JOINT INSTITUTE FOR NUCLEAR RESEARCH
Frank Laboratory for Neutron Physics

FINAL REPORT ON THE INTEREST PROGRAMME

Coexistence of Superconductivity and Ferromagnetism at Low-Dimensional Heterostructures

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Participation period:
February 14 – March 25, Wave 6

Dubna, 2022

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Abstract

By using the experimental method of Polarized neutron reflectometry we study the Coexistence of Superconductivity and Ferromagnetism at Low-Dimensional Heterostructures. We use Matlab to study the correlation between experimental and theoretical data associated with heterostructures containing superconducting and ferromagnetic layers for simulations of different nominal structures And also we researched changing of neutron and X-ray scattering properties from thickness, magnetization, and several layers using numerical methods. The proximity effect can be measured using neutron polarized reflectometry where scattered neutron beam intensity could be compared above and below the superconducting critical temperature to observe spin asymmetry existence. Tuning superconducting order parameter for S/F geometry has predicted the existence of the proximity effect and its inverse proximity effect. we conclude that there is a mutual influence of the ferromagnetic and superconducting layers in analyzed heterostructures.

1.Introduction:

1.1. Superconductivity and Ferromagnetism:

The superconductivity phenomenon was discovered in 1911 by Dutch physicist Heike Kamerlingh Onnes. Like ferromagnetism and atomic spectral lines, superconductivity is a phenomenon that can only be explained by quantum mechanics. It is characterized by the Meissner effect, the complete ejection of magnetic field lines from the interior of the superconductor during its transitions into the superconducting state. The occurrence of the Meissner effect indicates that superconductivity cannot be understood simply as the idealization of *perfect conductivity* in classical physics.

In 1986, it was discovered that some cuprate-perovskite ceramic materials have a critical temperature above 90 K (−183 °C). Such a high transition temperature is theoretically impossible for a conventional superconductor, leading the materials to be termed high-temperature superconductors. The cheaply available coolant liquid nitrogen boils at 77 K, and thus the existence of superconductivity at higher temperatures facilitates many experiments and applications that are less practical at lower temperatures.

Both superconductivity and superfluidity were some of the earliest examples of phase transition phenomena that happened due to the quantum nature of electrons due to the coherence of wavefunction of electrons in the ground state below their phase transition temperature (T.). Superconductivity and superfluidity transition were also early examples of new phases of matter discovered beyond thermodynamics phases of matter and ferromagnetic phases of matter which famously occurred below Curie's Temperature (Tc). In nature, there are only 4 materials that have Te beyond 273 K which are iron (1043 K), cobalt (1394 K), nickel (631 K), and Gadolinium (292 K).

Aside from zero resistance, superconductivity has perfect diamagnetism in the sense that there was no magnetic field inside the superconductor, and it expulsed external magnetic fields until certain field values (H). This property was named after its discoverer, the Meissner effect. Above H superconductivity vanished except for Type-II superconductor where above He superconductivity existed with supercurrent vortex where it pinned magnetic field until certain values.

Superconductivity vanished completely beyond it (He). This phase is called the Abrikosov phase, named after Russian scientist Alexei Abrikosov, who discovered it. All elemental superconductors have T, below 10 K such as Niobium (T-9.26K, H_{c} = 0.82T , type-II), Vanadium (T-5.03K, H_{e} = 1T , Type-II), and Lead (T.-7.19K, H = 0.08T , type II). The compound usually has a larger T. and H. and usually has type-II superconductivity such as Nb Ge (T_{c} = 23.2K, H_{c} = 37Gamma), MgB. (T-39K, H-74T), and Nb Sn (T.-K. H=30T).

All superconductors discovered until 1980 had a maximum T around 40K. After 1986 many superconductors with temperatures beyond 77K (boiling point of N_2) were discovered and were mainly cuprates-oxide-based superconductors. These superconductors were two-dimensional heterostructures and had perovskite lattice structures with orthorhombic geometry of the unit cell. The compound such as YBCO (T-95K). BSCCO (T-95K), and HgTiBaCaCaCuO (T=164K) are type two superconductors and are colloquially called High T. superconductors. In 21 century, iron-based superconductors were discovered such as FeSe (T-65K), SmFand eAs(O.F) (T.-55K). and CeFeAs(O.F) which was unprecedented due to incompatibility of superconductivity with ferromagnetism. All compounds mentioned in this paragraph are unconventional superconductors where microscopic phenomena which gave rise to superconductivity could not be described by BCS theory.

Microscopic theory BCS relied on the assumption that as the electron passes through crystal lattices it polarized the electron cloud in the lattice and in doing so it made lattice distortion which took time to relax. If a second electron passed the lattice before the distortion relaxed. then it would have lower energy because the lattice was polarized. As such, it would have attractive interaction between the two electrons forming the Cooper pair and such attractive force is called Froehlich interaction. Froehlich interaction is a phonon-mediated interaction between electrons with opposite spins which lowers the energy of the Cooper pair, and it has lower energy than the energy of two electrons combined, thus favoring pairing. The treatment of BCS theory would not be provided here. In superconductivity, the macroscopic explanation of zero resistivity and the Meissner effect can be invoked through à parameter known as supercurrent density which was derived from current density formulation in quantum mechanics,

$$\vec{J_s} = \frac{q^*}{2m^*}\left\{\Psi^*\left(\frac{\hbar}{i}\vec{\nabla} - \frac{q^*}{c}\vec{A}\right)\Psi + \left[\left(\frac{\hbar}{i}\vec{\nabla} - \frac{q^*}{c}\vec{A}\right)\Psi\right]\Psi^*\right\}$$

Where q = -2|el is a charge of the Cooper pair, m*= 2m, is the effective mass of the Cooper pair and A is vector potential which is added to the momentum operator due to gauge invariance of the momentum operator in the presence of the magnetic field. Ψ is a superconducting order parameter which is introduced by Russian scientists Vitaly Ginzburg and Lev Landau and took the form of

$$\Psi(\vec{r}) = |\Psi(\vec{r})|e^{i\theta(\vec{r})} = [n_s^*]^{\frac{1}{2}}e^{i\theta(\vec{r})}$$

Where n*s is the number of Cooper pairs in the sample which is easily in the order of 10^23. If we input this order parameter into supercurrent density and use some Algebra, the following supercurrent equation can be described.

$$\vec{J_s} = -\frac{\vec{A}}{\Lambda_s c} + \frac{\hbar}{q^*\Lambda_s}\vec{\nabla}\theta$$

Where A, = m*/(q*n*s). If we assume that Cooper pair density is independent of time, then time differentiation of the supercurrent equation would yield,

$$\vec{E} = \Lambda_s \frac{\partial \vec{J_s}}{\partial t}$$

Since m* is so small and n*s so large, then the value of current is nearly infinite. If we took the curl of the supercurrent equation and invoke a little bit of differentiation and maxwell equation,

$$\vec{\nabla} \times \vec{J_s} = -\frac{1}{\Lambda_s c}\vec{B}$$

Knowing, that $\vec{\nabla} \times \vec{H} = (4\pi/c)\vec{j}$ and $\vec{B} = \hat{\mu}\vec{H}$

$$\vec{\nabla} \times \vec{\nabla} \times \vec{B} = \vec{\nabla}(\vec{\nabla} \cdot \vec{B}) - \nabla^2\vec{B}$$

Since $(\vec{\nabla} \cdot \vec{B}) = 0$

$$\nabla^2\vec{B} = \frac{1}{\lambda_s^2}\vec{B}$$

Where λs, is the superconducting penetration depth and it has the values of

$$\lambda_s = \left(\frac{\Lambda_s c^2}{4\pi\hat{\mu}}\right)^{\frac{1}{2}} = \left(\frac{m^* c}{4\pi\vec{\nabla} q^* n_s^*}\right)^{\frac{1}{2}}$$

The above equation that relates zero resistivity and superconducting penetration depth is called as first and second London equation. The solution for the second London equation can be written as

$$B(z) = B(0)e^{\left(-\frac{z}{\lambda_s}\right)}$$

This manifests that the magnetic field is suppressed inside the superconductor, at least not until penetration depth which has ordered in several dozens or hundreds of nm. For this reason, it was thought until the early 1980s that superconductivity would not happen if it was composed of magnetic materials, hence the discovery of iron-based superconductors and the superconductivity-ferromagnetism proximity effect initially baffled many scientists. Further derivations of the London equation would give coherence length or the Cooper pair correlation length in absence of any scattering. In short, electrons in any distance below this length could interact as pair.

$$\xi_o = \frac{h v_F}{\pi \Delta_o}$$

Where v_F is Fermo velocity and $\Delta_o = m^* v \ \delta v$

Ferromagnetism:

