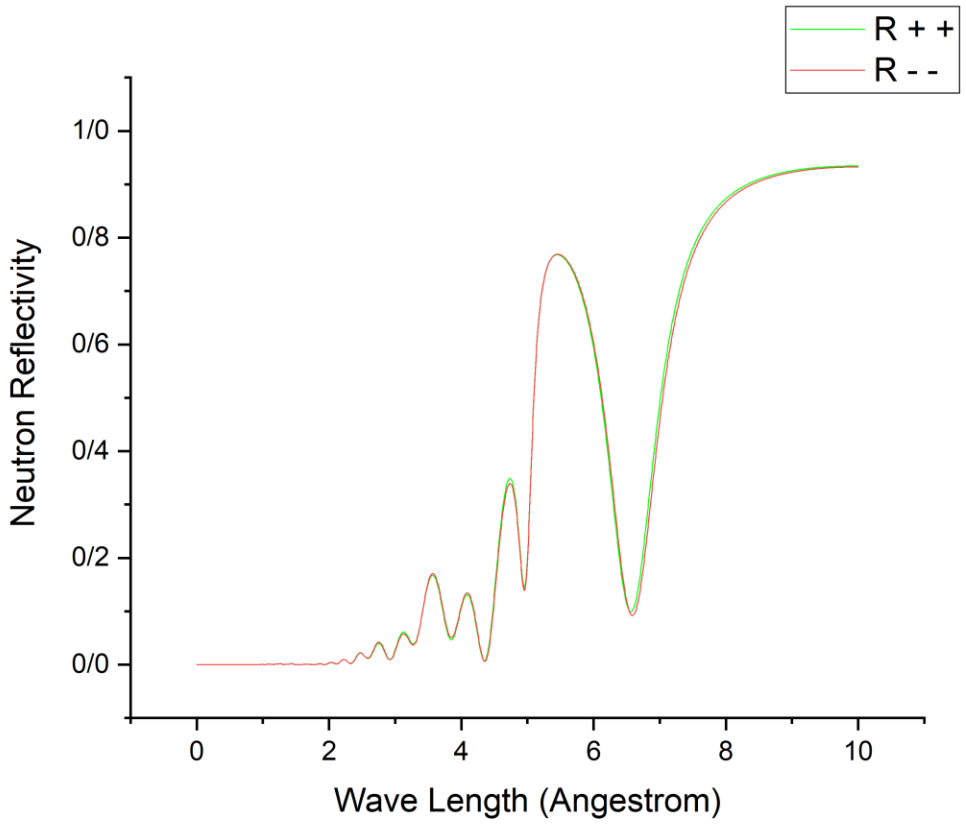


Figure No. 11 - Collinear case: Mz(Gd) = 1000 , Mx(Gd)=0, My(Gd)=0

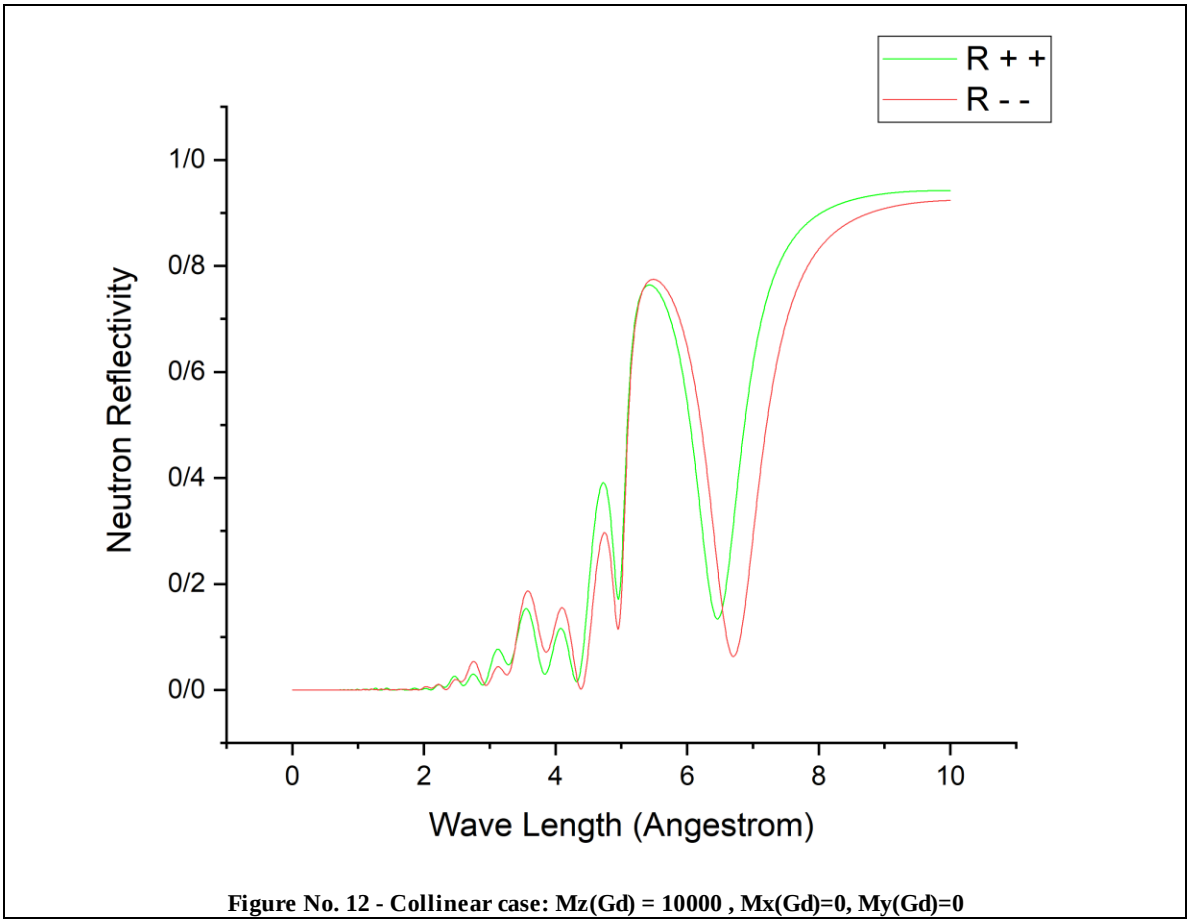


Figure No. 12 - Collinear case: Mz(Gd) = 10000 , Mx(Gd)=0, My(Gd)=0

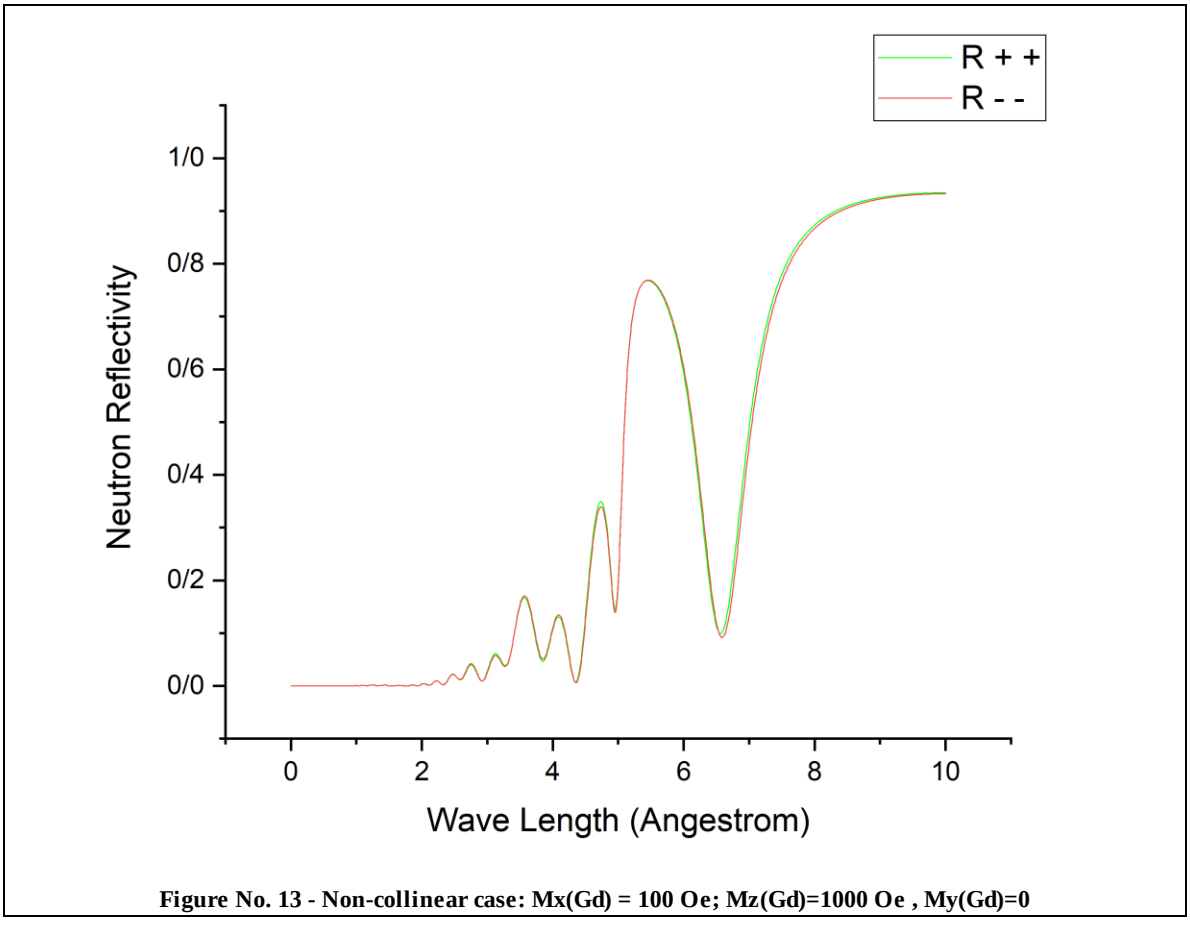


Figure No. 13 - Non-collinear case: Mx(Gd) = 100 Oe; Mz(Gd)=1000 Oe , My(Gd)=0

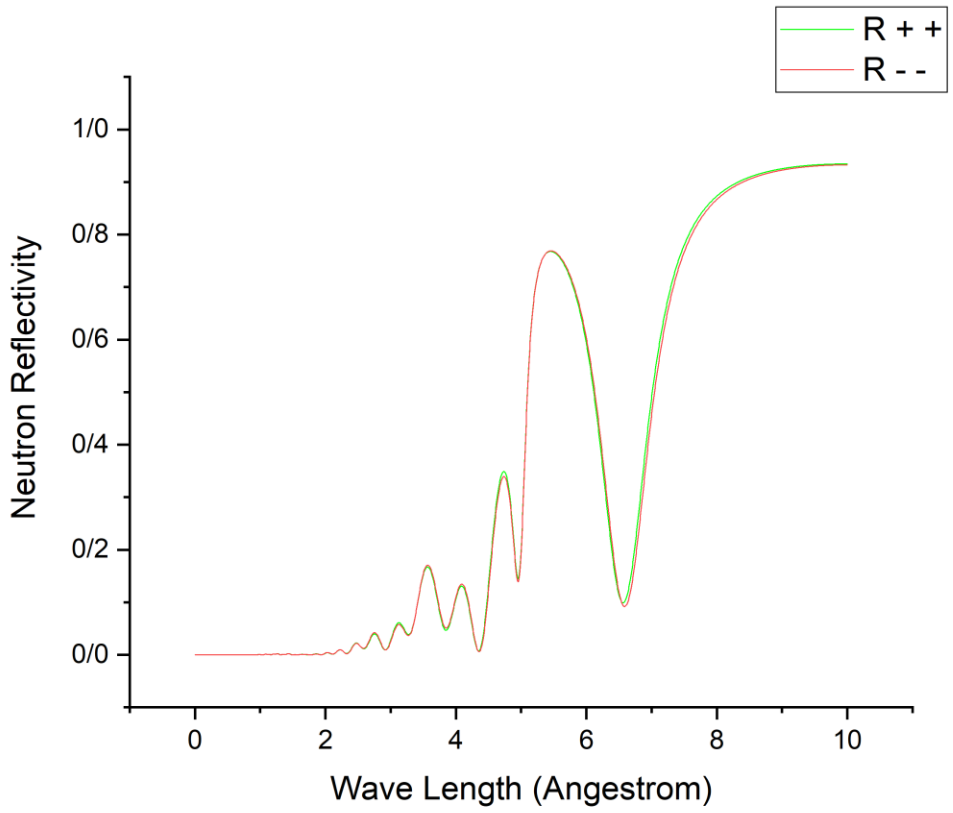


Figure No. 14 - Non-collinear case: Mx(Gd) = 500 Oe; Mz(Gd)=1000 Oe , My(Gd)=0

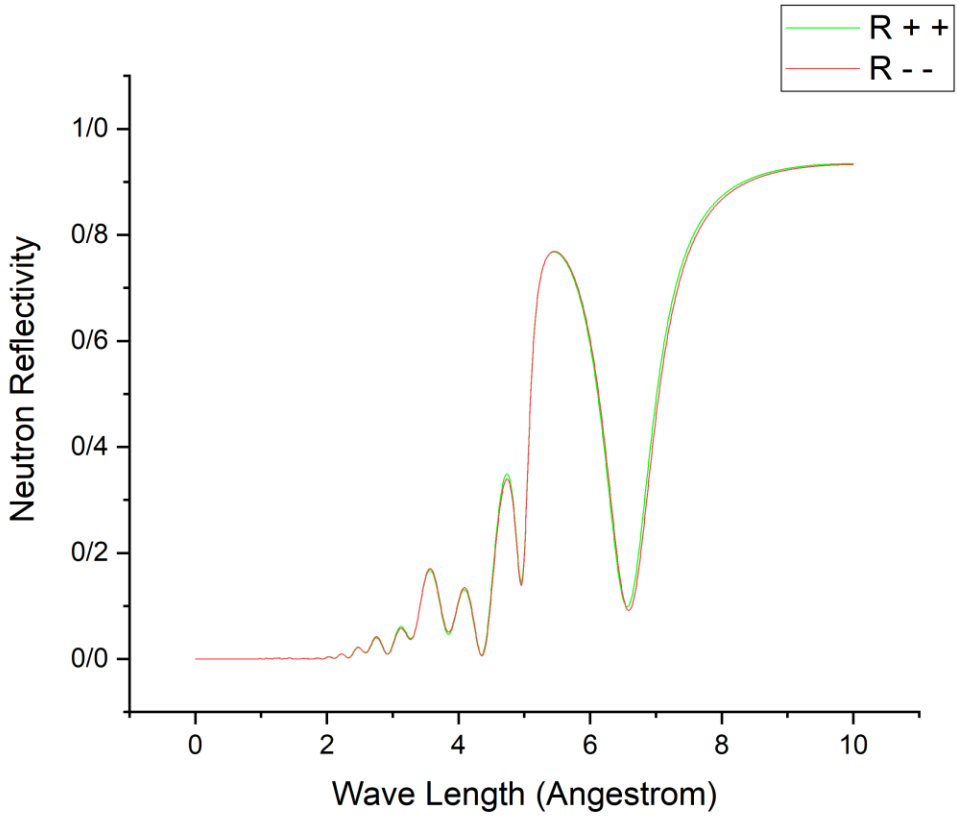


Figure No. 15 - Non-collinear case: Mx(Gd) = 1000 Oe; Mz(Gd)=1000 Oe , My(Gd)=0

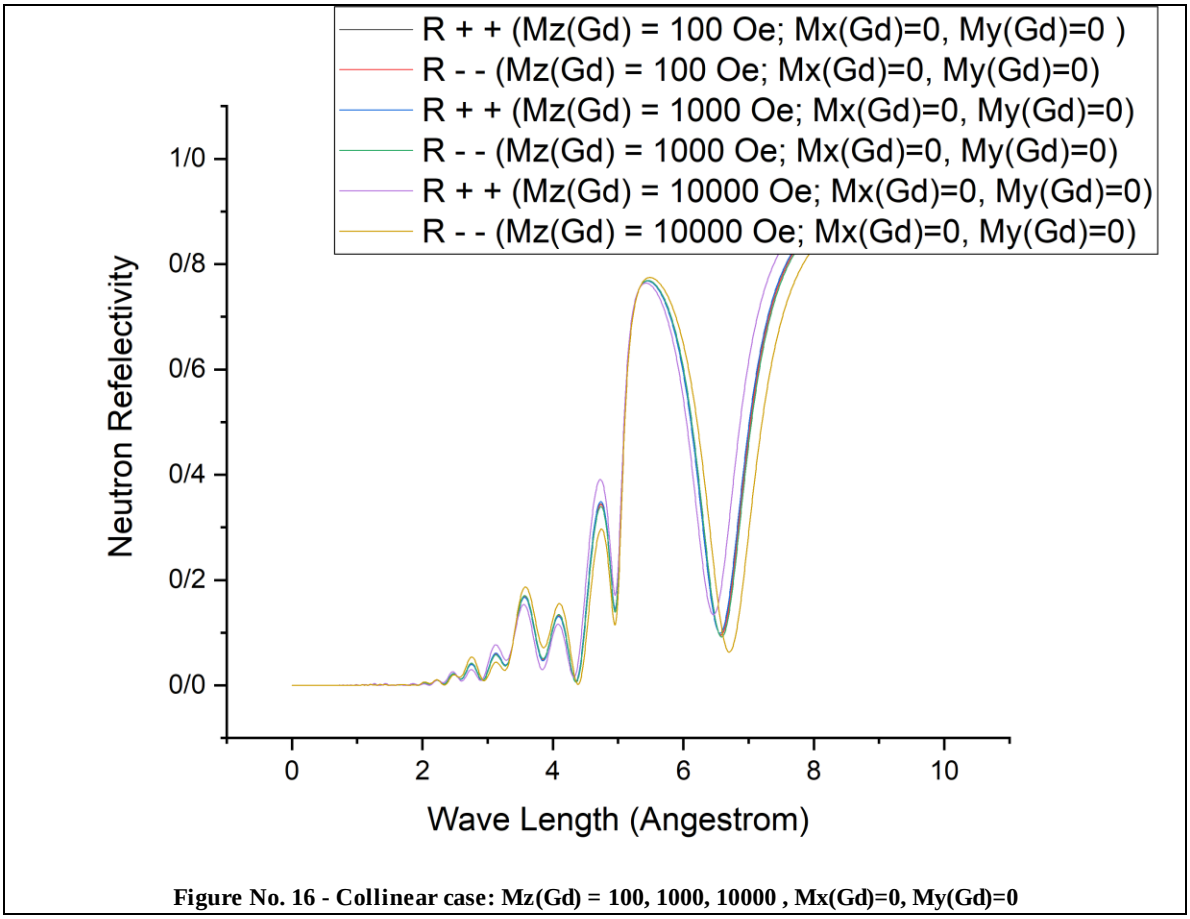


Figure No. 16 - Collinear case: Mz(Gd) = 100, 1000, 10000 , Mx(Gd)=0, My(Gd)=0

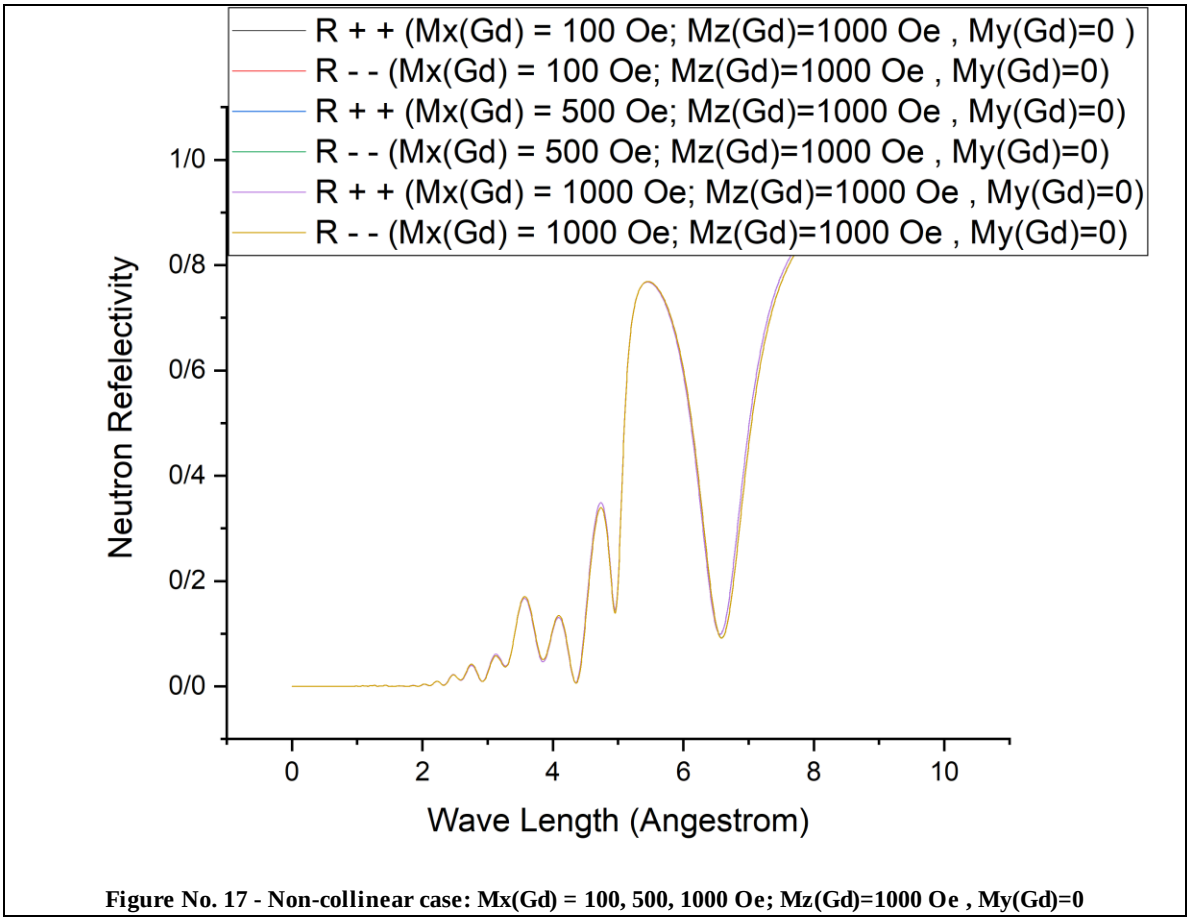


Figure No. 17 - Non-collinear case: Mx(Gd) = 100, 500, 1000 Oe; Mz(Gd)=1000 Oe , My(Gd)=0

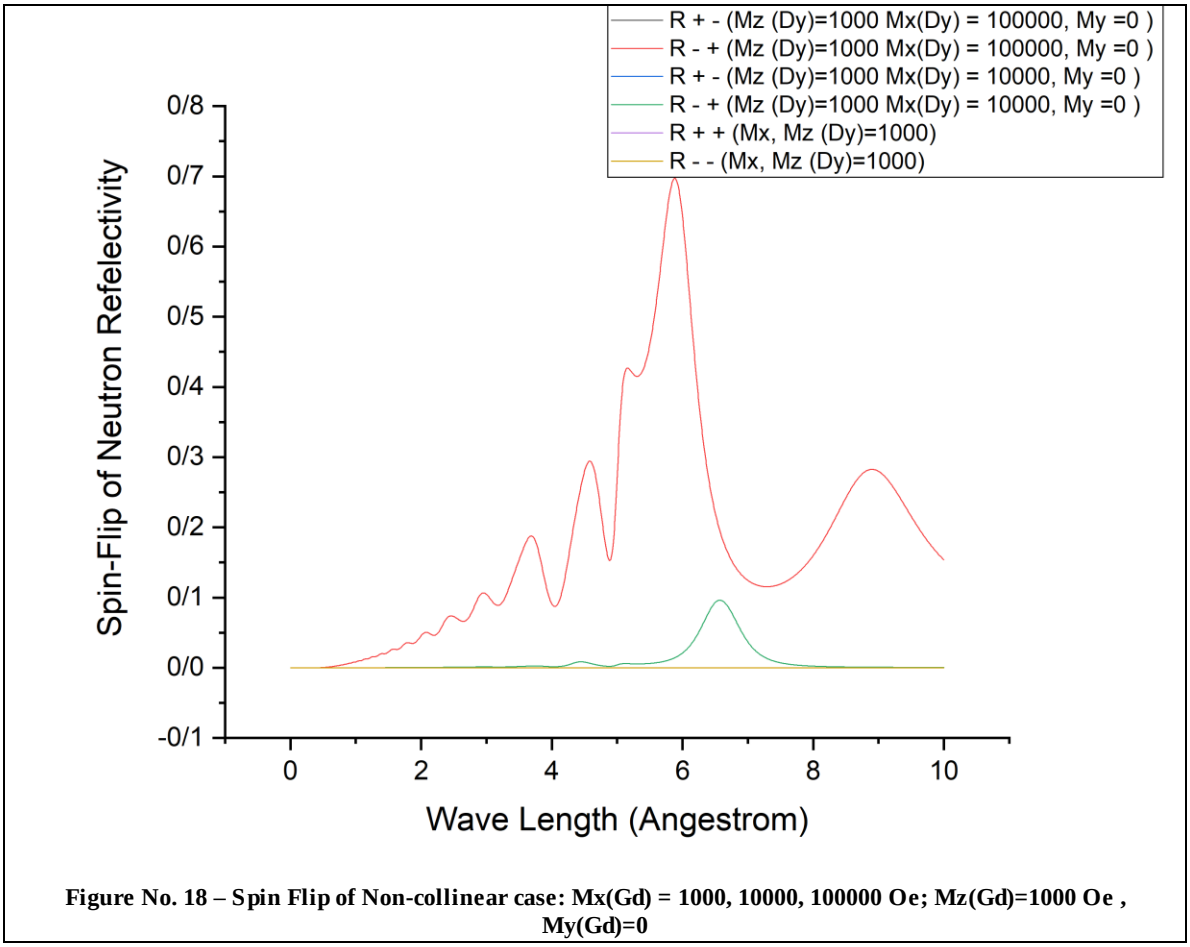


Figure No. 18 – Spin Flip of Non-collinear case: Mx(Gd) = 1000, 10000, 100000 Oe; Mz(Gd)=1000 Oe , My(Gd)=0

Conclusion:

As you can see in the figures No. 10 to 17 with the increase of Mz (Mx, My remaining constant), neutron polarization occurs and the green and red graphs that display "plus" and "minus", will appear. Also, for Mz=1000 and the change of Mx, considering the constancy of the reflectivity graphs, it can be concluded that the polarized neutrons that have occurred remain constant. According to Figure No.18, as Mx increases, Spin Flip also increases and peaks appear. It is worth mentioning, that until Mx=1000, Spin Flips were zero.

Task 4. Comparing structures with different thickness (Calculation of neutron and X-ray reflectivity)

Scope:

Based on the following relationships, it can be concluded that the thickness affects the reflectivity, so in this task we intend to investigate this effect.

Method of Calculation:

First, using MATLAB code for different thicknesses of gadolinium in the following model, he drew neutron reflectivity change diagrams and then using X-ray reflectivity software (which is specific for evaluating secondary radiations due to the impact of thermal neutrons on elements) we have evaluated the changes in X-ray intensity for the same thicknesses.

