ISSN 2072-5981

Magnetic Resonance in Solids

Electronic Journal

Volume ***16,***
Issue ***2***
Paper No ***14208,***
1-14 pages
2014

http://mrsej.kpfu.ru
http://mrsej.ksu.ru

Established and published by Kazan University
Sponsored by International Society of Magnetic Resonance (ISMAR)
Registered by Russian Federation Committee on Press, August 2, 1996
First Issue was appeared at July 25, 1997

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* In Kazan University the Electron Paramagnetic Resonance (EPR) was discovered by Zavoisky E.K. in 1944.

The peculiarities of the operation of the superconducting spin valve†

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(Received: February 28, 2014; accepted: April 19, 2014)

Review of our recent results concerning the spin valve effect are presented. Using the spin switch design F1/F2/S proposed theoretically that comprises a ferromagnetic bilayer (F1/F2) as a ferromagnetic component, and an ordinary superconductor (S) as the second interface component, we have realized for the first time a full spin switch effect for the superconducting current. For CoO_x/Fe1/Cu/Fe2/In multilayered systems with varying Fe2-layer thickness we observed the sign-changing oscillating behavior of the spin valve effect $\Delta T_c = T_c^{\mathrm{AP}} - T_c^{\mathrm{P}}$ (here T_c^{AP} and T_c^{P} are the superconducting transition temperatures for antiparallel and parallel orientations of magnetizations of the F1 and F2 layers, respectively). We have also studied the angular dependence of T_c for the spin valve system CoO_x/Fe1/Cu/Fe2/Pb. We found that this dependence is nonmonotonic when passing from the parallel to the antiparallel case of mutual orientation of magnetizations of the Fe1 and Fe2 layers and reveals a distinct minimum near the orthogonal configuration. The analysis of the data in the framework of the superconducting triplet spin valve theory gives direct evidence for the long-range triplet superconductivity arising due to noncollinearity of the two magnetizations.

PACS: 74.45+c, 74.25.Nf, 74.78.Fk

Keywords: superconductor, ferromagnet, proximity effect

1. Introduction

During the last decades the growing attention to the ideas and experiments concerning the development of elements of superconducting spintronics is clearly seen (see, e.g. [1, 2]). In these works, in particular, the interest in the superconductor/ferromagnet/superconductor (S/F/S) heterostructures as possible elements of quantum logics was emphasized [3]. An element of a qubit [4, 5] is based on the Josephson π-contact [6, 7]. Under certain conditions such contact can be implemented in the S/F/S thin-film heterostructure. The S/F contact itself is of long-standing fundamental interest (see, e.g., [8]).

The antagonism of superconductivity and ferromagnetism 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

†This paper is originally written by authors on the occasion of eightieth birthday of Professor Boris I. Kochelaev.

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 ξ_F. For pure Fe the value of ξ_F is less than 1 nm (see, e.g., [9]). All details of the S/F proximity effect are well described in reviews [10–13]. One can see from these reviews that many questions concerning physics of the S/F proximity are already clarified. Nevertheless, some theoretical predictions still need to be confirmed. First, that is the implementation of the full spin valve effect based on the S/F proximity effect. The results we present in this paper may be considered as the first positive example in this field. Second, this is the long-range triplet component in the superconducting condensate which should be generated in the S/F systems and exist in the presence of the singlet suiperconductivity only. All experiments indicate the existence of the triplet superconductivity in the S/F/S systems. Nevertheless, the origin of the generation of the triplet superconductivity is not always confirmed by experiments.

The physical origin of the spin switching based on the S/F proximity effect relies on the idea to control the pair-breaking, and hence the superconducting (SC) 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. The possibility to develop a real switch based on the S/F proximity effect has been theoretically substantiated in 1997 by Sanjiun Oh *et al.* [14]. They proposed the F1/F2/S layer scheme where an S-film is deposited on top of two F-layers. The thickness of F2 should be smaller than ξ_F to allow the superconducting pair wave function to penetrate into the space between F1- and F2-layers. Two years later a different construction based on an F/S/F trilayer was proposed theoretically by Tagirov [15] and Buzdin *et al.* [16, 17]. 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., [18–21]). However, the difference in T_c between the AP and P orientations $\Delta T_c = T_c^{\mathrm{AP}} - T_c^{\mathrm{P}}$ (here T_c^{AP} and T_c^{P} are the superconducting transition temperatures for antiparallel and parallel orientations of magnetizations of the F1 and F2 layers, respectively) turns out to be smaller than the width of the superconducting transition δT_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.* [14] with a $[\mathrm{Fe/V}]_n$ antiferromagnetically coupled superlattice instead of a single F1/N/F2 trilayer [22, 23] is not actually the spin switch device because the system can not 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 ΔT_c can reach up to 200 mK at the superconducting transition width $\delta T_c \sim 100$ mK. Comparison of the results obtained for both proposed constructions of the spin switches gives grounds to suppose that the scheme by Oh *et al.* may be more promising for the realization of the full spin switch effect. Later on a set of asymmetric construction was proposed [24–26]. It is necessary to note that they are not still experimentally tested.

Recently [27] it was shown that precise analysis of the processes taking place in the course of the penetration of the Cooper pair from the S- into the F-layer predicts the generation of the triplet component in the superconducting condensate in the F-layer. Within the homogeneous ferromagnet such a component has zero spin of the Cooper pair ($S_z = 0$). It is certainly can not be experimentally detected. At the same time the magnetic inhomogeneity leads to $S_z = \pm 1$ [27]. In this case the triplet component can be indicated through the anomalously deep penetration of the superconducting condensate into the ferromagnet. This component should manifest itself in the systems with noncollinear orientation of magnetizations in the F-layer [28] as well as in the systems with spatial or momentum dependence of the exchange field [29]. The series of the experiments was performed (see, e.g., reviews [12, 13, 30]) which shows the anomalous deep penetration of the superconducting condensate into the F-layer typical for the triplet superconductivity.

In the following we review the results of our recent studies of the spin valve effect [31–35].

2. Results and discussion

2.1. Full spin valve for the superconducting current in a superconductor/ferromagnet thin film heterostructure

We have fabricated a set of samples MgO(001)/CoO_x/Fe1/Cu/Fe2/In which show a full switching between the SC and normal states when changing the mutual orientation of the magnetizations of F1 and F2 layers. In this construction MgO(001) is a high quality single crystalline substrate, cobalt oxide antiferromagnetic (AFM) layer plays a role of the bias layer which pins the magnetization of the F1 layer; Fe stands for the ferromagnetic F1- and F2-layers; Cu as a normal metallic N-layer which decouples the magnetizations of F1- and F2-layers; finally In is an S-layer.

