Peculiarities of magnetic states in Ferromagnet/Superconductor heterostructures due to the proximity effects

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Abstract. The peculiarities of magnetic states in ferromagnet (F)/superconductor (S) heterostructures due to the proximity effects are investigated with the use of polarized neutron reflectometry. An anomalously strong reduction in the magnetization of the F layer due to superconductivity is observed in the S/F/S system. The dependence of the effects on an external magnetic field is studied for the first time. The enhanced neutron standing waves are used for registration of weak signals.

1. Introduction

The term “proximity effect” in heterostructures usually means the appearance of the properties of one phase in another, caused by the contact of these phases. In the physics of ferromagnet (F)/superconductor (S) proximity effects there are a number of intriguing predictions, which call for further clarification and experimental verification [1]. In particular, it was predicted that due to the proximity effects various scenarios of the influence of the superconducting phase transition on the magnetic properties of the layered F/S systems are possible: the formation of inhomogeneous ordered magnetic structure (cryptoferromagnetic), formation of the domain structure, magnetization “leakage” from F to S layer (inverse proximity effect). The magnetization of the system is reduced as a result. Experimentally the problem has been studied using the ferromagnetic resonance method [2, 3] and by the polarized neutron reflectometry (PNR) [4 – 7]. The effect of the reduction of the F layer magnetization and changes of magnetic profiles of the system below the superconducting transition were observed, but the understanding of physical mechanisms of these observations is not complete.

In this paper we present the results of investigations of F/S and S/F/S layered structures with the use of the polarized neutron reflectometry. We study the effects in dependence to an external magnetic field, which has not been done before. The due attention has been paid to the study of the quality of layers and interfaces, as it is known that considerable and developed interface roughness can disguise physical effects and change the interpretation. In the heterostructures with high structural quality the effects can be weak. We propose that in such a case the regime of enhanced neutron standing waves should be used in the polarized neutron reflectometry.

2. Field dependence of magnetization reduction

In our recent paper [7] we reported on an anomalously strong reduction of magnetization in the S/F/S structure \( \text{Nb}(50 \text{ nm})/^{57}\text{Fe}(3.9 \text{ nm})/\text{[Si}(3.4 \text{ nm})/\text{Mo}(3.4 \text{ nm})]_{40}/\text{Si} \). Here the Nb layer and Si/Mo
structure are two superconductors with transition temperatures $T_{C-Nb} = 9.2$ K and $T_{C-Si/Mo} = 4.5$ K, respectively. The F phase between them was presented by the iron layer with Curie temperature $T_K = 55$ K. Such a small temperature was explained by high intermixing with neighboring layers. The PNR measurements were performed with the time-of-flight polarized neutron reflectometer REMUR at the IBR-2 reactor (JINR, Dubna) at temperatures $T = 15$ K, 5 K and 2 K in the magnetic field $H = 0.5$ kOe. It was obtained that the magnetization decreases below $T_{C-Nb}$ and $T_{C-Si/Mo}$ from 1.9 kOe at 15K to 1.3 kOe (≈30%) at 5K and to 0.1 kOe (≈95%) at $T = 2$K. A row of Abrikosov vortices was observed in the center of the Nb layer at 2 K. A possible reason for the absence of vortices at 5 K can be the shunting of the magnetic flux by the F layer with non-zero magnetization, which leads to the reduction of intensity of the magnetic field in the Nb layer. The estimations give $H \approx 150$ Oe, which is much less than $H_{C1} \approx 360$ Oe.

The experiments have been repeated recently on the reflectometer ADAM at ILL at $H = 225$, 750 and 970 Oe [8]. At $H = 750$ and 970 Oe we observed similar behavior as at $H = 500$ Oe. However, at $H = 225$ Oe no change of spin asymmetry has been observed. Here spin asymmetry $S = [R^- - R^+] / [R^- + R^+]$ is the ratio of difference to the sum of two neutron reflectivities in different spin states. With the assumption that the magnetic part of the optical potential is much less than the nuclear one, it can be easily shown that the spin asymmetry is proportional to the magnetization profile of the structure. In Fig.1 the temperature evolution of the integrated absolute value of the spin asymmetry weighted on statistical error $f = \int [S(Q)/\delta S(Q)]dQ$ is shown. The normalization to the statistical error helps to avoid artificial effects in the case when the spin asymmetry is small. We use modulus of $S(Q)$ because for this sample the spin asymmetry is an oscillating function and therefore simple integration would give a value close to zero. Thus, one can say that the parameter $f$ is somehow proportional to the absolute value of magnetization of the whole sample.

