A Spin Valve Core Structure based on the Fulde-Ferrell Larkin-Ovchinnikov Like State: Studies on Bilayers and Trilayers of Superconductors and Ferromagnets

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Abstract. Interference effects of the superconducting pairing wave function in thin film bilayers of Nb as a superconductor (S) and Cu41Ni59 as ferromagnetic (F) material lead to critical temperature oscillations and reentrant superconductivity for increasing F-layer thickness. The phenomenon is generated by the Fulde-Ferrell Larkin-Ovchinnikov (FFLO) like state establishing in these geometries. So far detailed investigations were performed on S/F bilayers. Recently, we could also realize the phenomena in F/S bilayers where the S-metal now is grown on top of the F-material. Combining both building blocks yields an F/S/F trilayer, representing the core structure of the superconducting spin valve. Also for this geometry we observed deep critical temperature oscillations and reentrant superconductivity, which is the basis to obtain a large spin switching effect, i.e. a large shift in the critical temperature, if the relative orientation of the magnetizations of the F-layers is changed from parallel to antiparallel.

1. Introduction

The superconducting spin-valve consists of a superconducting thin film (S) sandwiched by two ferromagnetic layers (F). The magnetization direction of one F-layer is pinned by an antiferromagnet (AF) against the rotation of the magnetization direction in an external magnetic field. Theory predicts a dependence of the superconducting transition temperature, \( T_c \), on the relative magnetization direction of the F-layers in this AF-F/S/F structure \[1\]. For a parallel alignment of the magnetizations, \( T_c \) is lower than for the antiparallel one. Thus, one should be able to switch the superconducting state off and on by changing the magnetization direction of one of the layers relative to the other one.

The underlying physics is the S/F proximity effect, in which, contrary to the well-studied S/N case (with N a normal conducting, non-magnetic material), the superconducting pairing wave function does not simply decay into the ferromagnet, but oscillates in addition. The reason is that a quasi-one dimensional Fulde-Ferrell Larkin-Ovchinnikov (FFLO) \[2,3\] like state is generated in the ferromagnetic layer \[4,5\].

While the FFLO state in bulk material is restricted to a very narrow range of extreme parameters \[7\] and is hard to realize \[8,9\], it is induced from the superconductor into the ferromagnetic film in layers of superconducting and ferromagnetic material. In this state, the Cooper pairing occurs by combining electrons with antiparallel spin as in the BCS theory \[10\]. However, their momenta, although being in opposite directions, do not have the same absolute value. This yields the oscillating pairing wave function mentioned above.

The oscillation of the superconducting pairing wave function leads to interference phenomena, if the thickness of the F-material has a limited value and the superconducting pairing wave function is reflected at the outer border of the F-material of a (e.g.) F/S bilayer \[11\]. Depending on the thickness of the F-layer this interference may be constructive or destructive. Thus, by acting back on the S-layer,
the critical temperature oscillates as a function of the thickness $d_F$ of the F-layer for a given thickness $d_S$ of the S-layer. If $d_S$ is thin enough, even an extinction of the superconducting state with a subsequent recovery, i.e. a reentrant superconducting state, is predicted [12].

Starting with Nb/Ni bilayers [13], we studied the phenomenon in detail experimentally on F/S (Nb/Cu$_{41}$Ni$_{59}$) [14,15] and S/F (Cu$_{41}$Ni$_{59}$/Nb) [16] bilayers, where the sequence of film growth is different, and F/S/F Cu$_{41}$Ni$_{59}$/Nb/Cu$_{41}$Ni$_{59}$ trilayers [17]. The later form the core of the superconducting spin valve. In all cases dealing with Cu$_{41}$Ni$_{59}$ alloy as F-metal, deep critical temperature oscillations were observed. Moreover, also the reentrant superconducting state could be observed. These are the key conditions for a functioning spin valve with a large $T_c$ shift [15]. In the present paper we summarize the most important results of these investigations.

2. Sample Preparation and Characterization

All sample series discussed in the present work were fabricated using our wedge technique described in detail in Refs. [13-17]. First, the Si(111) substrate is covered by an amorphous Si buffer layer. In the case of an S/F bilayer, then the S-material (Nb) is deposited by dc magnetron sputtering, applying a full power operating magnetron, which is moved by a dc motor drive along the 80x7mm$^2$ substrate, thus, spraying it homogenously with the S-material. Next, utilizing the intrinsic gradient of the sputtering rate by placing the substrate out-of the symmetry axis of the sputtering characteristics, a wedge shaped F-metal (Cu$_{41}$Ni$_{59}$) layer is deposited by rf magnetron sputtering. Finally, the F-layer is covered by a thin silicon cap layer, to protect the wedge like basis sample against a degradation at atmospheric conditions. Then, a series of samples is cut perpendicular to the thickness gradient of the wedge.

To get F/S bilayers, the S-layer is grown on top of the wedge type F-layer, which means different growth conditions compared to the deposition of S/F bilayers, described above. In the case of F/S/F trilayers, two different types of sample series were produced, namely the single wedge geometry, with a flat bottom F-layer and a wedge type top F-layer (see sample FSF1 No. 5 in Fig. 1, for critical temperature measurements see Ref. [17]) and the double wedge geometry, with both F-layers being wedge-like. In all cases the S-layer has a constant thickness. For S/F and F/S bilayers, we also investigated a different type of samples, where an S-wedge is covered by a physically infinite F-layer of constant thickness, to get the critical thickness of the superconducting material below which superconductivity is suppressed [15,16].

