Full spin switch effect for the superconducting current in a superconductor/ferromagnet thin film heterostructure

P. V. Leksin, 1 N. N. Garif’yanov, 1 I. A. Garifullin, 1 J. Schumann, 2
H. Vinzelberg, 2 V. Kataev, 2 R. Klingeler, 2 O. G. Schmidt, 2 and B. Bückner 2

1 Zavoisky Physical-Technical Institute, Kazan Scientific Center of Russian Academy of Sciences, 420029 Kazan, Russia
2 Leibniz Institute for Solid State and Materials Research IFW Dresden, D-01171 Dresden, Germany

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Superconductor/ferromagnet (S/F) proximity effect theory predicts that the superconducting critical temperature of the F1/F2/S or F1/S/F2 trilayers for the parallel orientation of the F1 and F2 magnetizations is smaller than for the antiparallel one. This suggests a possibility of a controlled switching between the superconducting and normal states in the S layer. Here, using the spin switch design F1/F2/S theoretically proposed by Oh et al. [Appl. Phys. Lett. 71, 2376 (1997)], that comprises a ferromagnetic bilayer separated by a non-magnetic metallic spacer layer as a ferromagnetic component, and an ordinary superconductor as the second interface component, we have successfully realized a full spin switch effect for the superconducting current.

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The antagonism of superconductivity (S) and ferromagnetism (F) consists of strong suppression of superconductivity by ferromagnetism because ferromagnetism requires parallel (P) and superconductivity requires antiparallel (AP) orientation of spins. The exchange splitting of the conduction band in strong ferromagnets which tends to align electron spins parallel is larger by orders of magnitude than the coupling energy for the AP alignment of the electron spins in the Cooper pairs in conventional superconductors. Therefore the singlet pairs with AP spins of electrons will be destroyed by the exchange field. For this reason the Cooper pairs can penetrate into an F-layer only over a small distance. The characteristic distance of decay of the pairing function in the F-layer is \( \xi_F = (4\hbar D_F/I)^{1/2} \), where \( D_F \) and \( I \) are the diffusion coefficient and the exchange splitting of the conduction band in the F-layer, respectively [1]. For pure Fe the value of \( \xi_F \) is less than 1 nm (see, e.g., [2]).

The physical origin of the spin switch effect based on the S/F proximity effect relies on the idea to control the pair-breaking, and hence the superconducting transition temperature \( T_c \), by manipulating the mutual orientation of the magnetizations of the F-layers in a heterostructure comprising, e.g., two F- and one S-layer in a certain combination. This is because the mean exchange field from two F-layers acting on Cooper pairs in the S-layer is smaller for the AP orientation of the magnetizations of these F-layers compared to the P case. Historically, the first paper devoted to the realization of the spin switch effect by manipulating the mutual orientation of the magnetizations of the F-layers has been published by Deutscher and Meunier in 1969 [2]. They studied FeNi/In/Ni trilayer and obtained a surprisingly large difference in \( T_c \) between the AP and P orientations of the magnetizations. The reason for this effect has not been clarified up to now. Clinton and Johnson [4] have developed a superconducting valve which uses the magnetic fringe fields at the edges of the F film of a \( \mu \)m size. These fringe fields can be varied in magnitude by changing the mutual orientation of the magnetization of two F layers separated by a nonmagnetic (N) spacer layer. In their F/N/F/S construction a dielectric interlayer between the F/N/F valve and the bridge in the S-layer with a width of 1 \( \mu \)m was formed artificially. Thus, a direct contact between magnetic and superconducting parts of the sample was absent similar to the case studied in Ref. [3]. The possibility to develop a switch based on the S/F proximity effect has been theoretically substantiated in 1997 by Oh et al. [5]. They proposed the F1/N/F2/S layer scheme where an S-film is deposited on top of two F-layers, F1 and F2, separated by a thin metallic N-layer. The thickness of F2 should be smaller than \( \xi_F \) to allow the superconducting pair wave function to penetrate into the N-layer. Two years later a different construction based on a trilayer F/S/F thin film structure was proposed theoretically by Tagirov [6] and Buzdin et al. [6, 7]. Several experimental works confirmed the predicted influence of the mutual orientation of the magnetizations in the F/S/F structure on \( T_c \) (see, e.g., [8-12]). However, the difference in \( T_c \) between the AP and P orientations \( \Delta T_c \) turns out to be smaller than the width of the superconducting transition \( \delta T_c \) itself. Hence a full switching between the normal and the superconducting state was not achieved. Implementation of a design similar to the F1/N/F2/S layer scheme by Oh et al. [5] with a [Fe/V] \( n \) antiferromagnetically coupled superlattice instead of a single F1/N/F2 trilayer [13, 14] cannot be switched from the AP to P orientations of the magnetizations instantaneously. At the same time the analysis of the temperature dependence of the critical field has shown that implicitly \( \Delta T_c \) of this system can be as large as 200 mK at \( \delta T_c \sim 100 \) mK.