![Figure 1](image_url)  

**Figure 1.** $f = \int [S(Q)/\delta S(Q)]dQ$ at different magnetic fields. The details see in the text.

The performed measurements show that the reduction of the average magnetization of the F layer due to the influence of superconductivity depends on the external magnetic field. This process develops at the fields $H \geq 0.5$ kOe in two stages: after the first superconducting transition in the Nb layer and then after the second transition in the [Si/Mo] layer. At the field $H = 0.5$ kOe we observe an anomalously strong reduction. The suppression of magnetism by superconductivity decreases with increasing $H$. The analysis of the diffuse scattering, field and temperature dependence of the
magnetization leads us to the conclusion that there are two mechanisms of the magnetization reduction: the appearance of the domains oriented by the magnetic field and the suppression of the magnetization inside each domain by the superconducting exchange energy. At $T < T_C$ a part of electrons in the F layer due to the proximity effect forms Cooper pairs and therefore does not participate in the ferromagnetic alignment.

3. Field dependence of the magnetic structure at the S/F interface

In this section we consider the details of the magnetic structure at the S/F interface at different external magnetic fields. The peculiarities of the magnetic state can be clarified with the use of the regime of neutron standing waves in a reflectometry experiment. This regime allows us to scan the spatial vicinity of an interface to detect magnetic non-collinear or scattering regions such as domains, clusters, vortices, etc. [9]. The study of the structure Pd(1.5 nm)/V(39 nm)/Fe(3 nm)/

$20 \times [V(3 \text{ nm})/Fe(3 \text{ nm})]/\text{MgO}$ has been done on the reflectometer D-17 at ILL (Grenoble) at $T = 3K$ and 7 K at magnetic fields $H = 0.2$, 0.7, 1.0, 1.5 and 4.5 kOe. The periodic structure $20 \times [V/Fe]$ (PS) plays a role of a standing wave generator. The superconducting critical temperature and magnetic field (applied parallel to the surface) in the S layer are $T_C(0) = 3.7$ K and $H^C_2(0) \approx 22$ kOe respectively. In Fig. 2 the spin asymmetries at magnetic fields $H = 0.7$ and 1.5 kOe are shown. The peak at $Q = Q_B = 1 \text{ nm}^{-1}$ corresponds to the 1st order Bragg reflection from the PS. The oscillations at $Q = [0.1 \pm 1] \text{ nm}^{-1}$ are mainly defined by the magnetic state at the S/F interface. At $H \leq 1$ kOe no significant changes of the spin asymmetry at the superconducting transition have been observed. From the spin asymmetries at $T = 7$ K it follows that the growth of peak at $Q = Q_B$ with increasing magnetic field takes place. This growth points to the process of magnetization of the PS. Starting from $H = 1.5$ kOe the magnetization of the S/F interface also takes place. When the V(39nm) layer goes into the superconducting state (see curves at $T = 3$ K) spin asymmetry $S(Q)$ at 1.5 kOe looks the same as spin asymmetries below 1 kOe. The same effect has been observed at higher field $H = 4.5$ kOe. Thus, one can draw a conclusion that the magnetization of the S/F interface is suppressed during the transition into the superconducting state of V(39 nm). The magnetic state of a periodic structure is not changed meanwhile.

![Figure 2](image)

Figure 2. Spin asymmetries at $T = 7$ K (open circles) and $T = 3$ K (solid circles) at different magnetic fields.

The intensity of diffuse scattering from magnetic inhomogeneities in the regime of standing waves (i.e. at $Q = Q_B$) can be written as: $J_{\text{diff}}(Q) \sim \int n(z, Q) \sigma_{\text{diff}}(z) \, dz$ [9]. Here $\sigma_{\text{diff}}(z)$ is the cross-section of diffuse scattering at a depth $z$, $n(z, Q)$ is the neutron density at a given depth and $Q$. For the latter we
can write an approximate form: \( n(z, Q) = (1 + R^{1/2})^2 - 4\sin^2[(Q\Delta z + \alpha(Q - Q_B)]/2\). Here \( R \) is the reflectivity at \( Q_B \), the factor \( \alpha \) depends on the number of repetitions \( N \) and thickness of the period \( D \) of the PS as \( \alpha = ND/2 \). The expression for \( n(z, Q) \) shows that at a given \( Q \) the neutron density has its maximum at a certain distance \( \Delta z = \alpha(Q_B/Q - 1) \) from the S/F interface. Thus, the diffuse scattering intensity provides information on the distribution of inhomogeneities at a depth \( \Delta z \). Its maximum at certain \( Q \) corresponding to the maximum of the diffuse scattering cross-section is situated at \( \Delta z \). The expression for the diffuse scattering can be simplified for the case when the function \( \sigma_{\text{diff}}(z) \) is locally limited around the S/F interface at a distance much smaller than a period of the standing wave. For this case we can omit integration. In Fig.3 the normalized \( z \)-distribution of inhomogeneities \( \kappa_D = \sigma_{\text{diff}}(\Delta z)/\sigma_{\text{diff}}(0) \) at different magnetic fields is presented. From Fig. 3 we can conclude that at \( H = 1.5 \) kOe the distribution maximum is closer to the F layer while at \( H = 4.5 \) kOe it is closer to the S layer. This observation can be interpreted in the following way. At \( H = 1.5 \) kOe there is an inhomogeneous magnetic state of the F layer, which becomes homogeneous at higher fields. At \( H = 4.5 \) kOe (\( \geq H_C \)) we have a homogeneous magnetic state in the F layer and a developed vortex state in the S layer.