Ferromagnetism is a physical phenomenon in which certain electrically uncharged materials strongly attract others. Two materials found in nature, lodestone (or magnetite, an oxide of iron, Fe_3O_4) and iron, can acquire such attractive powers, and they are often called natural ferromagnets. They were discovered more than 2,000 years ago, and all early scientific studies of magnetism were conducted on these materials. Today, ferromagnetic materials are used in a wide variety of devices essential to everyday life—*e.g.,* electric motors. Ferromagnetism is a kind of magnetism that is associated with iron, cobalt, nickel, and some alloys or compounds containing one or more of these elements. It also occurs in gadolinium and a few other rare-earth elements. In contrast to other substances, ferromagnetic materials are magnetized easily, and in strong magnetic fields, magnetization approaches a definite limit called saturation. When a field is applied and then removed, the magnetization does not return to its original value—this phenomenon is referred to as hysteresis (*q.v.*). When heated to a certain temperature called the Curie point (*q.v.*), which is different for each substance, ferromagnetic materials lose their characteristic properties and cease to be magnetic; however, they become ferromagnetic again on cooling. The magnetism in ferromagnetic materials is caused by the alignment patterns of their constituent atoms, which act as elementary electromagnets. Ferromagnetism is explained by

the concept that some species of atoms possess a magnetic moment—that is, that such an atom itself is an elementary electromagnet produced by the motion of electrons about its nucleus and by the spin of its electrons on their axes. Below the Curie point, atoms that behave as tiny magnets in ferromagnetic materials spontaneously align themselves. They become oriented in the same direction so that their magnetic fields reinforce each other. Lastly,
Superconductivity is a very widely studied phenomenon in condensed most matte
physics both on the scale of theoretical research and industrial applications.
Meanwhile, ferromagnetism is the alignment of the magnetic moments of a material's unpaired electrons producing a net magnetic moment for the material.
ferromagnetism is the reason for the existence of so-called permanent magnets
Proximity Effect and Inverse Proximity in Superconductor-Ferromagnet

As previously mentioned, ferromagnetism could destroy superconductivity due to the existence of the external field, as such for a long time it was thought that ferromagnets were not compatible to become superconductors. Vitaly Ginzburg formulated this problem which explained the problem and concluded that ferromagnet would suppress superconductivity. In Frohlich interaction, minimization of energy in superconductors happened because the interaction of an electron with momentum k_1 gave rise to lattice polarization (phonon) with momentum k and it interacted with the second electron with momentum K_2 and its respective phonon with momentum K2. The interaction is such that,

$k_1 + k_2 = k_{12} + k_2 = 0$

From the equation above, it was clear that interaction only occurs if and only if the electron occurred to have opposite momentum. In a broader term, interaction in cooper pair is maximized if the electron occurred to have opposite total momentum and hence opposite spin. After the invention of the BCS theory, it was firmly believed that in a conventional superconductor, the exchange interaction of ferromagnet would align the spin of the cooper pair and as such decrease the coherence of superconducting states by increasing the pairing energy inside the superconductor. However, Paul Anderson displayed that ferromagnetism would not arise inside a superconductor because spin alignment happened in the short range, and it mainly happened for ion-core and the effect was negated by long-range interaction of coherence length. Instead, Anderson argued that non-ordered magnetic would appear in superconductors and this new state was called cryptoferromagnet. In unconventional superconductors such as UGe, ferromagnetism is observed as bulk properties of the sample. This occurred due to the pairing of three electrons forming triplet pairs and such could not be treated using BCS theory.

Following the definition of superconductor where the material has zero electric resistance and expelling external magnetic field, it was easy to explain why ferromagnetism would not arise in superconductivity. The Meissner effect prevents the field to enter and affect the Cooper pair. However as mentioned before, the magnetic field could enter the superconductor and caused the existence of the superconductor vortex. Cooper pair would not be easily affected by external magnetic field if the penetration depth of magnetic field (A) is smaller than superconductivity coherence length (50). This assumption was true for bulk superconductors but in 2-D heterostructures consisting of a ferromagnet (F) and Superconductors (S) layers, cooper pair of superconducting layers could penetrate in a damped situation in a ferromagnet (proximity effect) and vice versa where ferromagnetic ordering happened inside the superconductor (inverse proximity effect). Both effects happened due to the small dimensional thickness of the layer compared to superconductor penetration depth (2) as well as coherence length (5), as a result, a magnetic field could penetrate well inside the superconductor and vice versa, electrons in superconductors could perturb and affect electron in ferromagnetic layer and gave rise to pairing in ferromagnet layer. There are many effects related to this phenomenon like spatial oscillations of the density of states of the electron, and a nonmonotonic dependence of the critical temperature of the S/F layer/multilayers.

In a zero magnetic field, an electron paired in Cooper pair is formed by two electrons with opposite momenta. However, in the presence of the magnetic field, Zeeman splitting occurs, and the momentum of the electron would shift by the value of +8k = µgH/VF and as such we would get k_2 + k_2 = 28k # 0. Cooper pair in this state is called FFLO state named after Fulde, Ferrel, Larkin, Ovchinnikov. The treatment of the FFLO state is performed using Ginzburg Landau order parameter functional, which is tuned to the geometry of the system and very difficult to be parameterized. For a very simple S/F layer the functional took the form of

$$a\psi - \gamma\frac{\partial^2\psi}{\partial^2 x} + \frac{\eta}{2}\frac{\partial^4\psi}{\partial x^4} = 0$$

The solution for the above equation is oscillating order parameter =, exp(ax) with a complex wave vector k = k_1+ik_2 where k_1 and k_2 are momenta for each electron. If we assume that the order parameter in the superconductor is real, then the order parameter of the ferromagnet is also real,

$$\psi(x) = \psi_i \exp(-k_1 x)\cos(k_2 x)$$

The above equation showed that superconductor order perimeter exists in ferromagnet albeit in an oscillating and damped form. Nonetheless, FFLO states showed that the proximity effect could occur in the S/F layer. Since order parameters are correlated with the density of states. this equation also showed how the superconductivity density of states exists and oscillates. inside ferromagnet. A superconductor, which is in contact with ferromagnet, received magnetization with the value of

$$\delta M = 2\mu_B N_0 \pi k_B T \sum_{n>0} \boldsymbol{g}(\boldsymbol{x}, \boldsymbol{\omega_n})$$

Where g is normal Green function over x and Matsubara frequency.

1.2. Proximity effects in superconductor-ferromagnet structures

When there are two different materials next to each other due to close contact they can affect each other. So the phenomena which originate from them (in the case of this research ferromagnetism and superconductivity) can affect each other, mix or coexist. This effect includes all interactions on the interface between two materials and in most cases, it is a consequence of the exchanging of electrons between two materials. Due to the superconductivity of one of these layers, we can call these pairs of electrons Cooper pairs.
On the surface (ie it can be said that in the contact area due to inhomogeneity of these structures) of the ferromagnetic and superconducting layer there is a mutual influence of ferromagnetism and superconductivity caused by the finite value of characteristic coherent length and London penetration depth. Change of the density of the superconductive electrons isn't fast, so there is a minimum length on which that change can occur without destroying of superconductive state and it is called coherence length. The second fundamental length is related to the characteristic distance in which the magnetic field can penetrate into the superconducting region – London penetration depth λ_L. I in the case of mutual interaction of ferromagnetism and superconductivity, two effects are defined: the proximity effect refers to the influence of the superconductivity on ferromagnetism, and the inverse proximity effect refers to the opposite affection of ferromagnetism on superconductivity. Both effects are the consequence of the interaction of free electrons in ferromagnetic material and conduction electrons in the superconducting layer close to the contact interface. These electrons are exchange-coupled. Occurred Cooper pairs of electrons induce superconductivity in a ferromagnetic material by penetration in it. In the exchange field, a ferromagnet the up-spin electron decreases its energy by *h*, while the down-spin electron energy

increases by the same value. To compensate this energy variation, the up-spin electron increases its kinetic energy, while the down-spin electron decreases its kinetic energy. Analyzed structures in this research are of nanometer dimensions and the F layers are several nanometers thick because the interplay between S and F phenomena occur at a nanometer scale. Also, in general, ferromagnetism is much "stronger" than superconductivity so it could destroy superconductivity by an exchange mechanism. But depending on conditions and temperature, both superconductivity and ferromagnetism can destroy one another. In the ferromagnetic state, the exchange field doesn't let electrons pair by aligning all Cooper pair spins in the same direction. This effect is called the paramagnetic effect. In addition to this effect, many different effects can occur in S/F systems. One of them is the cryptoferromagnetic effect which refers to the formation of nonuniform domain structure in the S layer. The coherence length is larger than the period of the domain ferromagnetic structure but the period is larger than the interatomic distance. It can be concluded that the period of occurrence of these domains is really small. The existence of the cryptomagnetic state is confirmed by showing that there occurred a decrease in the average magnetization in the F layer. One can say that domain structural organization is the consequence of the coexistence of ferromagnetism and superconductivity inside one sam layer. It can be said that from the magnetic side it looks like ferromagnetism but from the superconductivity side like antiferromagnetism. Besides these effects many other can occur in the S/F structures, some of them are damped oscillatory behavior of the Cooper pair wave function in the ferromagnet, spatial oscillations of the electron density of states, a spontaneous vortex phase, π Josephson junctions in S/F/S systems, a nonmonotonic dependence of the T_c of S/F multilayers and bilayers on the ferromagnet layer thickness, and triplet superconductivity. Maybe the most important of them is the damped oscillatory behavior of the Cooper pair wave function in the ferromagnet which results in a nonmonotonic dependence of the T_c of S/F multilayers on the thickness of the ferromagnetic layer, as well as in the formation of π junctions in S/F/S interfaces.
It should be noted that the length of the interface of the F and S layers in investigated structures is comparable to the coherent lengths of superconductivity and ferromagnetism. Close to this interface occurs changing of properties of both layers. For the experimental study of these properties, electrical resistance and magnetic field (behaving of magnetic moments) are usually measured. But due to changes in the properties, spatial magnetic and superconducting profiles must be determined. They can be successfully measured by polarized neutron reflectometry

