Magnetic proximity effect in [Nb/Gd] superlattices seen by neutron scattering

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(Dated: February 21, 2019)

We have used spin-polarized neutron reflectometry to investigate the magnetization profile of superlattices composed of ferromagnetic Gd and superconducting Nb layers. We have observed a partial suppression of ferromagnetic (F) order of Gd layers in [Gd(dF)/Nb(25nm)]12 superlattices below the superconducting (S) transition of the Nb layers. The amplitude of the suppression decreases with increasing dF. By analyzing the neutron spin asymmetry we conclude that the observed effect has an electromagnetic origin - the proximity-coupled S layers screen out the external magnetic field and thus suppress the F response of the Gd layers inside the structure. Our investigation demonstrates the considerable influence of electromagnetic effects on the magnetic properties of S/F systems.

Artificial heterostructures with alternating superconducting (S) and ferromagnetic (F) layers are currently attracting great attention due to a diverse set of proximity effects [1–5], including the Larkin-Ovchinnikov-Fulde-Ferrell phase, π-phase superconductivity and triplet pairing. These effects show how ferromagnetism influences the superconducting properties of the S/F heterostructures. Converse proximity effects in which superconductivity influences ferromagnetism have received less attention. Such magnetic proximity effects are expected in systems where the F and S transition temperatures, TF and Tc, are comparable, which is the case for heterostructures of cuprate high-Tc superconductors and ferromagnetic manganates [6–9], and for some bulk compounds [10–12]. However, because of the chemical and electronic complexity of these materials, simple model systems for magnetic proximity effects are highly desirable.

For most S/F heterostructures composed of elemental metals or alloys, TF greatly exceeds Tc. In such systems, one still expects significant magnetic proximity effects if the effective energy EF ∼ TFdF/dS becomes comparable to ES ∼ Tc, where dF (dS) are the thicknesses of the F (S) layers [1]. The first to indicate such a possibility were Anderson and Suhl [13]. They considered systems consisting of S and F phases and came to the conclusion that a homogeneous magnetic phase above Tc may become inhomogeneous below Tc. Such a transition, which they called cryptoferromagnetism (CFM), would depress the effective exchange field of ferromagnetism, thus enabling the co-existence of superconductivity and magnetism. Later on the concept of CFM was further investigated in the theoretical work of Buzdin and Bulavinov [14] and Bergeret et al. [15]. Recently Zhu et al. reported the observation of an increased coercivity below Tc in GdN/Nb/GdN trilayers [16]. The authors interpreted this increase as a superconductivity-driven antiferromagnetic (AF) alignment of the GdN layers.

The ability to control the magnetic state by superconductivity is attracting attention also in applied research on superconductor spintronics [17, 18] including such new approaches as neuromorphic computing [19–21]. At the moment, most research efforts are focused on simple S/F structures such as bilayers and trilayers [22–24]. However, both the superconducting and the magnetic properties of more complex S/F systems, such as [S/F]n superlattices, may qualitatively differ from the properties of their S/F unit cells thus opening up perspectives for novel functionalities. An essential difference in behavior is expected when the thickness of the S and/or F layer becomes comparable with the coherence length of superconductivity in the S (ξS) or F (ξF) layers [25–29]. Preparation of such superlattices requires proper choice of materials with thin F and S layers, transparent S/F interfaces, and uniformity of the layer characteristics throughout the entire structure.

Our recent study of Nb(25nm)/Gd(dF)/Nb(25nm) trilayers has shown that high quality structures with highly transparent S/F interfaces and rather high correlation length ξF = 4nm can be grown [30]. Moreover gadolinium is a weak ferromagnet with TF = 293K which in combination with Nb, the strongest elemental superconductor with Tc = 9.3K, allows for preparation of S/F systems with EF ∼ ES. In this work we report on a study of the magnetic and superconducting properties of superlattices of composition [Gd(dF)/Nb(25nm)]12. The superlattices were deposited on 25x5mm2 (1102) Al2O3 substrates and covered by a Nb(5nm) capping layer. Later on we cut ~5x5mm2 pieces for magnetization and transport measurements (details of the sample preparation can be found in Ref. [30]). The thickness of the Gd layers was chosen to be dF = 0.5ξF (sample 1), 0.75ξF (sample 2) and 1.25ξF (sample 3). Fig. 1 shows an X-ray scattering map measured on sample 2 at the NREX neutron/X-ray reflectometer (details can be found in Ref. [31]). The
specular channel $\theta_2 = \theta_1$ exhibits more than 15 Bragg peaks arising from diffraction on the superlattice with period $D = d_F + d_S$, demonstrating high repeatability of a Gd/Nb bilayer in the $z$-direction. In addition we have detected diffuse scattering for $\theta_2 \neq \theta_1$ in the form of tilted lines around the specular Bragg peaks. These Bragg-like sheets indicate a high statistical correlation of the in-plane roughness profiles of Nb/Gd interfaces in the periodic structure [32].

