Nano-structures Based on Superconducting Nb and Ferromagnetic CuNi Alloy for Elaboration of Spin-valve Core

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***Abstract* — The main goal of our research group is the elaboration of superconducting spin-switch (valve) based on Ferromagnetic/Superconductor/Ferromagnetic core. We could realize all building blocks necessary for the fabrication of the core structure of the superconducting spin valve, consisting of two mirror symmetric bilayers [4]. In other words, the spin valve consists of a F/Š/F trilayer, which can be regarded as a package of a F/S and S/F bilayer so that Š=2S in the trilayer [16, 17]. For such a trilayer, the theory [4] predicts that the critical temperature depends on the relative orientation of the magnetization of the ferromagnetic layers. To enable a reversal of one of the magnetizations of the layers with respect to the other by an external magnetic field, the coercive forces of the F layers have to be different due to either intrinsic properties or to an antiferromagnetic pinning layer delivering an exchange bias. The main points of our study are presented here.**

***Index Terms* — hetero-structures, nano-structures, nonuniform LOFF-superconductivity, spin valve, ultra-thin films.**

I. INTRODUCTION

The interplay of mutual antagonist states of matter – singlet superconductivity and ferromagnetism is in the focus of experimental and theoretical investigations for last decades [1, 2, 3] The exotic quasi- one dimensional Fulde-Ferrell-Larkin-Ovchinnikov (FFLO) state can be realized in artificially layered S/F nanostructures, i.e. in S/F layers with film thickness in the nanometer range. It results in many new effects promising for application [3]. Superconducting spin valve effect [4] having a prominent potential for implementations to commercial cryogenic devices attracts special attention of the experimentalists for recent decade [5, 6, 7, 8]. However, the scale of the effect is still not enough for the device implementation.

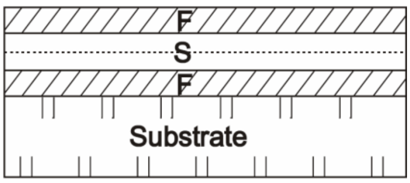

Figure 1 The core of superconducting spin-valve device-Ferromagnet/Superconductor/Ferromagnet

Fig.1 demonstrates the core of spin switch structure (F/S/F) consisting of 2 bilayers – F/S and S/F from both theoretical [9, 10, 11] and technological point of view. In metallic F/S/F trilayers the overall Tc is reduced by the usual proximity effect with the adjacent ferromagnetic layers [12], but the amount of reduction depends on the relative magnetization orientation in the F layers. This magnetization orientation dependence occurs when the Cooper pair size ξS is comparable with or smaller than the thickness of the superconducting layer, so that the pairs are influenced by both F layers simultaneously. The strong correlation between the scale of superconducting critical temperature (Tc) oscillations from the thickness of F layer (dF) in F/S/F structure {Tc(dF)} and the scale of spin-valve effect was mentioned in [4, 9, 13]. The most spectacular oscillatory effect predicted by the theory for quite some time, the re-entrance of the superconducting state, could be detected convincingly only very recently for S/F bilayers[14]. Although the sequence of the layers S/F and F/S is considered by existing theoretical models as equal [4, 9], it has not been experimentally confirmed. The growth conditions, being rather different for nano-layers formed on the substrate or on one of the layers, could affect superconducting (ferromagnetic) properties of the layers. The first prime aim of the project is to detect large scale oscillation of Tc from dF for F/S bilayers fabricated at the same technological conditions and for the same material as S/F bilayers in our preliminary work [14]. The combination of the technological parameters used for fabrication of F/S and S/F bilayers with the largest scale of Tc from dF oscillation should possess to apply knowledge, collected from bilayers, to Tc oscillations in F/S/F trilayers to build a core of superconducting spin-valve device structure. The other important aim is the study of the influence of an external magnetic field on the Tc oscillations and the re-entrance phenomenon. These investigations will result not only in the knowledge of optimized parameters to demonstrate the Tc-oscillation phenomenon in F/S structures and to drive the F/S/F spin-valve, but also in a deep understanding of the concurrence of exchange and thermal energies and their importance in a theoretical description of the observed phenomena.