1.3. Neutron reflectometry:

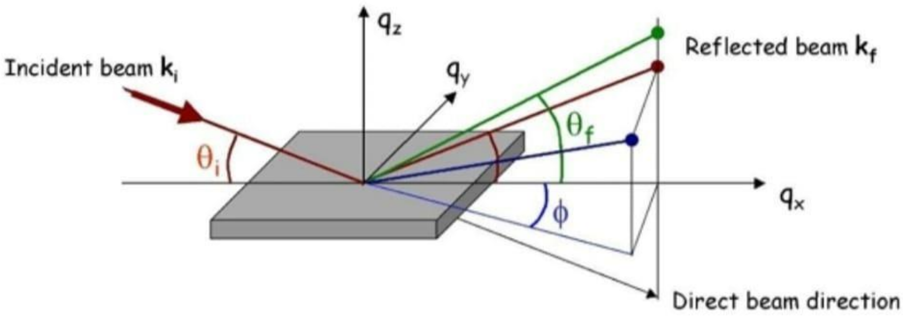
Neutrons are noncharged particles of big mass and they have spin. They can interact with nuclei and penetrate deep into the sample. Due to their spin, they can interact with magnetic dipoles in the material so it is possible to obtain a spatial magnetic profile of the analyzed sample.

Neutron reflectometry is a technique for determining the structure of thin films based on neutron diffraction on the nuclei of the atoms in the analyzed structure. The sample is irradiated with a highly collimated neutron beam which reflects on it. The intensity of the reflected beam is being measured as a function of the angle of a wavelength of neutrons. From this dependence, it is possible to get information about the thickness, density, and roughness of the surface of the sample. Neutrons are especially suitable for use in analyzing the S/F structures due to that that thermal neutron wavelength corresponds to the characteristic coherent length and London penetration depth in superconductors and ferromagnets. Also, spatial profiles can be easily obtained due to the high penetration power of neutrons. Wavelengths of used neutrons for obtaining analyzed results are between 1 and 20 Å.

Measurement can be performed in specular and off-specular modes. In specular mode, the angle of the incident beam is equal to the angle of the reflected beam. Neutron reflection is described in terms of a momentum transfer vector Q:

$$Q = \frac{4\pi}{\lambda}\sin(\theta_i), \qquad \text{- (1)}$$

where λ is the neutron wavelength and θi the angle of the incident beam. The momentum transfer vector denotes the change of neutron momentum after reflection on the sample ($\boldsymbol{Q} = \boldsymbol{ki} - \boldsymbol{kf}$). Off-specular reflectometry gives rise to diffuse scattering and involves momentum transfer within the layer and is used to determine lateral correlations within the layers, such as those arising from magnetic domains or in-plane correlated roughness.



Scheme of the neutron reflection on the sample

Standard polarized neutron reflectometry is used for determining the spatial magnetic profile. Neutrons can have two different polarizations in a magnetic field. The positive one is the direction of the applied magnetic field and the negative one is in a direction opposite to the magnetic field. Due to the different interactions of these neutrons with nuclei of the atoms in the heterostructure and the field, spin asymmetry and neutron polarizability can occur. So we can say that there are two states of neutrons, based on which there can be four different reflectivities:

$$r = \begin{pmatrix} r_{++} & r_{+-} \\ r_{-+} & r_{--} \end{pmatrix}. \qquad \text{- (2)}$$

Magnetic moments of neutrons can be aligned in the direction of the magnetic field (+ state) and the opposite direction (-state).

Due to differences in values of reflectivities from Eq. 2., spin asymmetry can occur and it can be calculated by the following equation. The polarization of neutrons can be changed by spin flippers in the neutron spectrometer before and after the reflection on the sample.

$$Spin\ asymmetry = \frac{r_{++} - r_{--}}{r_{++} + r_{--}}. \qquad \text{-(3)}$$

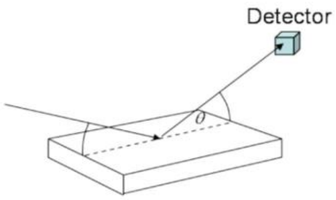
In the heterostructures and on the interface between its layers, the interaction potential is the sum of the interaction potentials of all elements close to the interface which penetrate from one layer to another. So, it is not possible to obtain just by standard neutron reflectometry a good magnetic and elemental profile of the structure. For this, it is necessary to obtain the potential profile of the interaction of neutrons with all elements in the sample which is possible by registering secondary radiation too. In the neutron reflectivity experiment, the absorption can take place so we can say that sum of reflectivity and transmission is less than 1 so it is possible to analyze emitted particles or beams due to occurring absorption. It can give us additional information about the structure, already mentioned earlier.

Different types of radiation can be registered, but the most common are charged particles, gamma quanta, and fission fragments which give more detailed information about the elemental/isotopic profile of the interface. Simultaneous registration of reflected neutrons and secondary radiation makes it possible to determine the good spatial profiles of isotopes in the examined material.

In S/F heterostructures, there is a modification of the magnetic and superconducting properties which affects the change in the spatial magnetic profile and distribution of the magnetization in the material so the neutron reflectivity is a powerful technique for determining these changes and is very useful for explaining the impact of these phenomena one to each other.

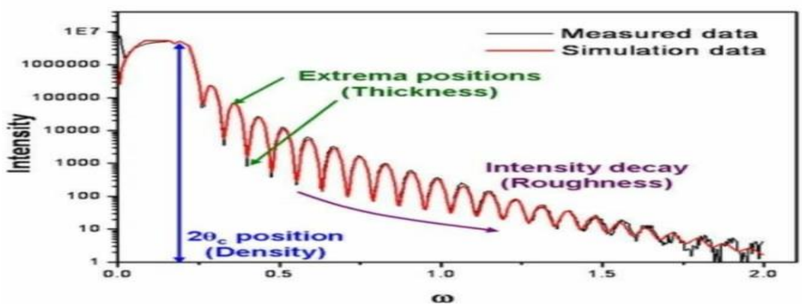
1.4. X-ray reflectivity:

X-ray reflectivity is based on measuring the reflection of an X-ray beam from a surface in a specular direction which means that the angle of the reflected beam is equal to the angle of the incident beam. The surface of the material can be perfectly flat but if it isn't the reflection will deviate from that predicted by Fresnel's law of reflectivity. Due to this unevenness, analyzing the deviations, a surface density profile can be obtained.



X-ray reflectivity

Below the critical angle of total external reflection, X-rays penetrate only a few nanometers into the sample. Above this angle, the penetration depth increases rapidly. At every interface where the electron density changes, a part of the X-ray beam is reflected. The interference of these partially reflected X-ray beams creates the oscillation pattern observed in reflectivity experiments. From reflectivity curves, one can determine layer parameters such as thickness and surface density gradients and layer density, interface, and surface roughness



Typical XRR pattern of a thin film on a substrate. The density, thickness, and roughness of the film are obtained from the critical angle, period of oscillations, and intensity decay, respectively

Measured reflectivities of neutrons and X-ray beams are presented below. The investigated initial structure is Al2O3/Nb(100nm)/Gd(3nm)/V(70nm)/Nb(15nm), where Al2O3 is a substrate. There is a heterostructure of the S/F/S sandwich, where superconducting materials are niobium and vanadium between which is the ferromagnetic layer of gadolinium with a thickness of 3nm. Due to the mutual impact of superconductivity and ferromagnetism, both superconducting layers (V and Nb) are divided into two sublayers. Because ferromagnetism from the Gd layer effects and changes magnetization profiles in both superconducting layers. Following is a discussion on the influence of different grazing angles of the neutron beam, collinear and non-collinear magnetization, ferromagnetic layer thickness, different ferromagnetic materials, superlattice, and influence of roughness on the neutron and X-ray reflectivity profiles.