To study the magnetic properties of our superlattices we used Polarized Neutron Reflectometry (PNR), which has been widely used for the study of S/F systems [6–9,33,36]. PNR allows measurements of the depth profile of the in-plane magnetization with nanometer depth resolution. Fig. 2a shows typical spin-up $R^+$ and spin-down $R^-$ reflectivity curves measured on sample 1 at $T = 7$K (that is, in the normal state above $T_c$) and $H = 4.5$Koe after cooling the sample in the same field. The neutron reflectivity exhibits six Bragg peaks positioned at $Q_n \approx 2\pi n / D$. The difference of $R^+$ and $R^-$ clearly indicates the presence of an in-plane magnetic moment. Using the Born approximation one can show that the spin asymmetry at the $n$-th Bragg peak $S_n \equiv [R^+(Q_n) - R^-(Q_n)]/[R^+(Q_n) + R^-(Q_n)]$ is proportional to the magnetic contrast $M_{Gd} - M_{Nb}$ of a unit cell, where $M_{Gd,Nb}$ is the in-plane magnetization of a Gd and Nb layer, respectively. In the first approximation we may neglect the small magnetization of the Nb layers ($M_{Gd} \gg M_{Nb}$) and write $S_n \sim M_{Gd}$. We were able to reproduce the experimental curves with a model based on 12 identical pairs with $d_F = 1.7$ nm and $d_S = 25.0$ nm and magnetization of Gd of $4\pi M_{Gd} = 2.5$ kG. Within the measurement error of 10%, this value agrees with 2.7kG which can be calculated using the saturation magnetic moment $m_{sat} = 134$ emu measured by a Superconducting Quantum Interference Device (SQUID) magnetometer (inset in Fig. 2a) at the same temperature and magnetic field.

Fig. 2b shows the superconducting phase diagram of sample 1 measured by a standard four-point electrical resistivity technique. This phase diagram allowed us to determine $T_c = 5.5$K and also the superconducting coherence length $\xi_S = 2\pi / \sqrt{2\pi H_{c2}(0)} = 11.6$nm. The latter value is in agreement with the previously reported value for Nb films [30].

Fig. 3 shows the field and temperature behavior of the spin asymmetry at the first Bragg peak $S_1$. Above $T_c$ we used the following protocol. First the sample was magnetized for a short time in the maximum magnetic field $H_{max} = 4.5$Koe. Then the field was released to zero and $S_1(H)$ was measured for ascending magnetic field from 5Oe to $H_{max}$ (black curve in Fig. 3a). Then the field was released to zero and the sample was cooled down to $T = 3.3$K in zero magnetic field. After this we repeated $S_1(H)$ by first raising the magnetic field from 5Oe to $H_{max}$ (red curve in Fig. 3a) and then lowering it to $H = 5$Oe (green curve in Fig. 3a). The $S_1(H)$ curve above $T_c$ repeats qualitatively the behavior of the upper branch of the macroscopic magnetic moment (inset in Fig. 2a): the $S_1(H)$ curve grows from remanence to $H \sim 2$Koe and then approaches saturation. The corresponding curve below $T_c$ is somewhat suppressed in the range of fields between remanence and saturation. The suppression is maximal around $H \approx 700$ Oe. The descending curve in turn is different at small fields close to zero. In order to check whether this difference is related to the superconducting state we measured the temperature dependence $S_1(T)$ using the following protocol. Above $T_c$ the sample was magnetized to saturation for a short time and then the field was released to zero and the sample was cooled down to 3.3K in zero field. Then a field of $H = 661$Oe was applied and $S_1(T)$ was measured by first heating the sample to $T = 7$K (black curve in Fig. 3b) and then cooling it back in the same field to $T = 3.3$K. One can see that the amplitude of $S_1$ is indeed suppressed below $T_c$ if the sample is cooled in zero field. We have conducted similar measurements for the other two samples and observed that the magnitude of the suppression is inversely proportional to $d_F$ (see the inset in Fig. 3b). For sample 3 with $d_F = 1.25\xi_F$ the effect is small but non-vanishing.