II. S/F TYPE SAMPLE

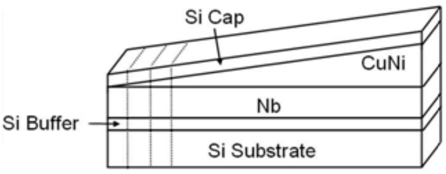

Figure 2. A sketch of S(const)/F(wedge) structure.

Figure 2 shows our wedge technique to get samples of different thickness prepared in the same run, i.e. with the same surface and boundary properties. For this purpose, first, a superconducting layer of constant thickness (here Nb), then the wedge, i.e. a film of steadily increasing thickness of the F-material (here CuNi), are deposited. The wedge is obtained utilizing the off-symmetry mounting of the long substrate (70-80 mm) and the intrinsic spatial gradient of the deposition rate of the magnetron sputtering setup. The whole Nb/CuNi structure is wrapped by amorphous silicon. On one side the amorphous silicon protects the structure from oxidation in the atmosphere, on the other side protects from absorbed gases and contaminations on silicon substrate. Cutting of the S/F-wedge into stripes results in samples with constant S but different F-layer thickness, which are then separately measured to determine their critical temperature.

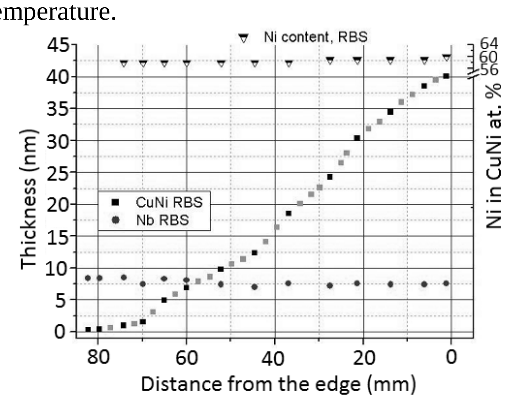

Figure 3. The graph shows a linearly increasing thickness of the copper nickel layer from around 1 to 40 nm in S/F(wedge) sample. The niobium thickness is around 7.8 nm and nearly constant. At the top of the graph is the atomic nickel content in the CuNi alloy.

In Figure 3 the thickness of the S and F layers resulting from RBS investigations are plotted, demonstrating constant S and steadily increasing F-layer thickness for the series of specimens investigated. At the top of the graph is the atomic nickel content in the CuNi alloy. There is a slight variation as discussed in the sample preparation section. It's important to note thanks to improving of the sample preparation technique we could adjust the thickness control to 0.2 nm and achieved a superconducting critical temperature of the Nb layer as high as 6K for a single Nb layer of thickness ~7 nm; and the RF magnetron sputtering (but not DC which is commonly applied for metallic targets) with carefully adjusted power has been used to deposit a Cu1-xNix alloy wedge, with minimal deviation of the deposited layer composition from the target one (it can be seen at the top of the graph, fig. 3). With this improved sample preparation technique we deposited Nb films at thicknesses close to the critical one (see shadowed area at Fig. 4), and choosing x=0.59, we, recently, could not only increase the amplitude of the Tc oscillations to a magnitude never reported before, but also presented experiments which for the first time convincingly demonstrate the re-entrant phenomenon of Tc as a function of the F-layer thickness [16], data are given in Fig. 5. All six solid curves in Fig. 4, Fig. 5a and Fig. 5b have been fitted simultaneously, except choosing slightly different values of the superconducting coherence length for the S15, S16, S21, S22 and S23 sample series. For Fig. 4 the same parameters as for S15 were used.

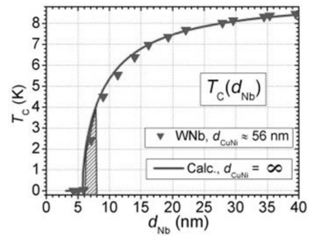

Figure 4 allowed detecting a value of critic thickness of Nb-layer in case of electric contact with massive ferromag-netic CuNi alloy (56 nm), the solid curve is the theoretical calculations Lenar Tagirov.