Methods and Goals:

The main purpose of this project was to study and determine X-Ray Reflectivity and Neutron Reflectivity using both experimental results and simulation from heterostructures that were composed of niobium (Nb). gadolinium (Gd), Vanadium (V), sapphire substrate (ALO), and other elements Main tasks are

Processing of raw-data spectra with Spectra Viewer software, Data fitting with the physical model by Matlab software, and Calculation of modeling reflectivity curve depending on different parameters. In this project, there were several software which was utilized to simulate and calculate numerical value. To open and extract numerical data of the neutron reflectometry experiment, Spectra Software was used. X'Pert Reflectivity from PAN analytical was being used to simulate X-Ray reflectivity from heterostructures layers. In addition to that, numerical calculation and simulation of neutron reflectivity were performed using MatLab using Lemur.m file provided by Dr. Zhaketov. Finally. plotting and several calculations were carried out using OriginLab.

2. Tasks :

2.1. Task 1

In the first task, experimental data of two neutron reflectometry experiments (T=1 (above T.) and T=1.5K(below T c)) was extracted from Spectra Software and its numerical. value (both during spin flipper on (-) and off (+)) was exported to Origin. Then, the reflectivity of the samples could be determined by knowing that R + =I + /I dir and R - =I - /I dir where R denotes reflectivity for flipper on (-) and flipper off (+) condition. L dir was reference spectra or Maxwell spectra from the experiment. After that, it was imperative to calculate spin asymmetry from the experimental data using the equation SA=(R + -R -)/(R + +R -). Where SA symbolizes spin asymmetry for each wavelength. The wavelength of the experimental data can be determined by the equation below,

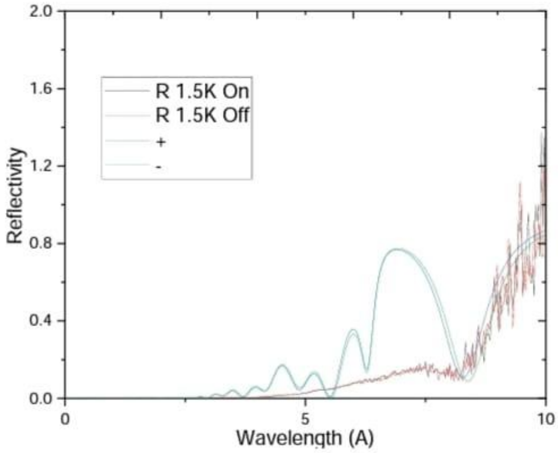
$$\lambda[\text{Å}] = \frac{(N + 35 - 12) * 3.956 * 256}{34030}.$$

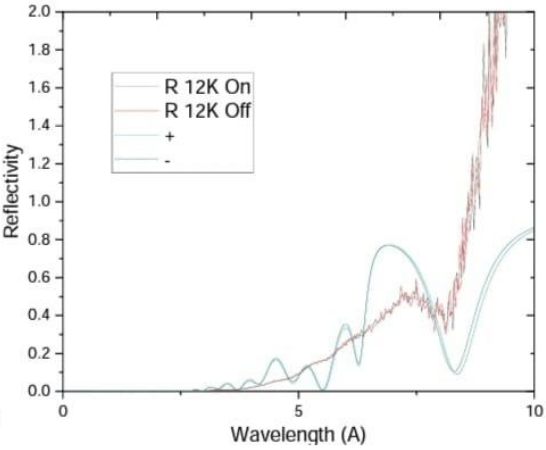
After performing calculation and plotting of experimental data, neutron reflectivity data were compared with simulation using the lemur program in Matlab. The simulation was because below Tc superconducting layers close to the ferromagnet layer were magnetized and at the same time ferromagnetism in the ferromagnetic layer was suppressed due to the proximity effect while above

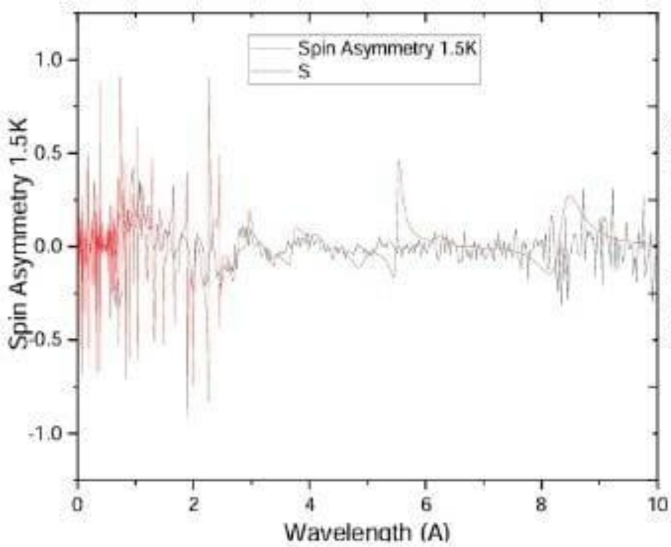
Tc ferromagnetism was not suppressed, and the superconducting layer was not magnetized. Reflectivity data (flipper on (-) and flipper off (+) condition) from simulation and their respective spin asymmetry were compared with experimental data. The fitting was carried out by determining the correct layer number and magnetization value that occurred inside the layers. In the simulation, the following layers and 7.6mrad grazing angle of neutron were the basis for simulation.

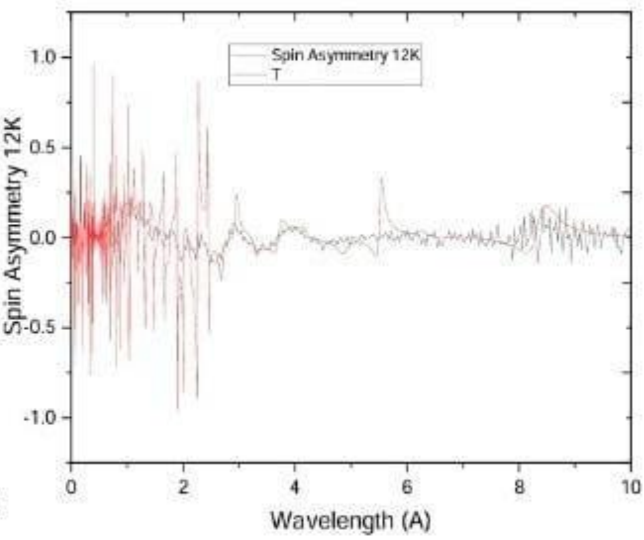
Al2O3/Nb/Gd/V/Nb

Result,





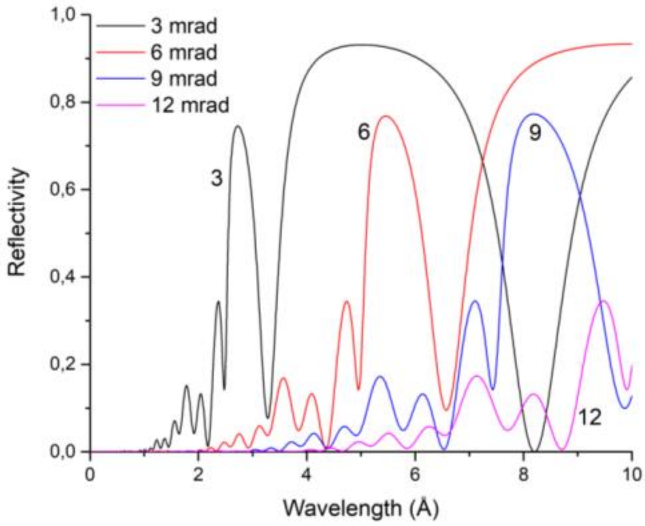




The above layers were simulated by assuming that in superconducting temperature, there is ferromagnetism in V and Nb layers with proximity layers with the width of 100 Angstrom, while in the non-superconducting state, there were no additional layers of V and Nb and it was assumed heat magnetization of Gd at that temperature was around 3000 Oe. The difference between experimental data and simulation happened due to systematic error in simulation. hampered by insufficient information on the initial value of Nb and V thickness. Nonetheless, spin asymmetry of experimental result and simulation closely matched and due to differing asymmetry values, in either case, it was assumed that spin asymmetry value arises in superconductivity layers due to ferromagnetic suppression of Gd layers.