- Comparing structures with different thickness by calculation of neutron and X-ray reflectivity for following data:

  Al2O3 / Nb(100nm) / Gd(3nm) / V(70nm) / Nb(15nm),No Magnetization and θ = 6 mrad

  Al2O3 / Nb(100nm) / Gd(6nm) / V(70nm) / Nb(15nm), No Magnetization and θ = 6

  Al2O3 / Nb(100nm) / Gd(12nm) / V(70nm) / Nb(15nm), No Magnetization and θ = 6

The figure of the results related to this problem are presented as follows:

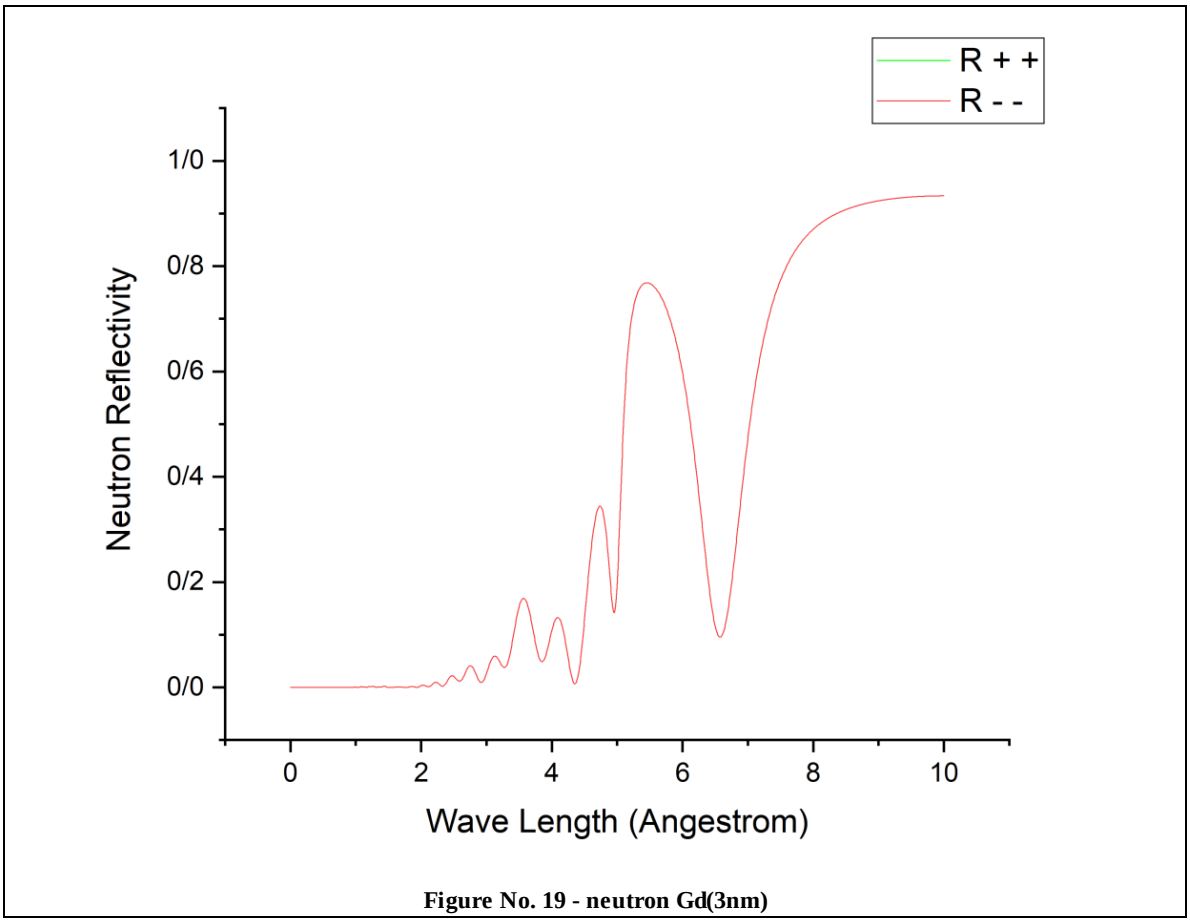


Figure No. 19 - neutron Gd(3nm)

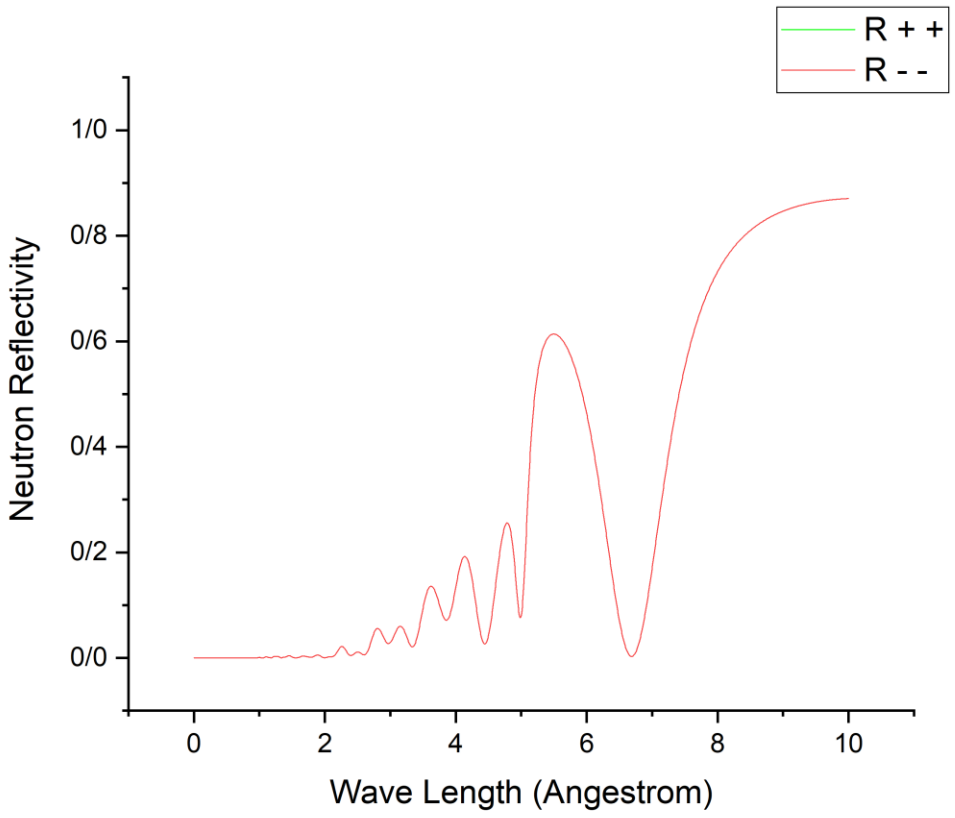


Figure No. 20 - neutron Gd(6nm)

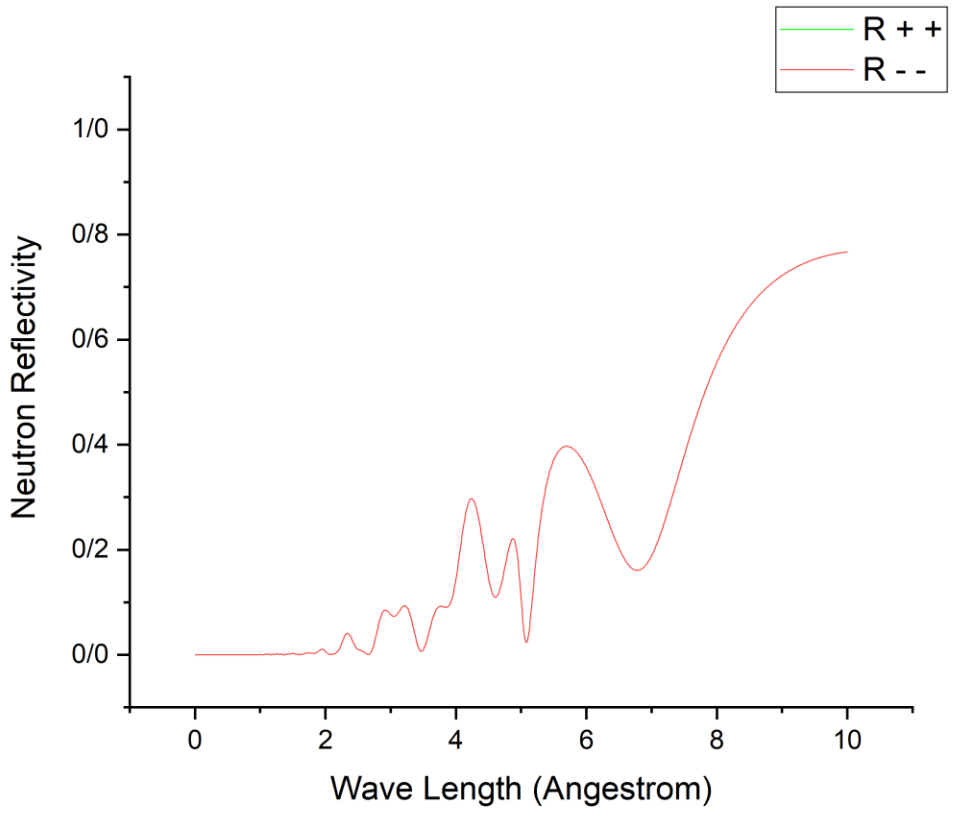


Figure No. 21 - neutron Gd(12nm)

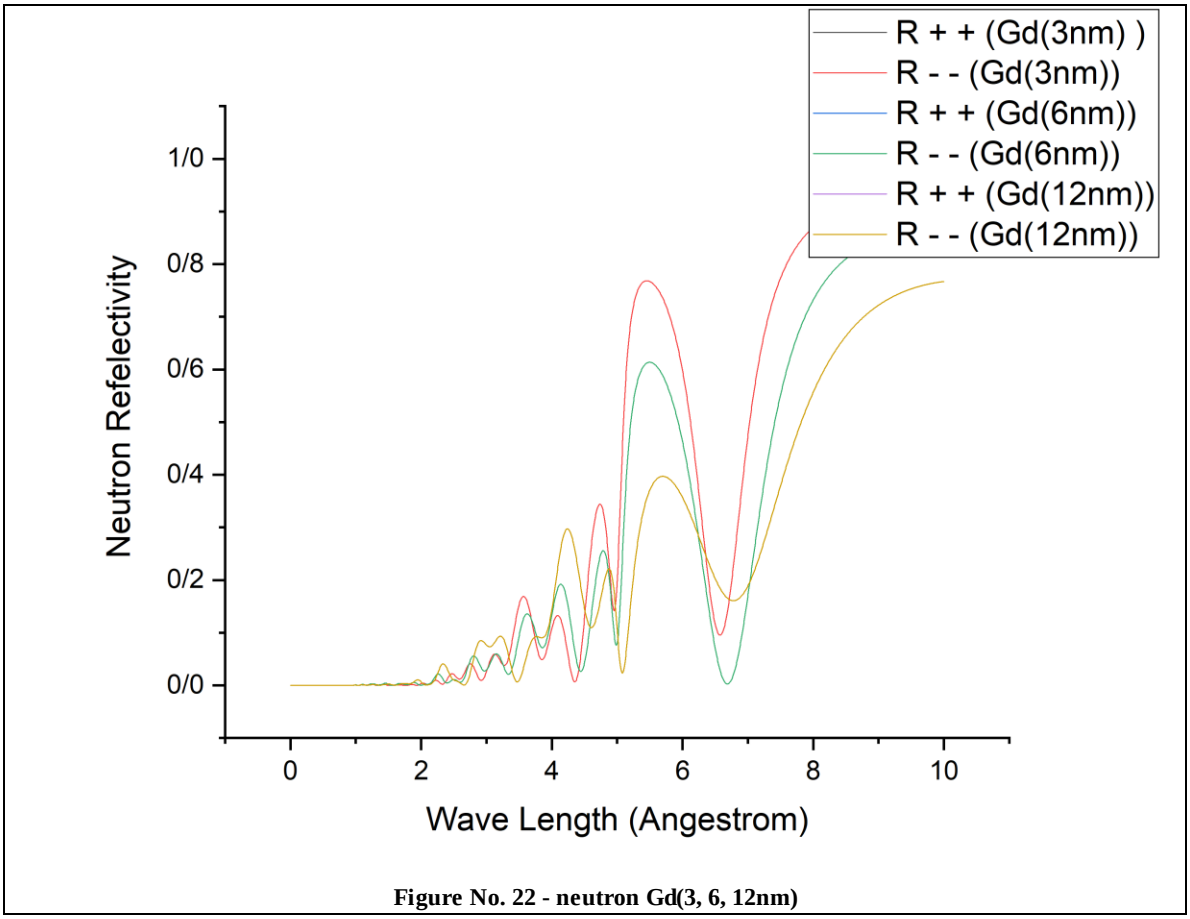


Figure No. 22 - neutron Gd(3, 6, 12nm)

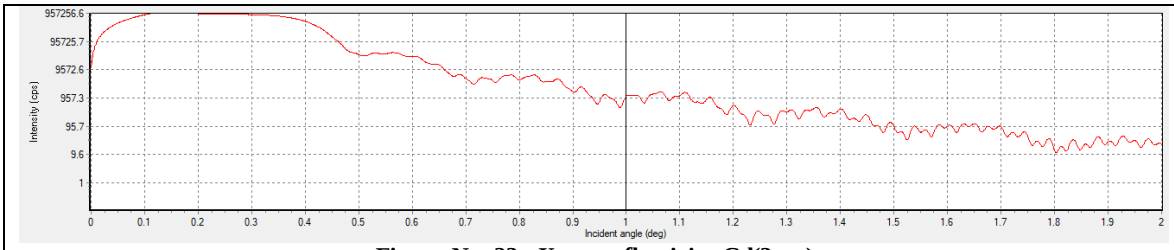


Figure No. 23 - X-ray reflectivity Gd(3nm)