The sample preparation was done by electron beam evaporation on room temperature substrates at the base pressure $2 \cdot 10^{-8}$ mbar. 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.

Before starting the measurements of the superconducting transition temperature at different mutual orientation of magnetizations of the F-layers the in-plane magnetic hysteresis loops of sample #3 in the direction of the magnetic field along the easy axis was measured by a SQUID magnetometer and is shown in Fig. 1a. This step is necessary to obtain the Fe-layers' magnetization behavior and to determine the magnetic field range where AP and P states can be achieved.

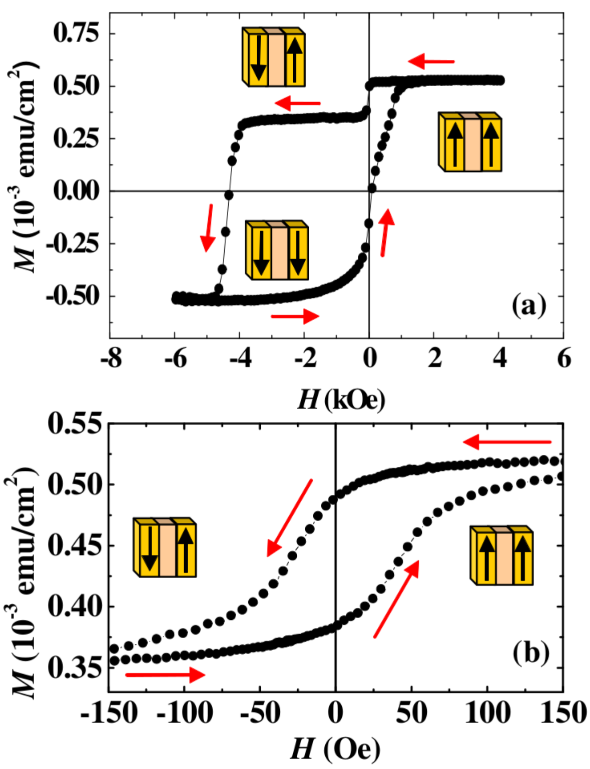

Figure 1. (Color online) (a) Magnetic hysteresis loop for sample #3 CoO_x(4 nm)/Fe1(2.4 nm)/Cu(4 nm)/Fe2(0.5 nm)/In(230 nm). Panel (b) shows part of the minor hysteresis loop for sample #3, obtained when decreasing the magnetic field from +4 kOe down to −1 kOe and increasing it up to +1 kOe. The amplitude of the minor hysteresis loops is proportional to the thickness of the free F2 layer. Coercive and saturation fields are the largest for the sample #3 and sharply decrease with increasing d_{Fe2}. [31, 33]

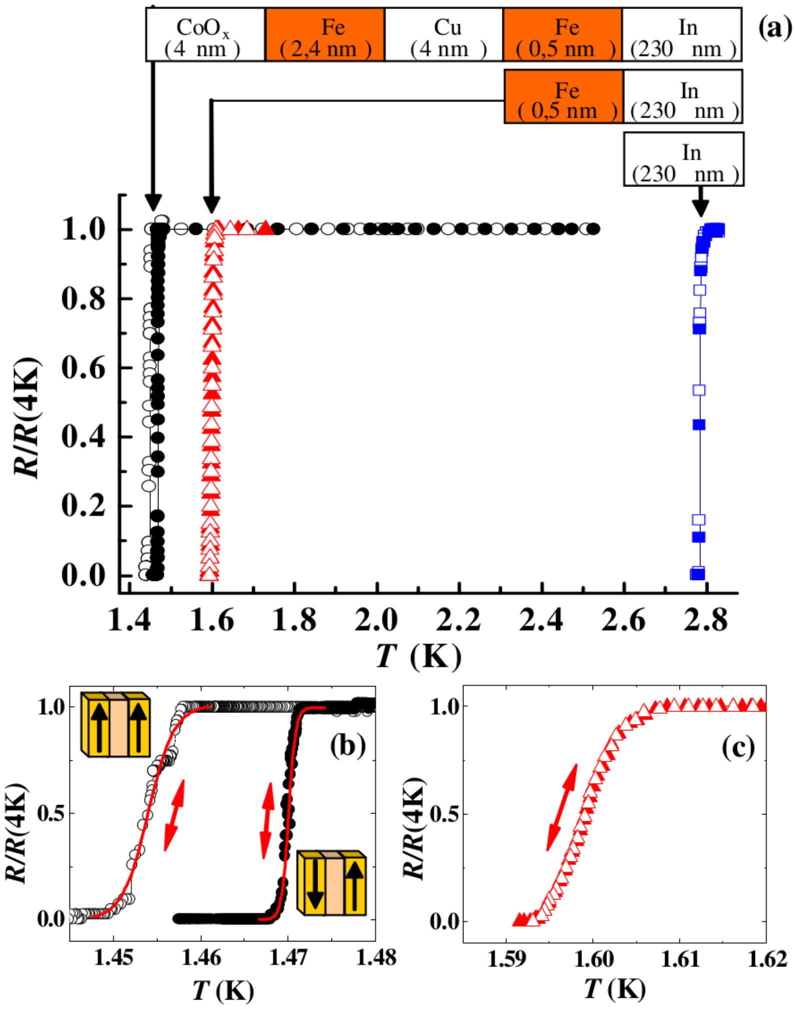

Figure 2. (Color online) (a) Overview of the resistivity transition curves. The spin valve sample #3 CoO_x(4 nm)/Fe1(2.4 nm)/Cu(4 nm)/Fe2(0.5 nm)/In(230 nm) is shown by black open ($H_0 = +110$ Oe) and closed ($H_0 = -110$ Oe) circles (see also panel (b) for detailed view). For the reference sample #2R Fe2(0.5 nm)/In(230 nm) the data are depicted by red open ($H_0 = +110$ Oe) and closed ($H_0 = -110$ Oe) triangles (see also panel (c) for detailed view). For the pure In sample #1 In(230 nm) the data are presented by blue open ($H_0 = +110$ Oe) and closed ($H_0 = -110$ Oe) squares [31].