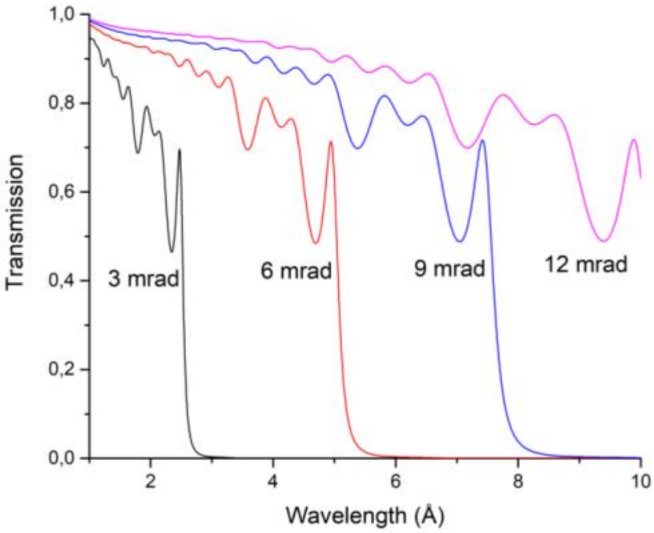
2.2. Task 2

Comparing reflectivity at different grazing angles (calculation only neutron reflectivity) First, we analyze the influence of neutron beam grazing angle on the neutron reflectivity on Al2O3/Nb(100nm)/Gd(3nm)/V(70nm)/Nb(15nm) heterostructure. Chosen grazing angles are 3, 6, 9, and 12 mrad.



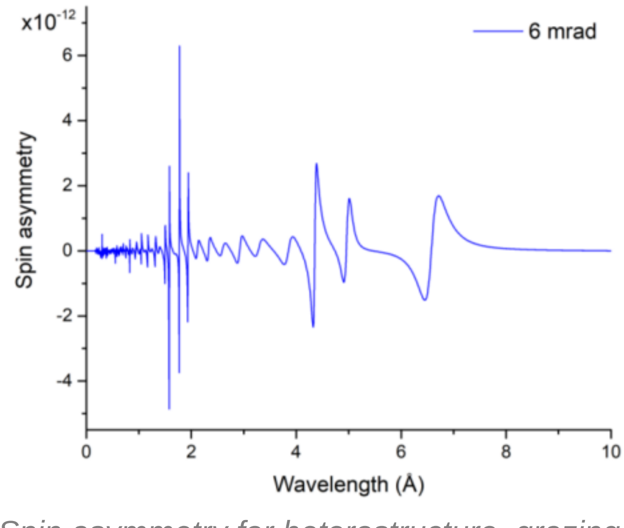
The reflectivity of Al2O3/Nb(100nm)/Gd(3nm)/V(70nm)/Nb(15nm) heterostructure for grazing angles of 3, 6, 9, and 12 mrad.

Due to reflection, neutrons change their momentum which depends on the incident angle of the neutron beam, Eq. 1. So with the bigger angles, there is a smaller wavelength of reflected neutrons so the reflectivity spectra are moving to the higher values of the neutron wavelengths as can be seen in the above figure. We can see that with the increase of the grazing angle, the intensity of reflectivity decreases, pink line in the above figure corresponds to the grazing angle of 12 mrad. The same trend occurs for the transmission spectra, which are given in the below Figure.



Transmission of Al2O3/Nb(100nm)/Gd(3nm)/V(70nm)/Nb(15nm) heterostructure for grazing angles of 3, 6, 9, 12 mrad.

In the 1st figure, it can be seen just one reflectivity curve for all samples because R++ and R−− are almost the same. There is no spin asymmetry because there is no magnetization in the system and so R−+ and R+− are equal to zero. The below figure shows spin asymmetry for the case of 6 mrad grazing angle. For all analyzed grazing angles spin asymmetry is of 10-12 orders of magnitudes so it can be concluded that it is negligibly small.

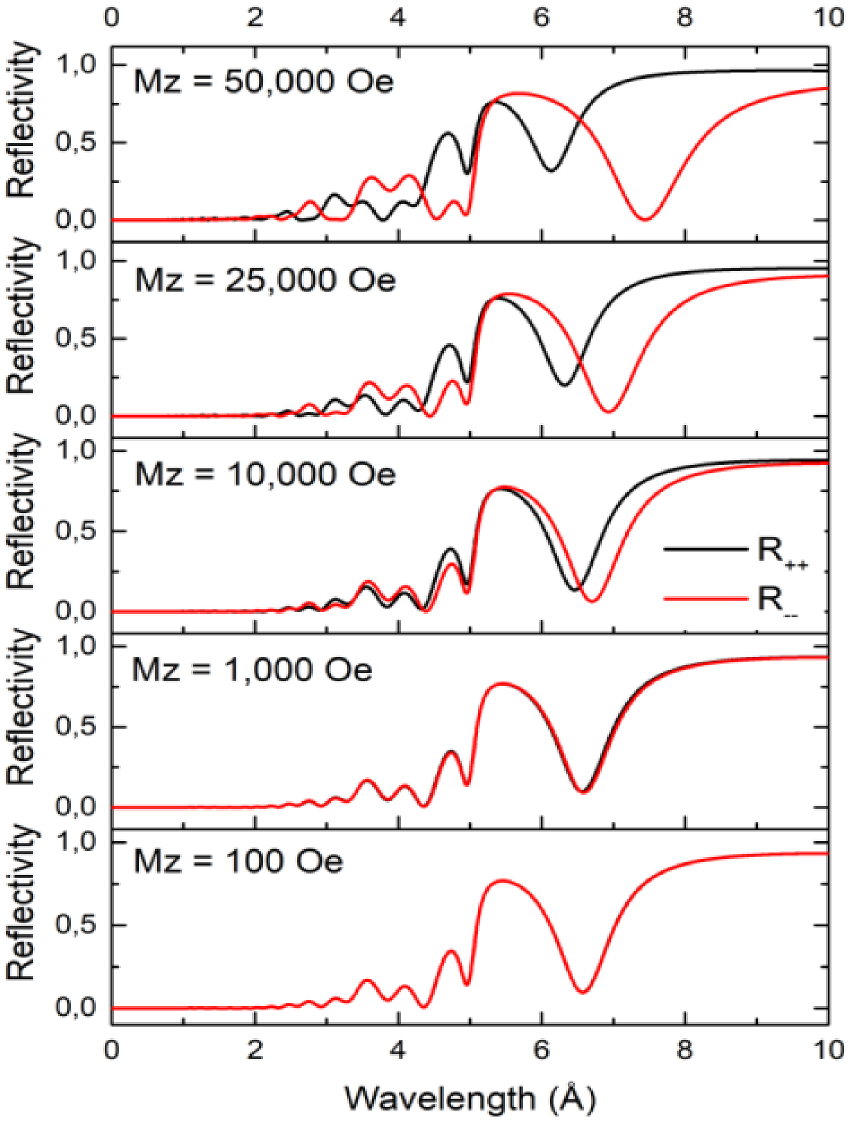


Spin asymmetry for heterostructure, grazing angle of 6 mrad.

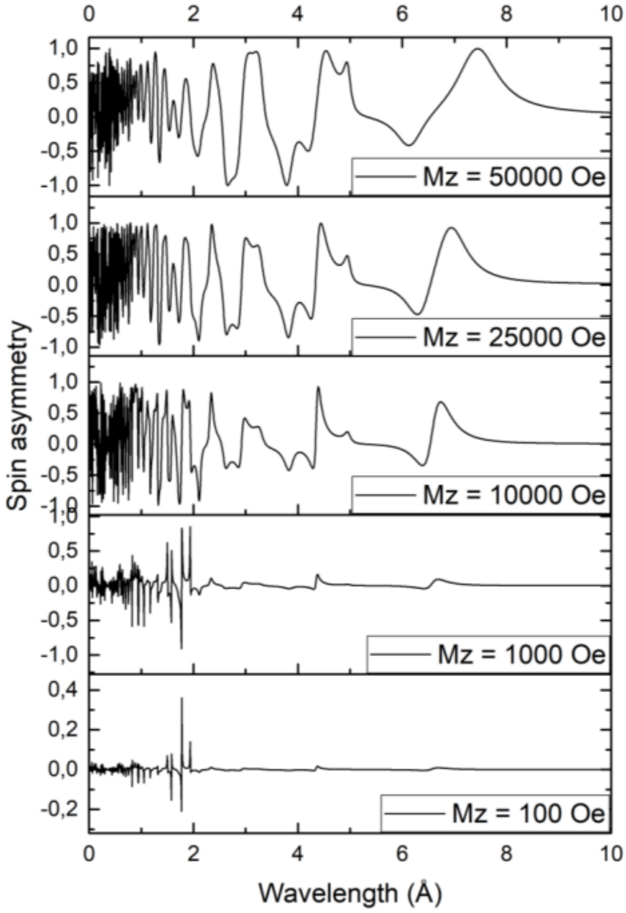
2.3. Task 3

Comparing reflectivity at different magnetization (calculation only neutron reflectivity)