**Figure 3.** Depth distribution of the inhomogeneities in the vicinity of the S/F interface at \( T = 7 \) K (open circles) and \( T = 3 \) K (solid circles). Solid and dashed lines show the position of the mass center above and below \( T_C \).

4. **New approach in reflectometry of proximity effects**

The influence of superconductivity on ferromagnetism in S/F heterostructures is usually pronounced for weak ferromagnets. This weakness can be achieved when a strong ferromagnet in the F layer is diluted or the thickness of the F layer is small (in the order of 1 nm). In both cases we have a weak magnetic signal in a reflectometry experiment to detect it by usual method. There is a possibility to enhance the signal in this case by using enhanced neutron standing waves [9].

For the experimental study we used a structure Cu(33 nm)/V(40 nm)/Fe(1 nm)/MgO, prepared by molecular beam epitaxy in RMKI KFKI (Budapest, Hungary) [10] on the basis of theoretical calculations [11]. Here S/F bilayer V(40 nm)/Fe(1 nm) is placed between Cu and MgO with high neutron scattering length densities to create regime of resonance enhanced neutron standing waves. The evaporation of iron directly onto the substrate allowed us to obtain structurally homogeneous F layer with extremely low roughness (\( \sigma \sim 1-2 \) monolayers). Superconducting transition temperature of the vanadium layer and Curie temperature of the iron layer are \( T_C = 3.5 \) K and \( T_K \geq 300 \) K, respectively.

The preliminary investigation of the sample was performed at the reflectometer N-REX+ (FRM2, Münich) at room temperature [10]. The neutron wave function in the structure was calculated with the
use of the neutron scattering length density profiles obtained in this experiment. It was obtained that
the neutron density around the S/F interface was $n_{S/F} = 50$ in comparison with the density $n_0 = 1$ in the
incident beam.

Figure 4 presents the results of the reflectivity measurements in spin-flip and non-spin-flip channels at $T = 10$ K and $H = 20$ Oe performed at the reflectometer ADAM (ILL). The magnetic field was in the sample plane and perpendicular to the magnetization of the F layer as shown in the inset to Fig. 4. The value of the resonance-enhanced spin-flip signal over the initial beam was 10%. This made it possible to measure the temperature dependence of the resonance peak with an accuracy of around 1% (Fig. 5a) and the diffuse scattering during 8-12 hours.

The decrease in the spin-flip intensity at $T < T_C$ (Fig. 5a) can be connected with the reduction of magnetization in the F layer. The increase of the diffuse scattering around $T = 5$ K just before $T_C$ (Fig. 5b) may be due to the appearance of short-range order clusters, which destroy the long-range magnetic order. The subsequent increase in the spin-flip intensity at $T < 2.5$ K ($\approx 0.6 T_C$) can be related to the appearance of induced magnetization in the S layer, which is antiparallel to the magnetization of the F layer and therefore noncollinear to the external magnetic field. The increase in the diffuse scattering around $T = 2$ K can be connected with the appearance of magnetic clusters in the S layer. Another possible mechanism of the presented behavior at $T = 2$ K consists in increasing of the reverse magnetization due to the increase in the exchange interaction below 5 K.

The presented results show the efficiency of the use of the regime of enhanced neutron standing waves for investigations of weak changes of magnetic states at S/F interfaces: in the given example we were able to detect a 4% decrease in the spin-flip intensity, which corresponds to a 2% decrease in the magnetization of the F layer.

5. Conclusions
The peculiarities of magnetic states in the F/S heterostructures due to the superconducting phase transition in the S layer depending on the external magnetic field have been investigated for the first time. In the S/F/S structure an anomalously strong reduction in the magnetization of the F layer up to 95% from $M_{sat}$ has been revealed. Both in the S/F and S/F/S structures a change in the nonhomogeneous domain state is observed below the superconducting temperature $T_C$. The reduction in the mean magnetization of a domain due to the superconducting exchange energy is suggested. Our results do not support the model of cryptoferromagnetic state.

It has been demonstrated that the details of the magnetic structure at a given spatial position, for example at the S/F interface, can be clarified with the help of the regime of neutron standing waves in

![Figure 4. Spin-flip and non-spin-flip reflectivity at $T = 10$ K. In the inset – scheme of the experiment: $H$ and $M$ – vectors of external field and magnetization of the F layer, solid line inside the structure shows neutron density inside the structure at resonance.](image-url)
the polarized neutron reflectometry. The regime of enhanced neutron standing waves is proposed for investigations of weak effects.

**Figure 5.** Temperature dependence of specular spin-flip (a) and diffuse (b) scattering around the resonance.

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