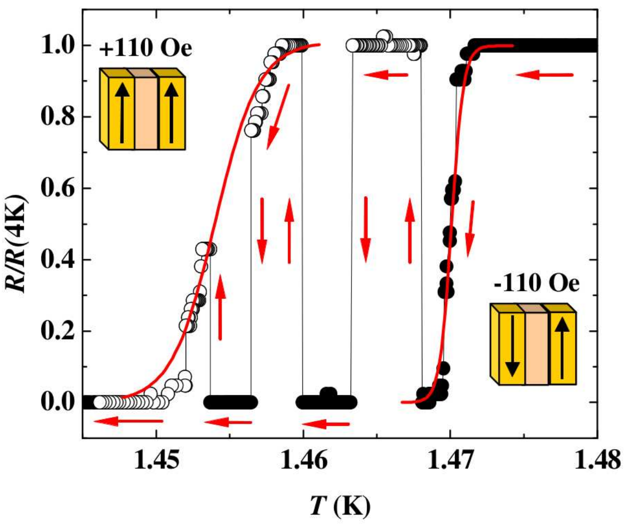

Figure 3. (Color online) Switching between normal and superconducting states in the spin valve sample #3 during a slow temperature sweep by applying the magnetic field $H_0 = -110$ Oe (closed circles) and $H_0 = +110$ Oe (opened circles) in the sample plane [32, 33].

The sample was cooled down in a magnetic field of +4 kOe applied parallel to the sample plane and measured at $T = 4\,\mathrm{K}$. Bearing in mind that the Néel temperature of the cobalt oxide is of the order of 250 K, after such cooling procedure the magnetization of the Fe1-layer turns out to be pinned by the exchange field of the AFM-layer. 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. The minor hysteresis loops on the low field scale were obtained with decreasing the field from +4 kOe down to −1 kOe and increasing it again up to +1 kOe. An exemplary loop for sample #3 is shown in Fig. 1b. 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 the 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 (see the magnetic hysteresis loops shown in Fig. 1).

Then at the in-plane magnetic field value of $H_0 = \pm 110\,\mathrm{Oe}$ the temperature dependence of the resistivity R was recorded. In the following we focus on the spin valve sample #3 CoO_x(4 nm)/Fe1(2.4 nm)/Cu(4 nm)/Fe2(0.5 nm)/In(230 nm) (see Fig. 2).

For this sample $\Delta T_c = T_c^{\mathrm{AP}} - T_c^{\mathrm{P}} = 19\,\mathrm{mK}$ (see Fig. 2b with an enlarged temperature scale). We also performed similar resistivity measurements of the reference sample #2R with only one Fe layer. For this sample we found $T_c = 1.60\,\mathrm{K}$, which does not depend on the magnetic field direction (see Fig. 2c). This T_c value is lower than that for the In single layer film (sample #1) and higher than for sample #3 (Fig. 2). 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 SC 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 ΔT_c decreases and becomes practically zero at 2.6 nm thick F2 layer.

The observed shift $\Delta T_c = 19\,\mathrm{mK}$ is not the largest one among the data published before (cf., e.g., Ref. [20], where $\Delta T_c \simeq 41\,\mathrm{mK}$ at $\delta T_c \sim 100\,\mathrm{mK}$). However, very importantly it is substantially larger than δT_c which is of the order of 7 mK for sample #3 at $H_0 = 110\,\mathrm{Oe}$. 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 ΔT_c and switching the magnetic field between +110 and −110 Oe. This central result of this part of our review is shown in Fig. 3. It gives straightforward evidence for a complete on/off switching of the SC current flowing through the sample.To the best of our knowledge this result is the first example of the realization of the full spin valve effect for the superconducting current for the layered system with the ideal contact between the layers.

2.2. Interference effects

We studied the dependence of ΔT_c on the thickness of the intermediate layer Fe2 for the series of the samples CoO_x(4 nm)/Fe1(3 nm)/Cu(4 nm)/Fe2(d_{Fe2})/In(230 nm) with the value of d_{Fe2} lying between 0.4 and 5.2 nm. The values of the switching field H_0 around ±100 Oe turn out to be optimal for the operation of our spin valve samples. Higher values of H_0 bring the system too close to the critical field H_c thus reducing strongly the T_c. On the other hand, a reduction of H_0 substantially below ~ 100 Oe has also a negative twofold impact. The final width of the minor hysteresis loop is related to the formation of magnetic domains in the F2 layer, whereas the width of the loop decreases with increasing the thickness of the layer d_{F2} (Fig. 4a and b). As soon as H_0 gets smaller than the field strength necessary to fully polarize the magnetization of the F2 layer in the P or AP configuration, the F2 layer enters the domain state. This yields a strong broadening of the width of the superconducting transition δT_c (Fig. 4c and d) which clearly correlates with the width of the hysteresis loop. The increase of δT_c is obviously related to the occurrence of the inhomogeneous stray field perpendicular to the In film which is induced by the domains. Just in this geometry superconductivity in the In layer is extremely sensitive to magnetic fields. The second negative effect is the reduction of the difference in the T_c for the two opposite directions of the applied field $T_c(-H_0) - T_c(H_0)$ (Fig. 4e and f), since due to the domains neither the AP nor P configuration can be fully reached at small H_0. Therefore the most optimal regime for the operation of the spin valve can be achieved in our samples by a *full suppression* of the domain state by application of a strong enough switching field H_0 which yet should not be too close to H_c.

By studying our spin valve samples in this optimal regime we have observed a remarkable change of the *sign* of the spin valve effect with increasing the thickness of the free F2 layer d_{Fe2} (Fig. 4f and Fig. 5). It is positive, as expected, in the thickness range $0.4\,\mathrm{nm} \leq d_{\mathrm{Fe2}} \leq 0.8\,\mathrm{nm}$.