Now, we analyzed the dependence on the strength of magnetization and its direction. Reflectivity spectra are shown the below Figure. Magnetization is just in the z-direction which is colinear magnetization, parallel to the surface of the sample, with values from 100 to 50,000 Oe. For the weakest Mz, a match of R++ and R-- can be seen, and there is no spin asymmetry, in the below figure. With the increase of the Mz, R++, and R— become more and more different in shape and values so spin asymmetry occurs. Higher magnetization has a stronger effect on the neutrons due to the interaction between external magnetization and neutron magnetic moment and thus reflectivity of differently polarized neutrons occurs as evidence of the presence of the magnetic moment

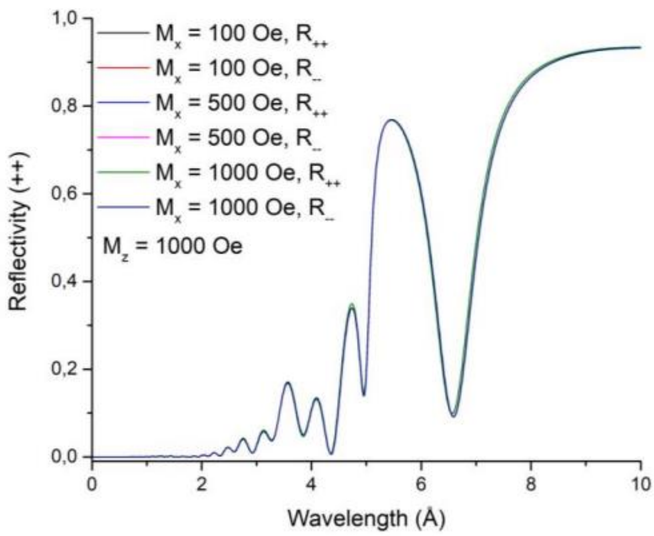


Reflectivities of the same heterostructure in different magnetizations. Magnetizations along x-axe are of 100, 1000, 10,000, 25,000, and 50,000 Oe.



Spin asymmetry for structures in different magnetic fields with Mz of 100, 1,000, 10,000, 25,000, and 50,000 Oe.

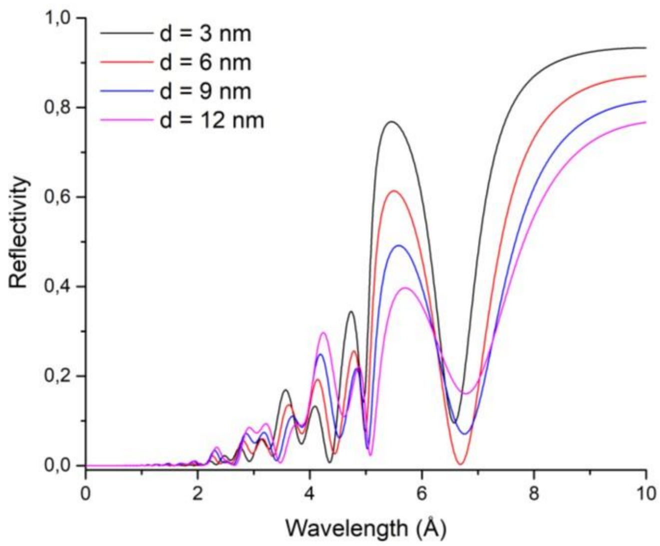
Reflectivities of investigated heterostructure are presented in the colinear magnetization of Mx = 100, 500, and 1000 Oe and Mz = 1000 Oe. The spin asymmetry didn’t occur. It can be concluded that the z component of magnetization has the main effect on the reflectivity and that the x component doesn't change the neutron reflectivity



Reflectivity for Mz = 1000 Oe, and values Mx of 100, 500, and 1,000 Oe.

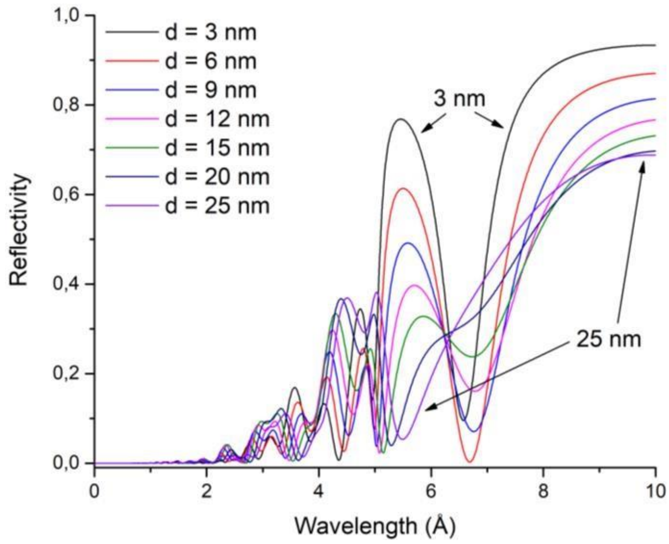
2.4. Task 4

Comparing structures with different thicknesses (calculation of neutron and X-ray reflectivity) The third parameter which we analyze is the thickness of the ferromagnetic layer of gadolinium. The calculation is performed for 3, 6, 9, 12, 15, 20, and 25 nm thick Gd layers. Due to the coherence length of superconductivity, this layer mustn't be too thick. The depth of the penetration of the superconductivity can be a few nanometers in the ferromagnetic layer. The below figure show reflectivities for the first four thicknesses.



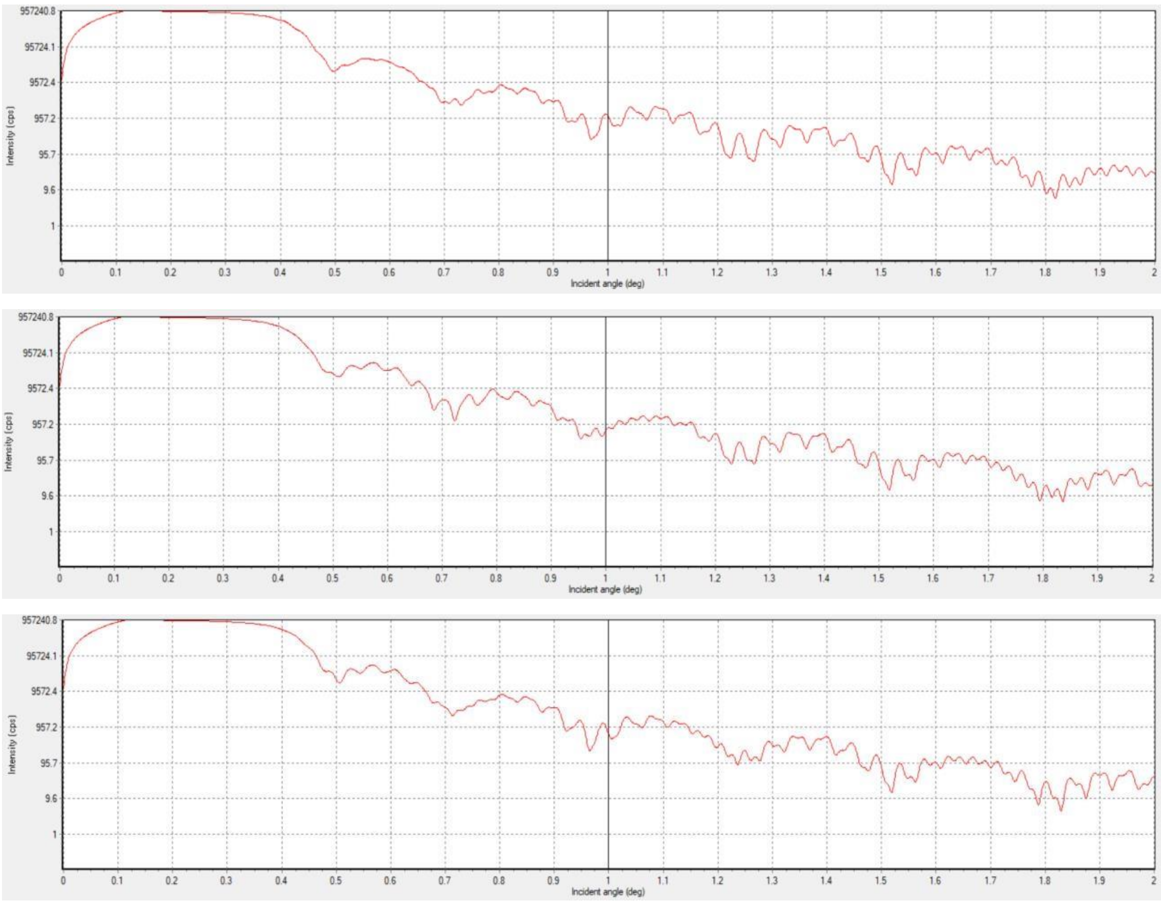
Reflectivity for heterostructure with the Gd layer thicknesses of 3, 6, 9, and 12 nm.