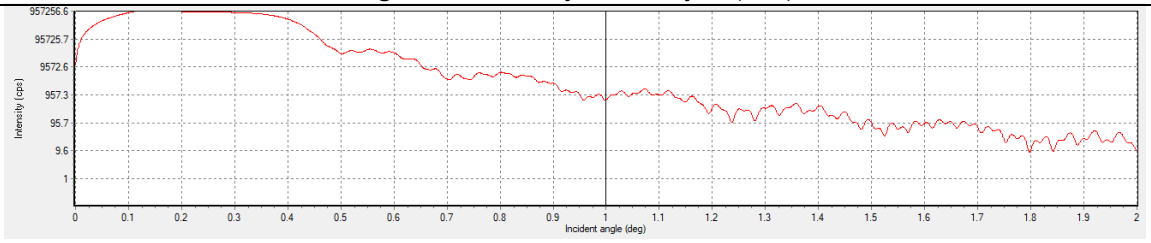


Figure No. 24 - X-ray reflectivity Gd(6nm)

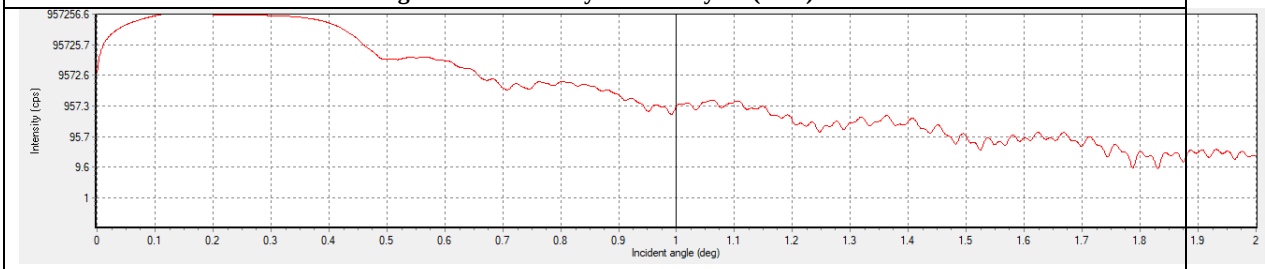


Figure No. 25 - X-ray reflectivity Gd(12nm)

Conclusion:

It can be concluded from the figures No. 19 to 22 that with the increase in the thickness of the Gadalonium element, the reflectivity also decreases, so that the maximum peak with a thickness of 3 nm is almost 2 times the maximum peak reflectivity with a thickness of 12 nm. be Also, based on figures No. 23 to 25, at a thickness of 12 nm and for an incident angle between 13 and 15 degrees, the X-ray intensity increases, and as well, regarding to the formula $Q = 4\pi\theta/Landa$, decreasing the intensity of x-ray is acceptable.

Task 5. Comparing structures with different Ferro magnets (Calculation of neutron and X-ray reflectivity)

Scope:

In this task, we intend to evaluate the effect of different ferromagnetic elements on reflectivity.

Method of Calculation:

First, using MATLAB code, we have drawn the reflectivity diagrams for the following models and ferromagnetic elements, Gd, Fe, Co, Ni, Dy (for a thickness of 3 nanometers) and then the amount of X-ray intensity produced by neutron radiation We have also drawn with the help of X-ray reflectivity software.

- Comparing structures with different Ferro magnets by calculation of neutron and X-ray reflectivity for following data:

  Al2O3 / Nb(100nm) / Gd(3nm) / V(70nm) / Nb(15nm),No Magnetization and $\theta = 6$ mrad

  Al2O3 / Nb(100nm) / Fe(3nm) / V(70nm) / Nb(15nm), No Magnetization and $\theta = 6$ mrad

  Al2O3 / Nb(100nm) / Co(3nm) / V(70nm) / Nb(15nm), No Magnetization and $\theta = 6$ mrad

  Al2O3 / Nb(100nm) / Ni(3nm) / V(70nm) / Nb(15nm), No Magnetization and $\theta = 6$ mrad

  Al2O3 / Nb(100nm) / Dy(3nm) / V(70nm) / Nb(15nm),No Magnetization and $\theta = 6$ mrad

The figure of the results related to this problem are presented as follows:

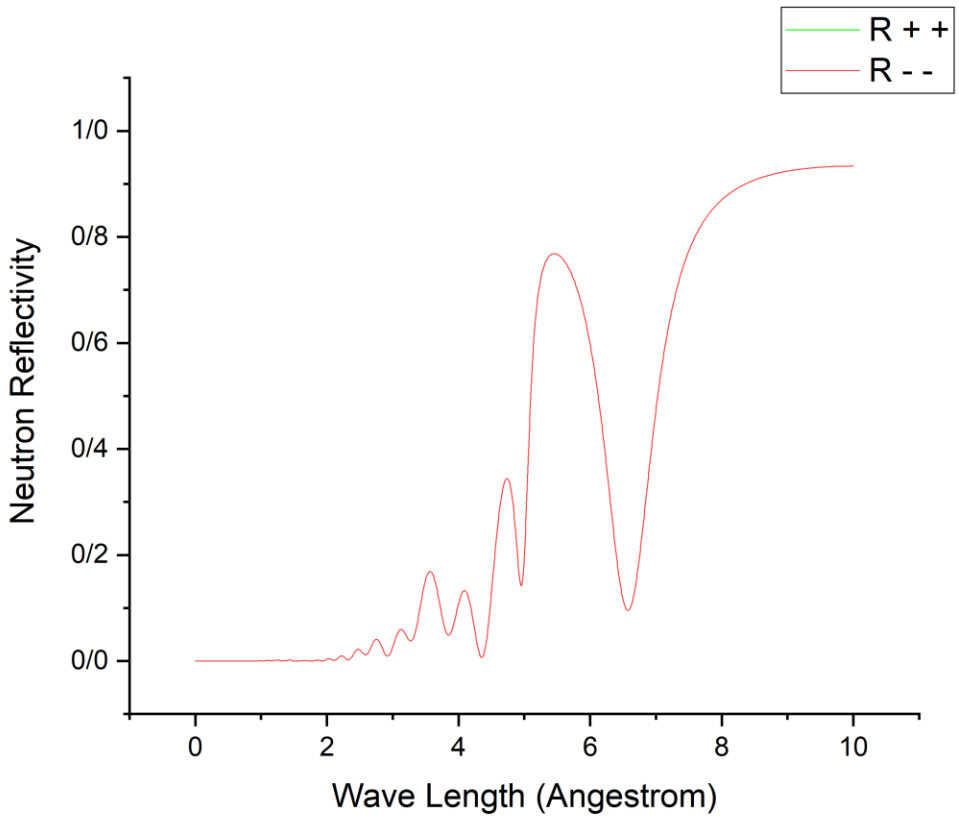


Figure No. 26 - calculation of neutron for Gd(3nm)

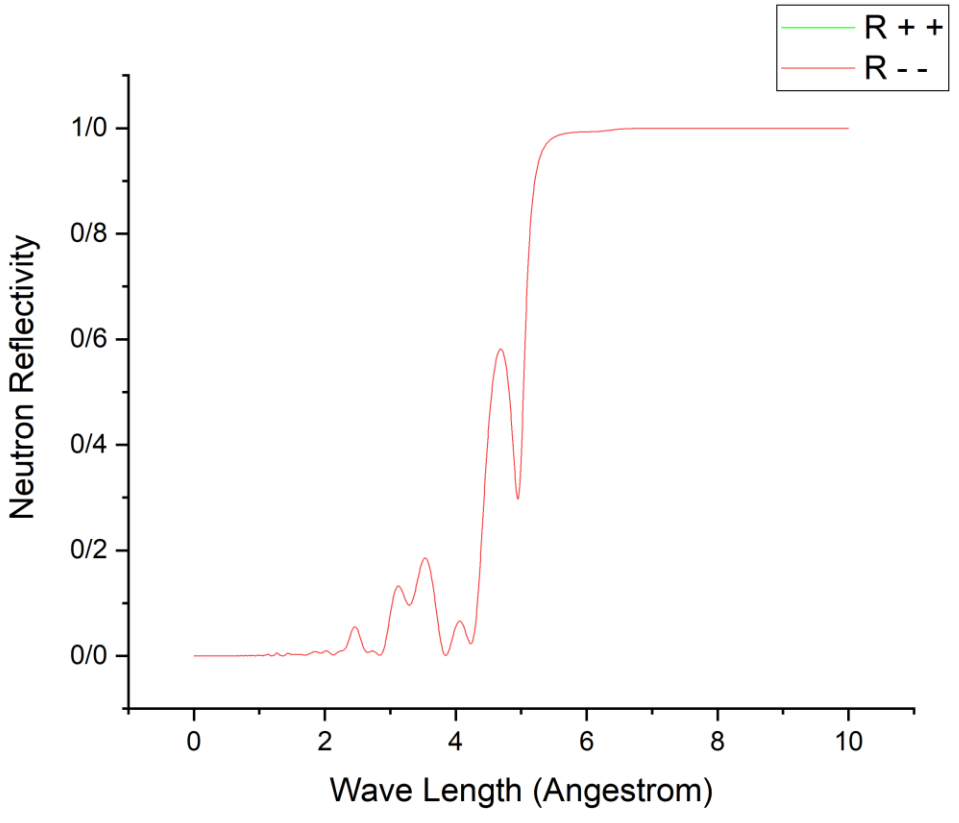


Figure No. 27 - calculation of neutron for Fe(3nm)

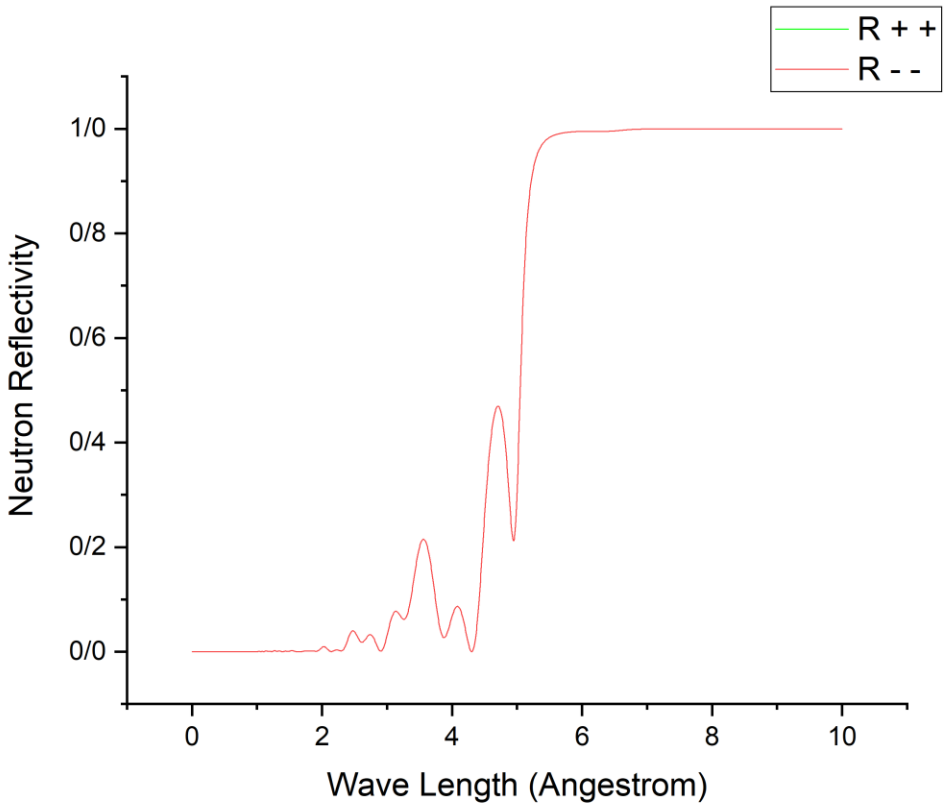


Figure No. 28 - calculation of neutron for Co(3nm)

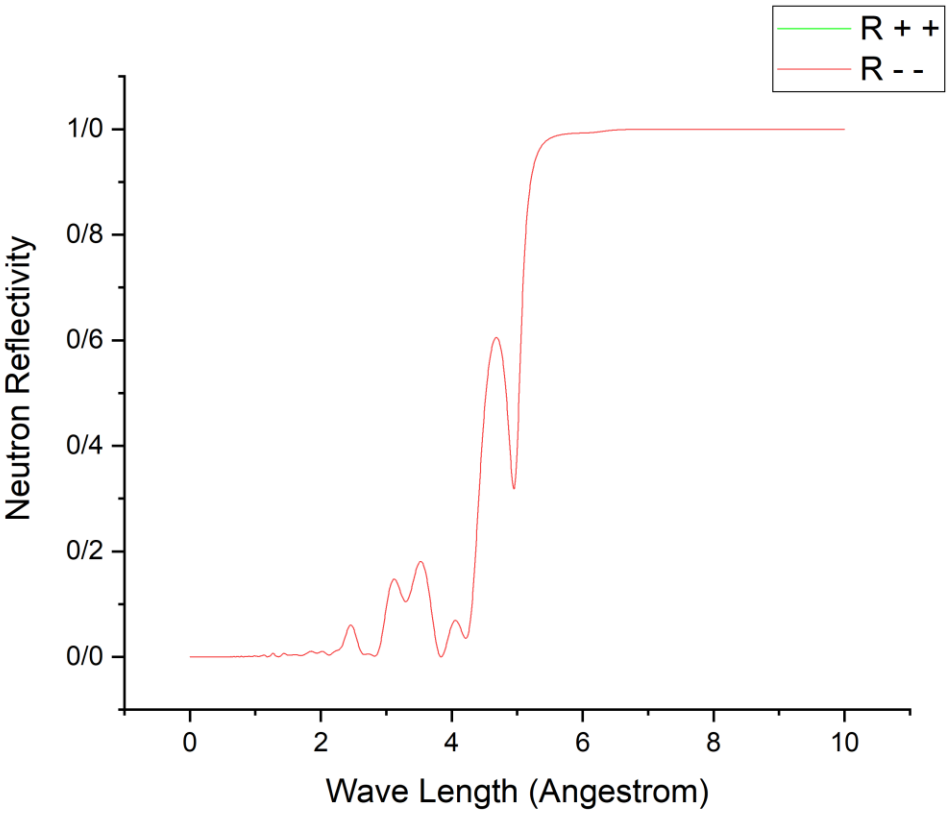


Figure No. 29 - calculation of neutron for Ni(3nm)