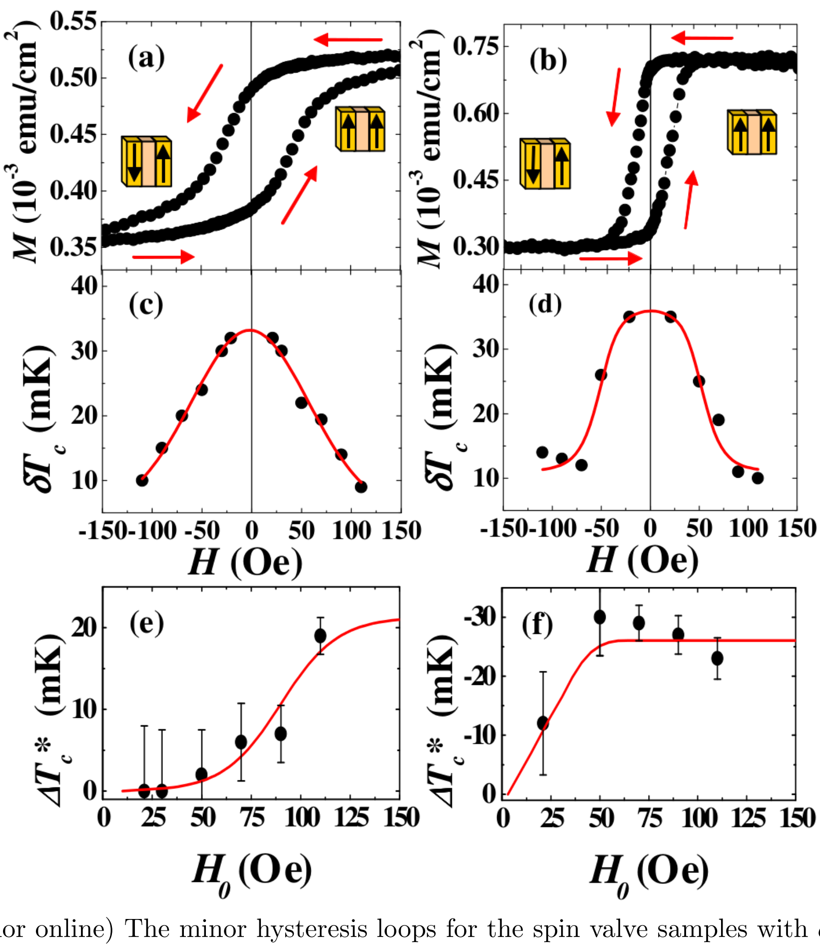

Figure 4. (Color online) The minor hysteresis loops for the spin valve samples with $d_{\mathrm{Fe2}} = 0.5$ nm (a) and $d_{\mathrm{Fe2}} = 1.3$ nm (b) and the dependencies of δT_c and ΔT_c^* on the magnetic field value: the panels (c) and (e) correspond to the sample with $d_{\mathrm{Fe2}} = 0.5$ nm and the panels (d) and (f) correspond to the sample with $d_{\mathrm{Fe2}} = 1.3$ nm [32, 33].

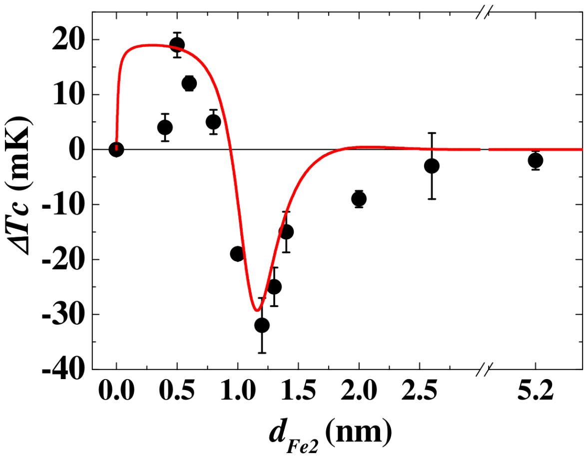

Figure 5. (Color online) The dependence of the T_c shift $\Delta T_c = T_c^{\mathrm{AP}} - T_c^{\mathrm{P}}$ on the Fe2-layer thickness d_{Fe2}. The applied switching field $H = \pm 110$ Oe. Theoretical curve (solid line) corresponds to the calculated function [W(0)-W(π)]/W(0) (see [38]) normalized to our experimental data [32,33]

Surprisingly, for a rather broad range of thicknesses $1\,\mathrm{nm} \leq d_{\mathrm{Fe2}} \leq 2.6\,\mathrm{nm}$ the spin valve effect has a negative sign, i.e. the T_c for the parallel mutual orientation of the magnetizations of the F1 and F2 layers is larger than for the antiparallel orientation. Moreover, the magnitude ΔT_c of this inverse effect is even larger than that of the positive direct effect (Fig. 5).

In the following we will discuss this striking observation with regard to three scenarios: (i) occurrence of magnetic domains in the F-layers; (ii) spin accumulation in the S-layer; (iii) quantum interference of the Cooper pair wave function in the S/F multilayer. "Old" theory of the spin valve effect based on the S/F proximity effect [14] predicts only the direct effect, i.e. $\Delta T_c > 0$. The inverse effect with $T_c^{\mathrm{AP}} < T_c^{\mathrm{P}}$ has been reported earlier for various systems. Its origin was discussed in terms of two, in fact conflicting, scenarios (i) and (ii). In model (i) magnetic domains may influence superconductivity in two different ways. Rusanov *et al.* [36] who studied Nb/Permalloy bilayers showed that the F-layer forms a domain state near its coercive field and the S-layer experiences a lowered average exchange field seen by the Cooper pairs. This yields a direct effect which may be called a Néel's domain wall induced enhancement of T_c.

Model (ii) is based on the giant magnetoresistance effect and predicts that the spin-polarized charge carriers should experience an enhanced spin-dependent reflection at the S/F interface in the AP state. Hence, they can accumulate in the S-layer which gives rise to a reduction of the superconducting energy gap, provided that the thickness of the S-layer is smaller than the spin diffusion length [37].

The first two scenario can be surely excluded in our case bearing in mind the field dependences of different parameters (Fig. 4). As to the third scenario indeed in a ferromagnetic layer the Cooper pair acquires a nonzero momentum due to the Zeeman splitting of electron levels and thus its wave function should oscillate in space (see, e.g., [10–13]). If the F-layer is sufficiently thin, the wave function reflected from the surface of the F-layer opposite to the S/F interface can interfere with the incoming one. Depending on the layer's thickness the interference at the S/F interface may be constructive or destructive. This should apparently lead to the enhancement of T_c or its decrease, respectively, thus naturally explaining our main result (Fig. 5).

Interestingly, there is a recent theory [38] where the same spin switch scheme F1/F2/S is considered. The starting points there do not strictly comply with the properties of our samples: F-layers were assumed to be weak ferromagnets, simplified boundary conditions were taken implying a 100 % transparency of the F2/S and F1/F2 interfaces for the electrons and superconductivity in the "dirty" limit in the F-layer ($l_F < \xi_F$) were assumed. Here l_F is the mean free path of conduction electrons. In our samples the F-layer made of iron is a strong ferromagnet with the penetration depth of the Cooper pairs into the F-layer $\xi_F \sim 1\,\mathrm{nm}$. In this case the transparency of the S/F interface should be reduced due to the exchange splitting of the conduction band in the F-layer [9]. Also the "dirty" limit is not realized owing to a small value of ξ_F. Finally the considered model does not involve the presence of the N-layer and assumes the F1-layer to be a half infinite ferromagnetic layer.