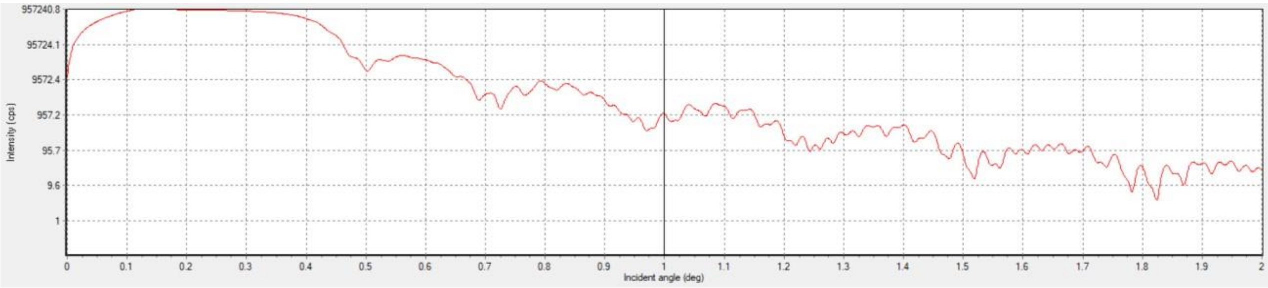
All calculated reflectivities are shown in Figure. For the Gd layer thicker than 20 nm there is a change in the shape of reflectivity due to the absorption of neutrons and by analyzing the emitted particles we could get a more detailed spatial magnetic profile of the structure. This is important because the ferromagnet induces magnetic moments in the surrounding superconducting layers and the profile of the magnetization in them is different close to the interface with the Gd layer.



Reflectivity for heterostructure with the Gd layer thicknesses of 3, 6, 9, 12, 15, 20, and 25 nm.

For thicknesses of Gd-layer of 3, 6, 9, and 12 nm. It can be seen that there are no flat surfaces between layers. This is expected due to the small thicknesses of our layers (a few nanometers).





.X-ray reflectivities for the heterostructure with the Gd layer thicknesses of 3, 6, 9, and 12 nm.

With the increase of thickness of the Gd layer, there is a change in the "height" of the extremums of the X-ray reflectivity spectra. According to the analysis of their shape and the difference in intensity of these peaks, we can check if does Gd layer has desired roughness and thickness (which is useful because it is hard to make desired "perfect" nanometer structures).

2.5. Task 5

Comparing structures with different ferromagnets (calculation of neutron and X-ray reflectivity)

As to this task, the reflectivity of the neutron, as well as X-Ray, were simulated with variations of ferromagnetic layers. There are 5 ferromagnets assumed to exist in the middle layers in each simulation (Gd. Fe, Co, Ni, Dy). It was assumed that there were no magnetizations and the grazing angle was constant, 6 mrad. Because each element has differing neutron scattering length densities, it was necessary to input neutron scattering length densities for each element. Data for neutron scattering length density was obtained from Neutron Activation and Scattering Calculator (nist.gov) for this task, neutron scattering length density could be summarized as follow,

Elements	Scattering Length Density (10 /Å)
Gd	(obtained from lemur program) / 4.150
Fe	8.024
Co	2.265
Ni	9.408
Dy	5.356

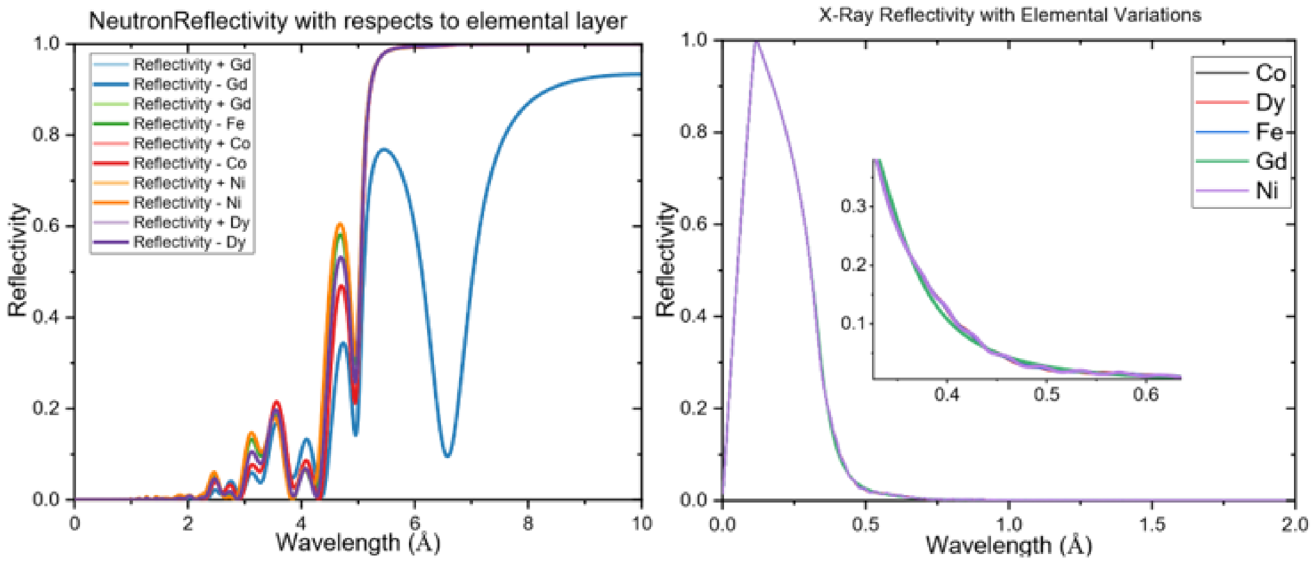
The layers being used for this task were as follow.

Al_2O_3/Nb (100 nm)/Gd (3 nm)/V (70 nm)/Nb (15 nm)

Al_2O_3/Nb (100 nm)/Fe (3 nm)/V (70 nm)/Nb (15 nm)

Al_2O_3/Nb (100 nm)/Co (3 nm)/V (70 nm)/Nb (15 nm)

Al_2O_3/Nb (100 nm)/NI (3 nm)/V (70 nm)/Nb (15 nm)
Al_2O_3/Nb (100 nm)/Dy (3 nm)/V (70 nm)/Nb (15 nm)



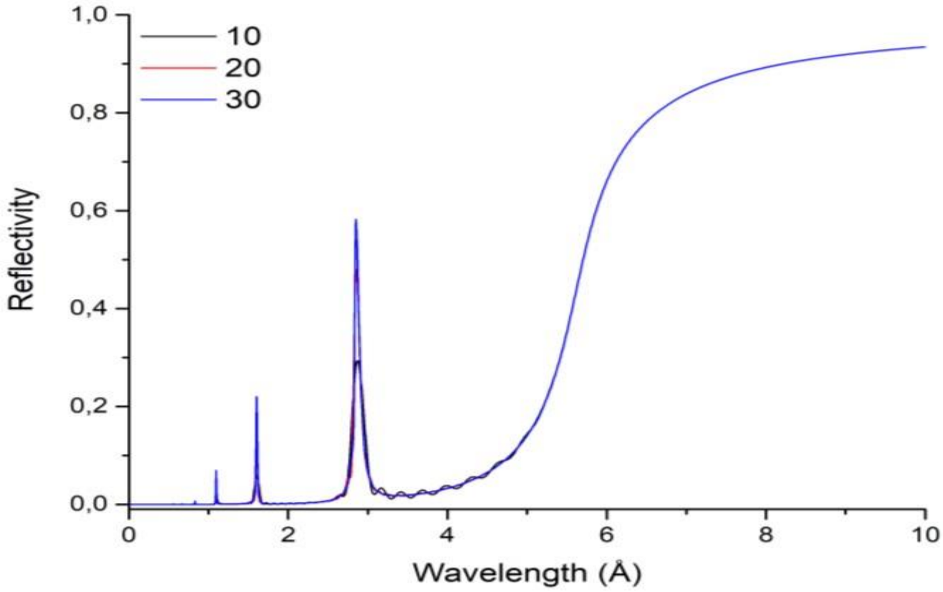
Simulation results of neutron and x-ray reflectivity.

The simulations show that there is no noticeable x-ray reflectivity due to elemental variations. As explained previously, this can be attributed to the location of the elements which were in the middle of heterostructures, and because of that, it did not contribute significantly to x-ray scattering. On the other hand, Neutron reflectivity has variational reflectivity in each peak of the upper left figure. Variational reflectivity below 5 Angstrom happened because ferromagnetism in the layer had various magnetic moment values. As such it has a differing effect on the proximity effect in the superconductivity. Gadolinium has a different neutron reflectivity value because in Gd/Nb layer, Er-Es and as such allow rather a large proximity effect where Gadolinium has superconductivity inside it. As a result, it screened out incoming neutron and has lower reflectivity.