The oscillation of the critical temperature measured for the S/F couple with S=Nb and F=CuNi, which we used in our first investigations, are clearly visible and well described by the Tagirov's theory (Fig. 5). For sample series with fixed dS= 14.1 nm, the Tc(dF) dependence only shows a slight suppression with a shallow minimum. A reduction of dS (to 8.3 nm and 7.8 nm) yields Tc(dF) curves with a strong suppression of superconductivity and a deep minimum. For even lower value of dS = 7.3 nm superconductivity vanishes for a certain range of dF and then restores again, i.e. reentrant superconducting behavior is observed and for value of dS= 6.2 nm for the first time the periodic-reentrant superconducting behavior phenomenon was detected [15], predicted by the theory of quasi-LOFF state.

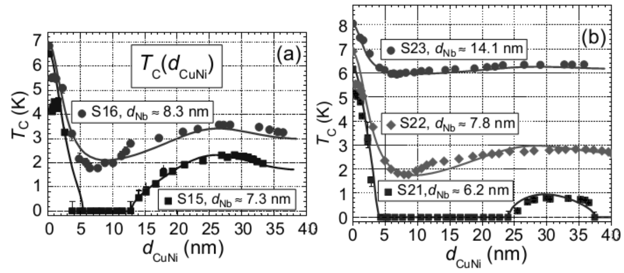

Figure 5 demonstrates the dependence of the superconducting transition temperature on the thickness of the $Cu_{41}Ni_{59}$ layer for five values of the Nb layer thickness, where dNb(S15) ≈7.3 nm, dNb(S16) ≈8.3 nm, dNb(S21) ≈6.2 nm, dNb(S22) ≈7.8 nm, and dNb(S23) ≈14.1 nm.

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III. F/S TYPE SAMPLES

The S/F bilayers may be regarded as the first building block of the superconducting spin valve. The next non-trivial problem on the way to the fabrication of the spin valve is the deposition of high quality Ferromagnet/Superconductor bilayers, where now the ferromagnet has to be grown on the Si substrate and the superconductor on top of the F material. As in previous cases to fabricate a series of F/S samples with variable layer thickness at the same run, with identical deposition conditions for all samples in the series, we applied our wedge technique described in detail [16, 17]. Immediately after, an ultra-flat superconducting niobium layer of constant thickness is deposited, making use of our custom, rotating target technique. Afterwards, the Cu41Ni59-wedge/Nb bilayer was coated by a thin amorphous Si cap-layer to prevent degradation at ambient conditions (see fig. 6).

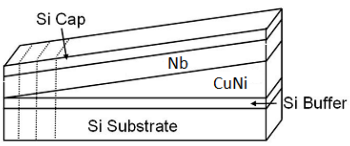

Figure 6. A sketch of F (wedge)/S(const) structure.

As a previous case for determination of the thickness of the Cu1-xNix and Nb nanolayers of the samples, Rutherford Backscattering Spectrometry (RBS) was applied. The results of such evaluation for series FS233 is shown in Fig. 7. The thickness of the niobium film is nearly constant at a value of dNb(FS 233) = 7.5 nm (-0.4 nm +0.3 nm). The Cu1-xNix layer thickness decreases from 32 nm to 1 nm. The nickel concentration in the Cu1-xNix alloy varies by +1 at.% to -2 at.% around the average value of 58 at.% with a slight decrease of nickel content towards the thin end of the wedge.

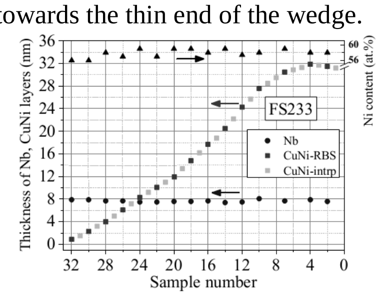

Figure 7. Results of the RBS measurements for the thickness of the $Cu_{1-x}Ni_x$ and Nb layers, respectively, together with the nickel content, x.