However, it is known that in practice the S/F proximity theories developed for the "dirty" limit deliver reliable results even beyond the domain of their applicability. Indeed, despite these differences we were able to obtain a reasonably good qualitative agreement between this theory and our experimental results as demonstrated by the fit curve to the experimental $\Delta T_c(d_{\mathrm{Fe2}})$-dependence in Fig. 5. An appreciable discrepancy with the experimental data point $d_{\mathrm{Fe2}} = 0.4\,\mathrm{nm}$ occurs most probably because at this thickness a transition from a continuous to an island like Fe film at even smaller thicknesses d_{Fe2} does take place. The fit parameters turned out to be quite realistic. Nevertheless, it is rather desirable to get independent parameters obtained from another source. As such source the $T_c(d_{\mathrm{Fe2}})$ dependence should give access to the parameters of the S-layer and enable an independent estimate of the parameters of the F-layers and of the S/F interface. This is due to the fact that the value of the spin valve effect is less than 30 mK and the influence of the Fe1-layer is negligible within the scale of variation of the $T_c(d_{\mathrm{Fe2}})$-dependence. A successful fitting of the $\Delta T_c(d_{\mathrm{Fe2}})$ dependence with this independent parameter set would provide an additional strong proof that interference of Cooper pair wave functions in the S/F proximity regime is responsible for the observed effects in the studied spin valve heterostructures.

The $T_c(d_{\mathrm{In}})$ dependence measured on a set of the samples with a fixed thickness of the Fe layer of 3 nm and varying In thickness d_{In} is shown in the inset of Fig. 6. It reveals a remarkably strong reduction of T_c with decreasing the d_{In} with a critical thickness for the vanishing of superconductivity $d_{\mathrm{In}}^{crit} \simeq 140\,\mathrm{nm}$.

A detailed dependence of the superconducting transition temperature T_c on the thickness of the Fe2 layer d_{Fe2} with the thickness of the In layer fixed at 230 nm is plotted in the main panel of Fig. 6. Comparing with the In stand alone film, the T_c drops sharply when introducing the Fe1 and Fe2 layers in the sample structure. A careful look on the data points for samples with $d_{\mathrm{Fe2}} \geq 0.5\,\mathrm{nm}$ reveals damped oscillations of the superconducting transition temperature as a function of d_{Fe2}. With increasing d_{Fe2} the T_c first increases up to a local maximum at 2.2 K, then slightly decreases, passing through a local minimum of 2.0 K at $d_{\mathrm{Fe2}} \simeq 1\,\mathrm{nm}$, after that slightly increases again to a second local maximum at $d_{\mathrm{Fe2}} \simeq 1.3\,\mathrm{nm}$ and finally saturates. Though these features are small, they cannot be ascribed to some sample artefacts or measurement uncertainties, but rather demonstrate a real physical property of the studied set of samples. The width of the superconducting transition for this series of samples does not exceed 30 mK. This means that the er-

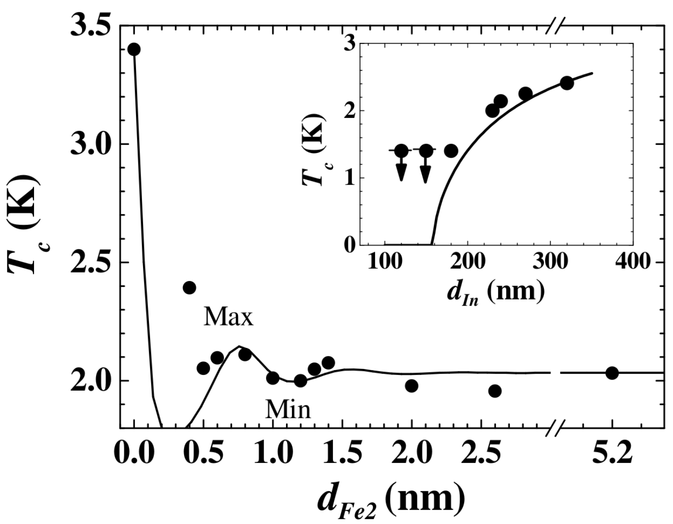

Figure 6. Superconducting transition temperature T_c versus the Fe2-layer thickness d_{Fe2} for the spin valve system CoO_x/Fe1/Cu/Fe2/In with a fixed thickness of the In layer $d_{\mathrm{In}} = 230\,\mathrm{nm}$. Inset shows the dependence of T_c on d_{In} for the set of the samples with a fixed thickness of the Fe layer $d_{\mathrm{Fe2}} = 3\,\mathrm{nm}$. Solid lines are theoretical fits (see the text) [33].

ror bars are smaller than the size of the data points in Fig. 6. Since we do not observe any broadening of the superconducting transition curve, the lateral length scale of the thickness fluctuations should be smaller than the superconducting coherence length. In this case the roughness parameter is irrelevant for the $T_c(d_{\mathrm{Fe2}})$ curve in Fig. 6. In addition our magnetization and ferromagnetic resonance results showing well defined sharp hysteresis loops and narrow resonance lines suggest that the Fe2 layer in our samples is continuous and uniform within the whole area of the film, at least down to 0.5 nm (the exceptional behavior of the sample with $d_{\mathrm{Fe2}} = 0.4$ nm is likely due to the island type of growth of the Fe2 layer at such small thickness). The easy axis of the Fe layers always lies in the plane of the film.

The proximity effect in bilayer S/F systems is usually described by the Usadel equations [39] valid in the "dirty" limit of the superconducting layer ($l \ll \xi_0$). For the interpretation of our results we can use a theory for the S/F proximity effect by Tagirov [40]. It takes the finite transparency of the interface explicitly into account and is applicable for the case of a magnetic layer made of a strong ferromagnet. Such approach has been used successfully before, e.g., for the description of the oscillating behavior of the T_c in Pb/Fe bilayers by Lazar *et al.* [9]. Following the procedure described in [9] we can produce a theoretical curve drawn by a solid line in Fig. 6 which models reasonably well the experimental $T_c(d_{\mathrm{Fe2}})$ dependence. In particular, the small oscillations of T_c in the range 0.5 nm $< d_{\mathrm{Fe2}} <$ 2 nm are well reproduced. Having established the set of parameters of the S/F proximity theory by Tagirov [40] for the $T_c(d_{\mathrm{Fe2}})$ dependence, we can revisit the analysis of the oscillating sign-changing dependence of the spin valve effect $\Delta T_c(d_{\mathrm{Fe2}})$ with the aid of the theory [38] (Fig. 5). Since the setups of these two theories are very different one should take care on the unification of the parameters. We have used these values as initial values in the modelling of the experimental dependence of the spin valve effect $\Delta T_c(d_{\mathrm{Fe2}})$ in the framework of the theory. Indeed, the best possible and quite satisfactory result of the modelling (Fig. 5) has been obtained by a minor variation of these initial values.