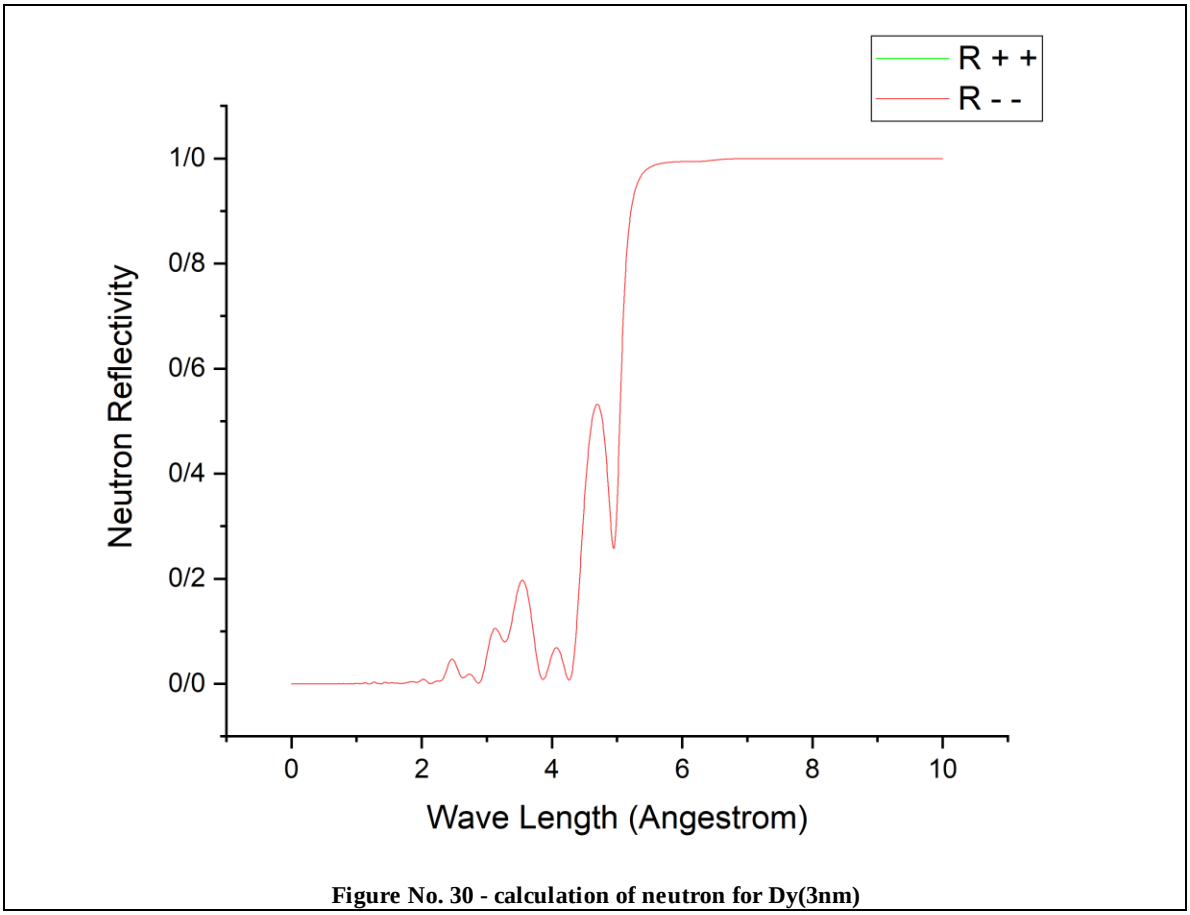


Figure No. 30 - calculation of neutron for Dy(3nm)

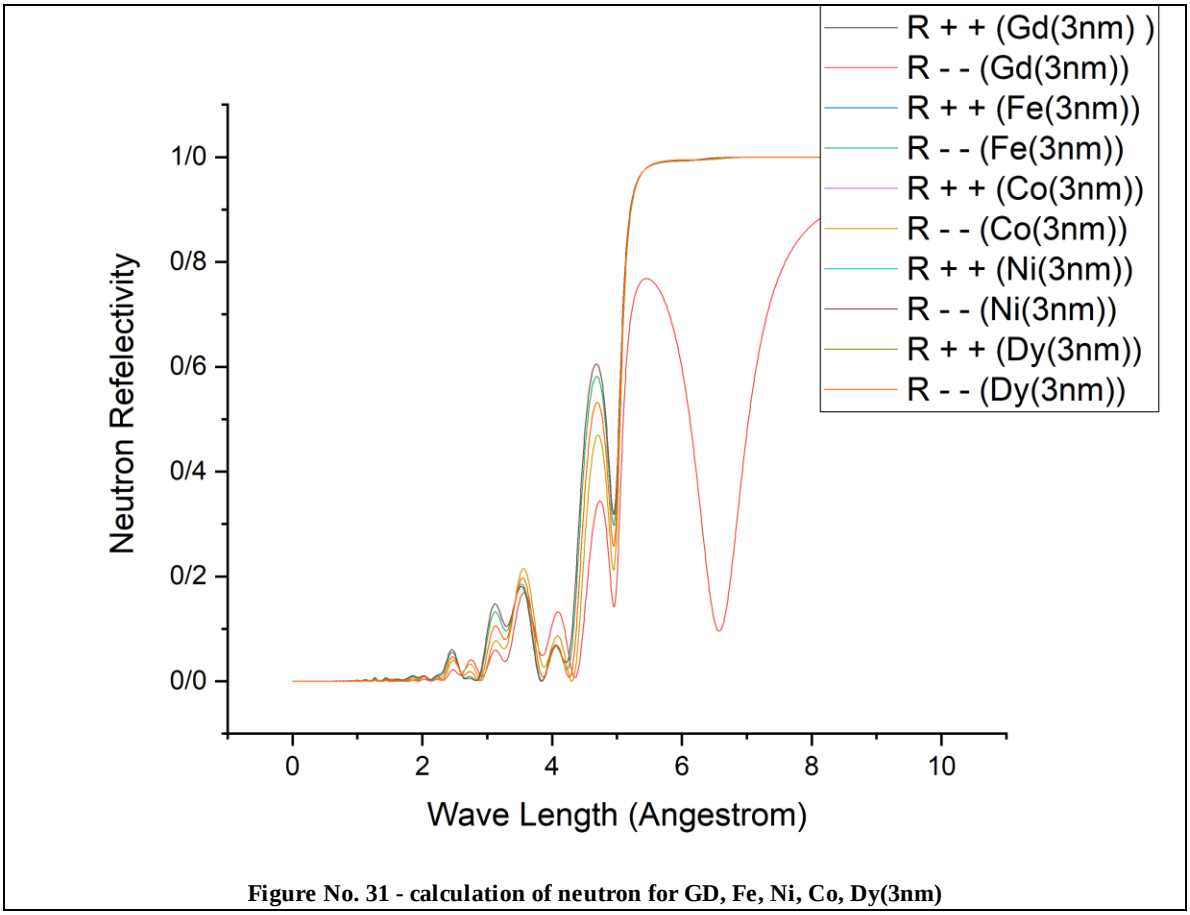


Figure No. 31 - calculation of neutron for GD, Fe, Ni, Co, Dy(3nm)

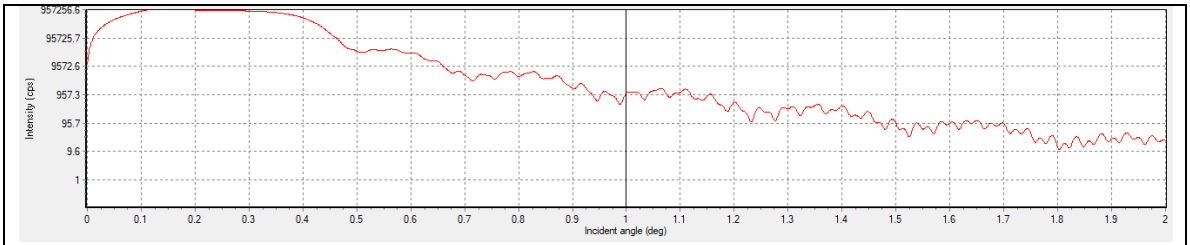


Figure No. 32 - X-ray reflectivity for Gd(3nm)

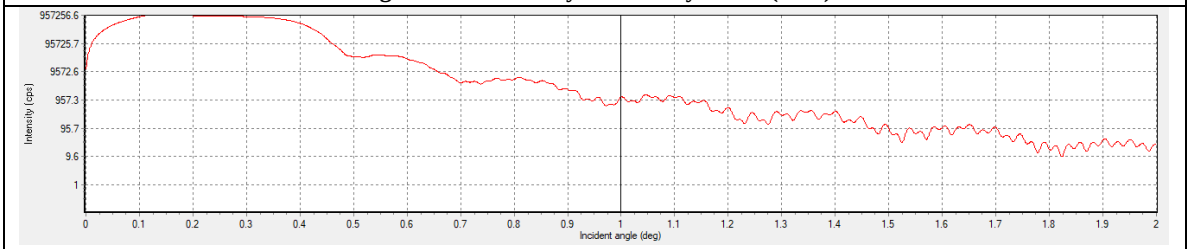


Figure No. 33 - X-ray reflectivity for Fe(3nm)

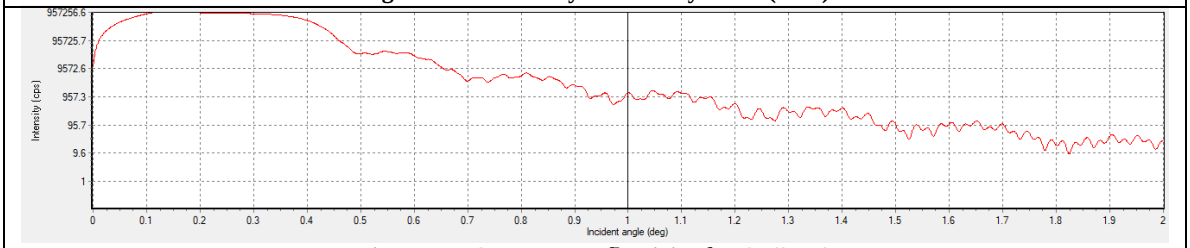


Figure No. 34 - X-ray reflectivity for Co(3nm)

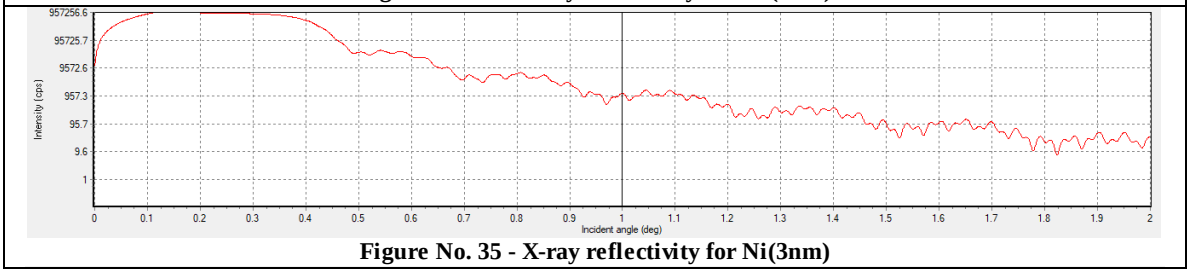


Figure No. 35 - X-ray reflectivity for Ni(3nm)

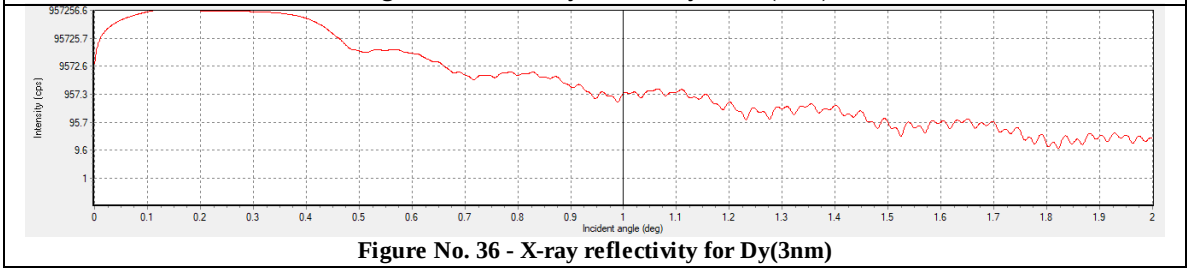


Figure No. 36 - X-ray reflectivity for Dy(3nm)

Conclusion:

As you can see in the graphs No. 26 to 31, the highest maximum peak value is related to the diagram of Gadolinium element and also the lowest maximum peak value is related to cobalt element and then iron and the reflectivity diagram of iron, cobalt and nickel elements are similar to each other. In addition, based on the observation of graphs No. 32 to 36, it can be concluded that almost all of them are similar and there is no change in X-ray reflectivity between different ferromagnetic elements.