We have thus observed a suppression of the spin asymmetry of the first Bragg peak below $T_c$ after zero-field cooling. The effect takes place in an intermediate range of magnetic fields between remanence and saturation. However, we did not observe any additional Bragg peaks below $T_c$ at these magnetic fields. Thus AF alignment or any other modification of the magnetic period can be excluded in our structure. Moreover in our previous study of Nb(25nm)/Gd(dF)/Nb(25nm) trilayers we did not observe any statistically significant change of the spin asymmetry below $T_c$ [30]. All these observations point to an electrodynamical origin of the effect. Indeed for $d_F \sim \xi_F$ two adjacent S layers are expected to be coupled by the proximity effect. This means that the whole sample is a superconductor with thickness $D_S = 12D \approx$
300 nm which is larger than the magnetic screening length $\lambda_{Nb} \sim 120$ nm in niobium films. Such a thick superconductor is able to expel a certain amount of external field. As a consequence, the central Gd layers feel less magnetic field than applied outside and hence their response is smaller. If the sample is cooled in a magnetic field, then magnetic flux is trapped around the Gd layers and the effect is smaller or not seen at all. This model also explains the existence of the effect in the intermediate range of magnetic fields where the derivative $dM/dH \neq 0$. Moreover, the same mechanism explains why we didn’t see the effect in Nb(25 nm)/Gd/Nb(25 nm) trilayers - the total thickness of the superconductor $D_S = 50$ nm is not enough to expel a significant amount of magnetic flux.

In order to qualitatively describe the suppression of the spin asymmetry we have fitted the neutron data measured on sample 1 above and below $T_c$ in magnetic field $H = 0.8$ kOe (Fig. 4a,b). Above $T_c$ we used a model with twelve identical Gd layers. The best fit was obtained for $4\pi M_{Gd} = 1.4$ kG. To fit the data below $T_c$ we used the following procedure. For the given value

FIG. 2. (a) Experimental (dots) neutron reflectivity curves measured on sample 1 at $T = 7$ K and $H = 4.5$ kOe. Solid lines show model curves. Inset: Magnetic hysteresis loops measured at $T = 7$ K by a SQUID magnetometer. (b) Temperature dependencies of the upper critical field measured with external field applied parallel (black) and normal (red) to the surface.

FIG. 3. (a) Magnetic field dependent spin asymmetry at the first Bragg peak, $S_1$, measured above $T_c$ (black) and below $T_c$ (red and green). The black and red curves were measured in ascending magnetic field from zero to $H = 4.5$ kOe. The green curve was measured in descending field from saturation to zero. The sample was cooled in zero field below $T_c = 5.5$ K. Open symbols show the difference $\delta S_1 = S_1(7$ K$) - S_1(3$ K$)$. (b) Temperature dependence of $S_1$ measured in the magnetic field $H = 661$ Oe where the maximum of $\delta S_1$ was observed. The black curve was measured by increasing the temperature after ZFC and after this the red curve was measured with decreasing temperature. The inset shows the $\delta S_1^{MAX}(d_F)$ dependence.
of λ we calculated the value of the local magnetic field \( H(z) \) at the position of every Gd layer using the well-known expression for the Meissner effect in a superconducting film of thickness \( D_S \) and applied magnetic field \( H_0 \): 
\[
H(z) = H_0 e \kappa (z/\lambda) / ch(D_S/2\lambda).
\]
This value is considerably larger than the cryptoferromagnetism predicted in [13–15]. In analyzing Gd layers. This effect is to some extent similar to thus suppressing the ferromagnetic response of the inner thick (magnetic) superconductor. As a thick superconductor it is able to screen the applied magnetic field, thus suppressing the ferromagnetic response of the inner Gd layers. This effect is to some extent similar to the cryptoferromagnetism predicted in [13–15]. In analogy to CFM it leads to a transition from homogeneous magnetic order above \( T_c \) to inhomogeneous order (along z in our case) below \( T_c \), and hence to a suppression of the averaged magnetic moment. Similar to CFM, the effect takes place for a weakened ferromagnet (\( d_F < \xi_F \)) and strengthened superconductor (\( D_S > \lambda \)). Our investigation shows that electromagnetic effects may play a significant role in S/F systems and should be taken into account when considering proximity effects in S/F systems. Note that recent theoretical work came to a related conclusion [15]. Our results demonstrate the potential of elemental S/F multilayers as simple model systems for ferromagnetic superconductors.

The authors would like to thank G. Logvenov, G. Christiani, A. Melnikov and S. Mironov for fruitful discussions. This work is partially based on experiments performed at the NREX instrument operated by the Max Planck Society at the MLZ), Garching, Germany, and supported by DFG collaborative research center TRR 80. Research in Ekaterinburg was performed within the state assignment of Minobrnauki of Russia (theme "Spin") No. AAAA-A18-118020290104-2) and was partly supported by RFBR (Project No. 19-02-00674).

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FIG. 4. Experimental spin asymmetry measured on sample 1 above and below $T_c$. (b) The best-fit model spin asymmetries for the magnetization depth profiles depicted in (c). (d) Dependence of the goodness of fit versus magnetic field penetration depth.

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