We have also performed the preliminary experimental study of the dependence of the spin valve effect on the thickness of the Fe1-layer [35]. In the theory [38] this layer was considered as half-infinite. We observed the monothonic increase of ΔT_c when decreasing the Fe1-layer thickness from 5 down to 1 nm at two fixed values of d_{Fe2}. We extended the oprevious theory [38] for arbitrary thickness of the outer F-layer (Fe1 in notation of the present paper). The theoretical curve for $\Delta T_c(d_{\mathrm{Fe1}})$ and experimental data seem to be consistent. Note that the theory also predicts peculiarities of the spin valve effect at very small d_{Fe1} (due to interference features of the oscillatory proximity effect in the F-part), however we do not focus on them since we do not have experimental data for such small thicknesses due to island growth of films.

2.3. Triplet superconductivity

Theory [38] predicts inevitable arising of the long range superconducting triplet component in the studied scheme of the spin valve.

In this part of our review the experimental evidence for generation of this component in the superconducting condensate in the multilayer spin valve heterostructure CoO_x/Fe1/Cu/Fe2/Pb where Lead stands instead of Indium is presented [34]. The basis of the present work has been formed by our earlier studies of the superconducting spin valve effect in the multilayer system CoO_x/Fe1/Cu/Fe2/In [31]. According to the theory [38] the activation of the triplet channel should be visible in an additional suppression of T_c for noncollinear arrangements of $\mathbf{M}_{\mathrm{Fe1}}$ and $\mathbf{M}_{\mathrm{Fe2}}$. Unfortunately, such experiment on the CoO_x/Fe1/Cu/Fe2/In system turned out to be unrealizable under well-controlled conditions owing to a low value of T_c for indium and its extreme sensitivity to small out-of-plane tilting of the external magnetic field. In this respect Lead has much better superconducting critical parameters, which has determined its choice as an S-layer in the present work. The triplet contribution manifests itself as an observation in all spin valve samples of a very special dependence of the superconducting critical temperature T_c on the angle α which the magnetization of the Fe2 layer $\mathbf{M}_{\mathrm{Fe2}}$ controlled by an external field makes with the magnetization of the pinned Fe1 layer. Examples of such dependences for selected spin-valve samples of different thickness d_{Fe2} are shown in Fig. 7. One can see that when changing the mutual orientation of magnetizations by a gradual in-plane rotation of the magnetic field from the P ($\alpha = 0°$) to the AP ($\alpha = 180°$) state T_c value does not change monotonically but passes through a minimum. Importantly, for the reference sample consisting of just one Fe layer the angular variation of T_c lies within the error bars (not shown). In the following we argue that a characteristic minimum in the $T_c(\alpha)$ close to $\alpha = 90°$ is a fingerprint of a long-range triplet superconducting component. The curve shown in Fig. 7 (left side panels) by dashed line displays imaginary monotonous angular dependence of T_c without taking into account for the triplet superconductivity. This is reference curve. Deviations δT_c of the actual T_c from the reference curves are, as the figures demonstrate, beyond the experimental error bars.

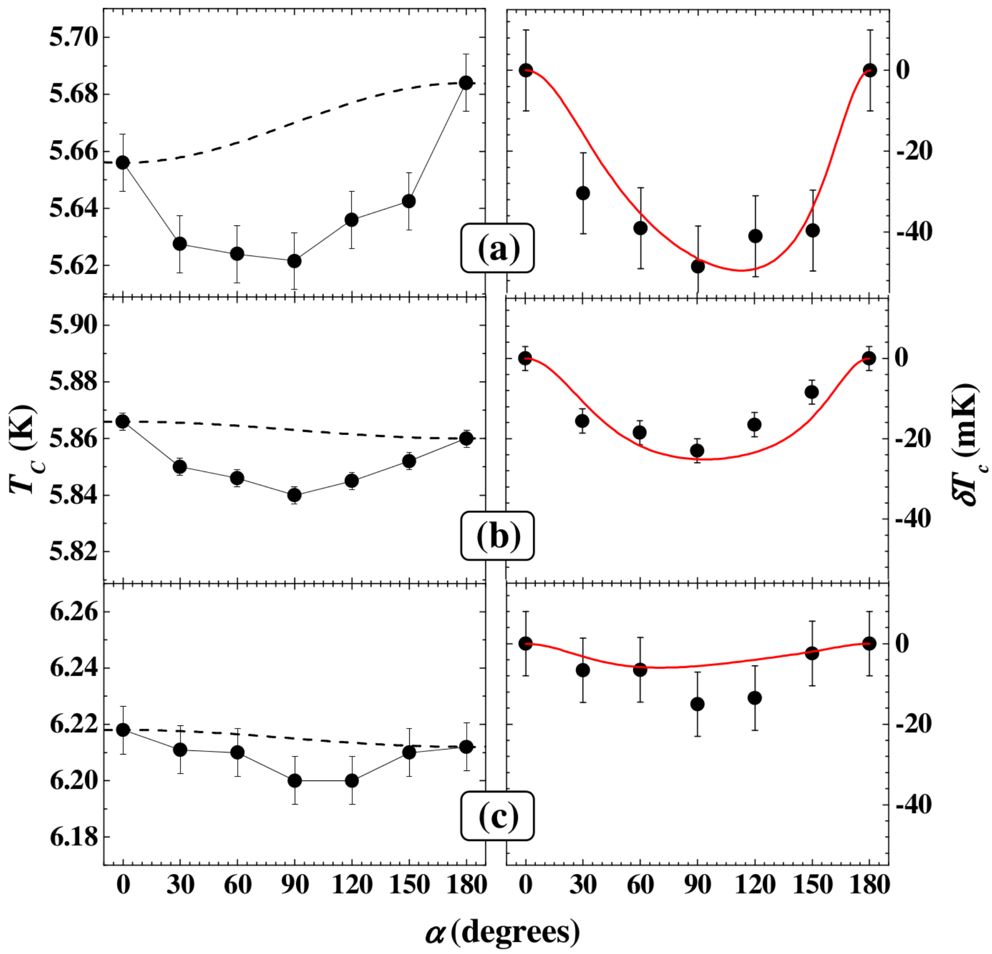

Figure 7. (Color online) Left: dependence of the T_c on the angle between magnetizations of the Fe1 and Fe2 layers measured in a field $H = 1\,\mathrm{kOe}$ for the samples with $d_{\mathrm{Fe2}} = 0.6\,\mathrm{nm}$ (a), $1.0\,\mathrm{nm}$ (b) and $1.5\,\mathrm{nm}$ (c). Dashed lines are the reference curves calculated without taking into account for triplet superconductivity. Right: deviations δT_c of the actual T_c values from the respective reference curves. Solid lines are theoretical results for δW (see the text) [34].