The resistance measurements of the F/S bilayers were performed like for S/F type of samples. In Fig. 8 the dependence of the superconducting transition temperature on the thickness dCuNi of the ferromagnetic alloy is shown for different fixed thicknesses dNb of the superconducting layer. For dNb= 7.5 nm (sample series FS233) the critical temperature Tc shows a non-monotonic behavior, beginning with a decrease, then a minimum is reached, and subsequently the critical temperature rises again. If the thickness of the flat superconducting layer is further decreased to dNb= 6.8 nm a quite unusual behavior is observed (sample series FS2406). For increasing dCuNi the critical temperature initially steeply drops towards zero at dCuNi ≈4.0 nm, i.e. the superconductivity is fully suppressed. After a further increase of dCuNi to a value of about 17.4 nm the superconducting state recovers. This is a reentrant behavior of the superconducting state predicted by theory [10]. In sample FS712, dNb is further reduced to an average value about 6.3 nm. It is close to the critical value dNbcr ~6.2 nm below which the superconductivity of Nb film is fully suppressed if the Cu41Ni59 alloy layer is sufficiently thick. As a result, upon increasing dCuNi, the critical temperature Tc steeply drops to zero and remains vanishing for any dCuNi ≥4 nm.

Our results for F/S bilayers have been achieved for different thicknesses of the functional layers compared with the S/F bilayers.

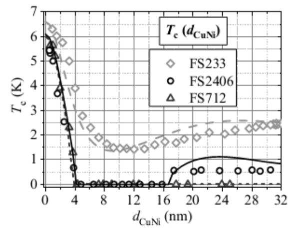

Figure 8. Critical temperature Tc of $Nb/Cu_{41}Ni_{59}$ bilayers as a function the thickness dCuNi.

IV. F/S/F –TYPE SAMPLES

The next big breakthrough was the combining of two shapes S/F and F/S type in single F/S/F sample [18] – the core structure of superconducting spin valve.

Applying our wedge technique, described in detail earlier, we fabricated two types of sample series (Fig. 9). In the first one, (a "single wedge" geometry, see Fig. 9 a)) the bottom F-layer ($Cu_{41}Ni_{59}$ grown on the Si buffer layer) has a constant thickness as well as the subsequently grown S-layer. The F-layer on top has a wedge like shape. The second type ("double wedge" geometry, see Fig. 9 b)) of sample series consists of two $Cu_{41}Ni_{59}$ wedges separated by a Nb layer of constant thickness.

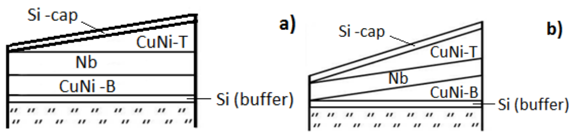

Figure 9. The F/S/F geometry consists of two copper-nickel layers which enclose a niobium film: a) single wedge geometry: Bottom $Cu_{41}Ni_{59}$ layer ("CuNi-B") kept at a constant thickness. Top $Cu_{41}Ni_{59}$ layer ("CuNi-T") wedge-type. b) Double wedge geometry: Both $Cu_{41}Ni_{59}$ layers are wedges. The symmetric situation is drawn, where the slope of both wedges is the same. In both geometries the thickness of the Nb layer is constant.

How in previous cases – for composition and thickness analyses the RBS was performed. It's important to note – the structures comprise two practically identical CuNi layers, and the technology of measurement and treatment of dates was slightly modified and combined with TEM data (for a little more details see [18]).

In Fig. 10 a) the results of a RBS evaluation are shown for the single wedge geometry (sample series FSF1). The thickness of the layers (CuNi-Top, CuNi-Bottom and Nb) has been plotted as a function of the sample number, i.e. the distance from the thick end of the $Cu_{41}Ni_{59}$ alloy wedge. The bottom CuNi alloy film as well as the Nb film have nearly constant thickness. In Fig. 10 b) a similar representation is shown for the double wedge geometry. Again, the Nb film has a constant thickness, whereas the thickness of the CuNi alloy wedges change continuously along the sample series. It is remarkable that for a given sample the thickness of both CuNi alloy films is always nearly equal.