Comparison of the results obtained for both proposed constructions of the spin switches gives grounds to sup-
pose that the scheme by Oh et al. may be the most promising for the realization of the full spin switch effect for the superconducting current in an S/F thin film heterostructure. In order to get a maximum spin switch effect we concentrated our efforts on the optimization of the materials’ choice and of the specific geometry of the F1/N/F2/S scheme by Oh et al. and have fabricated a set of samples which show a full switching between the superconducting and the normal state when changing the mutual orientation of the magnetizations of F1 and F2 layers.

For the layer sequence AFM/F1/N/F2/S to be deposited on the single crystalline MgO substrate (Fig. 1) we have chosen the following set of materials: cobalt oxide for the antiferromagnetic (AFM) layer that plays a role of the bias layer which pins the magnetization of the F1 layer; Fe for the ferromagnetic F1- and F2-layers; Cu as a normal metallic N-layer; and finally In for the S-layer. While the Fe/Cu/Fe trilayer as a ferromagnetic component looks ordinary, the choice of the S-component in our scheme is of primary importance. Considering that only few superconducting materials do not form a solid solution or intermetallic compounds with iron (they are Pb and In) and therefore do not form the otherwise unavoidable intermixed interface region we decided to concentrate on indium. The sample preparation was done by electron beam evaporation in an ultra-high vacuum chamber within a closed vacuum cycle. The base pressure was $2 \times 10^{-8}$ mbar. All deposition experiments were carried out on room-temperature substrates. The thickness of the growing films was measured by a quartz crystal monitor system. The Co oxide films were prepared by a two-step process consisting of the evaporation of a metallic Co film followed by the plasma oxidation converting Co into CoO$_x$ layer.

The residual resistivity ratio $RRR = R(300\text{K})/R(4\text{K})$ for all studied samples is similarly high for all studied samples (see Table 1) evidencing a high purity of the deposited In layers.

The indium film in our samples is a type I superconductor with parallel and perpendicular critical fields $H^\parallel_{\text{c}} \sim 220\text{ Oe}$ and $H^\perp_{\text{c}} \sim 20\text{ Oe}$, respectively, at $T = 2\text{ K}$. In view of this anisotropy we have taken care to avoid the appearance of the perpendicular component of an external field larger than 2 Oe. This means that we adjusted the sample plane position with an accuracy better than 2 degrees relative to the direction of the dc external field. The easy axis of the magnetization which is induced by residual magnetic fields in our vacuum system was directed parallel to the long axis of the sample. The parameters of the studied samples are shown in Table 1. Along with the spin switch samples #3 – 5 we prepared for control purposes an indium thin film sample (#1) and a reference sample comprising an indium layer and only one F-layer (#2R).

In our work we put emphasis on the experimental determination of both the hysteresis magnetization behavior and the current in-plane transport measurements, enabling a correlation between both properties.

In a first step the in-plane magnetic hysteresis loops of sample #3 in the direction of the magnetic field along the easy axis were measured by a SQUID magnetometer and is shown in Fig. 2a. This step is necessary to find out the Fe-layers’ magnetization behavior and to determine the magnetic field range where AP and P states can be achieved. The sample was cooled down in a magnetic field of +4 kOe applied parallel to the sample plane and measured at $T = 4\text{ K}$. The magnetic field was varied from +4 kOe to - 6 kOe and back again to the value of +4 kOe. Both limits correspond to the orientation of the magnetizations of the F1- and F2-layers parallel to the applied field. For the studied sample by decreasing the field from +4 kOe to the field value of the order of +50 Oe the magnetization of the free F2 layer starts to decrease. At the same time the magnetization of the F1-layer is kept by the bias CoO$_x$ layer until the magnetic field of -4 kOe is reached. Thus, in the field range between -0.3 and -3.5 kOe the mutual orientation of two F-layers is antiparallel. Below $H = -3.5\text{ kOe}$ the magnetization of the F1 layer starts to change its value and at the field of the order of -4.5 kOe magnetizations of both Fe layers become parallel. This corresponds to a further step-like decrease of the total magnetization. Qualitatively similar hysteresis loops were obtained for samples #4 and 5. The minor hysteresis loops on the low field scale obtained with decreasing the field from +4 kOe down to -1 kOe and increasing it again up to +1 kOe are shown in Figs. 2b, 2c and 2d for samples #3, 4 and 5, respectively. It is necessary to note that later when performing

| Sample | Layers’ thickness (nm) | $R_{RRR} \text{ Fe}$ | $R_{RRR} \text{ In}$ | $R_{RRR} \text{ CoO_x}$ | $\Delta T_c$ (mK) | $\beta T_c$ (mK) |
|--------|------------------------|-----------------------|---------------------|-----------------|-----------------|-----------------|
| 1      |                        |                       |                     |                 |                 |                 |
| 2R     | 0.5 230 15 4           | 0.5                   | 230                 | 15              | 0 ± 3           |                 |
| 3      | 4 2.4 4 0.5 230 47     | 4                     | 2.4                 | 0.5             | 7               | 19 ± 2         |
| 4      | 4 2.9 4 0.6 230 41     | 4                     | 2.9                 | 0.6             | 13              | 12 ± 3         |
| 5      | 4 2.6 4 2.6 230 44     | 4                     | 2.6                 | 2.6             | 50              | -2 ± 8         |

TABLE I: Experimental parameters of the studied samples.
the transport measurements, the magnetic field did not reach values beyond \pm 1 \text{kOe}.