Fig. 1: Cross sectional TEM images of an F/S (Cu$_{41}$Ni$_{59}$/Nb) bilayer (left) and an F/S/F (Cu$_{41}$Ni$_{59}$/Nb/Cu$_{41}$Ni$_{59}$) single wedge geometry trilayer (right).
The determination of the thickness of the different layers was performed by Rutherford Backscattering Spectrometry (RBS). Moreover, for S/F and F/S bilayer samples also of the alloy content of the F-layer was evaluated by this method [13-16]. Cross sectional Transmission Electron Microscope (TEM) images of F/S and F/S/F specimens are shown in Fig. 1, demonstrating the perfect smooth boundaries between the layers. A detailed analysis of the High Resolution Electron Microscopy (HRTEM) images allows getting some knowledge about the growth direction of the layers [16]. More detailed information is obtained from electron diffraction patterns performed in the TEM [17].

3. Superconducting Properties

The S/F and F/S bilayers are the building blocks of the F/S/F trilayer, forming the core structure of the AF-F/S/F spin valve. This core structure may be regarded as a mirror symmetric F/S-S/F arrangement. This means, that the thickness of the S-material in the trilayer is twice compared to the respective bilayers.

Results of $T_c(d_F)$ curves for different fixed thicknesses of the S-material are shown in Fig. 2. In all cases deep critical temperature oscillations could be realized. For sufficiently thin S-layers, also the reentrant, and moreover, evidence for a multi-reentrant superconducting state, is observed. The curves are described by the theory [12], elaborated for the different types of sample series and parameterized for the use of measurable physical parameters [13,15,17].

**Table**

| Series | $T_{c_{0,Nb}}$ (K) | $\xi_0$ (nm) | $N_x y_x$ | $N_z y_z$ | $T_F$ | $l_x/\xi_{10}$ | $\xi_{10}$ (nm) |
|--------|--------------------|--------------|----------|----------|------|----------------|----------------|
| S22    | 6.85               | 6.5          | 0.22     | 0.61     | 1.1  | 10.7           |
| S21    | 6.2                | 6.1          | 0.22     | 0.85     | 1.1  | 11.2           |
| FS233  | 6.6                | 6.0          | 0.19     | 0.81     | 0.6  | 11.6           |
| FS2406 | 6.1                | 6.0          | 0.2      | 0.88     | 0.6  | 10.4           |
| FS3    | 8.0                | 7.0          | 0.22     | 0.68     | 0.68 | 10.8           |
| FS5    | 7.8                | 6.4          | 0.21     | 0.83     | 1.0  | 12.8           |

The $T_c(d_{CuNi})$ denotes the sum of the thicknesses of the bottom (B) and top (T) layers, $d_{CuNi} = d_{CuNi-B} + d_{CuNi-T}$. The Table summarizes the fitting parameters used for the theoretical curves, as defined in the text. For samples FSF3 and FSF5, the upper rows refer to the top ferromagnetic layer whereas the lower ones to the bottom F-layer.

**Fig. 2:** Critical temperature of S/F and F/S bilayers and F/S/F trilayers (S=Nb, F= Cu$_{41}$Ni$_{59}$) as a function of the F-layer thickness for different constant S-layer thicknesses $d_S$, according to Refs. [15-17]. In the case of F/S/F trilayers $d_{CuNi}$ denotes the sum of the thicknesses of the bottom (B) and top (T) layers, $d_{CuNi} = d_{CuNi-B} + d_{CuNi-T}$. The Table summarizes the fitting parameters used for the theoretical curves, as defined in the text. For samples FSF3 and FSF5, the upper rows refer to the top ferromagnetic layer whereas the lower ones to the bottom F-layer.
The fit parameters of the theory are: $\xi_S$, the superconducting coherence length in the S-metal, as defined e.g. in Eq. 1 of [15]; $\xi_{FS}$, the coherence length for Cooper pairs in a F-metal; $l_F$, the mean free path of conduction electrons in a F-material; $N_{cF}/N_{cS}$, the ratio of Sharvin conductances at the S/F interface, and $T_f$, the interface transparency parameter. Moreover, $T_{c0,Nb}$ is the critical temperature of a free standing Nb film of the given thickness, according to Fig. 5 of Ref. [15]. Furthermore, $\xi_{BCS} = 42$ nm was used for the BCS coherence length [18]. The fit parameters are of similar size, except $l_F/\xi_{FS}$, which is especially large for the S/F bilayers and for the top layer of sample series FSF5. Since $\xi_{FS}$ is similar for all samples, this means that $l_F$ is larger for these samples, where the F-film is grown on top the S-layer.

3. Conclusion
So far, the building blocks (i.e., S/F and F/S bilayers) of the superconducting spin valve core could be realized and even the F/S/F core structure itself. All of them show the required critical temperature dependence on the F-material layer thickness to get a functioning AF-F/S/F spin valve.

The next step is to realize a sufficient exchange bias between the bottom F-layer and the AF-sublayer. Then the spin valve is expected to exhibit critical temperature shifts in the Kelvin range if the magnetization direction of the top layer is rotated into the opposite direction of that of the bottom layer, as already theoretically calculated [15].

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