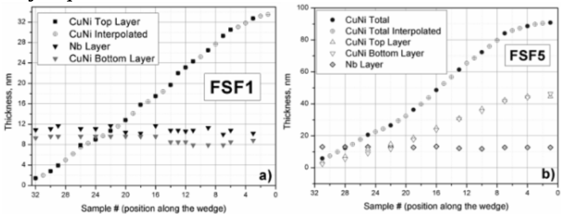

Figure 10. Results of the RBS measurements for the thickness of the two $Cu_{1-x}Ni_x$ and Nb layers between them, respectively. Some of the data points of the $Cu_{41}Ni_{59}$ are interpolated values between two RBS measurements; a) - Single wedge sample series FSF1; b) Double wedge sample series FSF5.

For both types of sample series for a larger thickness of the S-layer an oscillation of the critical temperature is observed. Reducing the thickness of the Nb layer, then yields a reentrant superconducting behavior (see Fig. 11).

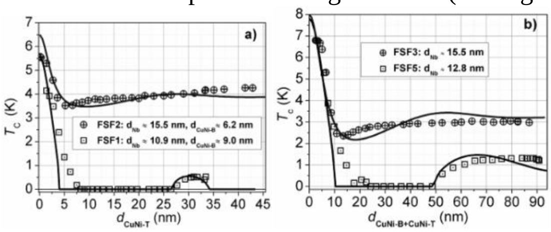

Figure 11. Transition temperatures, Tc, of the investigated $Cu_{41}Ni_{59}$/Nb/$Cu_{41}Ni_{59}$ trilayer samples as a function of the thickness dCuNi of the $Cu_{41}Ni_{59}$ alloy layers. a) Single wedge sample series FSF1 and FSF2. Critical temperature as a function of the increasing thickness of the top layer dCuNi-T. b) Double wedge series FSF3 and FSF5 plotted as a function of the sum of the thickness of the bottom and top layer, dCuNi-B+dCuNi-T.

For samples FSF3 and FSF5 with a double wedge geometry a comparison with results obtained on S/F and F/S bilayers is possible, if we divide the Nb thickness by a factor of two. Since in average dNb= 15.5 nm and 12.8 nm for sample FSF3 and FSF5, respectively, we have to compare the behavior with bilayers having a Nb layer thickness of 7.8 nm and 6.4 nm. For S/F and F/S bilayers indeed an oscillation of Tc is observed for samples with a niobium thickness of about 8 nm, whereas the Nb thickness of about 6 nm leads to reentrant superconducting behavior [15-18]. Moreover, also the range of dCuNi, AV =(1/2)(dCuNi-B + dCuNi-T) in which superconductivity vanishes in the reentrant case of sample series FSF5, is comparable to S/F and F/S bilayers. In the case of sample series FSF3 the minimum of the critical temperature is close to dCuNi, AV=6.8 nm, in agreement with S/F bilayers [16].

For the single wedge sample series FSF1 and FSF2 a direct comparison with bilayer measurements is only possible for the CuNi alloy thickness of the upper layer which is equal to the constant lower layer. For FSF1 and FSF2 this thickness is 9 nm and 6.2 nm, respectively, while dNb=10.9 nm and 15.5 nm, yielding 5.5 nm and 7.7 nm, if dividing by two. For the FSF1 series the critical temperature is zero for dCuNi-T=9 nm of the top layer, because the sample series shows reentrant behavior. For the FSF2 series there is a minimum of the critical temperature at dCuNi-T =6.2 nm. This sample series shows an oscillation of the critical temperature. In spite of the thickness of the Nb layers these observations coincide with the behavior of S/F bilayers [16].

V. CONCLUSION

These experiments represent an important step towards the superconducting spin valve, which needs a Ferromagnet/Superconductor/Ferromagnet core structure with an oscillatory or, more optimal, a reentrant superconducting behavior to realize the theoretically predicted spin switch effect.

Thus, we now have elaborated and successfully applied the basic experimental technique, necessary to start the proposed investigations. Together with the theoretical calculations of L. Tagirov and A. Buzdin, this opens the possibility to come to a deep understanding of the FFLO-like state in S/F layered systems.

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