The angular dependences of this deviation are shown on the right side panels (a), (b), and (c) of Fig. 7.

The dependence of the maximal deviation of T_c demonstrates monothonic decrease with increasing d_{Fe2}. Theory demonstrates good agreement with theoretical results.

Bearing in mind that the effect is not observed for the reference sample CoO_x/Cu/Fe/Pb with a single iron layer we interpret our finding as evidence for long-range triplet superconductivity that arises in the spin-valve samples with a noncollinear geometry of magnetizations of the Fe1 and Fe2 layers.

The suppression of T_c in the S layer of an S/F1/F2 proximity system studied in our work takes place due to "leakage" of Cooper pairs into the F-part. In this language, the generation of the long-range triplet superconductivity at noncollinear magnetizations opens up an additional channel for this leakage, hence T_c should be suppressed stronger. Note that the triplet superconducting correlations are generated from the singlet ones (conversion due to the exchange field), reducing the amplitude of the singlet component in the S layer and thus "draining the source" of superconductivity in the whole system. This effect is substantial since the magnitudes of the proximity-induced singlet and long-range triplet superconductivity can be of the same order near the interface of the S-layer (if the thickness of the adjacent F-layer is smaller than its coherence length).

Finally we mention, that earlier indications for long-range superconductivity in an F layer have been detected through the proximity-induced conductance [41, 42] even before the theoretical works have appeared. Recently the occurrence of the odd in the Matsubara frequency triplet superconductivity in the S/F/S systems, predicted in Ref. [28] was inferred from the experiments on Josephson junctions through observation of the anomalously deep penetration of the Cooper condensate into the F layer (see e.g. [43–49]). We note that our experiments are advantageous in that they address the primary SC parameter of the spin valve, the behavior of T_c, which is directly affected by the spin-triplet component. And, finally, recently [50] for the construction of the spin valve similar to the studied by us an evidence for generation of long range triplet pairing was also obtained.

3. Conclusions

In the present works we were able to realize experimentally the idea by Sanjiun Oh *et al.* [14] for the first time. We have also presented experimental evidence for the oscillating behavior of the spin valve effect in a ferromagnetic/superconductor multilayer F1/N/F2/S with a varied thickness of the ferromagnetic F2-layer. We have observed the direct spin valve effect for F2-layer thicknesses smaller than the decay length ξ_F of the Cooper pair wave function in the F2-layer and the inverse spin valve effect for larger thickness up to $2.5 \cdot \xi_F$. The analysis of the data suggests that the inverse spin valve effect is likely caused by the interference effects for the superconducting pairing function reflected from both surfaces of the F2-layer. And, finally, we have observed a remarkable nonmonotonic dependence of T_c on the angle between the directions of the magnetization $\mathbf{M}$ in the Fe1 and Fe2 layers. The T_c passes through a clear minimum near the orthogonal orientation of $\mathbf{M}_{\mathrm{Fe1}}$ and $\mathbf{M}_{\mathrm{Fe2}}$ which is not expected in the case of singlet superconductivity. We argue that this particularly strong suppression of T_c in the orthogonal geometry is due to an enhanced "leakage" of the SC Cooper pairs into the F layer occurring via the long-range spin-triplet channel.

Acknowledgments

This work was partially supported by RFBR (Grants No. 13-02-01389-a, 13-02-97037-r-Povolzhje-a and 14-02-00350-a), by Programs of the RAS, by the Ministry of Education and Science of the Russian Federation, and the program “5top100”.

References

1. Ioffe L.B., Geshkenbein V.B., Feigel’man M.V., Fauchère A.L., Blatter G. *Nature* **398**, 679 (1999)

2. Feigel’man M.V. *Physics-Uspekhi* **42**, 823 (1999) [*Uspehi Fiz. Nauk* **169**, 917 (1999), in Russian]

3. Ryazanov V.V. *Physics-Uspekhi* **42**, 825 (1999) [*Uspehi Fiz. Nauk* **169**, 920 (1999), in Russian]

4. Ryazanov V.V., Oboznov V.A., Veretennikov A.V., Rusanov A.Yu. *Phys. Rev. B* **65**, 020501 (2001)

5. Veretennikov A.V., Ryazanov V.V., Oboznov V.A., Rusanov A.Yu., Larkin V.A., Aarts J. *Physica B* **284-288**, 495 (2000)

6. Ryazanov V.V., Oboznov V.A., Rusanov A.Yu, Veretennikov A.V., Golubov A.A., Aarts J. *Phys. Rev. Lett.* **86**, 2427 (2001)

7. Kontos T., Aprili M., Lesueur J., Grison X. *Phys. Rev. Lett.* **89**, 137007 (2002)

8. Ryazanov V.V., Oboznov V.A., Bol’ginov V.V., Prokof’ev A.S., Feofanov A.K. *Physics-Uspekhi* **47**, 732 (2004) [*Uspehi Fiz. Nauk* **174**, 795 (2004), in Russian]

9. Lazar L., Westerholt K., Zabel H., Tagirov L.R., Goryunov Yu.V., Garif’yanov N.N. Garifullin I.A. *Phys. Rev. B* **61**, 3711 (2000)

10. Izjumov Yu.A., Proshin Yu.N., Khusainov M.G. *Physics-Uspekhi* **45**, 109 (2002) [*Uspehi Fiz. Nauk* **172**, 113 (2002), in Russian]

11. Buzdin A.I. *Rev. Mod. Phys.* **77**, 935 (2005)

12. Bergeret F.S., Volkov A.F., Efetov K.B. *Rev. Mod. Phys.* **77**, 1321 (2005)

13. Efetov K.B., Garifullin I.A., Volkov A.F., Westerholt K. *Magnetic Heterostructures. Advances and Perspectives in Spinstructures and Spintransport.* Series Springer Tracts in Modern Physics, V. 227, Berlin, Springer (2007), P. 252 [arxiv: cond-mat/0610708v2]