2.6. Task 6.

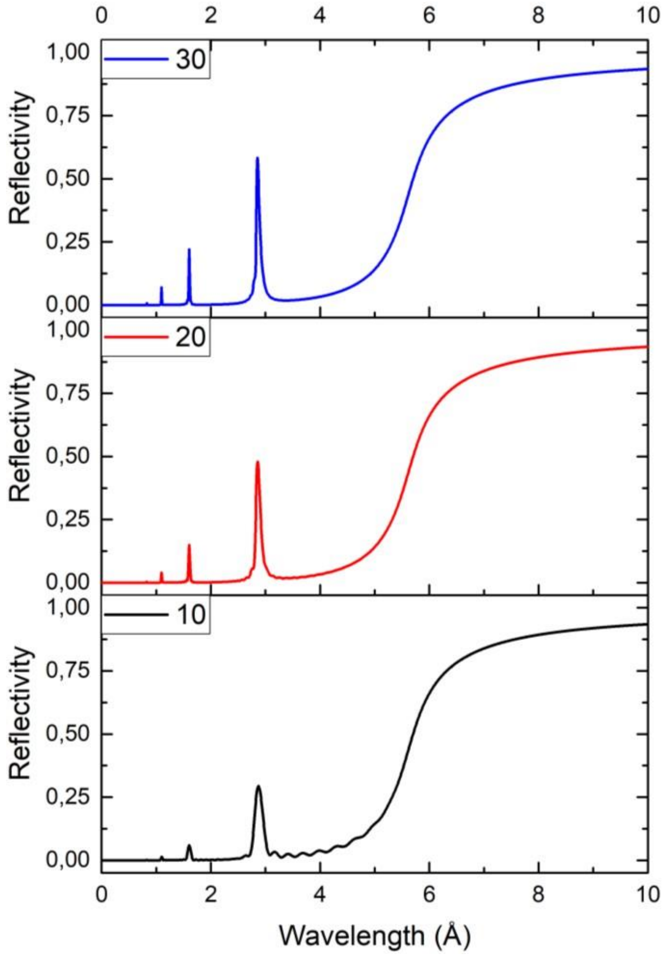
Superlattice (calculation of neutron and X-ray reflectivity)

In this topic , we analyze Al_2O_3/[Nb(25nm)/Gd(3nm)]$_n$/Nb(15nm) heterostructures with n = 10, 20, and 30 number of Nb(25nm)/Gd(3nm) bilayers. Obtained reflectivities of neutrons and X-ray beams are given in the following Figures. The neutron reflectivity spectra were given in two ways to make it easier to see the difference between them and to make the results clearer.



The reflectivity of investigated heterostructure with different superlattices.

Superlattice is made by the different number of bilayers of Nb(25nm)/Gd(3nm) layers. The more pairs of these layers exist in the structure, the more intense the reflection maxima due to the stronger coupling of ferromagnetism and superconductivity which is happening due to the pairing of electrons from these layers into pairs. One of the electrons is in the F layer and the second one is in the Slayer but with opposite oriented spin. From the below figure, it can be seen that for the n = 30, the reflectivity curve is smoother.



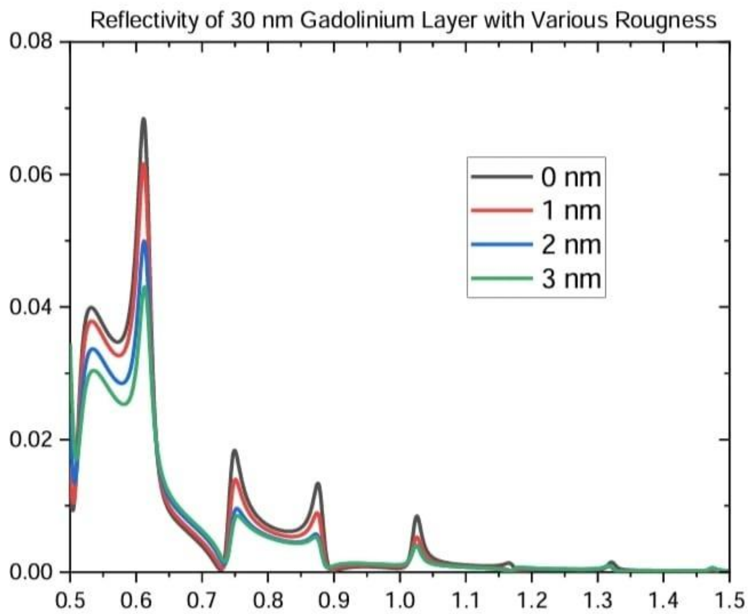
Comparison of the reflectivities of investigated heterostructure with different superlattices.

X-ray reflectivity showed an interesting result. A layer of heterostructures has different. reflectivity than multiple layers (superclusters) of heterostructures. The upper figure showed that for multiple layers (4 and beyond), there are no significant changes in reflectivity. intensity, but they had a larger reflectivity value compared to a layer of heterostructures. This happened because X-Ray scattering depended on electronic density on the surface of the sample. The larger the number of electrons, the more we could expect that there should be larger value scattered beam intensity. Furthermore, adding more layers to the samples means that there were more beams to reflect X-Ray, hence we found a larger reflectivity value for multiple layers samples.

The reason why we didn't observe any variations of reflectivity with the addition of more layers had to do with X-Ray penetration depth which was quite low for a sample that has a large surface electronic density of states such as transition metal. In contrast with X-Ray, the addition of more heterostructures layers gave rise to a larger value of reflectivity because neutron has a larger penetration depth than X-Ray (it does not interact with electron cloud around the d nucleus), as to when we add more layers to the samples, then we would expect to have a more nuclear magnetic moment which could scatter neutron. The result is consistent with what we already know so far about neutrons and their behaviors.

2.7. Task 7.

Influence of roughness (calculation only X-ray reflectivity)



Simulation Results

The simulation of x-ray reflectivity of gadolinium layer with various roughness closely matched the theoretical prediction of X-Ray reflectivity. From the figure, we could observe that from 0.5 to 1.5 Angstrom, the reflectivity of each sample varied with several noticeable peaks at 0.55, 0.66, 0.75, 0.9, and 1.025 Angstrom. For a small angle scattering, the intensity of the scattered beam decreased exponentially for a larger beam wavelength. This happened because, from Bragg's equation of scattering, a coherent scattering only occurs if and only if the beam wavelength has some correspondence with the object being observed, so for a larger wavelength, the scattered beam would have a lowered intensity.

X-ray scattering experiments often relied on the fact that detectors could only detect scattered beams that arrived at it, including superposition of beam due to lattice scattering of multilayers samples. If samples have some defects or inhomogeneity, then scattered beams would display reduced intensity because of destructive interference of reflected beams. This explains why in the results above, for a larger value of roughness, reflectivity became smaller. The reason is that for a larger value of roughness more beams were getting destructive interference and at the same time scattered specularly to more directions, thus reducing the intensity of beams that arrived at the sample

3. Conclusion:

In this report, different variations of the parameters and properties of the Al_2O_3/Nb(100nm)/Gd(3nm)/V(70nm)/Nb(15nm) heterostructures are studied from the side of the ferromagnetic/superconducting proximity effect by neutron and X-ray reflectivity. Due to the complexity of the mutual impact of F and S in examined heterostructures, we can conclude that there are present both proximity and inverse proximity effects.
Comparing neutron and X-ray reflectivity spectra, we discussed the influence of the grazing angle of the incident neutron beam, colinear and non-collinear magnetization, thickness of the ferromagnetic layer, different ferromagnetic materials, roughness, and the size of the superlattice on the S/F proximity effect in this structure. There is a big influence on the grazing angle which should be less than 12 mrad. The presence of the magnetization in the system induce spin asymmetry - the difference between R_{++} and R_{--}reflectivities. Increasing the thickness of the ferromagnetic layer can change the shape and intensity of the reflectivity spectra and due to it, there is no significant S/F proximity effect due to short coherence and London penetration lengths. Different ferromagnetic materials give much different reflectivity spectra due to different interactions of elements with neutron and X-ray beams due to different electronic and nuclei structures. The number of the sublayers in the superlattice affects the intensity of peaks in neutron reflectometry but it doesn't give big changes in X-ray reflectivity curves. The roughness of the Gd layer is studied by X-ray reflectometry and it can be obtained from the height of the reflectivity curve peaks.

Finally, it can be concluded that polarized neutron reflectometry is a powerful technique for analyzing the S/F proximity effects in S/F heterostructures.

4. Acknowledgment:

I would like to express my thanks to my Supervisor Dr. Vladimir Zhaketov, Frank Laboratory of Neutron Physics and Joint Institute for Nuclear Research "Interest Programme" for giving me a great opportunity to excel in my learning through this project "Coexistence of Superconductivity and Ferromagnetism at Low-Dimensional Heterostructures"

I have achieved a good amount of knowledge through the research & the help that I got from my Supervisor Dr. Vladimir Zhaketov.

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