For the transport study we used another system which also enables a very accurate control of the real magnetic field acting on the sample. This field was generated by a high homogeneous electromagnet. The magnetic field value was measured with an accuracy of \pm 0.3 Oe using a Hall probe. The temperature of the sample was monitored by the 230 \Omega Allen-Bradley resister thermometer which is particular sensitive in the temperature range of interest. Therefore the accuracy of the temperature control within the same measurement cycle below 2 K was better than \pm 2 \div 3 \text{mK}. In order to study the influence of the mutual orientation of the magnetizations on \(T_c\) we have cooled the samples down from room to a low temperature at a magnetic field of 4 kOe applied along the easy axis of the sample just as we did it when performing the SQUID magnetization measurements. For this field both F-layers’ magnetizations are aligned parallel (see the magnetic hysteresis loops in Fig. 2). Then at the in-plane magnetic field value of \(H_0 = \pm 110 \text{Oe}\) the temperature dependence of the resistivity \(R\) was recorded. In the following we focus on the spin valve sample \#3 (see Fig. 3). For this sample the difference in \(T_c\) for different magnetic field directions is clearly seen (see Fig. 3b with an enlarged temperature scale). The superconducting transition temperature for the AP orientation of the magnetizations occurs at a temperature exceeding the \(T_c\) for the P orientation of the sample by 19 mK. We also performed similar resistivity measurements of the reference sample \#2R with only one Fe layer (see Table 1). For this sample we found \(T_c=1.60 \text{K}\), which does not depend on the magnetic field direction (see Fig. 3c). This \(T_c\) value is lower than that for the In single layer film (sample \#1) and higher than for sample \#3 (Fig. 3). This means that \(T_c\) is suppressed by the F2 layer and in turn is sensitive to the influence of the F1 layer separated from the superconducting In layer by a 0.5 nm thick F2 Fe layer and 4 nm thick Cu layer. As can be expected from the the S/F proximity theory, with increasing the thickness of the free F2 layer \(\Delta T_c\) decreases and becomes practically zero for the 2.6 nm thick F2 layer (see Table 1).

The observed shift of the superconducting transition temperature \(\Delta T_c=19 \text{mK}\) is not the largest one among the data published before (cf., e.g., \(\Delta T_c \simeq 41 \text{mK}\) at \(\delta T_c \simeq 100 \text{mK}\) in Ref. [11]). However, it is very impor-
ducting transition width $\delta T_c$ is significantly larger than the superconducting transition width $T_c$, which is as small as $\sim 7$ mK for sample #3 at $H_0=110$ Oe. This opens a possibility to switch off and on the superconducting current flowing through our samples completely within the temperature range corresponding to the $T_c$-shift by changing the mutual orientation of magnetization of F1 and F2 layers. To demonstrate this we have performed the measurements of the resistivity of sample #3 by sweeping slowly the temperature within the $\Delta T_c$ interval and switching the magnetic field between $+110$ and $-110$ Oe. This central result of our study is shown in Fig. 4. It gives straightforward evidence for a complete on/off switching of the superconducting current flowing through the sample. To the best of our knowledge, this is the first ever example of the realization of a full spin switch design for a superconducting current in F/S structures with a perfect contact at each interface.

For sample #3 (as well as for sample #4) the obtained result is in agreement with the expectation based on the S/F proximity effect theory, namely that $T_c$ for the AP orientation of magnetizations is higher than $T_c$ for the P orientation. In the end it is not surprising, since all necessary prerequisites to realize the theoretical idea of Oh et al. [5] are fulfilled in this sample: (i) -the thickness of the F2-layer is smaller than the coherence length $\xi_F$; (ii) -owing to the absence of the intermixed region an atomically sharp highly transparent metallic interface between the S- and the F-layer has been achieved. The combination of (i) and (ii) makes a penetration of the Cooper pair wave function into the F2-layer and further into the N-layer possible. Finally, the high quality of the iron layers yields magnetization hysteresis curves with sharp well defined steps enabling a well controlled switching of the mutual orientation of the magnetization of the F-layers by application of relatively small magnetic fields. This is essential for the control of the decay of the Cooper pair wave function.

In summary, we have presented the first, to the best of our knowledge, experimental realization of the spin switch for the superconducting current in that we have achieved a complete switching on and off a superconducting current flowing through a sample by changing the mutual orientation of magnetizations of the F layers. Our results provide a compelling experimental confirmation of theoretical predictions by Oh et al. [5] for the spin switch device thereby suggesting the S/F proximity effect as the operation principle of our switch.

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* ilgiz.garifullin@yahoo.com

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