Task 6. Super-lattice
(Calculation of neutron and X-ray reflectivity)

Scope:

In this task, we intend to evaluate the reflectivity changes for the determined super lattices of this specific layer coupling by coupling Gadolinium and Niobium elements as a specific layer and repeating them several times.

Method of Calculation:

At first, using MATLAB code for the following models and as shown in the figure below (for 10, 20, and 30 couples of Gadolinium and Niobium layers), we have drawn the graphs of their reflectivity changes. And continue using the X-ray reflectivity software, we have drawn the graph of the changes in radiation intensity, for this number of layers.

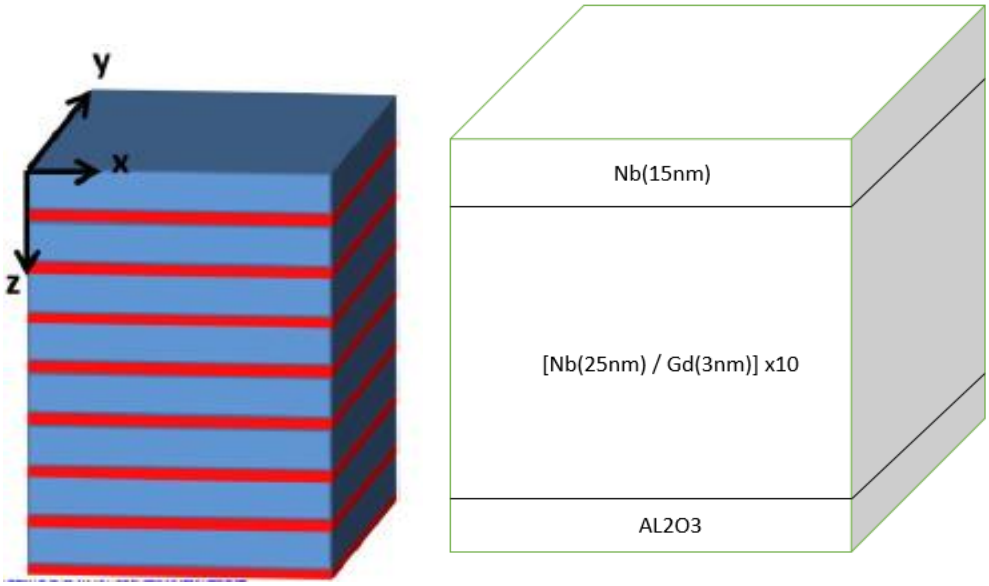


Figure No. 31 – Samples of super lattice models

- Super-lattice by calculation of neutron and X-ray reflectivity for following data:

  Al2O3 / [Nb(25nm) / Gd(3nm)] x10 / Nb(15nm) ,No Magnetization and $\theta = 6$ mrad

  Al2O3 / [Nb(25nm) / Gd(3nm)] x20 / Nb(15nm) ,No Magnetization and $\theta = 6$ mrad

  Al2O3 / [Nb(25nm) / Gd(3nm)] x30 / Nb(15nm) ,No Magnetization and $\theta = 6$ mrad

The figure of the results related to this problem are presented as follows:

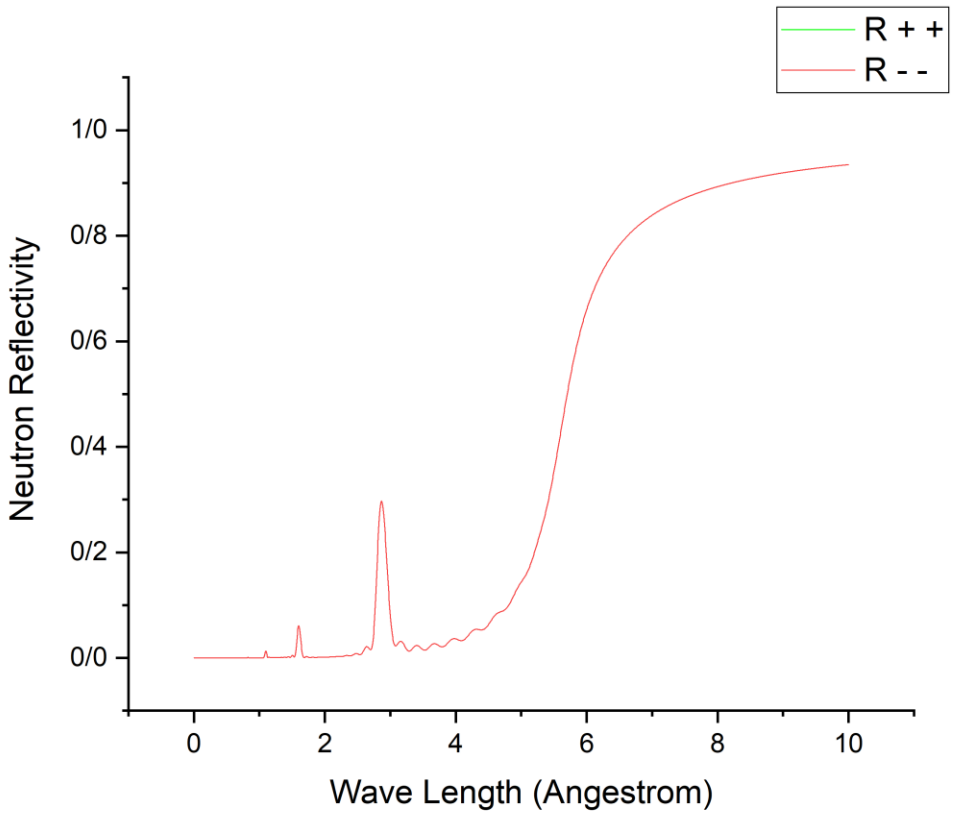


Figure No. 37 - calculation of neutron for [Nb(25nm) / Gd(3nm)] x10

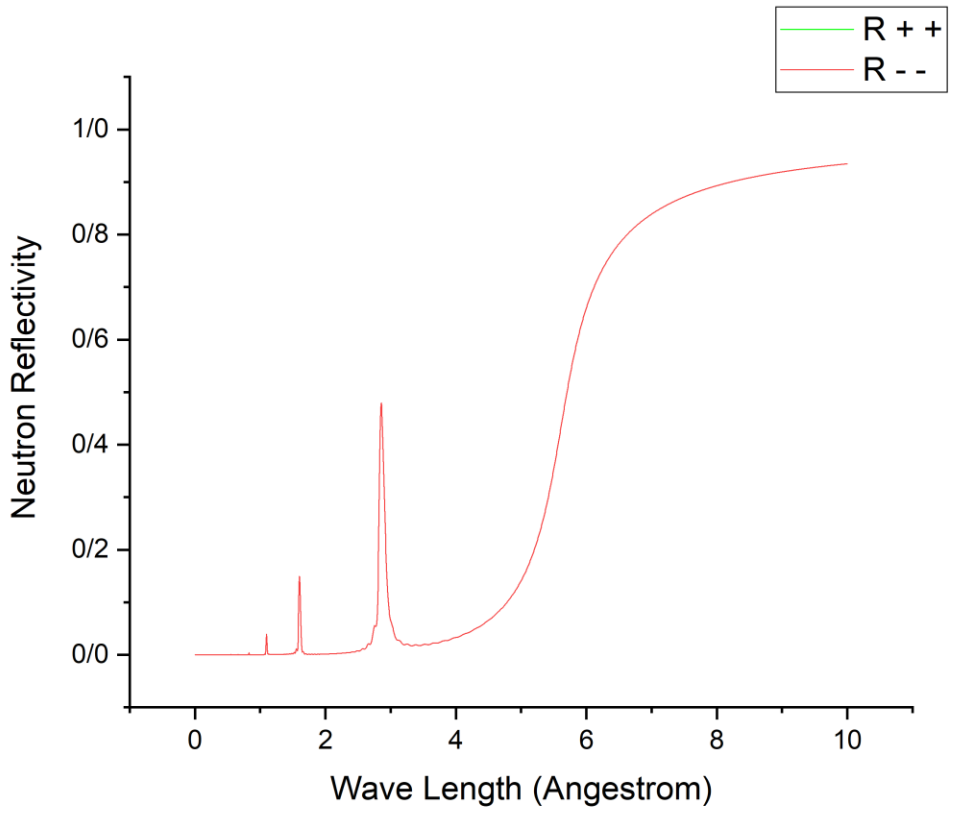


Figure No. 38 - calculation of neutron for [Nb(25nm) / Gd(3nm)] x20

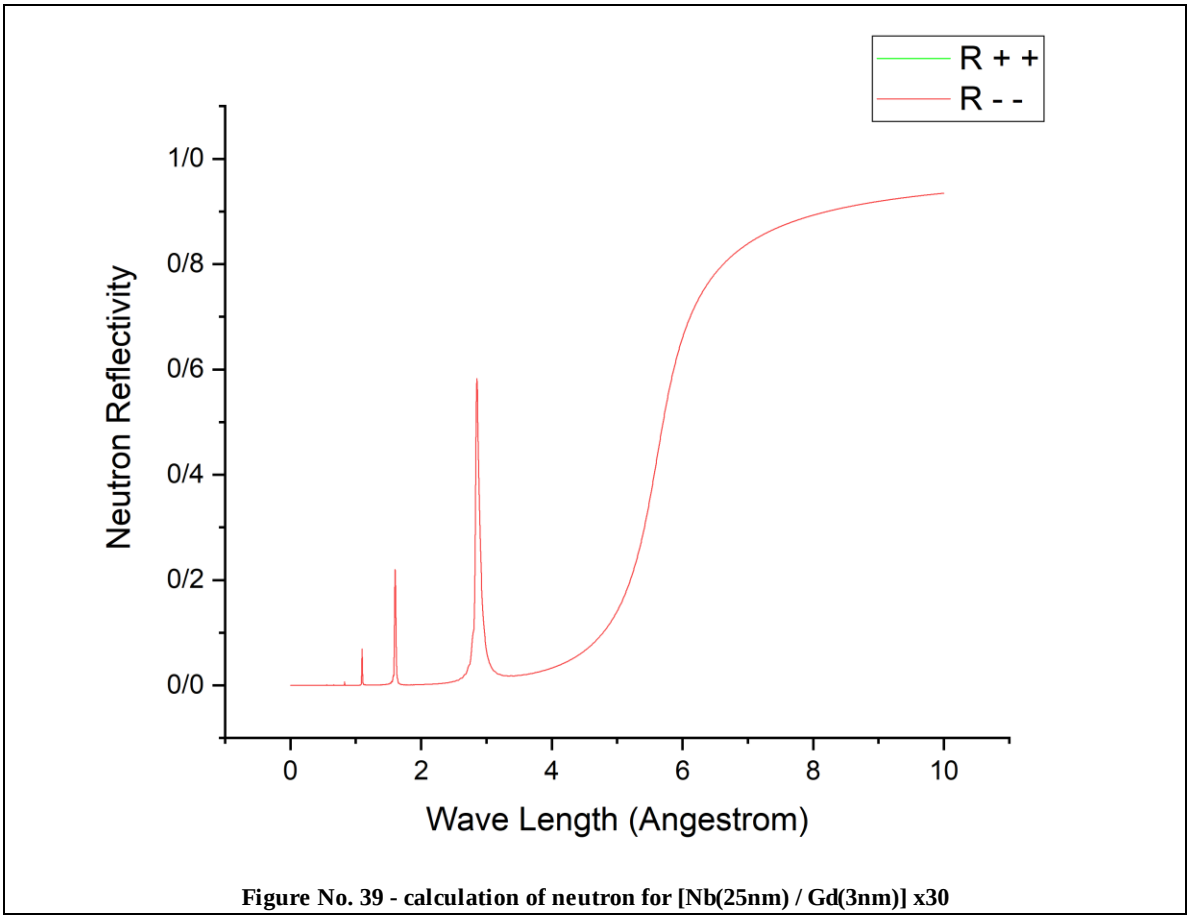


Figure No. 39 - calculation of neutron for [Nb(25nm) / Gd(3nm)] x30

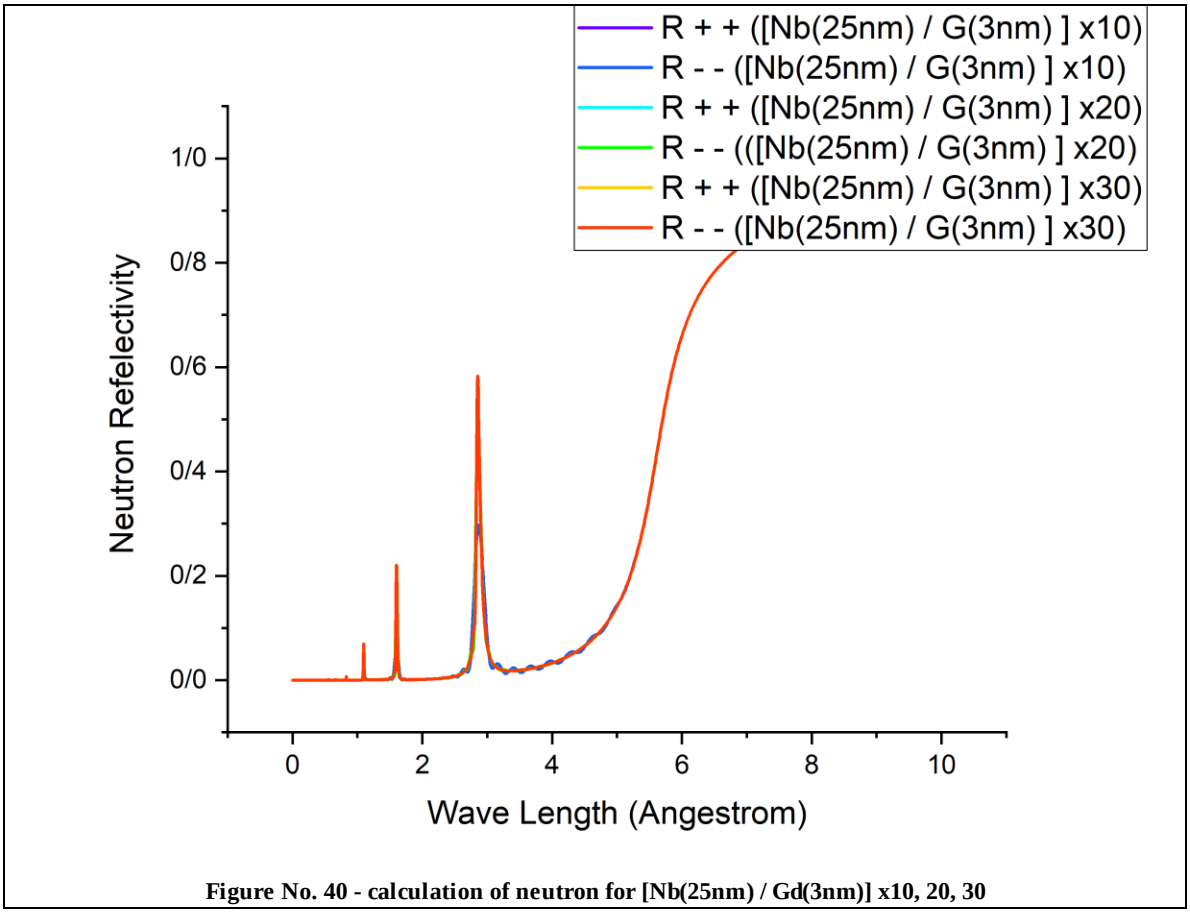


Figure No. 40 - calculation of neutron for [Nb(25nm) / Gd(3nm)] x10, 20, 30

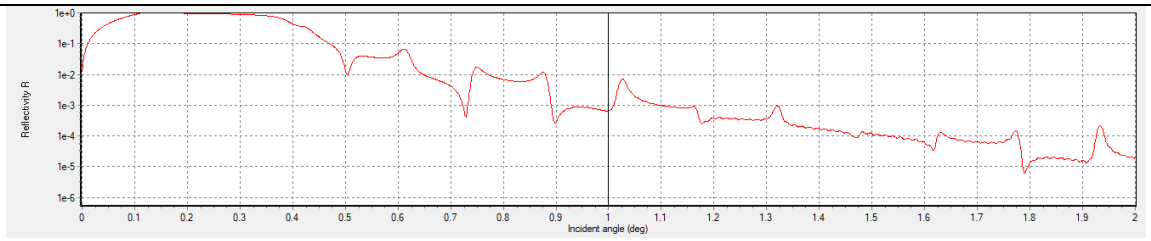


Figure No. 41 - X-ray reflectivity for [Nb(25nm) / Gd(3nm)] x10

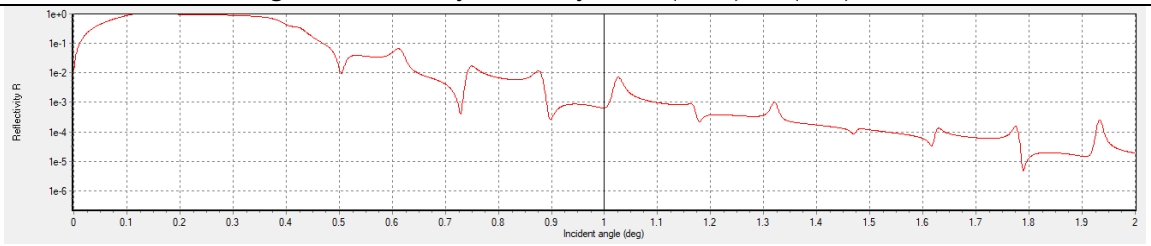


Figure No. 42 - X-ray reflectivity for [Nb(25nm) / Gd(3nm)] x20

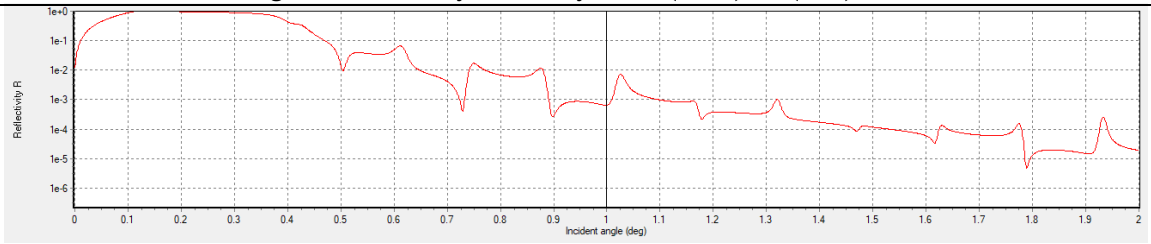


Figure No. 43 - X-ray reflectivity for [Nb(25nm) / Gd(3nm)] x30

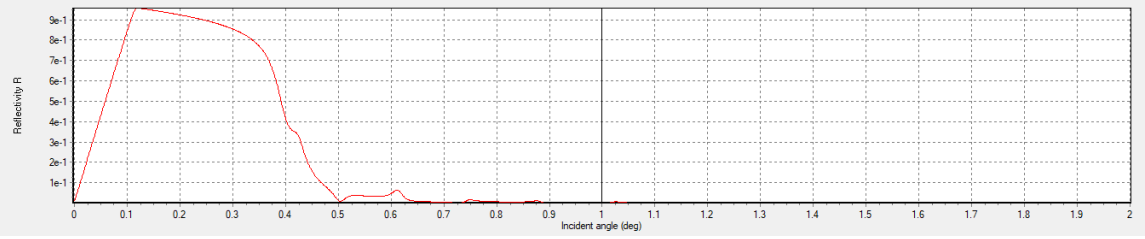


Figure No. 44 - Linear X-ray reflectivity for [Nb(25nm) / Gd(3nm)] x10

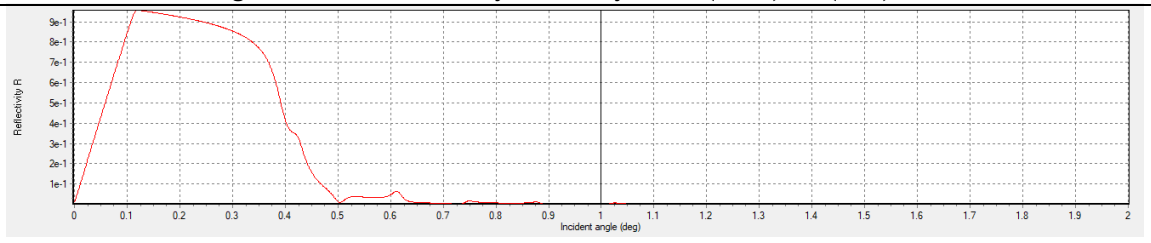


Figure No. 45 - Linear X-ray reflectivity for [Nb(25nm) / Gd(3nm)] x20

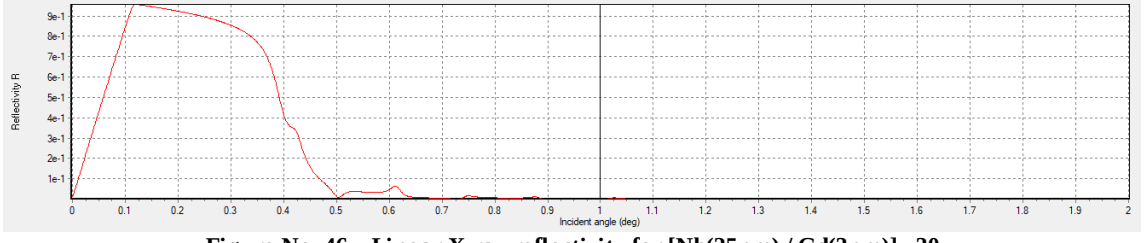


Figure No. 46 – Linear X-ray reflectivity for [Nb(25nm) / Gd(3nm)] x30

Conclusion:

According to the graphs No. 37 to 40, it can be concluded that the more the number of pairs of layers increases, the maximum peak value of the reflectivity also increases, so that for super lattice = 30, the maximum peak value is almost 2 times It is more than super lattice = 10. And also by looking at the graphs in the figures 41 to 46 it can be concluded that the intensity of X-rays has not changed, only to a very small extent in incident angel = 18 deg., the intensity of X-rays has decreased for super lattice = 30 Is and moreover, the X-ray diagrams become more smooth as we increase the repetition of Nb-Gd layer. In addition, in order to better display the neutron reflectivity peaks, graphs No. 44 to 46 have been drawn as linear graphs.

Task 7. Influence of roughness (Calculation only X-ray reflectivity)

Scope:

In this task, we intend to evaluate X-ray intensity changes due to reflectivity based on different roughness.

Method of Calculation:

Using X-ray reflectivity software and for the following model, we have evaluated the roughness of 0, 1, 2 and 3 for the gadolinium element (super lattice = 20).

- Influence of roughness by calculation only X-ray reflectivity for following data:

  Al2O3 / [Nb(25nm) / Gd(3nm)] x20 / Nb(15nm)
  And
  Roughness of Gd layers = 0, 1, 2, 3 nm

The figure of the results related to this problem are presented as follows:

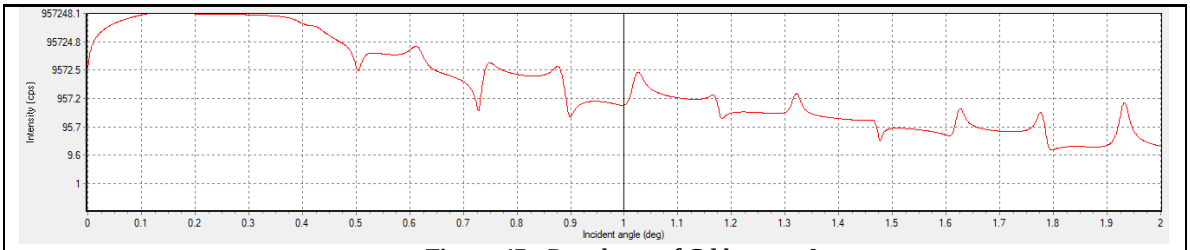


Figure 47 - *Roughness of Gd layers = 0, nm*

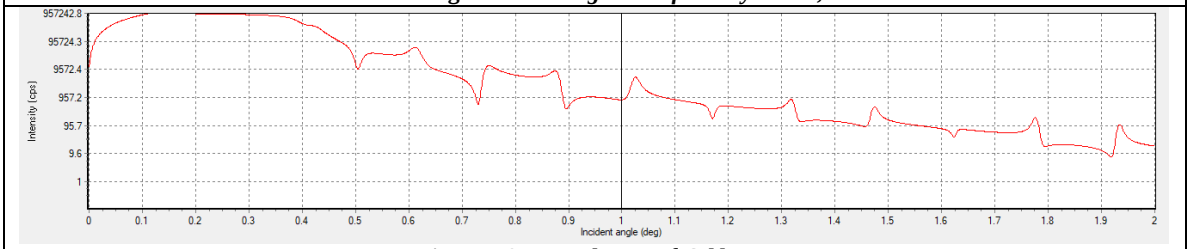


Figure 48 - Roughness of Gd layers = 1, nm

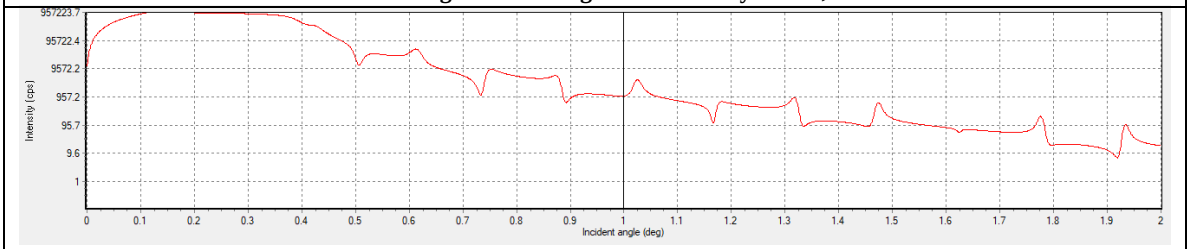


Figure 49 - Roughness of Gd layers = 2, nm

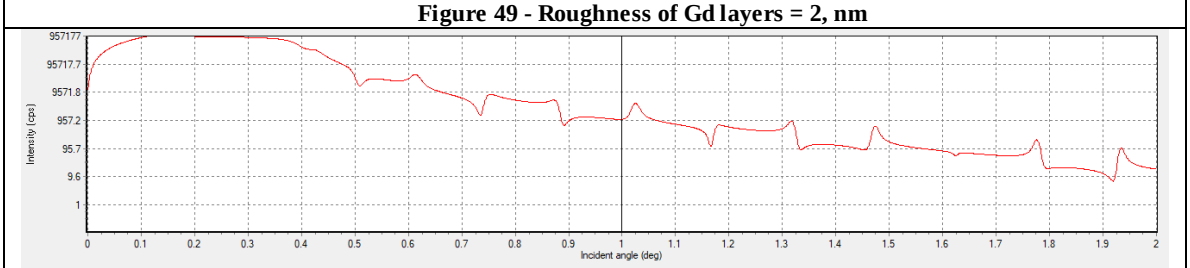


Figure 50 - Roughness of Gd layers = 3, nm

Conclusion:

By observing the graphs number 47 to 50, we can conclude that with the increase in roughness, the peaks and fluctuations of the graph related to the X-ray intensity for different incident angels decrease and the graph looks smoother.