14. Oh S., Youm D., Beasley M.R. *Appl. Phys. Lett.* **71**, 2376 (1997)

15. Tagirov L.R. *Phys. Rev. Lett.* **83**, 2058 (1999)

16. Buzdin A.I. , Vedyaev A.V., Ryzhanova N.N. *Europhys. Lett.* **48**, 686 (1999)

17. Baladié I. , Buzdin A., Ryzhanova N., Vedyayev A. *Phys. Rev. B* **64**, 054518 (2001)

18. Gu J.Y., You C.Y., Jiang J.S., Pearson J., Bazaliy Ya.B., Bader S.D. *Phys. Rev. Lett.* **89**, 267001 (2002)

19. Potenza A., Marrows C.H. *Phys. Rev. B* **71**, 180503(R) (2005)

20. Moraru I.C., Pratt Jr.W.P., Birge N.O. *Phys. Rev. Lett.* **96**, 037004 (2006)

21. Miao G.-X., Ramos A.V., Moodera J. *Phys. Rev. Lett.* **101**, 137001 (2008)

22. Westerholt K., Sprungmann D., Zabel H., Brucas R., Hjörvarsson B., Tikhonov D.A., Garifullin I.A. *Phys. Rev. Lett.* **95**, 097003 (2005)

23. Nowak. G., Zabel H., Westerholt K., Garifullin I., Marcellini M., Liebig A., Hjörvarsson B. *Phys. Rev. B* **78**, 134520 (2008)

24. Proshin Yu.N., Zimin A., Fazleev N.G., Khusainov M.G. *Phys. Rev. B* **73**, 184514 (2006)

25. Avdeev M.V., Proshin Yu.N. *JETP* **117**, 1101 (2013) [*Zh. Eksp. Teor. Fiz.* **144**, 1251 (2013), in Russian]

26. Avdeev M.V., Tsarevskii S.L., Proshin Yu.N. *Journ. Korean Phys. Soc.* **63**, 466 (2013)

27. Bergeret F.S., Volkov A.F., Efetov K.B. *Phys. Rev. Lett.* **86**, 4096 (2001)

28. Volkov A.F., Bergeret F.S., Efetov K.B. *Phys. Rev. Lett.* **90**, 117006 (2003)

29. Mel'nikov A.S., Samokhvalov A.V., Kuznetsova S.M., Buzdin A.I. *Phys. Rev. Lett.* **109**, 237006 (2012)

30. Efetov K.B., Garifullin I.A., Volkov A.F., Westerholt K. *Magnetic Nanostructures. Spin Dynamic and Spin Transport.* Series Springer Tracts in Modern Physics, Vol. 246, Hartmut Zabel, Michael Farle (Eds.), Springer-Verlag, Berlin Heidelberg (2013), P. 85–118

31. Leksin P.V., Garif'yanov N.N., Garifullin I.A., Schumann J., Vinzelberg H., Kataev V., Klingeler R., Schmidt O.G., Büchner B. *Appl. Phys. Lett.* **97**, 102505 (2010)

32. Leksin P.V., Garif'yanov N.N., Garifullin I.A., Schumann J., Vinzelberg H., Kataev V., Klingeler R., Schmidt O.G., Büchner B. *Phys. Rev. Lett.* **106**, 067005 (2011)

33. Leksin P.V., Garif'yanov N.N., Garifullin I.A., Schumann J., Vinzelberg H., Kataev V., Klingeler R., Schmidt O.G., Büchner B. *Phys. Rev. B* **85**, 024502 (2012)

34. Leksin P.V., Garif'yanov N.N., Garifullin I.A., Fominov Ya.V., Schumann J., Krupskaya Y., Kataev V., Schmidt O.G., Büchner B. *Phys. Rev. Lett.* **109**, 057005 (2012)

35. Leksin P.V., Kamashev A.A., Garif'yanov N.N., Garifullin I.A., Fominov Ya.V., Schumann J., Hess C., Kataev V., Büchner B. *JETP Lett.* **97**, 549 (2013) [*Pis'ma Zh. Eksp. Teor. Fiz.* **97**, 478 (2013), in Russian]

36. Rusanov A.Yu., Hesselberth M., Aarts J., Buzdin A.I. *Phys. Rev. Lett.* **93**, 057002 (2004)

37. Rusanov A.Yu., Habraken S., Aarts J. *Phys. Rev. B* **73**, 06050 (2006)

38. Fominov Ya.V., Golubov A.A., Karminskaya T.Yu., Kupriyanov M.Yu., Deminov R.G., Tagirov L.R. *JETP Lett.* **91**, 308 (2010) [*Pis'ma Zh. Eksp. Teor. Fiz.* **91**, 329 (2010), in Russian]

39. Usadel K.D. *Phys. Rev. Lett* **25**, 507 (1970)

40. Tagirov L.R. *Physica C* **307**, 145 (1988)

41. Giroud M., Courtois H., Hasselbach K., Mailly D., Pannetier B. *Phys. Rev. B* **58**, R11872 (1998)

42. Petrashov V.T., Sosnin I.A., Cox I., Parsons A., Troadec C. *Phys. Rev. Lett.* **83**, 3281 (1999)

43. Sosnin I., Cho H., Petrashov V.T., Volkov A.F. *Phys. Rev. Lett.* **96**, 157002 (2006)

44. Keizer R.S., Goennenwein S.T.B., Klapwijk T.M., Miao G., Xiao G., Gupta A. *Nature* **439**, 825 (2006)

45. Wang J., Singh M., Tian M., Kumar N., Liu B., Shi C., Jain J.K., Samarth N., Mallouk T.E., Chan M.H.W. *Nature Physics* **6**, 389 (2010)

46. Robinson J.W.A., Witt J.D.S., Blamire M.G. *Science* **329**, 59 (2010)

47. Anwar M.S., Czeschka F., Hesselberth M., Porcu M., Aarts J. *Phys. Rev. B* **82**, 100501 (2010)

48. Khaire T.S., Khasawneh M.A., Pratt, Jr W.P., Birge N.O. *Phys. Rev. Lett.* **104**, 137002 (2010)

49. Sprungmann D., Westerholt K., Zabel H., Weides M., Kohlstedt H. *Phys. Rev. B* **82**, 060505 (2010)

50. Zdravkov V.I., Kehrle J., Obermeier G., Lenk D., Krug von Nidda H.-A., Müller C., Kupriyanov M.Yu., Sidorenko A.S., Horn S., Tidecks R. Tagirov L.R. *Phys. Rev. B* **87**, 144507 (2013)