Task 8. Structure with helicoidally magnetic (Calculation only of neutron reflectivity)

Scope:

In this task, we are going to evaluate the amount of reflectivity changes for the element with helicoidally magnetic properties (ie element Dy).

Method of Calculation:

Using the MATLAB code, we have divided the 3 nm thick plate related to the Dy element into 20 smaller layers (that is, 20*1.5=30), and continue we have drawn the diagram of neutron reflectivity changes for Mz=Mx=100, 1000, 10000.

- Structure with helicoidally magnetic by calculation only of neutron reflectivity for following data:

  Al2O3 / Nb(100nm) / Dy(3nm) / V(70nm) / Nb(15nm)
  And Separate Dy-layer to 20 sublayers, with Mz and Mx modulate helicoid

The figure of the results related to this problem are presented as follows:

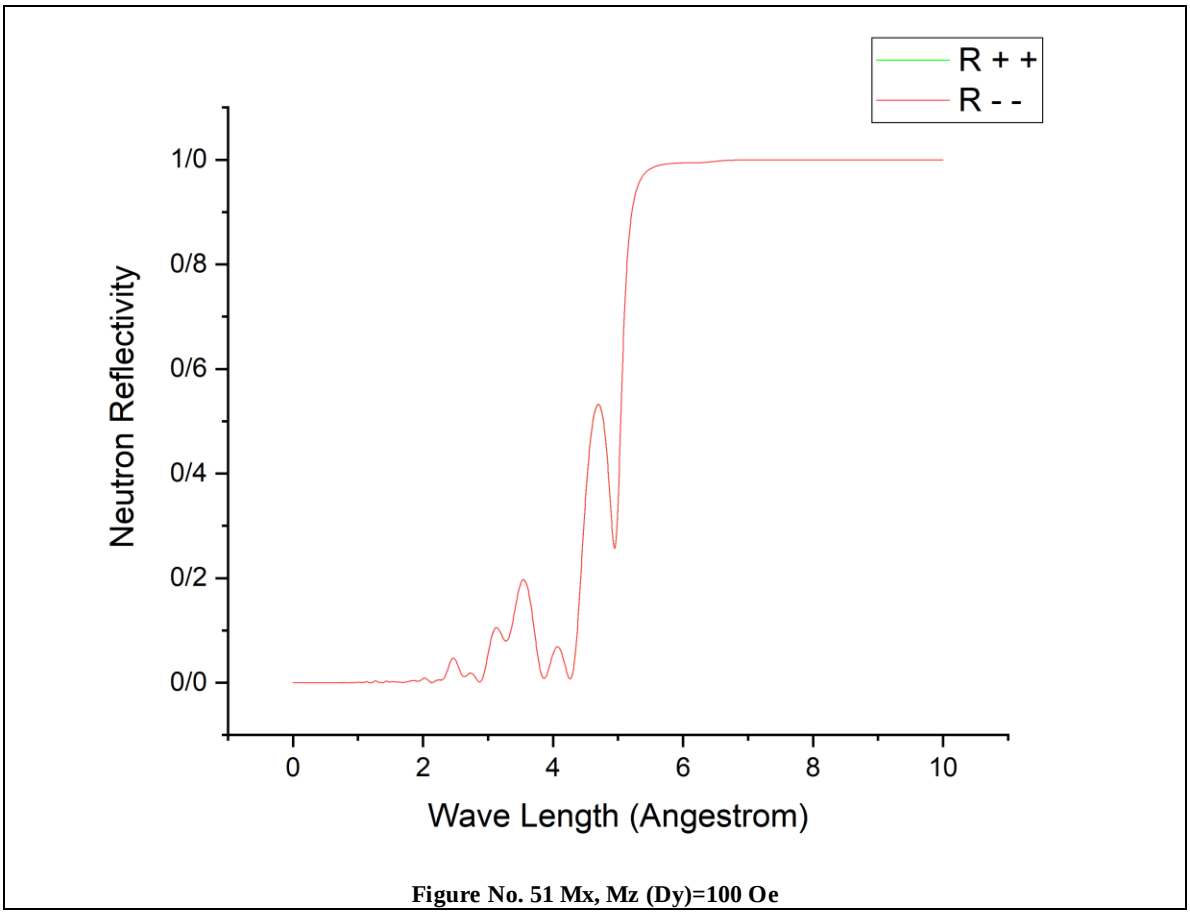


Figure No. 51 Mx, Mz (Dy)=100 Oe

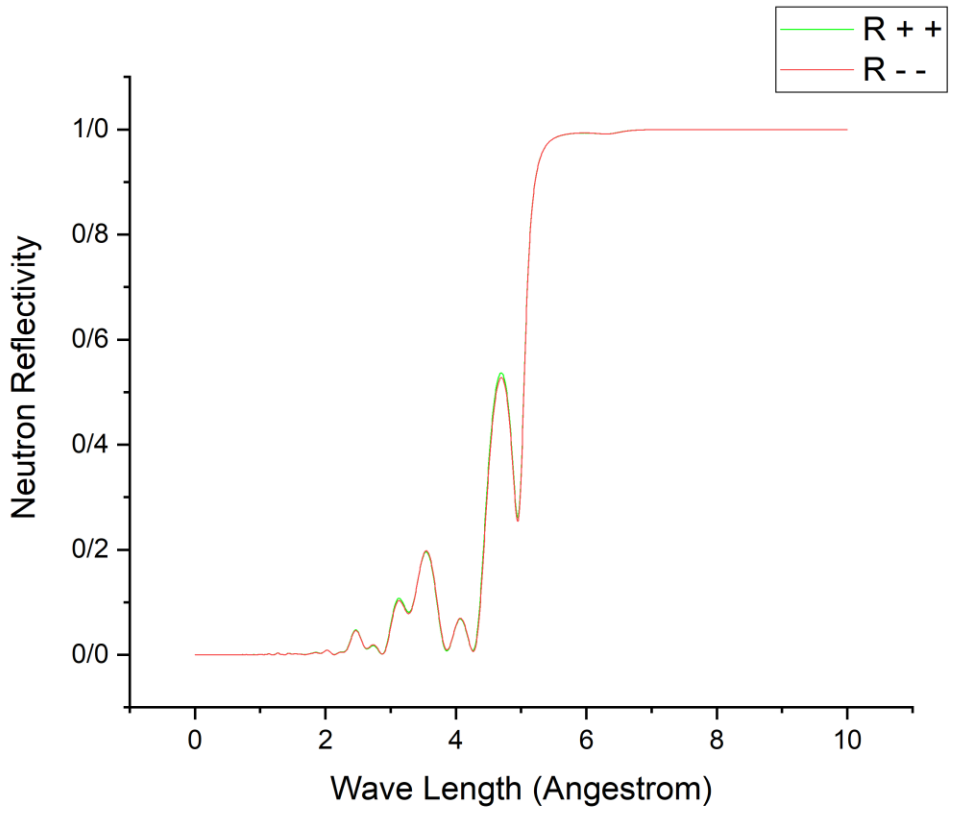


Figure No. 52 Mx, Mz (Dy)=1000 Oe

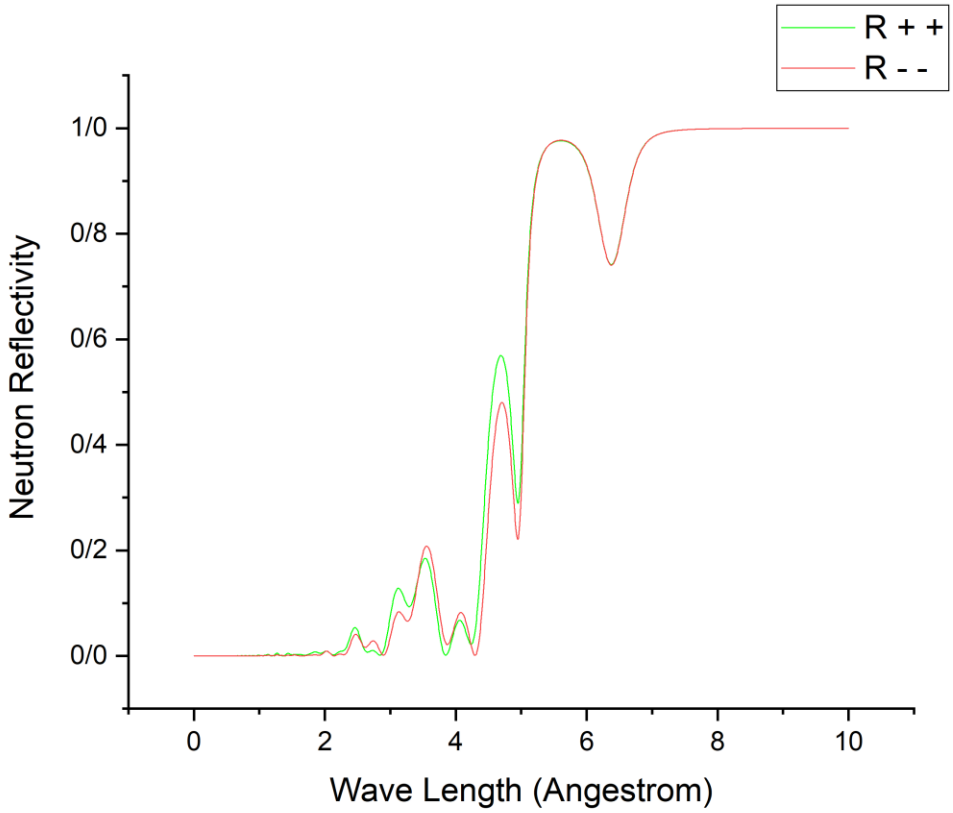


Figure No. 53 Mx, Mz (Dy)=10000 Oe

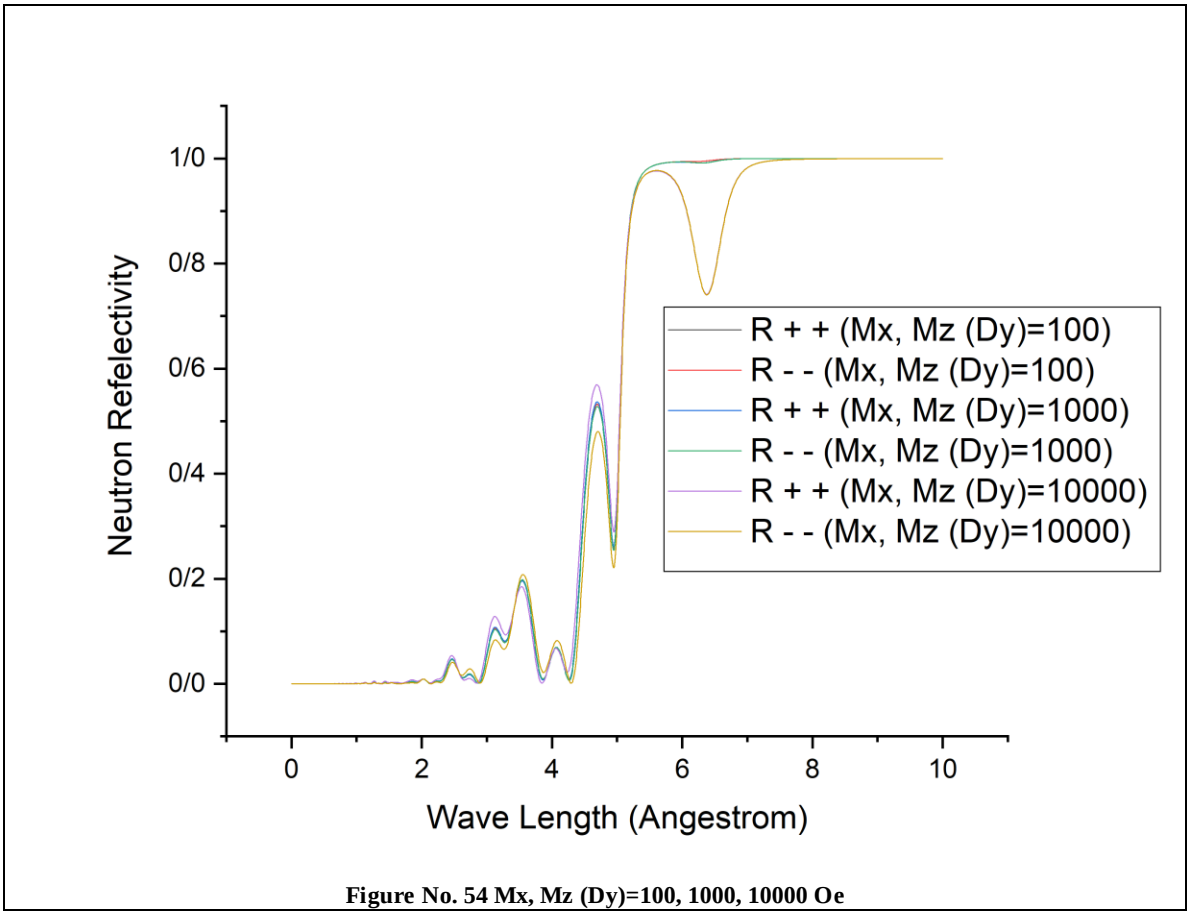


Figure No. 54 Mx, Mz (Dy)=100, 1000, 10000 Oe

Conclusion:

By looking at the figures No. 51 to 54 we can conclude that with the increase in the magnetic content of the Dy element, the neutrons are polarized and the green and red graphs (R + + and R - -) can be separated from each other. It is necessary to explain that, based on different sources (especially the article "Magnetic rare earth super lattices"), has been taken the functions Mx= MCosθ, Mz= MSinθ, are used to evaluate the changes.

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